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IJPRA ISSN 2766-2748 INTERNATIONAL JOURNAL OF PHYSICS RESEARCH AND APPLICATIONS

Short Review

Various Theories of Fast and Ultrafast Magnetization Dynamics

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Submitted: August 07, 2024
Approved: November 20, 2024
Published: November 21, 2024

How to cite this article: Fähnle M. Various Theories of Fast and Ultrafast Magnetization Dynamics. Int J Phys Res Appl. 2024; 7(2): 154-158. Available from: https://dx.doi.org/10.29328/journal.ijpra.1001101

Keywords: Laser-induced magnetization; Spintronics; Femtosecond timescales; Magneto-optical effects; Phonons; Spin dynamics

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Abstract

The background of my paper is that magnetization dynamics is a very important subject of basic and technological research. The purpose of the paper is to review various theories of magnetization dynamics. There are many important technological applications of magnetization dynamics.

Introduction

The fundamental knowledge to understand magnetization dynamics is the basis of knowledge of magnetism and the physics of dynamical processes. The research on magnetization dynamics is very interesting from a fundamental point of view because the underlying physical processes were not really understood in former research. It is also interesting from a technological point of view because fast and ultrafast magnetization dynamics can be applied in future magnetic computers. The highlight of this paper is that for the first time, various theories of fast and ultrafast magnetization dynamics are being reviewed. The technique to generate fast magnetization dynamics is to excite the system by the application of external magnetic fields or spin-polarized currents the technique to generate ultrafast magnetization dynamics is the application of femtosecond laser pulses. The paper contains references to former papers on magnetization dynamics by other authors, most of them by phenomenological approaches. In this paper, various theories of fast and ultrafast demagnetizations dynamics based on basic physical principles, and not based on phenomenological approaches were reviewed.

I. Fast magnetization dynamics

The observed phenomena of magnetization dynamics are typically subdivided into two categories, according to the respective time scale.

In the region between several picoseconds (ps) to nanoseconds (ns) the magnetization dynamics is close to the adiabatic limit. In this case, the magnetization dynamics is so small that the electrons are always in their ground state configuration for the momentary magnetization configuration. Examples are the dynamics of domain walls or the gyrotropic precession of magnetic vortices. In the case of ultrafast magnetization dynamics after femtosecond (s) laser pulses there are changes of the magnetization on a time scale of several hundred fs, and the system is not close to the adiabatic limit.

The fast magnetization dynamics is described in the work of Jonas Seib [1].

The fast magnetization dynamics is often a dissipative magnetization dynamics. Thereby it is distinguished between direct and indirect damping. In the indirect damping for the indirect damping, there is a transfer of energy and angular momentum from the considered magnetic mode to other magnetic degrees of freedom. This happens, e.g., during ferromagnetic resonance, in which the homogeneous ferromagnetic mode generates by non-linear effects other non-homogeneous modes. In contrast, when energy and angular momentum go from the magnetic system to non-magnetic degrees of freedom, then this is called direct damping. Examples are the radiation of electromagnetic waves generated by the time-dependent.

Magnetization, or relaxation processes due to dipole-dipole interactions, and the damping of the spin magnetization by spin-orbit couplings. Without spin-orbit coupling the operator of the complete spin moment commutes with the Hamilton operator, and the magnetic spin moment is conserved.

The theories of direct damping can be further subdivided. On the one hand, there are mechanisms by which the transfer

of energy and angular momentum from the magnetic degrees of freedom occurs directly to the lattice degrees of freedom. Thereby the spin-orbit coupling is described in a phenomenological manner by the magnetoelastic interaction of the magnetization with the strain field of the lattice [2]. A time-dependent magnetization generates due to this magnetoelastic interaction a time-dependent strain field, which generates a modification of the phonon modes by phonon scatterings at scattering centers.

Another category of theories describes mechanisms in which in an intermediary step electronic degrees of freedom are excited, which then relax by interactions with the lattice. An example is the excitation of curly currents by a time-dependent magnetization field. The relaxation of the curly currents is due to Ohmic dissipation. An example for such a theory is the breathing Fermi-surface model. There the dissipation of the magnetization dynamics is described in two steps. A slightly non-adiabatic situation is modelled, in which the magnetization dynamics primarily generate electron-vacancy pairs, which then relax by electron-phonon scatterings and by electron scatterings at defects.

