Dilute ferromagnetic semiconductors: Physics and spintronic structures

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This review compiles results of experimental and theoretical studies on thin films and quantum structures of semiconductors with randomly distributed Mn ions, which exhibit spintronic functionalities associated with collective ferromagnetic spin ordering. Properties of p-type Mn-containing III-V as well as II-VI, IV-VI, V2VI3, I-II-V, and elemental group IV semiconductors are described paying particular attention to the most thoroughly investigated system (Ga,Mn)As that supports the hole-mediated ferromagnetic order up to 190 K for the net concentration of Mn spins below 10%. Multilayer structures showing efficient spin injection and spin-related magnetotransport properties as well as enabling magnetization manipulation by strain, light, electric fields, and spin currents are presented together with their impact on metal spintronics. The challenging interplay between magnetic and electronic properties in topologically trivial and non-trivial systems is described, emphasizing the entangled roles of disorder and correlation at the carrier localization boundary. Finally, the case of dilute magnetic insulators is considered, such as (Ga,Mn)N, where low temperature spin ordering is driven by short-ranged superexchange that is ferromagnetic for certain charge states of magnetic impurities.

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I. INTRODUCTION

It has been appreciated for a long time that materials systems combining the tunability of semiconductors with the spin contrast specific to ferromagnets offer a rich spectrum of outstanding properties which are attractive per se as well as open prospects for an entirely new set of functionalities. From the materials physics perspective there appear three main roads bridging semiconductors and ferromagnets. The first of them is to use hybrid structures consisting of ferromagnetic metals and semiconductors or oxide semiconductors [Zutic et al. 2004]. Here, as an example of commercially relevant development, one can quote the trilayer structure FeCoB/MgO/FeCoB. In this magnetic tunnel junction, owing to specific symmetry mismatch of wave functions at high quality interfaces, the tunneling resistance increases more than sevenfold at room temperature when magnetization of two ferromagnetic layers becomes antiparallel [Ikeda et al. 2008]. The second possible strategy is to turn ferrimagnetic oxides, such as (Zn,Ni)Fe$_2$O$_4$ showing spontaneous magnetization up 800 K, into good semiconductors by mastering carrier doping and interfacing of these compounds with main stream semiconductors and metals. The third road is to develop semiconductors supporting spontaneous spin polarization, preferably, up to above the room temperature.

This review focuses on epitaxially grown Mn-containing III-V but also II-VI, IV-VI, V$_2$-VI$_3$, I-II-V, and elemental group IV semiconductors, in which randomly distributed spins of Mn ions show collective ferromagnetic ordering. While the list of the dilute ferromagnetic semiconductors studied so-far is long, certainly the most extensively investigated compound is (Ga,Mn)As [Ohno et al. 1996] and, accordingly, a large part of this review is devoted to this system. It has been demonstrated over the last decade or so that owing to previously unavailable combination of quantum structures and ferromagnetism in semiconductors, the engineered structures of these systems show a variety of new physical phenomena and functionalities. In fact, a series of accomplishments in this field accounts, to a large extent, for spreading of spintronic research over virtually all materials families.

At the same time, however, over the course of several years the studies of semiconductors showing ferromagnetic features have emerged as one of the most controversial fields of today’s condensed matter physics and materials science. It becomes increasingly clear that there are four principal reasons for this state of the matter:

- Challenging the natural assumption [fulfilled in, e.g., Mn-based II-VI dilute magnetic semiconductors (DMSs)] that the transition metal (TM) impurities substitute randomly distributed cation sites, it appears that depending on epitaxy conditions, co-doping with shallow dopants, and post-growth processing, the magnetic ions can assume interstitial positions and/or aggregate, also with other defect centers, which affects crucially magnetic properties.

- A number of growth and processing methods exposes the studied samples to contamination by magnetic nanoparticles and whenever they reside (in the film or substrate volume, at the surface or interface) can determine the magnetic response of the system, particularly in the thin film form.

- These materials, including (Ga,Mn)As, show simultaneously intricate properties of mismatch semiconductors alloys [such as Ga(As,N)] and of doped semiconductors on the localization verge (such as GaAs:Si). The controversies here echo much dispute, and by some regarded as still unsettled questions, of whether in the relevant range of concentrations the impurity levels (derived from N or Si in...
the above two examples) are dissolved in the band continuum or form resonant or band gap states, respectively.

- Considerable effort has been devoted to describe DMSs from first principles (ab initio), employing various implementations of the density-functional theory (DFT), particularly involving the local spin-density approximation (LSDA) and its variants. It becomes increasingly clear that inaccuracies of this approach, such as the placement of $d$ levels too high in energy and the underestimation of the band gap, have twisted the field, for instance, by indicating that the double exchange dominates in (In,Mn)As and that ferromagnetism exists in intrinsic (Zn,Co)O.

As emphasized in this review, the present understanding of the field and, in particular, the progress in resolving the above controversial issues as well as a successful modeling of spintronic functionalities are built on two experimental and two conceptual pillars:

- advanced nanoscale characterization allowing to assess the location and distribution of magnetic ions, dopants, defects, and carriers;
- comprehensive spectroscopic data providing information on the position of levels introduced by TM ions, their spin and charge states, as well as the coupling to band and/or local states;
- careful consideration of host band structure, taking into account thoroughly interband and spin-orbit couplings, confinement effects, as well as the presence of surface and edge states in topologically non-trivial cases;
- realization that the realm of quantum (Anderson-Mott) localization underlines transport and optical phenomena in carrier-controlled ferromagnetic DMSs.

According to the accumulated insight, most of magnetically doped semiconductors and semiconductor oxides, which exhibit ferromagnetic features can be grouped into two main classes:

1. Uniform DMSs, in which ferromagnetic behavior originates from randomly distributed TM cations. In most cases [the flagship example being (Ga,Mn)As] the spin-spin interactions are mediated by a high density $p$ of delocalized or weakly localized holes. The confirmed magnitude of the Curie temperature $T_C$ approaches 190 K in Ga$_{1-x}$Mn$_x$As [Olejnik et al. 2008, Wang et al. 2008b] and Ge$_{1-x}$Mn$_x$Te [Fukuma et al. 2008, Hassan et al. 2011] with saturation magnetization (in moderate fields, $\mu_B H \lesssim 5$ T) corresponding to less than 10% of Mn cations. In the absence of itinerant carriers other coupling mechanisms, such as ferromagnetic superexchange in (Ga,Mn)N [Bonanni et al. 2011, Sawicki et al. 2012], can account for ferromagnetic spin ordering. Since with no carriers the coupling is short ranged, the $T_C$ values reach only about 13 K at $x \approx 10\%$ in Ga$_{1-x}$Mn$_x$N [Stefanowicz et al. 2013].

2. Heterogenous DMSs, specified by a highly non-random distribution of magnetic elements. Here, ferromagnetic-like properties persisting typically to above room temperature are determined by nanoregions with high concentrations of magnetic cations, brought about by chemical or crystallographic phase separation [Bonanni and Dietl 2010]. To this family belong also numerous materials systems, in which ferromagnetic-like properties – persisting up to high temperatures – appear related rather to defects than to the presence of TM-rich regions [Coey et al. 2008].

The studies of the compounds belonging to the first class is undoubtedly the most mature. On the one hand, significant advances in epitaxy and post-growth processing allowed one to develop a class of ferromagnetic semiconductors, primarily (Ga,Mn)As, showing textbook thermodynamic and micromagnetic characteristics, despite inherent alloy disorder and a relatively small concentration of the magnetic constituent. More importantly, the progress in controlling and understanding of these materials has provided a basis for demonstrating novel methods enabling magnetization manipulation and switching as well as spin injection, sensing of the magnetic field, and controlling of the electric current by magnetization direction, the accomplishments having now a considerable impact on the metal spintronics [Ohno 2010]. At the same time, over the course of the years, ferromagnetic DMSs, particularly their magnetic phase diagrams $T_C(x,p)$ and micromagnetic properties, have become a test bench for various theoretical and computational methods of materials science.

In contrast, the control, understanding, and functionalization of the second class of materials systems is in its infancy. However, one may expect a number of developments in the years to come as the availability of materials systems with modulated semiconductor and metallic ferromagnetic properties at the nano-scale, which persist up to above the room temperature, opens new horizons for basic and applied research.

Our aim here is to survey various properties of uniform Mn-based ferromagnetic DMSs, which we refer to as dilute ferromagnetic semiconductors (DFSs). As seen in the Table of Contents, the main body of the present review consists of three major parts.

First we discuss epitaxial growth and nanocharacterization of DFSs (Sec. II). We put a particular emphasis on the question of the position and spatial distribution of magnetic ions, which is essential in understanding pertinent properties of any DMSs. We also touch upon the issue of a non-uniform carrier distribution.
In the second part (Secs. III-VI), we present various outstanding spintronic capabilities of DFSs and their quantum structures with nonmagnetic semiconductors. In particular, we describe how hole-mediated ferromagnetism allows for magnetization manipulation and switching not only by doping or co-doping but also by strain, electric field, and light (Sec. III). Next the suitability of these systems for spin injection to non-magnetic semiconductors is discussed (Sec. IV). We also show that in addition to properties specific to semiconductor quantum structures, these materials exhibit functionalities presently or previously discovered in magnetic multilayers, including magnetization switching by an electric current and various magnetotransport phenomena (Sec. V) as well as inter-layer coupling, exchange bias, and ferromagnetic proximity effect (Sec. VI).

Finally, in the third part (Secs. VII-X), we present results on quantitative theoretical studies of thermodynamic, micromagnetic, and spintronic properties of DFSs. We start this part by describing the present understanding of the electronic structure of these systems and exchange coupling between localized spins and itinerant carriers (Sec. VII). Equipped with this information, we present theoretical models of superexchange (Sec. VIII) and carrier-mediated ferromagnetism in DFSs (Sec. IX). Exploiting detailed information on the band structure effects, spin-orbit coupling, and p-d hybridization provided by extensive spectroscopic studies on relevant DMSs, these models allow for a computationally efficient interpretation of experimental findings with no adjustable parameters (Sec. X). Along with emphasizing success of this experimentally constrained approach to the understanding of basic properties and spintronic capabilities of DFSs, we indicate unsettled issues awaiting further experimental and theoretical investigations.

We conclude our review by discussing possible future directions in basic and applied studies of magnetically doped semiconductors (Sec. XI).

In this review, we purposively refrain from describing a historical perspective, intermediate or disproved/unconfirmed developments, and a variety of qualitative considerations that have been put forward but not yet shaped into the form allowing for a quantitative verification vis-à-vis experimental results with no adjustable parameters. We refer readers interested in a survey of various models proposed over the course of the years to explain the nature of electronic states and ferromagnetism in these systems to review articles on the theory of DFSs from the perspective of model Hamiltonians (Jungwirth et al. 2006a) and ab initio approaches (Sato et al. 2010; Zunger et al. 2010). A short paper presenting the topic in a condensed and tutorial way as well as explaining origins of various exchange mechanisms is also available (Bonanni and Dietl 2010). Earlier book chapters review thoroughly the pioneering works on II-VI (Dietl 1994; Furdyna and Kossut 1988) and III-V (Matsukura et al. 2002) DMSs. Two other surveys present successes and limitations of Drude-Boltzmann type models in describing abundant experimental results on transport (Jungwirth et al. 2008) and optical (Burch et al. 2008) phenomena in (Ga,Mn)As and related systems. Accordingly, we only briefly discuss these phenomena here, also realizing that there are not yet theoretical frameworks allowing for the quantitative description of absolute values of dc or ac conductivity tensor components in the regime of quantum localization, even in the absence of p-d coupling (Belitz and Kirkpatrick 1994; Lee and Ramakrishnan 1985).

II. GROWTH AND CHARACTERIZATION

A. Growth methods and diagrams

Some of DFSs can be grown by the thermal equilibrium Bridgman method, a primal example being IV-VI alloys, particularly p-Pb_{1-x}Sn_{x}Mn_{x}Te [Eggenkamp et al. 1993; Story et al. 1986], in which cation vacancies supplied a large concentration of holes mediating ferromagnetic coupling between Mn spins. The same growth technique delivered ferromagnetic Zn_{1-x}Mn_{x}Te:P [Kepa et al. 2003], in which P acceptors provided holes after appropriate annealing. Interestingly, the Bridgman method was successfully used to obtain rhombohedral Bi_{2-y}Mn_{y}Te_{3}, a ferromagnetic topological insulator, in which Mn ions that introduced both spins and holes, were found to be randomly distributed up to at least x = 0.09 (Hor et al. 2010). At the same time, solid state reaction was employed to synthesize polycrystalline p-Ge_{1-x}Mn_{x}Te [Cochrane et al. 1971], up to x = 0.5 and p-Li(Zn_{1-x}Mn_{x})As up to x = 0.15 (Deng et al. 2011), in which holes originated presumably from cation vacancies and Li substituting Zn, respectively.

However, rapid progress in the search for ferromagnetic DMSs stems, to a large extent, from the development of methods enabling material synthesis far from thermal equilibrium, primarily by molecular beam epitaxy (MBE) (Ohno 1998), but also by pulsed-laser deposition (PLD) (Fukumura et al. 2005), metalorganic vapor phase epitaxy (MOVPE) (Bonanni 2007), atomic layer deposition (ALD) (Lukasiewicz et al. 2012), sputtering (Fukumura et al. 2005), ion implantation (Pearson et al. 2003), and pulsed-laser melting of implanted layers (Scarpulla et al. 2008; Zhou et al. 2012). These methods have a potential to provide high-quality DMS films with a concentration of the magnetic constituent beyond the solubility limits at thermal equilibrium. Moreover, the use of these methods offers unprecedented opportunity for considering physical phenomena and device concepts for previously unavailable combination of quantum structures and ferromagnetism in semiconductors.

Figure 1 outlines the growth phase diagram of (Ga,Mn)As (Matsukura et al. 2002; Ohno 1998; Van Esch et al. 1997), which appears to be generic to a wide class of DMSs. Because of low solubility of TM impurities, typically a fraction of a percent, and the asso-
B. Importance of nanocharacterization

As already mentioned in Introduction, the rich materials physics of ferromagnetic DMSs stems to a large extent from non-anticipated forms of distributions and lattice positions assumed by magnetic ions, defects, and carriers in these systems as well as from their sensitivity to contamination by ferromagnetic nanoparticles. Importantly, rather than being specific to a given DMS, these striking properties depend sensitively on the employed substrate, growth conditions, co-doping, and post-growth processing. Four issues, relevant to DFSs, can be called into attention here.

1. Attractive interactions between magnetic impurities and their limited solubility can result in the highly non-random distribution of TM atoms over cation sites (chemical phase separation) or in TM precipitation in the form of compounds or elemental inclusions (crystallographic phase separation). Typically, the TM-rich nanocrystals formed in these ways dominate the magnetic response of the system. They are either randomly distributed over the film volume or tend to accumulate near the surface or interface. Atom diffusion on the growth surface is typically faster than in the bulk, which facilitates aggregation of magnetic cations to the form of TM-rich nanocrystals during the epitaxy.

2. Even if nanocrystals are not assembled, the attractive force between TM cations can enhance the concentration of nearest neighbor cation-substitutional TM dimers. Moreover, since on the surface (comparing to bulk) certain crystal directions are not equivalent, the dimers—if stable during the entire growth of misfit dislocations. Adapted from Matsukura et al. 2002.

FIG. 1 (Color online) Schematic diagram of the temperature window for growth of dilute magnetic semiconductor Ga$_{1-x}$Mn$_x$As by low temperature molecular beam epitaxy. With the increase of the Mn content $x$ the window shrinks and the concentration of Mn interstitials Mn$_1$ increases. The magnitude of biaxial strain is determined by the substrate lattice constant even beyond the critical thickness for the formation of misfit dislocations. Adapted from Matsukura et al. 2002.

The lowering of the substrate temperature to the 200–300°C range (Boeck et al., 1996; Munekata et al., 1989; Ohno et al., 1992; 1996; Van Esch et al., 1997) makes it possible to surpass the thermal equilibrium solubility limit and, at the same time, to maintain the two-dimensional coherent growth, as witnessed by the smoothness of the surface and the persistence of electron diffraction stripes over the entire process of the film deposition. The use of a cracker effusion cell for the anion source (Campion et al., 2003) as well as a careful adjustment of the ratio between cation and anion fluxes (Myers et al., 2006) allows one to minimize the concentration of point defects, such as As-antisite donors, which tend to form during low temperature MBE.

Importantly, owing to low deposition temperatures, strain associated with lattice mismatch to the substrate remains unrelaxed even for film thicknesses exceeding critical values for the formation of misfit dislocations under thermal equilibrium conditions. A uniformly strained (Ga,Mn)As film with the thickness of 6.8 µm was obtained employing (001) GaAs substrate for which lattice mismatch was $\Delta a/a \approx 0.4\%$ (Welp et al., 2004). The use of substrates with various lattice parameters and crystallographic orientations allows one to fabricate DFS films with tailored magnetic anisotropy characteristics (see, Sec. III.B).

Additionally, the low-temperature (LT) epitaxy process makes it possible to increase substantially the electrical activity of shallow impurities. For instance, by assisting MBE growth with nitrogen plasma, it was possible to introduce a sizable concentration of holes indispensable to mediate ferromagnetic coupling between Mn spins in (Zn,Mn)Te (Ferrand et al., 2000, 2001) and (Be,Mn)Te (Hansen et al., 2001; Sawicki et al., 2002). Another relevant approach is to employ the concept of modulation doping, successfully applied in (Cd,Mn)Te/(Cd,Mg,Zn)Te:N (Boukari et al., 2002; Haury et al., 1997), and also examined in the case of (Ga,Mn)As/(Al,Ga)As:Be (Wojtowicz et al., 2003a).

In addition to III-V and II-VI DFSs, the MBE method has been employed for deposition of (Ge,Mn) (Park et al., 2002) and (Ge,Mn)Te (Fukuma et al., 2008; Hassan et al., 2011; Knoff et al., 2009; Lim et al., 2011).

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growth process can assume a directional distribution that lowers alloy symmetry and, hence, modify magnetic anisotropy.

3. The upper limit of achievable carrier density in a given host is usually determined by the mechanism of self-compensation. In the case of hole doping the effect consists of the appearance of compensating donor-like point defects once the Fermi level reaches an appropriately low energy in the valence band. These defects not only remove carriers from the Fermi level but can form with TM ions defect complexes characterized by non-standard magnetic properties. In fact, TM ions can form complexes also with other defects or impurities.

4. Even for a perfectly random distribution of magnetic ions and carrier dopants, due to the relevance of quantum localization effects, there appear significant nano-scale spatial fluctuations in the hole density. Because of the relationship between carriers and magnetism, the value of magnetization ceases to be spatially uniform. Another source of inhomogeneity are space charge layers often forming at the surface or interface.

These outstanding properties of DMSs can be addressed by ever improving nanocharacterization tools involving synchrotron, electron microscopy, ion beam, and scanning probe methods. Some of experimental techniques relevant to DMSs have recently been reviewed [Bonanni, 2011]. This collection contains also useful information about the methodology of magnetic measurements on thin DMS films.

Next the above issues are described in some details paying particular attention to the data obtained for (Ga,Mn)As. Enlisted are also methods allowing to determine the concentration of holes and Mn ions if their distribution is, at least approximately, random.

C. Solubility limits and Mn distribution

It is well known that the phase diagrams of a number of alloys exhibit a solubility gap in a certain concentration range. Particularly low is the solubility of Mn in II-VI compounds, where Mn atoms remain distributed randomly over the substitutional cation sites up to concentrations often exceeding 50% [Furdyna and Kossut, 1988, Pajączkowska, 1978], even if the alloy is grown close to thermal equilibrium, as in the case of, e.g., the Bridgman method.

The large solubility of Mn in II-VI compounds can be associated to the truly divalent character of Mn whose d states little perturb the sp$^3$ tetrahedral bonds as both the lower d$^5$ (donor) and the upper d$^6$ (acceptor) Hubbard levels are respectively well below and above the band edges (Dietl, 1981, 2002, Zunger, 1986). This qualitative picture is supported by first principles computations, showing a virtual absence of an energy change associated with bringing two Zn-substitutional Mn atoms to the nearest neighbor cation sites in (Zn,Mn)Te, $E_d = 21$ meV [Kuroda et al., 2007].

According to the pioneering ab initio work [van Schilfgaarde and Mryasov, 2001] and to the subsequent developments [Da Silva et al., 2008; Kuroda et al., 2007; Sato et al., 2005; Ye and Freeman, 2006], a strong tendency to form non-random alloys occurs in the case of DMSs in which TM-induced states are close to the Fermi energy and thus contribute significantly, via the $p$-$d$ hybridization, to the bonding as well as can supply or trap carriers. For instance, the pairing energy of two Ga-substitutional Mn atoms is computed to be $E_d = -120$ meV in GaAs and $-300$ meV in GaN [van Schilfgaarde and Mryasov, 2001].

However, as already mentioned in Sec. [II.A], a sufficiently low magnitude of substrate temperature prevents the formation of hexagonal MnAs or zinc-blende Mn-rich (Mn,Ga)As nanocrystals in (Ga,Mn)As grown by MBE. Indeed, according to the newly developed three-dimensional atom probe technique (3DAP) that allows one to obtain 3D maps of elements’ distribution with a 1 nm resolution, the Mn distribution is uniformed along the growth direction and in-plane, without any evidence for Mn aggregation in the sample volume or Mn segregation at the interface, as shown in Figs. [2] and [3]. However, within the attained resolution, the presence of short range correlations, that is a formation of dimers of trimers, cannot be confirmed or ruled out. It is unclear at present to what extent this new method provides accurate information on the absolute values of the particular element concentration. The present data, as they stand, suggest a surplus of As and Mn in the studied slices.

The absence of Mn aggregation in (Ga,Mn)As obtained by low temperature MBE was confirmed by cross-sectional scanning tunneling tomography [Richardella et al., 2010].

In the case of wurtzite (wz) (Ga,Mn)N grown by MOVPE [Bonanni et al., 2011] and MBE [Kunert et al., 2012] a range of nanocharacterization methods indicate the absence of Mn aggregation in films grown under carefully adjusted conditions. A remarkable difference between (Ga,Mn)N and (Ga,Fe)N [in which the same methods reveal the formation of Fe-rich nanocrystals (Bonanni et al., 2008; Navarro-Quezada et al., 2011)] was explained by LSDA ab initio studies in terms of the repulsive ($E_d = 170$ meV) and attractive ($E_d = -120$ meV) interactions between the nearest neighbor cations pairs of Mn and Fe, respectively, on the growth surface (0001) of wz-GaN [Gonzalez Szwacki et al., 2011].
D. Formation of Mn dimers

Figure 4 presents the nearest neighbor Mn dimers residing on GaAs (001) surface along two crystallographic directions. As seen, in the [110] case the two Mn ions are connected by the same As atom, whereas there is no such As atom for the dimer along the [110] axis, implying that these two directions are not equivalent on the surface, in contrast to bulk dimers, for which there is an As bridge for these two cases – one below, one above the dimer plane. Furthermore, since the Mn-Mn interaction discussed in the previous subsection is brought about by p-d hybridization, one may expect much stronger attractive force for the [110] pair comparing to the [110] case. Indeed, \textit{ab initio} computations show that the corresponding difference in $E_d$ is as large as 1.0 eV (Birowska et al., 2012), much higher than growth temperature. Thus, if barriers for Mn diffusion along the surface are sufficiently small, a non-volatile asymmetry in the pair distribution will set in in the whole film during the epitaxy. A group theory analysis demonstrated that the corresponding lowering of symmetry leads to the appearance of two additional terms in the $kp$ Hamiltonian, which are of the form of effective biaxial and shear strains, $\epsilon_{xx}$ and $\epsilon_{xy}$, respectively (Birowska et al., 2012).

The asymmetry in the dimer distribution invoked by the above model has not yet been directly confirmed by any nanocharacterization method. However, it was suggested (Birowska et al., 2012) that strain associated with the formation of dimers along the [110] direction can trigger stacking faults propagating in the (111) and (111) planes, as observed in (Ga,Mn)As by high resolution electron transmission microscopy (Kong et al., 2005) and synchrotron x-ray diffraction (Kopecky et al., 2011).

E. Self-compensation – Mn interstitials and Mn complexes

In many cases TM impurities rather than residing in the substitution sites, prefer to occupy interstitial positions, the case of TM-doped Si (Zunger, 1986), but also of Mn in GaAs, as suggested theoretically (Mašek and Máca, 2001) and found experimentally (Yu et al., 2002). According to combined Rutherford backscattering (RBS) and particle-induced x-ray emission (PIXE)
concentration knowing only Mn concentrations (as indicated that it may not be possible to determine the hole concentration, $p$, with the total Mn content $N_I$). The presence of Mn in (Ga,Mn)As was confirmed by extended x-ray absorption fine structure (EXAFS) spectroscopy (Bacewicz et al., 2005) and transmission electron microscopy (TEM) (Glas et al., 2004). The interstitials appear to enlarge the (Ga,Mn)As lattice constant, according to x-ray diffraction (Mack et al., 2008) and theoretical studies (Masek et al., 2003).

Whilst the Mn impurity in the cation-substitutional site acts as a single acceptor in III-V compounds, it becomes a double donor in the interstitial position of the GaAs lattice (Masek and Maca, 2001) and GaAs structures deposited at low temperatures (Sec. III.A), lowers the value of $p$ further on,

$$p = N_0(x - 3x_1) - zN_D,$$

where $z = 1$ and $z = 2$ for the single and double donors, respectively, of the concentration $N_D$. This formula indicates that it may not be possible to determine the hole concentration knowing only Mn concentrations (x and $x_1$).

It appears natural to assume that mobile positively charged interstitials will occupy a void position next to the negatively charged Mn$_{Ga}$ acceptors, as shown in Fig. 5. However, this conclusion appears in variance with the TEM studies indicating that Mn$_I$ occupies preferably a tetrahedral position with As as the nearest neighbors (Glas et al., 2004). Also EXAFS spectroscopy (d’Acapito et al., 2006) has not yet provided evidences for the formation of Mn$_I$-Mn$_{Ga}$ dimers. On the other hand, the observation by x-ray magnetic circular dichroism (XMCD) of some non-ferromagnetic Mn inside (Ga,Mn)As films has been assigned to such dimers (Kronast et al., 2006). This issue, as well as the strength of exchange couplings between band holes and Mn$_I$ in various positions, have not yet been settled theoretically (Blinowski and Kacman, 2003) and experimentally. At the same time, a strong antiferromagnetic interaction is expected for Mn$_I$-Mn$_{Ga}$ dimers (Blinowski and Kacman, 2003). The corresponding formation of spin singlets, not coupled to holes by an exchange interaction, could explain a reduction in the concentration of Mn spins,

$$x_{eff} = x - 2x_1,$$

contributing to ferromagnetic order in as-grown (Ga,Mn)As (Chiba et al., 2008b; Edmonds et al., 2005; Potashnik et al., 2002; Stefanowicz et al., 2010a; Wang et al., 2004). We note that, as discussed in Secs. III.A and III.B, antiferromagnetic superexchange between substitutional Mn ions can reduce $x_{eff}$ and $T_C$ further on.

F. Non-uniform carrier distribution

A spontaneous formation of a spatially non-uniform (modulated) carrier and magnetization distribution has been persistently suggested theoretically in the context of magnetic semiconductors (Nagaev, 1993) and considered also for DFSs (Timm, 2006).

If magnetic ordering is mediated by carriers, spatially inhomogeneous magnetization can result from a non-uniform distribution of carrier density. One origin of such inhomogeneity is the formation of space charge layers at the interfaces or surfaces of DFSs, the effect examined quantitatively in gated metal-insulator-semiconductor structures of (Ga,Mn)As (Nishitani et al., 2010; Sawicki et al., 2010). It was also argued (Proselkov et al., 2012) that Coulomb repulsion between surface and interstitial donors produces a gradient in the concentration of Mn$_I$ and, thus, in the hole density and $T_C$, as seen in neutron (Kirby et al., 2006) and magnetization (Proselkov et al., 2012) studies.

Furthermore, according to the physics of disorder-driven quantum localization in doped semiconductors (Altshuler and Aronov, 1985; Lee and Ramakrishnan, 1985), density of electronic states at the Fermi level $\rho_F$ does not exhibit any critical behavior in the vicinity of the Anderson-Mott transition. In contrast to $\rho_F$, the local density of states (LDOS) shows critical fluctuations in the vicinity of the transition, as recently visualized by scanning tunneling spectroscopy in (Ga,Mn)As (Richardella et al., 2010). These fluctuations lead to a nanoscale electronic phase separation into regions with differing hole concentrations, the effect explaining (Dietl, 2008b) a surprising appearance of Coulomb blockade peaks in the conductance of gated nanoconstrictions (Schlapps et al., 2009; Wunderlich et al., 2006).

The electronic phase separation is expected to be enhanced by competing long-range ferromagnetic and short-range antiferromagnetic interactions (Dagotto et al., 2001), particularly in the instances when carrier density is relatively low, as in II-VI and compensated III-V DFSs. These phenomena give rise to the coexistence of ferromagnetic with paramagnetic or superparamagnetic regions, even if the distribution of magnetic ions is perfectly uniform. Such a co-existence was seen in XMCD studies (Takeda et al., 2008) as well as in direct magnetic measurements (Ferrand et al., 2001; Oiwa et al., 1998; Sawicki et al., 2010), as shown in Fig. 6.
FIG. 5 (Color online) Location of Mn (full circles) in GaAs films (Ga - open circles with central dots; As - open circles) grown by low temperature molecular beam epitaxy as seen by particle-induced x-ray emission (PIXE): Ga-substitutional, interstitial, and Mn-rich small clusters incommensurate with the GaAs lattice. A tetrahedral interstitial position with cations as the nearest neighbors is shown but the experiment does not exclude that Mn occupies a tetrahedral position close to anions or a hexagonal interstitial site. From Yu et al. 2008.

