Ultrafast Demagnetization After Femtosecond Laser Pulses: Transfer of Angular Momentum from the Electronic System to Magnetoelastic Spin-Phonon Modes

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Abstract When exciting a ferromagnetic film with a femtosecond optical laser pulse, then there is a partial demagnetization within a few hundred fs. This means that the angular momentum of the electronic system is transferred to the lattice. This transfer is calculated by describing the lattice degrees of freedom as spin-phonon eigenmodes which have a sharp angular momentum. Possible other lattice degrees of freedom are mentioned but not considered explicitly. For Ni and Fe the calculated amount of transfer of angular momentum to the spin-phonon eigenmodes is rather small.

Keywords Ultrafast demagnetization · Angular momentum · Spin-phonon modes

1 Introduction

When a ferromagnetic film is excited by a laser pulse of length of 30–100 fs, the sample is partially demagnetized within a few hundred fs with a following remagnetization on a longer time scale [1, 2]. Recently, it has been discussed both theoretically [3, 4] and experimentally [5] that very short pulses (5–6 fs) of higher intensity lead to a demagnetization on time scales of the order of just 5 to 35 fs. At present, this is the fastest possible microscopic manipulation of the magnetization in solids. Such ultrafast processes are of great importance for advanced data storage and data manipulation devices based on magnetism, if remagnetization to the original state is avoided. This is the case for ultrafast all-optical switching of ferromagnetic compounds [6] or antiferromagnetically coupled ferromagnetic multilayers [7], by which the magnetization can be switched in a reproducible manner by a femtosecond circularly polarized optical laser pulse. The two effects, ultrafast demagnetization and all-optical switching, are certainly related to each other. Therefore, it is of great interest to figure out the mechanisms of ultrafast demagnetization.

Although ultrafast demagnetization is investigated worldwide both experimentally and theoretically since 1996, a detailed understanding of the underlying physical processes is still lacking and several models compete. A possible mechanism is the super-diffusive transport of excited electrons mainly with majority spin orientation from the conducting ferromagnetic film to a conducting substrate [8]. In the following, we consider the situation with a non-conducting substrate for which the super-diffusive transport may lead to an inhomogeneous distribution of the magnetization in the film but not to a reduction of the total magnetic moment of the film. Other possible mechanisms are spin-flip scatterings of the excited electrons at other electrons [9, 10], at phonons [11], or at magnons [12, 13]. The spin-flip scatterings change the electronic spin- and orbital-angular momentum, leading to the demagnetization. Thereby, the electronic angular momentum has to be transferred to other degrees of freedom.

To describe the transfer of angular momentum, one had to take into account in principle explicitly the coupling of
the sample to the whole surroundings. However, a simplification arises because on the fs time scale, a process in the ferromagnetic sample can lead to reactions only within a very limited spatial distance from the sample. In a first step, one therefore could try to model the system by a Hamiltonian which contains all degrees of freedom of the lattice of the sample, i.e., to assume that the electronic angular momentum is mainly transferred to the lattice. An example for lattice degrees of freedom would be local elastic twists in the sample as reactions on local torques, twists which have certainly small amplitudes but which may have angular momentum which add up in the whole sample. However, these degrees of freedom are very complicated to be described mathematically. Another example for lattice degrees of freedom are collective motions of the atoms, which on an appropriately long time scale are described by phonons, and the scattering of electrons at phonons has been discussed in the literature (see, e.g., [11]). So far, however, only the change of the occupation numbers of the electronic states by electron-phonon scatterings and hence the contribution of electron-phonon scatterings to ultrafast demagnetization has been calculated. By these theories, also the occupation numbers of phononic states have been determined. The transfer of the angular momentum from the electronic system to the lattice via electron-phonon scattering has never been calculated explicitly. The reason is that usually the lattice dynamics are described by linearly polarized phonons which do not have a sharp angular momentum and for which the expectation value of the angular momentum is zero [14]. Therefore, it is not possible to calculate the change of the phononic angular momentum from the above discussed change of the phononic occupation numbers. The first idea to cope with this problem is to construct circularly or elliptically polarized phonon states with non-zero angular momentum by a linear combination of linearly polarized transverse phonon states [14]. But linear combinations are stationary states only if the two combined states are degenerate, which is the case only for special phonon wave vectors \( \mathbf{q} \) in high-symmetry directions of the phonon-Brillouin zone, and the formalism of electron-phonon scatterings requires stationary states. In isotropic systems like amorphous materials, the degeneracy is given for arbitrary \( \mathbf{q} \), and therefore it is possible to generate stationary phonon states with well-defined angular momentum [15]. But this does not help to describe the angular momentum transfer during demagnetization experiments which are mainly performed for crystalline ferromagnetic films. In the present paper, the collective dynamics of the ferromagnetic lattice are described by magnetoelastic spin-phonon modes which will be introduced in the next section and which carry a well-defined angular momentum. Within the framework of an electron-spin-phonon scattering Hamiltonian, we calculate for the first time explicitly the transfer of angular momentum from the electronic system to the lattice during ultrafast demagnetization.

