TOPICAL REVIEW

Magnetic nanostructures

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Abstract

Characteristic results of magnetism in small particles, thin films and tunnel junctions are presented. As a consequence of the reduced atomic coordination in small clusters and thin films the electronic states and density of states are modified. Thus, magnetic moments and magnetization are affected. Generally, in clusters and thin films magnetic anisotropy plays a special role. In tunnel junctions the interplay of magnetism, spin currents and superconductivity are of particular interest. In ring-like mesoscopic systems Aharonov–Bohm-induced currents are studied. Results are given for single transition metal clusters, cluster ensembles, thin films, mesoscopic structures and tunnel systems.

(Some figures in this article are in colour only in the electronic version)
In general, the topology of the nanostructure affects strongly the electronic structure and the orientation of the magnetization, its domain structure and magnetic relaxation. The electron energy spectrum gets discrete and quantum well states (QWS) occur in thin films. Note, in nanostructured thin films a magnetic domain structure is frequently present. The size of the domains and their relaxation depends on the atomic structure of the film, anisotropy and temperature. The reversal of a domain magnetization may be relatively slow, but may speed up at nonequilibrium, for example, due to hot electrons.

In growing thin films interesting nonequilibrium behavior may result from the interdependence of atomic morphology of the nanostructure and its magnetic domain formation. This may be seen by observing time-resolved properties of growing nanostructures. Thus, it is of interest to study, time-resolved, the occurrence of uniform ferromagnetism in thin films as a function of film thickness and domain density and size, in general, on growth conditions for producing nanostructures [7, 8].

The electronic structure of nanostructures and mesoscopic structures can frequently be described approximately by a semiclassical Balian–Bloch-type theory. A few electronic orbitals close to the Fermi energy may determine predominantly the electronic structure. Then, as has been shown by extensions of such a theory by Stampfl, the electronic properties may be described rather well [16]. Besides other applications this offers interesting studies of Aharonov–Bohm effect-induced electric currents into mesoscopic ring structures of metals (and electrolytes). Note, in the case of ferromagnetic metals, such induced currents may be spin-polarized.

For tunneling the interplay of magnetism and superconductivity is of particular interest [10, 12, 17, 18]. The interface of singlet and triplet superconductors (the latter possibly caused by magnetism) is interesting. Furthermore, spin-polarized currents and Aharonov–Bohm effect-induced currents need to be studied and promise important applications. Between two magnets one expects, as a result of the spin continuity equation and the Landau–Lifshitz equation, Josephson-like spin currents [12].

In the following magnetic nanostructures having different geometry and tunneling are discussed. We focus mainly on characteristic results obtained during recent years by Bennemann et al. For comparison important results of other studies are referred to, see, for example, for clusters Sugano [3], Jellinek [5], for films Jensen [6, 7, 19–28], molecular-field-type theory for magnetism see Moran-Lopez et al. [29], for nonequilibrium behavior see Jensen [30, 31], for magnetooptics see the book Nonlinear Optics in Metals [32], for tunnel junctions studying the interplay of magnetism and superconductivity see Manske, Morr et al. [10, 11, 13, 14], for nonequilibrium physics Bennemann et al. [33] and for induced currents into mesoscopic rings via Faraday induction law (or the Aharonov–Bohm effect) see Bloch, Landaue, Althshuler et al. [34–37]. Photon-assisted tunneling deserves special attention, since it offers interesting quantum mechanical behavior, interference effects and nonequilibrium behavior, see Garcia and others [14]. In general, the discretization of the electron energy spectrum in small systems should exhibit interesting confinement effects.

### I.1. Magnetic clusters

While small clusters may have a complicated atomic structure, this will tend to become bulk-like for larger ones. Hence, approximately one may assume first liquid-like structures as the number of atoms \( N \) in the cluster increases and then bulk-like structures for larger clusters. The cluster volume \( V \) is given by \( V \sim N^3 \) and the surface area by \( S \sim N^{2/3} \) [1–5].

In figure 1 fcc- and bcc-like cluster structures are shown. These may approximate magnetic transition metal clusters. In such clusters DOS and magnetic moments are site-dependent.

In figure 2 the discretization of the electron energy spectrum of such clusters is illustrated. Levels are occupied up to the Fermi level \( \varepsilon_F \) and interesting odd and even effects occur as a function of cluster size (number \( N \) of atoms).

The magnetism of the cluster is characterized by its atomic shell \((i)\)-dependent magnetic moments \( \mu_i(N) \), by the magnetization \( M(T, N) \) and the Curie temperature \( T_c(N) \). These magnetic properties may be calculated using electronic theory and, for example, a tight-binding Hubbard-like Hamiltonian, or on a more phenomenological level a Heisenberg Hamiltonian including magnetic anisotropy. Thus, one may estimate the Curie temperature from

\[
T_c \sim a J q_{eff}(N),
\]

where \( a \) is a fitting parameter, \( J \) the interatomic exchange coupling integral and \( q_{eff} \) the effective coordination number [1, 2].

In figure 2 size effects for the electronic structure are sketched. Note, the screening of Coulombic interactions and
arranged on a lattice is shown. In general, the lattice sites
formation of magnetic moments, magnetization, size of
quantum dots, in particular magnetism and superconductivity
discretization might affect many properties in grains and
Ni
the BCS–BEC transition in superfluids.

between Cooper pairs in the case of superconductivity or, in general,
local moment magnetism, the size of Cooper pairs versus distance
discretization of the energy spectrum should affect itinerant versus
reflect this, in particular magnetism and superconductivity. The

n
affects magnetic activity. Also, the spacing of the
width of d-electron states varies with cluster size and thus
affects magnetic activity. Also, the spacing of the n electronic states is
\[
\delta \approx \varepsilon_F/n \approx (\hbar v_F/R)(k_F R)^{-2}(N(0)V)^{-1}.
\]
Here, \( \varepsilon_F \) is the Fermi energy and \( v_F \) and \( k_F \) the corresponding Fermi velocity and wavevector. \( V \) refers to the volume and \( N(0) \) to the density of states (DOS) at \( \varepsilon_F \). One expects for \( T > \delta \) that the discretization plays no great role. However, discretization might affect many properties in grains and quantum dots, in particular magnetism and superconductivity (formation of magnetic moments, magnetization, size of Cooper pairs, superfluid density, etc).

The local electron density of states of atom \( i \) is given by \( N_i(\varepsilon, N) \) and generally spin-dependent \( (N_{i\sigma}(\varepsilon, N)) \). This determines the occurrence and size of local magnetic moments. Their direction is determined by magnetic anisotropy. Note, besides spin magnetism also orbital magnetism occurs and is typically enhanced compared with the bulk one. Also generally the orbital magnetism is dependent on atomic site.

Mie scattering by spherical small particles is an interesting classical physical phenomena. It is of general interest to extend this to include magnetism. This may offer another optical method to determine magnetism in small particles (of growing interest regarding, for example, high density magnetic recording media). One expects that magnetism leaves a characteristic fingerprint on the Mie scattering profile. The magnetic field of the incident light couples to the cluster magnetization. This affects the Mie scattering, in particular the backscattering intensity [15].

This effect of magnetism on Mie scattering needs be studied more. It offers, for example, an interesting alternative to relatively difficult cluster beam deflection experiments by a magnetic field regarding the study of small-particle magnetism.

In figure 3 an ensemble of clusters (or quantum dots) arranged on a lattice is shown. In general, the lattice sites may be occupied by ferromagnetic (or paramagnetic) clusters or may be empty. Dependent on the cluster pattern one gets rich physical behavior. For example, while the single clusters are ferromagnetic, for larger spacing between the clusters one might get (for higher temperature) no global magnetization of the whole cluster ensemble. For dense spacings of the grains, dots and sufficient strong interaction amongst them, local and also global magnetization may occur as a function of temperature \( T \). Note, typically, \( T_C(N) > T_C \) where \( i \) refers to the cluster \( i \) and \( T_C \) to the Curie temperature of the ensemble or cluster lattice. Of course, antiferromagnetism may also occur. Note, exchange and magnetic dipole coupling may occur between grains. Also, regarding the magnetic state of a single grain, superparamagnetism must be taken into account.

In general, interesting phase diagrams are expected for an ensemble of magnetic grains (quantum dots). One gets for interactions \( J > 0 \) and \( J < 0 \) ferromagnetism or antiferromagnetism for the grain ensemble. For a mixture of grains, for example a mixture of superconducting and ferromagnetic ones, one may observe interesting behavior as for alloys and one may get Josephson currents between the dots and due to electronic charge transfer, odd–even occupation effects, etc, see [17, 18]; see also Coulomb blockade effects and interesting dependence of tunnel magnetoresistance (TMR) on the structure of the tunnel junction by Majumdar, Fujimori and Maekawa [38, 39].

An interesting example of an important nanostructure is an ensemble of quantum dots, an anti-dot lattice for example. In figure 4, an anti-dot lattice immersed in a medium is sketched. Here, the anti-dots repel electrons moving, for example, between the quantum dots. An external magnetic field \( B \) will change, deforming the electron orbits spin-dependently and this may yield interesting behavior of the electronic properties of the system. Sensitive quantum mechanical interferences appear in such nanostructures, see Stampfli [16].

Currents between two quantum dots and assisted by an external electromagnetic field exhibit interesting behavior and
et al. The reorientation transition of the magnetization at radius \( r \) and thickness \( d \), see the discovery by Grünberg\ et al\ [6].

Better experiments; for details see [6]. The scaling law thickness of the ferromagnetic film is often described by the electronic density of states (DOS). These may be spin-dependent. The extension of the Balian–Bloch theory by Stampfli can be used to calculate the currents induced in the anti-dot lattice. Here, \( a \) denotes the spacing of the anti-dots with radius \( d \). The electrons move between the dots and are repelled by the anti-dot potential and thus selectively the polygonal paths 1, 2, 3, etc., yield the most important contribution to the spin-dependent DOS, magnetoresistance, etc. Details of the theory determining the electronic structure of such mesoscopic systems are given by Stampfli et al [16].

Figure 4. Magnetic field effects on polygonal electron paths in an anti-dot lattice. Here, \( a \) denotes the spacing of the anti-dots with radius \( d \). The electrons move between the dots and are repelled by the anti-dot potential and thus selectively the polygonal paths 1, 2, 3, etc., yield the most important contribution to the spin-dependent DOS, magnetoresistance, etc. Details of the theory determining the electronic structure of such mesoscopic systems are given by Stampfli et al [16].

Figure 5. Thin magnetic film structures are shown. Typical configurations are given. (a) Thin ferromagnetic film of thickness \( N \) on a substrate, (b) two ferromagnetic films (FM1, FM2) on a substrate and (c) multilayer film structure. The ferromagnetic films are separated by a nonmagnetic one (NM). At the surface the magnetic film may be covered by nonmagnetic material (cap). Of particular importance is the interplay of magnetism of magnetic thin films of a film multilayer system.

As indicated in figure 4, a few electronic orbits may determine mostly the electronic structure, near the Fermi energy, see the theory by Stampfli et al [16]. Quantum mechanical interference causes characteristic oscillations in the electronic density of states (DOS). These may be spin-dependent. The extension of the Balian–Bloch theory by Stampfli can be used to calculate the currents induced in mesoscopic rings, etc, via the Aharonov–Bohm effect [35–37].

In summary, these are some cluster-like nanostructures. Regarding magnetism transition metal and rare-earth atoms, metal oxides, etc, are particularly interesting. The dependence of magnetic spin, orbital moments and spin correlations on particle size and temperature are of interest. Magnetic moments, Curie temperature and magnetization control the magnetic behavior [1, 2, 6, 19, 22–27].

1.2. Magnetic films

For ultrathin films one may get magnetic behavior which could differ drastically from the one for thick ferromagnetic films, see Jensen et al [6]. The latter are approaching as a function of film thickness \( d \) the bulk behavior (b). For the Curie temperature one finds \( T_c(d) \rightarrow T_c(N) \) as film thickness \( N \) increases. The increase of \( T_c(N) \) for increasing thickness of the ferromagnetic film is often described by the scaling law \( \Delta T_c/T_{cb} \propto N^{-\lambda} \), or, more empirically, by \( \Delta T_c/T_c(N) \propto N^{-\lambda'} \) with non-universal exponent \( \lambda ' \) which fits better experiments; for details see [6].

Very important is the dependence of currents across multilayer film structures on the relative magnetic configuration of the thin films. Upon changing, for example, for neighboring Fe and Cr films the ferromagnetically aligned film magnetization (↑) to an AF one (↑↓) causes a giant increase in the resistivity (GMR), see the discovery by Grünberg et al [40] and Fert et al [41]. The reorientation transition of the magnetization at the surface of thin magnetic films is related. Also this may be important, for example, regarding high density magnetic recording. Clearly, magnetic coupling of films and of tunnel junctions exhibits related behavior [10–13]. Interplay of superconductivity and magnetism is sensitively reflected by tunneling [11]. Generally, quantum interference devices, switching devices for currents and their optical manipulation (hot electrons, photon-assisted tunneling, etc), and also topological manipulation of electronic structure (discrete spectrum rather than a continuous one) in nanostructures (see Peeters et al [42]), offer interesting options for applications.

Note, in ultrathin films, one or two atomic layers thick, ferromagnetism results from magnetic anisotropy (spin–orbit coupling, etc.) suppressing two-dimensional magnetic fluctuations [6] and causing the Mermin–Wagner theorem to become ineffective.

Various film structures are shown in figure 5. Neighboring magnetic films may order parallel or antiparallel. This can be affected optically too. As is well known the magnetic coupling in multifilm structures is very important and sensitively affects, in particular, transport properties and electric currents between the films, see the giant magnetoresistance (GMR) and its discovery by Grünberg and Fert and corresponding studies by others [40, 41]. Manipulation of the magnetic configuration and currents by external fields (magnetic or electric) is interesting and permits studies of nonequilibrium behavior. Of course, ultrathin magnetic film behavior is related to that of tunnel junctions of the magnetic material. In general spin-dependent electron DOS and electron scattering control the behavior of the electrons. The magnetic coupling between films may yield ferromagnetic, antiferromagnetic (AF) and disordered-like (quasi-as a compromise) spin order of the neighboring films or at the interface. Many studies have shown that exchange interaction and magnetic anisotropy are important. Note, particular interesting interfaces are Cr/Fe and
Effective magnetic field \( \mathbf{B} \) orientation of the magnetization. One expects that the density of the magnetic pattern increases for perpendicular orientation of the magnetization at the surface. Note, magnetic domain structure and domain size is affected by the orientation of the magnetization.

Figure 6. The reorientation transition \( M_\perp \to M_\parallel \) of the magnetization at the surface or interface of films is shown. This transition may be induced by temperature \( T \), increasing film thickness \( N \) and film morphology and external magnetic field \( \mathbf{B} \). One expects that the density of the magnetic pattern increases for perpendicular orientation of the magnetization at the surface. Note, magnetic domain structure and domain size is affected by the orientation of the magnetization.

Figure 7. Illustration of a magnetic stripe domain phase of a thin film. Neighboring domains are antiferromagnetically oriented and separated by Bloch-type domain walls. The magnetic structure is generally controlled by the film morphology and magnetic anisotropy. Of course, dependent on this, different domain structures may result, see \([6, 7, 25–27]\). Magnetic anisotropy controls the orientation of the magnetization, \( \mathbf{M}(T, d, \ldots) \), at the surface of the film. Dependent on temperature \( T \), film thickness, structure and effective magnetic field \( \mathbf{B} \) the orientation of the surface magnetization \( \mathbf{M} \) may change from perpendicular \( M_\perp \) to parallel \( M_\parallel \). This reorientation transition, important, for example, for magnetic patterns at surfaces, magnetic recording, is illustrated in figure 6 \([6, 25–27]\).

Obvious, the orientation of the magnetization at surfaces is related to the magnetic domain structure occurring in thin films. This is also clear from the illustration of domain structure in figure 7. In view of the GMR effect the magnetic domain structure strongly controls transport properties. Thus, for example, time-dependent domain relaxations should be reflected in the electrical resistivity.

As expected on general physical grounds, dependent on film growth conditions, one gets for thin film growth on a substrate a variety of nanostructures. This is illustrated in figure 8. For a growing film the accompanied magnetism may not be at equilibrium, but changes as the film topology changes. One has a nonequilibrium situation and magnetic structure changes due to magnetic relaxation of domains \([7, 8]\). The latter may be relatively slow. Various patterns of magnetic domains occur in general, see the results using STM and SEMPA spectroscopy \([8]\). Then a reversal of local domain magnetization occurs on a ps (picosecond) to ns (nanosecond) timescale and changes the global film magnetization as a function of time, see, for example, studies by \([31]\).

Similarly, as in the case of cluster ensembles on a lattice, the magnetic domains resulting in nanostructured films may be dense or separated by larger distances. Correspondingly, the domains are predominantly coupled via exchange or dipolar interactions, for example. This then will be reflected in the global magnetization and size of magnetic domains and, in particular, magnetic relaxation during film growth.

In figure 9 the growth of a thin film resulting from atom by atom deposition at the surface is shown. For simplicity...
fast diffusion of chemisorbed atoms is assumed. Growing of a film occurs, since deposited atoms prefer to sit next to already deposited atoms, thus gaining optimal cohesion. As time progresses islands of deposited atoms coalesce and a nanostructured film is formed. Of course, via diffusion of the deposited atoms the film structure depends somewhat on the film structure of the substrate. Thus, one may get island formation, striped structures and quasi-uniform growth, see calculations by Jensen et al [6, 8, 9, 31]. It is remarkable that already simple growth models yield most of the observed structures during film growth.

Regarding magnetization this reflects the nanostructure and typically magnetic domains occur before global uniform film magnetization is present.

In thin films one expects quantum well states (QWS) which will change characteristically with film thickness. Such states were calculated, for example, by Luce et al [43]. For details see Bennemann in *Nonlinear Optics in Metals* [32, 43, 44]. The confinement in thin films causes the QWS. In FM films these states are spin-split, reflecting characteristically magnetism. In figure 10 QWS are sketched and their parity is indicated by (+) and (−). Magnetooptics (SHG: second harmonic light generation) will reflect magnetism sensitively. Thus, in particular for magnetic nanofilm structures with spin-split quantum well states (QWS), one will get interesting optical properties reflecting characteristically magnetism. This is illustrated in figure 11. Optical interference involving QWS causes oscillations in the MSHG as a function of film thickness.