A phenomenological equation of motion for the dissipative fast magnetization dynamics is the Gilbert equation [3]. Thereby the time-derivative of the magnetization $\mathbf{M}(\mathbf{r},t)$, is equal to the torque, which is exerted on the magnetization by an effective field $\mathbf{H}_{eff}$ and by a dissipative filed $\mathbf{H}_{diss} = -ǎ/(ϒM)\,\partial\mathbf{M}/\partial t$,

$$\partial\mathbf{M}/\partial t = -ϒ\,\mathbf{M}x(\mathbf{H}_{eff} + \mathbf{H}_{diss}) = -ϒ(\mathbf{M}x\mathbf{H}_{eff}) + 1/M\,\mathbf{M}x\,ǎ\,\partial\mathbf{M}/\partial t \quad (1)$$

The quantity ϒ is the gyromagnetic ratio, M is the modulus of the magnetization, and ǎ is a phenomenological scalar damping constant. The effective field is composed of the external field, the exchange field, the anisotropy field, and the dipole field.

The Gilbert equation is the simplest conceivable equation of motion for the damped precession of the magnetization. The precession term contains the torque of the effective field on the magnetization, generating the precession of the magnetization around the equilibrium direction, the damping term $\mathbf{H}_{diss}$, the strength of which is given by the constant ǎ, describes the relaxation of the magnetization in the direction of the equilibrium direction.

Already Gilbert suggested a generalized form of the equation of motion, with a Rayleigh dissipation function, with a non-local damping parameter that is no longer a scalar but a matrix. This matrix character causes an anisotropy of the damping. This generalized form has to be used for a more accurate description of the dissipative fast magnetization dynamics [4].

II. Ultrafast magnetization dynamics

The ultrafast magnetization dynamics after femtosecond laser pulses are described, e.g., [5,6].

The situation of the direct damping close to the adiabatic limit is described by the breathing-Fermi-surface model [5]. In an adiabatic situation, the electronic occupation numbers Are given by the Fermi-Dirac statistics with occupation number f_i In a not completely adiabatic situation the adiabatic occupation numbers have to be replaced by time-dependent non-adiabatic occupation numbers n_i (t) for the electron states I The non-adiabatic occupation number lag behind the adiabatic occupation numbers, and this is described by a relaxation-time ansatz with relaxation times τ_i,

$$dn_i\,dt = -1/\tau_i\,(n_i\,(t) - fi_i \quad (2)$$

where the f_i depends on the energy of the electron in the state I Of course these occupation numbers depend on the momentary state of the system. Often the approximation τ_i = is used, i.e., it is assumed that the occupation numbers for all electron state I are the same. In a nonadiabatic situation close to the adiabatic limit, in which the dynamics of the atomic magnetic moments are slow in comparison to the relaxation time τ, the solution of equation 2 is approximately given by

$$n_i\,(t) = f_i\,(t) - \tau\,df_i\,/dt \quad (3)$$

The results of this breathing-Fermi-surface theory for the ultrafast magnetization dynamics are given in the study by Keith Gilmore, et al. [7].

In y typical experiment on ultrafast magnetization dynamics, there is a femtosecond (fs) laser, which excites a ferromagnetic film. Below the film, there is a substrate, e.g., an isolating substrate, like MgO. As a result of the excitation, there is at least partial demagnetization of the film in about 100 fs, and a re-magnetization to the original state on a bit longer time scale. The laser frequency can be an optical frequency [8] or a THz frequency [9]. The typical experiment is a pump-probe experiment, i.e., the excitation is with a laser pulse (pump) at the system is investigated after a certain time, e.g., by MOKE, XMCD, etc. (probe). During de- and re-magnetization there is a transfer of angular momentum from the electron system (which carries the magnetization) to other degrees of freedom. Thereby the question is whether the total angular momentum is conserved or not. A precondition for the conservation of the angular momentum is an anisotropy of the Hamiltonian describing the system. In the experiment the isotropy is not exactly fulfilled, because the considered system with the magnetization is coupled to the surrounding. A typical specimen is a thin film on a dielectric substrate, which is fixed on a sample holder. It is sometimes argued that the whole world is isotropic and that therefore an angular momentum conservation holds when considering a Hamiltonian which takes into account all degrees of freedom of the world, which – however – is not possible. But on the time scale of a few hundred fs a process in the magnetic system can lead to a reaction outside the system only on a very limited scale of space and time, and therefore it suffices to consider the sample and just the direct neighborhood, and on this time

