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Laser induced ultrafast demagnetization in diluted magnetic semiconductor nanostructures

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Abstract. We present a dynamical model that reproduces the observed time evolution of the magnetization in diluted magnetic semiconductor films after weak laser excitation. Based on a many-particle expansion of the exact p–d exchange interaction, our approach goes beyond the usual mean-field approximation. Numerical results demonstrate that the hole spin relaxation plays a crucial role for explaining the ultrafast demagnetization processes observed experimentally. The influence of the laser power on the magnetization dynamics is also investigated.

PACS. 78.20.Ls Magnetooptical effects – 78.30.Fs III-V and II-VI semiconductors

1 Introduction

Ultrafast light-induced magnetization dynamics in ferromagnetic films and in diluted magnetic semiconductor (DMS) nanostructures is today a very active area of research. From the observation of the ultrafast dynamics of the spin magnetization in nickel films [1] and the analogous processes in ferromagnetic semiconductors [2], special interest has been devoted to the development of dynamical models able to mimic the time evolution of the magnetization on both short and long time scales. In III-V ferromagnetic semiconductors such as GaMnAs and InMnAs a small concentration of Mn ions is randomly substituted to cation sites so that the Mn–Mn spin coupling is mediated by the hole-ion p–d exchange interaction, allowing the generation of a ferromagnetic state with a Curie temperature of the order of 50 K [3]. The magnetism can therefore be efficiently modified by controlling the hole density through doping or by excitation of electron-hole pairs with a laser pulse.

III-Mn-V ferromagnetic semiconductors offer the advantage of providing a clear distinction between localized Mn impurities and itinerant valence-band hole spins, thus allowing the basic assumptions of the Zener theory to be satisfied [4]. Based on this hypothesis, a few mean-field models have been successfully applied for modelling the ground properties of DMS nanostructures. Ultrafast demagnetization in DMS is a phenomenon where the p–d exchange interaction cause a flow of spin polarization and energy from the Mn impurities to the holes, which is subsequently converted to orbital momentum and thermalized through spin-orbit and hole-hole interactions [5]. Since energy and spin polarization transfer is a many-particle effect, the mean-field Zener approach cannot provide a satisfying explanation of the ultrafast demagnetization regime that has been observed in DMS films [6,7]. A phenomenological approach able to take into account this energy flux was given in [1] where a model based on three temperatures was derived.

In this paper we derive a dynamical model based on the Green's function formalism and a many-particle expansion of the exact p–d exchange interaction via Feynman's diagrams. Our approach extends the Zener model beyond the usual mean-field approximation. In agreement with recent experimental results [7] our simulations show that, depending on the initial lattice temperature and the laser power, a decrease of a few percent of the total magnetization in a picosecond time scale is observed. Finally we present numerical results which highlight the crucial role played by the hole spin relaxation time due to the spin-orbit interaction in the demagnetization processes.

2 Time evolution model

We model the DMS film as a system consisting of heavy holes (with density N^h) and Mn ions (with density N^M and spin $S^M = 5/2$) strongly coupled by spin-spin interaction. We denote with $a^\dagger_{k,s}$ ($a_{k,s}$) and $b^\dagger_{\eta,m}$ ($b_{\eta,m}$) the creation (annihilation) operators of a hole with spin projection s and quasi-momentum k and an ion with

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spin projection m and spatial position R_η respectively. The time evolution of the system is governed by the Hamiltonian

$$\mathcal{H} = \sum_{k,s} \varepsilon_{k,s} a^\dagger_{k,s} a_{k,s} + \mathcal{H}_{pd}.$$

In the parabolic band approximation the kinetic energy of the holes reads $\varepsilon_{k,s} = E^h - \frac{\hbar^2 k^2}{2m^*}$ where E^h is the valence band edge. The Kondo-like exchange interaction $\mathcal{H}_{pd}$ is given by

$$\mathcal{H}_{pd} = \frac{\gamma}{V} \sum \left(b^\dagger_{\eta,m'} b_{\eta,m} a^\dagger_{k',s'} a_{k,s} \right) \mathcal{V}\, e^{i(k'-k)R_\eta}$$
$$\mathcal{V} = \mathbf{J}_{m',m}\, \boldsymbol{\sigma}_{s',s}$$

where the sum is extended over all indices, γ is the p–d coupling constant, V is the volume of the system, and $\boldsymbol{\sigma}$, $\boldsymbol{J}$ are the spin matrices related to the holes and to the ions respectively.