FIG. 6 (Color online) Magnetic field dependence of magnetization at 2 K for non-annealed films of Ga$_{1-x}$Mn$_x$As with the nominal value of $x$ ranging from 0.015 to 0.071. The magnetic field is applied perpendicular to the sample plane. The dashed lines are fits of the high field magnetization changes to the paramagnetic Brillouin function with the adjusted effective temperature $T_{\text{eff}} = 4$ K. The fitting implies that the concentration of paramagnetic ions is between 20\% ($x = 0.35$ and $0.43$) and 50\% ($x = 0.071$) of the total Mn composition. From Oiwa et al. 1998.

Since a characteristic relaxation time of superparamagnetic particles is slower than 0.1 $\mu$s, they would generate a ferromagnetic-like response in muon rotation experiments (Dunsiger et al. 2010).

G. Determination of carrier concentration

The determination of carrier density in DFS films from Hall effect measurements is highly challenging for three reasons.

First, the anomalous Hall effect often dominates, so that the evaluation of the normal Hall effect is only possible in high magnetic fields and at low temperatures, where spins of magnetic ions are saturated (Omiya et al., 2001). Under these conditions, taking into account the valence band structure of DFSs, the Hall resistance is expected to provide the hole concentration within an accuracy of about 20\% (Jungwirth et al., 2005). However, in this regime, a direct influence of the magnetic field on the hole magnetic moments reduces the hole spin polarization (Dietl et al., 2001b; ´Sliwa and Dietl, 2006, 2013) and, hence, the anomalous Hall effect, linearly in the magnetic field. This results in an overestimation of the hole concentration, particularly in the high hole concentration range, where the ordinary Hall resistance is relatively small. In an extreme case of (In,Mn)Sb, where the magnetic moment of holes is large, a sign reversal the Hall resistance in the magnetic field was observed (Mihály et al. 2008).

Second, additional corrections to the Hall resistance come from quantum localization phenomena (Altshuler and Aronov, 1985; Lee and Ramakrishnan, 1985), which eventually lead to the divergence of the Hall coefficient in the vicinity of the metal-insulator transition (Dietl 2008b), the effect persisting up to temperatures of the order of the acceptor binding energy (Fritzsche and Cuevas, 1960), about 1000 K in (Ga,Mn)As. Accordingly, the determined magnitude of carrier density directly from the Hall resistance, even at room temperature, is typically significantly underestimated at the localization boundary in DFSs such as (Ga,Mn)As (Satoh et al. 2001, Sheu et al. 2007).

Third, carriers accumulated at interfaces or substrate often contribute to total conductance. Under these con-
ditions, in order to determine the relevant Hall resistivity, magnetotransport measurements should be carried out over a wide field range and interpreted in terms of multichannel formulae (Bonanni et al., 2007). The presence of an electron layer at the (In,Mn)As/GaSb interface is thought to lead to an underestimated value of hole density in (In,Mn)As (Liu et al., 2004).

In view of the above difficulties other methods were successfully employed to determine hole density in (Ga,Mn)As: (i) electrochemical capacitance-voltage profiling (Yu et al., 2002); (ii) Raman-scattering intensity analysis of the coupled plasmon-LO-phonon mode (Seong et al., 2002), and (iii) infrared spectroscopy providing the hole concentration from dynamic conductivity integrated over the frequency (Chapler et al., 2013).

H. Determination of alloy composition

A routine, non-destructive, and accurate determination of an average alloy composition $x$ is by no means straightforward in the case of DFS thin films. The intensity of TM flux during the growth and the character of reflection high-energy electron diffraction (RHEED) or ex-situ secondary ion mass spectroscopy (SIMS) serve to evaluate the nominal TM concentration $x$. For the purpose of calibration the electron probe microanalysis (EPMA)—requiring usually films thicker than 1 μm—or the relation between the flux and thickness of the end compound, say, MnAs (Oliya et al., 2007b) have been employed. The calibration can also be used to establish the composition dependence of the lattice constant $a(x)$, which can readily be determined by x-ray diffraction (XRD) measurements. Here the sensitivity of $a(x)$ to the carrier and defect density (Mack et al., 2008; Matek et al., 2003; Potashnik et al., 2001; Sadowski and Domagała, 2004) has to be considered. Channeling Rutherford backscattering (c-RBS) (Kurnet et al., 2012; Yu et al., 2002) and particle induced x-ray emission (c-Pixe) (Yu et al., 2002) experiments also allow to determine Mn content.

Recently, a three dimensional atom probe technique (3DAP) is being developed (Kozuka et al., 2009), which together with already frequently used electron energy loss spectroscopy (EELS) (Jain et al., 2006) and energy dispersive x-ray spectroscopy (EDS) (Kuroda et al., 2007), have the potential to provide TM composition, also in the case of thin films.

In the case of DMSs the composition can also be assessed from, interesting by its own, magnetic measurements, the method requiring a modeling of magnetism. However, it is now appreciated that because epitaxial films are thin and the concentration of magnetic impurities is typically low, magnetic response of DMS layers can be significantly perturbed by spurious magnetic moments and a limited resolution of typical magnetometers (Sawicki et al., 2011). Accordingly, prior to deposition of DMS films, magnetic properties of the substrate have to be carefully assessed. Furthermore, results of magnetic measurements on DMSs samples should be systematically compared to data obtained for films nominally undoped with magnetic ions but otherwise grown, coalesced, and processed in the identical way as the DMS samples in question.

An example of the application of this technique for a ferromagnetic (Ga,Mn)As is illustrated in Fig. 7, where the findings revealed a rather high value of saturation magnetization, $M_{\text{Sat}} = 90 \pm 5$ emu/cm$^3$ (Chiba et al., 2007). However, assuming the magnetic moment of 5 μB per Mn ion [i.e., neglecting a small hole contribution (Sliwa and Dietl, 2006)], the magnitude of $M_{\text{Sat}}$ leads to the effective concentration of Mn spins $x_{\text{eff}}$ more than 2 times smaller than the nominal value $x = 0.20$ obtained for this film from a linear extrapolation of the Mn flux calibration for $x < 0.1$. This discrepancy, noted also by other groups (Mack et al., 2008; Wang et al., 2008b); is discussed further on in the subsection that follows (Sec. III.A).

The destructive influence of compensating donor defects, such as Mn in the hole and effective Mn concentrations lowers the magnitude of $T_C$ significantly. However, as discussed in Sec. III.A, the concentration of interstitials can be considerably reduced by low temperature annealing.

According to RBS-Pixe studies of (Ga,Mn)As mentioned above, in addition to Ga-substitutional and interstitial positions, Mn atoms assume locations incommensurate with the GaAs lattice, referred to as ”random”, which can involve a half of the total number of Mn ions (Chiba et al., 2008b). It has been suggested that the “random” incorporation corresponds, at least partly, to Mn gathered on the surface as a result of out diffusion of interstitial Mn occurring during the growth or annealing of thin layers (Chiba et al., 2008b; Yu et al., 2005). Such a scenario is supported by the study combining synchrotron XRD and a technique of x-ray standing-wave fluorescence at grazing incidence (Holy et al., 2006), which shows that (Ga,Mn)As consists of a uniform single-crystal film covered by a thin surface Mn-rich layer containing Mn atoms at random non-lattice sites. After annealing, the concentration of interstitial Mn and the corresponding lattice expansion of the epilayer are reduced, the effect being accompanied by an increase in the density of randomly distributed Mn atoms in the disordered surface layer (Rader et al., 2009), where Mn ions are oxidized (Edmonds et al., 2004); Olejnik et al., 2008; Schmid et al., 2008; Yu et al., 2005).

Another kind of a self-compensation mechanism was found in (Ga,Mn)N. In this material, the Mn acceptor level resides in the mid-gap region (Sec. III.A), so that a co-doping by shallow acceptors, such as Mg, is necessary to produce holes in the valence band. It turned out, however, that Mg-Mn complexes are formed in MOVPE grown GaN:Mn:Mg, hampering hole doping of the valence band (Devillers et al., 2012).
The normal- was measured at a photon energy of 1.82 eV and the saturation value of \( M_{\text{s}} \) is the effective Mn concentration \( c \). Similar results were recently obtained at 1.82 eV. All data are obtained at 20 K.

In this section we discuss the most prominent feature of DMSs, namely the existence of a strong interaction between subsystems of localized spins and effective mass carriers, which is the signature of DMSs (Dietl et al., 1994). Purdya and Kossut (1988). This interaction accounts for giant Zeeman splitting of bands, spin-disorder scattering, the formation of magnetic polaron, and the mediation by itinerant carriers of ferromagnetic coupling between localized Mn spins. As predicted theoretically (Dietl et al., 1997), and observed experimentally for (Zn,Mn)O:A (Andraezyk et al., 2001), this coupling is relatively weak in the case of electrons in DMSs. In contrast, ferromagnetic interactions between diluted spins are rather strong when mediated by delocalized or weakly localized holes (Fernandez et al., 2000). Hall et al., 1997; Jungwirth et al., 2010; Olmo et al., 1992, 1996; Sheu et al., 2007; Story et al., 1986). In fact, they can overcome competing short-range antiferromagnetic superexchange occurring between Mn\(^{2+}\) ions in DMSs. Thus, along with the dependence on the magnetic ion density \( x \), ferromagnetic properties of DMSs can be controlled by changing the net acceptor concentration as well as by gating (Sec. III.C) or illumination (Sec. III.E). Conversely, experimentally observing that \( T_\text{C} \) does not vary with \( x \) usually means that magnetic impurities are not randomly distributed (i.e., their local concentration does not depend on the average value \( x \)). Similarly, the lack of dependence on carriers’ density indicates that carriers may not account for ferromagnetic order. Several issues, discussed in more details further on, has to be taken into account in this context:

1. Similarly to other doped semiconductors, holes in DFSs undergo Anderson-Mott localization if their concentration is smaller than a critical value \( p_c \).

2. Due to a contribution of \( p-d \) coupling to the hole binding energy, the value of \( p_c \) is shifted to higher hole densities in DFSs, as compared to correspond- ing non-magnetic counterparts.

3. The carrier-mediated ferromagnetism appears already in the weakly localized regime \( p < p_c \) but no long-range and, thus, efficient ferromagnetic coupling takes place in the strongly localized regime, \( p \ll p_c \), where holes are tightly bound to the parent acceptors.

4. Deeply in the metallic phase, \( p \gg p_c \), ferromagnetic features show typically textbook thermodynamic and micromagnetic properties, despite disorder inherent to doped semiconductor alloys.

5. Because of the self-compensation mechanism (Sec. III.E), the introduction of a sizable acceptor concentration may not result in a correspondingly large hole concentration.

There is no quantitative theory for \( p_c \) but empirically its magnitude is typically within the range \( p_c^{1/3} a_B = 0.26 \pm 0.05 \), if the effective Bohr radius \( a_B \) is evaluated from the binding energy \( E_B \) of the relevant acceptor in the limit \( p = 0 \) according to one of the prescriptions...
Employing the latter for GaAs:Be and GaAs:Mn, for which $E_1 = 28.6$ meV [Fiorentini, 1995] and 112.4 meV [Linmarsson et al., 1997], one obtains $p_c = (2.3 \pm 1.6) \times 10^{-8}$ and $1.4 \pm 1 \times 10^{20}$ cm$^{-3}$, respectively. It is worth noting, however, that in DFSs $p_c$ also depends on the magnitude of magnetization and even on its orientation with respect to crystallographic axes [Pappert et al., 2006].

As shown in Fig. 8, $E_1$ increases rather dramatically on going from antimonides to nitrides through arsenides and phosphides in Mn-doped III-V compounds. The values of $E_1$ were primarily determined from optical data but also from transport studies in the strongly localized regime (Wolos et al., 2009), where at temperatures above the hopping regime the activation energy of conductivity $\epsilon_1 = E_1$. A deviation of $E_1$ from values expected for effective mass acceptors, known as a central cell correction or a chemical shift, was interpreted (Dietl et al., 2002, 2003; Edmonds et al., 2002; Mahadevan and Zunger, 2004; Sato et al., 2010) in terms of the hybridization-induced repulsion between $t_g$ states originating from Mn $d$ levels and $p$-like valence band states from which the acceptor state is built. The role of this mechanism increases with the decreasing cation-anion bond length and energy distance between the valence band top and Mn level, eventually resulting in a transition to the strong coupling limit, where the hole binding energy is dominated rather by the $p$-$d$ interaction than by the acceptor Coulomb potential. The emergence of an impurity band in the energy gap with increasing $p$-$d$ coupling was also captured by the dynamic mean-field approximation (Chattopadhyay et al., 2001).

Because of these differences in magnitudes of $E_1$, critical densities for the metal-insulator transition (MIT) vary significantly within the Mn-based III-V DMSs family (Dietl, 2008a; Wolos et al., 2009). For instance, a comparison of uncompensated Ga$_{1-x}$Mn$_x$As, Ga$_{1-x}$Mn$_x$P, and Ga$_{1-x}$Mn$_x$N with similar Mn content $x \approx 6\%$, shows that holes are respectively delocalized (Jungwirth et al., 2007), at the localization boundary (Scarpulla et al., 2005), and in the strongly localized regime where no carrier-mediated mechanism of spin-spin coupling operates (Sarigiannidou et al., 2006; Stefanowicz et al., 2013).

Except for (Ga,Mn)N, considerable hole conductivities $\sigma$ are characteristic to DFSs, where actually a correlation between the magnitudes of $\sigma$ and $T_C$ is seen, as shown in Fig. 10 for (Ga,Mn)As. In most situations, $\sigma(T)$ remains non-zero at $T \to 0$, implying metallic conductance. However, in some important cases, e. g., (Ga,Mn)As with $x \lesssim 2\%$ (Jungwirth et al., 2007; Sheu et al., 2007), (Ga,Mn)P (Winkler et al., 2011), and (Zn,Mn)Te:N (Ferrand et al., 2001), $\sigma(T)$ vanishes at $T \to 0$ but $T_C$ remains non-zero. Altogether, these data indicate that the ferromagnetism occurs not only on the metal side of the MIT but in a non critical way penetrates into the weakly localized regime, where high-temperature activation energy of conductivity $\epsilon_2 < E_1$ provides information on the distance between the mobility edge and the Fermi level (Fritzsche and Cuevas, 1986). However, on moving deeply into the insulator phase $T_C$ vanishes or at least becomes smaller than the explored temperature range down to 2 K in (Ga,Mn)N (Sheu et al., 2007).

The above mentioned detrimental effect of interstitials on the ferromagnetism of (Ga,Mn)As can be partly reduced by an annealing process (Chiba et al., 2003; Edmonds et al., 2002; Hayashi et al., 2001; Ku et al., 2003; Potashnik et al., 2001; Sørensen et al., 2003) that promotes the diffusion of the Mn$_3$ ions to the surface, where they partake in the formation of an antiferromagnetic MnO thin film (Edmonds et al., 2004b; Olejník et al., 2008; Schmid et al., 2008; Yu et al., 2005) or MnAs monolayer, if the surface is covered by As (Adell et al., 2007). This post-growth thermal treatment leads to a substantial increase in the magnitudes of conductivity, $T_C$, and spontaneous magnetization, to the values shown in Figs. 9, 7, and 10. A similar effect of low-temperature annealing is observed in (In,Mn)As (Hashimoto et al., 2002).

The efficiency of annealing appeared enhanced in nanodots (Eid et al., 2005) or nanowires (Chen et al., 2011). In contrast, the process of out diffusion was self-limiting if the annealing was performed in an oxygen-free atmosphere or the surface was covered by a cap (Chiba et al., 2003). The diffusion of Mn$_3$ towards the surface can already occur during the growth, the process being particularly efficient in thin samples (Chiba et al., 2008b; Yu et al., 2005). Accordingly, the concentration of Mn$_3$ in such samples is relatively low, below 2$, as shown in Fig. 10.

A natural question arises whether co-doping of (Ga,Mn)As by non-magnetic acceptors, say Be, could enlarge $T_C$ over the values displayed in Fig. 9. It could be expected that an antiferromagnetic character of carrier-mediated interaction, showing up when carrier density...
becomes greater than the magnetic impurity concentration, can drive the system towards a spin-glass phase, as observed in Pb$_{1-x-y}$Sn$_x$Mn$_y$Te (Eggenkamp et al., 1995). It turned out, however, that the presence of additional holes during the growth of (Ga,Mn)As layer increases the concentration of Mn interstitial donors (Mn$_i$) by the self-compensation mechanism. This diminishes net hole and Mn densities, so that $T_C$ gets actually reduced (Wojtowicz et al., 2003b; Yu et al., 2004). However, $T_C$ is increased if additional holes are transferred to (Ga,Mn)As layer after its epitaxy has been completed. Such engineering of ferromagnetism in Ga$_{1-x}$Al$_x$As/(Ga,Mn)As/Ga$_{1-y}$Al$_y$As quantum structures is presented in Fig. 11. As seen, modulation doping by Be in the back barrier diminishes $T_C$, as then the Fermi level assumes a high position during the growth of (Ga,Mn)As layer which results in the Mn$_i$ formation. In contrast, when Be is introduced in the front barrier, i.e., after the growth of (Ga,Mn)As, the concentration of Mn$_i$ is small and $T_C$ becomes high.
FIG. 12 (Color online) Effect of growth conditions on the Curie temperature in (Ga,Mn)As. The hole density \( p \) from the Hall effect at 300 K and the Curie temperature \( T_C \) are plotted as a function of As:Ga flux ratio for two wafers with different Mn concentrations, (a) 1.25%, and (b) 1.50%. Lines guide the eye. The stoichiometric region is shaded grey, where effects of disorder-induced hole localization (caused by roughness on the Ga-rich side and by As antisites on the As-rich side) are minimized. Adapted from Myers et al., 2006.

Not surprisingly, the magnitude of \( T_C \) in DFSs can be lowered by incorporating compensating donor impurities or donor defects. In the case of (Ga,Mn)As the effect was observed by co-doping with Sn (Satoh et al., 2001), Si (Wang et al., 2008c), Te and S (Scarpulla et al., 2008), As vacancies (Mayer et al., 2010), and As antisite defects (Myers et al., 2006; Sheu et al., 2007), as shown in Fig. 12. On the other hand, co-doping with donors during the epitaxy can facilitate the incorporation of Mn in the substitutional positions (Wang et al., 2008c), the effect particularly appealing if the donors could then be removed by post-growth processing (Bergqvist et al., 2011).

Figure 13 presents temperature dependence of resistivity (upper panel) and magnetization (lower panel) at various hole densities changed by irradiation of a Ga\(_{0.955}\)Mn\(_{0.045}\)As film by Ne\(^+\) ions. From Mayer et al., 2010.

\( T = 0 \), \( x_{\text{eff}} \), gets smaller. Similar results were obtained for hydrogenated samples (Thevenard et al., 2007) and a (Ga,Mn)P film irradiated with Ar\(^+\) ions (Winkler et al., 2011). These data support the scenario of the electronic and, hence, magnetic phase separation in the vicinity of the MIT in DFSs, the effect enlarged by competing ferromagnetic and antiferromagnetic interactions (see, Sec. II.F).

Also post-growth hydrogenation reduces the hole density and turns (Ga,Mn)As into a paramagnet (Farshchi et al., 2007; Goennenwein et al., 2004; Thevenard et al., 2007). This process is entirely reversible by annealing below 200°C (Thevenard et al., 2007). By employing local reactivation using confined low-power pulsed-laser annealing or by hydrogenation through a mask it was possible to pattern ferromagnetic structures with features size below 100 nm (Farshchi et al., 2007), as shown in Fig. 14.

The ferromagnetism can be weakened even if the net acceptor concentration is kept constant but additional disorder enhances hole localization. For example, partial anion substitution (As → P or As → N) (Stone et al., 2008) or structure roughness introduced by excess Ga.

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sub-micrometer regime by using suitably sized SiO2 features to block the hydrogenation. For this purpose, a 200 nm SiO2 layer was deposited via e-beam evaporation

Figure 3. C-AFM image of sub-micron Ga1/C0 xMnxAs features produced via II-VI DMSs, such as (Zn,Mn)Te:N, the effect described above for (Cd,Mn)Te:N [Haury et al., 1997], (Zn,Mn)Te:N [Ferrand et al., 2000], (Be,Mn)Te:N [Sawicki et al., 2002], and (Zn,Mn)Te:P [Kepa et al., 2003]. Similarly, ferromagnetism of Pb1−x−ySn x,y Mn yTe is brought about by holes originating from native defects—primarily cation vacancies—generated by post-growth annealing [Story et al., 1986]. Also in the case of II-VI DMSs, such as (Zn,Mn)Te:N, the effect of self-compensation challenges the progress in raising the Curie temperature, where the magnitude of achievable hole density by nitrogen doping decreases with the Mn concentration [Ferrand et al., 2001]. Furthermore, the MIT is shifted to higher hole concentrations, as the acceptor binding energy is enhanced by magnetic polaron effects [Ferrand et al., 2001; Jaroszyński and Dietl, 1985]. Moreover, in the strong coupling limit, the Mn ion can act as a hole trap, which hampers the possibility of obtaining holes in the valence band [Dietl et al., 2008a]. This situation takes presumably place in (Zn,Mn)TeO, as witnessed by the presence of a relatively large subbandgap absorption corresponding to the photoionization process: Mn2+ + ν → Mn3++ + h + e, where the hole is bound to Mn3++ and the electron transferred to the conduction band [Godlewski et al., 2010].

FIG. 14 (Color online) Conductance atomic force microscopy image of sub-micron (Ga,Mn)As features produced with selective hydrogenation. Inset shows a scan on a single feature. From Farshchi et al., 2007

FIG. 15 (Color online) Inverse magnetic susceptibility from SQUID measurements (squares) for two p-Zn1−xMnxTe samples with similar Mn composition x ≈ 0.045 but different hole concentrations. Solid lines show linear fit. The dotted line presents the dependence expected for an undoped sample with a similar Mn content. From Ferrand et al., 2001

B. Controlling magnetic anisotropy by hole density and strain

Together with magnitudes of TC and MSat, the character and strength of magnetic anisotropy determine possible functionalities of any ferromagnet. In the subsections below experimentally demonstrated manipulations with orientation of magnetization are discussed taking into account strain engineering by lattice mismatch to substrates and strain relaxation in nanostructures as well as by piezoelectric and elastic actuators. Microscopic theory of these phenomena is outlined in Sec. IX.C whereas its comparison to experimental findings is presented in Sec. X.D.

1. Magnetic anisotropy in films and nanostructures

Extensive magnetic [Sawicki, 2006], ferromagnetic resonance (FMR) [Cubukcu et al., 2010; Liu and Furdyna, 2006], magnetotransport [Glunk et al., 2009; Gould et al., 2008; Tang et al., 2003] and magnetooptical [Hrabovsky et al., 2002; Welp et al., 2003] studies of (Ga,Mn)As and (Ga,Mn)(As,P) films deposited coherently on (001) substrates (typically GaAs substrate or relaxed (Ga,In)As buffer layer) allowed to establish how the system energy depends on the direction of magnetization $\mathbf{M}$ at a given external magnetic field $\mathbf{H}$ and bi-axial strain imposed by lattice mismatch, quantified by a relative difference between the lattice parameter of the substrate and the free standing layer,

$$\epsilon_{xx} = \epsilon_{yy} = \Delta a/a; \quad \epsilon_{zz} = -2\epsilon_{xx}c_{12}/c_{11},$$

where the ratio of elastic moduli $c_{12}/c_{11} = 0.453$ in GaAs.
In a single domain state, according to the Stoner-Wohlfarth formalism, the functional of free energy density contains contributions from the Zeeman energy, shape (demagnetization) and crystalline magnetic anisotropies, \( F = F_Z + F_d + F_{cr} \). To determine spatial orientation of \( \vec{M} \), \( F \) is minimized with respect to \( \theta \) and \( \phi \) defined in Fig. 10. It has been established that in order to describe experimental data, \( F_{cr} \) has to contain at least three contributions (taken in the lowest order): cubic as well as in-plane and out-of-plane uniaxial anisotropy terms. Their relative magnitudes were found dependent on magnetization, hole density, and strain leading to a range of spectacular phenomena, such as spin reorientation transition on varying temperature (Kamara et al. 2012; Sawicki et al. 2004; Thevenard et al. 2006; Wang et al. 2006a; Welp et al. 2003), hole density (Khazen et al. 2008; Sawicki et al. 2004; 2005; Thevenard et al. 2005; 2006) or strain imposed by piezoelectric stressors (Bihler et al. 2008; Casiraghi et al. 2012; Overby et al. 2008; Rushforth et al. 2008). This strong sensitivity to strain means also that for anisotropy-related studies, samples should be mounted in a way minimizing thermal stress. Importantly, magnetization orientation can also be manipulated by gate voltage, electric current, and light, as described in Secs. III.C-G.

Taking \( F(\vec{M} \parallel [100]) \) as a reference energy the particular contributions to \( F \) in terms of assume then the form,

\[
F_Z = -\mu_0 M \vec{H} = -\mu_0 M H [\cos \theta \cos \theta_H + \sin \theta \sin \theta_H \cos(\phi - \phi_H)],
\]

\[
F_d = \frac{1}{2} \mu_0 M^2 m_z^2,
\]

and

\[
F_{cr} = K_C (m_x^2m_y^2 + m_y^2m_z^2 + m_z^2m_x^2) + K_{xy}m_xm_y + K_{zz}m_z^2,
\]

where we have introduced magnetization directional cosines \( m_x = \sin \theta \cos \phi \), \( m_y = \sin \theta \sin \phi \), and \( m_z = \cos \theta \). \( \theta_H \) and \( \phi_H \) are azimuthal and polar angles of \( \vec{M} \) and \( \vec{H} \), respectively (see, Fig. 16) and \( K_i \) are sample and temperature dependent fitting parameters (crystalline anisotropy energies) to experimental dependence \( \vec{M}(\vec{H}) \). These energies are related to the anisotropy magnetic fields, \( \mu_0 H_i = 2K_i/M \), describing the strength of the applied field allowing aligning of magnetization along the hard axes. As required by time reversal symmetry, \( F_d \) and \( F_{cr} \) are even functions of \( M \).

Because of a relatively low magnitude of spontaneous magnetization (typically \( \mu_0 M \lesssim 0.1 \) T), the strength of the shape anisotropy field, \( \mu_0 H_d = \mu_0 M \), is substantially smaller in DFSs than in ferromagnetic metals. In contrast, the magnitude of crystalline anisotropy is rather sizable. According to experimental studies referred to above, each of the three contributions to crystalline magnetic anisotropy, displayed in Eq. 7 shows a specific pattern:

**Cubic anisotropy** – Independently of epitaxial strain and hole density, the value of \( K_C \) was found positive in (Ga,Mn)As (showing that the cubic easy axis is along (100)) and corresponds to \( \mu_0 H_C \) of the order of 0.1 T at \( T \ll T_C \). It decays rather fast with temperature, \( K_C \sim M^4(T) \), consistently with the expected isotropy of linear response functions in cubic systems requiring that \( \partial^2 K_C / \partial M^2 \to 0 \) for \( M \to 0 \). In contrast, a negative value of \( K_C \) (corresponding to a (110) cubic easy direction) was reported for (In,Mn)As (Liu et al. 2003) and (Ga,Mn)P (Bihler et al. 2007).

**In-plane uniaxial anisotropy** – No such anisotropy, first observed in magnetotransport experiments on (Ga,Mn)As (Katsumoto et al. 1998), is expected for the \( D_{2d} \) symmetry group corresponding to biaxially strained (001) zinc-blende crystals. It was demonstrated that the corresponding anisotropy field was independent of the film thickness (Welp et al. 2004), pointing to the bulk, not surface or interface, origin of this anisotropy, the conclusion consistent with no effect of film thickness by etching on its presence (Sawicki et al. 2005). However, as noted in Sec. I.D according to theory (Birowski et al. 2012), a surplus of the Mn dimer concentration along the [110] direction comparing to the [110] case is expected.

1 Differing conventions of parameterizing \( F_{cr} \) exist in the literature. For instance, the cubic term is often decomposed into in-plane and perpendicular-to-plane components, which increases the number of fitting parameters but is a priori justified by symmetry in the presence of a biaxial strain.
This lowers the symmetry to $C_{2v}$ (even in the absence of any strain), for which distinct in-plane and out-of-plane uniaxial anisotropies are allowed. Experimentally, the value of an effective shear-like component $K_{xy}$ is usually positive (Zemen et al., 2009) (i.e., the corresponding easy axis points along [110] direction), and $\mu_0 H_{xy}$ is typically of the order of 0.02 T at $T = T_C$, so that it is smaller than $\mu_0 H_C$. It was found (Sawicki et al., 2005) that at appropriately high hole concentrations the uniaxial easy axis flips to the [110] direction, as shown in Fig. 17, the effect often occurring only at sufficiently high temperatures, $T_C/2 < T < T_C$ (Kopecký et al., 2011; Proseklov et al., 2012; Sawicki et al., 2005). Furthermore, since comparing to $K_C$, $K_{xy}$ decays slower with temperature, $K_{xy} \sim M^2(T)$, a spin reorientation transition (100) $\rightarrow$ [110] is observed on increasing temperature in the range $T \leq T_C/2$ (Kamara et al., 2012; Wang et al., 2005c; Welp et al., 2003), the effect illustrated in Fig. 18. A competition between cubic and in-plane uniaxial magnetic anisotropies were also found for (In,Mn)As (Liu et al., 2005) and (Ga,Mn)P (Bihler et al., 2007).