This then summarizes interesting nanostructures consisting of films. Film multilayers offer particularly interesting magnetic patterns. Neighboring films interact largely via exchange coupling and this controls the magnetic behavior of each film and the global one. For example, transport properties like magnetoresistance depend on the relative orientation of the magnetization of neighboring films. During film growth magnetic relaxation controlled by anisotropy plays an important role. This is reflected in the nonequilibrium magnetization of the film and its relaxation towards the equilibrium one.
Tunnel junctions involving magnetism are interesting microstructures, in particular regarding quantum mechanical behavior, switching devices and charge- and spin currents and their interdependence. For illustration, see figure 12.

Coupling of the magnetic order parameter phases, for both ferromagnets and antiferromagnets, on both sides of the tunnel medium yields Josephson-like (spin) currents (if the electron spin free path is long enough), driven by the phase difference; for details see the study by Nogueira et al [16]. Obviously, in general tunnel currents will depend on the magnetic state of the medium through which tunneling occurs and on spin relaxation. The effective spin coupling between material $N_1$ and $N_3$ may determine $S_{N_2}$ and the effective exchange coupling $J_{ex} = J_{ex}(x)$.

Note, from the continuity equation there follows for the spin current ($\vec{S}$) using the Landau–Lifshitz equation

$$\vec{j}_T \propto J_{ex} \vec{S}_1 \times \vec{S}_3 + \cdots .$$ (3)

Here, $\vec{S}_1$ and $\vec{S}_3$ are the vectorial spin operators representing the magnetism of $N_1$ and $N_3$. Of course, spin relaxation must be taken into account, in general, and it weakens the current (the ansatz $(1 + \tau \vec{J}_T \vec{S}_1 \times \vec{S}_3$ may take into account spin relaxation and damping effects).

Clearly, see figure 12, tunneling allows us to study magnetic effects: ferromagnetic versus antiferromagnetic configurations of the tunnel system $(\uparrow | T | \uparrow)$ or $(\uparrow | T | \downarrow)$ and the interplay of magnetism and superconductivity ($T = \text{SC}$), such as, for example, for junctions (FM/SC/FM) or (SC/FM/SC). Such junctions may serve as detectors for triplet superconductivity (TSC) or ferromagnetism (FM) and antiferromagnetism (AF). Of course tunneling is different for parallel or antiparallel magnetization of $M_1$ and $M_3$, see the GMR effect observed by Grünberg and Fert [40, 41]. Also importantly, tunnel junctions may serve to study Onsager theory on a molecular-atomistic scale.

Electron and Cooper pair transfer between two quantum dots exhibits interesting behavior, for example von Stückelberg oscillations due to the bouncing back and forth of electrons if an energy barrier is present. Then the assistance of photons is needed to overcome the energy barrier. The current (spin and charge current) may depend on the pulse shape and duration, see Garcia, Speer et al [14]. Of course, energy spectrum discretization due to confinement plays a role.

Summary. For reasons of illustration various interesting nanostructures like clusters, films and tunnel junctions with important magnetic effects (interplay of superconductivity and magnetism, Josephson-like spin currents, photon-assisted tunneling) are discussed. In the next section some theoretical methods useful for calculations are presented. Then results obtained in this way are given.

It is important to note that, for illustrational purposes in this review, the physics has been largely simplified. Mostly, the analysis could be improved. For example, tight-binding-type calculations could be improved using density functional theory (DF), molecular field theory by recent extensions using (Wannier-type) Green’s function methods, Bethe–Peierls approximation, Hubbard-type calculations by using dynamical mean-field theory and quantum field theory, in particular regarding phase transitions. However, it is likely this will not change the physical insights obtained from the simplified analysis. For more details see, for clusters. studies by Pastor et al and others [1, 5, 22–24], for mesoscopic structures, rings, etc, theory by Stampfli et al [16], for films, electronic and magnetic structure, research by Jensen et al [6, 7, 9, 25, 31], and for tunnel junction studies by Nogueira [12, 46], Morr et al [10, 11] and Maekawa et al [13]. For Aharonov–Bohm effect-induced currents into mesoscopic rings see Bloch [37] and recent studies [34–36] and the application of Balian–Bloch-type theory by Stampfli [16].

2. Theory

In general, a theory for nanostructures must allow for a local, atomic-like analysis of the electronic structure. Using the Hubbard Hamiltonian and tight-binding-type theory one may determine

(a) $N_{io}(\epsilon, \ldots)$, the electron density of states at an atomic site $i$ and for spin $\sigma$, dependent on the local atomic configuration surrounding atom $i$,

(b) $\mu$, the atomic-like magnetic moment, as a function of particle size (cluster size or film thickness),

(c) $M$, the magnetization and the Curie temperature ($T_c$).

Note, all properties result from the electronic Green’s function $G_{io}(\epsilon, \ldots)$. Including spin–orbit coupling ($V_{so}$) yields magnetic anisotropy. Thus, also

(d) orbital magnetism is obtained. Note, anisotropy and orbital magnetism become typically more important for nanostructures.
Alternatively to an electronic theory, one may use on a more phenomenological level the Heisenberg Hamiltonian including magnetic anisotropy to analyze magnetism in nanostructures.

2.1. Magnetism: electronic theory

To determine the size and structural dependence of the magnetic properties of small transition metal clusters the Hubbard Hamiltonian for d-electrons, which are expected to contribute predominantly, is used (see [1, 22, 24, 29]):

\[
H = \sum_{i<j} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + H',
\]

and effective on-site electron energies:

\[
\epsilon_{i\sigma} = \epsilon^0 + U \Delta n_i - \sigma J \frac{\mu_i}{2},
\]

Here, \(c_{i\sigma}^\dagger\) and \(c_{i\sigma}\) are the usual creation and annihilation operators for electrons on-site \(i\) and with spin \(\sigma\) and \(t_{ij}\) denotes the distance-dependent hopping integral. Note, \(i, j\) refer to atomic sites and includes orbital character (\(d_{e}\), \(d_{eg}\) orbitals, \(s, p\) orbitals). \(H'\) describes interactions (in the unrestricted Hartree–Fock approximation). The effective intra-atomic Coulomb interactions are denoted by \(U\) and the exchange interaction by \(J\) (note, we assume \(U_{++} = U_{++} = U_{++} = U_{++} - J\), hence \(J = U_{++} - U_{++}\) and \(U = (U_{++} + U_{++})/2\)). Here, \(U_{\sigma\sigma'}\) refers to electron spins \(\sigma, \sigma'\). \(E_{dk}\) is given by (1/2) \(\sum_{\sigma}(\epsilon_{i\sigma} - \epsilon^0)\langle n_{i\sigma}\rangle\) corrects for double counting as usual, see [24]. The charge transfer \(\Delta n_i\) is given by \(\Delta n_i = n_i - n^0\), \(n^0 = (1/n) \sum n_i\).

The quasi-local magnetic moment at site \(i\) is given by

\[
\mu_i \propto \langle n_{i\uparrow} - n_{i\downarrow}\rangle,
\]

with \(\langle n_{i\sigma}\rangle = \int e^{-\beta E_{i\sigma}} d\epsilon N_{i\sigma}(\epsilon - \epsilon_{i\sigma})\). Here, \(N_{i\sigma}(\epsilon)\) is the density of states (DOS) at site \(i\) for electrons with spin \(\sigma\) and at time \(t\).

This theory applies also to thin films. Then, \(i\) may refer to the film layer and one should keep \(\mu_i\) and \(p_i^{\uparrow\downarrow}\).

To calculate the magnetization \(M(T)\) one must take into account the orientation of the magnetic moments. Assuming a preferred magnetization axis one gets (Ising model), see [29]

\[
M(T) \simeq \sum [p_i^{\uparrow\downarrow} \mu_i^{\uparrow\downarrow}(T) + p_i^{\downarrow\uparrow} \mu_i^{\downarrow\uparrow}(T)]/\mu^0(0).
\]

The probabilities \(p_i^{\uparrow\downarrow}\) refer to finding moments \(\mu_i^{\uparrow\downarrow}\) pointing parallel or antiparallel to the preferred magnetization axis. For simplicity one may use the approximation \(p_i^{\uparrow\downarrow} = p^{\uparrow\downarrow}\). Assuming spherical-like clusters, then \(i\) refers to the atomic shell of the cluster and \(\mu_i^{\uparrow\downarrow}\) to the magnetic moment within shell \(i\) pointing in the direction of the magnetization (+) and in the opposite direction (−), respectively.

Then one determines the order parameter \((\alpha M)\):

\[
\mu_i = p_i^{\uparrow\downarrow} - p_i^{\downarrow\uparrow} \geq p^{\uparrow\downarrow} - p^{\downarrow\uparrow}
\]

(\(\mu_i \approx \mu\)) from minimizing the free energy:

\[
F = \Delta E - T S.
\]

Here, the entropy \(S\) is given by

\[
S \simeq -kN[p^{\uparrow\downarrow} \ln p^{\uparrow\downarrow} + p^{\downarrow\uparrow} \ln p^{\downarrow\uparrow}],
\]

and \(\Delta E = E(\mu) - E(0)\). The electronic energy is calculated using a Hamiltonian \(H\), for example equation (4). For calculating the Green’s functions the electronic energies are determined from

\[
\epsilon_{i\sigma}^{\uparrow\downarrow} \simeq \epsilon_{i\sigma}^0 - \sigma J \sum_j \mu_j^{\uparrow\downarrow} \mu_j^{\downarrow\uparrow}.
\]

Note, \(\epsilon_{i\sigma}^{\uparrow\downarrow} \simeq \epsilon_{i\sigma}^0 - \sigma \mu_i^{\uparrow\downarrow} \sum_{\sigma'} \sum_{\sigma''} [p^{\uparrow\downarrow}\mu_i^{\uparrow\downarrow}\mu_i^{\downarrow\uparrow}]\). The Curie temperature is given by \(M(T_c) = 0\).

A similar analysis can be performed using functional–integral theory as developed by Hubbard et al, see [2, 22, 24].

Note, as already mentioned, this theory can also be used for films. Then \(i\) may refer to the film layer, etc.

The magnetization can also be determined using the Bragg–Williams approximation. Assuming for simplicity Ising-type spins one finds for the magnetization (see [28])

\[
M_i(T) = \text{tanh}[\beta J \mu_i(z_0 M_i \mu_i + z_1 M_{i+1} \mu_{i+1} + z_{-1} M_{i-1} \mu_{i-1}) - \Delta h_i].
\]

Here, \(\beta = 1/kT, z_0, z_1, z_{-1}\) are nearest-neighbor coordination numbers and \(\Delta h_i\) denotes the Onsager reaction field. Referring to cluster shells (film layer) \(z_0\) gives the neighboring atoms of \(i\) within shell (layer) \(i\) and \(z_{-1}\) and \(z_1\) are the nearest-neighbor atoms in the shell (layer) below and above, respectively. It is

\[
\Delta h_i = (\beta J \mu_i)^2 M_i(z_0 \mu_i^2(1 - M_i^2) + z_1 \mu_i^2(1 - M_{i+1}^2) + z_{-1} \mu_{i-1}^2(1 - M_{i-1}^2)).
\]

Applying these expressions to films one has \(z_1 = 0\) (and \(\mu_i = 0\)) if \(i\) refers to the surface plane and \(z_{-1} = 0, \mu_i = 0\) if \(i\) refers to the film layer on a nonmagnetic substrate.

Note, for \(T \ll T_c\) equation (13) can be linearized yielding a (tridiagonal) matrix equation whose largest eigenvalue gives \(T_c(d)\). For the Hamiltonian \(H\) one may use the Heisenberg one, for example.

Orbital magnetism and anisotropy: adding spin–orbit interaction \(V_{so}\) to equation (4), \(H \rightarrow H + V_{so}\), and using

\[
V_{so} = -v \sum_{\alpha,\beta} (\vec{L}_i \cdot \vec{S})_a_b c_{a\alpha}^\dagger c_{b\beta},
\]

one may also determine magnetic anisotropy and also orbital magnetic moments \((\vec{L}_i)\). \((\vec{L}_i \cdot \vec{S})_a_b\) refers to intra-atomic matrix elements between orbitals \(\alpha, \beta\). Of course, the orbital moment \(\vec{L}_i\) depends on cluster atom \(i\) and on the orientation \(\delta\) of \(\vec{S}\) with respect to the structural axis: \(\vec{L}_i \rightarrow L_{i,\delta}\) (see [21]). In the case of films \(i\) refers to the film layer.

Note, in clusters and thin films, and in general at surfaces, the spin–orbit coupling and orbital magnetism are typically enhanced. Orbital magnetism may play an interesting role for nanostructures and interface physics.
2.2. Magnetism: Heisenberg-type theory

One may also calculate the magnetism in nanostructures, in particular, in thin films by assuming local spins and using then the Heisenberg-type Hamiltonian, including magnetic anisotropy (see Jensen et al. [6–9]). Regarding magnetic anisotropy see also studies by Jensen with details given in [26, 27, 31]). Note, due to magnetic anisotropy phase transitions occur also in two dimensions. Furthermore, as known, magnetic anisotropy controls in particular in thin films and at interfaces the magnetic structure-like direction of the magnetization, formation of domains, the domain size and shape.

Note, typically in bulk transition metals (in contrast to rare-earth) the exchange coupling dominates, but in nanostructures (at surfaces, etc) anisotropy is enhanced and must be taken into account. Generally magnetic anisotropy determines sensitively the magnetic behavior in thin films and in structures with reduced dimension, see [6]. The main magnetic anisotropies are the magnetocrystalline one (MCA), due to spin–orbit coupling and the orbital motion of electrons, the exchange anisotropy (MEXA), involving the exchange interaction, and in particular for layered film structures the magnetic dipole coupling (the origin of stray fields and the demagnetization field).

Note, the exchange anisotropy (MEXA) for most magnetic properties yields similar results as for (in second order) the single-ion MCA (when describing the Hamiltonian \( H_{\text{MCA}} \) in terms of spherical harmonics \( Y_{l,m} \)). Hence, both anisotropies, despite having as is well known different physical origins, may approximately be combined by using appropriate parameters. Then we determine the magnetic structure resulting from the quasi-local magnetic moments \( \mathbf{S}_i \) using the Hamiltonian

\[
H = -\frac{1}{2} J \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{2} A \sum_{i,j} \left( \frac{\mathbf{S}_i \cdot \mathbf{S}_j}{r_{ij}^3} - \frac{3 (\mathbf{S}_i \cdot \mathbf{r}_{ij})(\mathbf{S}_j \cdot \mathbf{r}_{ij})}{r_{ij}^5} \right) + H_{\text{anis}},
\]

(16)

with the exchange anisotropy Hamiltonian

\[
H_{\text{anis}} = -\frac{1}{2} K \sum_{i,j} S_i^x S_j^x - \frac{1}{4} D \sum_{i,j} (S_i^x S_j^y + S_i^y S_j^x).
\]

(17)

Here, \( K \) and \( D \) are uniaxial and quartic in-plane (exchange) anisotropy constants, respectively. Note, in \( H \) the first term is the Heisenberg exchange interaction and the second term refers to the magnetic dipole interaction, and \( A = \mu_0 (g \mu_B)^2 / a_0^3 \). \( \mathbf{S}_i \) denotes Heisenberg spins (for Ising model spin \( \frac{1}{2} \)) at site \( i \). The second term in equation (16), which results as an interaction amongst magnetic moments in classical electrodynamics, reduces the magnetization normal to the surface in particular at surfaces. It gives the demagnetization field and is called the shape anisotropy. Although this long range magnetic dipole coupling is usually much smaller than the exchange one, it is typically very important on a mesoscopic length scale and determines the magnetic domain structure.

The anisotropy Hamiltonian \( H_{\text{anis}} \) includes as discussed both the magnetocrystalline and exchange anisotropy, fitting the parameter correspondingly. Of course, the parameters, in particular \( K \) and \( D \), may depend on \( i \) and \( j \) (shell and film layer). Typically anisotropy is larger at surfaces and interfaces. The interplay of \( K \) and \( D \), which may depend on film thickness and temperature (\( K \to K_f \), where \( i \) refers to film layer), determines the normal and in-plane magnetization at surfaces and the reorientation transition (see [6]). Note, one has also higher-order anisotropy constants like \( K_4 \) due to noncollinearity of the spins (approximately \( K_4 \propto J^{-1} \)), see [6, 25, 26], which may play a role. It is also important to note that exchange and dipolar coupling have a different distance dependence. The parameters, in particular the anisotropy ones, depend in general on temperature. Of course, temperature induces changes of the atomic structure and this plays a role for thin films and for films during growth on a substrate.

Applying standard methods of statistical mechanics one gets the free energy \( F \), the magnetization and phase diagrams in terms of \( J \), and the anisotropy forces for clusters and thin films, and also for nanostructured films with magnetic domain structure. For a more detailed analysis see, in particular, [6, 9, 27, 30, 31]. The magnetic phase diagram (PD) follows from minimizing the free energy [6, 30].

From equation (16) follows the magnetic reorientation transition, for example as driven by temperature:

\[
M_\perp \rightarrow T_R M_\parallel
\]

(18)

occurring at temperature \( T_R \). Note, the temperature dependence of the effective anisotropy parameters and of the dipole coupling is mainly determined by the magnetization. Minimizing the free energy (anisotropy contribution to the free energy) gives the transition \( M_\perp \rightarrow M_\parallel \) at temperature \( T_R \), see [25, 26].

Note, the control parameters in equation (16) depend on atomic structure, morphology of the nanostructures and film thickness, for example. Thus, \( T_R \) is affected.

The above Hubbard-tight-binding-type electronic theory and the Heisenberg-type phenomenological theory including magnetic anisotropy permit a calculation of the magnetic properties of nanostructures. A basic understanding of the dependence of magnetism in nanostructures on spin–orbit coupling and orbital magnetic moments (see [21]) and on atomic configuration and on a more global morphology is obtained.

Note, for alloys one may extend the above theories using appropriate versions of the CPA-like analysis (CPA: coherent potential approximation).

2.3. Electronic structure of mesoscopic systems: Balian–Bloch-type theory

The important electronic structure (shell structure) of mesoscopic systems, like spherical clusters, discs, rings and (quantum) dots, can be determined using a relatively simple theory developed by Stampfl et al. extending the original work by Balian–Bloch (Gutzwiller) [16]. One assumes a square-well-like generally spin-dependent electron scattering potential \( (U_s) \). The dominant contribution to the electronic structure (near the Fermi energy and for electronic wavevector
$k_{\perp}^{-1}$ larger than the atomic distance) results from (interfering) closed electronic orbital paths. Then the key quantity of the electronic structure of a quantum dot system, the density of states (DOS), can be calculated from \((n = \text{number of atoms in the nanostructure})\) from the Green function \(G\). The Green function \(G(\vec{r}, \vec{r}')\) is derived by using multiple scattering theory, see Stampfli et al. Interference of different electron paths yields oscillations in the DOS, etc. This leaves a fingerprint on many properties.