scale the Hamiltonian, can be considered approximately as isotropic, so that angular momentum conservation is fulfilled in a very good approximation. Anyway, even for a non-isotropic situation it is interesting to know how much of the angular momentum of the considered system is conserved.

To describe the couplings and the angular momentum transfer between all possible subsystems one had to calculate the time-evolution of the Hamiltonian, which considers all possible interactions. To do this one has of course to perform approximations, because in the time-evolution of operator products of order n always at least terms of the order n+1 are found. For a solution, it is necessary to find a closed approximate form in order to make a finite calculation time possible. One has to consider the following scatterings: electron-electron scatterings. The electron-electron interaction is the strongest interaction in the system, but it is also the one which is most difficult to calculate. The electron-phonon scattering. Calculations have shown [5,6] that the electron-phonon scattering rates are too small to explain the observed demagnetization.

The electron-magnon scattering also contributes to the magnetization dynamics, as well as the electron-defect scattering. Under discussion is also a transport of the angular momentum out of the magnetic system, i.e., the transport of spin-polarized electron currents. To the substrate, a prominent example is the 'super-diffusion' of hot electrons (i.e., of electrons excited by the laser pulse) to the substrate. When an isolating substrate of the magnetic film is considered, then the transport of spin-polarized currents is strongly suppressed. However, it can come to a spin accumulation at the interface between the magnetic film and the substrate, which can modify the scattering rates at the interface. In the following, the transport of angular momentum out of the system is not considered.

The calculations have shown [10] that electron-electron scattering, electron-phonon scatterings, electron magnon scatterings, and electron-defect scatterings.- can not explain the experimentally observed demagnetization. In reference 10 therefore a totally new scattering was discussed, namely the scattering of electrons at local-elastic twist modes which are generated by the magnetoelastic interactions of magnons with the lattice. This required a revolutionary new construction of the corresponding Hamiltonian. The numerical calculations so far have to been performed but the hope is that the sum of all scatterings (electron-electron scatterings, electron-phonon scatterings, electron-magnon scatterings, electron-defect scatterings, and scatterings of electrons at the local elastic-twist modes can explain the experimentally observed changes of the magnetization, and that the total angular momentum of the system is conserved in a good approximation.

III. Ultrafast magnetization dynamics after femtosecond THz laser pulses

The ultrafast magnetization dynamics after femtosecond THz laser pulses is discussed extensively in reference 9. The study of ultrafast magnetization dynamics is crucial for our understanding of magnetic systems, as well as for developing ultrafast magnetic memory devices. The difficulty of the subject lies in the spin dynamics and its rule as a part of a larger picture of the barely understood structural dynamics.

As an example, when a ferromagnetic film is excited by a femtosecond (fs) laser pulse at least a partial demagnetization of the material occurs within about 100 fs, followed by a re-magnetization to the original state on a bit longer time scale. This was observed for the first time in reference 8 for optical laser pulses, and later also for THz laser pulses [9]. This phenomenon has led to intensive investigations in the field, both with experiments and theories. It has now become evident that the process is mediated by a complex puzzle of strongly entangled processes. To elaborate on them, it is essential to understand the physical mechanisms that can influence the sin angular moments of the electrons (which are responsible for the magnetization. In order to simplify the almost unfeasible task of tacking coupled structural dynamics, the responsible phenomena and their time scales are examined. This allows to decouple mechanisms that have small effects on each other.

