I. Ultrafast Magnetization Dynamics on the Nanoscale

Temporal spin behavior in magnetic systems at short length and time scales

- Time and length scales in magnetism
- Magnetically sensitive X-ray measurement techniques
- Ultrafast manipulation of the magnetization
- Instabilities in low-dimensional magnetism

Magnetism is responsible for one of the oldest inventions, the magnetic compass, and further applications have revolutionized our world, through, e.g., ferrite core memory to today’s high-density data storage. In modern magnetic storage devices, of the order of 400 gigabits can be written per square inch at a rate of one bit per two nanoseconds. Faster reversal, on the sub-ps time scale, has been demonstrated, but its origins still remain to be investigated.

The fact that many fundamental magnetic processes take place on the nanometer length and picosecond time scales, and the high magnetic sensitivity of resonant, circularly-polarized X-rays, make the SwissFEL a versatile instrument for state-of-the-art research in magnetism. Of particular current interest are the ultrafast disappearance, creation and modification of magnetic order and the novel quantum effects which arise in two-, one- and zero-dimensional systems. The Swiss-FEL, with coherent, high-brightness, circularly-polarized X-rays at energies resonant with the 3d-transition metal ions, corresponding to nm wavelengths, is capable of single-shot lensless imaging of nanometer-scale magnetic structures. Furthermore, the combination of high-energy, half-cycle THz pump pulses and the synchronized, sub-picosecond SwissFEL probe pulses will permit the investigation in real time of ultrafast magnetic interactions.
**Time and length scales in magnetism**

The rich variety of characteristic magnetic lengths and times accessible by the SwissFEL is summarized in Figure I.1 and Tables I.1,2 [1,2]. Time-scales $\tau$ are determined by the interaction energies $E$ via the Heisenberg relation $\tau \approx 1/\nu = h/E$, where Planck’s constant $h = 4.14 \times 10^{-15}$ eV s. For example, if the reorientation of a magnetic moment requires expenditure of an energy $E$, it will relax to its equilibrium orientation in a time of order $\tau = h/E$. Reference is made in the Tables to the following material-dependent constants: $J$ – magnetic exchange interaction, $K$ – crystalline anisotropy, $M_s$ – saturation magnetization and $k_F$ – electron Fermi momentum.

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**Table 1: Magnetic length scales**

<table>
<thead>
<tr>
<th>Parameter</th>
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<tbody>
<tr>
<td>Distance over which a diffusing electron maintains its polarization</td>
</tr>
<tr>
<td>Bloch domain wall width</td>
</tr>
<tr>
<td>Minimum size of a magnetic particle before superparamagnetic fluctuations destroy its ferromagnetism</td>
</tr>
<tr>
<td>The exchange length determines the diameter of a vortex core</td>
</tr>
<tr>
<td>Wavelength of the RKKY spin-density oscillations of the conduction electrons near a magnetic ion</td>
</tr>
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</table>

**Table 2: Magnetic time scales**

<table>
<thead>
<tr>
<th>Parameter</th>
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<tbody>
<tr>
<td>Atomic moment reversal due to field-or current-induced domain wall motion, with a velocity of $\sim 100$ m/s</td>
</tr>
<tr>
<td>Magnetization precession and damping, according to the LLG-equation (see Infobox)</td>
</tr>
<tr>
<td>Spin-orbit interaction between the electron spin and its orbital motion</td>
</tr>
<tr>
<td>Jahn-Teller interaction, which stabilizes an elastic distortion to avoid a degenerate electronic ground-state</td>
</tr>
<tr>
<td>Spin-wave energy, at intermediate wave-vector</td>
</tr>
<tr>
<td>Electrostatic crystal-field interaction of oriented 3d-orbitals with neighboring ions</td>
</tr>
<tr>
<td>Inter-electronic exchange energy</td>
</tr>
<tr>
<td>$J$ arising from the Pauli principle</td>
</tr>
<tr>
<td>Correlation energy, responsible within an atom for enforcing Hund’s rules</td>
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Fig. I.1: Time and length scales in magnetism.
XMCD: A simple example