Thus, for example, oscillations in the electronic structure of mesoscopic systems, in the magnetoresistance and in other properties of quantum dot systems can be calculated.

From the electronic Green function \(G\) one determines the (generally spin-dependent) density of states (DOS) \((\vec{r} \text{ and } \vec{r}' \text{ refer to spatial coordinates, } d^d \text{ to the dimensional integration: note that superscript } d = 2 \text{ refers to two-dimensional structures, for example})\). The DOS at energy \(E\) is given by

\[
N_\sigma(E, n) = \frac{1}{2\pi i} \int d^d r \{G_\sigma(\vec{r}, \vec{r}', E + i\epsilon) - G_\sigma(\vec{r}, \vec{r}', E - i\epsilon)\} \gamma_{\sigma,\gamma}\).
\]

One gets \((\bar{N} = \text{average DOS})\)

\[
N_\sigma(E, n) = \bar{N}_\sigma(E, n) + \Delta N_\sigma(E, n),
\]

where \(\Delta N_\sigma\) refers to the oscillating part of the DOS due to interference of the predominant closed electron paths in clusters, thin films and the ensemble of repelling anti-dots, see the theory by Stampfli et al [16]. Clearly the scattering can be spin-dependent (due to a potential \(U_\sigma\)) and can be manipulated by external magnetic fields \(B\) (see cyclotron paths, Lorentz force, etc) [16].

Under certain conditions regarding the potential felt by the electrons in the nanostructures (square-well-like potentials, etc, and states with \(k^{-1} > a\), where \(a\) is the interatomic distance) one gets for the DOS the result (see Stampfli et al [16] using extensions of the Baltic–Bloch theory)

\[
\Delta N_\sigma(E, n) \simeq \sum_l A_{l\sigma}(E, n) \cos(kL_l + \phi_{l\sigma}).
\]

Here, \(l\) refers to closed orbits (polygons) of length \(L_l\), \(k = \sqrt{|E|} + ik\) and \(\phi_{l\sigma}\) denotes the phase shift occurring for orbit \(l\) and electron spin \(\sigma\) and characterizing the scattering potential and the geometry of the system. The factors \(A_{l\sigma}\) are the weights by which each orbit contributes to the DOS. An external magnetic field affects the DOS \(\Delta N_\sigma(E, n)\) via path deformation and phase shifts resulting from magnetic flux (see the Aharonov–Bohm effect). This will induce electric currents \(j\) into ring-like metallic mesoscopic structures which are proportional to the DOS \((j_{AB}(E) \propto (1/R) \sum N'(E, i),\) where \(N'\) is the derivative with respect to phase and where \(R\) is the radius of the ring and \(i\) refers to the electron orbit).

Clearly, the DOS in particular \(\Delta N_\sigma(E, n)\) will be affected characteristically by the magnetism in various nanostructures and in quantum dot systems. For details of the Baltic–Bloch-like analysis see the theory by Stampfli et al [16].

Note, the electronic structure due to the interference of the dominant electronic states near the Fermi energy described approximately by equation (21) results, in addition to the one due to the atomic symmetry of the nanostructure (a spherical one for clusters, 2d symmetry for thin films, etc.). Thus, for example, one may find for magnetic clusters a phase diagram temperature versus cluster size which is dependent on magnetization and exhibits phases where the atomic (shell) structure and then spin-dependent electronic structure dominates. Of course, for sufficiently large clusters these structures disappear.

The interference of closed electron orbits in magnetic mesoscopic systems like spherical clusters, discs, rings, thin films and quantum dot lattices causes spin-dependent characteristic structures and oscillations in the DOS \(N(E, n)\) (yielding corresponding ones, for example, in the cohesive energy, occupation of d, s electron states, the Slater–Pauling curve for magnetism, magnetoresistance, etc).

(a) Spherical clusters. The properties of small spherical clusters follow from the DOS given approximately by

\[
\Delta N_\sigma = \Delta N_\sigma + \sum_{p=2}^{\infty} \sum_{l=1}^{N_\sigma} A_{l, p, \sigma} \cos \phi_{l, p, \sigma},
\]

where the second term gives the contribution of the various electron states (paths). The electron orbits are characterized by the number of corners \(p\) and \(t\) describes how many times the center of the sphere is circled. Here, \(\Delta N_\sigma\) results for \(p = 2t\) and turns out to be unimportant. For a detailed derivation of the amplitudes \(A_{l, p, \sigma}\) and phases \(\phi_{l, p, \sigma}\) see the analysis by Stampfli et al [16]. One gets

\[
A_{l, p} = 2R^2 a_{l, p} \sqrt{\sin^3(\pi t/p)/p} \times \exp[-(2p2R \sin(\pi t/p)] ,
\]

with \(a_{l, p} = \sqrt{t!R/\pi(-1)^l \cos(\pi t/p)}\). Note, the amplitudes decrease rapidly for increasing number \(p\) of corners. The most important contributions to the DOS result from triangular, square and lower polygon orbits. The structure in the DOS affects many properties (conductivity, magnetoresistance, spin susceptibility \(\chi\), spin-dependent ionization potential, etc).

(b) Circular rings. Circular rings with outer radius \(R_o\) and inner radius \(R_i\) are particularly interesting, since \(R_o/R_i\) controls which orbits are most important. For example, one may eliminate DOS contributions due to triangular orbits. One gets (see figure 13 for illustration)

\[
\Delta N = \Delta N_1 + \Delta N_2 + \Delta N_3,
\]

where \(\Delta N_1\) results for orbits inside the ring (with \(\pi t/p < \arccos(R/R)\) and which are also present for discs, \(\Delta N_2\) due to orbits scattered by the outer and inner surfaces of the ring and which circle around the center, and \(\Delta N_3\) results from ping-pong-like orbits between \(R_o\) and \(R_i\) In all cases one gets rapid oscillations (due to exponentials) and slower ones due to cos and sin functions.

Note, results for thin films follow for \(R_o, R_i \rightarrow \infty\).
number of corners and tangents are spin-polarized. The Aharonov–Bohm effect may induce currents. For ferromagnets these currents are the center, respectively, of the polygonal paths. In (c) the effect of an external magnetic field $B$ is shown. Note, in particular for rings magnetic flux quantization may occur. Also the phase shift due to the Aharonov–Bohm effect may induce currents. For ferromagnets these currents are spin-polarized.

(c) Quantum dots. The DOS of quantum dots is calculated similarly. Thus, one gets, for example, oscillations in the magnetoresistance $r_x$, since $r_x \propto N(E, n)$. Molecular fields and external magnetic fields $B$ will affect the electronic orbits, see figure 13 for illustration. For discs, rings and quantum dots one gets phase shifts $\Delta\phi$ due to path deformation and the flux through the polygons. Note, the Lorentz force deforms the orbits. Also the magnetic flux due to a vector potential $A$ perpendicular to the disc’s plane, for example, gives a phase shift:

$$\Delta\phi' = \oint A \cdot d\vec{r} = \vec{B} \cdot \vec{S},$$

where $S$ is the area enclosed by the orbit. Note, the structure in DOS due to Landau levels may be included already in $N(E, n)$.

(d) Circular disc and quantum dots. For circular discs and quantum dots one gets (for square-well-like potentials) in equation (22) that $N_0 \approx 0$ and for the second term that mainly orbits with $p = 2, 3$ are important, since $A_{t, p, \alpha} \sim 1/p^{1/2}$. Note, now $A_{t, p, \alpha} = (\alpha_{t, p, \alpha}/2)!/(\pi k_1 R p)$ and $A_{t, p} = \alpha_{t, p} R^2 \left[(1/\pi k_1 R p) \sin^2(\pi r / p)\right]^{1/2}$

$$\times \exp \left[-2p k_2 R \sin(\pi r / p)\right] + \text{(26)}$$

with $\alpha_{t, p} = 2$ for $p \neq 2t$ and $\alpha_{t, p} = 1$ for $p = 2t$ and phases $\Phi_{t, p} = p[2k_1 R \sin(p / R) - \pi / 2 + \delta_{t, p}] + \Delta / 4$ as for spheres. Phases $\delta_{t, p}$ result from (possibly spin-dependent) potential scattering at the surface of the system, see books on quantum mechanics [16]: note $\delta_{t, p} = -\pi$ for $U \to \infty$. The phase shift $\delta$ affects level spacing (and vice versa) and the appearance of the electronic shell structure.

Effect of magnetic field $B$. The magnetic field is taken to be perpendicular to the disc plane. Then one gets a change of the phase by

$$\Delta\Phi = \Delta\Phi_1 + \Delta\Phi_2,$$

where $\Delta\Phi_1 = p k R \Phi_{t, p}^0$, $\Phi_{t, p}^0 = 2 \arcsin(R \sin(\Phi_0^0) / R_c)$, the cyclotron orbit $R_c = k_c / B$ ($R_c \gg R$) is due to path deformation and $\Delta\Phi_2 = \Delta\Phi'$, see the previous equation, due to the enclosed flux (Aharonov–Bohm). The angle $\Phi_0^0$ refers to the center of the cyclotron orbit, see figure 13. Straightforward analysis gives

$$\Delta\Phi' = \pm BS\phi_c,$$

with

$$S_{t, \alpha} = S_0 \pm \Delta, \quad \Delta = (p / 2) R^2 \left[\phi_c^0 - \sin\phi_c^0\right]$$

Here, $2\phi_c^0 = (2\pi / p)t$, where $\phi_c^0$ is the angle seen from the center of the disc, see figure 13, and for the cyclotron orbit one assumes $R \gg R$. $t$ refers to clockwise and counterclockwise motion around the center of the orbit. $\Delta$ denotes the area resulting from path deformation due to field $B$. It is then

$$\Delta N_0(E, B) \propto \Sigma_{t, p} A_{t, p} \cos(S\Phi_0^0) \cos(S) \sin(S) \exp \left[-(2k_2 / B)\Phi_{t, p}^0\right].$$

where $\Phi_{t, p}$ denotes phase changes due to path deformation, see the previous expression. The factor $\cos(S\Phi_0^0)$ describes the Aharonov–Bohm effect plus interference effects and gives oscillations periodic in $B$.

Note, cos $BS$ causes oscillations which are periodic in $B$, and oscillations change with $(a - d)$. For large field $B$, $2R_c \leq (a - d)$, see figure 4, paths 1 and 3 disappear and only 2 remains (see Landau level oscillations with periodicity $1(B)$, since $S \propto (1/B^2)$. $\Delta N \propto \cos(2S)$. The DOS oscillations are $\Delta N \propto \cos(2S)$. The ratio $(d/a)$ characterizes, for example, the strength of anti-dot potential scattering. Of course, the oscillations in the DOS cause similar ones in the magnetoresistance, for example.

Anti-dot lattice: one assumes a 2d lattice of anti-dots scattering via a repulsive potential of the electrons. The lattice of anti-dots is illustrated in figures 4 and 14. Note, orbits 1, 2 and 3 are most important. The scattering by the anti-dot lattice causes phase shifts $\Phi_{t, p, r}$. Note, the area $S$ enclosed by the
orbits 1, 2 and 3 is nearly independent of magnetic field $B$; such orbits occur for $2R_c > (a - d)$ and $R_c \propto 1/B$. One assumes that orbits are dephasing for distances much larger than lattice distance $a$. The oscillations in the DOS are given by (performing calculations similarly as for discs, see similar paths in figures 13 and 14)

$$\Delta N_o(E, B) = \left(\frac{\sqrt{2}(a - d)}{k_1\pi}\right) \times \sum_{n=0}^{\infty} \left(\frac{\sin\varphi}{\sinh(4t + 1)\varphi}\right)^{1/2} \sin\varphi, \quad \varphi = \frac{\pi}{2},$$

where the eigenvalues $\{\varphi\}$ of the state $|m\sigma\rangle$ are determined by the Schrödinger equation. Then

$$N(E)_{\sigma} = \Sigma_{\sigma}\delta(E - E_{m\sigma}).$$

Note, in general for lower dimensions ($3 \rightarrow 2$) degrees of freedom are reduced and one expects stronger oscillations of the DOS, for example in a magnetic field. The interference of the orbits yields generally rapid and slow oscillations. The spin dependence is calculated from the spin-dependent scattering potential $U_{\sigma}$ at the surfaces of the system.

Note, a ring may be viewed as an anti-dot enclosed by the metallic ring. The electrons are repelled by a strong repulsive potential from the inner core of the ring. If a magnetic field $B$ is present, due to the flux quantization the electronic DOS reflects this and has fine structure as a result of the flux quantum $\phi_0 = 2\pi \hbar c/e$. Also, a beating pattern of the oscillations may reflect interference of several paths dominating the electronic structure, see the corresponding figures. These oscillations are also present in the electronic energy $E = \int dB N(E) + \cdots$. As also discussed later the Aharonov–Bohm effect may induce a current in rings, discs, etc., with interesting structure. (Note, this is also expected for rings of electrolytes. Then the induced currents may be reduced by a factor $m/M$, where $m$ and $M$ refer to the electron and ion mass, respectively.)

For further details of the analysis for rings and films see Stampfli et al [16]. In particular these may exhibit interesting behavior at nonequilibrium (due to hot electrons, for example, [32, 33]).

It is important to compare the results by Stampfli et al using the Balian–Bloch-type theory [16] with quantum mechanical calculations of the DOS. Then

Note, (31) takes into account that path reflections result from small anti-dots (rather than from spherical surfaces of discs with varying curvature). The oscillations of $\Delta N_o$ periodic in $B$ result for orbits 1, 2, 3 and 4 and change with $(a - d)$. For very small fields $B$, note $R_c \propto 1/B$, orbits become unstable and the large orbits become irregular due to dephasing. For increasing magnetic field $B$ such that $2R_c \leq (a - d)$ first orbit 3 and then 1 disappear and only 2 survives. This orbit causes then Landau level oscillations with period $(1/B)$ for increasing field $B$, since $S \propto R^2 \propto (1/B)^2$, flux $\sim B$ and $\Delta N \propto \cos(SB)$. The oscillation periods are expected to decrease for decreasing lattice constant $a$ and increasing magnetic field $B$ [16].

The oscillations in DOS $\Delta N_o(E, B)$ affect many properties, in particular the magnetoresistance. A crossover from periodicity proportional in $B$ to one in $(1/B)$ should occur, for example, for discs.

The theory by Stampfli et al can be used also for a system of quantum dots on a lattice, including then hopping between the dots. Furthermore, an ensemble of magnetic and superconducting quantum dots may exhibit interesting behavior regarding (quantization effects) magnetic flux, etc [17, 18].

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For further details of the analysis for rings and films see Stampfli et al [16]. In particular these may exhibit interesting behavior at nonequilibrium (due to hot electrons, for example, [32, 33]).

It is important to compare the results by Stampfli et al using the Balian–Bloch-type theory [16] with quantum mechanical calculations of the DOS. Then

$$N(E)_{\sigma} = \Sigma_{\sigma}\delta(E - E_{m\sigma}).$$

where the eigenvalues $E_{m\sigma}$ of the state $|m\sigma\rangle$ are determined by the Schrödinger equation.

2.4. Magnetooptics

Interesting magnetooptical behavior is exhibited by magnetic films. In addition to linear magnetooptics (Faraday effect, Kerr effect), see the review by Bader et al in [32], nonlinear optics and second harmonic light (SHG) (in particular nonlinear magnetooptics (MSHG)) is used intensively in recent years. Note, SHG and MSHG (note, spin–orbit coupling is the physical origin) is very surface-sensitive and reflects the magnetic properties of thin films very well. SHG requires a breakdown of inversion symmetry thus generated in nanostructures and at the surface, at the interface surface/substrate, or at the interface of the two films, see [32].

Thus, MSHG is best suited to study multifilm structures, their magnetic configuration and, very importantly, to study the dynamics and nonequilibrium magnetism. Note, in contrast to linear magnetooptics frequently MSHG exhibits a much larger Kerr effect and, in a more pronounced way, magnetism in general.

The symmetry sensitivity of SHG becomes obvious from the expansion of the electric field $E$-induced polarization $P = \chi^1 E + \chi^2 EE + \cdots$. Here, $\chi^1$ and $\chi^2$ denote the linear and nonlinear susceptibilities. The intensity of SHG light is given by $I \propto |\chi^2|^2$. SHG depends characteristically on the

Figure 14. Effect of a magnetic field on various closed electron orbits and paths: (a) for a ring, (b) for a lattice of anti-dots, which scatter the electrons by a repulsive potential. Note, in particular for a narrow ring, path 1 may dominate and magnetic flux quantization occurs. If the mean free path of the electrons is relatively large, as compared to the circumference of the ring, a magnetic field may induce a current driven by the phase shift due to the Aharonov–Bohm effect. On general grounds this current is expected to be spin-polarized for ferromagnets and to decrease proportional to $R^{-2}$ and for increasing temperature.
optical configuration at the surface and on the polarization of the incoming and outgoing light. This is described by the various elements of the susceptibility tensor $\chi_{ijkl}$. The optical configuration used for calculating the polarization dependence and dependence on the magnetization direction of SHG is illustrated in figure 15.

Figure 15 shows the optical configuration and SHG at surfaces (for light $\omega \rightarrow 2\omega$). As said, the $2\omega$ light is characterized by the response function $\chi_{ijkl}(\tilde{M}, \omega)$. Clearly, since the response depends on $\tilde{M}$, note $M_x$ and $M_y$ yield different optical responses $\chi_{ijkl}(\tilde{M})$. Hence the reorientation transition of the magnetization can be studied optically.

For thin films the nonlinear susceptibility and response function $\chi^2$ may be split into the contribution $\chi^2$ from the surface and $\chi^2$ from the interface. Then owing to the contribution $\chi^2 \chi^1$ to the SHG intensity $I \propto |\chi|^2$, the relative phase of the susceptibilities $\chi^1$ and $\chi^2$ is important. Furthermore, the magnetic contrast

$$\Delta I(2\omega, M) \propto I(2\omega, M) - I(2\omega, -M)$$

will reflect the film magnetism, since the susceptibility has contributions which are even and odd in $M$, see [32].

Note, high resolution interference studies are needed to also detect, for example, lateral magnetic domain structures of films. Also, polarized light reflects magnetism and in particular the magnetic reorientation transition, see figure 16 and the following results.