In this section we give a detailed derivation of the time evolution equations for the hole mean magnetization. The Mn ions system can be treated in a similar way. We define the hole Green's functions as

$${}^h\mathcal{G}_{k,s,k',s'}(t,t') = -\frac{i}{\hbar} \frac{\left\langle \Psi \left| \mathcal{T}\left[a_{k,s}(t) a^\dagger_{k',s'}(t') \right] \right| \Psi \right\rangle}{\langle \Psi | \Psi \rangle}, \quad (1)$$

where Ψ denotes the many-particle wavefunction containing both the holes and ions degrees of freedom. To compact notations hereafter we will use the collective indices $\mathbf{s} = (k,s)$ and $\mathbf{m} = (\eta, m)$. We introduce new time variables $T = (t'+t)/2$ and $\tau = t - t'$ and we derive the time evolution Dyson equation for the hole density matrix. In the limit $\tau \to 0^-$ we obtain

$$\left[i\hbar \frac{1}{2} \frac{\partial}{\partial T} + \varepsilon_{\mathbf{s}} - \varepsilon_{\mathbf{s}'} \right] {}^h\mathcal{G}^<_{\mathbf{s},\mathbf{s}'}(T,T) = \sum_{\mathbf{s}_1} \int dt_1$$
$$\times \Big[\Sigma^r_{\mathbf{s},\mathbf{s}_1}(T,t_1)\, {}^h\mathcal{G}^<_{\mathbf{s}_1,\mathbf{s}'}(t_1,T) + \Sigma^<_{\mathbf{s},\mathbf{s}_1}(T,t_1)\, {}^h\mathcal{G}^a_{\mathbf{s}_1,\mathbf{s}'}(t_1,T)$$
$$- {}^h\mathcal{G}^r_{\mathbf{s},\mathbf{s}_1}(T,t_1)\, \Sigma^<_{\mathbf{s}_1,\mathbf{s}'}(t_1,T) - {}^h\mathcal{G}^<_{\mathbf{s},\mathbf{s}_1}(T,t_1)\, \Sigma^a_{\mathbf{s}_1,\mathbf{s}'}(t_1,T) \Big] \quad (2)$$

where Σ is the hole self-energy term generated by the exchange interaction. In the above expression, the superscript r (a) denotes retarded (advanced). To derive equation (2) the Langreth theorem has been used [9]. We have applied our dynamical model to a semiconductor thin film with a thickness of the order of one hundred nanometers. In the limit of wide DMS films, we can assume that the Hamiltonian of the system is both time and space invariant. Furthermore the evolution system can be considerably simplified if we assume that the thermalization scattering processes of the hole gas are instantaneous processes without memory effects (Markovian approximation). For a (III, Mn)V DMS structure the time evolution of the carrier density matrix is at least one order of magnitude longer than the correlation time of the thermal bath. In this regime the Markovian approximation is fully satisfied [8]. Within this approximation equation (2) becomes

$$i\hbar \frac{1}{2} \frac{\partial n^h_{s,s}}{\partial T} = \frac{1}{N^h} \frac{1}{2\pi\hbar} \sum_{\mathbf{s}_1,k} \int \Big[{}^h\mathcal{G}^>_{\mathbf{s},\mathbf{s}_1}(T,T)\, \widetilde{\Sigma}^<_{\mathbf{s}_1,\mathbf{s}}(E,T)$$
$$- {}^h\mathcal{G}^<_{\mathbf{s},\mathbf{s}_1}(T,T)\, \widetilde{\Sigma}^>_{\mathbf{s}_1,\mathbf{s}}(E,T) \Big]\, dE, \quad (3)$$

where the tilde symbol denotes the Fourier transform of the Green's function in energy variables

$$\widetilde{\mathcal{G}}_{\mathbf{s},\mathbf{s}'}(E,T) = \int \mathcal{G}_{\mathbf{s},\mathbf{s}'} \left(T + \frac{\tau}{2}, T - \frac{\tau}{2} \right) e^{\frac{i}{\hbar}E\tau}\, d\tau,$$

and the mean spin hole density is defined as

$$n^h_{s',s}(T) = \frac{1}{N^h} \sum_k \left\langle a^\dagger_{k,s'}(T) a_{k,s}(T) \right\rangle = \frac{\hbar}{i} \frac{1}{N^h} \sum_k {}^h\mathcal{G}^<_{k,s,k,s'}.$$