This simple explanation of X-ray Magnetic Circular Dichroism is due to Gallani [22]. Consider an 8-electron atom, with the electron configuration \((1s^2\ 2s^2\ 2p^6)\) in an applied magnetic field \(B\). Due to the Zeeman splitting of the 2p states, the \(|l=1, m=1\rangle\) state is unoccupied. Using circularly-polarized X-rays, we excite the 1s→2p transition (see Fig. I.i1). The dipole selection rule requires that \(\Delta m = \pm 1\), and the conservation of angular momentum implies that for right-(RCP) and left-circular (LCP) polarization, \(\Delta m = +1\) and \(-1\), respectively. The RCP transition, to the empty \(|1,1\rangle\) state, results in strong absorption, while, because the \(|1,-1\rangle\) state is occupied, LCP X-rays are not absorbed. In this way, angular momentum conservation laws and selection rules lead to a circular dichroic, polarization-dependent absorption.

Fig. I.i1. A simple, 8-electron atom example of X-ray circular dichroism.

Magentically sensitive X-ray measurement techniques

X-ray Magnetic Circular Dichroism (XMCD)

XMCD is the X-ray equivalent of the magneto-optical Kerr effect; the quantity measured is the dependence of the absorption on X-ray helicity close to a magnetically-sensitive resonant absorption edge. An example is the spin-orbit-split 2p→3d transitions (\(L_2\) and \(L_3\) edges) of the transition-metal ions. The origin of X-ray magnetic dichroism is explained for a simple example in the Infobox. In contrast to the magneto-optical Kerr effect, XMCD involves a localized initial state, and hence provides chemical and orbital selectivity. If many-body effects can be neglected, XMCD sum-rules can yield both the orbital and spin magnetic moments \(\mu_L, \mu_B\) of the resonant ion (see Fig. I.2). Note that the XMCD contrast is a strong effect: at resonance, the dichroic X-ray interaction per atom can exceed that of 40 electrons! Application of the XMCD technique at the SwissFEL will profit from flexible wavelength tuning, and ultrafast pump-probe measurements will follow the individual time evolutions of the spin and orbital moments in the sample.

Magnetic-contrast holography

With the strong XMCD contrast of resonant magnetic scattering and the high coherence of the SwissFEL, it is possible to perform time-resolved lensless magnetic imaging (see also Chapter III). An elegant X-ray holographic method for thin magnetic films has been developed by Eisebitt et al. [4] (Fig. I.3). Since the experiment is performed at a synchrotron, spatial-filtering is required to produce transverse coherence, and a monochromator
is used to provide sufficient longitudinal coherence. The magnetic thin-film sample, which spans a hole in the sample support, transmits the resonant, circularly-polarized object beam, and close to the sample is a smaller hole, through which an undisturbed reference beam is transmitted. The two beams interfere on their way to the CCD area detector, where they produce a holographic image, and a simple Fourier transform is sufficient to reconstruct a real-space image of the sample.

A direct measurement of the electron polarization can be performed by using photoemission, where electrons are ejected from the sample surface, and where spin analysis is performed, e.g., via Mott scattering. For sufficiently high photon energies, band-structure-dependent final state effects can be neglected. The photoelectric cross-sections for the magnetic sub-shells of magnetic elements peak in the photon energy range 30–100 eV, which will be accessible by the SwissFEL. Spin-analysis using Mott detectors is highly inefficient, and laser-induced magnetic phenomena can be extremely fast (sub-ps). Thus the high peak intensity and the short pulse duration of the SwissFEL make it very attractive for such measurements. Furthermore, with a pulsed photon source, a simple electron time-of-flight (TOF) detector can be used to provide energy resolution (see Fig. I.4). One concern is that the high peak intensity of the SwissFEL will briefly produce a high density of low-energy photoelectrons near the sample surface, causing space-charge effects, which may distort the photoelectron energy distribution. However, since the interaction is to a good approximation electrostatic, the electron spin-polarization will not be affected.

Spin-polarized time-of-flight (TOF) photoelectron spectroscopy

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The three-temperature model of magnetism

The three-temperature model (Fig. I.i2) for magnetization dynamics [10] has been invoked to help explain the sub-ps demagnetization observed by Beaurepaire et al. [8]. The model assumes that the electron motion (el), the electron spins (mag) and lattice phonons (lat) represent three mutually-interacting thermal reservoirs. The internal equilibration of the reservoirs occurs at the plasma frequency (el), the spin-wave frequency (mag) and the phonon scattering rate (lat). Furthermore, the reservoirs interact with one another via the electron-phonon interaction (el-lat), the spin-lattice relaxation rate (mag-lat) and the spin-orbit interaction (el-mag).