In magneto-optics (MSHG) using different combinations of polarization of the incoming and outgoing light, see figure 15, one may analyze

$$\chi_{ijkl}(\tilde{M})$$

Regarding the dependence on magnetization, for the susceptibility odd in magnetization one writes

$$\chi_{ijkl}(\tilde{M}) = \chi_{ijkl}(M) + \cdots$$

Here, $M_m$ is the $m$th component of the magnetization $M$. Note, the susceptibility can be split into contributions odd ($o$) and even ($e$) in $M$, respectively, $\chi = \chi^e + \chi^o$. Thus, in particular the susceptibility as a function of the magnetization, $\chi_{ijkl}(M_n)$, changes characteristically for the spatial directions $c = x, y$ or $z$. For example, for incoming s-polarized light and outgoing p-polarized $2\omega$ light, see figure 15 for illustration, the nonlinear susceptibility $\chi_{ijkl}(M_n)$ dominates. Similarly $\chi_{ijkl}(M_n)$ dominates in the case of outgoing s-SHG polarization. For general analysis see, in particular, Hübner et al [44] and other chapters in [32].

Regarding susceptibility $\chi^o$ and $\chi^e$ note that further interesting behavior could result from terms which are higher order in the magnetization $M (\chi^o = \chi^o_0 + aM^2 + \cdots)$ and $\chi_{ijkl}$ reflects also the strength of the spin–orbit interaction. As discussed by Hübner et al in the case of

1. $\tilde{M} \parallel \tilde{x}$ (longitudinal configuration), the tensor $\chi_{ijkl}$ involves $\chi_{xzx}, \chi_{yzy}, \chi_{zzz}, \chi_{yzy}, \chi_{xzx}, \chi_{zzz}, \cdots$ and in the case of
2. $\tilde{M} \parallel \tilde{z}$ (polar configuration, optical plane $x, z$) elements $\chi_{xzx}, \chi_{xzy}, \chi_{xxx}$ and $\chi_{xxx}$ of susceptibility $\chi_{ijkl}$ occur.

For a longitudinal configuration and polarization combination $s \rightarrow p$ $\chi_{xxx}$ and for $p \rightarrow s$ $\chi_{xxx}, \chi_{zzz}$ are involved. Furthermore, in the case of a polar configuration and polarization combination $s \rightarrow p$ the element $\chi_{xxx}$ and for $p \rightarrow s$ the element $\chi_{xxx}$ occur. This demonstrates clearly that Nolimoke (MSHG) as well as MOKE can observe the magnetic reorientation transition and other interesting magnetic properties of nanostructures, see [32].

In figure 16 results by Hübner et al [32, 44] for the polarization dependence of SHG light are shown. Note, agreement with experiments is very good. Obviously magnetism is clearly reflected. Similar behavior regarding polarization dependence is expected for the magnetic transition metals Cr, Fe and Co. Due to $\langle d | z | d \rangle \approx \langle d | x | d \rangle$ for the dipole transition matrix elements one gets typically the same results for s- and p-polarized light. To understand the behavior of Cu, note $\langle s | z | s \rangle \approx \langle s | x | s \rangle \approx 0$. Curves (a) and (b) refer to wavelengths exciting and not exciting the Cu d-electrons, respectively. Generally as physically expected, $I_s(p-SH)_{NM} \rightarrow 0 \ I_s(p-SH)_{TM}$. Again for a
detailed discussion of the interesting polarization dependence see magneto-optics and the discussion by Hübner et al [44].

In particular, the interesting magneto-optical behavior of SHG and MOKE results also from the spin-dependent quantum well states (QWS) occurring in thin films, see figure 17. These states result from the square well potential representing the confinement of the electrons in a thin film. In magnetic films the resulting electron states are, of course, spin split. Characteristic magnetic properties follow. In contrast to band-like states for the electrons in the film only QWS show a strong dependence on film thickness. Thus, to study film-thickness-dependent (optical) behavior SHG involving QWS needs to be studied.

One expects characteristic behavior of the optical response, its magnitude and dependence on light frequency $\omega$ and $M$. Characteristic oscillations in the MSHG signal occur, since QWS involved in resonantly enhanced SHG occur periodically upon increasing film thickness, see figures 17 and 18 for illustration (note, SHG involving transitions $i \rightarrow j$ and with one of these states being a QWS).

Clearly, the QWS energies shift with varying film thickness. Then the SHG light intensity ($I_{2\omega}$) involving these states may oscillate as a function of film thickness and this in particular may reflect the magnetism of the film. Clearly, owing to the periodic appearance of QWS at certain energies for increasing film thickness, SHG involving these states may be resonantly enhanced and then oscillations occur as a function of film thickness.

A detailed analysis, see the calculation of QWS by Luce, Bennemann [43, 44], shows that the SHG periods depend on the parity of the QWS, the position of the QWS within the Fermi sea or above, and on the interference of second harmonic light from the surface and interface film/substrate, etc. If this interference is important, then SHG response is sensitive to the parity of QWS, the light phase shift at the interface and inversion symmetry of the film. If this interference is not important, then SHG response and oscillations are different, see figure 11 for illustration of the interference.

Thus, for an analysis of SHG involving QWS one may study: (a) $\chi^i \simeq \chi^s$: when interference is important. Different behaviors of SHG may then result if (1) QWS is involved as the final state, (2) as the intermediate state or (3) both QWS as the intermediate and final state matter. Note, for $\chi^i \chi^j \rightarrow (-1)$, for example due to inversion symmetric films or a phase shift $\pi$ at the interface, the destructive interference yields no SHG signal. Also, if a final QWS near the Fermi energy (and which may set the period of the SHG oscillation) has even parity, then in contrast to a QWS with odd parity no SHG signal occurs, since for the latter the product of the three dipole matrix elements is small.

(b) $\chi^i \neq \chi^s$ and interference is unimportant. Then different oscillation periods may occur. For example, if a QWS above the Fermi energy becomes available at film thickness $d_1$ for SHG then a first peak in SHG appears and then again at film thickness $2d_1$ when the previous situation is repeated, and so on. Of course, the strength of the signal depends on wavevector $k$ in the BZ (Brillouin zone), the unit cell in the reciprocal crystal lattice space, and DOS and frequency $\omega$ must fit the optical transition. In the case of an FM film then the resonantly enhanced SHG transitions are spin-dependent and an enhanced magnetic contrast $\Delta I$ may occur.

If occupied QWS below the Fermi energy are involved, then also oscillations occur, in particular due to DOS, see transition metals. If QWS below and above the Fermi energy cause oscillations then the period of the SHG may result from a superposition: as an example see the behavior of $x$Cu/Co/Cu(001) films.

Characteristic properties of film SHG are listed in table 1. Various properties are listed for the case of no interference ($\chi^i \gg \chi^s$) and strong interference ($\chi^i \approx \chi^s$) of light from the surface and interface. The first case is expected for a film system $x$Cu/Fe/Cu(001), for example, since the interface Cu/Fe dominates due to the QWS of the Cu film and the large DOS of Fe near the Fermi energy $\epsilon_F$ (see SHG transitions: Cu $\rightarrow_{\omega}$ Fe $\rightarrow_{\omega}$ QWS $\rightarrow_{2\omega}$ Cu). Thus, $\chi^i \gg \chi^s$ and, due to the QWS above $\epsilon_F$, one gets two or more SHG oscillation periods.

The case $\chi^i \approx \chi^s$ is expected for a film $x$Au/Co(0001)/Au(111), for example, since no Co d-states (see the band
The characteristics of SHG response from thin films. Its dependence on the film thickness involves QWS. The SHG oscillations reflect magnetic properties of the film.

| \( k \) selectivity | \( |\chi'| \gg |\chi^0| \) | \( |\chi'| = |\chi^0| \) |
|---------------------|---------------------------------|---------------------------------|
| \( (x \text{ Cu/Fe/Cu}(001) \text{ for example}) \) | Strong magnetic signal due to strong (magnetic) interface contributions | Weak SHG signal, from only a few \( k \) points and without strong interface contributions |
| \( \) | Sharp SHG peaks due to few contributing \( k \) points resulting in strong resonances | Doubled period and additional periods are frequency-dependent |
| \( \) | Strong frequency dependence of the SHG oscillation of the QWS in the \( k_{\perp} \) direction | MOKE period absent; doubled and additional SHG period visible |
| \( \) | MOKE period and larger periods visible; no exact doubling of the MOKE period | \( (x \text{ Au/Co}(0001)/\text{Au}(111) \text{ for example}) \) |
| \( \) | \( \) |
| \( \) | \( \) |

| No \( k \) selectivity | \( \) | \( \) |
|---------------------|---------------------------------|---------------------------------|
| \( \) | Strong magnetic signal, since strong interface contributes broad SHG peaks, since contributions come from many \( k \) points | Smaller magnetic contribution, since interface and (nonmagnetic) surface contributions are of the same magnitude |
| \( \) | Weak frequency dependence of the oscillation period | Broad, smooth peaks, since interference effects do not change magnitude |
| \( \) | MOKE period and larger periods present | SHG oscillation periods rather independent of the frequency, since the SHG signal is caused by the QWS near \( E_F \) |
| \( \) | \( \) |
| \( \) | \( \) |

If the interference of light from the surface and interface is negligible then different oscillations of the outgoing SHG light as a function of film thickness occur.

The weight of the optical transitions \( i \rightarrow j \rightarrow l \) changes as the film thickness increases, see the later discussion. Thus, (linear) MOKE and (nonlinear) Nonmoke oscillations occur.

Regarding optical properties, the morphology of the thin film and its magnetic domain structure should play a role in general.

Film multilayers. For magnetic film multilayers one expects interesting interfering transitions and magnetic optical behavior. Assuming for example the structure shown in figure 19, neglecting for simplicity QWS, one gets for SHG

\[
I(2\omega) \sim |\chi_s(\vec{M}) + \chi_{\text{int},1} + \chi_{\text{int},2} + \cdots|^2. \tag{39}
\]

Writing \( \chi = \chi^s + \chi^0, \chi^0 \sim M \), one has

\[
I(2\omega) \sim |\chi^s + \chi^0(\vec{M}) + \Sigma_i \chi^0_{\text{int},i} + \cdots|^2 \tag{40}
\]

for the AF structure. In the case of ferromagnetically aligned films it is

\[
I(2\omega) \sim |\chi_s + \Sigma_i \chi^0_{\text{int},i}(\vec{M})|^2. \tag{41}
\]

Note, to detect the magnetic domain structure, experiments must achieve high lateral resolution, via interferences for example, etc. Of course, surfaces with a mixture of domains with magnetization \( M_{\perp} \) and \( M_{\parallel} \) might yield a behavior as observed for the magnetic reorientation transition.

Table 1 lists some characteristic SHG properties. Here, MOKE refers to linear magnetooptics. In summary, the magnetization pattern of a multilayer system is clearly reflected by magnetooptics. The resolution of the optical response can be enhanced using interference effects.
type theory) that the magnetization in thin films changes
for heterogeneous structures to a film or a magnetic domain in
Here, $i$

The magnetic dynamics is described by (see the Landau–
Lifshitz equation)

\[
\frac{d\vec{M}}{dt} = -\left(\frac{\mu B}{h}\right)\vec{M} \times \vec{H}_{\text{eff}} + \frac{G}{M_s^2} \vec{M} \times (\vec{M} \times \vec{H}_{\text{eff}}). \tag{42}
\]

Here, $i$ refers to the magnetization of an elementary volume, or
for heterogeneous structures to a film or a magnetic domain in
a nanostructured film. $H_{\text{eff}}$ refers to a molecular field acting on $M_i$ and resulting, for example, from neighboring films
or magnetic domains. The first term describes precessional
motion and the second one relaxation. $M_s$ is the saturation magnetization (note, using $\vec{M} \times \vec{H}_{\text{eff}} \rightarrow \partial M_i / \partial t$ in the second
term on the rhs of equation (42) yields the Gilbert equation with
damping parameter $G$ describing spin dissipation).

One gets from equation (38) (and also from Boltzmann-
type theory) that the magnetization in thin films changes
during times of the order of (controlled by angular momentum
conservation), see [33]

\[
\frac{1}{\tau_M} \propto A(T_{el})|V_{11}|^2 \bar{N} \hat{N} + \cdots. \tag{43}
\]

Here, in the case of excited electrons $T_{el}$ may refer to the
temperature of the hot electrons, $V_{11}$ to the spin-flip scattering
potential causing changes in the magnetization (for example,
spin–orbit scattering or exchange interactions) and $\bar{N}$ is the
average DOS of the electrons [33].

Note, in the case of transition metals with many hot
electrons equation (43) may yield response times $\tau_M$ of the
order of 100 fs. Clearly, raising the temperature, $T \rightarrow T_{el}$
due to hot electrons, can speed up the magnetic dynamics.

The Landau–Lifshitz (LL) equation is generally important
for spin dynamics. One may use the spin continuity equation
($\partial_i M_i + \partial_j j_{ij,\sigma} = 0$) to determine the spin currents $\vec{j}_\sigma(t)$ (including the spin Josephson one) induced by magnetization
dynamics and then describe the latter by using the LL equation [12, 33, 34, 37].

As will be discussed one gets for magnetic tunnel
junctions, of course dependent on the spin mean free path $l_i$
(presumably best if $l_i > l_s$), the spin current ($j = -\partial F / \partial \phi$
where $F$ is the free energy and $\phi$ the phase, see Bloch [37])
driven by the phase difference $\Delta \Phi$ between the magnetization
of neighboring magnets of the form

\[
\vec{j}_\sigma \propto \partial \vec{M} / \partial t \sim \sin(\Delta \Phi). \tag{44}
\]

This follows from the LL equation and the spin continuity
equation (see nonequilibrium magnetism, [33]). Note, in
accordance with the GMR effect discovered by Grünberg and
Fert the tunnel current depends on the relative orientation of
the magnetization on the left-hand and right-hand side of the
tunnel junction [12].

2.6. Theory for magnetic films during growth: nonequilibrium
magnetic domain structure

The magnetic structure of thin films can be controlled by film
growth conditions, since magnetism depends on the atomic
structure of the film. Obviously this is of great interest for
engineering the magnetic properties of films [6–9, 27, 31].

During growth of the film one has a film structure
changing in time. This changes the magnetization. Thus, the
film magnetization also changes in time (note, magnetic
relaxation processes may occur somewhat time-delayed). Then

\[
\vec{M}(\vec{r}, t, \text{[atomic structure]}(t)) \tag{45}
\]
describes the nonequilibrium magnetization of growing films.
Clearly, the magnetic structure (domain structure) changes in
time $t$ as the atomic structure changes during growth of the
film. For simplicity one may use a kinetic MC simulation (MC:
Monte Carlo method) for molecular beam epitaxial film growth
(MBE) to calculate the film structure and the accompanying
magnetic one. The nonequilibrium behavior reflects sensitively
the magnetic properties of the film, relaxation processes, range
of magnetic forces and magnetic domain structure.
The atomic structure of the growing film is determined from a particular film growth model. For example, in the Eden-type growth model the atoms are randomly deposited (to sites \( i \)) with probability

\[ p_t = \alpha e^{-E_i/4kT}, \]  

(46)

where \( E_i \) is the atomic binding energy depending on the local coordination number \( z \) \( (E_i \approx -A\sqrt{z_i}) \). For an illustration see figure 9.

Different film growth is obtained by using a layer-dependent parameter \( A \). One may also take into account diffusion of the deposited atoms. This then yields irregular film structures during growth with varying clusters of deposited atoms (islands of different sizes and shapes).

The resulting magnetic structure consists typically of an ensemble of magnetic domains. The magnetic domains may not be at equilibrium and will respond to the time varying atomic structure of the growing film. This may occur time-delayed. Thus, successively atomic structure and magnetic structure change.

Magnetic relaxation flipping the domain magnetizations is described by using a Markov equation and an Arrhenius ansatz for the magnetic transition rates.

In summary, for each obtained atomic structure of the growing film one calculates the corresponding magnetic structure. Using the Heisenberg-type Hamiltonian including anisotropy it is obvious how, by changing the atomic coordination, the exchange interactions change and thus the magnetism. For simplicity one uses first equation (16). For details of the analysis see [7, 8, 31].

Spin relaxation assumes coherent rotation of the spins of each atomic cluster (deposited group of atoms). The magnetization (one may for simplicity an Ising model) of such atomic clusters is directed along the easy axis. Magnetic relaxation is (using Arrhenius-type rates for transitions \( M_i \rightarrow -M_i \)) given by the rate

\[ \Gamma_i = \Gamma_0 e^{-E_{i}^{b}/kT}. \]  

(47)

Here, \( E_{i}^{b} \) gives the energy barrier against switching of the domain magnetization. Note, if no energy barriers are present then one may use the Metropolis algorithm. The magnetic barrier energy of a cluster controlling the domain relaxation is, for example, given by

\[ E_{i} = N_{i}K_{i}(T)\cos^{2}\varphi - g\mu_{B}H_{i}S_{i}\cos\varphi + \cdots. \]  

(48)

The first term is due to magnetic anisotropy and the second due to the field \( H_{i} \) (interaction exchange, external magnetic field, dipolar field) acting on spin \( S_{i} \) [8, 31].

Generally an energy barrier \( E_{i}^{b} \) is present for the two magnetic orientations of magnetic cluster \( i \). One applies then as mentioned already a kinetic MC simulation during film growth to obtain the magnetic structure corresponding to a given atomic structure at time \( t \). Correlated relaxation of neighboring magnetic domains must be taken into account. Note, the time for each calculational step is set by the spin precession frequency \( \Gamma_0 \approx 10^{9} \text{--} 10^{12} \text{s}^{-1} \).

A successful analysis has been derived by Brinzanik et al [8, 31]. In their studies first the atomic structure of the film at time \( t \) is calculated using an Eden-type growth model. Then the corresponding magnetic structure is determined performing a kinetic MC simulation and using a Markov equation and for the system of magnetic domains (interacting via exchange and dipolar coupling) the energy

\[ E \approx -(1/4)\sum_{j}y_{ij}L_{j}S_{i} \cdot S_{j} = \sum_{ij}K_{j}N_{j}(S_{i}^{2})^{2} + \Delta E. \]  

(49)

Here, \( y_{ij} \) is the domain wall energy, \( L_{j} \) the island surface area and \( N_{j} \) the number of island atoms. The second term describes anisotropy. Also

\[ \Delta E = \sum_{i>j}(\mu_{i}/r_{ij}^{3})[S_{i} \cdot S_{j} - 3(S_{i}^{2} \cdot S_{j})(S_{i}^{2} \cdot S_{j})/r_{ij}^{2}] \]  

\[ - \sum_{ij}\mu_{i}\vec{B} \cdot \vec{S}_{i}. \]  

(50)

Here, for simplicity islands with \( N_{j} \) atoms and aligned magnetic moments are treated as particles with magnetic moments \( \mu_{i} \). The film magnetization results from averaging over the domain magnetization.