**Out-of-plane uniaxial anisotropy** — According to experimental studies of (Ga,Mn)As on $In_{0.5}Ga_{0.5}As$ (Glunk et al., 2009) and of (Ga,Mn)As$_{1-y}$P$_y$ on GaAs (Cubukcu et al., 2010) as a function of $y$ and, thus, epitaxial (biaxial) strain $\epsilon_z$, the anisotropy energy $K_{zz}$ can be decomposed into two contributions.

One is linear in $\epsilon_z$:

$$\mu_0 H_{zz} = A \epsilon_z,$$

corresponding to the in-plane and perpendicular-to-plane crystalline magnetic anisotropy for compressive and tensile strain, respectively. According to studies up to $|\epsilon_z| \approx 0.4\%$, $A$ can reach a magnitude of the order of $+1/T/\%$ (Cubukcu et al., 2010; Glunk et al., 2009), but decreases, or even changes sign when diminishing hole density at a fixed compressive strain (Khazen et al., 2008; Sawicki et al., 2004; Thevenard et al., 2005) or tensile strain (Thevenard et al., 2006). For hole concentrations $p \approx 10^{20} \text{cm}^{-3}$ corresponding to the vicinity of the spin reorientation transitions (001) $\rightarrow$ [011], the transition can occur on changing temperature (Sawicki et al., 2004; Thevenard et al., 2006). Similarly to (Ga,Mn)As with high hole concentrations, also (In,Mn)As shows perpendicular-to-plane orientation of the easy axis for a tensile strain, imposed by either (Ga,Al)Sb (Liu et al., 2004; Muneoka et al., 1993; Ohno et al., 2000) or InAs substrate (Zhou et al., 2012), whereas the easy axis is in-plane under a compressive strain (produced by an (In,Al)As substrate) (Liu et al., 2005). Interestingly, an opposite relation between the strain character and easy axis direction, consistent with the findings for (Ga,Mn)As with low carrier density, was observed for p-(Cd,Mn)Te:N (Kossaacki et al., 2004b), as shown in Fig. 19, as well as for (Al,Ga,Mn)As (Takamura et al., 2002) and (Ga,Mn)P (Bihler et al., 2007), presumably because of low net hole concentrations in all these cases.

Another contribution to $K_{zz}$ found in (Ga,Mn)As films is strain independent term leading to a non-zero out-of-plane uniaxial anisotropy term even for $\epsilon_z = 0$ (Cubukcu et al., 2010; Glunk et al., 2009). The corresponding value is of the order of $\mu_0 H_{zz} \approx 0.1 \text{T}$. Its positive sign means that this anisotropy, along with the demagnetization term, enlarges a tendency to the in-plane orientation of the easy axis. As already mentioned, this contribution, unexpected within the group theory for a zinc-blende alloys having a random distribution of constituents, is as-
signed to a surplus of [110] Ga-substitutional Mn dimers \cite{Birowska et al., 2012}.

Appropriately modified forms of $\mathcal{F}_{cr}$ were found to describe $\hat{M}(\hat{H})$ for (Ga,Mn)As grown on a (113)A GaAs substrate \cite{Dreher et al., 2010, Limmer et al., 2006, Stefanowicz et al., 2010a, Wang et al., 2005b}. In this case, however, four (not two) contributions to $\mathcal{F}_{cr}$ are allowed by symmetry and, in fact, describe magnetic and FMR data \cite{Stefanowicz et al., 2010a}. They correspond to cubic $K_C$, biaxial $K_{zz}$, and two shear-like, $K_{xy}$ and $K_{xz} = K_{yz}$, anisotropy energies (the axes of the coordinate system are taken along main crystallographic directions). Similarly to the case of (001) substrates discussed above, the spin reorientation transition from the biaxial $\langle 100 \rangle$ anisotropy at low temperatures to uniaxial anisotropy with the easy axis along the [110] direction at high temperatures is observed (around 25 K). As evidenced by investigations of the polar magneto optical Kerr effect, a declined orientation of the easy axes with respect to the film plane and the film normal allows the perpendicular-to-plane component of magnetization to be reversed by an in-plane magnetic field \cite{Stefanowicz et al., 2010a}.

A specific strain distribution in (Ga,Mn)As nanostructures, either in the form of nanobars patterned lithographically \cite{Humphmer et al., 2007, King et al., 2011} or shells deposited onto GaAs nanowires \cite{Rudolph et al., 2009}, was found to result in the easy axis orientation along the nanostructure long axis. The magnitude of the observed anisotropy field was much larger than expected for the corresponding shape anisotropy, pointing to the importance of crystalline and strain effects. In the case of rectangular nanobars the epitaxial in-plane strain is retained along the bar long axis but it is partly relaxed in the transverse direction, as confirmed by finite element calculations \cite{King et al., 2011, Wenisch et al., 2007} and observed by x-ray reciprocal space mapping \cite{King et al., 2011, Wenisch et al., 2007}. It was possible to rotate the easy axis by $90^\circ$ by nanopatterning \cite{King et al., 2011}.

Magnitudes of possible surface or interface magnetic anisotropies have not yet been assessed for DFSs.

2. Piezoelectric and elastic actuators

A strong sensitivity of magnetic anisotropy to strain makes it possible to manipulate magnetization directions by an electric field in hybrid structures consisting of a DFS film cemented to a piezoelectric actuator. This appealing method was successfully demonstrated for (Ga,Mn)As by applying a voltage-controlled strain along either $\langle 110 \rangle$ \cite{Casiraghi et al., 2012, Goennenwein et al., 2008, Rushforth et al., 2008} or $\langle 100 \rangle$ \cite{Bihler et al., 2008, Overby et al., 2008} directions of (Ga,Mn)As. In this way, rotation of the easy axis form either $\langle 110 \rangle$ or $\langle 100 \rangle$ directions by about $70^\circ$ was possible at appropriately selected temperature and magnetic field values, as shown in Fig. 20. Importantly, an elaborated sequence of applied magnetic fields and voltages was found to switch magnetization in an irreversible fashion, showing a road for developing a novel voltage-controlled memory cell \cite{Bihler et al., 2008}.

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{image1.png}
\caption{(Color online) Determination of easy directions in p-(Cd,Mn)Te modulation doped quantum wells with only the ground state hole subband occupied under compressive (a) and tensile epitaxial strain (b). Curie-Weiss behavior above Curie temperature was obtained from photoluminescence measurements in the magnetic field parallel and perpendicular to the growth axis (points). The line splitting, proportional to weak-field Mn magnetization, is presented in relative units. The straight lines are drawn through experimental points. Adapted from Kossacki \textit{et al.} 2004b.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{image2.png}
\caption{(Color online) Voltage-induced in-plane magnetization rotation at 50 K and in 10 mT, determined from longitudinal and transverse (Hall) resistances (open and full circles, respectively), in a Ga$_{0.955}$Mn$_{0.045}$As film cemented to a piezoelectric actuator, demonstrating a reversible change of magnetization direction by about $70^\circ$ by the application of a voltage. Magnetic field and magnetization angles ($\alpha$ and $\beta$, respectively) are measured in respect to the [110] direction of current and main expansion of the actuator. The hysteretic behavior is caused by the actuator. From Goennenwein \textit{et al.} 2008.)}
\end{figure}
Magneto-elastic properties of (Ga,Mn)As were also studied by determining eigenfrequencies of nanoelectromechanical resonator as a function of temperature and magnetic field orientation, as shown in Fig. 21 (Masmanidis et al., 2005). This experiment was described by considering a contribution $F_{\text{me}}$ to the system free energy associated with additional strain $\varepsilon_{ij}$ imposed by vibrations. By combining $F_{\text{me}}$ with an elastic energy of the host lattice, it was possible to determine how stress $\sigma_{ij}$ and, thus, the frequency of the (Ga,Mn)As nanoelectromechanical resonator should vary with the magnetic field orientation. It was found that the low temperature results can be described by first order magnetostriction coefficients, $\lambda_{100}$ and $\lambda_{111}$, but above 20 K a second order magnetostriction, characterized by a parameter $h_3$, had to be taken into account in order to describe the data.

C. Manipulation by an electric field

Since ferromagnetism in DFSs is hole-mediated, one can turn on and off the magnetic phase by controlling the number of holes in the system without changing the temperature, which can be done electrostatically by applying an electric field $E_G$ to the ferromagnetic semiconductor layer of interest. This was demonstrated using a thin (In,Mn)As (5 nm) as a channel layer of a metal-insulator-semiconductor field-effect transistor (MISFET) with a polyimide insulator (Ohno et al., 2000) and a modulation doped p-(Cd,Mn)Te 8 nm thick quantum well placed in the intrinsic region of a p-i-n diode (Boukari et al., 2002). An appropriate strain engineering resulted in the easy axis perpendicular to the layer plane and allowed to probe magnetization through the anomalous Hall effect (Ohno et al., 2000) and splitting of the luminescence line (Haury et al., 1997), as shown in Figs. 22 and 23, respectively.

Another effect of gating is the change of the coercive force $H_c$ at which magnetization reverses its direction; greater (smaller) $H_c$ for negative (positive) $E_G$. By using this phenomenon, a new scheme of magnetization rever-
sal, an electric-field assisted magnetization reversal, was demonstrated for \((\text{In,Mn})\text{As}\) \((\text{Chiba et al.}, 2003)\). Under a certain applied field \(H\), \(H_e\) is electrically modified from its original \(|H_e| > |H|\) to \(|H_e| < |H|\), thereby electrically triggering the magnetization reversal. Once \(T_C\) becomes high enough, this scheme can be useful for future magnetic field recording technology, where the magnetic anisotropy for retaining information becomes so large that it is almost impossible to change the magnetization direction by field alone.

Although there have only been limited success on \((\text{Ga,Mn})\text{As}\) \((\text{Nazmul et al.}, 2004)\), recent progress in low-temperature deposition of high-quality gate oxides by atomic layer deposition made it possible to observe electrical modulation of ferromagnetism in \((\text{Ga,Mn})\text{As}\) \((\text{Chiba et al.}, 2006a, 2010; \text{Sawicki et al.}, 2010), (\text{Ga,Mn})\text{Sb} \((\text{Hsiao Wen Chang et al.}, 2013)\), and \((\text{Ge,Mn})\) \((\text{Xiu et al.}, 2010)\). For such oxides, the gate-induced changes in the areal carrier density reach \(3 \times 10^{13} \text{ cm}^{-2}\), which for a typical value of the Thomas-Fermi screening length would result in the amplitude of the hole variation about \(3 \times 10^{20} \text{ cm}^{-3}\), and the correspondingly high modulation of \(T_C\). However, the Fermi level is often pinned by gap surface states, which limit the \(T_C\) changes to about \(20 \text{ K}\) in \((\text{Ga,Mn})\text{As}\) \((\text{Chiba et al.}, 2006a, 2010; \text{Nishitani et al.}, 2010; \text{Sawicki et al.}, 2010), even if a polymer electrolyte is employed \((\text{Endo et al.}, 2010a)\). A theoretical description of these and related data \((\text{Stolichnov et al.}, 2011)\) is discussed in Sec. X.A.2.

As elaborated in the previous section, also magnetic anisotropy, which determines the magnetization direction, depends on the hole concentration in DFSs. By applying an electric field and by using anisotropic magnetoresistance \((\text{Chiba et al.}, 2008a)\) as well as direct magnetization measurements \((\text{Chiba et al.}, 2008a; \text{Sawicki et al.}, 2010)\), the effect of the electric field on in-plane magnetization orientation was evidenced in MIS-FET of \((\text{Ga,Mn})\text{As}\) and high-\(k\) oxide as a gate insulator. As demonstrated by Hall effect measurements, a fourfold change in the value of the out-of-plane uniaxial anisotropy field was achieved by gating a ultrathin ferromagnetic \((\text{Ga,Mn})\text{As}/(\text{Ga,Mn})(\text{As,P})\) bilayer \((\text{Niazi et al.}, 2013)\).

An important variant of gating is the application of ferroelectric overlays allowing for a non-volatile and subnanosecond change in interfacial hole density, the method successfully employed to demonstrate the manipulation of \(T_C\) by an electric field in \((\text{Ga,Mn})\text{As}\) \((\text{Küster et al.}, 2009; \text{Stolichnov et al.}, 2011; \text{Stolichnov et al.}, 2008)\).

Manipulation of magnetism by gating was also demonstrated for \((\text{Ge,Mn})\) films \((\text{Park et al.}, 2002)\) and quantum dots \((\text{Xiu et al.}, 2010)\). In the former case, an enhancement and a reduction of \(T_C\) was demonstrated at 50 K by applying \(\pm 5\) V through an \(\text{SiN}_x\) gate insulator to a 60-nm thick \(\text{Ge}_9 \text{Ga}_{0.977} \text{Mn}_{0.023}\) film on Ge(001). Self-assembled \(\text{Ge}_{90.95} \text{Mn}_{0.05}\) dots were deposited on a p-Si substrate and covered by a 40-nm thick \(\text{Al}_2\text{O}_3\) gate insulator. In was shown by SQUID measurements that positive gate voltage up to 40 V reduced a saturation value of magnetization tenfold at 50 K and by 30% at 100 K.

Another interesting case constitutes magnetically doped topological insulator \((\text{Bi,Mn})_2(\text{Te,Se})_3\) showing in the bulk form hole-mediated ferromagnetism with \(T_C\) of 12 K \((\text{Checkelsky et al.}, 2012)\). In contrast, no conductivity and ferromagnetism were observed in few nm-thick flakes put on a \(\text{SiO}_2\)/doped-Si wafer, presumably because of cleavage-induced hole compensating defects. However, the application of a strong negative electric field across \(\text{SiO}_2\) allowed to restore hole conductivity and ferromagnetism characterized by \(T_C\) up to 12 K.

**D. Current-induced magnetization switching**

When current flows through a ferromagnetic layer, the current becomes spin-polarized. In magnetic tunnel junctions (MTJs), the flow of spins from one ferromagnetic electrode to the other across the tunnel barrier exerts a torque between the two electrodes, and its direction depends on the flow of spin, i.e., on the current direction. In sufficiently small MTJs, the torque can reach a threshold value above which magnetization reversal takes place. This is so-called current-induced magnetization switching (CIMS) that was observed in submicron \((\text{Ga,Mn})\text{As}\) MTJs \((\text{Chiba et al.}, 2004b; \text{Elsen et al.}, 2006)\), as shown in Fig. 24. The critical current density \(j_c\) for switching is of the order of \(10^4–10^5\) \(\text{A/cm}^2\), and can be qualitatively understood by Slonczewski’s spin-transfer torque model \((\text{Chiba et al.}, 2004b)\), although for the assumed value of the Gilbert damping constant \(\alpha_G\), the model resulted in an order of magnitude greater \(j_c\) than the observed one.

Another appealing method, particularly in the context of DFSs, is magnetization manipulation by an effective magnetic field produced by an electric current through...
spin-orbit coupling, as opposed to the Oersted effect. It is well known that in confined 2D systems there appear terms linear in k coupled to the electron spin. In particular, the corresponding Rashba field was shown to generate current-dependent shift of electron spin-resonance in an asymmetrical quantum well of n-Si [Wilamowski et al., 2007]. It was suggested theoretically (Bernevig and Vishik, 2005) and demonstrated experimentally that sufficiently strong current, via spin-orbit coupling in strained (Ga,Mn)As (see, Eq. 27), can serve to rotate magnetization by 90° (Chernyshov et al., 2009) or even by 180° (Endo et al., 2010b) in (Ga,Mn)As films.

E. Current-induced domain wall motion

Magnetic domains are formed in a ferromagnet as a result of competition between exchange energy (which tries to align all the spins) and magnetostatic energy (which tries to align magnets antiparallel). The transition region between the domains, in which localized spins (Mn spins of magnetization M in our case) gradually changes its direction, is called a domain wall (DW). Spin-polarized currents interact with DW and once a threshold \( j_c \) is passed can displace the DW, resulting in magnetization reversal of a region swept by the DW. Although such a current-induced DW motion has been of interest for many years in the context of metallic ferromagnets, the DW switching without assistance of a magnetic field was first demonstrated for (Ga,Mn)As films [Yamanouchi et al., 2004].

In microtracks of (Ga,Mn)As [Adam et al., 2009] Yamanouchi et al., 2006, 2004, 2007 and (Ga,Mn)(As,P) [Curiale et al., 2012; Wang et al., 2010a] with the easy axis perpendicular to the film plane, DW displacement under current pulses was monitored by magneto-optical Kerr microscopy. As shown in Fig. 25 a similar dependence of DW velocity \( v \) on the current density \( j \) were found for these two material systems [Curiale et al., 2012; Yamanouchi et al., 2006].

These findings were interpreted by the generalized Landau-Lifshitz-Gilbert equation containing, as displayed in Appendix, current-induced adiabatic and non-adiabatic spin torques, accounting for transfer of spin momenta from current carriers to Mn ions. In particular, [Wang et al., 2010a; Yamanouchi et al., 2006] assumed the dominance of the adiabatic spin torque, i.e., \( \beta_a / \alpha_G \ll 1 \), the assumption led to \( \{ \text{Tataru and Kolmo, 2004} \} \) \( v = A (j^2 - j_c^2)^{1/2} \) for \( j > j_c \). Here, in terms of spin current polarization \( P_s \) and DW width \( \delta_w \) (discussed theoretically in Secs. [X,E] and [X,F] respectively), \( A = g_{\mu_B} P_s / 2 e M \) and \( j_c = 2 e K / \delta_w / \pi \hbar P_s \), where in the perpendicular case the relevant magnetic anisotropy energy of DW spins is \( K = \mu_0 M^2 / (2 + 4\Omega / \pi t) \), where \( t \) is the film thickness. The experimental values of both \( A \) and \( j_c \), implied by the data in Fig. 25, are in quantitative agreement with this theory. Moreover, this approach explains why no current-induced DW displacement was observed for the in-plane easy axis [Tang et al., 2006], as \( j_c \) is then determined by \( K_{\pi} \gg K \). Within this model, DW motion at \( j > j_c \) is accompanied by in-plane Mn spin precession.

On the other hand, Curiale et al., 2012 (see also Adam et al., 2009) interpreted their data assigning non-zero values of \( j_c \) to extrinsic DW pinning and allowing for a large magnitude of the non-adiabatic spin torque (present due to spin-orbit interactions), \( \beta_a / \alpha_G \gg 1 \) (Garate et al., 2009; Hals et al., 2009; Zhang and Li, 2004). In this case, \( v = A j \), \( j > j_c \), the regime corresponding within this model to steady state DW flow without Mn spin precession.

In the subthreshold regime, \( j < j_c \), the DW velocity was found to decay exponentially when reducing \( j \), indicating that DW displacement proceeded through current-induced creep [Curiale et al., 2012; Yamanouchi et al., 2006, 2007]. Over the exploited range of \( j \) and \( T \), \( \sigma (j) \) assumed a scaling form \( \text{Yamanouchi et al., 2006, 2007} \) \( j = a - bj^{-\sigma} \), where \( a \propto (T_c - T)^{\beta} , b \propto (T_c - T)^{\gamma} \). Empirically \( \text{Yamanouchi et al., 2006, 2007} \), \( \mu = 0.4 \pm 0.1 \) and \( \sigma = 2 \pm 0.2 \) (the value of \( \rho \) is uncertain but probably around 1). Interestingly, for DW creep generated by a magnetic field \( H \), the scaling equation assumed a similar form (with \( j \) replaced by \( H \)). However, in agreement with theoretical considerations, the values of the exponents were different, emphasizing the unalike role of these two DW driving mechanisms (Yamanouchi et al., 2007).

Domain wall dynamics in the magnetic field, particularly mobility, pinning, and flexing were examined in (Ga,Mn)As and (Ga,Mn)(As,P) with perpendicular easy
axis by spatially resolved magnetooptical and Hall effects [Balk et al., 2011, Dourlat et al., 2008, Thevenard et al., 2011].

An issue obviously related to current-induced DW motion is DW resistance that, in general, consists of extrinsic $R^\text{ext}$ and intrinsic $R^\text{int}$ components. The former is brought about by a non-uniform current distribution associated with differences in magnitudes of conductivity tensor components $\sigma_{ij}(\mathbf{M})$ on the two sides of the DW. It was demonstrated [Chiba et al., 2006b, Roberts et al., 2007, Wang et al., 2010b, Xiang and Samarth, 2007], by solving the current continuity equation $\text{div}(\sigma_{ij}(\mathbf{M})\text{grad}V(x,y)) = 0$, that the extrinsic term dominates in (Ga,Mn)As with both perpendicular (Chiba et al., 2006b, Wang et al., 2010b) and in-plane easy axis (Tang et al., 2004) as well as explains the corresponding magnetoresistance (Xiang and Samarth, 2007).

Nevertheless, if DW cross sections $A$ were sufficiently small, $R^\text{int}$ could be revealed, as shown for 25 nm thick Ga$_{0.95}$Mn$_{0.05}$As bars of the width from 150 down to 4 μm and with the strain-induced perpendicular orientation of the easy axis (Chiba et al., 2006b, Wang et al., 2010b). For samples containing etched steps that pinned DWs, $R^\text{int}A \approx 0.5 \Omega \mu m^2$ (Chiba et al., 2006b) and 0.15 $\Omega \mu m^2$ (Wang et al., 2010b) for films with $T_C = 80$ and 122 K, respectively. These values are much larger than $R^\text{int}A$ evaluated from the measured magnitude of anisotropic magnetoresistance (AMR) for the Bloch DW in strained films in question. However, if DWs were pinned by linear defects, the value below experimental resolution, $R^\text{int}A = 0.01 \pm 0.02 \Omega \mu m^2$, was found (after subtracting the AMR contribution) for the sample with $T_C = 122$ K (Wang et al., 2010b). Theoretically predicted DW resistances in (Ga,Mn)As (see, Sec. X.F) are within this range.

F. Magnetization manipulation by light

It was demonstrated that light irradiation affects magnetic properties and, in particular, changes the magnitude of the coercive field in (In,Mn)As/GaSb heterostructures [Koshihara et al., 1997, Oiwa et al., 2001]. The effect was attributed to persistent photoconductivity, that is with the light-induced increase of hole density in (In,Mn)As associated with trapping of photoelectrons by deep levels, which was not reversible at a given temperature – in order to return to the original state, the sample had to be heated.

In the case of Mn-based II-VI DMS reversible tuning of magnetism by light was demonstrated in the case of modulation doped p-(Cd,Mn)Te/(Cd,Mg,Zn)Te:N heterostructures [Boukari et al., 2002, Haury et al., 1997], as depicted in Fig. 23 and discussed theoretically in Sec. X.A.3. Interestingly, illumination with photons of energies above the barrier band gap destroys ferromagnetic order if the magnetic quantum well (QW) resides in an undoped (intrinsic) region of a p-i-p structure. Here, the holes are effectively transferred from the QW to acceptors in the (Cd,Mg,Zn)Te barrier (Boukari et al., 2002, Haury et al., 1997). In contrast, illumination enhances the magnitude of spontaneous magnetization in the case of a p-i-n diode in which photoholes accumulate in the (Cd,Mn)Te QW (Boukari et al., 2002), as shown in Fig. 23.

Reversible changes of magnetization by circularly polarized light were witnessed by Hall effect measurements for (Ga,Mn)As and Mn $\delta$-doped GaAs [Nazmul et al., 2004, Oiwa et al., 2002]. The magnitude of the Hall voltage (and hence, presumably, the magnitude of magnetization along the growth direction) either increased or decreased depending on the helicity of impeding light.

All-optical switching of magnetization between two non-equivalent cubic in-plane directions was demonstrated in (Ga,Mn)As microbar employing a scanning laser magneto-optical microscope (Aoyama et al., 2010). Lithography-induced strain relaxation contributed significantly to the magnitude of uniaxial anisotropy. External magnetic field served to magnetize the sample along the harder cubic direction but was not applied during the switching. Light served primarily to elevate temperature to $T > T_C/2$ at which cubic and uniaxial anisotropy energies became nearly equal (Sec. III.B).

Another interesting case is (Ge,Mn)Te, which deposited at low temperature is amorphous and paramagnetic, presumably because dangling bonds associated with lattice point defects (vacancies) are reconstructed in the amorphous network and do not provide holes. A laser or electron beam triggers a local lattice recrystallization, allowing to pattern ferromagnetic nanostructures (Knoff et al., 2011).

Particularly informative and relevant for fast magnetization manipulation is subpicosecond magneto-optical two-color Kerr spectroscopy and related magnetization sensitive time-resolved methods. Here, we discuss experiments in which illumination generated incoherent magnetization dynamics; the data pointing to coherent magnetization precession are described in the subsequent subsection.

In the case of a Ga$_{0.98}$Mn$_{0.02}$As film with in-plane magnetization, circularly polarized 0.1 ps pulses with the fluence of 10 μJ/cm$^2$ resulted in a transient Kerr effect (Kimel et al., 2004). The determined spectral dependence of the Kerr effect was similar to that observed in a static magnetic field of 1 mT along the growth direction (Kimel et al., 2004).

Extensive time-resolved studies with linearly polarized pumping pulses were carried out for (Ga,Mn)As (Kojima et al., 2003, Wang et al., 2007b) and (In,Mn)As (Wang et al., 2005a), and revealed the presence of fast (< 1 ps) and slow (> 100 ps) processes. The fast component rapidly grew with pump power, saturated at high fluences (> 10 mJ/cm$^2$), and indicated a quenching of ferromagnetism on a subpicosecond timescale, also when the holes were excited via intra valence band transitions (Wang et al., 2008a). A detailed quantitative theoretical study (Cywiński and Sham, 2007) demonstrated that the
inverse Overhauser effect, that is dynamic demagnetization of Mn spins by sp – d spin exchange with photocarriers, accounted for the fast process, whose timescale was determined by carriers’ energy relaxation. In contrast, the slow component at low fluences (~10 \( \mu J/cm^2 \)) corresponded to a recovery of ferromagnetic order or even enhancement of \( T_C \) by an enlarged carrier density, the effect appearing on the timescale of spin-lattice relaxation and persisting up to photohole lifetime [Wang et al. 2007b]. However, a substantial rise of lattice temperature dominated at high fluences leading to a complete destruction of ferromagnetism [Cywiński and Sham 2007] [Wang et al., 2005a].

G. Coherent control of magnetization precession

In a series of experiments on (Ga,Mn)As trains of sub-picosecond pulses of light with photon energies near the band gap [Hashimoto et al. 2008] [Němec et al., 2012] [Qi et al., 2009] or picosecond strain pulses [Bombeck et al., 2013] [Scherbakov et al., 2010] triggered oscillations of Kerr rotation as a function of time. These findings were assigned to a tilt of the magnetization vector \( \vec{M} \), followed by coherent precession of \( \vec{M} \) around its equilibrium orientation. This tilt was brought about by illumination-induced modification of the magnetic anisotropy field \( \vec{H}_{\text{eff}} \) generated by a transient change of temperature [Qi et al., 2009] or strain [Bombeck et al., 2013] [Scherbakov et al., 2010]. Also evidences were found for the presence of non-thermal effects generated by light pulses, such as a transient torque produced by a burst of spin polarized photoelectrons [Němec et al., 2012] or an influence of photoholes on magnetic anisotropy [Hashimoto et al., 2008]. Altogether, studies of time-resolved Kerr rotation as well as of magnetization precession driven by an a. c. magnetic field [FMR, Sec. III.B] or electric current [Fang et al., 2011] have demonstrated that the Landau-Lifshitz-Gilbert equation (recalled in Appendix) describes adequately magnetization dynamics in DFSs. Actually, an explicit solution of this equation was derived providing a frequency and damping of magnetization precession in the Gilbert damping constant \( \alpha_G \) and anisotropy fields \( H_i \) specific to DFSs [Němec et al., 2013] [Qi et al., 2009].

In another study [Luo et al., 2010] magnetization precession in (Ga,Mn)As was found to be overdamped but polarization dependent transient out-of-plane component of the magnetization was visible. A transient out-of-plane magnetization was also detected for a linearly-polarized pump, if the sample was exposed to an in-plane magnetic field prior to optical measurements.

It was shown experimentally [Wang et al., 2009] and discussed theoretically [Kapetanakis et al., 2009] that excitations with near ultraviolet photons lead to coherent magnetization rotation in (Ga,Mn)As driven by photocarrier coherences and nonthermal populations excited in the (111) equivalent directions of the Brillouin zone.

A subsequent theoretical work proposed a protocol for all-optical switching between four metastable magnetic states in DFSs [Kapetanakis et al., 2011].