Note that the 3-temperature model is purely thermodynamic in nature and hence fails to account for the wavelength of the excitation. With a tunable source such as the SwissFEL, the dependence of bath temperature on wavelength can be investigated [12].

Ultrafast manipulation of the magnetization

Microwaves and terahertz pulses

Dynamic magnetic phenomena can be initiated by a short magnetic field pulse. Using a laser-triggered strip-line, a thin-film sample can experience a 0.01 T magnetic pulse with a rise-time of approximately 100 ps, corresponding to an excitation bandwidth of 10 GHz. In 1999, using pulses of relativistic electrons from the SLAC accelerator directed into the sample, Ch. Back et al. [6] produced multi-tesla fields lasting a few ps, which were large enough to initiate a full magnetization reversal within the plane (see Fig. I.5). Although during the field pulse the magnetization only rotates by approximately 10 degrees (see the Infobox on the Landau-Lifshitz-Gilbert equation of motion), as soon as it tips out of the sample plane, $M$ begins feel the large shape anisotropy from the proximity of the film surfaces.

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The design of the SwissFEL foresees a separate source of terahertz (THz) radiation, capable of delivering high-energy electromagnetic pump pulses which are synchronized with the X-ray probe pulses from the SwissFEL. The intensity (power/area) delivered by an electromagnetic pulse is $I = B_0^2 c / 2 \mu_0$, implying that a 100 $\mu$J, half-cycle THz pulse (0.5 ps) focused to 1 mm$^2$ will produce a peak magnetic field $B_0 = 1.3$ T, i.e., more than 100 times that of a strip-line, and with a THz excitation bandwidth. This offers the possibility of probing the ultimate limit of magnetization dynamics, which is at least a factor 1000 faster than conventional field-induced spin switching.
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A further possibility for rapidly perturbing the magnetic moments in a dynamic XMCD experiment is X-ray detected ferromagnetic resonance, using continuous-wave or pulsed microwave (GHz) radiation [7]. When resonant with the Zeeman-split energy levels of the magnetic ions, the microwaves excite damped magnetic precession, the details of which are sensitive to dynamic couplings and magnetization relaxation. If several magnetic species are simultaneously present in the sample, the elemental selectivity of XMCD allows the dynamics of each to be studied individually. The high peak brightness and excellent time resolution of the SwissFEL would be very beneficial for this technique, avoiding the present restriction to samples with very low damping.

**Laser-induced phenomena**

In 1996, Beaurepaire et al. published a very remarkable observation [8]: a Ni film exposed to an intense 60 fs pulse from an optical laser becomes demagnetized in less than a picosecond. Using the magneto-optical Kerr effect as probe, an ultrastark decrease is observed in the magnetization, followed by a slower recovery (see Fig. I.6). This observation, together with later measurements using other methods of detection, raised the fundamental question, as yet unanswered, of where the spin angular momentum of the electrons goes and how it can be transferred so quickly.

![Fig. I.6. Sub-picosecond demagnetization of a Ni film following an optical laser pulse, observed with the magneto-optical Kerr effect [8]. This observation stimulated much speculation on the as yet unanswered question of how angular momentum can be transferred so efficiently from the spin system to the lattice.](image)

The three-temperature model, which invokes separate temperatures to characterize the electron kinetic energy ($T_{el}$), the electronic spin order ($T_{mag}$) and the lattice vibrations ($T_{lat}$) (see Infobox), has been used to describe the demagnetization process. It is assumed that the laser pump pulse initially delivers energy to the electron reservoir $T_{el}$ and that each reservoir individually remains in equilibrium. But due to their relatively weak intercoupling, the three temperatures may differ significantly, giving rise to strong non-equilibrium effects which have not yet been investigated.

Microscopic models have difficulty in explaining how the laser excitation of the conduction electrons can cause such a rapid transfer of angular momentum away from the spin system or indeed what is its destination. Among the proposals put forward are: hiding the angular momentum in electronic orbits, or transferring it to the lattice via special hot-spots in the electron band structure with exceptional spin-orbit coupling or via an enhancement of the spin-orbit interaction by the presence of phonons [9].