Typically one gets first for the given nanostructured film magnetism which is not at equilibrium. A non-saturated magnetization is typically obtained. As time progresses magnetic relaxation processes and magnetization reversals of domains, for example, occur. These change the magnetization of the film. Thus, finally equilibrium magnetization is obtained. For growing film thickness one might get a uniformly magnetized film, for details see the analysis by Jensen et al [7, 8, 31].

2.7. Tunnel junctions: spin currents

Tunnel junctions \( (N_{1}\mid N_{2}\mid N_{3}) \) are interesting nanostructures regarding (ultrafast) switching effects, interplay of magnetism and superconductivity, and, in general, quantum mechanical interference effects. Note, \( N_{1} \) may refer to material which is ferromagnetic or superconducting, for example. The situation is illustrated in figure 20. On general grounds one may get spin currents driven by the phase difference between two magnetic systems, ferromagnets or antiferromagnets (whose order parameter is also characterized by a phase) [12, 13, 37, 45]. In ring structures Aharonov–Bohm-induced currents are of special interest [34–37].

Interesting tunnel junctions are shown in figure 20. In figure 20(a) is shown a two quantum dot system with the occupation \( n_{i}\alpha \) of the states with energy \( \varepsilon_{i}\alpha \). These control the tunnel current \( j_{i}^{\alpha} \). The position of the energies \( \varepsilon_{i}\alpha \) may depend on occupation (see Hubbard-like Hamiltonian: \( \varepsilon_{i}\alpha = \varepsilon_{i}^{\alpha} + U n_{i}\alpha + \cdots \)). Note, the occupations \( n_{i}\alpha \) and thus the current \( j_{i}^{\alpha} \) can be manipulated optically.

For a detailed discussion of currents between quantum dots see [14]. of tunnel currents involving superconductivity see [10, 11] and of tunnel currents affected by magnetism see [13]. In general, tunneling sensitively reflects quantum mechanical behavior and interplay of magnetism and superconductivity.

The tunnel system shown in figure 20(b) can be used as a sensor for triplet superconductivity. Then the configuration of the angular momentum \( \vec{d} \) of the triplet Cooper pairs
Figure 20. Illustration of various tunnel junctions. (a) A two quantum dot system with spin-dependent levels $\epsilon_i^{\sigma} (n_i^{\sigma}, t)$ is shown (for example, $\epsilon_i^{\sigma} = \epsilon_0^{\sigma} + U_{Ni}\sigma$). Two levels are separated by an energy barrier which can be overcome using an external electric field. (b) A triplet superconducting Josephson junction (SC1|FM|SC3) is sketched. Here, $\vec{d}$ refers to the angular momentum of the Cooper pairs, whose order parameter has the phase $\phi$. (c) A junction (FM1|SC|FM2) is illustrated. An external electrical field (potential $V$) shifts the electronic energy levels (bands). This may control the tunnel current (magnetoresistance), see Takahashi et al. Of particular interest is the interplay of spin current and superconductivity.

and of the magnetization $\vec{M}$ of the tunnel medium control characteristically the tunnel current. The Josephson current exhibits interesting behavior, for example upon rotating the magnetization relative to the angular momentum of the Cooper pairs.

In figure 20(c) is illustrated how tunneling can be used (with the help of a bias voltage) to control magnetoresistance and to determine the magnetization of a ferromagnet (see [13]). Note, upon changing $(\uparrow|N_2|\downarrow) \rightarrow (\downarrow|N_2|\uparrow)$ the magnetoresistance increases, see the GMR effect by Grünberg and Fert [40, 41], and accumulation of spin-polarized electrons occurs in $N_2$. The importance of the tunnel junction structure has been discussed by Takahashi, Maekawa, Majumdar et al [38, 39]. More studies are needed for the influence of energy discretization due to confinement on tunneling.

Furthermore, if $N_2$ becomes superconducting a competition between superconductivity and magnetization occurs for configuration $(\uparrow|N_2|\downarrow)$ as a result of accumulating nonequilibrium spin density in $N_2$. In figures 20(b) and (c) we assume $\tau_s > \tau_t$, where $\tau_s$ and $\tau_t$ refer to the spin diffusion and electron tunneling time, respectively. Then electrons keep spin while tunneling.

For tunnel configuration (FM1|TSC|FM2) one expects similar interesting tunneling behavior, for example regarding dependence on relative orientation of $M_i$ and angular momentum $\vec{d}$ of the triplet Cooper pairs as for junctions (TSC/FM/TSC), see figure 20(b).

Furthermore, the current $j^s_\mu$ expected for $\tau_s > \tau_t$ depends on relative magnetization of the two magnets and on the state of $N_2$ (normal, superconducting singlet or triplet).

Spin currents: using the continuity equation one can find the relationship between spin currents and magnetization dynamics for the magnetic tunnel junction illustrated in figure 12. One has

$$\partial_t M_i + \partial_\mu j^s_{i,\mu,\sigma} = 0. \quad (51)$$

This may give, under certain assumptions, straightforwardly the connection between magnetization and spin-polarized electron currents (induced by hot electrons, temperature gradients or external fields). Note, the magnetic dynamics (characterized by $\partial_t M_i$) may be described by the LL equation including damping. This generally yields a spin Josephson current between magnets.

Also, according to Kirchhoff the emissivity ($e$) of the junction is related to its (time-dependent) magnetization and magnetic resistance. It is

$$\frac{\Delta e}{e} \simeq \alpha (\text{GMR}), \quad (52)$$

where GMR denotes the giant magnetoresistance resulting for the junction if the configuration $(\uparrow|N_2|\uparrow)$ changes to $(\downarrow|N_2|\uparrow)$. This changes the emissivity to $\Delta e$. For studies of GMR see Grünberg, Fert and recent publications in particular in Phys. Rev. Lett., Phys. Rev. and other journals.

Viewing the phase of the magnetic order parameter $\phi_i$ (magnetization $M_i$)

$$\vec{M} = |M_i|e^{i\phi_i} \quad (53)$$
Similarly as the phase of the SC state, one gets for a junction (FM/FM/FM) a Josephson-like current \( j^i \) driven by the phase difference of the spin polarizations on both sides of the \( j^i \) tunnel junction. Using the continuity equation for the spins, integrating using a Gauß integral and the Landau–Lifshitz equation, one gets for the spin current

\[
\dot{J}_S = J_{S}^i (V) + \dot{j}_i^1,
\]

\[
\dot{j}_i^1 \propto \frac{dM_i}{dt} \propto \tilde{M}_L \times \tilde{M}_R + \cdots \propto \left[ |M_i| |M_j| \right] \sin (\phi_i - \phi_j) + \cdots.
\]

(54)

Here, L and R refer to the left-hand and right-hand side of the tunnel junction, respectively. \( j_0^i \) refers to the spin current due to the potential \( V \) and may result from the spin-dependent DOS. For details of the derivation of the spin Josephson current see [12].

Note, using the general formula \( j = -\frac{dE}{d\phi} \), where \( F \) is the free energy, one gets

\[
\dot{j} = -\left( e/\hbar \right) \Sigma_i \partial E_i / \partial \phi \tanh (E_i / kT).
\]

(55)

Then using \( E_i \propto \dot{J}_S^0 \tilde{S}_L \cdot \tilde{S}_R + \cdots \), one also gets

\[
\dot{j}_i^0 \propto \dot{J}_S^0 \sin (\phi_i - \phi_j) + \cdots
\]

(56)

Here, \( E_i \) gives the energy difference between opposite directions of the magnetization (molecular field).

Of course, such a Josephson-like spin current is expected on general grounds, since \( \phi \) and \( S^z \) are canonical conjugate variables and (approximately)

\[
[\phi, S^z] = i.
\]

(57)

Note, this holds for the Heisenberg Hamiltonian as well as for the itinerant magnetism described by the Hubbard Hamiltonian, for example. The commutator relationship suggests in analogy to BCS theory to derive the spin Josephson current from the Hamiltonian

\[
H = -E_i S_i^2 \cos (\phi_i - \phi_j) + \frac{\mu_B^2}{2C_i} (\tilde{S}_L^z - \tilde{S}_R^z)^2 + \cdots
\]

(58)

where again L and R refer to the left-hand and right-hand side of the junction, and \( C_i \) denotes the spin capacitance. In general, spin relaxation effects should be taken into account (magnetization dissipation, see the LL equation). Using then the (classical) Hamiltonian equations of motion (\( \dot{\phi} = \partial H / \partial S^z \), \( S^z = -\partial H / \partial \phi \)) one gets

\[
\Delta \phi = 2\mu_B V_i, \quad \dot{j}_i^1 = (2E_i S_i^2 / \mu_B) \sin \Delta \phi.
\]

(59)

Here, \( \Delta \phi = \phi_1 - \phi_2 \) and \( V_i = (\mu_B / C_i) (\tilde{S}_L^z - \tilde{S}_R^z) \). That \( \Delta \phi = 0 \) if \( M_L \parallel M_R \) and \( \Delta \phi \neq 0 \) if \( M_L \) is antiferromagnetically aligned relative to \( M_R \) can be checked by experiment. It is \( \dot{j}_i^1 \sim \sin (\Delta \phi_0 + 4M_i \tilde{l}) \). Note, details of the analysis for the ac-like effect are given by Nogueira et al [12].

Thus, interestingly the spin current in an FM/FM tunnel junction behaves in the same way as the superconductor Josephson current. Of course, as already mentioned magnetic relaxation (see the Landau–Lifshitz–Gilbert equation) affects \( \dot{j}_i^1 \). For further details see again Nogueira and Bennemann [12, 45] and further recent studies by Sudbo and others in Phys. Rev., Phys. Rev. Lett., etc [45].

Clearly, one expects that junctions involving antiferromagnets (AF), (AF/F) and (AF/AF) also yield such Josephson currents, since the order parameter for an AF one gets a \( S^z \) and \( \phi \) are conjugate variables. (Treat antiferromagnetism (AF) as consisting of two ferromagnetic (FM) sublattices.) Equation (57) should hold for both \( J > 0 \) and \( J < 0 \), see the Heisenberg Hamiltonian.

Note also \( J_{S}^0 \propto J_{S}^0 (\chi) \) is a functional of the spin susceptibility \( \chi \), since the effective exchange coupling between the L and R side of a tunnel junction is mediated by the spin susceptibility of system \( N \), see figure 12.

The analysis may be easily extended if an external magnetic field \( B \) is present. Then from the continuity equation one gets \( j_i^1 \sim \frac{dM_i}{dt} \) and \( dM_i = a \tilde{M}_L \times \tilde{M}_R - g \mu_B \tilde{B}_L \times \tilde{S}_L + \cdots \) (and similarly \( dM_i = a \tilde{M}_L \times \tilde{M}_R + \cdots \)). Alternatively one may use the Hamilton–Jacobi equations with the canonical conjugate variables \( S^z \) and \( \phi (S^z = \frac{d\phi}{d\phi}, \phi = -\frac{\mu_B}{\hbar} \phi_0 + \cdots \) and changing the Hamiltonian \( H \rightarrow H - g \mu_B (\tilde{B} \cdot \tilde{S} + \tilde{B} \cdot \tilde{S}^\dagger) \) to derive the currents \( j_i^1 \) and \( j_i^2 \).

Note, according to Maxwell’s equations, the spin current \( j_i^1 \) should induce an electric field \( E_i \) given by

\[
\partial_i E_i - \partial_i E_i = -4\pi \mu_B \partial_i \langle S^z \rangle = 4\pi j_i^1.
\]

(60)

Here, for simplicity we assume no voltage and \( \partial_i B = 0 \) for an external magnetic field \( B \). Of course, in view of the Maxwell equations’ spin dynamics, the time-dependent tunnel spin currents are accompanied by (polarized) light.

Tunnel junctions with spin and charge current: generally one gets both a charge current \( j_e = -e \tilde{N}_L \) and a spin current \( j_s = -\mu_B (\tilde{S}_L^z - \tilde{S}_R^z) \) and these may interfere. This occurs, for example, for SCM/SCM junctions, where SCM refers to nonuniform superconductors coexisting with magnetic order (see the Larkin–Ovchinnikov state) and for a (SC/FM/SC) junction with a ferromagnet between two superconductors. Then one gets after some algebra for the Josephson currents (see Nogueira et al [45])

\[
\dot{j}_i^1 = (j_i + j_2 \cos \Delta \psi) \sin \left( \frac{2\pi l}{\phi_0} H_n \right)
\]

(61)

and

\[
\dot{j}_i^2 = j_i \sin \Delta \psi \cos \left( \frac{2\pi l H_n}{\phi_0} \right).
\]

(62)

Here, \( \phi_0 \) is the elementary flux quantum, \( H_n \) an external magnetic field in the \( y \) direction, \( l = 2\lambda + d \), with \( \lambda \) being the penetration thickness and \( d \) the junction thickness. \( \Delta \psi \) and \( \phi \) refer to the phase difference of magnetism and superconductivity, respectively. The magnetic field \( H_n \) is perpendicular to the current direction.

In the following some special tunnel junctions are discussed:

1. (TSC/FM/TSC) junction. Regarding switching of tunnel current and analysis of triplet superconductivity the tunnel junction (TSC/FM/TSC) is of interest. Here, TSC refers to triplet superconductivity. The Josephson current then flows through low-energy Andreev states. Relative
orientation of the magnetization $\tilde{M}$ of the ferromagnet (FM) and $d$-vectors of the triplet superconductors, see figure 20(b), control the tunnel current. One gets, see [10, 11]:

$$j_{i}^{c} = -\frac{e}{h} \sum_{i} \frac{\partial E_{i}}{\partial \phi} \tanh \left( \frac{E_{i}}{kT} \right),$$  \hspace{1cm} (63)

where $\phi$ is the phase difference between the two superconductors and $E_{i}$ are the energies of the Andreev states and which are calculated using Bogoliubov–de Gennes analysis. Thus one derives an unusual temperature dependence of the Josephson current on temperature and even that $j_{i}^{c}$ may change sign for certain directions of $\tilde{M}$, although $\Delta \phi$ did not change.

(2) (SC/FM/SC) junction. As discussed recently by Kastening et al [10, 11] such junctions characteristically reflect magnetism. Again the tunnel current is carried by Andreev states. No net spin current flows from left to right, since the spin-polarized current through the Andreev states is compensated by the tunnel current through continuum states, as must be the case due to basic physics. In the case of strong ferromagnetism single electrons tunnel, while for weak ferromagnetism Cooper pair tunneling occurs. The Josephson current may change sign for increasing temperature without a change in the relative phase of the two singlet superconductors. Of course, dependent on coherence length, temperature and thickness of the FM and the strength of the FM one may get $j_{i}^{c} = 0$ for the Josephson current.

(3) (FM/SC/FM) junction. It is already obvious from figure 20(c) that, in the presence of an applied voltage $V$, junctions (FM1|SC2|FM3) carry currents which depend sensitively on the relative orientation of the magnetization of the two ferromagnets. If these are directed in opposite directions (AF configuration) one gets a maximal spin accumulation in the superconductor and thus one may suppress (at a critical voltage) superconductivity in singlet superconductors. As a consequence the magnetoresistance changes. Hence, such junctions exhibit currents:

$$J_{\sigma} = \sigma J_{c}(\tilde{M}_{1}, \tilde{M}_{3}, \Delta_{2}, T)$$  \hspace{1cm} (64)

which reflect sensitively superconductivity and ferromagnetism ($\Delta_{2}$ denotes the order parameter of SC$_{2}$).

Of course, the current $J_{\sigma}$ is affected by temperature gradients $\Delta T$ between the ferromagnets and resulting, for example, at nonequilibrium from hot electrons in one FM. This may cause interesting behavior.

Induced currents into rings. The equation for the current due to $\partial E_{i}/\partial \phi$, for the current driven by the phase dependence of the electronic energy, is of general significance. From it one may derive the currents induced in rings, discs, etc, if a magnetic field is present due to the Aharonov–Bohm effect. Note, $j = -\sum_{i} \frac{\partial E_{i}}{\partial \phi} \tanh \left( \frac{E_{i}}{kT} \right)$ and the electronic energies $E_{i}$ may be calculated for rings using the extension of the Balian–Bloch theory by Stampfl [16]. Thus one gets $j_{AB} \sim (1/2\pi R) \sum \left( \cdot \cdot \cdot \right) \sin(S_{i}B) \sin(\phi_{i} + \cdot \cdot \cdot)$, where $R$ refers to the radius of the mesoscopic ring, $i$ to the electronic orbits (characterized also by $t$, $p$, see Stampfl), $S_{i}$ is the area enclosed by the orbit $i$ and $B$ is the external magnetic field. The factor $(\cdot \cdot \cdot)$ in front of $\sin(\cdot \cdot \cdot)$ gives the weight by which the various orbits contribute and can be calculated straightforwardly, see Stampfl.

Note, one expects also in the case of electrolytes such induced currents, smaller due to the heavier mass of ions (and viscosity of the fluid) as compared to electrons. Of interest is the motion of charged impurities in superfluid He, for example.

Also a ring structure hollow at the center (and with magnetic flux $\Phi$) and outer ring superconducting and a neighboring inner ring ferromagnetic and which is also next to the hollow center may exhibit interesting behavior in the presence of an external magnetic field $B$.

2.8. Quantum dot systems

Interesting current patterns and interferences are also expected for the quantum dot grain structure shown in figures 3 and 20(a). Assuming a mixture of ferromagnetic and superconducting grains (clusters, quantum dots) one has the Hamiltonian

$$H = H_{T} - \langle \frac{z^{2}/C}{\sum_{i,j} N_{i} - N_{j}} \rangle + \frac{1}{\rho_{s}}.$$  \hspace{1cm} (65)

Here, $H_{T}$ denotes the tunneling Hamiltonian. This also includes spin Josephson currents of the form $I_{ij}^{s} \cos \varphi_{ij}$ and also Josephson Cooper pair current contributions resulting from the phase difference $\varphi_{ij} = \varphi_{i} - \varphi_{j}$ of the phases of the SC order parameter of grains $i$ and $j$. The electrostatic effects due to different charges of grains $i$, $j$ are given by the second term. $H'$ denotes the remaining effects, for example due to ferromagnetic grains. One expects characteristic differences for singlet and triplet superconductivity and interplay of spin currents $j_{sp}$ between magnetic grains and Josephson currents.