IV. SPIN INJECTION

Ferromagnetic semiconductors can be used as an epitaxially integrated spin-polarized carrier emitter into nonmagnetic structures working without or in a weak external magnetic field. Electrical spin injection from (Ga,Mn)As to nonmagnetic GaAs has been shown to be possible in a device structure integrated with a GaAs-based nonmagnetic light-emitting diode (LED) as a detector of spin-polarized holes [Ohno et al., 1998] [Young et al., 2002] or electrons in Esaki diodes [Johnston-Halperin et al., 2002] [Kohda et al., 2006, 2001] [Van Dorpe et al., 2005] [Van Dorpe et al., 2004]. By measuring circular polarization of electroluminescence, one can determine the spin polarization of injected carriers from (Ga,Mn)As. Because of carrier confinement in the LED emission region, the heavy hole subband is usually relevant in the radiative recombination process. Hence, according to corresponding selection rules, this method allows to detect carriers with spins polarized along the growth direction, which give rise to circularly polarized vertical (surface) emission [Fiedlerling et al., 2003] [Jonker et al., 2000] [Oestreich, 1999]. Since in the structures studied so far (Ga,Mn)As easy axis was in-plane, an out-of-plane magnetic field was applied to either orient Mn magnetization along the growth direction [Johnston-Halperin et al., 2002] [Kohda et al., 2006, 2001] [Young et al., 2002] or—in an oblique magnetic field configuration [Van Dorpe et al., 2005] [Van Dorpe et al., 2004]—to generate additionally a spin component along the growth direction by spin precession. In these experiments emission in \( \sigma^+ \) polarization prevails demonstrating antiferromagnetic coupling between holes and Mn spins in (Ga,Mn)As.

By employing an Esaki diode as spin-injector in the magnetic field tilted 45° out-of-plane, electroluminescence circular polarization \( P_{\text{EL}} \) reached the saturation magnitude of 21% for Ga\(_{0.92}\)Mn\(_{0.08}\)As with \( T_C = 120 \) K [Van Dorpe et al., 2004]. For this magnetizing field direction and the selection rules specified above, the determine value of \( P_{\text{EL}} \) leads to spin current polarization injected from (Ga,Mn)As, \( \Pi_{\text{inj}} \) = 40% at 4.6 K, where the experimentally determined depolarization factor \( T_C/\tau = 0.74 \) [Van Dorpe et al., 2004] is taken into account. A 6% anisotropy in \( P_{\text{EL}} \) was observed by rotating magnetization projection between [110] and [110] [Van Dorpe et al., 2005].

By the use of a three terminal device structures to

\[ \text{2 The selection rules assumed here imply } \Pi_{\text{inj}} \text{ twice smaller than that quoted originally.} \]
control bias voltages of an Esaki diode and a LED (spin-detector) independently, as shown in Fig. 26 the efficiency of the electron spin injection via band-to-band Zener tunneling from p-type Ga$_{0.945}$Mn$_{0.055}$As to n-type GaAs and then to LED was measured as a function of bias voltage. Emission in $\sigma^+$ polarization prevailed, confirming antiferromagnetic coupling between holes and Mn spins in (Ga,Mn)As. The values of $P_{\text{EL}}$ up to 32.4% were attained for a (Ga,Mn)As emitter with $T_C = 70$ K (Kohda et al. 2006). Since in this case the magnetic field is along the growth direction and $T_s/\tau = 0.64$ (Kohda et al. 2006), one obtains $\Pi_{\text{inj}} = 47 \pm 1\%$ at 10 K, where a 1% correction for a non-zero $P_{\text{EL}}$ without emitter current is taken into account.

Electrical injection and detection of spin-polarized electrons were demonstrated in a single wafer all semiconductor lateral structure, incorporating Ga$_{0.95}$Mn$_{0.05}$As/n$^+$-GaAs Esaki diodes acting as both spin injecting (or extracting) and spin detecting contacts to n-GaAs (Ciorga et al. 2009). Prior to processing, $T_C$ of (Ga,Mn)As was 65 K. Spin precession and the spin-valve effect were observed in the nonlocal signal. Figure 26(d) shows $\Pi_{\text{inj}}$ and $\Pi_{\text{ext}}$ for the reverse and forward bias $V_{\text{EB}}$, respectively, determined under the assumption that $\Pi_{\text{inj(\text{ext})}}$ is equal to the spin detection efficiency, which is strictly valid at $V_{\text{EB}} \rightarrow 0$. As seen, $\Pi_{\text{inj(\text{ext})}} = 51 \pm 2\%$ at 4.2 K in this case.

Further evidences for spin injection from (Ga,Mn)As were provided by studies of Andreev reflection (Braden et al. 2003; Panguluri et al. 2005; Piano et al. 2011), the spin-Seebeck effect (Jaworski et al. 2010), and spin pumping under FMR conditions (Chen et al. 2013). Andreev reflection was also detected in the case of (In,Mn)As (Geresdi et al. 2008; Panguluri et al. 2004).

Theoretically expected values of spin current polarization are presented in Sec. X.7.

V. SPINTRONIC MAGNETORESISTANCE STRUCTURES

A. Anisotropic magnetoresistance and Hall effects

Owing to the strong spin-orbit interaction and typically lower carrier densities compared to ferromagnetic metals, DFSs show sizable magnitudes of anisotropic magnetoresistance (AMR), planar and anomalous Hall effects as well as of related thermomagnetic (Jaworski et al. 2011; Pu et al. 2008, 2006) and magneto-optical phenomena in the subbandgap spectral region (Acbas et al. 2009). Magnetization-dependent transport effects have been playing a crucial role in determining magnetization magnitude and orientation in variety of DFSs, as discussed in Secs. III.B and III.C. Quantitative theory aiming at evaluation of conductivity tensor components $\sigma_{ij}(M)$ in (Ga,Mn)As-type DFSs, developed in the lowest order in disorder for films of (Ga,Mn)As and related systems, was already reviewed vis-à-vis results of extensive experimental studies (Jungwirth et al. 2008, 2006a; Nagaosa et al. 2010). An open and interesting question is how quantum localization and confinement will affect magnitudes of magnetization-dependent charge and heat transport in these ferromagnets. A breakdown of the proportionality between magnetization and the anomalous Hall effect found in thin and high quality (Ga,Mn)As films at low temperatures (Chiba et al. 2010) is just one example showing that considerable further effort will be devoted towards understanding of transport phenomena in DFSs.

B. Colossal magnetoresistance

A direct manifestation of interplay between magnetism and localization in magnetic semiconductors, as well as in DMSs and DFSs (Dietl 2008b), are colossal magnetoresistance (CMR) phenomena and a related effect of critical scattering (Novák et al. 2008). A peculiarity of CMR in DFSs is its strong dependence on the orientation of the magnetic field in respect to crystallographic axes (Gareev et al. 2010; Katsumoto et al. 1998). In general terms, magnetization rotation $\delta M$ results in a shift of the Fermi level, related to a change of anisotropy energy according to $\Delta E_F(\delta M) = d\Delta F_{\text{el}}(\delta M)/d\rho$. As $\epsilon_F$ controls the critical hole concentration $\rho_c$ corresponding to the metal-insulator transition (see, Secs. III.A and III.C), colossal effects are seen in transport (Gareev et al. 2010; Katsumoto et al. 1998) and tunneling (Pappert et al. 2006) for samples with hole densities close to $\rho_c$. Importantly, the influence of quantum localization persists well beyond the immediate vicinity of $\rho_c$. 

FIG. 26 (Color online) Spin injection from (Ga,Mn)As. (a) Schematic band diagram (a) of the three terminal device (b) allowing to bias independently the Esaki and light emitting diode ($V_{\text{EB}}$ and $V_{\text{CB}}$, respectively); (c) circular polarization $P_{\text{EL}}$ of light emitted along the growth and magnetization direction vs. $V_{\text{EB}}$ at various $T_C$ (adapted from Kohda et al. 2006). (d) Spin current polarization $\Pi$ for spin injection to n-GaAs ($V < 0$) or spin extraction from n-GaAs ($V > 0$) via contacts of the Esaki diode in non-local magnetotransport measurements (adapted from Ciorga et al. 2009).
C. Coulomb blockade

One of signs indicating that quantum-localization effects persist up to $p \gg p_c$ are signatures of the Coulomb blockade found in nanoconstrictions of $(Ga,Mn)As$ (Schlapp et al., 2009; Wunderlich et al., 2006), pointing to substantial nano-scale fluctuations in the hole density. Interestingly, in these experiments conductance oscillations were not only generated by sweeping the gate voltage but also by changing the direction of magnetization. The latter results from the dependence of the Fermi energy $\epsilon_F$ on $\delta \vec{M}$, as discussed in the previous subsection. This dependence leads to: (i) a charge redistribution within the nanoconstriction, as $\Delta \epsilon_F(\delta \vec{M})$ depends on $p$ that show spatial fluctuations (Wunderlich et al., 2006); (ii) changes in the localization and fluctuation landscape, as $\epsilon_F$ controls $p_c$. At sufficiently small values of $p - p_c(\vec{M})$, astonishingly large magnitudes of AMR were found in various nanostructures of $(Ga,Mn)As$ at low temperatures (Giddings et al., 2005).

D. Giant and tunneling magnetoresistance devices

Like in their metal counterpart, in trilayer structures of $(Ga,Mn)As$ the magnitude of vertical resistance increases if magnetization in the two ferromagnetic electrodes assumes an anti-parallel alignment, either if nonmagnetic central layer is of conductive p-GaAs (Chung et al., 2010) or forms a tunneling barrier, the case of AlAs (Chun et al., 2002) (Tanaka and Higo, 2001), GaAs (Chiba et al., 2004a), ZnSe (Saito et al., 2005) or paramagnetic $(Al,Mn)As$ (Ohya et al., 2009). In the latter case, the magnitude of tunneling magnetoresistance (TMR), $(R_{\uparrow\downarrow} - R_{\uparrow\uparrow})/R_{\uparrow\uparrow}$, attained 175% at 2.6 K for the barrier thickness $d = 4$ nm.

Do magnetic tunnel junctions (MTJs) of DFSs exhibit specific features? As shown in Fig. 27, the magnitude of TMR, grows (up to 76% at 8 K) when the width of the AlAs barrier diminishes down to 1.5 nm (Tanaka and Higo, 2001). At the same time, the values of TMR are seen to depend on the direction (in respect to crystallographic axes) of the magnetic field employed to reverse sequentially magnetization at a coercive force of particular ferromagnetic electrodes. In this family of phenomena, known as tunneling anisotropic magnetoresistance (TAMR), particularly spectacular is the case of the junction with one nonmagnetic electrode, e.g., $(Ga,Mn)As/AlO_2/Au$ (Gould et al., 2004), in which the vertical resistance decreases (by 3%) when rotating magnetization from an easy to a hard in-plane direction. Actually, a linear dependence between the decrease of the MTJ resistance (up to 10%) and the energy of magnetic anisotropy was found for various magnetization orientations in $(Ga,Mn)As/ZnSe/(Ga,Mn)As$ (Saito et al., 2005). Another characteristic feature is a fast decay of TMR magnitude with the bias voltage $V$ – in $(Ga,Mn)As/ GaAs/ (Ga,Mn)As$ at 4.7 K, the TMR magnitudes drops from 100% to 20% when $V$ increases to 0.1 V (Chiba et al., 2004a). These findings are compared to theoretical expectations in Sec. X.F.

FIG. 27 Junction resistance (a) and the magnitude of TMR measured for two in-plane directions of the magnetic field (b) at 8 K as a function of the barrier width for the MTJ structure show in the inset (b). Adapted from Tanaka and Higo 2001.

There are a range of MTJ properties escaping up to now from a straightforward quantitative modeling. In particular, $I(V)$ characteristics of MnAs/AlAs/(Ga,Mn)As $(Chun et al., 2002)$ and $(Ga,Mn)As/GaAs/(Ga,Mn)As$ MTJs (Pappert et al., 2006; Rüster et al., 2005) showed a Coulomb gap, a prominent manifestation of how important are correlation effects in quantum localization, as discussed in Secs. V.B and VII.C. Presumably, these effects, together with temperature dependent magnetization of Mn spins residing in a depleted interfacial layer, accounted for a threefold increase, up to 290%, of TMR between 4.7 and 0.3 K in an $(Ga,Mn)As/GaAs/(Ga,Mn)As$ MTJ (Chiba et al., 2004a). A question also arises to what extent the Coulomb gap affected the magnitude and magnetic anisotropy of tunneling thermopower in a GaAs:Si/GaAs/(Ga,Mn)As MTJ (Naydenova et al., 2011).
E. Double barrier structures

A series of works [Elsen et al., 2007; Muneta et al., 2012; Ohno et al., 1998; Tran et al., 2009] were devoted to tunneling phenomena in double barrier MBE-grown (Ga,Mn)As/AlAs/GaAs/AlAs/p-GaAs:Be structures deposited onto p'-GaAs:Be substrates. To minimize effects of inter-diffusion, few nm-thick GaAs separators were additionally inserted between AlAs barriers and the p-type electrodes. Except for the top (Ga,Mn)As electrode, all layers were deposited at high temperatures ($T_g \geq 600^\circ$C). As shown in Fig. 28, sharp peaks in the dynamic conductance $dI/dV$ as a function of bias voltage $V$ were observed [Ohno et al., 1998]. These and related results [Elsen et al., 2007; Muneta et al., 2012; Tran et al., 2009] led to a number of conclusions. In particular, the appearance of resonances for both polarities of bias voltage and the magnitudes of their relative distances demonstrated the presence of resonant tunneling via quantized hole subbands in the GaAs quantum well [Elsen et al., 2007; Ohno et al., 1998]. Second, the absence of positive bias of resonances corresponding to the hole ground state subbands in GaAs quantum well (HH1 and LH1) indicated, in accord with the TMR results discussed in the previous subsection (Sec. V.D), that the Fermi level of (Ga,Mn)As resides about 0.1 eV above the valence band top of GaAs [Elsen et al., 2007]. Finally, up to 5 times larger distances between resonances if holes were injected from (Ga,Mn)As (positive bias) than in the case when (Ga,Mn)As was a collecting electrode (negative bias) pointed to an asymmetry in the structure layout [Elsen et al., 2007; Muneta et al., 2012; Ohno et al., 1998]. This asymmetry was linked to a much lower value of the hole concentration ($p \approx 10^{18}$ cm$^{-3}$) and, thus, longer depletion length in GaAs:Be comparing to (Ga,Mn)As, leading to a rather different effective barrier width on the collector side for the two polarities [Elsen et al., 2007], as shown in Fig. 28.

In these devices three magnetic signatures were observed. First, a spontaneous and temperature dependent splitting of two peaks was revealed when holes were injected from (Ga,Mn)As, the effect visible in Fig. 28 [Ohno et al., 1998]. The temperature dependence of the splitting showed a Brillouin-type behavior with $T_C \approx 70$ K. Second, for the same bias, the magnitudes of resonance peaks were found dependent on magnetization orientation – they were reduced by about 10% when magnetization was turned from the easy axis [100] to the hard [001] direction [Elsen et al., 2007]. The effect was examined quantitatively [Elsen et al., 2007] within the $p$-$d$ Zener model exposed in Sec. X.F and the outcome is shown in Sec. X.F. Third, it was demonstrated that aforementioned magnetization rotation resulted in a shift of resonance positions for negative bias, the effect assigned to a decrease of the work function when magnetization was moved away from the easy direction [Tran et al., 2009]. Magnetic effects, were also detected by in structures containing on its top an additional AlAs barrier and a (Ga,Mn)As layer [Muneta et al., 2012]. Here, a change of series resistance associated with parallel and antiparallel arrangement of (Ga,Mn)As magnetizations led to a shift of resonance positions. These and related experiments demonstrate, therefore, how to gate electric current by manipulating with magnetization.

Double barrier structures deposited at low temperatures ($T_g \approx 230^\circ$C), in which the bottom GaAs:Be layer was replaced by (Ga,Mn)As, showed significantly different properties [Mattana et al., 2003]. In particular, the absence of resonances and the magnitude of TMR (about 40%) demonstrated that rather than resonant, sequential tunneling accounted for hole transport, the effect pointing out to a shortening of the phase coherence time below the dwell time in GaAs wells grown by LT-MBE [Mattana et al., 2003]. Even lower values of TMR (below 2%) were found in similar structures containing (In,Ga)As wells [Ohya et al., 2005], indicating that spin relaxation time became shorter than the dwell time in this case.

In another type of investigated structures, the GaAs quantum well in the original design (Fig. 28) was replaced by a (Ga,Mn)As layer of various thicknesses up to 20 nm grown by LT-MBE [Ohya et al., 2007a, 2010a]. In these devices, a resonance in $dI/dV$ was observed at $V \approx -0.1$ V, accompanied by one or two satellite features visible in $d^2I/dV^2$ at $V < 0$. A slight shift of the resultant oscillatory pattern was resolved on going from parallel to antiparallel magnetization orientation of the two (Ga,Mn)As layers, leading to a TMR-like behavior with a relative change of current for parallel and antiparallel magnetization orientations reaching 40%. It was demonstrated that the position and the magnitude of these os-
evant (Ga,In,Mn)As quantum well embedded by AlAs-rich barriers (or AlAs and the Schottky barrier underneath the Au film); (ii) particular features correspond to subsequent hole subbands starting from the ground state HH1 level, the assumption allowing to describe (with four adjustable parameters) the position and evolution of the features with the layer thickness; (iii) since a negative voltage has to be applied to reach the ground state subband in (Ga,In,Mn)As, the hole Fermi level is pinned by an impurity band located about 50 meV above the valence band top, implying that up to $10^{21}$ cm$^{-3}$ holes reside in a narrow band separated from the valence band in both (Ga,Mn)As and (In,Mn)As; (iv) the valence band states are entirely immune to the presence of Mn ions, which results in the lack of exchange splitting and high coherency of quantized states even for a 20 nm thick (Ga,Mn)As quantum well.

However, the above model was found questionable [Dietl and Sztenkiel 2011], particularly taking into account previous results on tunneling in (Ga,Mn)As [Richardella et al. 2010] as well as in double well structures involving (Ga,Mn)As layers [Elsen et al. 2007; Mattana et al. 2003; Ohno et al. 1998; Tran et al. 2009]. It was suggested [Dietl and Sztenkiel 2011] that the findings can be interpreted by assigning the features in $d^2I/dV^2$ at $T \ll T_C$ to sequential hole tunneling transitions from quantized hole subbands in the accumulation layer of GaAs:Be to continuum of states determined by quenched disorder in (Ga,Mn)As, followed by transitions to the top electrode. The features originating from quantum states in GaAs:Be can be resolved in this case since competing resonances associated with the quantum states in the well are washed out by disorder in (Ga,Mn)As. They appear at $V_R < 0$, where $|V_R|$ scales with the Mn concentration dependent valence band offset between (Ga,Mn)As and GaAs. This new interpretation, contradicting the presence of an impurity band, is consistent with: (i) the failure to observe the genuine impurity band directly by tunneling spectroscopy; (ii) the absence of resonant tunneling when the well consisted of disordered GaAs grown by LT-MBE [Mattana et al. 2003]; (iii) the presence of the features only for $V < 0$, in contrast to the case of GaAs well grown by HT-MBE, where resonances appeared for both bias polarities, as expected for resonant tunneling [Elsen et al. 2007; Ohno et al. 1998]; (iv) the non-occurrence of corresponding features when GaAs:Be was replaced by (Ga,Mn)As [Mattana et al. 2003]; (v) the evolution of the feature positions with the device resistance (showing correlation with the thickness of (Ga,Mn)As layers, see Fig. 29); (vi) the presence of TMR-like behavior but at the same time the lack of spin splitting of electronic states giving rise to the tunneling features (the splitting of the features is at least two orders of magnitude smaller than exchange splitting of states in any known uniform magnetic semiconductors with a corresponding magnitude of magnetization); (vii) the similarity of the results for wells of ferromagnetic (Ga,Mn)As grown on AlAs/GaAs:Be, and (In$_{0.53}$Ga$_{0.47}$,Mn)As and

cillations in $dI/dV$ and TMR vs. $V$ can be efficiently controlled by a third electrode biasing the (Ga,Mn)As quantum well [Ohya et al. 2010b], a valuable step towards development of a spin transistor. A similar oscillatory behavior of $d^2I/dV^2$ at $V < 0$ was also detected in simpler structures, in which the top barrier and the (Ga,Mn)As electrode were replaced by an Au film deposited directly onto the lower (Ga,Mn)As layer, resulting in the layout Au/(Ga,Mn)As/AlAs/p-GaAs:Be, where the (Ga,Mn)As layer exhibited $T_C$ up to 154 K [Ohya et al. 2011b]. Corresponding results were also obtained for structures in which (Ga,Mn)As was replaced by (Ga,In,Mn)As ($T_C$ up to 135 K) or (In,Mn)As ($T_C \approx 47$ K) and p-GaAs:Be by p-(Ga,In,Mn)As:Be; employing p-InP substrates in this case [Ohya et al. 2012].

These comprehensive investigations showed consistently that the oscillatory pattern in $d^2I/dV^2$: (i) appeared for $V < 0$; (ii) tended to spread and move towards higher voltages $|V|$ when the thickness of the bottom (Ga,Mn)As layer decreased, as shown in Fig. 29; (iii) did not reveal any temperature-dependent splitting of quantized hole states.

In order to interpret these findings it was suggested in these works that: (i) the oscillations in $d^2I/dV^2$ witness resonant tunneling via quantized subbands in the rela
(In,Mn)As grown on AlAs/In_{0.53}Ga_{0.47}As:Be.

In related structures (Ga,Mn)As/AlAs/(Ga,Mn)As/(Al,Ga)As/ GaAs:Be, where the AlAs and (Al,Ga)As barriers were 1.5 and 100 nm thick, respectively, negative dynamic resistivity features with various degree of sharpness were seen in the $I(V)$ dependence in a number of tested devices [Likovich et al., 2000]. These features underwent a shift to higher values of bias $V$ for antiparallel magnetization orientations, which resulted, in the most prominent case, in a TMR-like signal as large as 30%.

F. Read-write devices

Figure 30 highlights layout and operation principle of (Ga,Mn)As-based magnetic memory cells, in which AMR-related phenomena allowed for bit reading, whereas either an external magnetic field or spin-polarized electric current served to bit writing.

VI. INTERLAYER COUPLING, FERROMAGNETIC PROXIMITY EFFECT, AND EXCHANGE BIAS

A. Interlayer coupling

Low-temperature magnetotransport studies of (Ga,Mn)As/Al$_{y}$Ga$_{1-y}$As/ (Ga,Mn)As trilayer structures revealed ferromagnetic coupling between (Ga,Mn)As layers, whose strength decayed with temperature and Al content, $0.14 < y < 1$, in the 2.8 nm thick Al$_{y}$Ga$_{1-y}$As spacer [Chiba et al. 2000]. A ferromagnetic interlayer interaction was also found by neutron investigations of (Ga,Mn)As/GaAs superlattices [Kea et al. 2010; Sadowski et al. 2002], for the whole explored range of GaAs thicknesses, $0.7 < d < 7$ nm [Chung et al. 2010; Kea et al. 2001; Sadowski et al. 2002]. However, for GaAs:Be spacers with hole density of $1.2 \times 10^{20}$ cm$^{-3}$, the coupling was still ferromagnetic for $d = 1.2$ and 2.3 nm but became antiferromagnetic when increasing $d$ to 3.5 and 7.1 nm [Chung et al. 2010]. Since ferromagnetism is spatially inhomogeneous in (Ga,Mn)As, a long range dipole-dipole coupling can account for this observation [Kakazei et al. 2005].

In the case of MnAs/p-GaAs/(Ga,Mn)As, a ferromagnetic coupling was found, whose strength monotonically decayed with the thickness of the p-GaAs layer in the studied range $1 \leq d \leq 5$ nm [Wilson et al. 2010].

Theoretical modeling of interlayer coupling is discussed in Sec. X.B.

B. Ferromagnetic proximity effect

The Fe and Mn L$_{2,3}$ XMCD spectra recorded at room temperature for Fe/(Ga,Mn)As heterostructures demonstrated the presence of Mn spin ordering antiparallel to Fe spins extending 2 nm [Maccherozzi et al. 2008] or 0.7 nm [Olejnìk et al. 2010] into (Ga,Mn)As. The uncovered character of the ferromagnetic proximity effect was reproduced by DFT computations [Maccherozzi et al. 2008]. Interestingly, if the thickness of (Ga,Mn)As was reduced down to 5 nm, the ferromagnetic proximity effect allowed to shift up by 35 K the temperature range in which both spontaneous magnetization and spin injection to n-GaAs through a Fe/(Ga,Mn)As/n-GaAs Esaki diode could be detected [Song et al. 2011]. These robust spin selective contacts made it possible to probe electrically the spin Hall effect in n-GaAs [Ehrt et al. 2012].

C. Exchange bias

As mentioned above, the coupling of Mn ions in (Ga,Mn)As to an Fe overlayer is antiferromagnetic. Accordingly, below $T_C$ of (Ga,Mn)As, its magnetic properties can be described in terms of exchange bias, leading to enlarged coercivity and a history dependent shift of the hysteresis loop center away from the zero magnetic field. Such phenomena were noted for MnAs/(Ga,Mn)As [Wilson et al. 2010; Zhu et al. 2007] and Fe/(Ga,Mn)As [Olejnìk et al. 2010], and interpreted by the exchange spring model [Wilson et al. 2010]. Magnetization processes related to exchange bias were found and examined for MnO/(Ga,Mn)As heterostructures, in which Néel and Curie temperatures were comparable [Eid et al. 2004; Ge et al. 2007]. Similarly, MnO and MnTe exchange-biased [Ge,Mn]Te [Lim et al. 2012]. Furthermore, nanocrystalline precipitates of ferromagnetic MnAs in (Ga,Mn)As [Wang et al. 2006] and of antiferromagnetic MnTe in (Ge,Mn)Te [Lechner et al. 2010] resulted in an enhancement of the coercivity field.

VII. ELECTRONIC STATES

A. Vonsovsky’s model and Mott-Hubbard localization

Experimental results discussed in the subsequent subsections (Secs. VII.B.1-2) indicate that magnetic moments of Mn in DFSs are localized, not itinerant. According to the Vonsovsky model [Vonsovsky 1946], the relevant electron states can then be divided into two categories [Dietl 1981]: (i) localized magnetic d-like levels described by the Anderson impurity model [Anderson 1961] or its derivatives [Parmenier 1973]; (ii) effective-mass band states that can be treated within tight binding or kp methods [Luttinger and Kohl 1955], employed commonly for quantitative simulations of functionalities specific to semiconductors, their alloys and quantum structures. Importantly, pertinent properties of holes, for instance valence band lineups in semiconductor heterostructures, result from hybridization between anion p orbitals and cation d orbitals [Wei and Zunger 1987]. In the case of open d shells, this hybridization leads additionally to a strong p-d exchange interaction [Bhattacharjee et al. 1983; Dietl 1981] accounting for
outstanding spintronic properties of DFSs. There exists
also an \( s - d \) exchange interaction in DMSs but because
of its relatively small magnitude, the corresponding \( T_C \)
values were found to be below 1 K \cite{Andrearczyk et al. 2001, Dietl et al. 1997}.

In the next two sections (Secs. VIII and IX.A),
Vonsovskii’s electronic structure is employed to describe
spin-spin exchange interactions within the superexchange
and \( p-d \) Zener models. This is followed (Sec. X) by
discussing the applicability of this approach to a quanti-
tative description of spintronic functionalities of DFSs.
We note in passing that since implementations of den-
sity functional theories within local density approxima-
tions (LDA) cannot handle adequately the physics of the
Vonsovskov model, particularly the Mott-Hubbard local-
ization, other \textit{ab initio} approaches are being developed
for DFSs, for instance, incorporating into the LDA hy-
brid functionals \cite{Stroppa and Kresse 2009} or the dy-
namic mean-field approximation \cite{Di Marco et al. 2013}.

B. Mn localized magnetic moments

1. Magnetic resonances

In the case of III-V and also III-VI compounds as well
as group IV semiconductors, Mn ions introduce both
spins and holes. According to electron paramagnetic
studies in the impurity limit \( x \ll 10^{-3} \), the Landé factor
of neutral Mn acceptors (Mn\(^{3+}\)) in GaAs:Mn is \( g = 2.77 \)
\cite{Schneider et al. 1987, Szczynsko et al. 1999}, the value
consistent with a moderate binding energy \( E_1 = 110 \) meV
(see, Fig. 8) and an antiferromagnetic character of \( p-d \) ex-
change coupling between the hole spin \( J = 3/2 \) and the
Mn\(^{2+}\) center in a high spin \( S = 5/2 \) state \cite{Schneider et al. 1987}. Spin resonance in GaP:Mn \cite{Kresse et al. 1996} as well as Mn-related optical spectra in GaN:Mn \cite{Bonanni et al. 2011, Wolos and Kamińska 2008}, were
successfully described in terms of the group theory for
Ga-substitutional localized Mn\(^{3+}\) centers corresponding
to \( S = 2 \). Magnetization studies suggest that this spin
state of Mn ions persists up to at least \( x = 0.1 \) \cite{Kunert et al. 2012}.

In contrast, in GaN:Mn samples containing compen-
sating donor impurities, the character of hyperfine splitting
and \( g = 2.01 \pm 0.05 \) \cite{Bonanni et al. 2011, Graf et al. 2003a, Wolos and Kamińska 2008} demonstrated
the presence of Mn\(^{2+}\) ions \((S = 5/2, L = 0)\). Similar
spectra were found on increasing Mn concentrations in
(Ga,Mn)As \cite{Fedorych et al. 2002, Szczynsko et al. 1999} and
(In,Mn)As \cite{Szczynsko et al. 2001}. In the case of ar-
senides, however, the presence of Mn\(^{2+}\) spectra indicates
detaching of holes from individual negatively charged
Mn\(^{2+}\) acceptors at \( x \gtrsim 0.001 \) rather than compensation
by donors.

