Optical orientation in ferromagnet/semiconductor hybrids

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Abstract
The physics of optical pumping of semiconductor electrons in ferromagnet/semiconductor hybrids is discussed. Optically oriented semiconductor electrons detect the magnetic state of a ferromagnetic film. In turn, the ferromagnetism of the hybrid can be controlled optically with the help of a semiconductor. Spin–spin interactions near the ferromagnet/semiconductor interface play a crucial role in the optical readout and the manipulation of ferromagnetism.

1. Introduction
Nowadays a lot of effort is directed to the integration of magnetism into the semiconductor architecture of modern computers. The first approach is to construct a universal object combining ferromagnetic (FM) and semiconducting (SC) properties. Following pioneering research of the 1960s on Eu-based chalcogenides and Cd–Cr spinels, Ohno (1998) and Dietl et al (2000) initiated the recent active studies of new III–V-based FM semiconductors. The second approach—the so-called semiconductor spintronics—is based on the generation and manipulation of spin and spin currents entirely within non-magnetic semiconductors (Zutic et al 2004). The third approach (Prinz 1990) exploits ferromagnet/semiconductor (FM/SC) hybrid systems. One of the advantages is an additional degree of freedom consisting in the choice of a desirable FM/SC pair among paramagnetic semiconductors and a large number of FM materials. As demonstrated by Zakharchenya and Korenev (2005), another advantage is the possibility of both reading and controlling the magnetization of the unified system. As a result, the SC is not only a substrate for a FM to lie upon but also participates actively in information processing.

The goal of the review is to present the FM/SC hybrid as a strongly coupled spin system whose magnetism can be tuned optically. Special attention is given to those studies which discuss (or try to discuss) the effect of the FM–SC exchange and the feedback effect of the SC on the ferromagnetism of the unified spin system. I concentrate mostly on the optical tuning of the magnetism because this special issue is devoted mainly to the optics of semiconductor-based structures. Surely, it is possible to tune the FM/SC magnetism electrically. Zakharchenya and Korenev (2005) made an initial attempt to address this point.

I begin with the detection of ferromagnetism. For example, a FM film creates a static magnetic field. Although relatively weak, this magnetic field penetrates deep into a semiconductor. The electron spin interacts with the stray magnetic field by means of Zeeman interaction. Randomly distributed stray fields bring about the Larmor precession of semiconductor electron spins, thus leading to their spin relaxation. This effect can be used to distinguish between the magnetized and demagnetized states of the ferromagnet (section 2). The exchange interaction causes ferromagnetism and splits the energy levels of the ferromagnet itself by a large value (~1 eV). The wavefunction overlap between carriers and magnetic atoms of FM leads to a strongly coupled spin system. It is short range, so that the coupling is interfacial. The uniqueness of the FM/SC system lies in the electrical and optical tunability of the electrical and magnetic properties of the paramagnetic SC. This means that the ferromagnetism (for example, magnetic hysteresis loop) of the unified system can be tuned as well. As a result, the SC is not only a substrate for a FM to lie upon but also participates actively in information processing.

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FM atoms and semiconductor charge carriers in a FM/SC hybrid leads in equilibrium to the local spin polarization of semiconductor carriers—the so-called proximity effect. Under non-equilibrium conditions, the FM-induced spin polarization is transferred into the semiconductor over the macroscopic distance (∼10 μm) leading to the dynamical proximity effect (section 3). The main part of the paper (section 4) discusses the physics of the feedback of semiconductor charge carriers on the ferromagnetism (magnetic hysteresis loop, stray fields, Curie temperature) of the entire hybrid. The feedback opens up the possibility of tuning the magnetism via the optical excitation of charge carriers in a nearby semiconductor. As a result, the FM/SC hybrid represents an optically controlled elementary magnetic cell.

2. Zeeman interaction in the FM/SC hybrid

An electron with spin $\vec{s}$ and magnetic moment $-\mu_B g \vec{s}$ interacts with an external magnetic field $\vec{H}$. The Hamiltonian of Zeeman interaction

$$\hat{H}_Z = -\vec{\mu} \cdot \hat{\vec{H}} = \mu_B g \vec{s} \cdot \hat{\vec{H}},$$

where $\mu_B = |\mu_B| = 6.0 \mu\text{B}$ is the Bohr magnetron and $g$ is the electron’s $g$-factor. The interaction leads to the spin precession around $\vec{H}$ with a Larmor frequency, $\Omega = \mu_B g \vec{H}/\hbar$, according to the equation of motion $\vec{s} = [\Omega \times \vec{s}]$. The spin precession of particles is also affected by the local magnetic fields of other sources, and in general the total field is the vector sum of the fields.

2.1. Magnetostatic stray fields created by the ferromagnetic film in a semiconductor

Local stray magnetic fields generated by a FM film affect the semiconductor electron spin similarly to the external magnetic field. The distribution of stray fields in space depends strongly on the magnetic state of the FM film. The magnetic field outside the uniformly magnetized laterally infinite film is zero because the magnetic lines of force close at infinity. The field of finite lateral size $L$ creates (figure 1(a)) a magnetic field $H_A \approx 4\pi M d/L$ at point A ($d$ is the film thickness, $M$ is the magnetization). It penetrates deep $\sim L$ into the SC. For a Ni film with $M = 510$ Oe, $d = 10$ nm and $L = 0.1$ cm, the field $H_A \approx 0.06$ Oe. The stray field near the FM/SC interface is the strongest when the film is demagnetized, i.e. broken into a large number of domains with different orientations of magnetization. In this case both the direction and strength of the magnetic field vary in space with the characteristic scale determined by the domain size $\ell$ (figure 1(b)). For $\ell = 10 \mu$m, the field $H_A \approx 6$ Oe is hundred times larger than that of the uniformly magnetized FM. However, it decays depthward much faster—over the length $\gg \ell$.