A phenomenological treatment of the entire de- and remagnetization process using an atomic analog of the LLG equations, with the magnetization $M$ replaced by the atomic spin $S$, has been published by Kazantseva et al. [10]. These authors introduced an effective field acting on the atomic moments which includes a stochastic, fluctuating component, which they then related to the LLG damping constant $\alpha$ using the fluctuation-dissipation theorem (see Infobox).

From their atomistic numerical simulations (see Fig. I.7), Kazantseva et al. [10] find that the magnetization can be non-zero in spite of a spin temperature $T_{mag}$ which exceeds the Curie temperature of Ni ($T_C = 631$ K), leading them to question the concept of equilibration of the spin system and hence of a spin temperature. The authors find that the coupling constant which governs the post-pulse recovery of the magnetization is the same as that responsible for the ultrafast demagnetization. They explain the much slower recovery, and the fact that the recovery is slower for a more complete demagnetization, with the concept of a magnetic entropic barrier (see Chapter IV) – i.e., if the magnetization vanishes, it takes time for the system to reorganize itself.

In addition to this, now classic, example of ultrafast demagnetization, some systems show the phenomena of ultrafast magnetization. For example, intermetallic FeRh undergoes a transition at 360 K from a low-temperature anti-ferromagnetic phase to a high-temperature ferromagnetic phase, which is accompanied by an isotropic lattice expansion. This transition can be induced...
The Landau-Lifshitz Gilbert Equation

To describe the time-evolution of the magnetization subjected to a magnetic field $H$, one uses the Landau-Lifshitz Gilbert equation, which has the form:

$$\frac{d\mathbf{M}}{dt} = \gamma \mu_0 \mathbf{M} \times \mathbf{H} + \frac{\gamma \mu_0 \alpha}{M_s} \mathbf{M} \times (\mathbf{M} \times \mathbf{H})$$

where $\mathbf{M} = M_s \mathbf{e}$ is the magnetic moment, $\gamma = g \mu_B / \hbar$ is the gyromagnetic ratio ($\approx 1.76 \times 10^{11}$ rad s$^{-1}$ T$^{-1}$, for $g = 2$), and $\alpha$ is the damping constant. The resulting motion of the magnetization is that of a damped precession about the effective field $\mathbf{H}$ (see Fig. I.i3). The first term causes a precession with angular frequency $\omega = \gamma H$, and the second term exponentially damps the precession at the rate $\alpha \omega$. In a one-tesla field, the precession period is 36 ps, and the damping time $1/\alpha \omega$ may be a factor 100 longer.

Spin-torque: ultrafast switching by spin currents

Beyond the conventional switching by applied external fields, magnetization manipulation can also be achieved by using spin transfer from spin-polarized currents that flow in a magnetic structure. This leads to ultra-fast reversal of nano-pillar elements as well as current-induced domain wall motion. Here the magnetization dynamic timescales are not limited by the gyromagnetic ratio and can be potentially much faster.

Including a spin-polarized current ($\mathbf{u}$ is proportional to the current density and the spin-polarization of the current), the extended Landau-Lifshitz Gilbert equation now reads:

$$\frac{d\mathbf{M}}{dt} = \gamma \mu_0 \mathbf{M} \times \mathbf{H} + \frac{\alpha}{M_s} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}) - \frac{\alpha}{M_s} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}) + \beta \mathbf{M} \times (\mathbf{M} \times \mathbf{H})$$

with the last two terms accounting for the adiabatic and non-adiabatic spin-torque. The strength of the effect is given by $\beta$ and the non-adiabaticity parameter $\beta$.

Spin currents can be generated by spin-injection, spin-pumping and, on a femtosecond timescale, by exciting spin-polarized charge carriers with a fs laser [23]. Furthermore, heating the electron system with a short laser pulse will generate large temperature gradients, and the resulting spin currents and such spin-torque-induced ultra-fast magnetization dynamics can be ideally probed using the SwissFEL.
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PSI – SwissFEL Science Case

Fig. I.7. A frame from a phenomenological, atomistic simulation using the LLG and Langevin dynamics approaches [10]. After rapid demagnetization, the spin system must choose between many possible magnetized states and thus is confronted with an entropic barrier.

by an ultrashort laser pulse [11]. Preliminary results using 50 ps X-ray probe pulses from a synchrotron suggest that the establishment of ferromagnetic order precedes the increase in lattice constant, possibly answering the chicken-or-egg question as to which is the cause and which is the effect.

