Femtosecond demagnetization of ferromagnetic metal: cooperative precession of delocalized spins

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Abstract: A microscopic model of coherent Elliot-Yafet phonon inducing the delocalized spin precession is proposed to drive a cooperative femtosecond quenching of the magnetization in ferrromagnetic metal, beyond the phenomenological temperature model.

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1. Introduction

Spin order of a metallic ferromagnet is quenched in only a few hundred femtoseconds (fs) after the pulsed ultrashort laser irradiation [1]. A demagnetization time is typically 100-300 fs for ferromagnetic metals such as Ni or Fe. Technologically, this implies a potential application for the ultrahigh speed magnetic recording. Despite much experimental progresses during the last two decades, the underlying microscopic mechanism for the femtosecond demagnetization still remains highly controversial. Several mechanisms for the quenching of spin angular momentum have been proposed; Elliot-Yafet phonon scattering [2], Coulomb scattering [3], laser-induced scattering [4], relativistic effect [5] or spin current [6]. In the present study, based on the Elliot-Yafet spin-flipping phonon scattering, we investigate the delocalized (or itinerant) spin precession induced by the coherent Elliot-Yafet phonon and find the ultrafast demagnetization, consistent with the experimental observation.

2. Theory

The phenomenological temperature model describes the relevant dynamics by the electron temperature T_e (of "hot" electrons by the optical pumping) and the spin temperature T_s, or additionally the phonon temperature T_p. The basic physical picture behind the temperature model is, however, that an entity of the electron is artificially regarded as a spinless charge and a classical localized spin. Evidently, this is not correct for $3d$ ferromagentic metals under our consideration because a declocalized electron carries charge and spin simultananeously. Delocalized spins are attributed to the observed magnetism.

We now propose a novel theoretical model to capture the correct picture of delocalized spins. The Hamiltonian H comprises the transition metal H_{TM}, the Elliot-Yafet phonon scattering, and the optical pumping by the ultrashort laser pulse $A(\tau)$

$$H = H_{TM} + g\sum_{\mathbf{kq}}\sum_{\sigma} c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{kq}-\sigma}(b_{-\mathbf{q}} + b^{\dagger}_{\mathbf{q}}) + A(\tau)\sum_{\mathbf{kG}}\sum_{\sigma} c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k+G}\sigma}. \quad (1)$$

$c^{\dagger}_{\mathbf{k}\sigma}$ or $c_{\mathbf{k}\sigma}$ is the $3d$ band electron constructing $H_{TM} = \sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}\sigma} c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}$ with $\varepsilon_{\mathbf{k}\sigma} = \varepsilon_{\mathbf{k}} - \sigma\tilde{V}(n_{\uparrow} - n_{\downarrow})$ and $n_{\sigma} = \sum_{\mathbf{k}} \langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}\rangle$, where $\varepsilon_{\mathbf{k}}$ is the band energy and $\tilde{V}$ is the electron exchange energy. $b^{\dagger}_{\mathbf{q}}$ or $b_{\mathbf{q}}$ is the Elliot-Yafet phonon that induces the spin-flipping scattering and g is the scattering strength. The last term indicates the photoinduced interband transition with the reciprocal lattice vector $\mathbf{G}$. Within the linear response theory of the Neumann equation for the time-dependent density matrix, we could have coupled equations of motion of two essential quantities like charge density $\langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}\rangle$ and spin precession $\langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}-\sigma}\rangle$ incorporating the $\mathbf{q} = 0$ lattice displacement Q $(= \langle b_{-\mathbf{q}} + b^{\dagger}_{\mathbf{q}}\rangle)$.

$$i\frac{\partial}{\partial\tau}\langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}\rangle \approx g\langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}-\sigma}\rangle Q - g\langle c^{\dagger}_{\mathbf{k}-\sigma} c_{\mathbf{k}\sigma}\rangle Q \quad (2)$$

$$i\frac{\partial}{\partial\tau}\langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}-\sigma}\rangle \approx (\varepsilon_{\mathbf{k}\sigma} - \varepsilon_{\mathbf{k}-\sigma} - i\gamma_{\Delta})\langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}-\sigma}\rangle + g\langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}\rangle Q - g\langle c^{\dagger}_{\mathbf{k}-\sigma} c_{\mathbf{k}-\sigma}\rangle Q \quad (3)$$