2.2. Effect of stray fields on the optical orientation of semiconductor electrons

Dzhioev et al. (1994, 1995) observed the spin precession of SC electrons in static stray fields in a Ni/n-GaAs hybrid by the optical orientation method. The essence of optical orientation (Meier and Zakharchenya 1984) is the generation of spin-polarized conduction band electrons by circularly polarized light transferring the photon angular momentum into the spin system of semiconductor electrons. In turn, annihilation of the electrons emits circularly polarized light. The degree $\rho$ of the circular polarization of photoluminescence (PL) is equal to the projection of the electron ensemble-averaged spin $\vec{s}$ onto the registration direction $z$, usually coinciding with the normal to the structure plane (top of figure 2). An external magnetic field induces the precession of each electron spin around $\vec{H}$ with Larmor frequency $\Omega$. Under steady state conditions, however, the average spin does not change with time but deflects from the initial direction ($z$) decreasing the absolute value (figure 2). This leads to the Hanle effect—depolarization of PL with the magnetic field. The halfwidth $H_{1/2}$ of the magnetic depolarization curve is determined by the equality of frequency $\mu_B g H_{1/2}/\hbar$ and reciprocal non-equilibrium spin lifetime $1/T_s$. The longer the spin lifetime $T_s$, the smaller the magnetic field necessary to rotate spin and depolarize PL. As a result, we have the Hanle effect-based optical magnetometer whose sensitivity is determined by the $H_{1/2}$ value. As we discussed in subsection 2.1, the amplitude of the stray fields is pretty small (∼1 G), so the halfwidth should be narrow.
enough. Such a narrow halfwidth is realized in n-GaAs samples (see the review by Kavokin (2008) in this issue). Figure 3(a) compares the Hanle effects in n-GaAs when the film is previously magnetized (filled circles) and demagnetized (open circles). It is seen that the demagnetization decreases the degree $\rho$ with the maximum difference being achieved in zero external magnetic field.

The magnetic field strength $h_c$ making the magnetic moment of the sample vanish is referred to as the coercive force. It can be estimated by measuring the zero-field polarization $\rho(H = 0)$ after flipping a previously magnetized FM film by an external magnetic field of strength $H^*$. When the switching field $H^*$ is equal to the field $h_c$, the film is demagnetized and the polarization value $\rho(H = 0)$ is minimal. The data points for the upper dependence in figure 3(b) were measured after switching in the dark. The sharp dip corresponds to the coercivity $h_c = 90$ Oe. Indeed, the Hanle curve in the minimum coincides with the curve (open circles) in figure 3(a) for the demagnetized structure. In turn, the Hanle curve out of the dip coincides with the curve in figure 3(a) for the magnetized sample (filled circles). Remarkably, the coercive force value appeared to be 2.5 times larger than the coercivity $H_c \approx 40$ Oe measured in the same structure with the use of a superconductor quantum interference device (SQUID). The difference between the two techniques is that SQUID registers the total magnetic moment of the structure. It is mainly the magnetic moment of the nickel film whose thickness (40 nm) is much larger than that of the interface NiGaAs (a few nm). However, the contribution of the stray fields of nickel and the interface to the Hanle effect is not reduced to the sum of their magnetic moments. Indeed, Dzhioev et al (1997) have shown that the non-equilibrium electron spin diffuses into n-GaAs over the distance $L_s \approx 10 \mu m$, which is ten times longer than the 1 $\mu m$ size (Bochi et al 1995) of the nickel film domain. Therefore, the nickel stray fields decay quickly near the surface, and the basic mass of electrons does not ‘feel’ them. In contrast, the interface stray fields penetrate deep $\gg L_s$ into GaAs and dephase electron spins inside. Therefore, the Hanle effect magnetometer detects the FM interface NiGaAs rather than the Ni film itself due to the space selection of their stray fields.

The bottom curve in figure 3(b) was measured under illumination by a He–Ne laser. The position of the dip was shifted by 45 Oe. Thus, the coercivity of the interface decreased twice. This experiment appeared to be the first example of the optical control of ferromagnetism in FM/SC hybrids. It is discussed in detail in section 4.

To detect the small stray field $\sim 1$ Oe Dzhioev et al (1995) used n-type GaAs where at low temperature ($T = 4.2$ K) the halfwidth $H_{1/2} = 2$ Oe corresponds to a very long optical orientation lifetime $T_1 = 130$ ns. However, the next attempt (Crowell et al 1997) using the semi-magnetic semiconductor ZnCdMnSe (instead of GaAs) with a giant $g$-factor $g \approx 100$ was unsuccessful because the lifetime $T_1 = 15$ ps was too short, so $H_{1/2} \sim 1$ kOe. Recently, Meier et al (2006) have used electrons with a longer spin lifetime $T_1 \approx 1.5$ ns in the InGaAs SC quantum well to successfully observe the stray fields from Fe patterned stripes, where the stray fields were enhanced up to $\sim 200$ Oe.

3. Exchange interaction in FM/SC hybrids

3.1. Exchange interaction of magnetic atoms with semiconductor charge carriers at the FM/SC interface

The exchange interaction between magnetic atoms and semiconductor charge carriers is very important in FM/SC hybrids. Tunneling of SC carriers through the interface induces the equilibrium proximity effect—spontaneous ordering of electron spins localized near the interface from the semiconductor side. Equilibrium spin polarization decays into the semiconductor over the length of the order of the de
3.2. Equilibrium proximity effect

The proximity effect in the insulating FM/SC system was considered theoretically by Korenev (1996, 1997). Figure 4(a) shows a contact between a FM insulator and a semiconductor with t-electrons, i.e. the electrons trapped on deep isolated centers near the FM/SC interface. Tunneling into the ferromagnet leads to their exchange interaction with FM atoms and splits the states of t-electrons with spins along (↑) and opposite (↓) to the magnetization \( \vec{M} \). The energy (per unit area) of the exchange interaction between t-electrons with surface concentration \( n_t \) and mean spin \( \vec{S}_t \), and the uniformly magnetized ferromagnet (see the appendix) is

\[
E_{ex} = -J n_t \vec{S}_t \cdot \vec{m} = -\hbar n_t \vec{S}_t \cdot \vec{\omega},
\]

where \( J \) is the exchange constant determined by the FM/SC wavefunction overlap\(^1\) and \( \vec{m} \) is the unit vector along \( \vec{M} \). Exchange interaction (3.1) is equivalent to the interaction of each t-electron spin with an effective magnetic field collinear to \( \vec{m} \), and the vector \( \vec{\omega} = J \vec{m} / \hbar \) is the Larmor precession frequency of the t-electron spin in this field. In equilibrium the mean spin polarization of t-electrons at temperature \( T \),

\[
\vec{S}_t = \frac{1}{2} \frac{\vec{\omega}}{\omega} \tanh \left( \frac{\hbar \omega}{2kT} \right) = \vec{S}_t^{eq},
\]

directed along \( \vec{\omega} \).