### 2 Spin-Phonon Modes

In ref. [16], the angular momentum of phonons in a magnetic crystal was studied, in which the magnetism generates a magnetic field which can be represented by a vector potential \( \mathbf{A} \). The theory was for an ionic magnetic crystal where on an appropriately long time scale are described by phonons, and the scattering of electrons at phonons has been discussed in the literature (see, e.g., [11]). So far, however, only the change of the occupation numbers of the electronic states by electron-phonon scatterings and hence the contribution of electron-phonon scatterings to ultrafast demagnetization has been calculated. By these theories, also the occupation numbers of phononic states have been determined. The transfer of the angular momentum from the electronic system to the lattice via electron-phonon scattering has never been calculated explicitly. The reason is that usually the lattice dynamics are described by linearly polarized phonons which do not have a sharp angular momentum and for which the expectation value of the angular momentum is zero [14]. Therefore, it is not possible to calculate the change of the phononic angular momentum from the above discussed change of the phononic occupation numbers. The first idea to cope with this problem is to construct circularly or elliptically polarized phonon states with non-zero angular momentum by a linear combination of linearly polarized transverse phonon states [14]. But linear combinations are stationary states only if the two combined states are degenerate, which is the case only for special phonon wave vectors \( \mathbf{q} \) in high-symmetry directions of the phonon-Brillouin zone, and the formalism of electron-phonon scatterings requires stationary states. In isotropic systems like amorphous materials, the degeneracy is given for arbitrary \( \mathbf{q} \), and therefore it is possible to generate stationary phonon states with well-defined angular momentum [15]. But this does not help to describe the angular momentum transfer during demagnetization experiments which are mainly performed for crystalline ferromagnetic films. In the present paper, the collective dynamics of the ferromagnetic lattice are described by magnetoelastic spin-phonon modes which will be introduced in the next section and which carry a well-defined angular momentum. Within the framework of an electron-spin-phonon scattering Hamiltonian, we calculate for the first time explicitly the transfer of angular momentum from the electronic system to the lattice during ultrafast demagnetization.
where $B$ is the magnitude of the internal magnetic field produced by the magnetic atoms. In ref. [16], values of $\lambda$, in the range of several THz have been used.

The solution of the generalized eigenvalue equation (2) yields stationary magnetoelastic modes with complex polarization vectors $\mathbf{e}_{q,\lambda}$, which have a sharp non-zero angular momenta [16] which after quantization has the form

$$\ell_{q,\lambda}^z = \hbar \frac{\hat{v}_{q,\lambda}^z}{M} \mathbf{e}_{q,\lambda}.$$  

(6)

with

$$M = \begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}.$$  

(7)

Therefore, we are now in the position to calculate the change of the angular momentum of the lattice from the change of the occupation numbers for the magnetoelastic modes due to scattering of itinerant electrons during ultrafast demagnetization (see Section 3).

### 3 Scattering of Electrons at Spin-Phonon Modes

In a system with spin-orbit coupling the electronic states are not pure spin states but spin-mixed states. For given electronic wavevector $\mathbf{k}$ and band index $j$, the wavefunction is either dominant up (the contribution of the spin-up spinor is larger than the one of the spin-down spinor) or dominant down. We denote the dominant spin character by $s = \uparrow, \downarrow$. The electrons are scattered at phonons because the lattice-periodicity of the effective potential matrix $W(\mathbf{r}, \{ \mathbf{R}_n \})$ is broken by the displacements $\mathbf{u}(\mathbf{R}_n)$. The effective potential matrix in a spin-polarized system [17] is

$$W(\mathbf{r}, \{ \mathbf{R}_n \}) = \begin{pmatrix} V^\uparrow(\mathbf{r}, \{ \mathbf{R}_n \}) & 0 \\ 0 & V^\downarrow(\mathbf{r}, \{ \mathbf{R}_n \}) \end{pmatrix} +$$

$$+ \sum_l \frac{\hbar}{4m_e^2c^2} (\nabla_l V^\alpha(\mathbf{r}, \{ \mathbf{R}_n \}) \times \hat{\mathbf{p}}) \cdot \mathbf{e}_{q,\lambda},$$  

