Femtosecond spin dynamics in the time domain

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Recently magnetization dynamics on the time scale of 100 fs has been observed. In this article, we explain this ultrafast spin dynamics by a microscopic many-body theory. Our theory yields a speed limit of the order of 10 fs for the spin dynamics in Ni. The fundamental mechanism is the dephasing of the initial excited states. We investigate the dephasing effects in detail as a function of the number of electronic states and K points. We also calculate the density of states as a crosscheck of our theory, where the typical satellite structure of nickel is present. Moreover, the magnetic moment of the monolayer is found to be $0.88\mu_B$, which is also consistent with the experimental results. Finally, based on our Hamiltonian, we briefly discuss the origin of ferromagnetism in our model. © *1999 American Institute of Physics.* [S0021-8979(99)28208-4]

The phase transition from ferromagnetism to paramagnetism in a typical ferromagnetic system occurs when the system temperature is heated above the Curie point or compensation point, where ''spins flip''. This process is usually applied in conventional magneto-optical recordings. The time needed to reverse spins is around 100 ps–10 ns, which is basically set by the spin–lattice interaction. An earlier experiment did show that, in Gd, the characteristic time for establishing a thermal equilibrium between the lattice and the spin systems is 100 ± 80 ps.[1] The data are explained by a two-temperature model for the spin and electron systems. A similar time scale is also observed in Ni.[2] A theory based on electron paramagnetic resonance and magnetocrystalline anisotropy[3] yields a spin–lattice relaxation time of 48 ps for Gd and of 304 ps for Ni.[4] It has remained unclear since the first experiment on Gd whether this so-called characteristic time is a real upper limit for demagnetization in ferromagnetic materials. Beaurepaire *et al.*[5] were the first to investigate demagnetization on a short time scale, much shorter than the characteristic time scale of spin–lattice interaction. Using the pump–probe technique, they observed very sharp demagnetization within 1–2 ps, where heating of the spin by the lattice is not yet involved, which indicates that the concept of spin temperature is not well defined on the ultrafast time scale. This ultrafast demagnetization has motivated further experimental[2,6] as well as theoretical studies.[7,8] Scholl *et al.*[2] showed that there are two different mechanisms for spin relaxation taking place on the two different time scales (<1 and several hundred ps). Hohlfeld *et al.*[6] demonstrated that the classical $M(T)$ curve could be reproduced for delay times longer than the thermalization time of the electron subsystem alone but shorter than the electron–lattice relaxation time. This indicates that demagnetization on the femtosecond time scale is purely electronic. Recently Aeschlimann *et al.*[9] found that the spin-resolved inelastic lifetime is around 10 fs and is different for the majority and minority spins. In the following we will use Ni as an example to study the spin dynamics on the femtosecond time scale.

We begin with a generic Hamiltonian

$$H=\sum_{i,j,k,l,\sigma,\sigma',\sigma'',\sigma'''} U_{i\sigma,j\sigma',l\sigma''',k\sigma''} c^{\dagger}_{i\sigma} c^{\dagger}_{j\sigma'} c_{k\sigma''} c_{l\sigma'''} + \sum_{\nu,\sigma,K} \mathscr{E}_{\nu}(K) n_{\nu\sigma}(K) + H_{\mathrm{SO}}, \tag{1}$$

where $U_{i\sigma,j\sigma',l\sigma''',k\sigma''}$ is the electron interaction, which can be described in full generality by the three parameters Coulomb repulsion U, exchange interaction J, and the exchange anisotropy ΔJ.[10] $c^{\dagger}_{i\sigma}(c_{i\sigma})$ are the usual creation (annihilation) operators in the orbital i with spin σ ($\sigma=\uparrow,\downarrow$). $\mathscr{E}_{\nu}(K)$ is the single-particle energy spectrum for band ν. We take a nickel thin film geometry as an example. $n_{\nu\sigma}(K)$ is the particle number operator in momentum space. H_{SO} is the spin orbit coupling. This is a typical many-body particle problem. One cannot solve it without simplification. In order to obtain a tractable model, we first build a two-hole basis set. In this basis set, for each Ni atom the dimension of the Hilbert space is 66. The matrix elements of the electron correlation for each atom can be obtained analytically. For each K point, we embed the electron correlation in the crystal field as given by the band structure. This treatment of correlations is analogous to a frequency dependent self-energy correction although formally avoiding it. With this treatment, we are able to exactly diagonalize the Hamiltonian for each K point explicitly. In order to characterize the spin dynamics, we calculate the magneto-optical Kerr effect (MOKE) characterized by the off-diagonal susceptibility $|\chi^{(1)}_{xy}(\omega,t)|$, where t is the delay time between the pump and probe beams and ω is the probe frequency.