Wavefunctions of t-electrons penetrate deep into the FM in the case of contact with the metallic FM film (figure 4(b)). This induces the broadening of the split ↑ (↓) state by the \( \gamma_+ (\gamma_-) \) value, respectively. McGuire et al (2004) calculated the parameters \( J \), \( \gamma_+ \), and \( \gamma_- \) for the two-dimensional electron gas near the FM metal for different barriers at the interface. They found that the proximity effect is basically determined by

\[ 1 \quad \text{Constant } J \text{ can have any sign in spite of the ferromagnetic ordering inside the FM.} \]

3.3. Optically induced spin polarization of semiconductor electrons near the FM/SC interface

3.3.1. Dynamical proximity effect in insulating FM/SC hybrids. The optical orientation of t-electrons in the insulating FM/SC (figure 4(a)) was considered theoretically by Korenev (1997). The illumination of the hybrid by circularly polarized light with the energy \( h\nu \) of quanta greater than the SC energy gap \( E_g \) leads to the optical orientation of semiconductor conduction band electrons. It is assumed that the band carriers do not interact with the ferromagnet, so only the t-electrons, localized near the interface, undergo the exchange interaction with magnetic atoms. Under these conditions the mean spin \( \vec{S}_t \) of the electrons is parallel to the exciting beam and is determined by its helicity. The valence band holes are depolarized due to their fast spin relaxation. They recombine with t-electrons. The optically oriented conduction band electrons are trapped by the surface states, thus replacing the recombined t-electrons. As a result, the non-equilibrium polarization of t-electrons appears. The steady state mean spin \( \vec{S}_t \) obeys the Bloch equation

\[
\frac{\vec{S}_t - \vec{S}_t^{eq}}{\tau_s} + \frac{\vec{S}_t^{eq} - \vec{S}_t}{\tau_e} + \vec{S}_t \times \vec{\omega} = 0.
\]
The first term in equation (3.3) describes the generation of spin compensated by recombination with characteristic time \( \tau_s \). The second term takes into account the spin relaxation during time \( \tau_s \) to equilibrium, whereas the last term gives the precession with the Larmor frequency \( \tilde{\omega} \) around the exchange field of magnetic atoms. The solution of equation (3.3) is

\[
\tilde{S}_t = \frac{\tilde{S}_0}{\tau_s} + \frac{\tilde{S}_0 \times \tilde{\omega} T_s}{1 + \tilde{\omega}^2 T_s^2} + \frac{\tilde{\omega} \times [\tilde{\omega} \times \tilde{S}_0]}{1 + \tilde{\omega}^2 T_s^2} \]

(3.4)

where \( \tilde{S}_0 = \frac{\tilde{S}_0}{\tau_s} \); the optical orientation lifetime \( T_s \) is determined by the shortest of the lifetime and the spin relaxation time: \( 1/T_s = 1/\tau_s + 1/\tau_s \). It follows from equation (3.4) that even the non-polarized excitation \( (\tilde{S}_0 = \tilde{S}_c = 0) \) decreases the mean spin \( \tilde{S}_c \) due to the recombination of polarized electron-t-electrons with non-polarized ones. Thus, the non-polarized excitation decouples FM and SC.

In the case of the circularly polarized excitation, the mean spin of conduction band electrons \( \tilde{S}_c \) is not collinear to the magnetization \( \tilde{M} \). Then new components of the t-electron mean spin \( \tilde{S}_c \) appear: (i) component \( \tilde{S}_0 \) parallel to the conduction band electron spin \( \tilde{S}_c \), (ii) component \( \tilde{S}_0 \times \tilde{\omega} \) perpendicular to both \( \tilde{S}_c \) and \( \tilde{M} \), and (iii) component \( \tilde{\omega} \times [\tilde{\omega} \times \tilde{S}_0] \) directed from \( \tilde{S}_0 \) toward \( \tilde{M} \). The latter two components result from the precession of the non-equilibrium spin of t-electrons in the exchange field of the ferromagnet.

The possible role of optically oriented surface states has been discussed by Prins et al. (1996) in their experiments on the spin-dependent tunneling of spin-polarized electrons from GaAs into a FM Pt/Co multilayer sample.

### 3.3.2. Dynamical proximity effect in metallic FM/SC hybrids.

In the previous subsection we assumed that the proximity effect is absent for the conduction band electrons. However, if the Schottky barrier is transparent (for example due to the intentional doping of the semiconductor) the free electrons reach the interface and undergo exchange interaction with magnetic atoms. In this case, there is noticeable spin-dependent amplitude of transmission of the semiconductor electron into the ferromagnet. Similar to localized states the free electrons acquire the exchange field spin polarization over the length scale \( \sim \delta_{bh} \) due to the proximity effect with the FM film. However, out of equilibrium the conduction band electrons are able to transfer the non-equilibrium spin over the macroscopic distance into the semiconductor via drift–diffusion processes. Aronov and Pikus (1976) suggested to transfer the spin from the FM to the majority SC charge carriers by externally applied bias (earlier the possibility of the electrical spin orientation of minority SC carriers was discussed by Scifres et al. (1973)). The difference in electrochemical potentials of the FM (\( \mu_f \)) and SC (\( \mu_c \)) induces both the particle flux \( \tilde{q}_p \) (figure 5(a)) and the spin flux (current) due to the spin-dependent tunneling through the heterojunction. The efficiency of spin injection is determined by the processes at the FM/SC interface and is taken into account under the boundary condition for the normal component \( \tilde{n} \) of the spin current as proposed by Aronov and Pikus (1976):

\[
\tilde{J}^{\mu}_{st} = a \cdot \tilde{m} \]

(3.5)

The parameter \( a \) is determined by the exchange interaction near the FM/SC interface. It also depends on the applied bias (figure 5(a)). Electrical spin injection was first realized by Alvarado and Renaud (1992) and recently drew a lot of attention as reviewed by Zutic et al. (2004), Lou et al. (2007), and Jonker et al. (2007) are more recent examples.

The electrochemical potential difference can be created optically too. For example, absorption of non-polarized light creates SC electron–hole pairs whose distribution is determined by the quasi-Fermi levels for electrons \( \mu_e \) and holes \( \mu_h \) (figure 5(b)). The holes are attracted to the FM/SC interface, thus dragging the electrons. Part of the electrons transmits into FM; the other part reflects from the boundary. The bulk non-equilibrium spin polarization of SC electrons develops due to the spin-dependent extraction into the FM. The spin density \( \tilde{S} = n_c \tilde{S}_c \) of conduction band electrons with concentration \( n_c \) in the n-type SC is purely non-equilibrium because \( \tilde{S}_T = 0 \) in the bulk of the non-magnetic SC. The spin dynamics in the bulk of the SC can be described by Bloch equations taking into account diffusion, drift, spin relaxation and Larmor precession in an external magnetic field. The boundary condition looks similar to equation (3.5) where the parameter \( a \) is determined by the exchange interaction at the FM/SC interface and generation/recombination processes in the SC (figure 5(b)).