In grains the Cooper pair size and the distance between Cooper pairs can be manipulated by confinement (for example, via superfluid density $\rho_{s}$). This is expected to exhibit interesting behavior.

For a lattice-like array of quantum dots it is of interest to study phase ordering of the order parameter of the various dots $i$, its dependence on distance between dots, etc (see the related situation for superconductors when $T_{c} \propto \rho_{s}$, where $\rho_{s}$ is the superfluid density).

As speculated for two-band superconductors and as assumed for sc $q$-bits, (strong) phase coupling of two magnetic systems (quantum dots) may cause a covalent splitting-like process (mode–mode coupling-like process). Thus, for example, one expects for the combined phase-coupled system $N_{1}$ and $N_{3}$, see figure 20, the two covalent-like split states $|N_{1}N_{3}\rangle$ and $|N_{1}N_{3}\rangle$.

Light irradiation causes interesting responses, since the occupation of electronic states (by single electrons or Cooper pairs) can be manipulated.

Also note for a mixture of superconducting and magnetic quantum dots on a lattice (and the presence of an external magnetic field $B$) one may expect interesting quantum mechanical effects [17, 18].
These examples may suffice already to demonstrate the interesting behavior displayed by nanostructures involving tunneling. This holds also for currents across multilayers of magnetic films. Note, the general formula for the current $j = \frac{e}{4\hbar} F \phi$ can also be applied to transport across multilayer magnetic film structures, see GMR behavior, etc.

3. Results

Characteristic results are presented for clusters, films and tunnel junctions.

3.1. Small magnetic particles

3.1.1. Single magnetic clusters. In small clusters the local DOS exhibits the dependence of the electronic structure on the local atomic environment of a cluster atom. In figure 21 the local DOS is compared with the average DOS. Results were obtained by Pastor et al. using the tight-binding Hubbard Hamiltonian [1, 2].

The DOS is the most important property of the electronic structure and determines the thermodynamical and magnetic behavior ($\mu_n, M_n(T)$).

In table 2 results for the bond length $d_n$ ($d_b$ refers to bulk), cohesive energy $E_{coh}(n)$ and magnetic moments $\mu_n$ are given for Ni$_n$ clusters.

In figure 22 the dependence of the magnetic properties on cluster size is given. The variation of $\mu_n$ reflects the atomic structure (shell structure). These results were obtained by Mukherjee et al. using different cluster shell models for the magnetic moment $\mu(N)$. $\mu(N)$ is calculated by averaging over the shell (coordination)-dependent moments $\mu_i$ and where these are given by the local atomic coordination (varying with $N$). Crosses refer to experimental results, and solid and dashed curves refer to calculations by Jensen using different models for the dependence of the local magnetic moments $\mu_i$ on atomic coordination. Of course, for larger clusters $\mu_i \to \mu_b$.

In figure 23 results for $\mu_n$ of Fe$_n$ are given. The results were obtained using the Hubbard Hamiltonian and a tight-binding-type calculation by Pastor et al. [1, 2, 22, 23]. For Fe one expects a particular strong interdependence of magnetism and structure (see bcc versus fcc Fe). For each cluster one assumes a structure which yields the largest cohesion. The variation of $\mu_{Fe}$ as a function of $n$ is due to the interplay of changes in coordination number and bond lengths (note, approximately $W_n \propto d^{-3}$, where $W$ is the bandwidth and $d$ the bond length). Interestingly, one gets for the small clusters $\mu_{Fe}$ larger than $\mu_b$, magnetic moments which are larger than the bulk one, as expected. The sensitive dependence of the Fe
Table 2. Results for Ni$_n$ clusters by Pastor et al. $E_{\text{coh}}$ denotes the cohesive energy, $d_b$ the bond length and $\mu(i)$ the local magnetic moments.

| Structure | $E_{\text{coh}}$ (n) | $d_b$ | $\mu_n$ | $\mu(1)$ | $\mu(2)$ | $\mu(3)$ | $\mu(4)$ |
|-----------|----------------------|-------|---------|---------|---------|---------|---------|
| Ni$_2$    | 0.66                 | 0.92  | 0.95    | 0.95    | (0.52)  | (0.96)  | (0.96)  |
| Ni$_3$    | 0.83                 | 0.92  | 0.28    | 0.28    | (0.68)  | (0.27)  | (0.27)  |
| Ni$_4$    | 0.87                 | 0.92  | 0.92    | 0.87    | 0.95    | (0.70)  | (0.96)  | (0.90)  | (0.99)  |
| Ni$_5$    | 0.93                 | 0.94  | 0.51    | 0.51    | (0.81)  | (0.53)  | (0.53)  |
| Ni$_6$    | 0.96                 | 0.96  | 0.72    | 0.72    | (0.89)  | (0.96)  | (0.96)  |
| Ni$_8$    | 1.0                  | 0.96  | 0.96    | 0.96    | (0.90)  | (0.97)  | (0.97)  |
| Ni$_{13}$ fccc | 1.05            | 0.97  | 0.82    | 1.20    | 0.79    | (1.01)  | (0.82)  | (1.17)  | (0.79)  |
| Ni$_{13}$ icos. | 0.99          | 0.96  | 0.97    | 1.27    | 0.94    | (0.91)  | (0.96)  | (1.23)  | (0.94)  |
| Ni$_{15}$ fccc | 0.99            | 0.97  | 0.0     | 0.0     | 0.0     | (0.95)  | (0.0)   | (0.0)   | (0.0)   |
| Ni$_{15}$ icos. | 0.97         | 0.95  | 0.0     | 0.0     | 0.0     | (0.85)  | (0.0)   | (0.0)   | (0.0)   |
| Ni$_{19}$ fccc | 1.15           | 0.97  | 0.0     | 0.0     | 0.0     | (1.08)  | (0.0)   | (0.0)   | (0.0)   |
| Ni$_{19}$ icos. | 1.22          | 0.95  | 0.0     | 0.0     | 0.0     | (1.09)  | (0.0)   | (0.0)   | (0.0)   |
| Ni$_{19}$ fccc | 1.08           | 0.99  | 0.78    | 1.08    | 0.78    | (1.04)  | (0.78)  | (1.03)  | (0.78)  | (0.73)  |
| Ni$_{43}$ fccc | 1.14           | 0.69  | 0.68    | 0.77    | 0.69    | (0.61)  | (0.69)  | (0.61)  |

magnetic moments and magnetism on structure is of general importance for nanostructures and of material involving Fe and Fe under pressure, see [1, 2, 22–24].

One may also calculate using molecular-field-type methods the Curie temperature $T_c$ as a function of cluster size [23, 28, 29]. Results are shown in figure 24. Typically one gets that $T_c$ increases for increasing cluster size. Note, the exchange interaction $J$, $q_{\text{eff}}$ and $\mu_i$ may change as the cluster grows.

Orbital magnetism: clusters of other low-dimensional systems (surfaces, thin films) also exhibit interesting orbital magnetism. For example, for Ni clusters a remarkable enhancement of the average orbital moment as compared to bulk is observed. Note, one gets for the orbital moment of Ni (L(Ni$_7$)) $\approx 0.5 \mu_B$ while for bulk (L(bulk)) $\approx 0.05 \mu_B$. Using the tight-binding Hubbard Hamiltonian including the spin–orbit interaction ($H_{\text{so}} = -a \sum (\vec{L} \cdot \vec{S})_{\alpha, \beta} a^\dagger a$), Pastor, Dorantes-Davila et al [21] obtain for the average orbital moment (L$_{\delta}$) the results shown in figure 25. Here, $\delta$ refers to the cluster crystal axis. The important DOS $N_{\delta}^{\text{orb}}(\varepsilon)$ is determined self-consistently for each orientation $\delta$ of the magnetization with respect to structure. The s–o coupling connects spin-up and spin-down states (dependent on relative orientation of $\vec{M}$ and atomic structure). ($\vec{L} \cdot \vec{S}$)$_{\alpha}$ are intra-atomic matrix elements.

The results show that the orbital magnetic moment is an important contribution to the magnetic moment of clusters and quite generally to the one of the nanostructures of transition metals (TM). ($\vec{L}$) depends sensitively on band-filling. Also note, for example, ($L_z - L_x$), etc, reflect magnetic anisotropy. For Ni the orbital moment aligns parallel to the spin moment.

Mie scattering: for determining magnetism in small clusters besides Stern–Gerlach-type deflection experiments (as performed by de Heer et al [47, 49]) Mie scattering could be used. Extending the usual Mie scattering theory to magnetic clusters one calculates from the Maxwell equations with spin–orbit coupling, coupling the electromagnetic field and the cluster magnetization, and with a dielectric function $\varepsilon$ of the tensor form the Mie backscattering profile. Characteristic differences are obtained for Ni, Co and Fe. For example, magnetic Mie scattering is important for spherical clusters with radii approximately larger than 6 nm for Fe and 10 nm for Co; for details see [15].

Generally $M$ has a weak effect on the electromagnetic field. This is strongest when the size of the cluster is of
Figure 23. Dependence of average magnetic moment of Fe$_n$ on cluster size. Note the dependence of the results on the relaxation of the atomic structure, see calculations using an electronic theory by Pastor et al [22, 24]. For each $n$ a structure with largest cohesion is assumed. For Fe clusters one expects a sensitive dependence on structure and possibly a particularly strong interdependence of magnetism and structure. The vertical bars refer to experimental results for the depletion factor and which are assumed to be proportional to the average magnetic moment, at least approximately.

Figure 24. The Curie temperature $T_C$ ($M(T) = 0$) as a function of the number of cluster atoms. The calculations by Jensen et al assume two different cluster structures [6, 19, 48].

Figure 25. Orbital moment per atom ($L_z$) for a pentagonal bipyramid transition metal model cluster of seven atoms as a function of $d$-state filling (band-filling) $n_d$. Bulk next-nearest-neighbor atomic distances are taken. $\delta$ refers to the magnetization direction taken along (principal $C_n$ symmetry) cluster axis ($\delta = z$: full curves, $\delta = x$: dashed curves, for details see Pastor, Dorantes-Davila et al [21]). Note, full and open circles refer to calculations using different electron energies $\epsilon_{i\sigma}$.

$\theta = \pi/2$ the backscattering intensity $I(M = 0)$ is very small and then the changes due to the cluster magnetization are relatively large. In figure 26(b) it is $\epsilon_0 = -1.569 + i5.58$ and $\epsilon_{xy} = -0.059 + i0.122$ is used. Note, in figure 26(c) we use for the dielectric function $\epsilon_z = -4.592 + i7.778$ and $\epsilon_{xy} = -0.1613 - i0.0733$.

Mie scattering exhibits also characteristic differences regarding the angular dependence of the backscattering intensity for Cr, Fe, Co and Ni.

3.1.2. Ensemble of clusters. To determine cluster magnetism from the behavior of an ensemble of magnetic clusters in a magnetic field, for example, one must take into account the (mechanical) rotation of the clusters (due to the cluster production mechanism) and that the magnetization is tied to the cluster easy axis and cluster superparamagnetism. One expects in general strong reflection of the magnetic anisotropy of the cluster. An external magnetic field $H$ will tend to align the magnetic clusters. The (typically superparamagnetic) cluster magnetization points in the direction of the easy cluster axis. This is set by the anisotropy axis of the atomic cluster structure. This should be reflected then in the dependence of the cluster ensemble magnetization on an external magnetic field.

If the temperature $T_{bl}$ characterizes the energy which pins the cluster magnetization to its easy axis, different behavior is expected for the ensemble magnetization $\langle \mu \rangle$ for temperature $T > T_{bl}$ (Langevin behavior) and $T < T_{bl}$.

The behavior of a cluster ensemble in an external magnetic field can be studied by a (Stern–Gerlach-like) deflection experiment (using an external magnetic field to deflect the cluster beam), see calculations by Jensen et al [19, 20] and experiments by de Heer et al [47] and by Bloomfield et al [47, 49], etc. Note, in contrast to single-atom (molecule)
deflection metal clusters are deflected in only one direction (as a result of internal angular momentum transfer).

One expects that the rotating transition metal clusters, for example, behave largely as superparamagnetic particles and thus the deduced cluster beam magnetization should correspond to an ensemble average of particles (clusters) with random orientation of their magnetic moments. Note, this will lead generally to small cluster beam magnetization, even if the atomic magnetic moments are of the same order as the bulk ones. In addition the (beam) magnetization depends strongly on whether the cluster temperature is below or above the blocking temperature $T_{\text{bl}}$ (note $T_{\text{bl}} \approx nK_2/\ln(\tau/\tau_0)$, where $K_2$ characterizes the anisotropy energy per atom, $n$ the number of cluster atoms, $\tau$ the measurement time and $\tau_0$ the gyromagnetic
precession time). Clearly, for temperatures $T$ larger than $T_{\text{bl}}$ superparamagnetic (Langevin) behavior is expected. For temperatures below $T_{\text{bl}}$ cluster anisotropy determines the small beam deflection due to the small ensemble average over the spins of the superparamagnetic clusters pointing in different directions. Of course, superparamagnetic behavior depends on the cluster temperature.

In figures 27(a) and (b) results are given for the magnetization of a cluster beam, see [19, 20, 48] and experiments by de Heer et al [47] and Bloomfield et al [47, 49]. Note, as discussed in a magnetic field $H$ cluster anisotropy, characterized by the blocking temperature $T_{\text{bl}}$, and $H$ interfere. For temperatures $T < T_{\text{bl}}$ the internal anisotropy needs to be overcome to align the cluster magnetization via the external field. Figure 27(a) refers to calculations of the cluster beam magnetization by Jensen for temperatures below and above the blocking temperature (the dotted curve is the Langevin one) and figure 27(b) compares theoretical results by Jensen with experimental ones for Co by Bloomfield et al where the temperature is above the blocking temperature.

Results shown in figure 27 give the magnetization observed in (Stern–Gerlach-like) deflection experiments for a cluster beam, an ensemble of clusters in a magnetic field $H$. Obviously magnetic anisotropy within the cluster may determine strongly the magnetic behavior. As discussed the temperature $T_{\text{bl}}$ characterizes the pinning of the magnetization to the easy axis of each (rotating) cluster. Thermally activated depinning occurs for $T > T_{\text{bl}}$ (see calculations by Jensen et al [19, 20, 48]). Clearly, for temperatures larger than the blocking one Langevin-type behavior is expected. For lower temperatures one needs a certain strength of the external magnetic field ($\mu H \geq kT$) for unpinning the magnetization from its easy axis.

Note, interesting magnetic resonance effects are also expected for a cluster beam in an external magnetic field due to cluster rotation and magnetic anisotropy. According to the Bloch equation a minimum results for the averaged cluster beam magnetization for $\gamma H_{\text{ext}} < p \omega_{\text{rot}}$, where $\gamma$ is the gyromagnetic ratio, $H_{\text{ext}}$ the external magnetic field, $p$ the symmetry multiplicity of the anisotropy axis and $\omega_{\text{rot}}$ the rotation frequency, see the theory by Jensen, Bennemann [19, 48]. This has been observed by de Heer et al [47] and by Bloomfield et al [49].

Quantum dot lattices: typical results for a lattice of quantum dots or anti-dots are shown in the following figures. Of course, one expects that the density of states $N(E)$, generally spin-dependent if magnetism is involved, characteristically reflects the nanostructure.

In figure 28 results for the DOS oscillations are given which demonstrate their dependence on the dimension and geometry of the nanostructure. For a sphere mainly paths with $t = 1$ and $p = 3$ and 4 contribute. Their interference yields a beating pattern. For circular rings path 3 is the most important (see figure 13) and note the larger discrepancy between classical and quantum mechanical calculations. For circular discs note $A_i \sim k^{-1/2}$ (in contrast to spheres with $A_i \sim k^{1/2}$).

Approximately the spin-up and spin-down DOS are split by the molecular field $h_{\text{eff}}$ in the case of ferromagnetic nanostructures. It is of interest to study the interplay of superconductivity and magnetism and magnetic field $B$.

In figure 28 the DOS results obtained from the semiclassical Balian–Bloch–Stampfli theory [16] are compared with quantum mechanical calculations. The oscillating part of the DOS due to the interference of the dominant electronic paths is shown for various nanostructures. The quantum mechanical calculations were performed using the Schrödinger equation and Green’s function theory:

$$N(E, \ldots) = \frac{(\gamma/\pi) \int dE' \frac{N(E')}{(E - E')^2 + \gamma^2}}{66}$$

Of course, the parameter $\gamma$ must be chosen as usual such that it does not affect the structure in the DOS.

Note, in the case of magnetism one has a spin-dependent DOS $N_{\sigma}(E)$ and to simplify one may assume that spin-up and spin-down states are split by the molecular field $h_{\text{eff}}$ which may include an external magnetic field $B$. 

![Figure 27](image_url)
Bohm effect. In ferromagnetic rings the induced currents are reflected in the electron current induced by the Aharonov–Casher effect. The flux due to the external field changes the orbits and thus their interference. As a result DOS oscillations occur. Note, only a few closed electron orbits (1, 2, 3, etc) give the dominant contribution to the electronic structure, the DOS, see the theory by Stampfli et al [16]. The inset shows the lattice of anti-dots. Note, the scattering may be spin-dependent and the density of states is spin-split. Note $2R_c \geq (a - d)$ is assumed.

Also hollow particles (see coated nanoparticles, for example) can be treated similarly as rings. This is of interest, for example, to study confined and bent 2d electronic systems.

In figure 29 results are given for the oscillating part of the DOS as a function of energy and external magnetic field for a lattice of anti-dots which repel the electrons. The magnetic field $B$ changes the orbits and thus their interference. As a result DOS oscillations occur. Note, only a few closed electron orbits (1, 2, 3, etc) give the dominant contribution to the electronic structure, the DOS, see the theory by Stampfli et al and equations (19) and (21). The magnetic field causes path deformation and phase shifts. The flux due to the external field $B$ is $\phi = SB$ and for orbits 1, 2 and 3 $S$ is approximately independent of $B$ for a small field. The cyclotron orbit radius is $R_c = \frac{\hbar}{c B}$ and we assume for simplicity $2R_c > (a - d)$ and $R_c \gg R$. Then, $R_c \sim 1/l$ and the DOS depends on $(a/l)$ and $\Delta N \sim \cos SB$. This controls also the height of oscillations (note $S \propto R_c^2 \propto \frac{1}{B}$).