(8)

where $V^\alpha(\mathbf{r}, \{ \mathbf{R}_n \})$ denotes the effective potential of the spin-density functional theory that an electron with spin $\alpha$ feels at position $\mathbf{r}$. $\hat{\mathbf{p}}$ is the momentum operator and $\mathbf{e}_{q,\lambda}$ is the $l$-th Pauli matrix. In a system without spin-phonon interaction, the rate of transitions $W_{\alpha,k_s,k'_s,j,k,j'}$ between two states $\Psi_{j,k}$ and $\Psi'_{j',k'}$ by absorption or emission of a phonon with wavevector $\mathbf{q}$ (which yields $k'$) and polarization vector $\mathbf{e}_{q,\lambda}$ is given in the framework of Fermi’s golden rule by Eqs. (4), (5), (10) of [11]. Thereby, the rigid-ion approximation [11, 18] has been used in which the quantities $V^\alpha(\mathbf{r}, \{ \mathbf{R}_n \})$ are represented as

$$V^\alpha(\mathbf{r}, \{ \mathbf{R}_n \}) \approx \sum_{n=1}^N v_{0,n}^\alpha(\mathbf{r} - \mathbf{R}_n)$$  

(9)

where $N$ is the number of atoms and the $v_{0,n}^\alpha(\mathbf{r} - \mathbf{R}_n)$ are atomic potentials (which build the equilibrium potential $V^\alpha(\mathbf{r}, \{ \mathbf{R}_0,n \}) = \sum_{n=1}^N v_{0,n}^\alpha(\mathbf{r} - \mathbf{R}_0,n)$) which are rigidly displaced when bringing the atoms from $\mathbf{R}_0,n$ to $\mathbf{R}_n$. For the magnetoelastic phonon modes with complex $\mathbf{e}_{q,\lambda} = e^\imath e_{q,\lambda}$, the transition rate from the initial state $\Psi_{j,k}$ to the final state $\Psi'_{j',k'}$ is within Fermi’s golden rule

$$T_{j,k',j,k} = \frac{2\pi}{\hbar} \frac{1}{2NM M_\lambda q_{\lambda}} |\langle \Psi'_{j',k'} | \Omega_{k'} | \Psi_{j,k} \rangle|^2,$$

$$+ \frac{2\pi}{\hbar} \frac{1}{2NM M_\lambda q_{\lambda}} |\langle \Psi'_{j',k'} | \Omega_{k'} - i\Omega_{k} | \Psi_{j,k} \rangle|^2,$$  

(10)

where the first (second) part corresponds to the absorption (induced and spontaneous emission) of a phonon, and $b_{q,\lambda}$ is the Bose distribution function. Thereby, we have (with $\mathbf{r}_n = \mathbf{r} - \mathbf{R}_{0,n}$) the relation

$$\Omega_{k,\lambda} = -\sum_{\alpha} e^{i\mathbf{q} \mathbf{R}_{0,n}} (e^\imath e_{q,\lambda} \nabla_{\mathbf{r}_n}) \cdot \left( \begin{pmatrix} v_{0,\alpha}^\uparrow(\mathbf{r}_n) \\ 0 \\ v_{0,\alpha}^\downarrow(\mathbf{r}_n) \end{pmatrix} + \sum_l \frac{\hbar}{4m_e^2c^2} \frac{\partial v_{0,\alpha}^\alpha}{\partial \mathbf{r}_n} (\hat{\mathbf{L}}_m) \cdot \hat{\mathbf{e}}_{q,\lambda} \right),$$  

(11)

where $\hat{\mathbf{L}}_m = \mathbf{r}_n \times \hat{\mathbf{p}}$ is the angular momentum operator.

### 4 Non-Isotropy of the Total Hamiltonian

The electron-phonon scattering operator which leads to expression (10) is denoted as $\hat{H}_{el-ph}$. The electronic states $\Psi_{j,k}$ and the related energies $\epsilon_{j,k}$ are calculated ab initio with the relativistic version [19] of the linear-muffin-tin-orbital (LMTO) method [20] in the local-spin-density approximation (LSDA) [21, 22] and in atomic-sphere approximation (ASA) [20]. The corresponding Hamiltonian is denoted as $\hat{H}_{el}$. The spin-phonon modes are calculated from (2) and the corresponding Hamiltonian is denoted as $\hat{H}_{ph}$. The total Hamiltonian is

$$\hat{H} = \hat{H}_{el} + \hat{H}_{ph} + \hat{H}_{el-ph}.$$  

(12)

