Ultrafast spin demagnetization by nonthermal electrons of TbFe alloy film

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An ultrafast spin demagnetization process of an amorphous Tb$_{35}$Fe$_{65}$ alloy film has been investigated by means of an all-optical pump-probe technique. Interestingly, steplike demagnetization on a subpicosecond time scale is observed before a much slower change on a time scale of tens of picoseconds. The steplike demagnetization at the subpicosecond scale is explained by the extended three-temperature model considering the interaction between a nonthermal electron and a spin system. The characteristic of subpicosecond demagnetization of TbFe alloy film is expected to be very useful in the manipulation of the spin state in ultrafast regime. © 2009 American Institute of Physics. [DOI: 10.1063/1.3130743]

Recently, much attention has been focused on the ultrafast manipulation of spin states in magnetic materials as a promising means to boost the device speed in spintronics. Since ultrafast spin demagnetization has been reported in ferromagnetic Ni by the pioneering work of Beaurepaire et al., many studies have been done on ultrafast dynamics of various magnetic materials using an all-optical pump-probe technique. Most of these experiments have been carried out on in-plane magnetic anisotropy materials such as Ni, Fe, FeRh, CrO$_2$, FeCr$_2$S$_4$, and CdCr$_2$Se$_4$. Over the past few years, as the demand for high-density recording has increased, perpendicular magnetic anisotropy (PMA) materials have been researched intensively because nanosized bit having PMA has the advantage of excellent thermal stability due to the high anisotropy energy of these materials. Although the large magnetic anisotropy and the coercivity characteristics of PMA materials make ultrafast switching difficult, their potentials for application are considerable when combined with the optically induced ultrafast phenomena of spin demagnetization and optomagnetic spin switching. Therefore, an understanding of their ultrafast response is important in order to realize a high recording speed in such systems. In this work, we have investigated the optically induced ultrafast demagnetization dynamics of TbFe alloy film with characteristics that are sensitive to temperature and that are easily controlled by a composition. By utilizing simultaneous measurements of the transient reflectivity and the magneto-optical Kerr signal, we report the observation of the two distinct demagnetization processes existed in different time scales of subpicosecond and tens of picoseconds, which could be understood through the nonthermal electron-spin interaction prior to the lattice-spin interaction.

The measurement was performed using an all-optical pump-probe technique. The experimental setup is shown schematically in Fig. 1(a). The Ti:sapphire laser pulse has an average power of 280 mW with a full width at half maximum of 30 fs. The pulse was divided into pump and frequency-doubled probe pulses with photon energies of 1.6 and 3.0 eV, respectively. The optical path difference between the two pulses was controlled by a delay stage with a maximum resolution of 1 fs. The pump (s-pol.) and the probe (p-pol.) pulses were focused onto a spot with a diameter of 1.5 μm. The transient reflectivity and magneto-optical Kerr rotation were then measured simultaneously using a balanced detection. Additionally, the Kerr ellipticity was measured by inserting a quarter-wave plate in front of a polarizing beam splitter.