In order for Eqs.(2) and (3) to be meaningful, an average displacement should be nonzero, that is, Q should be the coherent phonon. Quantum mechanics of the coherent phonon tell us that Q should satisfy [7]

$$\frac{\partial^2}{\partial\tau^2}Q+\gamma_Q\frac{\partial}{\partial\tau}Q+\omega_0^2Q+g\sum_{\mathbf{k}\sigma}\langle c^\dagger_{\mathbf{k}\sigma}c_{\mathbf{k}-\sigma}\rangle=\bar{g}N(\tau), \tag{4}$$

where the photoexcited charge density $N(\tau)$ is determined by the rate equation $\frac{\partial}{\partial\tau}N=-\gamma_N N+\eta A(\tau)$ in the lowest approximation. Solving the coupled equations of Eqs.(2)-(4), we keep track of the magnetization $M(\tau)$ $(=\sum_{\mathbf{k}}[\langle c^\dagger_{\mathbf{k}\uparrow}c_{\mathbf{k}\uparrow}\rangle-\langle c^\dagger_{\mathbf{k}\downarrow}c_{\mathbf{k}\downarrow}\rangle])$ and the spin precession $\Delta(\tau)$ $(=\sum_{\mathbf{k}}\langle c^\dagger_{\mathbf{k}\uparrow}c_{\mathbf{k}\downarrow}\rangle)$. Now it is confirmed that the coherent phonon plays a role of the transverse magnetic field and induces the delocalized spin precession and eventually decreases the magnetization.

3. Results and discussion

As shown in Fig.1, coherent phonon is found to drive the ultrafast quenching of the magnetization in the time of $\sim$100 fs, being consistent with the experimental observation. Depending on the competition between γ_N (relaxation of photoexcited charge density) and γ_Δ (relaxation of spin precession), the demagnetization profile is determined.

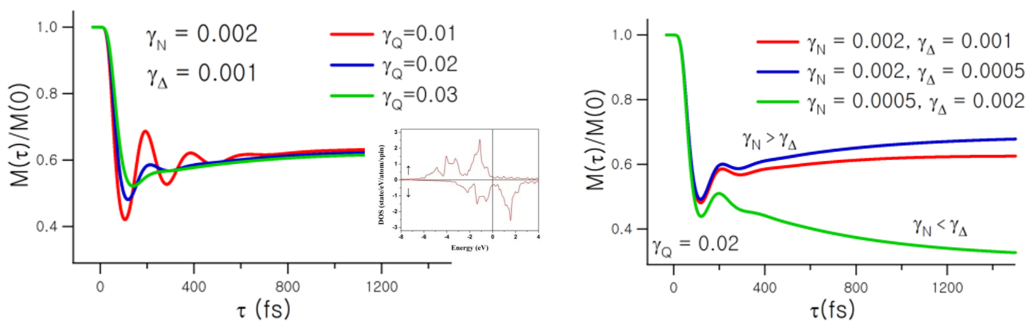

Fig. 1. Time-dependent magnetization of Fe after the ultrashort optical pumping. Inset of the left panel is density of states (DOS) of Fe, adopted in the calculation.

An analytic approach in the long time limit (i.e, except for the short time range within the pumping pulse length) enables to show $\partial M/\partial\tau\approx 4gQ(\tau)\mathrm{Im}[\Delta(\tau)]$, from which $\mathrm{Im}[\Delta(\tau)=0]$ would then give the extrema of $M(\tau)$. Therefore, when $\gamma_N>\gamma_\Delta$, the time $\tau_{\min}$ to reach the minimum of the magnetization $M(\tau)$ can be shown to be

$$\tau_{\min}=\frac{3\pi}{\sqrt{\omega_0^2-4\gamma_Q^2}}+\frac{4(\gamma_N-\gamma_\Delta)}{\gamma_Q\sqrt{\omega_0^2-4\gamma_Q^2}} \tag{5}$$

while, when $\gamma_N<\gamma_\Delta$, $M(\tau)$ approaches the minimum in the time scale of $\sim 1/\gamma_N$ asymptotically (See the right panel of Fig.1).

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