For increasing external magnetic field $B$ the oscillations in the DOS change. Interestingly, the contributions of some orbits may be nearly eliminated. The oscillation period in $B$ decreases for increasing $B$ and decreasing lattice constant $a$.

Note, an internal molecular field due to magnetism is expected to affect the DOS similarly as the external magnetic field. Note also the above oscillations in the DOS result from the confinement (closed electronic orbits) and additional structure may result from detailed atomic structure yielding the well-known electronic shell structure in clusters, etc.

Note, for orbit 4 (see the figure) and similar ping-pong orbits one has $\cos(SB) \approx 1$. Of course, for increasing scattering potential $U$ the amplitude and interference changes ($R_c \sim 1/B$). For decreasing field $B$ dephasing occurs and must be included.

For increasing radius the results for thin rings may be similar to the ones for planar surfaces of thin films. For thin films one may perform Balian–Bloch-type calculations as sketched in figure 30. Note, the structure in the DOS should be reflected in the electron current induced by the Aharonov–Bohm effect. In ferromagnetic rings the induced currents are spin-polarized. Spin–orbit coupling may cause spin-polarized induced currents. Of special interest is the additional effect of a magnetic field on the induced current. Also confinement discretization of the electron energy spectrum may play a role and in two-fluid superconductivity the coupling between the normal and superfluid part. Generally one expects the induced ring currents to decrease with increasing ring radius (dependent on scattering) and increasing temperature (see the formulae for $j$).

3.2. Thin magnetic films

Thin films are of central importance for engineering magnetic material on the nanoscale. Thin transition metal (TM) films, for example, exhibit magnetic properties of interest for many technical applications like magnetic recording. In the following typical results are given which show the dependence of film magnetism on the atomic structure, topology of the film and on film thickness. Magnetic anisotropy is very important. Magnetism is characterized by the magnetization as a function of film thickness and anisotropy by the orientation of the magnetization at the film surface and by the Curie temperature $T_c$. Nanostructured films exhibit characteristic magnetic domain structures, depending on film growth conditions (controlling the film morphology). Then depending on the distances between the domains various magnetic interactions controlling the global film magnetization come into play.

3.2.1. Magnetic structure. The magnetic properties of thin ferromagnetic films have been studied intensively. Due to anisotropy long range ferromagnetic order occurs even for ultrathin, quasi-2d films. Competing anisotropic forces determine the orientation of the magnetization relative to the atomic lattice. We use the Hamiltonian given in equation (16) with a local anisotropy:

$$H_{\text{anis}} = -\Sigma_i (K_2 \cos^2 \theta_i + K_4 \cos^4 \theta_i + K_s \sin^2 \theta_i).$$  (67)

Note, for $T > 0$ the in-plane anisotropy $K_s$ becomes important. After some standard algebra (see [9, 30]) one finds the energy per spin. One gets for

![Figure 29. Oscillations of the DOS as a function of magnetic field $B$ for orbits 1, 2 and 3 due to scattering by repelling anti-dots, see the calculations by Stampfli et al [16]. The inset shows the lattice of anti-dots. Note, the scattering may be spin-dependent and the density of states is spin-split. Note $2R_c \geq (a - d)$ is assumed.](image)

![Figure 30. Sketch of the important orbits for thin films. Here, one has to include transmission ($T$) and reflection ($R$) coefficients at the corners of the paths. Magnetic films yield spin-dependent DOS. Note $T$ and $R$ are spin-dependent at the Cu/Co interface.](image)
dashed curves indicate what happens if only uniform phases are
respectively. Phases III and IV refer to uniform magnetization with
exchange coupling and magnetic anisotropy. Phases I and II refer to
demagnetization energy.) The anisotropy constants \( K \) are given in equation (49); their physical significance is clear. The
Figure 31. Magnetic structure of thin films obtained at \( T = 0 \) due to
exchange coupling and magnetic anisotropy. Phases I and II refer to
stripe domain structure with perpendicular and canted magnetization,
respectively. Phases III and IV refer to uniform magnetization with
canted and in-plane magnetization, respectively. (\( E_o \) is the
demagnetization energy.) The anisotropy constants \( K \) are given in equation (49); their physical significance is clear. The
dashed curves indicate what happens if only uniform phases are
considered.

(1) Uniform magnetic phases:
\[
E_u(\theta) = -(9/2)J - K_2 \cos^2 \theta - K_4 \cos^4 \theta - K_s \sin^4 \theta
+ E_o(\cos^2 \theta - 1/3),
\]
where \( \theta \) denotes the angle between film normal and magnetization, and for
(2) Stripe domains with wall width \( b \) and domain periodicity \( 2L \):
\[
E_{\text{dom}}(\theta) = -J/2(q - \pi^2/bL)
+ K_2(\cos^2 \theta(1 - b/L) + b/2L)
+ K_4(\cos^4 \theta(1 - b/L)) + 3b/8L
+ K_s(\sin^4 \theta(1 - b/L) + 3b/8L) + \cdots.
\]
Here, \( \theta \) denotes the canting angle of the domain magnetization along the \( z \) axis \((\theta = 0)\). \( q = 4 \) for a square
monolayer. The domains have the periodicity \( 2L \) along the \( y \) direction and have a uniform magnetization along the \( x \) direction, inside the domains the magnetization is along \( \pm z \) direction. This domain configuration is assumed for
simplicity. One gets rich magnetic behavior for thin films. Magnetic
anisotropy is important.

Typical results for the magnetic structure are given in figure 31. Generally one gets interesting phase diagrams, see [6, 30].

Multilayers of magnetic films reflect how the magnetization of neighboring films interfere and affect each other, and
act like a molecular field on the magnetization of a particular film.

In figure 32 results are given for multilayer films Cu–Ni–Cu and Co–Cu–Ni. These display the interfilm exchange coupling and magnetic influence of a magnetic film on neighboring films [50]. Note, one finds for the Curie temperature \( T_c(\text{Co}) > T_c(\text{Ni}) \). Clearly, the Co film causes
that the magnetization of the Ni film is present at higher temperatures. This results from interlayer exchange coupling, see the experimental studies by Baberschke et al [50].

The results shown in figure 33 demonstrate the dependence of \( T_c \) on film structure. Obviously, interesting characterization of magnetic films is also given by their Curie temperature, see [28].

3.2.2. Reorientation transition of the magnetization. The reorientation transition of the magnetization at surfaces (and interfaces) is very important regarding applications, for example, imprinting a magnetic pattern at surfaces, but also regarding an understanding of magnetic anisotropy. Such a transition may be driven by temperature, film thickness
and film topology (and at nonequilibrium by hot electrons), see [25, 26]. Magnetooptically the reorientation transition is characterized by the linear response \( \chi_{ij} \) or the nonvanishing elements of the nonlinear susceptibility \( \chi_{ijkl} \), for example.

In the following typical results for the reorientation transition of the magnetization are presented. In figure 34 results are given (a) for a thickness-induced reorientation transition in bcc Fe films, (b) for a temperature-induced transition at \( T_R \) and (c) for the dependence of the phase diagram (PD) on anisotropy constants, \( K_4, K_4^\perp \) and \( K_4^\parallel \) are taken into account. The free-energy change \( \Delta F = F(T, \theta = 0) - F(T, \theta = \pi/2) \) is calculated with \( F = \Sigma_i K_2(i)(m(T)) \cos^2 \theta + \cdots \). Here, \( i \) refers to the film layer and \( \theta \) to the angle between film normal and direction of magnetization, see [6, 25, 26]. Note, \( \Delta F < 0 \) indicates a
perpendicular magnetization and \( \Delta F > 0 \) an in-plane one.

Of course, for thin films the magnetization pattern of the domains is important. The reorientation transitions, in particular at nonequilibrium may play a crucial role.

3.2.3. Magnetooptics. Magnetism of thin films and multilayers thereof is well determined by their magneto optical behavior. In particular SHG, sensing sensitively structural
symmetry, surfaces and interfaces, is very useful for studying magnetic properties of nanostructures. Magnetic thin films cause characteristic SHG reflecting magnetism, since the nonlinear susceptibility $\chi_{ijl}$ determining the light intensity $I(2\omega)$ of SHG depends on magnetization $\vec{M}$.

For the analysis one splits as usually the susceptibility into parts even and odd in $M$. Then the magnetic contrast

$$\Delta I(2\omega) \propto I(2\omega, M) - I(2\omega, -M)$$  \hspace{1cm} (70)

exhibits sensitively the magnetic behavior, see [32]. Resonantly enhanced SHG transitions of the ferromagnet whose states are spin-split are a sensitive measure of magnetism. Note, the interference of SHG from the film surface and interface (film/substrate) is of interest, in particular if magnetism is different at the surface and interface, see figure 11.

In table 1 characteristic features of the SHG response from thin films are given. We refer to weak interference of SHG when $\chi^i \gg \chi^s$ and to strong interference when $\chi^i \approx \chi^s$. Note, the wavevector $k$ selectivity is controlled by band structure. For the film system Cu/Fe/Cu the dipole matrix elements with d-states cause $\chi^i \gg \chi^s$. For Au/Co/Au films no Co d-states are involved and therefore $\chi^i \approx \chi^s$. The periodic appearance of QWS for increasing film thickness may cause oscillations in SHG.

In thin films (which may be described with respect to their electronic structure by potential wells) one gets QWS (quantum well states) and corresponding contributions to MSHG (magnetic second harmonic light). As supported by experiments QWS in thin films may contribute strongly to SHG (quasi-resonantly enhanced SHG). Since the occurrence of QWS is dependent on film thickness, one gets QWS involving characteristic oscillations in SHG as a function of film thickness. These oscillations depend, of course, on light frequency, magnetism, parity of the QWS, inversion symmetry of the film and interference of SHG from the surface and interface.

If interference of nonlinear light from the surface and interface is important, then SHG response depends on QWS parity. As a consequence only a period twice as large as for the linear Kerr effect optics appears. If this interference is not so important, then SHG exhibits typically two oscillation periods. For details of such behavior see Luce, Bennemann [12] and also the discussion given previously, see figure 18, etc.

SHG involving QWS is shown in figure 35. In figure 35(a) thin film QWS and their parity are sketched. The figures refer to the $\alpha$Cu/Fe(001) system. For other systems the analysis is similar. Figure 35(a) shows the energies of the QWS. The parity is indicated by (+) and (−). Note, at the Fermi energy a parity change occurs. The spin-polarized d-states of Fe are also shown. In figure 35(b) the dominant SHG contribution resulting from an occupied QWS below the Fermi energy is illustrated (the final state of the SHG transition is a d-state of Fe). In figure 35(c) SHG involving unoccupied QWS is shown. Note, states along the $\Gamma \rightarrow X$ direction in the Brillouin zone (BZ) are most important. Interference of surface and interface SHG is negligible. Dominant SHG transitions are indicated: (i) are possible for 6 ML and for 12 ML of Cu. This gives oscillations with a period of 6 ML. (ii) Possible only for 12 ML. Thus, one gets two different oscillations (which are frequency-dependent, at $k_1$ and $k_2$ in the BZ).

Note, for $\chi^i \chi^s \rightarrow -1$ one gets a cancellation of the thickness-dependent contribution to SHG due to QWS. If $\chi^i$ and $\chi^s$ become comparable the parity of QWS becomes...
Figure 34. The magnetic reorientation transition. (a) Transition $M_\perp \rightarrow M_\parallel$. The solid curve gives the magnetization of a uniform (single-domain) film occurring for increasing Fe film thickness and for FeAg and for FeCu films. (b) Free-energy change $\Delta F = F(M_\perp) - F(M_\parallel)$ per spin and for increasing temperature. Magnetic anisotropy is temperature-dependent and this causes the reorientation transition at $T_R$. An fcc(100) film is assumed, see Jensen et al [6]. (c) Phase diagram for the orientation of the magnetization is controlled by the anisotropy parameter $K_{\perp}^2(T, N)$ and $K_{\parallel}^2(T, N)$, see equation (49). Note, in the coexistence phase the energy minima of the states with perpendicular ($\perp$) and parallel ($\parallel$) magnetization are separated by an energy barrier. The PD results from minimizing the free energy ($K_{\perp}^2$ refers to $M_\perp$).

3.2.4. Nanostructured thin films with magnetic domains. General remarks: in the following the magnetic properties of generally irregular nanostructures at equilibrium and nonequilibrium are discussed. Generally one observes that the magnetic properties of nanostructured films depend on the film growth conditions. During growth irregular film structures with varying island sizes and shapes appear. Competing interactions between islands (exchange, dipole, etc) occur. Magnetic anisotropy controls the formation of domains, their size and shape and magnetic relaxation of the domains. The relaxations determine the transition from nonequilibrium to equilibrium. In view of the complex behavior one may use Monte Carlo simulations. To describe the transition from isolated islands to connected ones correlations of magnetic relaxation (of neighboring islands) must be taken into account. For example, cluster (island) spin flips will lead to a much faster magnetic relaxation than single spin flips; for details see Jensen et al [7, 8, 27, 31].

The main questions are: (1) what is the domain structure, (2) what is the time-dependent approach of the film magnetization towards equilibrium and (3) what are the controlling forces. If only short range interactions (exchange ones) are active then one expects that percolating atomic structures (coverage $\theta \geq \theta_p$) are necessary for long range global magnetic ordering. Ordering for coverages below $\theta_p$ might indicate dipolar interactions.

The time-dependent structural and magnetic behavior of films during growth reveals many interesting properties and, in particular, the important interplay of atomic structure and
formation of magnetic domains and global film magnetization. As the film grows the size of the domains, its surface roughness and the orientation of the domain magnetization change. Magnetic relaxation occurring typically on a ps to ns timescale causes a time-dependent magnetization in thin films which approaches only at relatively long times (via thermal activation, etc) the equilibrium magnetization. Depending on the domain size and distances between the domains exchange or magnetic dipole coupling may dominate and cause corresponding different relaxation. Thus, the approach of inhomogeneous ultrathin films towards equilibrium magnetization and its dependence on film topology requires careful study.

An important parameter controlling film behavior is the blocking temperature \( T_{\text{bl}} \) (\( T_{\text{bl}} \sim K N / k_B \ln(t_{\text{eq}} \Gamma_0) \sim 5K \)), for \( N \approx 1000 \) atoms resulting from magnetic anisotropy and the percolation coverage \( \theta_p \). While magnetic ordering due to exchange coupling occurs for \( T < T_c \sim J_x \), long range magnetic dipolar coupling causes ordering at \( T_{\text{dip}} \) for \( T < T_{\text{dip}} \sim (N \mu)^2 / R^3 \) (which for \( N = 1000 \) atoms gives about 5 K). \( R \) refers to the distance between domains. As mentioned due to its complexity, analysis requires generally Monte Carlo simulations, see Jensen et al [8, 9, 27, 31].

In figure 36 magnetic domain relaxation is sketched. The typical energy barriers against reversal of the direction of an island (domain) magnetization are shown. Also correlated magnetization changes of the neighboring domains are indicated.

The magnetic relaxation is calculated using a Monte Carlo method (MC), see [7, 31], and a Markov master equation:

\[
dP(x, t) / dt = -\Sigma \Gamma(x \to x') P(x, t) + \Sigma \Gamma(x' \to x) P(x', t),
\]

where \( P \) gives the probability for a spin state of a domain with spin \( x = S_1, S_2, \ldots \) at time \( t \). \( \Gamma(x \to x') \) denotes the transition rate for a domain magnetization change. The magnetization is calculated using \( \langle M(t) \rangle \propto \sum_i M_i(t) \), where \( i \) refers to the time steps taken when time averaging the magnetization of the ensemble of magnetic domains. Hereby possible coherent spin flips of connected domains are taken into account, see [7, 8, 31].

The Markov equation in combination with the MC statistical methods yields the time-dependent nonequilibrium magnetization and its relaxation towards the equilibrium one. The expectation value of the nonequilibrium magnetization at MC time \( t \) for a given film structure is then given by \( \langle M(t) \rangle = \sum_x P(X, t) M(X) \), where \( P(X, t) \) is the solution of the Markov master equation. Using the MC method the relaxation of \( \langle M(t) \rangle \) is calculated by averaging over \( R \gg 1 \) statistically independent MC runs \( r \), thus \( \langle M(t) \rangle \approx \bar{M}(t) = (1/R) \sum_{r=1}^R M_r \). For details of the MC type calculation see [8, 31].

Note, depending on the density of the domains and their distance, correlated relaxation of neighboring domains occurs. This is demonstrated in figure 36(c) where results obtained assuming cluster spin relaxation versus single-spin relaxation are shown.
Figure 36. Sketch of (a) coherently relaxing connecting magnetic domains. (b) Relaxing domains illustrating a model assuming coherent relaxation and successively single-domain relaxation used by Jensen et al [7, 8] (CSF: coherent spin flip of neighboring domains, SSF: single-domain spin flip). (c) Energy barrier of a domain on which a molecular field \( h_{\text{eff}} \) acts for magnetization reversal. The barrier energy \( \epsilon_i = E_b / K N \) controls the reversals of the domain magnetization and results from magnetic anisotropy (\( \sim K \)) and (interdomain, interlayer) molecular field \( h_{\text{eff}} \). (d) Test results for the relaxation of the magnetization using CSF approximation.

For a domain structured film the film magnetization is given by

\[
\vec{M} \propto \sum_i m_i n_i \langle \vec{S}_i \rangle,
\]

where \( \vec{M} \) points along the easy axis and \( i \) refers to the \( i \)th island with \( m_i \) aligned magnetic moments. Here, one averages over the magnetization directions of the domains. In figure 37 magnetic relaxation of thin films is shown (for details see Jensen et al) [7, 8]. Results obtained below and above percolation coverage \( \theta_p \) are compared. Note (for \( \theta > \theta_p \)) the strong influence of single island anisotropy \( K \) on relaxation. For increasing connectivity of the islands faster relaxation results from exchange interactions between islands. Note the temperature and film thickness dependence of the relaxation.

The MC results demonstrate that faster relaxation occurs for correlated spin flips (CSF), as expected of course. As
also expected, different behavior occurs below and above percolation coverage ($\theta_p$).

In figure 38 it is shown how the relaxation of the magnetization depends on the atomic film structure. The dependence of the remanent magnetization of a film on its growth mode (layer by layer or island type one) is given. Note the dependence of the results by Jensen et al. [9, 31] on anisotropy and temperature.