For all parts of the Hamiltonian, the positions of the atoms are fixed on the time average. The electronic part $\hat{H}_{el}$ thus describes the interaction of the electrons with a potential which is not isotropic. The phononic part $\hat{H}_{ph}$ contains the force-constant matrix $\Phi$ (Section 2), which for fixed atomic positions is also not isotropic. So, altogether, the Hamiltonian $\hat{H}$ is not isotropic. In reality, the demagnetization time is smaller than the inverse of a typical phonon frequency, and from this point of view the description of the lattice dynamics by phonons is not really adequate. However, it should be noted that to the knowledge of the authors...
all theory papers on ultrafast demagnetization start from a Hamiltonian which is not isotropic in the above discussed sense. Therefore, the total angular momentum is not conserved in such models. The hope is that such an anisotropic Hamiltonian is nevertheless a good model for the change of the electronic angular momentum due to spin-flip scatterings. And it is interesting to know how much of the electronic angular momentum is transferred to the lattice and hence the internal magnetic field is reduced, and therefore the value of the parameter $\lambda_e = \frac{g^B}{2M}$ decreases. We will see in the figures that then the rate of change of the phononic angular momentum also decreases beyond the value calculated for $t = t_s$. For the phonons, we have

$$b_{q,\lambda} = \left[ \exp \left( \frac{\hbar \omega_{q,\lambda}}{k_B T_{\text{ph}}} \right) - 1 \right]^{-1}$$

where $T_{\text{ph}}$ is the phonon temperature which is assumed to be not affected by the laser heating, i.e., $T_{\text{ph}} = 293$ K at $t = t_s$.

Using Boltzmann’s rate equations, the rate of change of the angular momentum of the electronic system per atom (which is dominated by the spin-angular momentum) is given by

$$\frac{dJ^c}{dt} = \frac{1}{\Omega_{BZ}^2} \sum_{jj'\lambda} \int_{BZ} d^3 k' \int_{BZ} d^3 k (S_{jj'}^{\lambda} - S_{jj}^{\lambda}) \cdot f(\epsilon_{jj'}^{\lambda}, \epsilon_{jj}^{\lambda}, T_{el}(t)) \left[ 1 - f(\epsilon_{jj'}^{\lambda}, \epsilon_{jj}^{\lambda}, T_{el}(t)) \right] T_{jj'}^{\lambda} \delta_{\lambda}$$

(14)

with $T_{jj'}^{\lambda}$ given by (10). $\Omega_{BZ}$ is the volume of the first Brillouin zone, and $S_{jj}^{\lambda}$ is the expectation value of the electronic spin angular momentum. For the rate of change of the angular momentum of the system of magnetoelastic phonons per atom, the Boltzmann theory gives

$$\frac{dJ^p}{dt} = \frac{1}{\Omega_{BZ}^2} \sum_{jj'\lambda} \int_{BZ} d^3 q \int_{BZ} d^3 k \cdot P_{q,jj'\lambda}$$

(15)

with

$$P_{q,jj'\lambda} \equiv \frac{2\pi}{\hbar} \frac{\hbar}{2NM_A o_{q,\lambda}} |\langle \Psi_{jj'}^{\lambda} | \Omega_{q}^{\lambda} + i\Omega_{\lambda}^{\lambda} | \Psi_{jj}^{\lambda} \rangle|^2$$

$$- b_{q,\lambda} f(\epsilon_{jj'}^{\lambda}, \epsilon_{jj}^{\lambda}, T_{el}(t)) \left[ 1 - f(\epsilon_{jj'}^{\lambda}, \epsilon_{jj}^{\lambda}, T_{el}(t)) \right] \cdot \delta(\epsilon_{jj}^{\lambda} + \hbar \omega_{q,\lambda})$$

$$+ \frac{2\pi}{\hbar} \frac{\hbar}{2NM_A o_{q,\lambda}} |\langle \Psi_{jj'}^{\lambda} | \Omega_{q}^{\lambda} - i\Omega_{\lambda}^{\lambda} | \Psi_{jj}^{\lambda} \rangle|^2$$

$$(b_{q,\lambda} - 1) f(\epsilon_{jj'}^{\lambda}, \epsilon_{jj}^{\lambda}, T_{el}(t)) \left[ 1 - f(\epsilon_{jj'}^{\lambda}, \epsilon_{jj}^{\lambda}, T_{el}(t)) \right] \cdot \delta(\epsilon_{jj'}^{\lambda} - \epsilon_{jj}^{\lambda} + \hbar \omega_{q,\lambda})$$

(16)

The first term of (16) describes the absorption of a phonon with wavevector $q$, and the second term describes the emission of a phonon with that wavevector. The integrals over $k, k'$ in (14) and over $k, q$ in (15) are replaced by sums over wavevectors on a discrete lattice. To evaluate the $\delta$-functions, it is necessary to replace them by their Gaussian identity with a smearing parameter $\sigma$. The calculations are made for different grids of wavevectors and for different
smearing parameters. We checked for convergence keeping the product $\sigma \cdot N_1$ constant where $N_1$ is the number of grid points in one direction. The largest grid was a $40 \times 40 \times 40$ $k$-point mesh in the first Brillouin zone.