The optically induced spin polarization of SC electrons was observed by Kawakami et al. (2001) in MnAs/n-GaAs and GaMnAs/n-GaAs heterostructures. Under illumination by linearly polarized light electrons acquire a non-equilibrium spin density \( \tilde{S} \), whose direction is collinear to the magnetization \( \tilde{M} \) of the nearby FM. The vector \( \tilde{M} \) lies in the plane due to the demagnetization field, which excludes the possible role of the MCD effect. The spin density \( \tilde{S} \) inside a semiconductor undergoes Larmor precession around an external magnetic field \( \tilde{H} \). Epstein et al. (2003) studied an optically induced dynamical proximity effect combined with an externally applied bias. They found that the value of the optically induced spin density \( \tilde{S} \propto \tilde{m} \) varies strongly with the bias: it is maximal at a forward bias. This result can be understood from figure 5(b). The forward bias (+ at ferromagnet) rectifies the band bending, thus increasing the electron spin current through the FM/SC interface. It results in increased spin density. Ciuti et al. (2002)
theoretically considered this effect as resulting from spin-dependent reflection/transmission off/through the interface. McGuire et al (2004) pointed out the inherent relation between the dynamic proximity effect and the strength of the FM–SC coupling across the interface.

3.3.3. Dynamic proximity effect for the non-collinear spins of the SC and FM at the interface. Consider the case when the spin density $\mathbf{S}$ near the FM/SC boundary is non-collinear to magnetization $\mathbf{M}$. It appears, for example, under optical orientation conditions, i.e. excitation by circular polarized light (figure 1). Optically oriented electrons reach the FM/SC boundary, partly going through (reflecting from) it with the change of their initial spin orientation. This problem reminds us of the well-known problem of scattering of the spin-polarized electron beam by a magnetic target (Kessler 1985). Besides the components $\mathbf{m}$ and $\mathbf{m} \times \mathbf{S}$ along magnetization, the reflected electrons acquire transverse components $\mathbf{m} \times \mathbf{S}$ and $\mathbf{m} \times (\mathbf{m} \times \mathbf{S})$, which describe the rotation of spin $\mathbf{S}$ around an effective exchange magnetic field and relaxation of $\mathbf{S}$ toward $\mathbf{M}$, respectively. It is reasonable to apply the well-known methods exploiting the density matrix to the FM/SC problem. Ciuti et al (2002) calculated the spin-dependent reflection coefficients for the FM/SC heterojunction within the effective mass approach. Earlier Brataas et al (2000, 2001) considered the spin transport through the paramagnet metal–FM junction for non-collinear polarizations. They deduced the boundary condition for the spin current:

$$
\mathbf{j}_n^S = a \cdot \mathbf{m} + b(\mathbf{m} \cdot \mathbf{S})\mathbf{m} + c[\mathbf{m} \times \mathbf{S}] + r[\mathbf{m} \times (\mathbf{m} \times \mathbf{S})]
$$

(3.6)

where the parameters $a$ and $b$ are determined by the spin-dependent conductance $g_1$ ($g_2$) for the electron spins along (opposite to) the FM magnetization, and the parameter $c$ ($r$) is given by the imaginary (real) part of the so-called mixing conductance ($g_1g_2$). The last two terms are similar to those in equation (3.4) for the insulating FM/SC hybrid (subsubsection 3.3.1).

Strictly speaking, not only the spin current but also the charge current through the FM/SC boundary is spin dependent. Johnson and Silsbee (1985) argued that the electron current at the FM–paramagnetic metal interface

$$
J = (g_1 + g_2)[\Delta \xi + \alpha(\mathbf{m} \cdot \mathbf{S})]
$$

(3.7)

determined not only by the electrochemical potential difference $\Delta \xi$ but also by the projection of $\mathbf{S}$ onto $\mathbf{M}$.

An experimental study of the effects given by the first term in equation (3.6) was discussed in subsubsection 3.3.2. To the best of my knowledge, no experimental evidence of the last three terms in equation (3.6) has been found in ferromagnet–semiconductor systems. Tserkovnyak et al (2005) reviewed both theoretical and experimental studies of all the terms in equation (3.6) in metallic FM–normal metal structures. The coefficients $a$, $b$ and $r$ are comparable. However, the coefficient $c$ is negligible in these systems due to dephasing: electrons enter the FM at different angles, thus spending different times to reach a given point inside the FM; as a result of precession in the FM exchange field they acquire different phases, which add destructively.

The spin-dependent charge current of optically oriented electrons, equation (3.7), was studied by Prins et al (1996), Isakov et al (2001) and Hirohata et al (2001). The authors modulated the spin density $\mathbf{S}$ of SC electrons via modulation of the helicity of photons. A magnetic field perpendicular to the structure plane brought the magnetization out of the film plane and induced the non-zero projection $(\mathbf{S} \cdot \mathbf{m}) \neq 0$. The signal of photocurrent through the interface was detected. At the same time, Prins et al (1996) and Isakov et al (2001) stated the strong influence of the MCD effect, i.e. the difference in absorption coefficients $\Delta \alpha \propto (\mathbf{S} \cdot \mathbf{m})$ of the left and right circular polarizations. Hence care should be taken for the quantitative interpretation of the results.

3.3.4. Dynamics of the dynamic proximity effect. Epstein et al (2002) observed the two-step dynamics of the nonequilibrium spin polarization of SC electrons due to the dynamic proximity effect. The electron spin polarization appears during the first 50 ps after excitation by linearly polarized laser pulse—the first step. The second step is the long-term spin relaxation of the order of nanoseconds. Bauer et al (2004) carried out calculation of the two-step dynamics for the case of the uniform spatial distribution of the electron spin inside the SC, when the drift–diffusion length is larger than the SC thickness. They found that the first step results from the fast withdrawal of photo-holes toward the interface under the action of a built-up electric field (figure 5(b)). In turn, this induces the spin-dependent transfer of electrons into the ferromagnet (spin extraction) accompanied by the accumulation of the non-equilibrium spin in the semiconductor. The non-equilibrium electron spin survives tens of nanoseconds at low temperature in n-type GaAs-based semiconductors (Dzhioev et al 1997). This explains the long-term stage of the spin dynamics. Gridnev (2007) calculated the nanosecond dynamics with the use of equation (3.6) for the case of a thick semiconductor when the spin density $\mathbf{S}$ is spatially non-uniform. He found the non-uniform distribution of spin density due to the Larmor precession and spin relaxation on its way into the interior of the semiconductor.