For still higher Mn concentrations \((x \gtrsim 0.02)\), ex-
tensive ferromagnetic resonance studies, carried out for
(In,Mn)As \cite{Liu et al. 2005}, (Ga,Mn)As \cite{Khanen et al. 2008, Liu and Furdyna 2006}, and (Ga,Mn)P \cite{Bihler et al. 2007}, pointed to the Landé factor \( g = 1.93 \pm 0.5 \)
at low temperature. A slight deviation from the value
\( g = 2.00 \) expected for Mn\(^{2+}\), suggests an admixture of
orbital momentum, brought presumably about by spin
polarized holes present in these DFSs below \( T_C \). The
value \( S = 5/2 \) was also evaluated from the neutron
scattering length in (Ga,Mn)As \cite{Kepa et al. 2001}. In
contrast, according to extensive magnetization measure-
ments \cite{Kunert et al. 2012, Sawicki et al. 2012}, trivalent
Mn\(^{3+}\) configuration dominates up to at least \( x = 0.1 \)
in Ga\(_{1-x}\)Mn\(_x\)N. This finding is consistent with a large
ionization energy of Mn acceptors in GaN (see, Fig. 8),
leading to strong localization of holes on individual Mn
ions even at high Mn content \( x \).

In the case of II-VI \cite{Dietl 1994, Furdyna and Kossut 1988} and IV-VI DMSs \cite{Bauer et al. 1992}, Mn ions sub-

FIG. 30 Memory cells working at 4.2 K involving three (a), two (b), and four (c) 200 nm wide and few µm long (Ga,Mn)As 

nanobars forming a junction and contacted to current leads (from Figielski et al. 2007, Pappert et al. 2007, and Mark et al. 2011, respectively). Due to strain relaxation magnetization is aligned along the nanobar long axes but its orientation in particular bars can be preselected by applying and removing an external magnetic field of an appropriate in-plane direction [bit writing in (a,b) and memory cell initiation in (c)]. Owing to the AMR effect associated with a domain wall in the junction [enhanced by carrier depletion in (b)], the value of two terminal resistance can tell relative magnetization directions in particular bars [bit reading in (a,b)]. In (c), magnetization direction along one of two cubic axes in the central disk (of diameter 650 nm) can be preselected by a current pulse along the pair of wires that are spin-polarized in the required direction [bit writing in (c)]. Tunneling resistance of the Au/AlO\(_x\)/(Ga,Mn)As MTJ deposited over the central disk depends on magnetization orientation in the disk [bit reading in (c)].
stitute divalent cations and assume Mn$^{2+}$ charge states characterized by a high spin and vanishing orbital momentum ($S = 5/2$, $L = 0$). This spin state was confirmed by ferromagnetic resonance studies on ferroelectric and ferromagnetic (Ge,Mn)Te (Dziawa et al., 2008).

2. High energy spectroscopy

It is worth recalling that ultraviolet and soft x-ray methods probe usually film regions adjacent to the surface, so that an adequate surface preparation is of paramount importance. With this reservation, we note that the picture presented above, namely that in all Mn-based DFSs but (Ga,Mn)N, Mn assumes single valent $2+$, $S = 5/2$ configuration, was strongly supported by photoemission and x-ray spectroscopy. In particular, Mn ions in both (In,Mn)As (Chiu et al., 2005), and (Ga,Mn)P (Stone et al., 2006) were measured the 2+ and Mn level by quantum hopping from As valence states.

Similarly, XMCD studies at the Mn $L$-edge corroborated the 2+ and $S = 5/2$ configuration of Mn ions in ferromagnetic (In,Mn)As (Okabayashi et al., 2002) and (Ga,Mn)As were found in a single valence state characterized by the Mn d electron count $n_d = 5.3 \pm 0.1$ (Okabayashi et al., 1999). A measurable enhancement over $n_d = 5$ can be interpreted as the presence of $p$-$d$ hybridization leading to a non-zero occupancy of the $d^6$ Mn level by quantum hopping from As valence states. Additionally, XMCD studies at the Mn $K$-edge corroborated the 2+ and $S = 2$ state of Mn in ferromagnetic (Ga,Mn)N. It was found, however, that surface donor defects turned adjacent Mn ions into divalent Mn$^{2+}$ states (Freeman et al., 2007), visualized also by photoemission studies (Hwang et al., 2005). Coexistence of Mn$^{2+}$ and Mn$^{3+}$ was also observed in x-ray absorption spectroscopy (XAS) (Sonoda et al., 2006). In contrast, in uncompensated samples of (Ga,Mn)N both $x$-$y$ x-ray absorption near-edge structure (XANES) (Bonanni et al., 2011) and x-ray emission spectroscopy (XES) (Devillers et al., 2012) pointed to 3+ charge state of Mn in GaN.

C. Anderson-Mott localization of carriers

As already mentioned in Sec. III.A, interplay between hole localization and hole-mediated ferromagnetism is arguably the most characteristic feature of DFSs. In particular, $p$-$d$ hybridization that accounts for exchange coupling between localized spins and itinerant holes, shifts at the same time the metal-insulator transition (MIT) to higher hole concentrations.

It is worth noting that current ab initio methods designed to handle disorder, such as the coherent potential approximation, are not capturing the physics of the Anderson-Mott localization. There are preliminary quantum Monte Carlo approaches aiming at elaborating computational schemes that might provide quantitative information in the regime of quantum localization in many body interacting systems (Fleury and Waintal, 2008). Nevertheless, for the time being it is safe to argue that the current theory of the Anderson-Mott MIT does not offer quantitative predictions on: (i) the magnitude of the critical hole concentration $p_c$ corresponding to the MIT; (ii) the absolute value of conductivities $\sigma_{ij}$, and (iii) the nature of excitations at high energies $\omega \gtrsim 1/\tau$. Empirically, some of these excitations exhibit single impurity characteristics, even on the metallic side of the MIT. This duality of behaviors is described phenomenologically within the so-called two-fluid model of electronic states (Paalanen and Bhatt, 1991), the approach exploited extensively to understand DMSs (Dietl, 2008b; Dietl et al., 2000), and now acquiring some theoretical support (Terletskaya and Dobrosavljević, 2011).

However, the current theory does provide quantitative and experimentally testable information on the values of critical exponents as well as on the dependence of $\sigma_{ij}(p)$ on dimensionality, frequency, temperature, magnetic field, spin scattering, and spin splitting in the metallic regime $k_F \ell > 1$, where $\ell$ is the microscopic mean free path (Altshuler and Aronov, 1985; Belitz and Kirkpatrick, 1994; Dietl, 2008b; Lee and Ramakrishnan, 1985). The appearance of these specific dependencies, known as quantum corrections to conductivity, heralds the failure of Drude-Boltzmann-like approaches in capturing the physics accounting for the magnitudes of $\sigma_{ij}(p)$. Importantly, a quantitative study of the quantum corrections can provide information on the thermodynamic density of states (DOS) $\rho_F = \partial p / \partial \epsilon_F$, which does not show any critical behavior across the MIT and assumes a value specific to the relevant carrier band (Altshuler and Aronov, 1985; Belitz and Kirkpatrick, 1994; Lee and Ramakrishnan, 1985). According to the same theory, with an accuracy of typically better than 20% (corresponding to a magnitude of the relevant Landau parameter of the Fermi liquid), the corresponding effective mass is equal to $m^*$ for low energy intraband charge excitations, provided by, e.g., cyclotron resonance studies.

In the region $k_F \ell < 1$, corresponding usually to $p \lesssim p_c$, renormalization group equations (Belitz and Kirkpatrick, 1994; Finkelstein, 1990; Lee and Ramakrishnan, 1985) can serve to assess the evolution of relevant characteristics, such as localization radius, dielectric constant, and one-particle DOS, with $p_c - p$. This DOS shows a Coulomb gap at the Fermi level on the insulator side of the MIT, $p < p_c$, which evolves into a Coulomb anomaly at $p > p_c$. The theory shows that in the weakly localized regime, the localization length $\xi$ is much longer than the effective Bohr radius $a_B$ of a single acceptor, so that band characteristics are preserved at distances smaller than $\xi$. Furthermore, because of large screening by weakly localized carriers, there are few, if any, bound
states associated with individual acceptors.

In the case of low dimensional systems (2D and 1D) a cross-over from the weakly to strongly localized regime, rather than the MIT, occurs at \( k_F \ell \approx 1 \). The absence of metallic regime means that, in principle, localization phenomena are relevant at any hole density.

D. Where do holes reside in DFSs?

As discussed in Sec. [III.A] the Fermi level is pinned by the deep Mn acceptor impurity band in (Ga,Mn)N, where charge transport might proceed only via phonon-assisted hopping. A broadly disputed question (Samarth, 2012; Wang et al., 2013) then arises, where do reside delocalized or weakly localized holes mediating ferromagnetic coupling in (Ga,Mn)As and related systems?

1. Photoemission

In addition to quantitative information on the \( d \) electron count (presented above) and \( p-d \) hybridization (discussed in Sec. [VII.E]), photoemission studies allow examining a shift of the Fermi level and modifications to the band structure introduced by Mn ions. In Fig. 31 the contribution of Mn 3d states to valence band DOS, obtained by angle integrated photoemission at various resonant excitation energies, are presented for (Ga,Mn)As containing above 5% of Mn. Corresponding findings are also shown for (In,Mn)As and (Cd,Mn)Te.

Several important conclusions emerge from these as well as from more recent spectra ([Kobayashi et al., 2013]. First, a general agreement between the (Ga,Mn)As data obtained for different energies of exciting photons (and, thus, absorption length) indicates that these results are not substantially affected by surface effects. Second, there is a considerable similarity between findings for (Ga,Mn)As, (In,Mn)As, and (Cd,Mn)Te: a center of gravity of the 3d-state contribution is at 4 eV below the Fermi level, and an additional local maximum appears deeper in the valence band. This indicates that the physics of \( p-d \) hybridization is similar in these systems. Third, the DOS magnitude decays to zero through a knee or a weak maximum at 0.2-0.4 eV on approaching the Fermi level, and an additional local maximum appears above the half-width 0.1-0.2 eV, as discussed in the subsections above and below (Secs. VII.C and VII.D.2 respectively). This conclusion was supported by angle-resolved photoemission studies on (In,Mn)As ([Okabayashi et al., 2002] and (Ga,Mn)As ([Gray et al., 2012]).

Another important finding of photoemission works was the demonstration that the total DOS tends to zero at \( \epsilon_F \) in (Ga,Mn)As ([Okabayashi et al., 1999] despite high values of hole concentrations. This reconfirmed the presence of a Coulomb anomaly at the Fermi level in this system, of the half-width 0.1-0.2 eV, as discussed in the subsections above and below (Secs. VII.C and VII.D.2 respectively). This conclusion was supported by angle-resolved photoemission studies on (In,Mn)As ([Kobayashi et al., 2013] which revealed a characteristic depression in DOS on approaching the \( \Gamma \) point.

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\(^{3}\) In one photoemission work a dispersion-less impurity-like band was detected above the valence band top in (Ga,Mn)As ([Okabayashi et al., 2001]. This observation was not confirmed by subsequent studies by the same group ([Kobayashi et al., 2013] Rader et al., 2004) and others ([Di Marco et al., 2013] Gray et al., 2012).
with respect to EEI is depending on the dimensionality of the sample can be observed in (Ga,Mn)As [19,20] even at 20 mK.

The size of the conductivity correction due to electroninteraction is depending on the diffusion constant //C0

/temperature dependency is

21D

N

PRL

et al.

et al.

et al.

et al.

et al.

et al.

et al.

The Anderson-Mott type of the MIT was well documented (Matsuda et al. 2011). According to the quantitative analysis (Dietl, 2008b; Dietl et al. 2006), they are less accurate for 1D systems. For 1D "..."

...corroborating that quantum interference, rather than trapping by individual impurities, accounts for hole localization in (Ga,Mn)As in the Mn concentration range relevant for ferromagnetism (Neumaier et al. 2009). Taking into account effects of disorder-modified hole-hole interactions, the magnitudes of slopes a in the dependence g(T) provide information on the thermodynamic DOS p(ETF) in the 1D and 3D cases (Altshuler and Aronov 1985; Lee and Ramakrishnan 1985). According to the quantitative analysis (Dietl 2008b; Neumaier et al. 2009; Sliwa and Dietl 2011), the values of a indicate that the effective mass of holes at the Fermi level in (Ga,Mn)As differs by less than by a factor of 2 comparing to that of the disorder-free GaAs valence band.

Since the external magnetic field has no effect on g(T) (Neumaier et al. 2008), the single-particle Anderson localization term, destroyed presumably by a demagnetizing field, does not contribute to the observed temperature dependence of conductivity in this ferromagnetic
The existence of sizable quantum localization effects indicates that the real part of intraband optical conductivity, $\sigma_1(\omega)$, in addition to a dispersion expected within the Drude theory for a GaAs-type complex valence band [Hankiewicz et al., 2004; Sinova et al., 2002], should show a significant drop with decreasing $\omega$ down to $\hbar \omega \approx k_B T$. The low energy gaps in DOS and conductivity not only share the same physical origin but involve the same energy scale and dispersion at low energies. Quantitative formulae describing the disappearance of quantum localization contributions to $\sigma$ with $\omega$ are theoretically known for a metallic case and low-energy excitations in a simple band, i.e., for $\omega \lesssim 1/\tau \lesssim \epsilon_F$ [Altshuler and Aronov, 1985; Lee and Ramakrishnan, 1985], where $\hbar / \tau$ is of the order of 0.1 eV in ferromagnetic Ga$_{1-x}$Mn$_x$As. Actually, interplay between Anderson-Mott effects and a Drude-like decay of intra-band conductance at high frequencies leads to a maximum in $\sigma(\omega)$, found at $\omega_m \sim 0.2$ eV in (Ga,Mn)As (Burch et al., 2008). Unfortunately, any detailed interpretation [Burch et al., 2008; Jungwirth et al., 2007; Kojima et al., 2007] of $\omega_m$ and its shift with $x$ or $T$ is rather inconclusive as no theory for $\sigma(\omega)$ is available in this cross-over regime, particularly for the complex valence band and in the presence of spin-disorder scattering. However, a comparison of $\sigma(\omega)$ across the MIT in (Ga,Be)As and (Ga,Mn)As (Chapler et al., 2011) provides a strong confirmation of the aforementioned persistence of localization effects even for $p \gg p_c$ in (Ga,Mn)As as well as demonstrates a similarity in the DOS and conductivity gaps in this DFS.

In contrast to $\sigma_1(\omega)$, an integral of $\sigma_1(\omega)$ over $\omega$ depends uniquely on the ratio of the hole density $p$ and a combination of the heavy and light hole masses, $m_{opt}$, for any strength of disorder according to the optical conductivity sum rule for intraband excitations [Sinova et al., 2002]. Figure 34 presents a comparison of $\sigma_1(\omega)$ and $\Delta N = \Delta p_{2D}/m_{opt}$ for (Ga,Be)As and (Ga,Mn)As determined at room temperature as a function of the gate voltage $V_{eff}$ that changes the hole concentration. The values of $\Delta N$ were obtained by the integration of $\sigma_1(\omega)$ up to a finite value $\omega_c$. A similar magnitudes of $\Delta N$ in both systems together with the evaluated value $m_{opt} \lesssim 0.42 m_0$ (Chapler et al., 2012) confirmed that the hole band of (Ga,Mn)As retains basic characteristics of the GaAs valence band for which $m_{opt}/m_0 = 0.25 - 0.29$ was theoretically predicted [Sinova et al., 2002].

This conclusion was corroborated by the magnitudes of room temperature thermoelectric power $S$ in (Ga,Be)As and (Ga,Mn)As examined in the same hole concentration range [Mayer et al., 2010], as shown in Fig. 35. It is worth noting that neglecting a phonon drag contribution, $S$ is proportional to a logarithmic derivative of conductivity over energy and, thus, to a first approximation, to thermodynamic DOS per one carrier. A large magnitude of $S$ would therefore be expected if the Fermi level were pinned by a large DOS of an impurity band [Heremans et al., 2012].

In summary, according to the data discussed in this and previous subsection, holes in (Ga,Mn)As and related DFSs reside in a host-like valence band that is, however, strongly affected by the proximity to the MIT. This conclusion is further supported by the outcome of photorefractive studies [Yastrubchak et al., 2011].

E. Experimental studies of p-d exchange energy

The energy distribution of Mn 3d states shown in Fig. 31 can serve to evaluate parameters of the Anderson Hamiltonian characterizing hybridization between p-like valence bands in tetrahedrally coordinated semiconductors and d states of Mn ions. This was carried out em-

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4 Some authors [Honolka et al., 2007; Mitra et al., 2010] analyzing $\sigma(T)$ up to 4 K, found that $\sigma(T) = \sigma_0 + AT^\alpha$, where $\alpha = 1/3$. This dependence was interpreted in terms of a renormalization group equation [Altshuler and Aronov, 1985] applicable close to the MIT, where $\sigma_0 < AT^\alpha$ and then $1/3 \leq \alpha \leq 1/2$ in the 3D case [Belitz and Kirkpatrick, 1994; Lee and Ramakrishnan, 1985]. Furthermore, the apparent value of $\alpha$ can be reduced above 1 K by a cross-over to the regime, where the effect of scattering by magnetic excitations onto quantum corrections to conductivity becomes significant [Dietl, 2008b].
Lines are calculated (neglecting phonon drag contribution) for the GaAs valence band [using the standard six band Luttinger model and parameters (Dietl et al. 2001b)], assuming that ionized impurity and acoustic phonon scattering dominates (upper and lower curve, respectively); the actual value of thermopower $S$ should lie between lines obtained for heavy and light hole bands in each case. From Slawa and Dietl 2011.

A configuration-interaction method to describe photoemission and x-ray absorption spectra in II-VI and III-V DMSs (Hwang et al., 2005; Mizokawa et al., 2002). By using the Schrieffer-Wolf transformation one then obtains the magnitudes of energies $N_0 |\beta |$ and $N_0 W$ (Benoit à la Guillaume et al., 1992), characterizing respectively spin-dependent and spin-independent parts of the local potential introduced by individual Mn ions. These energies are proportional to square of hybridization matrix element $V_{pd}$, and inversely proportional to distances of the Fermi level to $d^5$ and $d^6$ states, $\epsilon_d$ and $U - \epsilon_d$.

When the Mn potential is too weak to bind a hole, and a possible Coulomb contribution (existing in III-V DMSs) is screened by carriers, the first order perturbation theory (virtual-crystal and molecular-field approximations) describes the valence band offset and spin splitting of valence band states leading to, $dE_v(x)/dx = N_0 W$ and $H_{pd} = \beta s \bar{M}/g \mu_B$, where $M$ is magnetization of Mn ions and $g = 2.0$ is their Landé factor. If, however, the perturbation introduced by a single Mn impurity is so strong that a bound state appears, the influence of the Mn ion ensemble on the band valence structure has to be treated in a non-perturbative way (Dietl, 2008a). Reversed signs of the band offset and of spin splitting can appear in this strong coupling case. In this regime, in addition to $W$ and $\beta$, the spectrum at $k = 0$, for a given hole mass value $m^*$, is determined by at least one more parameter, namely, the spatial extend $b$ of the perturbation introduced by Mn or alternatively by the ratio of the corresponding potential well depth to its minimum value giving rise to a bound state, $U/U_c$ (Benoit à la Guillaume et al., 1992; Dietl, 2008a).

Exciton magnetospectroscopy has been the primary source of information on exchange splittings of bands, and thus on $\beta$ in DMSs without carriers. Magnetooptical studies of hole-doped (Cd,Mn)Te quantum wells (Boukari et al., 2002; Haury et al., 1997; Kossacki et al., 2004a) demonstrated that interband transitions are considerably affected by hole-hole interactions as well as by the Moss-Burstein shift that accounted for the sign inversion of MCD comparing to the case of undoped (Cd,Mn)Te (Haury et al., 1997). Despite that modulation doping was employed, evidences for scattering broadening of DOS were also found (Boukari et al., 2002). Polarization-resolved magnetoabsorption measurements in the band gap region of ferromagnetic (Ga,Mn)As (Szczytko et al., 1999), were also interpreted taking into account the Moss-Burstein shift and scattering broadening in the heavy hole band (Szczytko et al., 2001). In Fig. 30 the magnitudes of $N_0 |\beta |$ determined from high energy spectroscopy and interband magnetooptical studies are collected. The reversed signs of the apparent $N_0 |\beta |$ values determined from excitonic magnetoreflectivity within the molecular field approximation for (Zn,Mn)O and (Ga,Mn)N show that these systems are in the strong coupling regime (Dietl, 2008a). To a first approximation $|\beta | = -54$ meV nm$^3$ describes the available data for various tetrahedrally coordinated DMSs in the weak coupling regime (except for mercury chalco- genides (Furdyna and Kossut, 1988), where $|\beta |$ magnitudes appear somewhat smaller). It corresponds to $N_0 |\beta | = -1.2$ eV for (Ga,Mn)As, the value consistent with the results of photoemission (Okabayashi et al., 1999, 1998), magnetoabsorption (Szczytko et al., 2001), and the energy difference between the states corresponding to a parallel and antiparallel spin arrangement of a bound hole bound and a Mn ion in the limit of low Mn concentrations, $\Delta \epsilon = 8\pm3$ meV (Averkiev et al., 1987; LimarsÌon et al., 1997). The antiferromagnetic character of this coupling was also corroborated by a direction of current-induced spin torque in (Ga,Mn)As (Secs. III.D and III.E) and a sign of circular polarization in spin-LEDs (Sec. IV).

Since the pioneering studies of interband MCD in (Ga,Mn)As (Ando et al., 1998), this technique has been widely employed to asses effects of $p$-$d$ coupling onto the valence band of III-V DFSs. In particular, the theory of optical absorption and MCD involving six valence subbands and conduction band was developed for thin films of carrier-controlled DFSs (Dietl et al., 2001b), and showed to describe puzzling MCD data for (Ga,Mn)As (Beschoten et al., 1999) with one fitting parameter – the Mn-induced band gap offset. Its sign (corresponding to gap narrowing) and value (about 0.2 eV for $x = 5\%$) are consistent with a net magnitude of many body effects (Dietl et al., 2001b) and $p$-$d$ hybridization in the weak coupling limit (Dietl, 2008a) and, moreover, with their
A vast majority of nonmetallic TM compounds are antiferromagnets or ferrimagnets. In the absence of carriers, short-range antiferromagnetic coupling determines magnetic properties of DMSs containing Mn$^{2+}$ ions, the case of, for instance, Mn-based II-VI DMSs (Shapira and Bindilatti 2002). The relevant coupling between localized spins—the superexchange (Anderson 1950; Goodenough 1958; Kanamori 1959)—proceeds via p-d hybridization with bands of anions residing on the path between the TM spins in question. For this indirect interaction, to the lowest relevant order perturbation theory, the exchange energy $J_{ij}$ is proportional to $|V_{pd}|^4$ and decays fast with the distance $R_{ij}$ between magnetic ions.

In random antiferromagnets, such as intrinsic II-VI DMSs, frustration of interactions in spin triads and larger Mn clusters leads to spin-glass freezing. According to comprehensive studies, critical temperature $T_\text{f}$ is about 1 K at $x = 0.1$ and grows with $x$ as $T_\text{f} \propto x^m$, where $m = 2.3 \pm 0.1$ in Mn-doped cadmium and zinc chalcogenides (Twardowski et al. 1987). Scaling invariance (Ramam and Souletie 1982) implies then that the exchange energy $J_{ij}$ decays with the spin-spin distance as $R_{ij}^{-\lambda}$, where $\lambda = md = 6.8 \pm 0.3$ for 3D systems.

In these systems, it is usually possible to parametrize experimental values of magnetization $M(T, H)$ by the paramagnetic Brillouin function for $S = 5/2$ (Gaj et al. 1979),

$$M(T, H) = g\mu_B N_0 x_{\text{eff}} B_S \left[ \frac{g\mu_B H}{k_B(T + T_{AF})} \right], \quad (7)$$

where $x_{\text{eff}} < x$ and $T_{AF} > 0$ describe a reduction of magnetization by antiferromagnetic interactions. The values of these parameters increases with temperature (Spalek et al. 1986), $x_{\text{eff}} \rightarrow x$ and $T_{AF} \rightarrow -\Theta_0$, where

$$\Theta_0 = \frac{1}{3} S(S+1) \sum_j z_j J_j. \quad (8)$$

Here, the summation extends over the subsequent cation coordination spheres; $z_j$ is the number of cations in the sphere $j$, and $J_j \equiv J_{ij} < 0$ is the corresponding Mn-Mn exchange energy in the Hamiltonian $\mathcal{H}_{ij} = -J_{ij} S_i S_j$. The values of $J_{ij}$ were successfully modeled for II-VI Mn-based DMSs by combining ab initio and tight-binding-like approaches (Larson et al. 1988).

As found for ferromagnetic p-(Cd,Mn)Te (Boukari et al. 2002) and p-(Zn,Mn)Te (Fer rand et al. 2001) as for n-(Zn,Mn)O (Androyarzyk et al. 2002), the relevant coupling between localized spins—the superexchange proceeds via p-d hybridization with bands of anions residing on the path between the TM spins in question. For this indirect interaction, to the lowest relevant order perturbation theory, the exchange energy $J_{ij}$ is proportional to $|V_{pd}|^4$ and decays fast with the distance $R_{ij}$ between magnetic ions.

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the magnitude of antiferromagnetic superexchange is larger for the nearest neighbor Mn pairs than ferromagnetic coupling at carrier densities achievable by now in these systems. This means that the magnitude of $T_C$ is weakened by non-zero values of $T_{AF}$ and $x = x_{eff}$. Furthermore, it was found by Monte Carlo simulations (Lipińska et al., 2009) that AF interactions account for a relatively fast spin dynamics in a ferromagnetic phase, provided that the holes visit only a part of the region occupied by TM spins. In such a situation, occurring for instance in (Cd,Mn)Te quantum wells, acceleration of spin dynamics and the associated decrease of coercivity are brought about by TM flip-flops at the boundary of the hole wave functions, where the molecular field produced by the hole spins vanishes.

2. (Ga,Mn)As and related compounds

In such compounds, a strong antiferromagnetic interaction between Mn ions in an interstitial and a neighboring substitutional position reduces $x_{eff}$ substantially, particularly in non-annealed samples (see, Sec. II.E). Relevant information on possible interactions between pairs of substitutional Mn spins was provided by studies of donor compensated samples, in which carrier-mediated ferromagnetic interactions are strongly reduced but superexchange is expected to be left intact. A sizable decrease of $x_{eff}$ values at low temperature under such conditions in (Ga,Mn)As (see, Sec. III.A) confirmed the presence of intrinsic antiferromagnetic coupling between Mn spins. This conclusion was substantiated by ab initio (Chang et al., 2007; Kudrnovsky et al., 2004) and tight binding (Jungwirth et al., 2005) studies of DFSs, which demonstrated that the role played by antiferromagnetic superexchange can be rather significant — the magnitude of the corresponding exchange energy was evaluated to be about 50% of the ferromagnetic contribution for the nearest neighbor Ga-substitutional Mn pairs at $x = 6\%$.

Altogether, the accumulated data indicate that in the case of uncompensated III-V DFSs with Mn$^{2+}$ ions the value of $x_{eff}$ is controlled by interstitial Mn in the whole relevant temperature and magnetic field range. Furthermore, somewhat weaker but also short ranged antiferromagnetic interactions between substitutional Mn ions reduce the magnitude of net ferromagnetic spin-spin coupling and, thus, lower $T_C$ further on.

3. Antiferromagnetic interactions in (Ga,Mn)N

Optical (Graf et al., 2003) and photoemission investigations (Hwang et al., 2005) showed that the acceptor Mn$^{3+}$/Mn$^{2+}$ level appear in the mid-gap region in GaN (see, Fig. 8). Owing to correspondingly small effective Bohr radius, no indications of hole delocalization were found up to at least $x = 0.1$. This means that holes reside in the Mn$^{3+}$/Mn$^{2+}$ impurity band, subject to Mott-Hubbard localization at weak or strong compensation, and to Anderson-Mott localization if the impurity band is partly occupied. It was found that if Mn$^{2+}$ ions prevailed (due to residual donor impurities or defects such as nitrogen vacancies (Yang et al., 2009)), antiferromagnetic interactions between Mn$^{2+}$ ions controlled magnetic properties (Granville et al., 2010; Zajac et al., 2001), leading to spin-glass freezing at 4.5 K for $x \approx 0.1$ (Dhar et al., 2003). The corresponding magnitudes of $x_{eff}$ and $T_{AF}$ (Granville et al., 2010; Zajac et al., 2001) were then similar to those of II-VI DMSs.

B. Ferromagnetic superexchange

1. Double exchange vs. superexchange

The case of chromium spinels and europium chalcogenides demonstrates that ferromagnetism is possible without band carriers. According to the time-honored nomenclature, one distinguishes two kinds of ferromagnetic coupling mechanisms operating in the absence band carriers:

**Double exchange** — This mechanism contributes when relevant magnetic ions are in two different charge states (Anderson and Hasegawa, 1955; Zener, 1951) and if the system of TM electrons is on the metallic side or in the vicinity to the Anderson-Mott transition, the case of $e_\text{g}$ manganites (Dagotto et al., 2001). In this situation, collective ferromagnetic ordering is triggered by a lowering of electron kinetic energy (increase in the width of the d band), appearing if all ions assume the same spin direction, the arrangement making quantum hopping efficient. The double exchange can be regarded as a strong coupling limit of the p-d Zener model discussed in Sec. IX.A as sufficiently large p-d hybridization leads to the appearance of an impurity band in the energy gap (see, Secs. II.A and VII.B). Within this model $T_C$ attains a maximum if the impurity band is half-filled, so that in the case relevant here — the concentrations of Mn$^{2+}$ and Mn$^{3+}$ ions are approximately equal.