6 Results

The results for $\frac{dJ_{\text{el}}}{dt}$ and $\frac{dJ_{\text{ph}}}{dt}$ at $t = t_s$ and for the sum of the two quantities

$$\delta J = \left| \frac{dJ_{\text{el}}}{dt} \right| + \left| \frac{dJ_{\text{ph}}}{dt} \right|$$

are given in Fig. 1 for Ni and for Fe, for various values of the parameter $\lambda_z$. The equation for the parameter $\lambda_z$ is

$$\lambda_z = \frac{q B}{2M_A}$$

where $q$ is the charge of the ion (10e for Ni and 8e for Fe) and $B$ the magnetic field. Mössbauer experiments yield for the internal magnetic field in Ni and Fe the values 7.6 T for Ni [23] and 33 T for Fe [24]. This gives the values $\lambda_z = 0.06$ GHz for Ni and $\lambda_z = 0.23$ GHz for Fe. It is seen that in general only part of the change of $\frac{dJ_{\text{el}}}{dt}$ is compensated by a change of $\frac{dJ_{\text{ph}}}{dt}$. This corresponds to the fact that due to the non-isotropy of the Hamiltonian the total angular momentum need not be conserved. Only for certain values of $\lambda_z$, the total angular momentum is conserved. For these values of $\lambda_z$, the part $\hat{H}_{\text{el-\text{ph}}}$ of the Hamiltonian compensates the non-isotropies of the parts $\hat{H}_{\text{el}}$ and $\hat{H}_{\text{ph}}$. However, such values are unrealistic for Ni and Fe (see above).

Obviously, the total angular momentum is not conserved when using a non-isotropic Hamiltonian, and therefore the speed of demagnetization is not determined by the need for angular momentum conservation. It is determined by the
magnitude of the energy which is injected from the laser pulse to the electronic system and which drives the system out of equilibrium, the farther the larger this injected energy is. In the present paper, we have calculated the demagnetization rates for the time directly after the action of the laser pulse. The rates evolve with time, first because the deviation from an equilibrium situation becomes smaller, and second because the value of $\lambda_z$ decreases during demagnetization. Our guess is that the angular momentum transfer does change strongly with time. The calculated rate of change $\frac{dM}{dt}$ gives a rate of change $\frac{dM}{dt}$ of the electronic magnetic moment per atom which is about $2\mu_B \frac{dM}{dt}$. This can be compared to experimental values for $\frac{dM}{dt}$. From ref. [25], one can learn that for Ni after a laser fluence which raises the electron temperature to 1000 K, the demagnetization rate is $\frac{dM}{dt} = 0.1\mu_B/(100 \text{ fs atom})$, and from ref. [12], we get $\frac{dM}{dt} = 0.2\mu_B/(100 \text{ fs atom})$ for Fe, which compares to our calculated values for $\lambda_z = 0.06$ GHz of $\frac{dM}{dt} = 0.04\mu_B/(100 \text{ fs atom})$ for Ni and for $\lambda_z = 0.23$ GHz of $\frac{dM}{dt} = 0.013\mu_B/(100 \text{ fs atom})$ for Fe. Of course, we cannot expect a perfect agreement between our theoretical values and the experimental values because we considered from the various contributions to ultrafast demagnetization just the contributions of spin-flip scatterings of electrons at spin-phonons.

7 Conclusions

We have calculated for Ni and Fe the transfer of angular momentum from the spin system to the lattice after excitation of the system with a fs optical laser pulse. The transfer is due to the scattering of the excited electrons at the spin-phonon eigenmodes which carry a well-defined angular momentum. Thereby, we have used a Hamiltonian which is not isotropic as all the Hamiltonians used in the literature to describe ultrafast demagnetization. For realistic values of a parameter describing the strength of the spin-phonon interaction, only a very small part of the change of the electronic angular momentum is transferred to the lattice. For a more realistic description of the whole situation, one should include in the Hamiltonian other degrees of freedom to which the angular momentum can be transferred, in a first step other degrees of freedom for the lattice dynamics. We have given suggestions for more realistic lattice degrees of freedom.

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