4. Optical control of ferromagnetism in the FM/SC hybrid

It is well known that light, besides trivial heating, exerts a non-thermal influence upon the magnetic properties of FM semiconductors (Nagaev 1988), such as Curie temperature, magnetic hysteresis loop, magnetic anisotropy etc. Recently, similar phenomena have been reported in new GaAs-based FM semiconductors (Wang et al 2005). This section considers the physics underlying the optical control of ferromagnetism specifically for the FM/SC hybrids: magnetism of the hybrid (magnetic hysteresis loop and the orientation of magnetization vector in space) is tuned optically with the help of the semiconductor. The electrical manipulation of ferromagnetism was considered by Zakharchenya and Korenev (2005).
4.1. Increase of the exchange stiffness coefficient of the FM/SC with an electron accumulation layer

In the case of an electron accumulation layer, the exchange interaction of magnetic atoms with SC electrons is isotropic. Korenev (1996) considered the exchange interaction between magnetic atoms and SC electrons localized at deep centers near the FM/SC interface (subsection 3.2, figure 4(a)). Exchange favors the collinear orientation of the t-electron spin and the magnetic atom spins located within the localization area of the t-electron. The local enhancement of the FM ordering increases the exchange rigidity coefficient of the hybrid in equilibrium.

Quantitative analysis is based on equation (3.1) that should be modified in the non-uniform case (Korenev 1996, 1997). Assume that the magnetization \( \vec{m}(x) \) changes orientation in one direction \( x \) over the distance \( \delta \) (domain wall thickness) much larger than the localization radius \( a_t \). Within the macroscopic approach (see the appendix) the exchange energy per unit surface area is

\[
E_{ex}(x) = -\hbar n_t(x) \vec{S}_t(x) \cdot \vec{w}(x)
\]

(4.1)

where the frequency

\[
\vec{w}(x) = \frac{J}{\hbar} \left[ \vec{m}(x) + \frac{a_t^2}{2} \vec{m}_n(x) \right]
\]

(4.2)

contains in comparison with equation (3.1) an additional term with the second space derivative. It results from the smooth variation of \( \vec{m}(x) \) within the localization area \( \sim \pi a_t^2 \) and represents the first nontrivial term of expansion of the exchange in series in powers \( (a_t/\delta)^2 \ll 1 \). As a result, the mean spin of t-electrons also depends on \( x \). Equation (3.2) gives its equilibrium value with vector \( \vec{w}(x) \) being taken from equation (4.2). At low temperature, the mean spin \( \vec{S}_t(x) = \vec{w}(x)/2\omega(x) \). Substituting \( \vec{S}_t(x) \) and equation (4.2) into equation (4.1) one finds

\[
E_{ex}(x) = \varepsilon_0 + \frac{A_1}{2} \left( \frac{\partial \vec{m}}{\partial x} \right)^2
\]

(4.3)

where \( \varepsilon_0 = -|J|n_t/2 \) does not depend on the distribution of magnetization in space and the exchange stiffness coefficient \( A_1 = |J|n_t a_t^2/2 \); the identity \( \vec{m} \cdot \vec{m}_n = -(\vec{m}_n)^2 \) was used. Equation (4.3) shows that the exchange interaction increases the exchange stiffness of the hybrid by the \( A_1 \) value in agreement with qualitative discussion.

The increase of the exchange stiffness coefficient gives an additional contribution to the coercivity of the hybrid. However let us recall first the origin of coercivity.

4.2. Origin of coercivity (Chikazumi 1984)

The width of the magnetic hysteresis loop is often determined by the displacement of the domain walls separating adjacent domains. Space fluctuations of the anisotropy constant \( \Delta K(x) \) or the exchange stiffness coefficient \( \Delta A(x) \) about their average values \( K_0 \) and \( A_0 \) induce the fluctuations of the domain wall energy \( y(x) \sim \sqrt{[A_0 + \Delta A(x)]/K_0 + \Delta K(x)} \). The potential profile \( y(x) \) fixes the domain walls giving the source of coercivity. The fluctuation \( \Delta K(\Delta A) \) can be much less than the averaged constant \( K_0(A_0) \), and no visible change of the average magnetic anisotropy (exchange stiffness) takes place. However it is crucial for the coercivity. The coercivity \( h_c \) is determined by both the amplitude of fluctuations \( \Delta y \) and their characteristic length \( \Lambda \). An external magnetic field \( H \) exerts pressure \( P = 2MH \) on the 180°-domain wall. It is compensated by the pressure \( \frac{\partial y(x)}{\partial x} \sim \frac{\Delta K}{\Lambda} \) due to the fluctuations. The coercivity can be estimated by equating the pressure at \( H = h_c \) to the maximum restoring force \( 2Mh_c = \frac{\partial y(x)}{\partial x} \max \sim \frac{\Delta K}{\Lambda} \). We have taken into account that the maximum is reached when \( \Lambda \) is close to the domain wall thickness \( \delta = \sqrt{A_0/\pi K_0} \): pinning is negligible at \( \Lambda \gg \delta \), whereas the effect of fluctuations is averaged over the wall thickness at \( \Lambda \ll \delta \). We conclude that one should decrease the amplitude of the potential profile to decrease the coercivity. For small fluctuations \( (\Delta y \ll y) \), we estimate \( h_c \sim \Delta K/M \) for the fluctuations of the anisotropy constant and \( h_c \sim \Delta A/(M \cdot \delta^2) \) for the fluctuations of the exchange stiffness coefficient.

4.3. Optical control of the coercivity of the FM/SC with an electron accumulation layer

In the case of a two-dimensional electron accumulation layer, the exchange of magnetic atoms with SC electrons is isotropic and affects the exchange stiffness coefficient \( A_1 \) (subsection 4.1). The fluctuations \( \Delta n_t \) of the surface density \( n_t(x) \) of t-electrons create fluctuations \( \Delta A_1 = |J|\Delta n_t a_t^2/2 \). If \( \Delta n_t \sim n_t \), then the coercivity is enhanced in the dark by the \( \delta h_c(0) \sim A_1/M_{\text{surf}} \delta^2 \) value \( (M_{\text{surf}} = M \cdot d \) is the magnetic moment per unit surface area). Under illumination of the hybrid by light with power density \( W \) and photon energy \( h\nu > E_g \), the photo-excited holes move to the interface (figure 5(b)) and recombine with t-electrons, thus decreasing both \( n_t \) and coercivity (Korenev 1996):

\[
h_c(W) = h_0(0) + \frac{\delta h_c(0)}{1 + W/W_0},
\]

(4.4)

where \( W_0 \) is the characteristic power density depending on the absorption coefficient and the capture efficiencies of electrons and holes by t-centers; the parameter \( h_0 \) takes into account other contributions to coercivity.