Finally, in experiments with a direct connection to magnetic data storage, it has been found that ultrafast pulses of circularly-polarized laser light can, via the inverse Faraday effect, even reverse the direction of magnetization in a sample (see the optomagnetism Infobox). The above examples of the ultrafast manipulation of magnetization with optical pulses point to a rich variety of possibilities for SwissFEL pump-probe investigations for both fundamental science and practical applications. Ultrafast perturbation of a magnetic spin system is also possible using the spin-transfer torque phenomenon, which is the basis for spintronic schemes of data processing (see Infobox) [12].

Langevin dynamics and the fluctuation-dissipation theorem

In their numerical treatment of ultrafast magnetization recovery after an optical pulse, Kazantseva et al. [10] used the formalism of Langevin dynamics to relate the amplitude of a fluctuating magnetic field experienced by a local moment to the strength of the viscous damping it undergoes. The general relationship between fluctuations and damping is expressed in the fluctuation-dissipation theorem (FDT), which we derive here for the simple case of the one-dimensional Brownian motion of a particle [24].

Consider a particle with mass $M$ and velocity $v(t)$ which interacts with the local environment via both a viscous force, characterized by the mobility $\mu$, and a randomly fluctuating force $f(t)$. The resulting (Langevin) equation of motion is:

$$M\ddot{v}(t) + \frac{1}{\mu}v(t) = f(t)$$

or, with the definitions $\gamma = 1/M\mu$ and $A(t) = f(t)/\mu$:

$$\ddot{v}(t) + \gamma v(t) = A(t)$$

This equation has the steady-state solution (for $\gamma t >> 1$):

$$v(t) = \int_0^t e^{\gamma(t-u)}A(u)du$$

We require that $A(t)$ have a zero mean value ($\langle A(t) \rangle = 0$), a vanishing autocorrelation time and a time-independent variance ($\langle A(t_1)A(t_2) \rangle = A^2 \delta(t_1 - t_2)$, $A=\text{const}$).

The (steady-state) variance of the velocity is then:

$$\langle v^2(t) \rangle = e^{2\gamma t} \int_0^t du e^{\gamma(u-w)} \langle A(u)A(w) \rangle$$

$$= A^2 e^{2\gamma t} \int_0^t du e^{2\gamma w} = \frac{A^2}{2\gamma} \left(1 - e^{-2\gamma t} \right) = \frac{A^2}{2\gamma}$$

Making use of the equipartition theorem:

$$\frac{1}{2} M \langle v^2 \rangle = \frac{kT}{2}$$

we find the following condition:

$$\gamma = \frac{M}{2kT} A^2$$

or:

$$\frac{1}{\mu} = \frac{1}{2kT} \int_0^\infty \langle f(0)f(t) \rangle dt$$

This relation between the viscous and random forces is a special case of the more general FDT.
Optomagnetism

A large effort is presently being made to use intense, fs pulses of circularly-polarized light to write magnetic information in thin film media. The goal is to explore the limits of high-speed magnetic writing. The production of an effective magnetic field by a circularly-polarized light wave is called the inverse Faraday effect. This is the basis of optomagnetism, i.e. the manipulation of the magnetization by laser light.

A circularly-polarized light wave can produce an effective magnetic field as follows [25]:

\[ \mu_0 H_{\text{eff}} = \frac{\partial W}{\partial M} = \epsilon_0 E(\omega) E^*(\omega) \frac{\partial \epsilon}{\partial \epsilon} \]

An expression due to Onsager relates the off-diagonal elements of the dielectric tensor and the magnetization:

\[ \epsilon_{ij} = \alpha M + \beta M^3 + ... \]

The resulting effective field (see Fig. I.4):

\[ \mu_0 H_{\text{eff}} = \alpha \epsilon_0 E(\omega) E^*(\omega) \]

can be as large as 1 T.

Fig. I.4. Circularly polarized light can, in an optomagnetic medium, create a large effective magnetic field.

A demonstration of sub-ps magnetic writing has been made by Stanciu et al. [26] using 40 fs pulses in GdFeCo (see Fig. I.5), but fundamental questions remain regarding how the ultrafast transfer of angular momentum occurs.