Our previous results[7,8] showed that the speed limit of ultrafast spin dynamics is around 10 fs for a thin Ni film. Both material specific and experiment specific parameters affect the final results.[7,8] The exchange interaction and spin–orbit coupling influence the spin dynamics differently. We demonstrated that the fundamental mechanism responsible for the spin dynamics is the dephasing among the initial excited states. The number of states that are involved becomes very critical to the final observation of spin dynamics. It is well established that the loss of coherence among the

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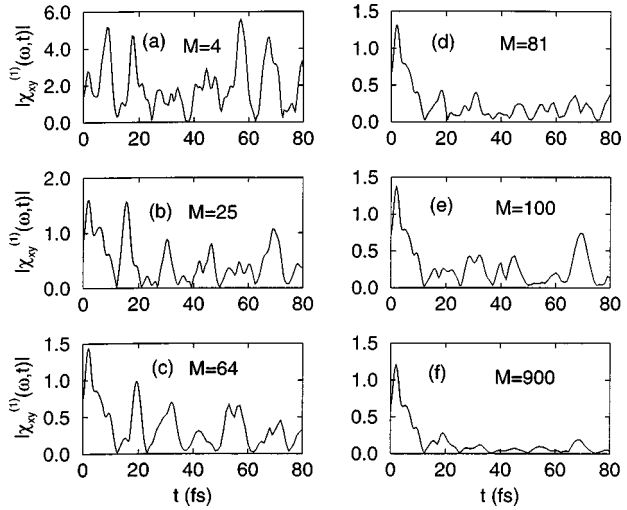

FIG. 1. Dephasing effect of ultrafast spin dynamics as a function of the number of K points M. As the number of K points increases, the coherence is gradually lost. The speed limit for the spin dynamics is around 10 fs. Here ω is 2 eV.

states is stronger if the number of states participating in the relaxation process is larger. Here we examine these effects by calculating $|\chi_{xy}^{(1)}|$ as a function of time t for a different number of K points.

The initial state is prepared 2 eV above the ground state, with a Gaussian broadening of 0.2 eV, which mimics a typical pump pulse width. We then probe the response after a delay time t. The probe frequency ω is 2 eV. Figure 1 shows the results of our calculation. In Fig. 1(a), the number of K points M is 4. Within 80 fs, one can observe several quasiperiodic oscillations of $|\chi_{xy}^{(1)}|$. The amplitude of the peak around 60 fs is roughly equal to that at 10 fs. This means that the coherence is largely kept among the initial states. In Fig. 1(b), we increase the number of K points to 25. $|\chi_{xy}^{(1)}|$ decreases within the first 10 fs, but comes back with almost the same amplitude at about 18 fs. Even at $t=30$ fs, the amplitude of the dominant peak exceeds half of the maximum of $|\chi_{xy}^{(1)}|$. Within 80 fs, five clear oscillations can be identified. Around 70 fs a large oscillation can be seen. All of these results reflect the fact that the coherence is still well preserved. If we increase M to 64, one can again see five clear oscillations, but their amplitudes slowly decrease with time, which indicates that the coherence is gradually lost. This trend continues as M is further increased to 81 [see Fig. 1(d)]: the loss of coherence is enhanced, but several small oscillations are still visible. When we increase M to 100 [Fig. 1(e)], the major change seen from Figs. 1(d)–1(e) is on a longer time scale. In Fig. 1(d) small humps have almost identical amplitudes while the amplitudes for the small humps are different in Fig. 1(e). The result becomes convergent when M is larger than 900. In Fig. 1(f), a pure decay of $|\chi_{xy}^{(1)}|$ with time can be seen, from this we estimate a spin relaxation time of around 10 fs. A detailed survey of Figs. 1(b)–1(f) reveals that, on a short time scale ($\leqslant$10 fs), the basic features of $|\chi_{xy}^{(1)}|$ are rather similar. The big difference occurs on a longer time scale. This is understandable since what is observed physically is essentially proportional to

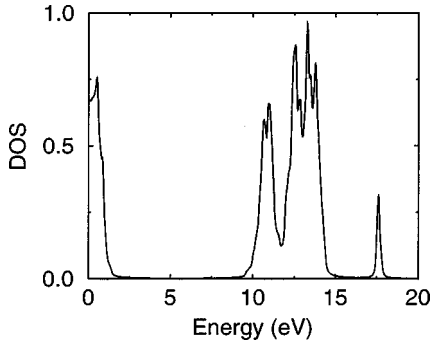

FIG. 2. Density of states for Ni thin film using a balanced band structure. The satellite structure appears around 10–14 eV for our unscreened value of 12 eV for the U parameter.