The optical control of the coercivity of the FM/SC hybrid has been demonstrated experimentally by Dzhioev et al (1994, 1995) in a Ni/n-GaAs structure. We have already considered this system in subsection 2.2 and found that the optically oriented electrons represent a sensitive detector of weak stray fields of the Ni–GaAs interface rather than the Ni film itself. The important feature of the Ni/n-GaAs hybrid consists in the optical tunability of the interface ferromagnetism. Figure 6 shows that the illumination by the He–Ne laser \((h\nu = 1.96 \text{ eV})\) decreases the interface coercivity by half (compare also the lower dependence in figure 3(b) with the upper one). However, illumination does not affect the coercive force of the nickel film. The effect we called photocoercivity is not sensitive to the light polarization and takes place only in the illuminated region. Photocoercivity takes place at a low power density (a few mW cm⁻²) and is not related to the heating of the sample:
the heating of the hybrid by passing the dc electric current across the FM/SC heterojunction, with dissipated power being ten times larger than the power of light, kept the coercive force unchanged. Spectral measurements have shown that the photoexcitability is due to the effect of the semiconductor on the FM interface: it diminishes if the photon energy is below the energy gap of GaAs. The solid line in figure 6 is calculated with the use of equation (4.4) for $h_1 = \delta h_c(0) = 45$ Oe, $W_0 = 8$ mWcm$^{-2}$. In spite of the good agreement, a few problems remain for the future. The first is the origin of the states that localize electrons near the interface. Their energy-space location and concentration are hardly controllable. The best choice for future studies seems to be the quantum well structure (4.2) with gate control of the charge carrier concentration in the well. Another situation arises from the contact of the FM/SC hybrid the interface magnetism is strikingly different from what we understand by ‘interface’. However, I have no information on the research of magnetism of Ni-rich GaAs.

Calculations of Chantis et al (2007) have shown that even in the ideal (i.e. with an abrupt interface) Fe/GaAs hybrid the interface magnetism is strikingly different from that of the bulk Fe. Zhao et al (2007) observed interface ferromagnetism in the Fe/AlGaAs heterostructure by a time-resolved magnetization-induced second harmonic generation technique. This technique enables one to selectively detect interface magnetism due to the reduced symmetry of the interface. They found that the Fe interface ferromagnetism is decoupled from the bulk Fe film measured with the use of the Kerr effect. However, no effect of AlGaAs excitation on interface magnetism was reported.

4.4. Surface anisotropy in the FM/SC with a hole accumulation layer

Another situation arises from the contact of the FM/SC with a hole accumulation layer. For example, it takes place in inversion layers, with strong band bending (figure 7), or in the case of the p-type quantum well near FM. Size quantization leads to anisotropic exchange interaction (subsection 3.2) with the energy per unit area,

$$E_{ex} = -J_h n_h \delta S_z^2 \hbar \omega_z, \quad (4.5)$$

and frequency $\omega_z = J_h m_z / \hbar$. At low temperature, the holes are completely spin-polarized due to the proximity effect (subsection 3.2) with the mean spin $S_z^h = \omega_z / (\omega_z^2)$ The exchange energy is $E_{ex} = -J_h n_h \delta S_z^2 \hbar \omega_z$. One can see that the exchange interaction can be considered as a peculiar magnetic surface anisotropy with the easy axis being directed along the $\zeta$-axis (perpendicular anisotropy). This is in striking contrast with the case of an electron accumulation layer (subsection 4.1). If the exchange constant and hole concentration are strong enough, then the orientational transition is possible (Korenev 2003): the magnetic moment leaves the plane and becomes oriented along the normal.

4.5. Photocoercivity in the FM/SC with the hole accumulation layer

Similar to subsections 4.2 and 4.3, the fluctuations of exchange coupling between magnetic atoms and SC holes induce fluctuations of the anisotropy constant $\Delta K \sim J_h n_h$ increasing the coercivity in the dark by $\delta h_c(0) \sim J_h n_h / M_{sat}$. The illumination of the hybrid may decrease the exchange constant $J_h$ due to band flattening, thus decreasing the overlap of the hole wavefunctions with magnetic atoms (figure 7).

Oiwa et al (2001) demonstrated the optical control of the coercive force of the p-InMnAs FM grown on a nonmagnetic GaSb semiconductor. The authors observed that the photoinitiation of the hybrid decreased the coercive force. This effect was the strongest under illumination by light with photon energy larger than the energy gap of the nonmagnetic semiconductor GaSb, thus pointing to the important role of SC. Strong band bending in GaSb (similar to figure 7) takes place near the FM/SC interface in the dark. This leads to accumulation of a substantial number of holes in the vicinity of the interface. The authors explained the photocoercivity by accumulation of a substantial number of holes in the vicinity of the interface.
4.6. Photo-induced change of the Curie temperature

Light can affect the Curie temperature $T_C$ too. For example, the photo-excited carriers may change the exchange constants of ferromagnets. The light-induced increase in the Curie temperature $\Delta T_C$ is well known in FM semiconductors (Nagaev 1988). A simple estimation shows that this effect is pretty small: $\Delta T_C \approx (n/N_{\text{tr}})T_C \approx 0.1$ K if the concentration of magnetic atoms $N_{\text{tr}} \approx 10^{22}$ cm$^{-3}$, $T_C = 1000$ K and the concentration of photocarriers $n \approx 10^{18}$ cm$^{-3}$.

It is reasonable to expect a similar small change in the FM/SC hybrids. However, Koshihara et al. (1997) claimed the photo-induced FM order in the p-InMnAs/GaSb hybrid to be persistent up to 35 K. The authors measured the magnetization component normal to the sample plane versus the magnetic field with the use of SQUID and the anomalous Hall effect. They found non-hysteretic behavior of magnetization in the dark. In contrast, contrast with photon energy larger than the energy gap of GaSb induced clear hysteresis. The authors interpreted this effect as a paramagnet–ferromagnet transition of the magnetic semiconductor InMnAs due to the transfer of SC photo-holes into InMnAs FM. In other words, the change of ferromagnetism by the holes excited as due to the change of ferromagnetism by the holes excited in the GaSb and transferred into the InMnAs layer. Another explanation follows from the above discussion: one could take into account the weakening of the FM/SC exchange coupling near the interface.

4.7. Effect of circularly polarized light on magnetization

The circularly polarized light transfers the angular momentum $\pm h$ per photon. Hence its absorption magnetizes the sample. For example, it excites spin-oriented charge carriers (Meier and Zakharchenya 1984). Ordinarily, their concentration is much smaller than that of magnetic atoms, so the direct magnetization by light is inefficient. A stronger effect consists in the appearance of an effective magnetic field proportional to the degree $P_e$ of the circular polarization of light. If the field value is larger than coercivity, then the sample is magnetized even in the absence of the external magnetic field. Van der Ziel et al. (1965) proposed the inverse Faraday effect: the circularly polarized light acts on atoms in non-absorbing media as a magnetic field lifting Kramer’s degeneracy. This field is relatively weak: $H_{\text{eff}} \approx 0.01$ Oe at power density $10^7$ W cm$^{-2}$ for Er$^{2+}$:CaF$_2$. Therefore, optical pulses heating the magnetic system close to $T_C$ should perform the magnetization. Stanciu et al. (2007) reported the optical magnetization of the ferrimagnetic GdFeCo.