Instabilities in low-dimensional magnetism

In the previous Section, measurements and simulations were discussed of ultrafast magnetization phenomena in three dimensions; here possibilities are considered for using the SwissFEL to investigate the quantum-fluctuating behavior of low-dimensional magnetic systems [13].

In many magnetic insulators, magnetic moments interact through an exchange of electrons between neighboring sites. Such exchange interactions are short-ranged. If these interactions are isotropic, such systems can be described by the well-known Heisenberg Hamiltonian, which is given by:

\[ H_{\text{Heisenberg}} = -J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j \]

where \( J \) is the exchange energy, and the summation is over nearest-neighbor spins; If \( J \) is positive, the spins \( \vec{S}_i \) and \( \vec{S}_j \) tend to align ferromagnetically. For an ordered magnetic phase, the temperature-dependent change in the saturation magnetization can be calculated [14] in this model as \( M_s(0) - M_s(T) \approx N_{sw}(T) \), where

\[ N_m(T) \propto \frac{k^{d-1}}{\exp[\epsilon(k)/k_B T]} dk \]

is the density of spin-waves excited at the temperature \( T \). For \( d=3 \) dimensions, this integral is proportional to \( T^{3/2} \), giving the well-known Bloch 3/2-law.

For dimensions lower than \( d=3 \), the expression for \( N_{sw}(T) \) diverges, implying that fluctuations will prevent the occurrence of long-range magnetic order. This is a fundamental result, which has been rigorously proven by Mermin and Wagner [15], and which means that many types of magnetic systems do not order at any finite temperature.

Some systems are disordered even at zero temperature, where thermal fluctuations are absent, due to the presence of quantum fluctuations in the ground state. This can happen if a static arrangement of magnetic moments is not an eigenstate of the Hamiltonian, causing quantum fluctuations to generate a new type of ground state. These disordered systems form ferromagnetically or antiferromagnetically-coupled spin liquids, and their quantum fluctuations, as described by the intermediate scattering function \( S(Q,t) \) (see chapter V), represent a particularly rich field of investigation for the SwissFEL.

Two-dimensional case (\( d=2 \))

As an example of the dynamics of a 2d-magnetic structure, consider the case of an infinite in-plane anisotropy (\( S_z=0 \)): the so-called \( xy \)-model: \( H_{\text{xy}} = -J \sum_{\langle ij \rangle} (S_i^x S_j^x + S_i^y S_j^y) \)
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As for the 2d-Heisenberg model, there is no magnetic order at finite temperature in the xy-model. However, it is found that spin correlations cause the formation at low temperature of a disordered array of magnetic vortices, with radii of order \( R \). The cost in exchange energy incurred by the formation of such a vortex is \( \pi J \ln(R/a) \), where  \( a \) is the lattice constant, and the gain in entropy represented by the varying position of the vortex center is \( 2k_B \ln(R/a) \). Hence the free energy \( F = \ln(R/a) (\pi J - 2k_BT) \) becomes negative below the Kosterlitz-Thouless transition, at the temperature \( T_{KT} = \pi J / 2k_B \), which separates a non-vortex from a vortex phase. At low temperatures, the \( S=1/2 \) layered perovskite ferromagnet \( K_2CuF_4 \) is approximately described by the xy-model, going through a Kosterlitz-Thouless transition at 5.5 K.

A further example of (quasi) 2d-magnetic dynamics is that of vortex core reversal in thin magnetic nanostructures (see Infobox).

One-dimensional case (\( d=1 \))

Magnetism in one dimension in the zero-temperature limit is particularly interesting, because it arises from quantum fluctuations. Consider first the isotropic \( J>0 \) Heisenberg model for a one-dimensional chain of \( N \) spins \( S=1/2 \), with periodic boundary conditions. The (ferromagnetic) ground state can be represented as \( |\Psi_0 \rangle = |\uparrow\uparrow\uparrow...\uparrow\rangle \). In the Bethe Ansatz, the excited states of the system are built up as a superposition of states with discrete numbers of flipped spins. If we confine ourselves to single-spin (\( r=1 \)) excitations:

\[
|n\rangle = |\uparrow\uparrow\uparrow...\uparrow\downarrow\uparrow...\uparrow\rangle
\]

(here the \( n^{th} \) spin has been reversed), we can write the excited state as

\[
|\Psi_i \rangle = \sum_{n=1}^N a(n) |n\rangle
\]