The self-energy appearing in equation (3) can be estimated by means of the usual diagrammatic Feynman approach. We have included in the diagrammatic expansion the Hartree and the ladder contributions. Within this approximation the first integral of equation (3) gives (and similarly for the other term)

$$\frac{1}{2\pi} \sum_{k_1,s_1,k} \int {}^h\mathcal{G}^>_{k,s,k_1,s_1}(T,T)\, \widetilde{\Sigma}^<_{k_1,s_1,k,s}(E,T)\, dE =$$
$$\sum_{m,m',s'} \mathcal{V}(m,m',s,s') \mathcal{V}(m',m,s',s) \Xi^h \sum_\eta f_b(\eta) \quad (4)$$

where we have defined

$$\Xi^h = e^{-\frac{\Delta E_{MF}}{k_B T}} \sum_{k',k} f_a\, h\, \delta(\Delta\mathbb{E})$$
$$f_b = \langle b^\dagger_{\eta,m} b_{\eta,m} \rangle \left(1 - \langle b^\dagger_{\eta,m'} b_{\eta,m'} \rangle \right)$$
$$f_a = \langle a^\dagger_{\mathbf{s}} a_{\mathbf{s}} \rangle \left(1 - \langle a^\dagger_{\mathbf{s}'} a_{\mathbf{s}'} \rangle \right)$$
$$h = \frac{1 + e^{\left[\gamma s' N^M \langle M \rangle + \varepsilon_k\right]/k_B T}}{1 + e^{\left[\gamma s' N^M \langle M \rangle + \varepsilon_{k'}\right]/k_B T}}. \quad (5)$$

We have assumed that the non-diagonal elements of the density operators vanish. According to the Zener model the ground state of the system can be estimated by taking into account only the mean field interaction between the holes and the magnetic ions. The hole gas experiences a mean magnetic field equals to $\overline{M} = \langle M \rangle N^M$ and in turn generates a mean field acting on the ions system equals to $\overline{S} = \langle S \rangle N^h$ where $\langle M \rangle = \sum_{m=-5/2}^{5/2} m\ n^M_m$ and $\langle S \rangle = \sum_{s=-1/2}^{1/2} s\ n^h_s$. The mean-field contribution to the total energy is thus $\Delta\mathbb{E} = \varepsilon_{k'} - \varepsilon_k + \gamma \left[(s'-s)\, \overline{M} + (m'-m)\, \overline{S} \right]$. By converting the sum over k in equation (5) by the corresponding integral with respect to the

energy variable $E = \varepsilon_k$ we obtain

$$\frac{\Xi^h}{V^2} = e^{-\frac{\Delta E_{MF}}{k_B T}} \int f_a \; h \; \rho(E)\rho(E - \Delta E_{MF}) \, dE \quad (6)$$

where $\Delta E_{MF} = \gamma \left[(s' - s)\,\overline{M} + (m' - m)\,\overline{S}\right]$ and ρ denotes the hole density of states. In the limit $\gamma\overline{S} \ll \gamma\overline{M} \ll \varepsilon_k$ we have

$$\frac{\Xi^h}{V^2} \simeq N^h \left(\frac{2m^*}{\hbar^2}\right) \sqrt[3]{3\pi^2 N^h} e^{-\frac{\Delta E_{MF}}{K_B T}} n_s^h \left(1 - n_{s'}^h\right).$$

Finally, inserting Ξ^h in equation (4) and averaging out over the quasi-momentum k and the randomly distributed positions R_η, we obtain

$$\frac{dn_s^h}{dt} = 2\xi N^M \frac{s}{|s|} \sum_{m=-S^M}^{S^M - 1} \left(S^M - m\right) \times \left(S^M + m + 1\right) \left(\mathcal{Z}_{m,m+1}^{1/2,-1/2} - \mathcal{Z}_{m+1,m}^{-1/2,1/2}\right) \quad (7)$$

$$\frac{dn_m^M}{dt} = 2\xi N^h \sum_{\sigma=\pm 1} \left(S^M - \sigma m + 1\right) \times \left(S^M + \sigma m\right) \left(\mathcal{Z}_{m,m-\sigma}^{-\sigma/2,\sigma/2} - \mathcal{Z}_{m-\sigma,m}^{\sigma/2,-\sigma/2}\right)$$

$$\mathcal{Z}_{m,m'}^{s,s'} = n_m^M \left(1 - n_{m'}^M\right) \; n_s^h \left(1 - n_{s'}^h\right) e^{-\frac{\Delta E_{MF}}{k_B T}}$$

$$\xi = 2\pi\gamma^2 \frac{m^*}{\hbar^3} \sqrt[3]{3\pi^2 N^h}. \quad (8)$$

In the above expression, the mean ion spin density is defined as $n_m^M = \frac{1}{N^M} \sum_\eta \langle b_{\eta,m}^\dagger b_{\eta,m} \rangle$.