Effective magnetic fields can be created under absorption of circularly polarized light. In this case, optically oriented carriers undergo a strong exchange interaction with magnetic atoms. Merkulov and Samsonidze (1980) considered theoretically the domain wall motion under the action of circularly polarized light exciting the FM semiconductor with perpendicular anisotropy. The magnetic circular dichroism (MCD) effect leads to the optical orientation of carriers: a larger electron concentration is excited in domains with one orientation of $M$ with respect to the domains with the opposite $M$. Then the size of one type of domain increases at the expense of the others. Merkulov and Samsonidze (1980) solved the dynamic Landau–Lifshitz equation and found that the domain wall moves as if it were an effective magnetic field whose value and direction are determined by the helicity of light. Nagaev (1988) reviewed early experiments in FM semiconductors on this topic.

4.7.1. Effective magnetic fields in FM/SC with an electron accumulation layer. Korenev (1997) considered theoretically the action of circularly polarized light on the domain wall in the FM/SC hybrids. He argued that the effective field $\vec{H}_{\text{eff}}$ appears as a result of a pressure exerted by the optically oriented semiconductor electrons on the domain wall. Both the usual optical orientation of carriers in the non-magnetic semiconductor and the MCD effect result in the appearance of the effective field whose value and direction are determined by the angular momentum $h\vec{P}_e$ of the photon. Imagine a thin FM film with perpendicular anisotropy grown on the semiconductor surface with distribution $M(x)$ in a 180° wall varying in one direction $x$ (figure 8). Below we shall assume the usual distribution of $M(\vec{r})$ within the domain wall $m_x(x) = \cos \theta(x)$, $m_y(x) = \sin \theta(x)$, $m_z(x) = 0$, and $\cos \theta(x) = -\tanh(x/\delta)$ (Chikazumi 1984). Let the exchange interaction between magnetic atoms and SC t-electrons be small enough not to affect either the domain wall distribution or the thickness $\delta$. The domain wall centered at the position $x = x_0$ undergoes pressure due to the exchange interaction between magnetic atoms and SC electrons:

$$P = 2M_{\text{sat}}H_{\text{eff}} = -\frac{\partial}{\partial x_0} \left[ \int_{-\infty}^{+\infty} E_{\text{ex}}(x - x_0) \, dx \right]$$

$$= h \int_{-\infty}^{+\infty} \frac{\partial \delta(x - x_0)}{\partial x_0} \, dx$$

(4.6)
where the exchange energy per unit surface area $E_{ex}(x)$ and $\bar{\omega}(x)$ are given by equations (4.1) and (4.2), respectively. Space derivation in equation (4.6) should be performed under fixed $n_t \bar{S}_t$ in spite of the fact that it may (and really does) depend on $\bar{m}(x)$ distribution. This is because the calculation of force in open systems should be performed under fixed external conditions (Landau and Lifshits 1979). In our case, the spin system of t-electrons is an external system with respect to the FM one.

In equilibrium the force will be absent. Indeed, the substitution of the equilibrium value $\bar{S}_t$ from equation (3.2) into equation (4.6) gives zero. Hence, the non-equilibrium spin density of t-electrons $n_t \bar{S}_t$ should be calculated. In the MCD case, the mean electron spin is in equilibrium and completely spin-polarized at low temperature, whereas the concentration of t-electrons is different in different domains:

$$n_t(x) = n_t(1 + \gamma \bar{P}_t \cdot \bar{m}(x)), \quad \bar{S}_t(x) = \bar{\omega}(x)/2\omega(x). \quad (4.7)$$

The parameter $\gamma$ characterizes both the value and sign of dichroism. Substituting equation (4.7) into equation (4.6), we reproduce the Merkulov and Samsonidze (1980) result deduced from the Landau–Lifshitz dynamic equation:

$$H_{eff} = \gamma P_t A_1 / 3M_{surf} \delta^2. \quad (4.8a)$$

Alternatively, one may consider the case of optically oriented electrons when the non-equilibrium spin $\bar{S}_t$ is governed by equation (3.4), but the concentration $n_t = \text{const}(x)$ is the same in different domains (no MCD effect). Taking into account that only the projection $\bar{S}_t \cdot \bar{\omega}/\omega^2$ of the spin $\bar{S}_t$ of t-electrons is conserved in the case of strong exchange $\omega T_t \gg 1$, we obtain from equations (3.4) and (4.6)

$$H_{eff} = 2S_t T_s A_1 / T_t 3M_{surf} \delta^2. \quad (4.8b)$$

Note that expressions (4.8a) and (4.8b) for the field $\tilde{H}_{eff}$ contain a small parameter $\delta^2/\delta^2 \sim 10^{-2} - 10^{-3}$ (for typical values of localization radius $a_t \sim 1$ nm, and domain wall thickness $\delta \sim 30$ nm), so for the point defect $H_{eff} = 0$. This is the result of the flexibility of the semiconductor electron spin system adjusting the direction of the mean spin to $\bar{M}$.

4.7.2. Effective magnetic fields in FM/SC with a hole accumulation layer. The effective field value increases drastically if the direction of the t-electron spin is fixed; thus the spin precession in the ferromagnet exchange field is absent. Suppose that a p-type accumulation layer with hole concentration $n_h$ is formed in the semiconductor (or the quantum well filled with holes) near the FM/SC interface. Due to the reduced symmetry, the hole spin states are split into heavy and light hole doublets as discussed in subsections 3.2 and 4.4. Then the hole spin $\bar{S}_h = m_h/2$ is fixed along the $z$-direction. Taking into account the expression for the Larmor frequency $\omega_l = J_0 m_z(x)/\hbar$ and using equation (4.6), we get for the MCD case

$$H_{eff}^{h} = \gamma P_t J_0 m_h / 6M_{surf}, \quad (4.9a)$$

and for the case of the heavy hole optical orientation

$$H_{eff}^{h} = S_h T_s J_0 n_h / M_{surf}. \quad (4.9b)$$

One can see that the $H_{eff}^{h}$ field is indeed greatly $(\delta^2/a^2$ times) enhanced due to the rigidity of the hole spin system.