![FIG. 1. (Color online) (a) Experimental configuration for the all-optical pump and probe measurement technique. The red line is for the pump and the blue line is for the probe beam. (b) The Δθ/θ (red circle) and Δκ/κ (blue triangle) signals are shown at 297 K with a pump-beam intensity of 9.9 mJ/cm$^2$. (c) Reflectivity (red rectangle) and Kerr signal (green diamond) differences between 297 and 310 K at a pump-beam intensity of 9.9 mJ/cm$^2$.](appliedphysicsletters/v94/i19/a94_192506_1_fig1.jpg)
It should be emphasized that extreme care was exercised to minimize several possible causes of artifacts in this experiment, as a magneto-optical signal in an ultrafast regime can be ascribed to optical artifacts and not to the magnetism.\textsuperscript{2-4,7} One of the main causes is apparently bleaching effect, as extensively described elsewhere.\textsuperscript{15} Therefore, we have conducted the dichroic pump-probe measurement of the normalized Kerr rotation ($\Delta \theta / \theta$) and ellipticity signals ($\Delta e / e$) at 297 K, as seen in Fig. 1(b). This graph clearly indicates that $\Delta \theta / \theta$ (red) and $\Delta e / e$ (blue) are identical, which strongly supports that the Kerr signal can be regarded as a genuine magnetic signal even at the subpicosecond scale. Although the bleaching effect is suppressed in the Kerr signal, it is possible that an artifact of a small portion of the transient reflectivity can be included as a form of the peak in the ultrafast regime. In order to remove this, we have carefully nullified the offset in a balanced detector. To check the extent of the artifact in the Kerr signal, we have demonstrated the transient reflectivity and Kerr signals at 297 and 310 K, as shown in Fig. 1(c). Here, two reflectivity signals are almost same except the peak region as observed in a signal difference curve (red rectangle). In contrast, the Kerr rotation signal is sensitive to temperature. Comparing the signal level of the Kerr difference (green diamond) and the transient reflectivity difference at the peak region, we confirmed that the contribution of optical artifacts to the magneto-optical signal was controlled to less than the noise level.

In Fig. 2, we demonstrate the transient reflectivity (red circle) and the Kerr rotation signals (blue triangle) of TbFe alloy film. In the inset, the signals on a shorter time scale are presented. The major reason why a transient reflection signal is seen already at a few hundreds of femtoseconds before the pump pulse arrives is due to the actual broadening of two pulses in passing through transmission optics. As indicated by the characteristics of the transient reflectivity shown in the inset, the electrons excited by the pump pulse become internally thermalized less than 500 fs via electron-electron scattering. Thereafter, the electron and lattice systems thermally equilibrate within only a few picoseconds, which is noticed by the fact that the transient reflectivity decays exponentially after the peak and remains at a constant value. Subsequently, the energy absorbed in the lattice system is transferred to the spin system, which is thermalized in several tens of picoseconds, as seen in Fig. 2. This rather slow thermalization of the spin system might be ascribed to its amorphous structure with insufficient intermediate phonon channels.

The most intriguing observation in this figure is that a step-like change ($\Delta \theta_t$) in the Kerr signal arises in the middle of the thermalization process of excited electrons. Its slope is noticeably larger than the subsequent one, indicating the existence of a different demagnetization process on this subpicosecond time scale. In order to explain the step-like demagnetization that occurs before electron thermalization, it is necessary to assume the subpicosecond energy transfer channel. Therefore, we have adopted extended three-temperature heat rate equations considering a nonthermal electron system as follows: \textsuperscript{8}

\[
\frac{\partial \rho_{\text{nth}}}{\partial t} = -\alpha \rho_{\text{nth}} - \beta \rho_{\text{nth}} - \eta \rho_{\text{nth}} + P(t),
\]

\[
C_e(T_e) \frac{\partial T_e}{\partial t} = \nabla(\sigma \nabla T_e) - G_{el}(T_e - T_s) - G_{el}(T_e - T_s) + \alpha \rho_{\text{nth}},
\]

\[
C_l(T_l) \frac{\partial T_l}{\partial t} = -G_{el}(T_l - T_e) - G_{el}(T_l - T_s) + \beta \rho_{\text{nth}},
\]

\[
C_s(T_s) \frac{\partial T_s}{\partial t} = -G_{el}(T_s - T_e) - G_{el}(T_s - T_l) + \eta \rho_{\text{nth}}.
\]