3 Demagnetization processes in DMS

In this section we discuss the spin dynamical processes in a DMS arising from a femtosecond laser pulse. We assume that before the laser is turned on, the ion-hole system is at equilibrium with the phonon bath at the lattice temperature T^L, so that the ground state can be well described by the Zener-type model described in [11]. The laser excitation generates a non-thermal electron-hole pairs distribution.

By means of the Coulomb hole-hole interaction, the hole distribution undergoes a quasi-instantaneous thermalization (within a few tens of femtoseconds) towards a Fermi-Dirac distribution with temperature and chemical potential T^h and μ^h respectively [12,13]. In particular, we consider an excitation by a monochromatic laser pulse tuned at the energy E_l and having a pump fluence P_f. To estimate the energy E_{ex} transferred initially from the electromagnetic field to the kinetic energy of holes and electrons, following [8], we assume that the fraction of the laser pulse energy imparted to the holes is 1/4 of the photon energy. The total injected kinetic energy is thus $E_{ex} = n_{ex}^h E_l' \eta$ with $E_l' = E_l - E_g - (\varepsilon_c^1 + \varepsilon_v^1)$ and $\varepsilon_c^1, \varepsilon_v^1$ are the first eigenvalues of the valence and conduction bands. n_{ex}^h is the density of photo-created particles and η is the ratio of kinetic energy absorbed by the electron gas which can be estimated in the spherical band approximation as $\eta = m_\parallel^{HH}/(m_\parallel^c + m_\parallel^{HH})$ [14] with $m_\parallel^{HH}$ ($m_\parallel^c$) the effective mass of the heavy hole (electron) in the parallel direction of the sample.

After the laser excitation, the ions system strongly interact with the out-of-equilibrium hole gas by means the $\mathcal{H}_{pd}$ exchange interaction. Since the p–d interaction conserves the total angular momentum, it cannot by itself change the total magnetic moment but it can only redistribute the spin polarization from one system to another. Net relaxation of the magnetization requires another independent process, which has been recognized to be the hole spin-orbit interaction that relaxes the net hole spin polarization in a sub-picosecond time scale. Short spin relaxation time of holes in DMS is thus essential for the explanation of measured changes of magnetization. It is worth noticing that no direct ion spin relaxation phenomena are present at the femtosecond time scale. Thus, despite the fact that the spin-orbit mechanism acts only on the hole subsystem, it is able to give rise to an ultrafast decrease of the total magnetization of the system (due to the difference between the concentration of the hole gas and the Mn impurity, the contribution of the hole gas to the total magnetization is almost negligible).

By means of a standard relaxation model, we include both the spin-orbit mechanism and the cooling of the kinetic energy of the excited holes driven by the phonons. The corresponding equations read

$$\left.\frac{\partial n_s^h}{\partial t}\right|_{so} = \frac{n_s^h - \overline{n_s^h}}{\tau_{SO}} \quad (9)$$

$$\frac{\partial T^h}{\partial t} = \frac{T^h - T^L}{\tau_L} \quad (10)$$

where $T^h(t)$ and T^L are the temperatures of the holes and the lattice, $\overline{n_s^h}(n_m^M, T^h)$ is the self-consistent quasi-static equilibrium hole spin distribution computed from the Zener-type model of [11]. The temperature relaxation rate $\tau_L^{-1} = \tau_{OP}^{-1} + \tau_{AP}^{-1}$ takes into account both the acoustic phonon scattering with $\tau_{AP} = 200$ ps and the optical phonon scattering with $\tau_{OP} = 1$ ps for $T^h > 50$ K and $\tau_{OP} = \infty$ for $T^h < 50$ K [13].

4 Numerical results

In order to study the time evolution of the mean magnetization of a GaMnAs/GaAs DMS heterostructure occurring after the interaction with a linearly polarized femtosecond laser pulse, we have applied our time dependent model constituted of equations (7), (8) along with equations (9), (10). Based on the experiment of [7], we consider a sample consisting of a 73 nm $Ga_{0.925}Mn_{0.075}As$ layer deposited on a GaAs buffer layer and a semi-insulating GaAs substrate. The background hole density is 10^{20} cm^{-3}. For the details of the chemical composition of the sample we refer to [7].