**Ferromagnetic superexchange** — According to Anderson-Goedenoogh-Kanamori rules (Anderson, 1958; Goodenough, 1958; Kanamori, 1959) superexchange is ferromagnetic for certain charge states of TM ions and bond arrangements. This mechanism outperforms the double exchange if all magnetic ions are in the same charge state or if TM electrons are strongly localized, so that the impurity bandwidth is rather determined by disorder than by quantum hopping.

It was found (Blinowski et al., 1996) employing a tight binding approximation that superexchange is ferromagnetic in the case of Cr$^{2+}$ ions in a tetrahedral environment, $J_{ij} > 0$. Experimental studies of (Zn,Cr)Se (Karczewski et al., 2003) and (Zn,Cr)Te (Saito et al., 2003)
revealed indeed the presence of ferromagnetism in these systems, which however was largely determined by aggregation of Cr cations (Karczewski et al. 2003; Kuroda et al. 2007). Ferromagnetic coupling mediated by bound holes in the strongly localized regime was also predicted within a tight binding approximation for a neutral or singly ionized pair of substitutional Mn acceptors in GaAs, neglecting on-site Coulomb repulsion $U$ and intrinsic antiferromagnetic interaction between $\text{Mn}^{2+}$ spins (Strandberg et al. 2010).

2. Ferromagnetic superexchange in (Ga,Mn)N

Extensive nanocharacterization of wz-(Ga,Mn)N obtained by MOVPE (Bonanni et al. 2011; Stefanowicz et al. 2010b) and MBE (Kunert et al. 2012; Sarigiannidou et al. 2006) demonstrated that under carefully adjusted growth conditions, within an experimental margin below 10%, all Mn ions are distributed randomly over Ga-substitutional positions, and assume a 3+, $S = 2$ charge and spin state, characterized by a non-zero value of orbital momentum. Empirically, $M(T, H)$ in the paramagnetic region, $x \lesssim 1\%$, and for the magnetic field perpendicular to the wurtzite c-axis is well described by the Brillouin function for $S = 2$ with the Mn Landé factor $g \simeq 2.5$. This electronic configuration is analogous to Cr$^{2+}$ and, indeed, ferromagnetic coupling between Mn spins was found in (Ga,Mn)N (Bonanni et al. 2011). Actually, owing to the absence of competing antiferromagnetic interactions and the high magnitude of cation density $N_0 = 4.39 \times 10^{22}$ cm$^{-3}$, the largest magnitude of magnetization ever reported for any DMS was observed for Ga$_{0.995}$Mn$_{0.095}$N, $\mu_0 M \simeq 190$ mT at $\mu_0 H = 6.5$ T (Kunert et al. 2012). Importantly, ferromagnetic ordering was found at low temperatures in these samples. According to the magnetic phase diagram displayed in Fig. 37, $T_C \simeq 13$ K at $x = 0.1$ and $T_C(x) \propto x^{m}$, where $m \approx 2.2 \pm 0.2$ (Sawicki et al. 2012; Stefanowicz et al. 2013). Since such a value of $m$ was observed for spin-glass freezing in II-VI DMSs (see Sec. VIII.A.1), it was concluded that the superexchange is the dominant spin-spin coupling mechanism.

The superexchange scenario was substantiated by the evaluation of the exchange integral $J_{ij}$ as a function of the Mn-Mn distance within the aforementioned tight binding theory (Bliokh et al. 1996) and then $T_C(x)$ by Monte Carlo simulations (Sawicki et al. 2012; Stefanowicz et al. 2013). Within this approach, Mn ions were described in terms of Parmenter’s (Parmenter 1973) rotationally invariant generalization of the Anderson Hamiltonian for the relevant electronic configuration of the TM taking into account the Jahn-Teller distortion (Gosk et al. 2005; Stroppa and Kresse 2009), whereas the host band structure was modeled by the $sp^3s^* $ tight binding approximation, employing the established parametrization for GaN in the cubic approximation. Other parameters of the model were taken from infrared and visible (Graf et al. 2003b) as well as photoemission and soft x-ray absorption spectroscopy (Hwang et al. 2003) of (Ga,Mn)N.

As shown in Fig. 37, the dependence of $T_C$ on $x$ in insulating (Ga,Mn)N with Mn$^{2+}$ ions is well reproduced by the impurity band theory in question (stars). The examination of the critical behavior around $T_C$ provided information on the variance $\Delta x$ of macroscopic inhomogeneities in the Mn distribution, evaluated to be $\Delta x \approx 0.2\%$ (Stefanowicz et al. 2013).

IX. THEORY OF CARRIER MEDIATED FERROMAGNETISM

This section presents the $p$-$d$ Zener model (Dietl et al. 2000) and its limitations. This model of ferromagnetism in p-type DFSs is built exploiting information summarized in Sec. VII on the relevant electronic states and coupling between localized Mn spins and itinerant holes. The presence of competing antiferromagnetic interactions is considered making use of findings presented in Sec. VIII. The model is parametrized by a small set of independently determined material parameters, it is numerically efficient and univocal. As shown in Sec. X the $p$-$d$ Zener model explains qualitatively, and often quantitatively, a palette of comprehensive results concerning ferromagnetic characteristics and their control in films, heterostructures, and nanostructures of (Ga,Mn)As, p-
which describe, for a given magnetization profile \(\vec{M}(\vec{r})\), the free energy densities of the Mn spins in the absence of any carriers and of the carriers in the presence of the Mn spins, respectively. A visible asymmetry in the treatment of the carriers and of the spins corresponds to an adiabatic approximation: the dynamics of the spins in the absence of the carriers is assumed to be much slower than that of the carriers. Furthermore, in the spirit of the virtual-crystal and molecular-field approximations, the classical continuous field \(\vec{M}(\vec{r})\) controls the effect of the spins upon the carriers. Now, the thermodynamics of the system is described by the partition function \(Z\), which can be obtained by a functional integration of the Boltzmann factor over all magnetization profiles \(\vec{M}(\vec{r})\),

\[
Z \sim \int \mathcal{D}\vec{M}(\vec{r}) \exp\{-\int d\vec{r}\mathcal{F}[\vec{M}(\vec{r})]/k_B T\}, \tag{10}
\]

the approach developed in the context of DMSs for bound magnetic polarons (Dietl et al. 1993, Dietl and Spalek 1983), and directly applicable for spin physics in quantum dots as well.

In the mean-field approximation, which should be valid for spatially extended systems and long-range spin-spin interactions, a term corresponding to the minimum of \(\mathcal{F}[\vec{M}(\vec{r})]\) is assumed to determine \(Z\) with a sufficient accuracy, the conclusion supported by Monte Carlo simulations discussed in Sec. [X.T].

If the energetics is dominated by spatially uniform magnetization \(\vec{M}\), the spin part of the free energy density in the magnetic field \(\vec{H}\) can be written in the form

\[
\mathcal{F}_S[\vec{M}] = \int_0^{\vec{M}} d\vec{M}_o \cdot \vec{h}(\vec{M}_o) - \vec{M} \cdot \vec{H}. \tag{11}
\]

Here, \(\vec{h}(\vec{M}_o)\) denotes the inverse function to \(\vec{M}_o(\vec{h})\), where \(\vec{M}_o\) is the available experimentally macroscopic magnetization of the spins in the absence of carriers in the field \(\vec{h}\) and temperature \(T\), whose anisotropy is typically weak for Mn\(^{2+}\) ions in the orbital singlet state. As discussed in Sec. [VIII.A] it is usually possible to parametrize \(M_o(h)\) by the Brillouin function \(B_S(T, H)\) that takes the presence of intrinsic short-range antiferromagnetic interactions into account. Near \(T_C\) and for \(H = 0\), \(M\) is sufficiently small to take \(M_o(T, h) = \chi(T) h\), where \(\chi(T)\) is the magnetic susceptibility of localized spins in the absence of carriers. Under these conditions,

\[
\mathcal{F}_S[M] = M^2/2\chi(T), \tag{12}
\]

which shows that the increase of \(\mathcal{F}_S\) with \(M\) slows down with lowering temperature, where \(\chi(T)\) grows. Turning to \(\mathcal{F}_C[M]\) we note that owing to the giant Zeeman splitting of the bands proportional to \(M\), the energy of the carriers, and thus \(\mathcal{F}_c[M]\), decreases with \(|M|\), \(\mathcal{F}_c[M] - \mathcal{F}_c[0] \sim -M^2\). Accordingly, a minimum of \(\mathcal{F}[M]\) at non-zero \(M\) can develop in \(H = 0\) at sufficiently low temperatures signaling the appearance of a ferromagnetic order.

It had been postulated (Dietl et al. 2000), and checked employing 40 orbitals' tight binding approximation (TBA) (Werpachowska and Dietl 2010a), that the minimal Hamiltonian necessary to properly describe effects of the complex structure of the valence band in tetrahedrally coordinated semiconductors upon \(\mathcal{F}_c[M]\) is...
the Luttinger six bands’ $kp$ model with the Bir-Pikus strain terms, supplemented by the $p-d$ exchange contribution taken in the virtual-crystal and molecular-field approximations,

$$\mathcal{H}_{pd} = \beta \mathbf{k} \cdot \mathbf{\tilde{M}} / g_\mu_B.$$ (13)

This term leads to spin splittings of the valence subbands, whose magnitudes—owing to the crucial role of the spin-orbit coupling—depend on the magnitude and direction of the hole wave vectors $\tilde{k}$ in a complex way even for spatially uniform magnetization $\tilde{M}$. Within this formalism, the spin-orbit interaction results from the $p$-type symmetry of the periodic parts of the Bloch wave functions and the corresponding spin-orbit splitting of the valence band at the $\Gamma$ point of the Brillouin zone into $J = 3/2$ and $J = 1/2$ hole subbands, the $J = 3/2$ subband exhibiting an additional splitting in the presence of confinement and/or strain. This effect is distinct from the $\tilde{k}$-dependent Dresselhaus or Rashba spin-splitting appearing in the conduction band in the absence of inversion symmetry or if the cubic symmetry is perturbed, respectively. The incorporation of the spin-orbit interaction into the valence band model is essential (Dietl et al., 2000), as it controls the magnitude of $T_C$ and accounts for magnetic anisotropy in DFSs with Mn in the high spin $2+\text{charge state for which single-ion magnetic anisotropy}$ is small according to magnetic resonance studies (Fedor-rych et al., 2002; Qazzaz et al., 1995).

It would be technically difficult to incorporate such effects to the RKKY model, as the spin-orbit coupling leads to non-scalar terms in the spin-spin Hamiltonian. At the same time, the indirect exchange associated with the virtual spin excitations between the valence subbands, the Bloembergen-Rowland mechanism (Dietl, 1994), is automatically included. The model allows for strain, confinement, and was developed for both zinc-blende and wurtzite materials (Dietl et al., 2001b). Furthermore, the direct influence of the magnetic field upon the hole spectrum was taken into account (Dietl et al., 2001b; Jungwirth et al., 2006b; Sliwa and Dietl, 2006). The aforementioned Stoner enhancement was described by introducing a Fermi-liquid-like parameter $A_F$ (Dietl et al., 1997; Haury et al., 1997; Jungwirth et al., 1999), which enlarges the Pauli susceptibility of the hole liquid, typically by 20% in the 3D case. No disorder effects were taken into account on the ground that their influence on thermodynamic properties is relatively weak except for the strongly localized regime. Obviously, a more elaborated parametrization of the valence band is necessary in many cases. For instance, the eight bands’ model was employed to compute the infrared (Hankiewicz et al., 2004) and Hall conductivity (Werbachowska and Dietl, 2010b) in (Ga,Mn)As, whereas the multi-orbitals’ tight-binding approaches served to describe $T_C$ (Jungwirth et al., 2005; Vargaftman and Meyers, 2001; Werbachowska and Dietl, 2010b) and interlayer coupling (Sankowski and Kacman, 2005) in GaAs/(Ga,Mn)As superlattices.

Having the hole energies, the free energy density $\mathcal{F}_c(\tilde{M})$ was evaluated according to the procedure suitable for Fermi liquids of arbitrary degeneracy, i.e., taking the carrier entropy into account. By minimizing $\mathcal{F}[\tilde{M}] = \mathcal{F}_S[\tilde{M}] + \mathcal{F}_c[\tilde{M}]$ with respect to $\tilde{M}$ at a given $T$, $\tilde{H}$, and hole concentration $p$, Mn spin magnetization $M(T,H)$ was obtained as a solution of the mean-field equation,

$$\tilde{M}(T, H) = x_{eff} N_0 g_\mu_B \beta M_S[0] \left[ \frac{g_\mu_B (-\partial \mathcal{F}_c[\tilde{M}] / \partial \tilde{M} + \tilde{H})}{k_B (T + T_{AF})} \right],$$ (14)

where the peculiarities of the valence band structure, such as the presence of various hole subbands, spin-orbit coupling, crystalline cubic and strain-induced anisotropies are hidden in $\mathcal{F}_c[\tilde{M}]$.

2. Theory of the Curie temperature

Near the Curie temperature $T_C$ and at $H = 0$, where $M$ is small and the free energy is an even function of $M$, one expects $\mathcal{F}_c(M) - \mathcal{F}_c(0) \approx -M^2$. It is convenient to parameterize this dependence by a generalized carrier spin susceptibility $\tilde{\chi}_c$, which is related to the magnetic susceptibility of the carrier liquid according to $\chi_c = A_F (g_\mu_B)^2 \tilde{\chi}_c$. In terms of $\tilde{\chi}_c$,

$$\mathcal{F}_c(M) = \mathcal{F}_c[0] - A_F \tilde{\chi}_c M^2 / (2 g_\mu_B)^2.$$ (15)

By expanding $B_S(M)$ for small $M$ and introducing the spin susceptibility of the magnetic ions in the absence of carriers, $\tilde{\chi}_S = \chi / (g_\mu_B)^2$, one arrives at the mean-field formula making it possible to determine $T_C$,

$$A_F \beta^2 \tilde{\chi}_S(T_C, q) \tilde{\chi}_c(T_C, q) = 1,$$ (16)

where $\beta$ should be replaced by the $s-d$ exchange integral $\alpha$ in the case of electrons and $\tilde{q}$ denotes the Fourier component of the magnetization texture, for which $T_C$ attains the highest value. For the spatially uniform magnetization $q = 0$, in terms of $x_{eff}$ and $T_{AF}$,

$$T_C = T_F - T_{AF},$$ (17)

where $T_F$ is given by

$$T_F = x_{eff} N_0 S(S+1) A_F \tilde{\chi}_c(T_C) \beta^2 / 3k_B.$$ (18)

with the cation concentration $N_0 = 4/a_0^3$, $4/(\sqrt{3}a_0^3)$, and $8/a_0^3$ for the zinc-blende, wurtzite, and elemental diamond-structure DFSs, respectively. As discussed in Sec. V11E for holes in tetrahedrally bound semiconductors the exchange integral $\beta = -54$ meV nm$^3$, except perhaps for mercury chalcogenides, in which the value of $\beta$ appears somewhat smaller (Furdyna and Kossut, 1988). For other lattice structures, different combinations of hybridization matrix elements describe an appropriate exchange integral characterizing coupling between carriers and localized spins (Dietl et al., 1994).
In the 3D case, typically, \( A_F \approx 1.2 \). For a strongly degenerate carrier liquid \( |\epsilon|/k_B T > 1 \), \( \chi_c = \rho_s/4 \), where \( \rho_s \) is the total DOS for intra-band spin excitations at the Fermi level, typically reduced by spin-orbit interactions from DOS for charge excitations \( \nu_e \). An analytic form of \( \rho_s \) was derived for the four band Luttinger model \cite{Ferrand2001}. In the absence of spin-orbit interactions and in the 3D case it is given by \( \rho_s = \nu_e = m^*_D \kappa_F/\pi^2\hbar^2 \). In this case and for \( A_F = 1 \), \( T_F \) assumes the well-known form, obtained already in the 1940s in the context of carrier-mediated nuclear ferromagnetism \cite{Frolich1940} and in the 1970s in the context of DMSs \cite{Pashitskii1979}. In general, however, \( \chi_c \) has to be determined numerically by computing \( F_c(M) \) for a given band structure and degeneracy of the carrier liquid.

The above model predicts \( T_F \) to be much higher for holes than for electrons for two reasons: (i) the density of states \( \rho_s \) is typically lower in the conduction band (though effects of spin-orbit interactions are weaker); (ii) the \( s-d \) exchange integral \( \alpha \) is typically over 4 times smaller than the \( p-d \) integral \( \beta \). Section 4 presents a comparison of these predictions to experimental data.

As described earlier, \( T_F \) can be computed by minimizing the free energy, and without referring to the explicit form of the Kohn-Luttinger amplitudes \( u_{i,k} \). Since near \( T_C \) the relevant magnetization \( M \) is small, \( \chi_c \) can also be determined from the linear response theory. The corresponding \( \rho_s \) assumes the form \cite{Dietl2001b},

\[
\rho_s = \lim_{q \to 0} \sum_{i,k} \frac{|(u_{i,k}|s_M|u_{i,k+q})|^2 f_i(k)[1 - f_j(k + q)]}{E_j(k + q) - E_i(k)},
\]

where \( s_M \) is the component of spin operator along the direction of magnetization and \( f_i(k) \) is the Fermi-Dirac distribution function for the \( i \)-th valence band subband. A quantitative analysis demonstrated that typically a 30% contribution to \( T_C \) originates from interband polarization (the Bloembergen-Rowland mechanism) involving light and heavy hole subbands \cite{Dietl2001b}. This formalism was extended to \textit{bulk} states in topological insulators, in which \( u_{i,k} \) for both the valence and conduction bands have a \( p \)-like symmetry, so that appreciable \( T_F \) can be expected from interband polarization even if the Fermi energy resides in the band gap \cite{Yu2010}.

### B. Theory of carrier-controlled Curie temperature in reduced dimensionality and topological insulator systems

In thin films, heterostructures, and superlattices, owing to the formation of interfacial space charge layers, the hole density and corresponding Curie temperatures \( T_C(\nu_e) \) are non-uniform even for a uniform distribution of acceptors and donors. The role of non-uniformity in the carrier distribution grows on reducing the thickness \( t \) of magnetic layers, and is particularly relevant in those structures in which \( \nu_e(p) \) can be tuned electrostatically, for instance, by the gate voltage. When \( t \) is larger than the phase coherence length \( L_\phi \), the region with the highest \( T_C \) value determines \( T_C \) of the whole structure. If, however, \( t < L_\phi \) the value of local magnetization \( M(z) \) and \( T_C \) are determined by the distribution \( \nu_e(z) \) across the \textit{whole} channel thickness. In this regime two situations were considered:

If disorder is strong, \( \ell < t \), the scattering broadening makes dimensional quantization irrelevant although quantum mechanical non-locality remains important. Under these conditions the magnitude of layer’s \( T_F \) can be expressed as \cite{Nishitani2010, Sawicki2010},

\[
T_F = \int dz \nu_e[p(z)] \int dz \nu_e[z^2/\int dz \nu_e(z)^2],
\]

where \( T_F(p) \) is to be determined from the relevant 3D model and \( \nu_e(z) \) is to be evaluated from the Poisson equation taking into account the pinning of the Fermi energy by surface states. It was predicted within the \( p-d \) Zener model that the energy position \( E_z \) of surface states should strongly affect the efficiency of \( T_C \) tuning by the gate voltage \( V_G \) (see, Ohno 2013 and Sec. X).

The opposite limit of a weak disorder \( \ell > t \), relevant to modulation-doped \textit{II-VI} heterostructures, was also considered \cite{Dietl1997, Haury1997}. Owing to a typically large confinement-induced splitting between heavy and light hole subbands, only one ground state heavy hole subband is occupied, for which the \( p-d \) exchange is of the Ising type, \( \mathcal{H}_{pd} = -N_0 \beta s_z S_z \), so that

\[
T_F = N_0 x_{eff} S(S + 1) A_F \beta^2 m^*/(12\pi^2\hbar^2 k_B L_W),
\]

Here \( m^* \) denotes the in-plane effective mass and \( L_W \) is an effective width of the region occupied by carriers relevant to ferromagnetism given by \cite{Dietl1999, Haury1997}.

\[
L_W = 1/\int dz |\phi(z)|^4.
\]

where \( \phi(z) \) is an envelope function of the relevant 2D subband. As seen, owing to a step-like form of DOS in the 2D case, \( T_F \) does not depend on the hole density in this case. The expression for \( T_F \) was generalized further on to the case of arbitrary degeneracy of the hole liquid and by including effects of disorder \textit{via} scattering broadening of DOS \cite{Boukari2002}.

The case of high carrier concentrations leading to the occupation of several hole subbands in (Ga,Mn)As based multilayer structures was also considered by incorporating an LSDA approach to two \cite{Giddings2008, Jungwirth1999} and four \cite{Fernandez-Rossier2002} band \( kp \) models, whereas the multi-orbitals’ tight-binding approaches served to describe \( T_C \) \cite{Vurgaftman2001, Werbach2001, Song2005} in GaAs/(Ga,Mn)As superlattices.
Theoretical approaches were developed allowing to evaluate Curie temperature for ferromagnetic ordering of magnetic impurities mediated by Dirac carriers at the surface of 3D topological insulators (Abanin and Pesin, 2011; Liu et al., 2009). An Ising type of exchange was assumed, \( H_{ex} = -N_0 J_x \delta_x S_x \), leading to a gapped dispersion given by,
\[
e(\vec{k}) = \pm [(J_z M/2g_\mu_B)^2 + (\hbar v_F k)^2]^{1/2}, \tag{23}
\]
where \( v_F \) is the Fermi velocity. For such a case (cf. Liu et al., 2009), in our notation,
\[
T_F = N_0 x_{eff} S(S + 1) A_F r J_x^2 (E_c - |\epsilon_F|)/(24\pi^2 k_B^2 v_F^2 L_W), \tag{24}
\]
where \( N_0 = 12/(\sqrt{3}a^2c) \) and \( 4/a_0^3 \) in hexagonal III-V (e.g., (B_1-x,Mn_x)Se) and cubic II-VI or IV-VI (e.g., Sn_1-x,Mn_x)Te) compounds, respectively; \( r \) is the number of Dirac cones at a given surface; \( E_c \) is a cut-off energy associated with the termination of the Dirac surface band, and \( L_W \) is the penetration depth of Dirac carriers related to their envelop function according to Eq. 22.

A formalism suitable to evaluate \( T_F \) determined by bulk states of topological insulators is presented at the end of Sec. IX.A.2.

The formation of spin-density waves is expected in the case of carrier-mediated ferromagnetism in 1D systems (Dietl et al., 1999).

C. Theory of magnetization and hole polarization

The mean-field Eq. 14 allowed to determine Mn magnetization \( M(T,H) \), particularly \( M(T) \) at \( T \leq T_C \) (Dietl et al., 2001b) and \( M(H) \) at \( T_C \) (Sliwa and Dietl, 2011). The same formalism also provided quantitative information on the value of thermodynamic hole spin polarization (Dietl et al., 2001b),
\[
P = \frac{2g_\mu_B}{\beta r} \frac{\partial F_c(M)}{\partial M}, \tag{25}
\]
which, despite the spin-orbit interaction, can approach 90% in the relevant range of hole and Mn densities in (Ga,Mn)As but gets reduced down to about 50% at high hole densities (Dietl et al., 2001b).

Furthermore, hole magnetization \( M_e \), which determines the magnitude of spontaneous magnetization \( M_e(T) = M(T) + M_v(M) \) was evaluated taking into account the effect of a magnetic field on the valence band (Dietl et al., 2001b; Sliwa and Dietl, 2006, 2013). It was found that holes reduce (Ga,Mn)As magnetization by about 10%, so that the value of the magnetic moment per one Mn ion can be taken as \( \mu \approx 4.5 \mu_B \) in ferromagnetic samples of (Ga,Mn)As weakly compensated by donors. The hole contribution is, therefore, about 2 times smaller than would be in the absence of spin-orbit coupling and for fully spin-polarized hole gas.

Theoretical studies of magnetic stiffness discussed in Sec. IX.E made it possible to evaluate a reduction of \( M(T) \) by spin wave excitations (König et al., 2001; Werbachowska and Dietl, 2010a).

D. Theory of magnetic anisotropy and magneto-elasticity

Since Mn^{2+} ions are in the orbital singlet \( ^6A_1 \) state in DFGSs, a single-ion magnetic anisotropy is small (Edmonds et al., 2006; Fedorych et al., 2002), so that the dominant contribution comes from spin-orbit effects within hole band states (Dietl et al., 1997, 2000). Owing to an interplay of the spin-orbit interaction with the crystal structure anisotropy, strain, and confinement the characteristic crystalline magnetic anisotropy fields \( H_o \) are typically larger than the shape term \( \mu_o H_A = \mu_o M_o \approx 0.13 T \) for a (Ga,Mn)As thin film with \( x_{eff} = 10\% \) (Dietl et al., 2001b). Similarly, in the case of (Ga,Mn)As nanobars, crystalline magnetic anisotropy determined by strain distribution specific to free standing strained nanostructures dominates over the shape term (Hümpfner et al., 2007).

Accordingly, the theoretically expected character and magnitude of crystalline magnetic anisotropy were obtained by considering how the carrier free energy density \( F_c(\vec{M}) \) depends on the direction of the magnetization vector \( \vec{M} \) with respect to crystallographic axes at various values of epitaxial strain (Dietl et al., 2000). Following subsequent detail studies for (Ga,Mn)As epilayers (Abolfath et al., 2001; Dietl et al., 2001b), further theoretical analysis of anisotropy energy coefficients \( K_i \) and anisotropy fields \( H_i \) were carried out for the canonical (001) films (Zemen et al., 2009) as well as, additionally, for an arbitrary (11n) substrate orientation (Stefanowicz et al., 2010a), the accomplishments discussed vis à vis experimental findings in Sec. X.D. The formalism was developed for an arbitrary form of the strain tensor \( \epsilon \) and it is valid as long as non-linear strain effects are not significant. It was checked that terms linear in products of \( k_i \) and \( \epsilon_{ij} \) can be neglected (Stefanowicz et al., 2010a).

A sizable strength of crystalline magnetic anisotropy and related magneto-elastic phenomena, comparable to ferromagnetic metals despite much smaller magnetic ion concentrations, comes from a large spin-orbit splitting of the valence band (about 0.3 eV for arsenides and 1 eV for tellurides), greater than the kinetic energy of holes (Dietl et al., 2001b).

E. Theory of micromagnetic parameters and spin wave dispersion

Similarly to other ferromagnets, a description of magnetization processes for various orientations of the external magnetic field \( \vec{H} \) as well as the understanding of the domain structure require information not only on the magnetic anisotropy but also on the exchange stiffness. These two micromagnetic characteristics correspond to energy penalties associated with (i) deviation of magne-
tization orientation from an easy direction, as described above and (ii) local twisting of magnetization from its global direction, respectively.

The exchange stiffness $A$ and the related spin wave dispersion $\omega(q) = Dq^2$, where $D = 2g\mu_B A/M$, were theoretically determined for DFSs by examining the $q$ dependent part of the hole spin susceptibility $\xi_C(q)$ at a given average Mn magnetization $M$. [Brey and Gómez-Santos 2003; Köning et al. 2001; Werbachowska and Dietl 2010a]. In general, taking the presence of a spin-orbit interaction into account, $D$ is a tensor and, moreover, terms linear in $q$ can appear. Their magnitude and possible effects were analyzed within a multi-orbital tight binding model for thin films of (Ga,Mn)As (Werbachowska and Dietl 2010a). A magnetic cycloid ground state was predicted for a few monolayer thick (Ga,Mn)As films.

These works made it possible to evaluate the width of the Bloch domain wall,

$$\delta_w = \pi (A/K)^{1/2}, \quad (26)$$

which is the shortest length scale of the micromagnetic theory. It was found (Dietl et al. 2001a) that over the relevant range of material parameters $\delta_w \gtrsim 15$ nm stays much longer than a mean distance between holes and Mn ions in ferromagnetic (Ga,Mn)As. This evaluation substantiated the validity of the continuous medium approximation, employed in the approach exposed in this chapter. Moreover, it pointed out that the time honored micromagnetic theory, presented for (Ga,Mn)As-type ferromagnets in Appendix, and corresponding software packages, are also applicable to these systems.

The Gilbert damping parameter $\alpha_G$ due to particle-hole excitations in the (Ga,Mn)As valence band was evaluated first neglecting (Sinova et al. 2004; Tserkovnyak et al. 2004), and then taking into account quantitatively important vertex corrections within the four band model (Garate and MacDonald 2009). A monotonic decrease of $\alpha_G$ with the hole scattering rate was found. Within a similar model, a magnitude of non-adiabatic spin torque $\beta_w$ was evaluated and found to be of the order of one (Hals et al. 2009). It would be interesting to find out how a finite value of spin-orbit splitting between $\Gamma_8$ and $\Gamma_7$ bands as well as localization and correlation effects will affect these conclusions.

A verification of the present theory by examining the temperature dependence of magnetization and specific heat is presented in Sec. X.C whereas Sec. X.E contains a comparison of experimental results to theoretical predictions on the domain structure and spin wave excitations. Section II.E contains information on experimental determination of $\beta_w$.