Let us estimate the $H_{eff}^{h}$ field value for the FM/SC coupling constant $J_0 = 0.1$ eV and the hole surface concentration $n_h = 10^{12}$ cm$^{-2}$. We take $M_{surf} \approx 2\mu_B N_{ion}d \approx 2 \times 10^{15}$ Bohr magnetons per unit area for the FM film thickness $d = 1$ nm and the concentration of FM atoms $N_{ion} \approx 10^{22}$ cm$^{-3}$. For the MCD case, we find from equation (4.9a) that $H_{eff}^{h} = 1400$ Oe under the favorable condition $\gamma = 1$ and for the $100\%$ circularly polarized light ($P_t = 1$). For the case of the optical orientation of holes, we get a value 4200 Oe for $2S_t T_s / T_t = 1$. We conclude that the exchange coupling with holes looks very promising for the optical control of ferromagnetism of the hybrid.

Oiwa et al (2002) observed the magnetization of the GaMnAs FM film by circularly polarized light in the GaMnAs/GaAs hybrid. The authors explain it by the photo-creation inside GaMnAs of spin-oriented holes, which dynamically polarize the Mn spins. However, a very sharp spectral dependence correlates with the excitation of paramagnetic GaAs rather than FM GaMnAs whose spectrum is very smooth due to strong disorder. This fact provides strong evidence of the crucial role of GaAs excitation in the optical magnetization of GaMnAs. Therefore, one could also consider the possible role of optically oriented holes in GaAs and their exchange with magnetic atoms as discussed in this section.

5. Summary

Spin–spin interactions in the FM/SC hybrid lead to a strongly coupled spin system of the ferromagnet and semiconductor. On the one hand, they induce the proximity effect—spin polarization of semiconductor electrons. Hence semiconductor electrons monitor the magnetic state of the ferromagnet. On the other hand, the magnetic properties of the unified system differ drastically from those of the FM film alone. The magnetism of the entire system can be controlled optically. As a result, the hybrid constitutes an elementary magnetic storage with the semiconductor being not only a substrate but also an active participant in information processing. An additional degree of freedom consisting in the choice of a desirable FM/SC pair among paramagnet semiconductors and a large number of FM materials provides many possibilities. The ultimate goal is the discovery of FM/SC hybrids with optical control of magnetism at room temperature. The most promising for this purpose seems to be the FM/SC system operating on the optically tunable proximity effect. The best practical choice could be related to the semiconductor quantum well grown near the ferromagnet with gate control of both the exchange coupling and the charge carrier concentration in the well.
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Appendix

Equations (3.1) and (4.1) can be derived from the Hamiltonian

\[ \hat{H} = \frac{1}{N_0} \sum_{i,j} \hat{S}_i \cdot \hat{S}_j \delta(\vec{r}_i - \vec{r}_j) \]

describing the isotropic exchange interaction between t-electrons and magnetic atoms with spin \( I \) and concentration \( N_0 \). Here \( \hat{S}_i \) and \( \hat{S}_j \) are the operators of spins of the \( i \)th t-electron and \( j \)th magnetic atom located at \( \vec{r}_i \) and \( \vec{r}_j \), respectively. Deep centers are assumed to be isolated \((n_i a_i^2 \ll 1)\).

Assume that the localization radius of t-electron \( a_i \) is much larger than the distance \( a_i \) between magnetic atoms. In this case ferromagnet can be considered as a continuous medium with macroscopic magnetization \( \vec{M}(\vec{r}) \). Replacing the spin density operator \( \hat{S} = \sum_i \hat{S}_i \cdot \hat{I}_i \delta(\vec{r}_i - \vec{r}_j) \) with its classical value \( \vec{S}(\vec{r}) \equiv -NI_0 \hat{m}(\vec{r}) \) (the sign is negative because the spin of the magnetic atom is usually antiparallel to its magnetic moment) we arrive at the mean-field Hamiltonian

\[ \hat{H}_S = -J \sum_i \vec{S}_i \cdot \vec{m}(\vec{r}_i) \]

by averaging the mean-field Hamiltonian with the ground state orbital wavefunction \( \Phi_0(\vec{r}) \) of the t-electron with coordinate \( \vec{r} \) located near the \( i \)th center. It is reasonable to consider the magnetization to be a slowly varying function, because the characteristic distance \( \delta \approx 30 \text{ nm} \) is much larger than \( a_i \approx 1 \text{ nm} \). Then the integral in equation (A.1) can be expanded in series in powers \((a_i/\delta)\). If the magnetization \( \vec{m}(x) \) varies only along the \( x \)-direction, then we have up to the second-order terms

\[ \hat{H}_S = -J \sum_i \vec{S}_i \cdot \left( \vec{m}(x_i) + \frac{a_i^2}{2} \vec{m}''(x_i) \right) \]

(A.2)

Here magnetization and its second derivative are taken at the position \( x_i \) of the \( i \)th center. Localization radius is determined by the relation \( a_i^2 \equiv \int (x - x_i)^2 \Phi_i^2(\vec{r}) \, d\vec{r} \). Equation (A.2) has a form \( \hat{H}_S = h \sum_i \vec{S}_i \cdot \vec{a}_i \) where the vector \( \vec{a}_i \) is given by equation (4.2) and summing over all paramagnetic centers. Introducing the spin density operator of t-electrons \( \vec{S}(\vec{r}) \equiv \sum_i \vec{S}_i \delta(\vec{r} - \vec{r}_i) \) we can present it in the equivalent form

\[ \hat{H}_S = h \int \vec{m}(\vec{r}) \cdot \vec{S}(\vec{r}) \, d^3\vec{r} \]

(A.3)

We can further treat the problem macroscopically if the number of centers within the characteristic area \( \geq \delta^2 \) is very large: \( n_1 \delta^2 \gg 1 \). Replacing in this case the spin density operator \( \vec{S}(\vec{r}) \) by its classical value \( n_i(\vec{r}) \vec{S}_i(\vec{r}) \), we arrive at the quasiclassical expression for the total interaction energy:

\[ E_{\text{Total}} = \int E_{\text{exc}}(\vec{r}) \, d^3\vec{r} = h \int n_i(\vec{r}) \vec{S}_i(\vec{r}) \cdot \vec{a}(\vec{r}) \, d^3\vec{r} \]

(A.4)

This justifies equation (4.1) for the exchange energy per unit area \( E_{\text{exc}}(\vec{r}) \), which reduces to equation (3.1) for the uniform \( \vec{M} \).

The limits of applicability of the macroscopic description bring about the inequality \( 1/a_i^2 \gg 1/a_j^2 \gg n_i \gg 1/\delta^2 \). For \( a_i = 3 \times 10^{-8} \text{ cm} \), \( a_j = 10^{-7} \text{ cm} \) and \( \delta = 3 \times 10^{-6} \text{ cm} \), it is satisfied in the wide concentration range \( n_i \in [10^{13} \ldots 10^{14}] \text{ cm}^{-2} \).