$e^{i(E_i-E_j)t/\hbar}=e^{i\Delta_{ij}t/\hbar}$. On a small time scale (small t), the states which have dominant contributions to the spin dynamics are those which have big energy differences Δ_{ij} between them. Even with a small number of K points, the relevant states are already present. That is why there is no significant difference between small and large numbers of K points. On a long time scale (large t), on the other hand, a small Δ_{ij} also contributes greatly to the final results. In the case of a small number of K points, a large number of states with small Δ_{ij} are absent. Thus the difference is rather conspicuous between small and large numbers of K points on a longer time scale.

Next we would like to check whether our Hamiltonian can describe the essential properties of Ni. Ni is a typical ferromagnetic material. The exchange interaction is very important. The density of states (DOS) exhibits a satellite structure. In Fig. 2, we show the DOS for Ni. One can see that the satellite structure appears above 10 eV for a generic set of parameters of nickel, which is consistent with the satellite structure found at 6 eV in Ni photoemission experiments.[11] It is important to note that this satellite structure results from Coulomb interaction and exchange interaction. In noble metals, an independent electron model accounts for the DOS well. But in transition metals, the inclusion of Coulomb and exchange interactions is definitely necessary as one can again see here. Because of this, theoretical treatments of transition metals are very difficult and are often limited. The beauty of our formalism is that we can reproduce even these essential properties visible in excited-state experiments such as photoemission.

In the following, we will show that by starting from a balanced band structure we are able to obtain a ferromagnetic ground state. It has been well established that a prerequisite to acquire a ferromagnetic ground state is a nonzero Coulomb interaction U and exchange interaction J. We can simply check this by setting both U and J to zero. By doing so, we find that the ground state is a singlet, i.e., a paramagnetic state, which contradicts the ferromagnetic nature of Ni. Once we use the generic sets of U and J of Ni, we obtain a triplet as its ground state, from which we find a magnetic moment of the monolayer, $0.88\mu_B$ (μ_B is the Bohr magneton). This magnetic moment is larger than that in the bulk material, which is consistent with the experimental observation.[12] Moreover, we are able to pinpoint some basic

features that actually show how Coulomb interaction and exchange interaction bring about the ferromagnetic state. We find that for our model it is in general not true that any nonzero Coulomb interaction or exchange interaction would result in a ferromagnetic phase. Actually there is a threshold which the Coulomb interaction or exchange interaction has to overcome before either leads to a ferromagnetic phase. The threshold is different for U and J due to their different natures. To investigate the origin of ferromagnetism quantitatively, we first set J and ΔJ equal to zero and examine the sole effect of U while keeping the hopping integrals as they are in order to get a correct band structure for Ni. We found that all the phases are ferromagnetic if U is larger than the threshold $U_c = 1.09\,\mathrm{eV}$. We also examine how J influences the ferromagnetic phase. Analogously we set the other interaction parameters, U and ΔJ, equal to zero. It is found that the ground state of the system becomes ferromagnetic if J is larger than $J_c = 0.29\,\mathrm{eV}$.

In conclusion, starting from a generic Hamiltonian, we calculated the ultrafast spin dynamics in ferromagnetic Ni. It was found that the theoretical speed limit is around 10 fs. The decay of the optical response is due to the dephasing among the initial states. This can be shown by changing the number of K points. For a small number of K points, such as $M = 25$, the coherence is well preserved. From $|\chi_{xy}^{(1)}|$, one can see several clear oscillations within 80 fs. Upon an increase of M, dephasing occurs. When one increases M to 81, a clear decay is observed. A sufficient number of K points is necessary in order to attain convergent results. The number of K points has a great impact on the long time tail of $|\chi_{xy}^{(1)}|$. On a short time scale, the basic features are similar. We carefully checked whether our Hamiltonian describes the ferromagnetic Ni film reasonably well. First of all, we correctly reproduced the satellite structure of the DOS, which is an excited-state many-body feature. The ground state is ferromagnetic with a magnetic moment of $0.88\mu_B$. We also briefly investigated the origin of the ferromagnetic phase. It was found that nonzero Coulomb interaction and exchange interaction are crucial to the ferromagnetic state. But their respective roles are different. Numerical results showed that there are different threshold values, U_c and J_c, for U and J to lead to a ferromagnetic phase.

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