F. Limitations of the mean-field $p$-$d$ Zener model

Material parameters – The model is parametrized by the lattice constant $a_0$; spin-orbit splitting $\Delta_0$; Luttinger parameters $(\gamma_1, \gamma_2, \text{and} \gamma_3$ in the zinc-blende case when the six band Luttinger Hamiltonian is employed); exchange integral $\beta$; Landau’s Fermi liquid parameter $A_F$, and–for non-zero strain–by elastic moduli $c_{ij}$ and two deformation potentials of the valence band, $b$ and $d$. Two of these parameters, $\beta$ and $A_F$, are known by now with an accuracy not better than 10%, which leads to the accumulated error in calculated $T_C$ values of the order of 25%. Additionally, a quantitative verification of any DFS theory is challenging because of difficulties in assessing real hole and Mn concentrations that, moreover, are often non-uniformly distributed over the film volume, as discussed in Sec. I.A.

Thermodynamic magnetization fluctuations – The question how various corrections to the mean-field and continuous medium approximation affect theoretical values of $T_C$ was addressed in some details [Brey and Gómez-Santos 2003; Jungwirth et al. 2002; 2005; Popescu et al. 2006; Timm and MacDonald 2005; Yildirim et al. 2007]. It was found that the mean-field $p$-$d$ Zener model remains quantitatively valid for (Ga,Mn)As and related systems, typical lowering of $T_C$ values by magnetization fluctuations being below 20%, though a value of 30% was found in the most recent study (Yildirim et al. 2007), if a correction for the classical spin approximation adopted in that work is taken into account. According to Monte Carlo simulations for the 2D case, the fluctuations of magnetization diminish $T_C$ by a factor of 2 in the absence of competing antiferromagnetic interactions, whereas in the presence of these interactions, a net quantitative correction to the mean-field approximation is much reduced (Lipińska et al. 2009).

Antiferromagnetic interactions – According to results presented in Sec. VIII.A carrier-mediated interactions compete with short-range superexchange coupling between Mn ions in cation-substitutional and/or interstitial positions. As discussed above, the presence of these antiferromagnetic interactions can be incorporated into the $p$-$d$ Zener model by introducing two parameters, $x_{\text{eff}} < x$ and $T_{\text{AF}} > 0$. Additionally, the short-range antiferromagnetic interaction enhances the importance of the antiferromagnetic portion of the RKKY coupling leading, for hole densities comparable to the concentration of localized spins, to a further reduction of $T_C$ values comparing to those expected from Eq. [17] (Ferrand et al. 2001). Actually, in this limit, $p \gtrsim N_0x_r$, randomness of the interaction type (ferro vs. antiferro) associated with RKKY oscillations can drive the system towards a spin-glass phase rather than towards a ferromagnetic ground state expected within the mean-field approximation (Eggenkamp et al. 1995). The stability of the ferromagnetic phase is, however, much enhanced in III-V and II-VI DFSs by multiband structure and strong anisotropy of the valence band (Timm and MacDonald 2005).
Kondo effect – The theory is developed for Mn concentrations high enough that magnetic ordering temperature is higher than the Kondo temperature, evaluated for II-VI p-type DMSs to be of the order of 1 K [Dietl et al., 1997].

Effects of disorder and localization – The understanding of the interplay between carrier-mediated ferromagnetism and carrier localization is an emerging field of research [Dietl, 2008b; Richardella et al., 2010; Sawicki et al., 2010; Sheu et al., 2007]. The relevant questions here are how the presence of spins affect carrier localization and how carrier-mediated ferromagnetism is influenced by localization. According to experimental investigations of (Ga,Mn)As [Matsukura et al., 1998] and (Zn,Mn)Te [Ferrand et al., 2001], the magnitude of $T_C$ is similar to other thermodynamic properties, showing no critical behavior at the MIT. A non-critical behavior of $T_C$ across the MIT stems from the scaling theory of the Anderson-Mott transition. This theory implies that an average hole localization length, which diverges at the MIT, remains much greater than the mean distance between acceptors for the experimentally important range of hole densities. Thus, holes can be regarded as band-like at the length scale relevant to coupling between magnetic ions. Hence, the spin-spin exchange interactions are effectively mediated by the itinerant carriers, so that the $p$-$d$ Zener model can serve to evaluate $T_C$, also on the insulator side of the MIT as long as holes remain only weakly localized. This view was supported by results of inelastic neutron scattering of nearest neighbor Mn pairs in p-(Zn,Mn)Te [Kepa et al., 2003]. In this experiment, the hole-induced change in the pair interaction energy shows the value expected for the band carriers despite that the studied sample was on the insulator side of the MIT.

As already mentioned, disorder introduces a certain lifetime broadening of DOS, the effect equivalent to the lowering of $T_C$ by a finite mean free path within the RKKY theory and being captured within, e. g., the coherent potential approximation [Jungwirth et al., 2005]. The broadening can also be phenomenologically introduced to the $p$-$d$ Zener model [Boukari et al., 2002; Dietl et al., 1997], typically diminishing the magnitude of DOS at $\epsilon_F$ and, thus of $T_C$. It was suggested [Dietl et al., 2001b] that this effect would destroy theoretically predicted oscillations in the magnitude of the cubic anisotropy field as a function of the Fermi level (hole concentration), as their period is smaller than the expected broadening energy of relevant $k$ states. It is important to note that in contrast to the one-particle DOS which determines, for instance, photoemission spectra and tunneling current [Altshuler and Aronov, 1985; Pappert et al., 2006; Richardella et al., 2010], the DOS for intra-band excitations, relevant to $T_C$ does not exhibit any interaction and disorder-induced Coulomb anomaly at the Fermi energy as well as does not show a critical behavior across the MIT.

Importantly, not only ferromagnetic correlations but also disorder effects, particularly, carrier localization depends on the strength of the $p$-$d$ interaction in DFSs. This dual effect of the $p$-$d$ coupling is sketched in Fig. 38 (Dietl, 2008a). According to results presented in Secs. VII and VII.E on going from the weak to the strong coupling regime, i. e., to materials with a short bond length (nitrides, oxides), the magnitude of $p$-$d$ hybridization and, hence, the TM binding energy $E_i$ get progressively enhanced, which shifts the critical hole density $p_c$ for the MIT towards correspondingly higher values, narrowing the hole concentration range where the carrier-mediated ferromagnetism can appear [Dietl, 2008a]. At the same time, however, according to Eq. 18, the magnitude of the characteristic ferromagnetic temperature $T_F$ increases with $N_0$, that is when the cation-anion distance diminishes [Dietl et al., 2000]. It is still an open question whether the MIT and, thus, the region of high $T_C$ values can be experimentally achieved in nitrides and oxides.

Another consequence of carrier localization is the presence of static nano-scale fluctuations in the local DOS, discussed in Sec. II.F. These fluctuations, typically accompanied by competing ferromagnetic and antiferromagnetic interactions, lead to a static phase separation into regions differing in the magnitude of hole density and, therefore, in the strength of ferromagnetic correlations [Dietl, 2007; Dietl et al., 2000; Sawicki et al., 2010]. Such an electronic phase separation leads to the appearance of randomly oriented ferromagnetic bubbles that start to develop at $T^* > T_C$ [Geresdi et al., 2008; Matthias et al., 2002] and tend to order at $T < T_C$ [Sawicki et al., 2010]. Within this picture, the localization-induced disappearance of carrier-mediated ferromagnetism proceeds via a growing participation of superparamagnetic regions, leading to melting away of the percolating ferromagnetic cluster. Eventually, when

![Fig. 38 (Color online) Schematic dependence of $T_C$ on the magnetic ion concentration and density of hole states at the Fermi level for a weak and a strong $p$-$d$ hybridization. Higher values of $T_C$ are predicted within molecular field and virtual crystal approximations (VCA) for the strong coupling. However, the region where the holes are not localized and mediate the spin-spin interaction is wider in the weak coupling case. From Dietl, 2008a](image-url)
antiferromagnetic interactions will start to dominate, a spin-glass phase will set in at low temperatures (see, Sec. VIII.A). While excellent micromagnetic properties are expected deeply in the metallic regime, where the p-d Zener model should be quantitatively correct, the diminished volume of ferromagnetic regions at lower hole densities makes the model only qualitatively valid. Alternatively, in weakly compensated DFS samples, a ferromagnetic superexchange or double exchange are expected to appear in the strongly localized regime (see, Sec. VIII.B).

X. COMPARISON TO EXPERIMENTAL RESULTS

In this section a detailed comparison of experimental and theoretical results is presented for III-V and II-VI DFSs, for which relevant material parameters have been already determined. It is expected that further works on other compounds will allow for a quantitative description of magnetism also in those systems.

A. Curie temperature

1. Chemical trends in III-V DFSs

In Fig. X.A.1 the highest values of $T_C$ found to date in p-type Mn-based III-V DMSs are reported (Abe et al., 2000; Chen et al., 2009; Olejník et al., 2008; Scarpulla et al., 2005; Schallenberg and Munekata, 2006; Wang et al., 2008b; Wojtowicz et al., 2003a), and compared to the early predictions of the p-d Zener model (Dietl et al., 2000; Jungwirth et al., 2002) for fixed values of the Mn and hole concentrations. We see that the theory reproduces the chemical trends and describes semi-quantitatively the absolute values of $T_C$. The observed trend reflects a decrease of the p-d exchange energy $N_0\beta$ for larger cation-anion distances as well as an enhanced role of the competing spin-orbit interaction in materials with heavier anions. However, a comparison of (In,Mn)As and (Ga,Sb)Mn or (Ga,Mn))As and (Ga,Mn)P in Fig. X.A.1 indicates that the values of hole effective masses in particular compounds are relevant, too.

2. Curie temperatures in (Ga,Mn)As and related systems

Figure 39 presents the experimentally established values of $T_C$ in a representative series of annealed (Ga,Mn)As thin films as a function of saturation magnetization $M_{Sat}$ determined at low temperatures, compared to the expectation of the mean-field p-d Zener model. In order to generate the theoretical curve (solid line), the calculation scheme and the set of standard material parameters proposed previously (Dietl et al., 2001b, 2000) are employed. It is assumed that hole density and the effective Mn concentration are equal and related to $M_{Sat}$ according to $p = N_0 x_{eff} = M_{Sat}/\mu$, where $\mu = 4.5\mu_{textB}$ takes into account a contribution of holes to the total magnetization. A similar comparison is shown in Fig. 41 where the theoretical curve was obtained by tight-binding theory within coherent potential and mean-field approximations, adjusting parameters to reproduce the empirical band structure of GaAs and spin-splitting of (Ga,Mn)As (Jungwirth et al., 2005).

The comparison of experimental and theoretical results shown in Fig. 40 allows to draw several important conclusions concerning limitations of the p-d Zener model, listed in Sec. IX.F. In particular, higher experimental than theoretical $T_C$ values at low $M_{Sat}$ stem, presum-
from nano-scale magnetization fluctuations at the localization boundary making that a portion of Mn spins does not participate in the ferromagnetic order. Under these conditions, an experimentally determined average value of $M_{\text{Sat}}$ is smaller than relevant $M_{\text{Sat}}$ corresponding to the ferromagnetic percolation cluster setting up at $T_C$. In the high $M_{\text{Sat}}$ range, in turn, the magnitude of $T_C$ saturates faster with $M_{\text{Sat}}$ than expected theoretically. In addition to a correction for the effect of thermodynamic magnetization fluctuations, two other effects appear to come into play in this regime.

First, a relative importance of short range antiferromagnetic interactions between Ga-substitutional Mn ions increases with the Mn concentration. As discussed in Sec. VIII.A, these interactions do not affect $x_{\text{eff}}$ in (Ga,Mn)As but make $T_{AF} > 0$. Since a dependence $T_{AF}(x_{\text{eff}}, T)$ is unknown for (Ga,Mn)As, guided by results for II-VI DMSs and (Ga,Mn)N (Sec. VIII), one can assume $T_{AF} = (M_{\text{Sat}}/A)^m$, where $A$ is a fitting parameter and $m = 2.3$. The dashed line in Fig. 40 has been obtained with $\mu_0 A = 10.6$ mT.

Second, it is rather probable that Mn interstitials are not entirely removed by annealing in this range of Mn content (see, Secs. II.E and II.A). If this is the case, the hole concentration is diminished according to Eqs. 1 and

$$p = N_0 x_{\text{eff}}$$

In particular, for $M_{\text{Sat}} = 71$ mT, which corresponds to $x_{\text{eff}} = 6.1\%$, the $p$-$d$ Zener model with antiferromagnetic interactions reproduces $T_C = 170$ K for $x_1 = 1.8\%$ (if $N_D = 0$), meaning that $x = 9.7\%$. A systematically observed saturation in values $x_{\text{eff}}$ and $T_C$ for $x > 10\%$ (Chiba et al. 2007, Mack et al. 2008, Ohya et al. 2007b) suggests that substitutional incorporation of Mn is particularly difficult in such heavily Mn doped (Ga,Mn)As samples.

Finally, it should be recalled referring to Fig. 11 that the agreement between the experimental and theoretical values could presumably be improved further on by noting that the hole concentration $p$ is underestimated and overestimated by the Hall effect measurements in the range of low and high concentrations of Mn acceptors, respectively, as discussed in Sec. II.G.

In view of the above discussion, particularly welcome are studies of $T_C$ as a function of hole density in a single sample, as such a dependence is virtually independent of poorly known values of $T_{AF}$ and background concentrations of compensating donors, $N_0 x_1$ and $N_D$. According to numerical results for the $p$-$d$ Zener model (Dietl et al. 2001b),

$$\gamma = d \ln T_C / d \ln p = 0.6 - 0.8$$

in the relevant region of hole densities. This prediction was confirmed experimentally by tracing the dependence $T_C(p)$ in (Ga,Mn)As films irradiated by ions that produce hole compensating donor defects, the original data for (Ga,Mn)As shown already in Fig. 13. As depicted in Fig. 42, the $T_C(p)$ results point to $\gamma = 0.5 - 1.0$, in agreement with the theory.

However, detailed studies of changes in $T_C$ induced by the gate voltage $V_g$ in metal-insulator-semiconductor (MIS) structures of (Ga,Mn)As (Nishitani et al. 2010, Sawicki et al. 2010) led to an entirely different value,

$$\gamma = d \ln T_C / d \ln (V_g) = 0.19 \pm 0.02$$

As shown in Fig. 43, this finding was elucidated...
by the p-d Zener model generalized to the case of a non-uniform hole distribution obtained by solving the Poisson equation in thin (Ga,Mn)As layers (Eq. 20), in which the Fermi level at the surface is pinned in the gap region by surface states [Stolichnov et al., 2010; Sawicki et al., 2010].

However, a much higher value, $\gamma \gtrsim 1$, was observed for MIS structures of (Ga,Mn)Sb, as shown in Fig. 44(a) [Hsiao Wen Chang et al., 2013]. According to theoretical evaluations from Eq. 20, the efficiency of $T_C$ tuning by $V_G$ and, hence, the magnitude of $\gamma$ depends strongly on the energy position of surface states in respect to the valence band top. Since in (Ga,Mn)Sb, in contrast to (Ga,Mn)As, the Fermi level is pinned in the valence band by surface states, a large value of $\gamma$ is theoretically expected, as depicted in Fig. 44(b).

A relation between $T_C$ and film resistance $R$, changed by ferroelectric overlayers, was determined to be $\delta = -d\ln T_C/d\ln R = 0.35 \pm 0.05$ for a number of thin (Ga,Mn)As films [Stolichnov et al., 2011]. Information on changes in hole mobility is needed to compare $\delta$ and $\gamma$.

Finally, results of a study of $T_C$ as a function of hydrostatic pressure $P$ [Gryglas-Borysiewicz et al., 2010] were found consistent with the diagram in Fig. 38. $T_C$ increases with $P$ according to the p-d Zener model at a high hole concentration but it decreases in a sample close to the localization boundary. An increase of $T_C$ with $P$, of the magnitude corroborating the p-d Zener model, was expected, as depicted in Fig. 38(b).

Relative Curie temperature $T_C$ as a function of sheet hole concentration $p_s$ (10^15 cm$^{-2}$)

(a) 

In (Ga,Mn)As films (Stolichnov et al., 2010) and in Ga$_{1-x}$Mn$_x$P, $x \approx 0.038$, obtained by ion implantation and pulsed-laser melting [Winkler et al., 2011], respectively. Dotted lines show limiting trends in $T_C \propto p^\gamma$, where $\gamma = 0.5$ and $1$, in accord with the theoretical anticipation, $\gamma = 0.6-0.8$ [Dietl et al., 2001b].

For comparison, the ab initio approach providing the data shown in Fig. 40 predicted ferromagnetic ordering to vanish already entirely ($T_C = 0$) for hole density reduced twofold by compensation [Sato et al., 2010]. After Winkler et al., 2011

Finally, results of a study of $T_C$ as a function of hydrostatic pressure $P$ [Gryglas-Borysiewicz et al., 2010] were found consistent with the diagram in Fig. 38. $T_C$ increases with $P$ according to the p-d Zener model at a high hole concentration but it decreases in a sample close to the localization boundary. An increase of $T_C$ with $P$, of the magnitude corroborating the p-d Zener model, was expected, as depicted in Fig. 38(b).
a value consistent with its independent magnetooptical evaluation [Kossacki, et al. 2004a]. A more detailed analysis, combining solving of the Schroedinger equation for a given Mn distribution with Monte Carlo simulations for competing FM and AF interactions, confirmed the presence of scattering broadening of DOS, and the associated reduction of $T_F$ at low hole concentrations [Lipińska, et al. 2009]. Furthermore, the simulations explained a specific shape of $M(H)$ in terms of fast Mn dynamics, even below $T_C$, caused by AF coupling to Mn spins residing beyond the region penetrated by holes.

A particularly relevant is the question whether the Zener theory can be extended to n-type DMSs. So far an indication of ferromagnetism was found by the observation of resistance hysteresis in n-Zn$_{1-x}$Mn$_x$Te with $x = 3\%$ and $n = 1.4 \times 10^{20}$ cm$^{-3}$, which persisted up to 160 mK [Andrearczyk, et al. 2001]. Such a value of $T_C$, factor of 20 lower than in p-type Zn$_{1-x}$Mn$_x$Te with a similar Mn content, is—in fact—expected theoretically from Eq. [16] for similar values of $A_F$ and $\rho_h$ in both systems, one order of magnitude difference between $\alpha^2$ and $\beta^2$ implies that the Mn spin susceptibility in the absence of carriers, $\chi_S(T_C)$, has to be greater by a similar factor, which was realized by lowering temperature below 200 mK.

### 3. Curie temperatures in II-VI DFSSs

Since Mn in II-VI DMSs does not provide any carriers, it is possible to vary the Mn and hole concentration independently as well as to prepared modulation-doped quantum wells, in which the mean free path is longer than the well width. However, a relatively strong short-range antiferromagnetic (AF) coupling between Mn spins in II-VI DMSs, reduces $T_C$ significantly, as discussed in Sec. [VI].

The corresponding values of $x_{AF}(x)$ and $x_{eff}(x)$ were determined from magnetization or spin-splitting studies for undoped DMSs, confirming that at low Mn concentrations $x \lesssim 5\%$, $x_{eff} = x (1-x)^{12}$, as AF coupling between the nearest neighbor Mn pairs is there essential. As shown in Fig. 45, taking the presence of the AF interactions into account, the magnitudes of $T_F$ can be described quantitatively in p-(Zn,Mn)Te [Ferrand, et al. 2001].

In the case of 2D quantum wells of p-(Cd,Mn)Te, the data shown in Fig. 45 substantiated the validity of Eq. [21] and, in particular, the dimensional enhancement of DOS and thus of $T_F$ in the range of low carrier densities. The parameter $A_F = 2.0$ was adjusted to explain the magnitude of $T_F$ [Boukari, et al. 2002; Haury, et al. 1997], a value consistent with its independent magnetooptical evaluation [Kossacki, et al. 2004a]. A more detailed analysis, combining solving of the Schroedinger equation for a given Mn distribution with Monte Carlo simulations for competing FM and AF interactions, confirmed the presence of scattering broadening of DOS, and the associated reduction of $T_F$ at low hole concentrations [Lipińska, et al. 2009]. Furthermore, the simulations explained a specific shape of $M(H)$ in terms of fast Mn dynamics, even below $T_C$, caused by AF coupling to Mn spins residing beyond the region penetrated by holes.

### B. Interlayer coupling

Detailed theoretical studies of interlayer exchange energy were carried out for (Ga,Mn)As/GaAs superlattices within the multi-orbital TBA [Sankowski and Kacman, 2005]. The case of the (Al,Ga)As spacer was also considered. This theory predicted the regions of hole and Mn densities as well as spacer thicknesses and Al concentrations, in which an antiferromagnetic interaction between (Ga,Mn)As layers should appear. However, only ferromagnetic coupling has so-far been observed experimentally for undoped spacers, as discussed in Sec. [VI]. It would be interesting to find out whether the parameter space where the antiferromagnetic interaction exists would become narrower if disorder and hole redistribution between particular LT-grown layers were incorporated into the theory in a self consistent manner. Furthermore, the role of dipole-dipole interactions is to be considered, too.

### C. Magnetization and specific heat

It appears not easy to separate experimentally the hole contribution $M_s$ to the total magnetization $M$ and, in particular, to verify whether it reduces $M_s$ by 10%, as predicted theoretically. However, measurements of XMCD at the As K-edge did provide the values of spin and orbital magnetic moments [Freeman, et al. 2008; Wadley, et al. 2010] in agreement with the theory of hole magnetization [Sliwa and Dietl, 2013]. Furthermore, a
recent study (Ciccarelli et al., 2012), using (Ga,Mn)As as a gate for the Coulomb blockade in an Al dot, allowed to determine the dependence of the Fermi level position on the magnetic field. By using thermodynamic relations \( \epsilon_F = -\partial F_c / \partial p \) and \( M_c = -\partial F_c / \partial H \), where \( F_c \) is the carrier free energy we obtain \( \partial \epsilon_F / \partial H = \partial M_c / \partial p \). For \( x = 3\% \) and assuming \( x_I = 0 \) or 0.5% the theory (Sliwa and Dietl, 2006, 2013) leads to \( -\partial M_c / \partial p = 14 \) or 15 \( \mu eV/T \), respectively, in good agreement with the experimental value of \( -\partial \epsilon_F / \partial \mu_0 H = 18 \pm 3 \mu eV/T \) for Ga\(_{0.97}\)Mn\(_{0.03}\)As in the magnetic fields saturating Mn spins, \( \mu_0 H > 7.5 \) T (Ciccarelli et al., 2012).

It appears that there are three main ingredients underlying the temperature dependence of spontaneous magnetization in (Ga,Mn)As-type DFSs, which we discuss

• The mean-field Eq. 14 allowed to determine TM magnetization \( M(T,H) \). It was predicted that the dependence \( M(T) \) should evolve from the Brillouin-like convex form at high hole densities towards a concave shape at the Fermi energy smaller than the low temperature spin splitting of the carrier band (Dietl et al., 2001b). Such a change in the magnetization behavior on reducing carrier density at a given Mn concentration was indeed observed in (Ga,Mn)As (Mayer et al., 2010). However, the concave shape is also expected, and commonly observed (Sheu et al., 2007), when the proximity of the Anderson-Mott localization results in the formation of superparamagnetic-like regions, whose magnetization grows relatively slowly on decreasing temperature (Sawicki et al., 2010).

• It was shown that away from the localization boundary, the dependence \( M_s(T) \) obeys the Bloch \( T^3/2 \) law, demonstrating the importance of spin wave excitations (Potashnik et al., 2002). However, the magnitude of the spin wave stiffness \( D \) which was obtained in this way for a large series of samples, was by about factor of 2 greater than expected theoretically (Werpachowska and Dietl, 2010a). This discrepancy was resolved (Werpachowska and Dietl, 2010a) by taking into account the presence of the spin gap brought by the anisotropy field (Eq. (A4)). As shown in Fig. 46 the theory described the experimental dependence \( M_s(T) \) with no adjustable parameters.

• A complex temperature dependence of magnetization was revealed in samples for which a combination of strain, hole and Mn density values resulted in the temperature-induced spin reorientation transition (Sawicki et al., 2004, 2005; Wang et al., 2005c). A simple single-domain model was found to describe both \( \bar{M}_s(T) \) in the whole temperature range and a critical-like behavior of a.c. magnetic susceptibility in the vicinity of the transition (Wang et al., 2005c). Within the Ginzburg-Landau approach, spin wave stiffness \( D \) controls also the lambda-like anomaly of specific heat \( C(T) \) near \( T_C \). As shown in Fig. 47 the theory (Sliwa and Dietl, 2011) described the experimental data (Yuldashev et al., 2010) reasonably well, particularly assuming that owing to uniaxial anisotropy, the Ising universality class applies to (Ga,Mn)As.
D. Magnetic anisotropy and magneto-elastic phenomena

A comparison of experimental data summarized in Sec. III.B to the theoretical model (Sec. IX.C) developed assuming literature values of deformation potentials and elastic moduli (see, e.g., Stefanowicz et al., 2010a), leads to a number of conclusions concerning particular contributions to bulk crystalline magnetic anisotropy in DFs.

**Cubic anisotropy:** As anticipated taking disorder into consideration (Dietl et al., 2001b), the experimental value of the cubic anisotropy field does not show any noticeable oscillatory behavior as a function of the hole concentration, expected within the disorder-free theory. At the same time, the observed order of magnitude \( \mu_0 H_c \approx 0.1 \) T at \( T \ll T_c \) and temperature dependence are consistent with the predicted amplitude of oscillations (Abolfath et al., 2001; Dietl et al., 2001b; Stefanowicz et al., 2010a; Zemen et al., 2009). However, why the cubic easy axis assumes predominantly (100) orientations in (Ga,Mn)As, whereas the (110) directions are preferred in (In,Mn)As and (Ga,Mn)P has not yet been theoretically explained.

**In-plane uniaxial anisotropy:** The sign and values of \( \mu_0 H_{xy} \) and \( \mu_0 H_{zz} \) found experimentally confirm the existence of a theoretically expected surplus of Mn dimers oriented along the [110] direction for (001) (Ga,Mn)As (Birowska et al., 2012), though the dimer formation can depend sensitively on the surface reconstruction, partial pressure of As, growth rate and temperature. The corresponding lowering of symmetry is described by effective strains \( \epsilon_{xy} \) and \( \epsilon_{zz} \) that can be incorporated into the \( p-d \) Zener model. The in-plane uniaxial anisotropy field \( \mu_0 H_{xy} \) obtained is this way for (Ga,Mn)As (Stefanowicz et al., 2010a; Zemen et al., 2009) is shown in Fig. 18 as a function of the hole concentration for two values of spontaneous magnetization \( M(T) \). In this range, \( \mu_0 H_{xy} \) is linear in \( \epsilon_{xy} \), so that the magnitudes of \( p \) and \( M \) corresponding to the spin reorientation transitions [i.e., \( H_{xy}(p,\Delta) = 0 \)] do not depend on the actual asymmetry in the Mn dimer distribution, which can vary from sample to sample, depending on epitaxy conditions. These theoretical results are in accord with directions of spin-reorientation transitions [110] \( = [110] \) observed as a function of either hole density or temperature in both high (Sawicki et al., 2005) and low hole concentration range, where gating was employed to vary hole concentrations (Chiba et al., 2008a; Sawicki et al., 2010).

Furthermore, the theory confirms a weaker temperature dependence of \( H_{xy} \) comparing to \( H_c \), which according to theoretical results shown in Fig. 19 for (Ga,Mn)As leads to the spin reorientation transition from (100) to [110] on increasing temperature, in agreement with the experimental observations (Kamara et al., 2012; Wang et al., 2005c; Welp et al., 2003).

The same approach was successfully applied to explain the direction and the magnitude of easy axis rotation as a function of the voltage applied to a piezoelectric actuator containing a (Ga,Mn)As film cemented to its surface along one of (110) directions (Rushforth et al., 2008). In this case a real and known strain \( \epsilon_{xy} \) is imposed by the actuator.

Theoretical description of the magnitude of \( K_{zz} \) within the \( p-d \) Zener model for (113) (Ga,Mn)As (Stefanowicz et al., 2010a) pointed also to the presence of a symmetry lowering perturbation. The current theory of
dimer-related magnetic anisotropy for (001) (Ga,Mn)As (Birowska et al. 2012) has not yet been extended to other orientations of the substrate, so that this finding awaits for a theoretical interpretation.

Out-of-plane uniaxial anisotropy. As exemplified in Fig. 50, the theory (Dietl et al. 2001b) describes quantitatively the magnitude anisotropy field $\mu_z H_{zz}$ in (001) (Ga,Mn)As at 4 K as well as its dependence on the epitaxial strain $\epsilon_{zz}$, as determined by various groups (Cubukcu et al. 2010; Glunk et al. 2009) for samples with typical hole and Mn concentrations, $3 \times 10^{20} \lesssim p \lesssim 6 \times 10^{20}$ cm$^{-3}$, $0.035 \lesssim x_{\text{eff}} \lesssim 0.065$. At given $p$, $H_{zz}$ varies linearly with spontaneous magnetization $M_s(T)$ (at not too high $M_s$), which accounts for the dependence of $H_{zz}$ on temperature. A good agreement between the experiment and theory for magnetic anisotropy generated by epitaxial strain was also found for (113) (Ga,Mn)As (Ste-fanowicz et al. 2010a).