In figure 39 ordering in thin films is characterized. Depending on the density of the domains exchange and dipolar coupling act differently. Clearly the percolation coverage is an important control parameter for the magnetic behavior and range of magnetic interactions. At low coverage dipole coupling may dominate and then ordering occurs at low temperatures.

In figure 40 the dependence of magnetic behavior and of domain structure on film morphology is shown. Results were obtained by Jensen et al using a kinetic MC simulation and a Markov equation for the relaxation of the magnetic domains is given. Film structures resulting from an Eden growth model and resulting magnetic domains are shown [7, 8, 31]. Regarding calculations an area of 500 × 500 lattice constants is taken which contains about 1250 islands. $\lambda$ refers to different spin cluster sizes [7, 8, 27, 31].

Of course, magnetic ordering in thin films depends sensitively on topology and film thickness and the dominant magnetic coupling between domains. Results presented demonstrate this.

The results presented in figure 41 for the domain size (and results on their roughness and surface) are very important regarding understanding the magnetic behavior of irregular thin films and applications.

In figure 41 results are given for the magnetic domain structure in thin films having different thicknesses. Of course, the magnetic domain structure changes and the size of the domains increases as the film thickness increases.

In figure 42 results for magnetic domains and their size dependence on film thickness and temperature are given. These were obtained using the previously discussed analysis by Jensen et al [27]. Note, as a function of temperature one may get a maximum in the domain size. The reason is that for low temperatures domain growth is hindered by energy barriers and at higher temperatures thermal activation disintegrates domains.

Note, generally for increasing film thickness the domains which are first isolated begin to merge and form larger connected areas, still markedly affected by the atomic nanostructure of the film. The roughness of the domains is affected by the nanostructure and decreases for thicker films and for increasing temperature. Of course, this is expected on general physical grounds.

3.3. Tunnel junctions: magnetic effects

Nanoscaled tunnel junctions offer interesting physics, in particular regarding quantum mechanical effects and spin-dependent currents, separation of charge and spin currents
and their interdependence, and interplay of superconductivity and magnetism. Also one may use such nanostructures (FM|M|FM), $M = \text{normal state, superconducting state, etc}$) as interesting fast switches and magnetoresistance devices. Note, the Josephson tunnel current is carried by Andreev states, see [10, 11]. In the following some examples are discussed. Note, different behaviors may result depending on relaxation times $\tau_{CP}$, $\tau_s$ and $\tau_t$, referring to Cooper pairs, spin diffusion and tunneling transport time, respectively.

For a tunnel junction

$$ (\text{TSC}/\text{FM}/\text{TSC}), $$

where TSC denotes a triplet superconductor and FM a ferromagnet, one gets interesting behavior of the Josephson current $j_J$ as a function of temperature, $\alpha$ and $\theta$. The angles $\alpha$ and $\theta$ determine the direction of the magnetization of the ferromagnet and the direction of the TSC order parameter, see figure 20(b) for illustration. The Andreev states are determined from $H \psi_j = E \psi_j$ using the Bogoliubov–de Gennes method.

As shown by figure 43 the Josephson current may change sign as a function of temperature and may be switched between zero and a finite value upon varying the angles $\alpha$, $\theta$. The unconventional change of sign of $I_J$ (without change of phase) for increasing temperature results from the changing occupation of the Andreev states and from the factor $\partial E_i / \partial \phi$ in the expression for the tunnel current. Note, this sign change is different from the one observed for singlet superconductors with a ferromagnetic barrier in between. The behavior of $I_J$ for rotating magnetization, as a function of $\alpha$, suggests using such junctions as switching devices. Note, for increasing $\phi$ the current changes more drastically.

Of particular interest is the behavior of $I_J$ as a function of phase $\phi$ (for $T = 0$) shown in figure 43(b). Then only the states $E_i < 0$ are occupied. For (1) $M \parallel d_{L,R}$, $\alpha \neq \pi/2$, the ferromagnet (FM) couples Andreev states, and (2) for $M \perp d_{L,R}$ the Andreev states are not coupled by the FM and spins are well defined.

Clearly the above junction would be a sensitive probe to detect triplet superconductivity, see the results by Mort et al [11].

It is also of interest to replace FM by a normal metal and to study a junction (TSC/SC/TSC).

Another tunnel system is one involving singlet superconductors:

$$ (\text{SC}/\text{FM}/\text{SC}). $$

The unconventional change of sign of $I_J$ (without change of phase) for increasing temperature results from the changing occupation of the Andreev states and from the factor $\partial E_i / \partial \phi$ in the expression for the tunnel current. Note, this sign change is different from the one observed for singlet superconductors with a ferromagnetic barrier in between. The behavior of $I_J$ for rotating magnetization, as a function of $\alpha$, suggests using such junctions as switching devices. Note, for increasing $\phi$ the current changes more drastically.
The FM is approximately represented by a barrier potential, scattering the electrons spin dependently (g refers to the magnetic scattering strength and z to the nonmagnetic one). Again, depending on nonmagnetic and magnetic scattering of the tunneling electrons the Josephson current may change sign as a function of temperature T and furthermore as a function of the relative phase of the two superconductors, see figures 43 and 44. (Note, $I_0 = I_1 + I_1$, $I = I_2 - I_1$, the Josephson current $I_1$ is solely carried by the Andreev states, and the total spin current $I_s = 0$, since spin current through Andreev states is canceled by the one through the continuum states.) (Note FM may also permit TSC.)

For $z \gg g$ one gets $I_1 = (e\Delta_0/h)\frac{\sin \phi}{2\varepsilon}$ and nonmagnetic current is dominated by Cooper pairs. For $g \gg z$ one has $I_1 = - (e\Delta_0/h)\frac{\sin \phi}{2\varepsilon}$ and current is carried by single electrons. Note then the phase shifts by $\pi$. Results assume for the thickness of the tunnel junction $d$ to be smaller than the coherence length, otherwise the situation is more complicated.

Note, the sign change of the Josephson current as a function of temperature shown in figure 45 results not from a transition of the junction from a $\pi$-state at low temperature to a 0-phase at high temperature, but from a change in the population of the Andreev states (while the relative phase between the superconductors remains unchanged).

Also interestingly, the total spin polarization of the two superconductors’ ground state changes from $(\langle s \rangle = 0$ to $1/2$.

Of course, as mentioned already interesting results are also expected for (FM,SC/FM) junctions. The tunnel current depends on the superconducting state, singlet versus triplet. Also a spin current is expected for $\tau_s > \tau_T$, where times refer as before to times without spin flip and tunneling time. Singlet superconductivity may block tunneling. For a triplet superconductor the angle between Cooper pair angular momentum $d$ and $M$ may control the tunneling.

Transport between quantum dots: an interesting case of electron transport (electron pump model) between two quantum dots involving possibly Coulomb and spin blockade (see the Hubbard Hamiltonian with spin-dependent on-site interaction) is illustrated in figure 20(a). Driving the current with a pulsed (polarized) external field to overcome an energy barrier one gets spin-dependent charge transport with von Stückelberg oscillations due to the bouncing back and forth of the electrons between the two quantum dots.

In figure 46 results are given for the photon-assisted tunneling between two quantum dots, see [14]. The applied electromagnetic field is given by $V(t) = V_0 \cos(\omega t)$. Note the dependence of the charge transfer on the duration of the applied light pulse and the Rabi oscillations due to the bouncing back and forth of the electrons. The Rabi oscillation frequency is $\Omega = 2\omega J_N (E = V_0/\hbar \omega)$. Here $J_N$ is a Bessel function of order $N$, where $N$ refers to the number of photons absorbed to fulfill resonant condition $N\hbar \omega = \sqrt{\Delta \epsilon^2 + 4\omega^2}$.

Different frequencies are used which cause resonant absorption of one, two and three, and possibly more, photons. Of course, this affects the charge transfer. The time-resolved analysis of the occupation of the electronic states shows that the system, if connected to reservoirs, acts as an electron pump. Before action of the external electromagnetic field the initial state is $n_1 = 1$ and $n_2 = 0$. After the pulse is over oscillations disappear and one gets again the initial state via transferring one electron to the right quantum dot and the left reservoir donating one electron to the left quantum dot, see Garcia et al [3]. Spin-dependent quantum dot electron states cause spin currents.

Of interest also is the coherent control of photon-assisted tunneling between quantum dots and its dependence on the shape of the light pulse, see Grigorenko, Garcia et al [14]. The shape of the external electric (or magnetic pulse) may be optimized to get a maximal charge or spin current between the quantum dots connected to two metallic contacts.

Note, these results are also of interest for fermion or boson systems on optical lattices, its dynamics and
Figure 42. Average magnetic domain size as a function (a) of coverage (film thickness) and (b) temperature. The film growth model is described in the text (see Jensen et al). The percolation threshold is $\theta_p \sim 0.9$ ML.

Figure 43. Tunnel junction (TSC|FM|TSC), where FM and TSC refer to a ferromagnet and triplet superconductor, respectively, and $\phi$ to the phase of the superconductor: (a) Josephson current $I_J$ as a function of temperature (changing sign), (b) $I_J$ as a function of $\alpha$, the angle between magnetization $\vec{M}$ and direction normal to the current. Note $I_J$ may be switched between zero and a finite value.

Figure 44. Josephson current $I_J$ at $T = 0$ for a junction (SC/FM/SC) as a function of phase $\phi$ of the superconductor for several values of the scattering strength $g$ and $z$ of the magnetic and nonmagnetic potential, respectively ($g = 0, 1/3, 2/3, 1$ from left to right, $z = 0, 1/3, 2/3, 1$ from bottom to top), see results by Kastening, Morr et al. Note the ferromagnet is approximately represented by a potential barrier.
interaction with external fields. Optical manipulation of molecular binding, in particular boson formation induced by an electromagnetic field is an option. In intense fields nonlinear behavior may be particularly interesting. Furthermore, on optical lattices one may study in particular the interplay of magnetism and lattice structure, the transition from local, Heisenberg-like to itinerant behavior of magnetism. Important parameters for the occurrence of ferromagnetism (antiferromagnetism) are Coulomb interactions and particle hopping (kinetic energy) between lattice sites, and possibly also spin–orbit coupling. Note, as indicated by Hund’s rules, ferromagnetism could occur already for a Fermi liquid with strong enough repulsive Coulomb interactions, quasi-irrespective of the lattice structure, since spin polarization (together with the Pauli principle) may minimize the repulsive Coulomb interactions. In the case where AF dimerization of neighboring lattice sites yields boson formation and resulting BEC condensation optical influence, also of the magnetic excitations, is an important option.

In figure 47 the possibility of manufacturing optically an ultrafast switching device is sketched. Note, in general changing the magnetization in nanostructures generates light in accordance with the Maxwell equations. Via hot electrons fast changing temperature gradients may yield interesting and novel behavior of tunnel junctions. Possibly in such a way Onsager theory can be tested for nanostructures.

For further discussion of the interesting physics realized by tunnel systems, the interplay of spin and charge currents (see the continuity equation for the spin density) and of accompanying light (see the Maxwell equations), one should study the literature, see [12, 33].

3.4. Currents in mesoscopic structures

As mentioned already for mesoscopic structures like discs, quantum dots and rings, for example, one might get interesting topological effects and persistent currents driven by phases resulting from the Aharonov–Bohm effect [34, 36, 37] or Faraday induction law. One may derive such currents induced by a magnetic field \( B \) from \( j = -\frac{d}{dt} F \), where \( F \) refers to the free energy [37]. (Note, approximately \( \Delta F \approx \int \varepsilon N(\varepsilon) f(1 - f) \cdots \), involving electron states close to the Fermi energy, and where the DOS results from the special orbits taken into account in the Balian–Bloch-type theory by Stampfl.)

From symmetry considerations one gets \( \Delta F \propto \cos BS \), where \( BS \) is the flux enclosed by a ring for example, and for

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![Figure 45](image1.png)

**Figure 45.** Josephson current \( I_J \) of a tunnel junction (SC/FM/SC) as a function of \( \frac{T}{T_c} \) for \( \phi = \pi/2 \) and nonmagnetic scattering strength \( z = 0.5 \) and several values of \( g \), the magnetic scattering strength.

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![Figure 46](image2.png)

**Figure 46.** Charge transfer between two quantum dots exhibiting Rabi (von Stückelberg) oscillations due to bouncing electrons between the quantum dots. (a) Dependence of transferred charge on pulse duration of external field and for different frequencies \( \omega_1, \omega_2, \omega_3 \) which cause resonant tunneling by absorption of one, two and three photons, respectively; (b) time-dependent occupation of quantum dot states, see results by Garcia et al. Note, for spin-dependent quantum dot states (magnetic quantum dot), see figure 20(a), the results depend on the light polarization and may involve spin-dependent currents.

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![Figure 47](image3.png)

**Figure 47.** Illustration of a tunnel junction which may act as an ultrafast switching device due to optical excitation of (hot) electrons. Magnetization changes as the electronic temperature changes.
the current \[ j \propto \sin BS(\cdots). \] (75)

Here, the factor denoted by \((\cdots)\) describes the detailed structure of the various electronic orbits and electron DOS, and the phase shift \(\Delta \phi = \pm BS\) results from circling the center (of the ring, disc, etc) clockwise or counterclockwise. Note, as in the case of superconductors, flux quantization may occur. Dynamics due to \(B(t)\) or \(M(t)\) is, of course, reflected in the induced current. Also electron scattering causes dephasing, etc, and affects the current, see Stampfli [16].

The induced currents into the ring due to the Aharonov–Bohm effect with details of their oscillations can be calculated from

\[ j = \frac{\partial F}{\partial \phi} = -\frac{e}{h} \sum \partial E_i / \partial \phi \tanh(E_i/kT) \] (76)

and using the Balian–Bloch-type theory developed by Stampfli et al for calculating \(E_i(\phi)\) and taking into account clockwise and anticlockwise circling of the center. For narrow rings one expects that mainly orbits 1, 2 and 3 are important. A magnetic field \(B\) causes the phase shift \(\Delta \phi = \pm \phi_1 + \phi_2\) due to path deformation and flux, respectively (\(\Delta \phi_2 = BS\)), where \(S\) refers to the area enclosed by the orbit and \(\pm\) to clockwise or counterclockwise circling of \(B\), see the Aharonov–Bohm effect. Thus, one gets for the current induced, for example, by the field \(B\) in a narrow ring the expression

\[ j \propto \sum_i \alpha_i \sin \phi_0 \sin \phi_i (\cdots) + \cdots. \] (77)

Here, \(\phi_0 = BS\) denotes the quantized flux (in units of \(hc/e\)) of the orbit characterized by \(i\) and \(p\), and \(\phi_i\) results from the path deformation due to the field \(B\). Note, the factor \((\cdots)\) causes rapid oscillations described by exponentials, see Stampfli et al [16]. Equation (77) describes the Aharonov–Bohm effect. The behavior of superconductors with normal and superconducting component and of spin-polarized induced currents might be interesting.

Interference of contributions from different electron paths may yield a beating pattern for the current oscillations. Generally the oscillations resemble those discussed for the DOS, see figure 28. The coupling of the induced currents is given by the terms \(j_i, A_i\), where \(A_i\) refers to the vector potential of \(B\).

One gets generally that the induced current decreases at least linearly with the ring radius \(R\) (\(j \propto 1/\sqrt{R}\)) [34, 35, 37]. (Note \(\Delta F = Uj\) [37].) Electron scattering, in particular diffusive motion, may cause the induced current to decrease faster than linear in \(R\), possibly to decrease as \(R^{-2}\) or so.

The fine structure of the current oscillations reflects the quantization of the enclosed flux. In accordance with the le Chatelier principle the magnetic field associated with the induced current should counteract the external field \(B\).

Of course, dephasing expected for strong scattering of the electrons and large rings (and discs) weakens and possibly destroys the current. Also for ferromagnets (Ni, Fe, etc) one expects spin currents (due to DOS effects, etc). For spin currents spin–orbit coupling may also play a role, see the Aharonov–Casher effect [34, 36, 12], and the splitting of the electronic spectrum due to the Zeeman term \(\mu_B \sigma \cdot \vec{B}\) [16].

It would be interesting to find out the occurrence of such induced currents also in electrolytes. Even if weaker as compared to electronic currents as expected these would be very important from first principles, etc.

As noted, a ring shows similar behavior as a (repulsive) anti-dot covered by a thin film. It would also be of interest to study a ring of graphene, due to the remarkable properties of graphene, or conducting nanotubes. To study the interplay of electric and magnetic fields (Maxwell equations at action in nanostructures) double rings are of interest. Generally the discussion and results for superconducting cylinders, rings, SQUIDs and tunnel junctions, see [17, 18], can be frequently applied somewhat to the topologically closed mesoscopic structures in the case of the relatively long free path of the electrons (see the dominant paths in Balian–Bloch–Stampfli theory [16]).

This demonstrates the interesting properties of nanostructures, their potential for studying the interplay of magnetism and superconductivity, of electron and Cooper pair currents (see the two-fluid model for superconductors), manipulating the transition BSC \(\iff\) BEC topologically (see Peeters et al [38]), nonequilibrium physics and for studying the Onsager theory in mesoscopic structures.

4. Summary

Magnetic effects in nanostructures have been discussed. Typical properties of clusters, cluster ensembles, single films and film multilayers and microscopic tunnel systems are presented. The interface of singlet–triplet superconductors needs to be studied and might offer interesting behavior (the triplet state may occur due to magnetism). For energetic reasons in corresponding tunnel junctions (TSC/SC/TSC) and (SC/TSC/SC) currents, in particular Josephson currents, may occur. Note, the general formulae \(j = -\frac{\partial F}{\partial \phi}\), see equation (63), apply. For example, in the tunnel medium 2, see figure 12 and junction (SC/TSC/SC), triplet Cooper pairs \(|\uparrow\downarrow\rangle, |\downarrow\downarrow\rangle\) or \(|\uparrow\downarrow\rangle + |\downarrow\downarrow\rangle\) may carry at least partly the current. Which triplet CP state cooperates best with singlet CP may depend in an interesting way on the superfluid density, spin–orbit coupling, magnetism, etc.

Due to the reduced dimension and system size external fields may change the physical behavior of nanostructures sensitively. Spin-dependent transport on a nanoscale, involving optical manipulation, temperature gradients, etc, see the corresponding Onsager theory, offers interesting possibilities. In general, confinement effects (energy discretization) are interesting [42], also Aharonov–Bohm-induced (spin) currents, hopping like conductivity (single electron, Cooper pair transport), etc. Transport between quantum dots should take into account different and discrete numbers of Cooper pairs and Coulomb effects. Also (strong) nonequilibrium behavior of nanostructure needs to be studied [33].

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