As Born and Oppenheimer pointed out, a separation of atomic mode dynamics and the dynamics of the surrounding electrons can be made. This is because electrons will adiabatically follow any comparatively slow changes in the lattice. That is, the typical dynamics of phonons are on the picosecond time scale, whereas the electron dynamics in metals can occur on a short time scale (as the attosecond time scale). Similarly, a separation of electronic charge and sin degrees of freedom is reasonable, due to the time scales that are observed for the dynamics of the spins (about 100 fs after laser pump excitations), which are slower than the time scales for the electrons (about 10 fs).

The dynamics of the magnetization in a stimulated material can be affected by a number of combinations of the mechanism described above. In particular, there may be different mechanisms of light-matter interactions. The two extremes are now elaborated on as direct coupling of the laser pulse to the electronic spins, and/or intermediate change of phonon excitations which induced magnetization dynamics.

In the direct scenario, the local electromagnetic field of the stimulus couples with the electronic angular momentum, such that there is only a minimal heating of the electron system. In the indirect channel, the laser photos do not primarily change the magnetization (as in the direct channel). Instead, the photon energy is transferred to the sample in the form of an increased electron and atomic temperature. This process can excite electron-vacancy pairs, lattice phonons, and magnons which will show sin-flip scatterings with the electrons, responsible for the modification of the magnetization, by modifying the electronic angular momenta.

Demagnetization processes were in the are reported for optical laser pulses. The changes in the magnetization are mediated through the indirect channel. The result is a non-equilibrium state, induced because the electronic temperature is raised above the phonon temperature, and an imbalance exists between the chemical potentials of electrons with different spins. This difference is the driving force for the ultrafast demagnetization. The system then evolves through the above-discussed spin-flip scatterings, leading to changes in the orientations of the atomic magnetic moments. As a result, a temporary demagnetization is observed, followed by a re-magnetization to the original state with a balance of the chemical potentials, and with a thermalization of electrons and phonons. Furthermore, in samples on a metallic substrate, there is also the contribution of a super-diffusion process (see section II). In this process, the excited spin carriers are transferred to the metallic substrate.

Furthermore, the total effective electromagnetic field exerts a Zeeman torque on the atomic magnetic moments, This minute contribution on the magnetization **M**(t) is a coherent oscillation in time, following the action of the electromagnetic wave of the laser pulse. However, excited electrons with the above-discussed spin-flip scatterings also alter **M**(t) incoherently in time, leading to fluctuations of the phase which dephase the spin motion. Therefore, a precession motion is typically not observed after optical laser pulse excitations, and the precession motions give only a minute contribution to the change of the magnetization.

Optical laser pulse demagnetization of the magnetic films has been extensively examined, and the majority of the studies report results on indirect spin excitations (see section II). To increase the understanding of all the potential interactions, experiments have been performed with THZ laser stimuli. The THz cycle oscillates on a similar time scale as the motional speed of electronic spins, in contrast to optical pulse stimuli, which oscillate on a much faster time scale.

At low field amplitudes, the THz laser the electromagnetic waves of the laser pulse are expected to show direct and coherent couplings to the electron spin dynamics. This interaction leads to the above-discussed precession motion of **M**(t) due to the Zeeman torque, leading to a selective control of the magnetic phase. Moreover, THz photon energies are three orders of magnitude smaller than optical photon energies, inducing significantly smaller heating at low-field amplitudes, and reducing the possibility for spin-flip scatterings between electrons and excited quasiparticles. The direct coupling of the THz electromagnetic field with the magnetization was first reported for a ferromagnetic Co film in the work of Carlo Vicario, et al. [11], in which a coherent phase-locked demagnetization was observed under THz laser pulse excitations.

In contrast, after THz laser excitations with large amplitudes of the electromagnetic wave there is damage to the indirect mechanism, i.e., there are excitations of electrons that show spin-flip scatterings, leading to fluctuations of the phase and to a dephasing of the magnetic system, so that no precession could be observed as after optical laser pulses.

At low field amplitudes, one can observe a linear response of the magnetization on the intensity of the pump electromagnetic field. The THz pump fluence needed to get a complete demagnetization is similar to the optical counterpart, which therefore suggests that the frequency of the pump pulse does not play a major role in the demagnetization process. To support this argument, it was found that the THz pulses which lead to a complete demagnetization induce a heating of the sample which is comparable to the Curie temperature. This shows that the sample heating is the major cause of the demagnetization process.