It is then a straightforward exercise to compute from the Schrödinger equation (for convenience, written in terms of the raising and lowering operators \( S_\pm \)) the excited-state energy \( E_i \), and one finds, for large \( N \), that excitations exist with arbitrarily small excitation energies \( E_1 - E_0 \); i.e., the excitation spectrum is gapless. Higher level excitations, involving multiple spin flips \( r = 2, 3, 4, ... \), become increasingly cumbersome to handle, but the gapless spectrum is retained (Figure I.8a shows the analogous result for the 1d-antiferromagnetic spin \( 1/2 \) chain [16]).

Magnetic vortex core switching

The magnetic vortex is a very stable, naturally-forming magnetic configuration occurring in thin soft-magnetic nanostructures. Due to shape anisotropy, the magnetic moments in such thin-film elements lie in the film plane. The vortex configuration is characterized by the circulation of the in-plane magnetic structure around a very stable core of only a few tens of nanometers in diameter, of the order of the exchange length. A particular feature of this structure is the core of the vortex, which is perpendicularly magnetized relative to the sample plane. This results in two states: “up” or “down”. Their small size and perfect stability make vortex cores promising candidates for magnetic data storage.

A study by Hertel et al. [27] based on micromagnetic simulations (LLG equation) has shown that, strikingly, the core can dynamically be switched between “up” and “down” within only a few tens of picoseconds by means of an external field. Figure I.6 below simulates the vortex core switching in a 20 nm thick Permalloy disk of 200 nm diameter after the application of a 60 ps field pulse, with a peak value of 80 mT. Using field pulses as short as 5 ps, the authors show that the core reversal unfolds first through the production of a new vortex with an oppositely oriented core, followed by the annihilation of the original vortex with a transient antivortex structure. To date, no experimental method can achieve the required temporal (a few tens of ps) and spatial (a few tens of nm) resolution to investigate this switching process. The combination of the high-energy THz pump source and circularly-polarized SwissFEL probe pulses will allow such studies.

Fig. I.6. Numerical simulation [27] of the proposed switching process of a magnetic vortex core. The SwissFEL will provide the spatial and temporal resolution required to visualize this process.
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Fig. I.8: a) The spin-wave dispersion relation for a 1d $S=1/2$ antiferromagnetic chain [16]. The shaded region corresponds to two-magnon excitations. b) The dispersion relation for a 1d Peierls spin $1/2$ chain [13], showing the opening of an energy gap $E_g$. Note that the Brillouin zone is reduced by half.

One of the simplest ways for a material to avoid magnetic order and develop macroscopic quantum correlations is through the creation of an energy gap $E_g$ in the excitation spectrum. Since $E_g$ is of the order of the exchange interaction, the gap introduces a time-scale for fluctuations which is typically on the order of femtoseconds. One such phenomenon is the spin Peierls effect. This is related to the better-known charge Peierls metal-insulator transition (see Chapter V).

In the spin-Peierls effect, a uniform 1d, $S=1/2$ spin chain undergoes a spontaneous distortion, causing dimerization, and hence the appearance of two different exchange couplings $\Delta J$ (see Fig. I.9). For $\delta J$ sufficiently large, $S=0$ singlet pairs are formed on the stronger links, implying a non-magnetic state and a finite energy gap to the excited states (see Fig. I.8b). The Peierls state is stable if the resulting lowering of magnetic energy more than compensates for the elastic energy of the lattice distortion. Note that the distortion is a distinctive feature which is visible with hard X-ray diffraction. The spin-chain compound CuGeO$_3$ is an inorganic solid which undergoes a spin-Peierls transition at 14 K.

Fig. I.9. Schematic representation of the spin-Peierls transition, caused by a dimerization of neighboring sites along a 1d $S=1/2$ Heisenberg chain.

A more subtle quantum effect also leads to an energy gap in the excitation spectrum of an antiferromagnetic Heisenberg chain of integral spins [17]. As conjectured by Haldane, neighboring $S=1$ spins can be resolved into two $S=1/2$ degrees of freedom, effectively forming singlet bonds (see Fig. I.10). This valence bond state is responsible for the existence of a Haldane energy gap, since long wavelength spin excitations cannot be generated without breaking the valence bonds. A consequence of the Haldane mechanism is a spatial correlation function for magnetic excitations which decays exponentially with distance, compared with the power-law dependence in the case of gapless excitations. An inorganic material which demonstrates the Haldane phenomenon is Y$_3$BaNiO$_5$.