By decreasing hole density down to $p \approx 10^{20}$ cm$^{-3}$, a temperature-dependent spin reorientation transition takes place from in-plane to perpendicular easy axis orientations (see, Sec. III.B), in agreement with theoretical predictions within the six bands’ kp model (Dietl et al. 2001b), as shown in Fig. 51. The perpendicular alignment of the easy axis for compressive strain is actually expected within a kp four band model of the valence band, which is valid at low hole concentrations. This model implies the perpendicular and in-plane orientation of the total orbital momentum $\vec{J}$ of holes for compressive and tensile strain, respectively, explaining the corresponding alignment of Mn spins in (Al,Ga,Mn)As (Takamura et al. 2002) and (Ga,Mn)P (Biiller et al. 2007) in the low hole concentration regime.

According to theoretical results displayed in Fig. 51 at high hole densities $p \gtrsim 10^{21}$ cm$^{-3}$, a subsequent spin reorientation transition between in-plane and perpendicular-to-plane magnetic anisotropy is expected theoretically for (Ga,Mn)As (Sawicki et al. 2004; Wer-pachowska and Dietl 2010b; Zemen et al. 2009), which has not yet been found experimentally.

In the case of patterned nanobars, a starting point of a theoretical analysis was a strain distribution, as determined by finite element computations and x-ray diffraction for nanobars patterned along various crystallographic directions from two different (Ga,Mn)As wafers (King et al. 2011). A linear dependence of the magnetic anisotropy energy on strain allowed to develop theory in terms a mean strain over the bar cross-section. As a whole, the theory confirmed that strain relaxation accounts for the alignment of the easy axis along the long edge of nanobars, as observed (Hümpfner et al. 2007; King et al. 2011). However, while the computed magnitude and temperature dependence of magnetic anisotropy characteristics were found in a satisfactory agreement with the data for nanobars patterned from one wafer, there were significant quantitative discrepancies in the case of another series of nanobars (King et al. 2011). It might be that the single domain approximation breaks down in some samples with highly non-uniform strain distribution.

Another important effect of strain in zinc-blende crystals is the appearance in the hole dispersion of terms
linear in $k$, coupled to the hole total orbital momentum $\vec{J}$, given within the four band model by [Ivchenko and Pikus, 1995],

$$\mathcal{H}_{k_e} = C_0 \hat{\psi} \vec{J} + C_6 \hat{\psi} \vec{J}.$$  

(27)

Here $C_i$ are relevant $kp$ parameters (deformation potentials); $\hat{\psi}_x = k_y \epsilon_{xy} - k_z \epsilon_{xz}$ [and cyclic permutations (c. p.)] and $\psi_x = k_y (\epsilon_{yy} - \epsilon_{zz})$ (and c. p.), where $\epsilon_{ij}$ denote the sum of the deformation-induced and effective components of the strain tensor. Remarkably, the form of $\mathcal{H}_{k_e}$ implies that electric current, by leading to a non-zero value of $\langle \vec{k} \rangle$ in its direction, generates an effective magnetic field that can orient hole spins and, thus, serve to switch the direction of magnetization. Such an effect was demonstrated experimentally (Chernyshov et al., 2009; Endo et al., 2010b), and interpreted within this kp formalism.

Finally, we note that the theory (Dietl et al., 1997; Kossacki et al., 2004b) readily explains why in p-type (Cd,Mn)Te quantum wells under compressive strain the easy axis is along the growth direction $z$ (Fig. 19), as for this strain configuration and confinement the heavy hole subband is occupied, for which $J_z = \pm 3/2$ and, thus, $s_z = \pm 1/2$. In contrast, for tensile strain light hole subband is involved, so that $J_z = \pm 1/2$, and hence the hole spins and, thus, the easy axis assume the in-plane orientation.

E. Domain structure, exchange stiffness, and spin waves

Macroscopically large magnetic domains were observed in (Ga,Mn)As with in-plane magnetic anisotropy (Welp et al., 2003), which confirmed excellent micromagnetic properties of this system. High-resolution electron holography technique provided direct images of domain wall magnetization profiles of such films (Sugawara et al., 2008). The Néel type domain walls were found of the width ranging from approximately 40 to 120 nm, the values consistent with the magnitude of $(A/K_C)^{1/2} \approx 40$ nm computed within the p-d Zener model for the studied films (Sugawara et al., 2008).

In the case of a 200 nm thick (Ga,Mn)As film under tensile strain imposed by an (In,Ga)As substrate, with the easy axis perpendicular to the plane, periodic stripe domains were revealed by a micro-Hall scanning probe (Shono et al., 2000). It was found (Dietl et al., 2001a) that the values of the energy $K_u$ of uniaxial magnetic anisotropy and of the exchange stiffness $A$, both determined from the p-d Zener model, lead to the low-temperature width of the stripes $W = 1.1 \mu$m, which compares favorably with the experimental value $W = 1.5 \mu$m. However, the data suggest that a decrease of $A$ with temperature is slower than expected theoretically.

More recently, the values of $M_s$, $K_u$, and $A$ were determined for a series of (Ga,Mn)As and (Ga,Mn)(As,P) 50-nm films with perpendicular magnetic anisotropy by combining magnetometry and ferromagnetic resonance with Kerr microscopy that allowed to determine the period of stripe domains (Haghgoo et al., 2010). As shown in Fig. 52, the low temperature value of $D = 2g\mu_B A/M$ deduced from the data agrees with the expectations of the p-d Zener model for $T \ll T_C$. On the other hand, a decrease of $A$ and, thus of $D$ with temperature for this sample is faster in this case (Haghgoo et al., 2010) than expected theoretically. Accordingly, the magnitude of $D$ obtained for a series of samples at $T = 0.4T_C$, even enlarged by 20% implied by temperature variation of $M_s$ between $T = 0.4T_C$ and $T \ll T_C$, are lower than theoretically predicted (Fig. 52).

Spin-wave signatures were clearly resolved in ferromagnetic resonance (FMR) (Bihler et al., 2009; Fedorych et al., 2002; Liu et al., 2007) and pump-probe differential magnetic Kerr experiments (Wang et al., 2007a) on (Ga,Mn)As films. However, a quantitative interpretation of resonance energies in terms of dimensional quantization of the spin wave spectrum was so far possible only assuming the presence of long range inhomogeneities along the growth direction, taken in the form of a triangular (Bihler et al., 2009) or a parabolic well (Liu et al., 2007). Similarly, heavily debated are effects of pinning, magnetic anisotropy, and modes at surfaces and interfaces (Bihler et al., 2009; Liu et al., 2007; Wang et al., 2007a). The experimental values of spin-wave stiffness $D$ obtained by various experiments show a rather large

![FIG. 52 (Color online) Spin-wave stiffness obtained from time-resolved magnetooptical studies of Ga$_{1-x}$Mn$_x$As at 15 K (solid circles, Némeč et al., 2013) and from widths of domain stripes (Haghgoo et al., 2010) measured at 4 K (empty square) or determined by extrapolation to low temperatures the values obtained at $T = 0.4T_C$ (solid squares) for (Ga,Mn)(As,P) on GaAs and (Ga,Mn)As on (In,Ga)As (solid triangle). Solid and dotted lines represent zero-temperature p-d Zener modeling with no adjustable parameter for $p = 0.3N_0x$ and $p = N_0x$, respectively, in (Ga,Mn)As (Werpaschowska and Dietl, 2010a). Zero-temperature spin wave stiffness obtained by ab initio computations is shown for a comparison (dashed line, Bouzerar, 2007).](image-url)
dispersion [Werpachowska and Dietl 2010a].

Recently, spin-wave-like resonances were detected by time-resolved magneto-optics on a series of thin annealed (Ga,Mn)As samples [Némeč et al. 2013]. Energy differences between particular resonances and scaling with sample thickness indicated that dimensional quantization of bulk spin waves in spatially uniformed ferromagnetic slabs was observed. The values of $D$ obtained in this way are larger by a factor of about 2 than expected within the $p$-d Zener model, as shown in Fig. 52.

F. Spintronic structures

Modeling of spin-injection efficiency, tunneling magnetoresistance, and domain wall resistance in various structures of DFSs is particularly appealing from the theoretical perspective as useful results can be obtained neglecting disorder entirely. Below we present a quantitative outcome of the disorder-free Landauer-Büttiker coherent transport theory combined with either multiband $kp$ or multiorbital tight-binding approaches, the latter better handling the inversion symmetry breaking (Dresselhaus terms) and interfacial Rashba effects as well as the tunneling via $\vec{k}$ points away from the zone center. Within the employed formalisms, the holes are assumed to reside in the GaAs-like valence band, subject to the $p$-d exchange interaction treated within the virtual crystal and molecular field approximations with the standard values of the $sp-d$ exchange energies, $N_0\alpha = 0.2$ eV and $N_0\beta = -1.2$ eV. As shown in this section, a number of prominent spintronic characteristics, including spin injection efficiency, the magnitude of TMR and TAMR effects as well as domain-wall resistance (reviewed in Secs. IV V.D and III.E respectively), are captured by such modeling, though some aspects of experimental results, such as zero-bias anomaly in tunneling spectra (Chun et al. 2002; Pappert et al. 2006; Richardella et al. 2010), a strong temperature dependence of TMR magnitude at $T \ll T_C$ (Chiba et al. 2004a) or very large values of TAMR in nanostructures (Giddings et al. 2005; Riister et al. 2005) or junctions (Pappert et al. 2006) point to the importance of correlation effects at the localization boundary, which have not been taken into account in theoretical models developed up to now.

1. Spin current polarization

Spin current polarization $\Pi_{\text{inj}}$ in Esaki diodes was computed within the forty orbitals' $sp^5d^3s^*$ tight-binding model (Sankowski et al. 2006, 2007; Van Dorpe et al. 2005). Figure 53 presents theoretical values of $\Pi_{\text{inj}}$ at low bias for various Mn and hole concentrations for a Ga$_{1-x}$Mn$_x$As/n-GaAs Esaki diode with the depletion region consisting of 4 double-monolayers and assuming $n = 10^{19}$ cm$^{-3}$ in GaAs. As the magnitude and the dependence of hole spin splitting on the orientation of $\vec{k}$ with respect to $\vec{M}$ are different in particular valence band subbands, the predicted values of $\Pi_{\text{inj}}$ vary strongly not only with $x_{\text{eff}}$, $p$, and $n$ but also with the angle $\theta$ between $\vec{j}$ and $\vec{M}$.

As mentioned in Sec. IV, three experiments (Ciorga et al. 2009; Kohda et al. 2006; Van Dorpe et al. 2004) carried out at $T \ll T_C$ and $\theta = 45^\circ, 0^\circ, 90^\circ$ for samples with nominal Mn content $x = 0.08, 0.057, 0.05$ and $T_C = 120, 70, 65$ K led to $\Pi_{\text{inj}} = 0.4, 0.47, and 0.51$, respectively. The values of $T_C$ for particular samples imply a certain degree of compensation by interstitial Mn and/or antisite defects, so that $x_{\text{eff}} \leq x$ and $p < N_0x$. Taking this into account, we conclude that the theoretical results summarized in Fig. 53 are consistent with the experimental findings, though a quantitative comparison requires more accurate information on the magnitudes of $x_{\text{eff}}$ and $p$. Furthermore, the theory (Van Dorpe et al. 2005) explained a decay of $\Pi_{\text{inj}}$ to zero at the bias of the order of 0.2 V (Ciorga et al. 2009; Kohda et al. 2006; Van Dorpe et al. 2004) (see, Fig. 26). Since spin injection is a surface-sensitive phenomenon, the theory (Sankowski et al. 2007; Van Dorpe et al. 2005) predicted a 6% difference in $\Pi_{\text{inj}}$ for $M \parallel [110]$ in comparison to the case $M \parallel [110]$ at $x_{\text{eff}} = 0.08$ and $p = 3.5 \times 10^{10}$ cm$^{-2}$, the effect found experimentally (Van Dorpe et al. 2005). This difference is about factor of 10 greater than that generated by effective shear strain (see, Sec. X.D), $\epsilon_{xy} \lesssim 0.1\%$ (Sankowski et al. 2007).

It is worth noting that the magnitudes of $\Pi_{\text{inj}}$ depicted in Fig. 53 are only approximately equal to the values of spin current polarization $P_c$ provided by Andreev reflection or domain wall velocity. With this taken into account, the theoretical results on $\Pi_{\text{inj}}$ are consistent with...
et al. FIG. 54 Computed magnitudes of TMR

$P_{\text{kp}}$ eight band polarized electrons was predicted theoretically, within an $2. Magnetic tunnel junctions

$x$ in double-barrier InAs/AlSb/Ga

The tight binding model discussed in the previous subsection in the context of spin current polarization was also successfully employed to describe low temperature magnitudes of TMR in tri-layer structures with (Ga,Mn)As electrodes and AlAs

2. Magnetic tunnel junctions

Figure 54 shows the magnitudes of TMR, for the hole concentration $10^{20}$ cm$^{-3}$, current along the [001] direction, and magnetization along the [100], [001], and [101] directions (TMR$_x$, TMR$_z$, and TMR$_{zx}$, respectively). The data are shown for two values of hole spin polarization $P = 0.75$ and 0.89 corresponding to, at $T \ll T_C$, $x_{\text{eff}} \simeq 0.042$ and 0.066, respectively. The values expected from Julliere’s formula TMR = $2P^2/(1+P^2)$ are shown by horizontal lines. From Brey et al. 2004

available data on $P_c$ (Curiale et al. 2012).

An effective and tunable by bias injection of spin polarized electrons was predicted theoretically, within an eight band $kp$ model, for interband resonant tunneling in double-barrier InAs/AlSb/Ga$_{1-x}$Sb/AlSb/InAs heterostructures (Petukhov et al. 2003).

3. Domain wall resistance

A spherical four band model (Nguyen et al. 2006) and a twenty orbitals’ $sp^3s^*$ tight-binding approximation (Oszwaldowski et al. 2006) were employed to evaluate the intrinsic domain-wall resistivity $R_{\text{int}}$ in unstrained (Ga,Mn)As within the disorder free Landauer-Büttiker formalism. These studies demonstrated that owing to the spin-orbit interaction $R_{\text{int}}$ > 0 even if the domain-wall width is much longer than the de Broglie wavelength of holes at the Fermi level. Various crystallographic orientation of (Ga,Mn)As nanowires containing either Bloch

FIG. 55 (Color online) Comparison of experimental (a) and computed (b) values of differential conductance $dI/dV$ (thin lines) and TAMR = $(I_{[001]} - I_{[100]})/I_{[100]}$ (thick lines), where the indices [001] and [100] describe magnetization orientation, as a function of bias $V$ in a resonant tunneling diode with a (Ga,Mn)As hole emitter, grown along the [001] direction. Experimental and theoretical data come into agreement if renormalization of $V$ and TAMR values by a factor of 5 implied by a series resistance is taken into account. The computation was performed for indicated values of average hole energy $\epsilon$ and in plane momentum $k_x$. From Olsen et al. 2007.

Theoretical studies were put forward to examine TMR and TAMR in double barrier structures within a six band $kp$ theory. It was demonstrated that spin-dependent resonant tunneling could dramatically enhance TMR in resonant tunneling diodes containing both emitter and collector of (Ga,Mn)As (Petukhov et al. 2002). This prediction has not yet been confirmed experimentally presumably because it is not easy to ensure coherent tunneling in RTDs grown by LT MBE. (Mattana et al. 2003). In contrast, the six band $kp$ formalism explained a character of TAMR oscillations as a function of bias in a double barrier structure with one (Ga,Mn)As electrode, as depicted in Fig. 55 (Olsen et al. 2007).

The intrinsic domain-wall resistivity $R_{\text{int}}$ was determined by the theory reproduced a fast decrease of TMR with the device bias as well as it indicated that the magnitude of TAMR should not exceed 10% under usual strain conditions and for hole densities corresponding to the metal side of the metal-to-insulator transition (Sankowski et al. 2007).
or Néel domain walls were considered (Oswwaldowski et al. 2006). It was found that the computed values $10^{-2} \lesssim R_{\text{int}}^2 \lesssim 10^{-3} \Omega \mu m^2$ depending on hole density, are at least one order of magnitude smaller than the ones determined experimentally for domain walls pinned by etched steps (Chiba et al. 2006b; Wang et al. 2010b; see, Sec. [II.F]) but consistent with the much lower value $R_{\text{int}}^2 \approx 0.01 \pm 0.02 \Omega \mu m^2$, found in the case of the wall pinning by linear defects (Wang et al. 2010b).

XI. SUMMARY AND OUTLOOK

A series of accomplishments presented in this review has documented a prominent role of dilute ferromagnetic semiconductors, especially (Ga,Mn)As, in bridging science and technology of semiconductors and magnetic materials. Indeed, several spintronic functionalities revealed in DFSs are now extensively explored in ferromagnetic metals, examples include electrical spin injection to semiconductors, electric-field control of magnetism, single domain wall motion by spin-torque transfer in the absence of a magnetic field, and tunneling anisotropic magnetoresistance in sequential, resonant, and Coulomb blockade regimes. Conversely, the Stoner-Wohlfarth, Landau-Lifshitz-Gilbert, and Berger-Slonczewski formalisms, developed initially for magnetic metals, have been successfully employed to describe spintronic characteristics of DFS-based ferromagnets.

An outstanding aspect of DFSs is that input parameters to these formalisms can be theoretically evaluated by incorporating exchange coupling between carriers and localized spins into the computationally efficient $k\cdot p$ or tight binding approaches, employed routinely to model semiconductor properties and devices. As emphasized in this review, the $p-d$ Zener model describes semi-quantitatively, and often quantitatively, a number of thermodynamic and micromagnetic properties of tetrahedrally coordinated DFSs containing delocalized or weakly localized valence band holes, including the Curie temperature in various dimensionality systems, Mn spin and hole magnetization, anisotropy fields, and exchange stiffness as a function of hole and Mn ion concentrations. It remains to be seen whether progress in the experimental determination of these concentrations will bring experimental and theoretical results even closer.

Furthermore, a number of findings reviewed here documented substantial progress in assessing the role played by Anderson-Mott localization, the competition between ferromagnetic and antiferromagnetic interactions, solubility limit, self-compensation, and the transition to the strong coupling case with decreasing of the lattice parameter — the challenges that had been identified (Dietl et al. 2000) as possible obstacles on the way to synthesize a DFS supporting ferromagnetic order up to above room temperature. In particular, as currently known, the influence of antiferromagnetic interactions and self-compensation limit $T_C$ to about 200 K so far in (Ga,Mn)As. At the same time, the importance of quantum localization in (Ga,Mn)As and related systems makes a quantitative description of static and dynamic conductivities difficult as there is no appropriate theory, even for non-magnetic semiconductors, in this regime. The strong coupling, in turn, shifts the insulator-to-metal transition to a non-achievable Mn and hole concentration range as of today in nitrides and oxides, so that rather than hole-mediated coupling, ferromagnetic superexchange accounts for $T_C$ up to 13 K in Ga$_{0.9}$Mn$_{0.1}$N with the Fermi level residing in the Mn acceptor impurity band. A striking consequence of solubility limits is a self-organized assembling of magnetic nanocrystals inside a semiconductor host by chemical or crystallographic phase separation. These heterogeneous magnetic systems have apparent $T_C$ usually well above room temperature and, accordingly, a number of groups look for possible spintronic functionalities of such nanocomposites.

Is it then possible to obtain a high $T_C$ uniform DFS? The view that the $p-d$ Zener mechanism can result in the robust ferromagnetism is supported by the case of double perovskite compounds, such as Sr$_2$CrReO$_6$, where this mechanism leads to magnitudes of $T_C$ as high as 625 K, despite the fact that the distance between localized spins is as large as 0.6 – 0.7 nm (Serrate et al. 2007), much greater than the separation of 0.5 nm between next nearest neighbor cations in GaN and ZnO. In light of this estimate, the search for a room temperature uniform DFS will continue to be an active field of research. Here, in addition to 3$d$ TM impurities in various hosts, other spin dopants will be considered, including 4$d$ TMs and elements with open $f$ shells as well as spin carrying defects. However, independently of the progress in achieving a high $T_C$ system, (Ga,Mn)As and related compounds as well as a (Ga,Mn)N-type of ferromagnets will continue to constitute an important playground for exploring novel phenomena, functionalities, and concepts at the intersection of semiconductor physics and magnetism.

In addition to current interest in various magnetically doped semiconductors, oxides, and organic materials, a lot of attention will be devoted to four emerging families of compounds: (i) high Curie temperature ferromagnetic spinel oxides and Heusler compounds such as Mn$_2$CoAl (Ouardi et al. 2013), awaiting mastering of defect and carrier control; (ii) high Néel temperature semiconductors, e. g. LiMnAs, for antiferromagnetic spintronics (Jungwirth et al. 2011); (iii) topological insulators, in which ferromagnetism might be mediated by Dirac electrons, e. g., (Bi,Mn)$_2$Te$_3$ (Checkelsky et al. 2012); (iv) derivatives of FeAs- and CuO-based superconductors, such as (K,Ba)(Zn,Mn,Fe)$_2$As$_2$ compounds (Zhao et al. 2013) for studies of interplay between $p-d$ Zener ferromagnetism, antiferromagnetic superexchange, and superconductivity. Here, nanocharacterization protocols, elaborated over the recent years for DMSs (Bonnmann 2011), will play the essential role in the meaningful development of new materials.

One may anticipate that an increasing number of
studies will be devoted to hybrid structures combining DFSs with other ferromagnets, antiferromagnets, superconductors, and topological insulators as well as to nanostructured systems, such as magnetically doped rings, nanowires, nanoconstrictions, quantum dots, and colloidal nanocrystals, including possibly more complex structures, such as nano electro mechanical systems. Recent progress in the fabrication of (Zn,Mn)Te/(Zn,Mg)Te core/shell nanowires (Wojnar et al., 2012) allows one to search for exotic ground states in modulation-doped 1D magnetic systems. Similarly, studies of magnetic quantum dots (Abolfath et al., 2007) and colloidal nanocrystals (Beaulac et al., 2008; White et al., 2008) bridge on the one hand the physics of bound magnetic polaron and carrier-controlled ferromagnetism, and on the other, the electronic and nuclear magnetism.

In this review, we primarily focussed on properties and functionalities resulting from the collective spin phenomena. Another ultimate limit of DMSs research constitutes works on manipulation of a single TM spin. This field of solotronics (Koenraad and Flatté, 2011) began in DMSs by exploiting a single Mn in II-VI (Cibert et al., 2008; Goryca et al., 2009) and III-V (Kudelski et al., 2007) self-assembled quantum dots. Another single-impurity phenomenon, not yet explored in the context of DMSs, is the Kondo effect expected in the case of a localized spin coupled by an antiferromagnetic exchange to a Fermi sea of carriers.

On the theoretical side, enduring progress in the reliability of ab initio methods is expected. From the DFS perspective, in addition to the issue of results’ convergence as a function of the energy cut-off, density of d points, and the number of atoms in the supercells, three experimentally relevant challenges, of differing numerical complexity, can be given: (i) the incorporation of the spin-orbit interaction that significantly affects the band structure of DFSs and accounts for magnetic anisotropy effects – some progress in that direction has already been reported (Mankovsky et al., 2011); (ii) the improved treatment of the exchange-correlation functional, so that more realistic values of the band-gap and d-level positions can be predicted as well as Mott-Hubbard localization of d electrons handled adequately – here also improved computational schemes are being implemented (see, e. g., Di Marco et al., 2013; Stroppa and Kresse, 2009); (iii) the development of methods that would be able to tackle Anderson-Mott localization phenomena, such as the appearance of the Coulomb gap in the density of states at the Fermi level.

In retrospect, striking properties and functionalities found in DFSs, not only influenced semiconductor and metal spintronics but, to a large extent, have accounted for a spread of spintronic research over many other materials families. Search for novel magnetic semiconductors not only provided the concept of magnetism without magnetic ions in oxides, and demonstrated a surprising influence of the Fermi energy upon the position and distribution of magnetic ions in semiconductors. Research on topological aspects of the anomalous Hall effect in bands coupled by spin-orbit interactions paved the way for uncovering spin Hall effects and topological matter. It might be, therefore, expected that studies of magnetically doped semiconductors, insulators, and organic materials will continue to bring unanticipated and inspiring discoveries in the years to come.

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Appendix A: Micromagnetic theory

As surveyed in this review, the structure and motion of magnetic domains as well as a number of FMR and time-resolved magneto-optical experiments on (Ga,Mn)As and related compounds have demonstrated that the time-honored micromagnetic theory of ferromagnets applies to these systems. The use of the continuous medium approximation inherent to this theory is justified by a sizable width of domain walls that is much larger than an average distance between Mn spins. The micromagnetic theory provides a spatial and temporary evolution of magnetization in given values of a magnetic field and spin current. The magnitudes of magnetic anisotropy fields $H_i$ and exchange stiffness $A$ constitute input parameters in the static case, whereas in the dynamic situation the Landé factor $g$ and the Gilbert constant $\alpha_G$ enter additionally into theory. For a specific case of current-induced domain wall motion, non-adiabatic spin torque $\beta_\alpha$ is one more input parameter. As discussed in Sec. IX the magnitudes of these material parameters can be theoretically evaluated in a rather straightforward way, which constitutes a unique aspect of DFSs.

The starting point of the micromagnetic theory is the Landau-Lifshitz-Gilbert (LLG) equation according to which the dynamics of the local spin direction $\vec{m} = \vec{M}/M$ is determined by a competition of a torque, due to an ef-
ffective magnetic field $\mathbf{H}_{\text{eff}}$ and/or an electric current $\mathbf{j}$, with a damping term, characterized by the Gilbert constant $\alpha_G$,

$$\dot{\mathbf{m}} = -\gamma \mathbf{m} \times \mathbf{H}_{\text{eff}} - (1 - \beta_w \mathbf{m} \times (\mathbf{r}_f \cdot \nabla)\mathbf{m} + \alpha_G \mathbf{m} \times \dot{\mathbf{m}}). \quad (A1)$$

Here $\gamma = g \mu_B / h$, $\mathbf{H}_{\text{eff}}$ is given by a variational derivative, $\dot{\mathbf{H}}_{\text{eff}}(\mathbf{r}) = -\delta \mathcal{F}[\mathbf{M}(\mathbf{r})] / \delta \mathbf{M}(\mathbf{r})$; $\beta_w$ describes the magnitude of a non-adiabatic out-of-plane current torque brought about by the spin-orbit interaction, and $\mathbf{r}_f = g \mu_B P_c / (2eM)$, where $P_c$ is the current spin polarization, and $M$ is localized spin magnetization. Interestingly, by the Onsager reciprocity relations, not only the spin current in the presence of a spin texture $\dot{\mathbf{m}} = \dot{\mathbf{m}}(\mathbf{r})$ induces magnetization dynamics $\mathbf{m}(t)$, but also $\dot{\mathbf{m}}(r, t)$ can generate an electric current (Hals et al. 2009). While the LLG equation (together with initial conditions) allows to determine $\dot{\mathbf{m}}(r, t)$, its static limit $\dot{\mathbf{m}} = 0$ is also interesting, as it makes it possible to find expected spin textures $\dot{\mathbf{m}}(\mathbf{r})$ under stationary conditions, i.e., the domain structure.

It is seen from the form of the LLG equation that only contributions to $\dot{\mathbf{H}}_{\text{eff}}$, which can have components perpendicular to $\mathbf{m}$, are relevant. These include external and the shape dependent dipolar (demagnetization) fields coming from surrounding magnetic moments, $\mathbf{H}_{\text{ext}} = \mathbf{H}_{\text{A}}[\mathbf{M}(\mathbf{r})]$, as well as the crystalline anisotropy field $\mathbf{H}_a$ and the magnetization stiffness,

$$\mathbf{H}_{\text{ss}} = -\frac{2A}{M} \nabla^2 \mathbf{m}. \quad (A2)$$

This term originates from the free energy change $\delta \mathcal{F}$ associated with a twisting of $\mathbf{M}(\mathbf{r})$. To the lowest order in the gradient of the magnetization components in an isotropic medium, it has a general form,

$$\delta \mathcal{F} = A |\nabla \mathbf{m}|^2, \quad (A3)$$

where $A$ is called the exchange stiffness, as its magnitude scales with the strength of the exchange coupling between the spins. In the absence of an electric current and damping, these equations lead to the precession modes (spin waves) of the form,

$$\hbar \omega(q) = g \mu_B H_0 + D q^2, \quad (A4)$$

where the magnetic field $H_0$ determining the spin gap is an appropriate sum of $H_0$, $H_3$, and $H_4$, and $D = 2g \mu_B A / M$, as reviewed elsewhere (Liu and Furdyna 2006). Boundary conditions as well as additional modes and spin-wave pinning at surfaces and interfaces in thin films and other nanostructures will modify this dispersion relation. Furthermore, since the precession involves the system of coupled Mn and hole spins, the apparent Mn Landé factor $g$ will be reduced or enhanced for antiparallel and parallel orientation of corresponding magnetizations, respectively (Liu and Furdyna 2006; Sliwa and Dietl 2006), as mentioned in Sec. VII.B.1.

In the static and single domain approximation, for which $\dot{\mathbf{m}}$ is time and position independent, its direction points along $\mathbf{H}_{\text{eff}}$ and is determined by minimization of $\mathcal{F}$ in respect to spherical angles $\theta$ and $\phi$, as discussed in Sec. III.B for the specific case of (001) and (113) substrates.

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