Here, $\rho_{\text{nth}}$ denotes the energy density stored in the nonthermal electron system and $\alpha$, $\beta$, and $\eta$ represent the energy transfer rate to the electron, lattice, and spin system, respectively. $P$ is the pumping source that activates the heat rate equation. $G_{el}$, $G_{el}$, and $G_{el}$ are the free parameters implying the coupling strength between the systems. For simplicity, $G_{el}$ was assumed to be negligible, as the electron-spin relaxation process via spin-orbit coupling appears to have a small probability for $\Delta \theta_t$, judging from that its steep slope completes in the vicinity of the thermalization time scale of electrons.\textsuperscript{16} Moreover, TbFe thin film has such a low level of thermal conductivity, $\sigma = \sigma_0 |T_e|/T_s$, $\sigma_0 = 6 \text{ W m}^{-1} \text{K}^{-1}$, to affect the ultrafast dynamics,\textsuperscript{17} and the excess energy in irradiation area is already spread out homogeneously to whole film depth through a nondiffusive, ballistic transport of nonthermal electrons within the temporal width of a pump pulse.\textsuperscript{18} Therefore, the heat diffusion term was also neglected. For this simulation, we used the following values for the heat capacity parameters: $\gamma = 2.2 \times 10^2 \text{ J m}^{-3} \text{K}^{-2}$ in $C_e = \gamma T_e$, $C_l = 2.3 \times 10^6 \text{ J m}^{-3} \text{K}^{-1}$, and $C_s = 0.6 \times 10^6 \text{ J m}^{-3} \text{K}^{-1}$.\textsuperscript{19} The values of free parameters $G_{el}=6.6 \times 10^{17} \text{ W m}^{-3} \text{K}^{-1}$, $G_{el}=0.4 \times 10^{17} \text{ W m}^{-3} \text{K}^{-1}$, $\alpha = 6.9 \times 10^{-1} \text{ ps}^{-1}$, $\beta = 1.7 \text{ ps}^{-1}$, and $\eta = 1.6 \times 10^{-1} \text{ ps}^{-1}$ were determined.

In Fig. 3, we plot the calculated transient temperature profiles of the electron, lattice, and spin systems with time, together with the energy density profile of the nonthermalized electron system. As seen in Fig. 3, the simulation results are in good agreement with the experimental ones in two aspects. One is that the signal of the spin system lags behind the signal of the electron system by several hundreds of femtoseconds, as the energy transfer rate from the nonthermal electron to spin system is smaller than that to the electron system. The other is that the step-like change is well produced due to nonthermal electrons that last up to near the thermalization time of electrons.

From the interaction time scale of the nonthermal electron system with the spin system, we believe that Elliott-Yafet-type nonthermal electron spin-flip scattering seems to be the most probable microscopic origin for the ultrafast de-

![Fig. 2. (Color online) Transient reflectivity (red circle) and magneto-optical Kerr rotation signals (blue triangle) at 297 K with a pump-beam intensity of 3.3 mJ/cm². The inset shows the same signals on a shorter time scale.](image-url)
FIG. 3. (Color online) Evolution of the calculated electron (dashed orange), lattice (dotted magenta), and spin (solid blue) temperature together with the energy density change of the nonthermal electron system (dot-dashed green) adopting the extended three-temperature model.

The demagnetization observed in TbFe film. One might consider the direct transfer of the angular momentum from the light to the spin system as a possible origin for the steplike demagnetization. But it cannot account for the fact that the transient reflectivity is followed by a steplike change with a finite time delay. Besides, an electron-spin relaxation process via spin-orbit coupling is improbable for subpicosecond demagnetization since the steplike change is completed in the vicinity of the thermalization time scale of electrons.

In conclusion, we have investigated the ultrafast spin demagnetization dynamics of Tb$_{35}$Fe$_{65}$ thin film using a time-resolved all-optical pump-probe technique. Unusually, steplike demagnetization has been observed even at a subpicosecond scale. With an extended three-temperature model calculation, we have identified that this ultrafast behavior is attributed to an energy transfer from a nonthermal electron to a spin system. The direct channel opening between these two systems makes possible to improve spin demagnetization time scale. Therefore, TbFe alloy film is considered to be a desirable system to control the spin state in the subpicosecond regime.

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