Fig. I.10. The Haldane mechanism in a 1d $S=1$ Heisenberg chain, which produces a valence bond state and a gapped excitation spectrum.

The Haldane mechanism is also used to describe the dynamic behavior of finite 1d $S=1$ antiferromagnetic chains, as investigated in Mg-doped Y$_3$BaNiO$_5$ by inelastic neutron scattering [18]. The finite chains are generated by the non-magnetic Mg impurities, and the ends of the chains represent $S=1/2$ impurities with a strong nano-scale correlation, with the result that the Haldane gap becomes a function of chain length. In an applied magnetic field, the triplet spin excitations undergo Zeeman splitting, eventually becoming a new ground state (see Fig. I.11). Thus the Zeeman transitions are hybrid excitations with both local and cooperative properties. They therefore serve as probes of the quantum correlation functions, which are otherwise difficult to access. The temperature and field ranges for such studies vary with the material, but effects can be observed in many systems at $T \sim 1$ K and $B \sim 1$ T. Some quantum magnets show magneto-electric interactions [19], which may allow perturbation of the quantum states by electric or optical pulses. In this case, it will be possible to probe the temporal evolution of macroscopic quantum correlations in a pump-probe experiment at the SwissFEL.
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Quantum Phase Transition

Fig. I.11: The occurrence in a singlet spin liquid at high magnetic field of a quantum phase transition [18].

Zero-dimensional case (d=0)

The extreme brightness of the SwissFEL allows measurements of magnetic phenomena on dilute samples consisting of isolated nanoparticles, with effectively zero dimension. A recent realization of such nanoparticles is the single-molecule magnet, manganese acetate (Mn12Ac) shown in Fig. I.1, in which 12 magnetic Mn ions, with a total spin $S=10$, are held in close proximity by an organic molecular framework. Another example is the creation of magnetic nanodots by sub-monlayer deposition onto a high-index surface of a metal (see Fig. I.12). If they have a magnetic anisotropy above the superparamagnetic limit, such nanoparticles may exhibit room-temperature ferro- or antiferromagnetic order, and undergo sub-nanosecond quantum tunnelling between different magnetization directions [21]. Details of this tunnelling, including field-enhancement of the rate, are an attractive topic in ultrafast magnetization dynamics, suitable for study with the SwissFEL.

Fig. I.12. Cobalt nanodots created [20] by the deposition of 0.3 monolayers on the gold (788) surface. The inset shows the distribution of dot sizes $s$ around the mean size $S=70$ atoms.
Summary

- Present magnetic storage devices are limited to densities of 400 gigabits/square inch (i.e. one bit/40 × 40 nm²) and reading/writing times of 2 ns, while standard synchrotron-based magnetic studies are limited by the 100 ps pulse duration. The SwissFEL will investigate magnetic phenomena well beyond these values.

- Fundamental magnetic interactions occur on the nanometer and sub-picosecond length and time scales – which are very well matched to the capabilities of the SwissFEL.

- With its provision of circularly-polarized radiation, high transverse coherence and femtosecond pulse duration, the SwissFEL will accommodate the X-ray measurement techniques of particular relevance to ultrafast magnetism: X-ray magnetic circular dichroism (XMCD), magnetic-contrast holography and time-of-flight, spin-analyzed photoemission.

- A synchronized terahertz pump source at the SwissFEL will provide magnetic field pulses with an amplitude of approximately 1 tesla and a duration of approximately 1 picosecond, for high-bandwidth excitation in ultrafast pump-probe experiments.

- Synchronized femtosecond pulses of optical laser light will also be available for pumping at the SwissFEL. These cause rapid heating of the electrons of the sample, which, through as yet not understood interactions, may demagnetize a sample, cause a transition from an antiferromagnetic to a ferromagnetic state or reverse the magnetization direction. Pump-probe XMCD experiments at the SwissFEL will follow the ultrafast behavior of the spin and orbital moments during these processes.

- Low-dimensional systems fail to develop long-range magnetic order. The quantum fluctuations of such spin liquids represent a rich field of application for the SwissFEL.
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References