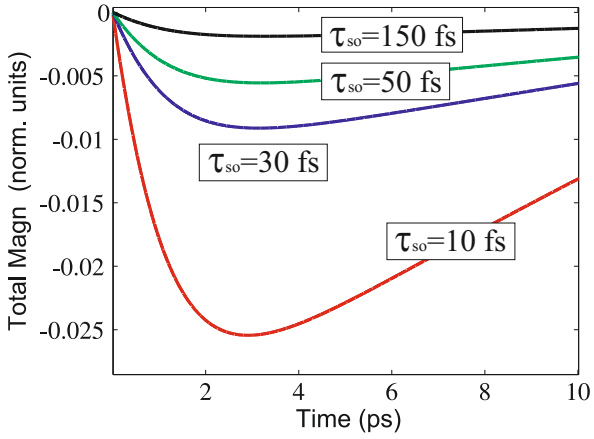

Fig. 1. (Color online) Time evolution of the total magnetization for different values of τ_{SO}. The lattice temperature is $T_L = 70$ K and the laser pump fluence is 1 μJ cm^{-2}.

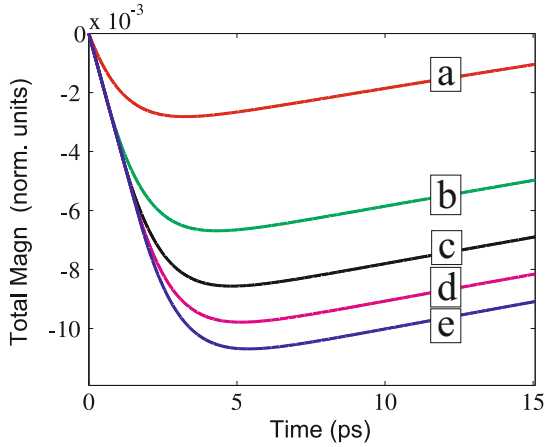

Fig. 2. (Color online) Time evolution of the total magnetization for different values of the laser power: (a) $P_f = 1$ μJ cm^{-2}, (b) $P_f = 10$ μJ cm^{-2}, (c) $P_f = 20$ μJ cm^{-2}, (d) $P_f = 30$ μJ cm^{-2}, (e) $P_f = 40$ μJ cm^{-2}. The lattice temperature is $T_L = 70$ K and $\tau_{SO} = 200$ ps.

In Figure 1 the time evolution of the total magnetization of the sample for different values of τ_{SO} is depicted. These results clearly demonstrate the critical role played by the spin-orbit relaxation time in the demagnetization processes. Estimation of τ_{SO} for a hole system is a rather difficult task since a first principle approach involves the evaluation of the projection of the hole eigenstate on the Bloch basis far beyond the validity of the kp theory. Our results relate the spin relaxation time to the partial demagnetization of the sample (which is measured) and therefore, can provide an indirect way to estimate the order of magnitude of τ_{SO}. This finding is in agreement with the arguments put forward in [8,15].

The total demagnetization processes become independent on τ_{SO} ("spin bottleneck") when $\tau_{SO} < \tau_{sd}$ where τ_{sd} is the characteristic time of the spin polarization exchange between the holes and the ions. Contrary to the findings of [8], our model does not exhibit any saturation effect of the total demagnetization when τ_{SO} is of the order of 10 fs. We ascribe this difference to the different excitation regime used. Indeed, Cywiński and Sham in [8] have considered a strong excitation regime for which the Mn-ion spin temperature increased toward the Curie temperature. Thus, the total magnetization of the sample is destroyed and τ_{sd} grows until nearly 10 fs where the spin bottleneck is observed. In our case (weak laser excitation) this phenomenon should be observed in a sub-femtosecond regime. However, this ultrafast relaxation regime is not consistent with the approximations underlying our dynamical model.

Finally in Figure 2 we show the time evolution of the total magnetization for different laser pulse fluences (in these simulations τ_{SO} is fixed at 200 ps). We note that the total demagnetization displays a non-linear behaviour with respect to the laser power even in the weak excitation regime under consideration (here the laser pump fluence is: 1 μJ cm^{-2} $< P_f <$ 50 μJ cm^{-2}).

In summary, in order to describe the strong spin-spin scattering regime observed in DMS, we have derived a dynamical model that goes beyond the usual mean-field approximation. This model is based on a many-particle expansion of the p–d exchange interaction in terms of single-particle density functions. Since the experimental value of the hole spin-orbit relaxation time in GaMnAs/GaAs DMS heterostructure is not yet firmly established, numerical results reveal that this quantity plays a crucial role in the demagnetization processes. Furthermore, in the weak excitation regime the demagnetization exhibits a nonlinear behavior with respect to the power of the laser beam.

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