With the recent advances of the THz technology, one can get laser electromagnetic fields of several Tesla. This gives now the possibility to get a complete precession reversal of **M**(t). From a technological perspective, such a complete precession reversal of the magnetization would be a milestone, because it gives the chance to increase the speed of magnetic processors. A complete magnetization reversal can be also obtained by optical laser pulses, but the cost for this is a permanent modification of the film properties, which makes the process inappropriate for magnetic processors. The hope is (and this should be confirmed experimentally) that a complete reversal of **M**(t) with the use of THz laser pulses does not lead to permanent modification of the film properties.

IV. Density-matrix theory of ultrafast demagnetization

In former theories of ultrafast demagnetization after femtosecond laser pulses a combination of Fermi's golden rule of time-dependent perturbation theory with a band theory for the electron states was used, at best in the sense of a Hubbard model for the electron states. Thereby the effect of electron correlations in the sense of a density-matrix theory is neglected. It has been shown that these electron correlations have an essential effect on the ultrafast dynamics of nonmagnetic systems. In the present paper, the effects of these correlations in a magnetic system are investigated, namely in the ultrafast demagnetization after femtosecond laser pulses (described in sections I and II). Thereby the direct interaction channel is considered, namely the change of the magnetization by spin-flip scatterings of the electrons (excited by the laser pulse) with phonons. We apply the density-matrix theory [12]. The former theories the Markov approximation used, i.e., it is assumed that the system does not have a memory. Furthermore, a long-time approximation was used in these theories, i.e., the Sine function of the time-dependent perturbation theory is replaced by Dirac's delta functional. In the present theory, two steps are performed. In the first step, we also apply the Markov approximation and the long-time approximation, in the second step the full

density matrix theory is used. The comparison of the results of the two theories yields the effect of the electron correlations in the sense of a density matrix theory. A simple model for the electron band structure was used, and a simple model for the electron-phonon scattering matrix elements. The electron correlations in the sense of a density matrix theory are described by the expectation values $<C_{k,l}^{+} C_{k,j\#l}>$, where the operators $C_{k,l}^{+}$ and $C_{k,l}$ are operators which create electrons in the state /k,l>, where k is the electron wave vector and l is the band index. These expectation values are non-zero only when there is a transition of the electrons between different bands l and j#l., for instance, by spin-flip scattering processes. In the present paper, only transitions between neighboring bands are considered. In the former theories of ultrafast demagnetization a combination of Fermi's golden rule of the time-dependent perturbation theory with a band model for the electronic states was used at best in the sense of a Hubbard model. In the Hubbard model, there are also electron correlations. They describe the effect of Coulomb interactions between the electrons, given by the Hartree potential, and the deviations of the real Coulomb interaction energy from the mean Coulomb interaction energy (described by the Hartree potential). These correlations c are described by the expectation values $<C_{k,l}^{+} C_{k,l}>$ which are different from the above-discussed expectation values describing the electron correlations in the sense of the density matrix theory. The full density-matrix theory yielded a surprising result. In contrast to the essential effect of electron correlations in the fast dynamics of non-magnetic systems, their effect in a magnetic system, namely in the ultrafast demagnetization after femtosecond laser pulses, is just marginal. This is very interesting from a fundamental point of view for all people working on ultrafast magnetization dynamics, and it is also interesting from a practical point of view because the result justifies the combination of Fermi's golden rule with a band theory for the electron states, which does not include electron correlations in the sense of a density-matrix theory. Such a theory was used in all former publications on ultrafast dynamics after femtosecond laser pulses. It should be investigated whether this very interesting result for the present magnetic system (i.e., the ultrafast demagnetization after femtosecond laser pulses) holds also for other dynamical magnetization processes.

Conclusion

In this paper, various types of theories for fast and ultrafast magnetization dynamics were reviewed. The techniques used to generate fast and ultrafast magnetization dynamics were described. It has been explained that research on fast and ultrafast magnetization dynamics is very interesting from a fundamental point of view and from the view of technological applications.

Declaration: I, Manfred Fähnle, am the only author, and I have written the whole paper.

Data availability: Data is available on request.

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