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Energy relaxation dynamics in ferromagnetic Co film with femtosecond transient absorption

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ABSTRACT

Femtosecond time-resolved pump-probe technology has been widely used in studying ultrafast dynamics in various materials. In the present study a Co thin film on MgO, as a typical ferromagnetic material, was selected for investigating the ultrafast energy relaxation dynamics related to spin feature of materials. The energy relaxation dynamics of this thin film was measured by femtosecond transient absorption. The results can be well stimulated by a three-temperature model, in which electron, spin and lattice were involved, indicating that spin plays an important role in the energy relaxation. From this model, the hot electron temperature from the transient absorption under different pumping laser energy was calibrated which can reach 638, 724, 841 and 935 K with the energy of 3.2, 4.3, 6.0 and 7.5 J/m respectively. In additional, three time constants were obtained by a global analysis of the transient absorption, and attributed to electron–spin (250~350 fs), electron–lattice (10~20 ps), and spin–lattice interaction (300~400 ps), respectively. By analyzing the dynamics of the ferromagnetic material after laser excitation, the rich physics governing ultrafast energy relaxation dynamics are elucidated, and the temperature references for femtosecond laser manipulation of this material are provided.

1. Introduction

Understanding the microscopic processes of electron relaxation dynamics in ferromagnetic materials is one of the most challenging and interesting issues for the further development of advanced spin devices. The hot electron generated by a femtosecond optical pulse relaxes its energy to the spin and lattice due to electron–spin and electron–lattice interactions. During this process, the temperature exchange among the electron, spin and lattice reservoirs can be investigated and the characteristic times of the microscopic interactions which govern the basic metallic properties can be deduced. Using transient transmission and magneto-optical Kerr techniques, Beaurepaire et al. studied the relaxation dynamics of electron and spin systems in ferromagnetic Ni excited by femtosecond pulses and described the experimental results by using a three-temperature model involving energy exchange among electron, phonon and spin thermal reservoirs more than twenty years ago [1]. Since then, much attention has been focused on the ultrafast manipulation of spin states in magnetic materials by using this technique [2–7]. However, the microscopic processes respond to the spin state changes such as electron and spin dynamics of the ferromagnets have not been unambiguously identified yet. A complete understanding of such processes would be extremely helpful for further technical applications in data read-write processes.

Most of the experimental approaches applied in this field employed magneto-optic Kerr effect and used optical [4,8] or X-ray circular polarization dichroism [3,9,10] as the probe source. However, femtosecond transient absorption technique, as a simple experimental method for measuring the electron relaxation dynamics of materials [11–13], can be useful for exploring the dynamics of ferromagnetic materials with ultrafast temperature change induced by well-controlled laser pulses. The remain question is to map the ultrafast dynamics of the excitation energy transfers into electron, spin, and lattice after the initial optical pulse interacted with the electron system in the materials. The hot electron released the excess energy to the spin and lattice to reach the equilibrium condition. Therefore, it important to show the hot electron temperature and its equilibrium time using theoretical and experimental data. In fact, these energy transfers will lead to ultrafast change of the spin and lattice temperatures which induce efficiently the demagnetization dynamics of materials. In this work, we investigate

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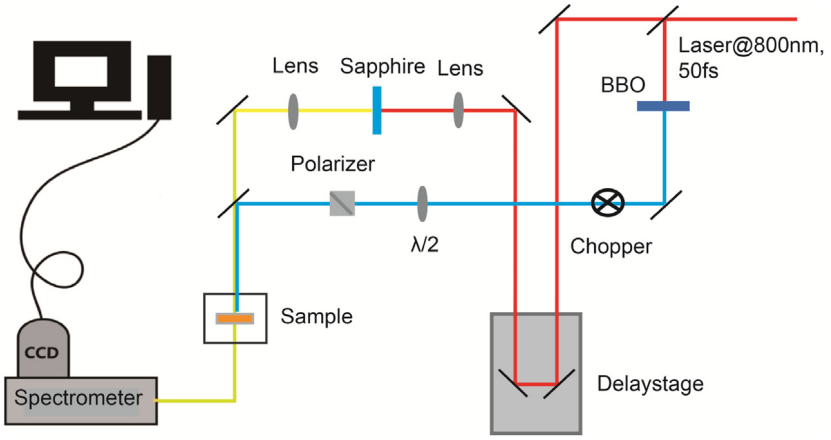

Fig. 1. Experimental setup for the femtosecond transient absorption measurement.

the energy relaxation dynamics after electron excitation in Co ferromagnetic films with femtosecond transient absorption scheme. We take Co thin film as the example since this ferromagnetic material shows great potential for various applications of magnetic devices due to its high Curie temperature and large saturation magnetization [14]. We observe the energy relaxation of thermal electrons through the interaction of electron–spin, electron–lattice and spin–lattice interaction. The time constants for the decays of electron–spin, electron–lattice, and spin–lattice are determined respectively from the measured evolutions of the electron temperatures under different laser pulse energies from comparison with the three-temperature model. The decay time of electron–spin decreases with increasing the pumping laser energy, the time of spin–lattice interaction increases with the increase of pumping laser energy with the analysis error bars.

2. Experimental details

The ferromagnetic Co sample with a 35 nm thickness on a 0.5 mm MgO substrate was deposited by a magnetron sputtering. The MgO used in our experiment is a transparent substrate, we do not observe any absorption signal of MgO substrate in the optical spectral range used in the experiment. The femtosecond transient absorption technique used in this work (Fig. 1) has been described in our previous study [15]. Experimentally, we used a Ti: sapphire laser system (Libra-USP-HE, Coherent Inc.) with a central wavelength of 800 nm, a repetition rate of 1 kHz, and a pulse duration of 50 fs. A 400 nm pump beam generated from a barium boron oxide crystal by the fundamental laser output was used to optically excite the electronic system in the sample. The pumping laser energies ranging from 3.2 to 7.5 J/m^2 irradiated onto the sample surface with a diameter of ~1.2 mm. It is well known that relaxation of hot electrons produced by the laser can drive the temperature changes of spin and lattice which are related directly to the observed decay of the transient absorption signals. To probe these relaxations, we used a white-light continuum in the range of 450 to 650 nm, generated by the fundamental laser pulse passing through a sapphire crystal. A delay stage was used to change the distance of light path of the probe beam and obtain the time delay. All the transient absorption measurements were performed at room temperature.

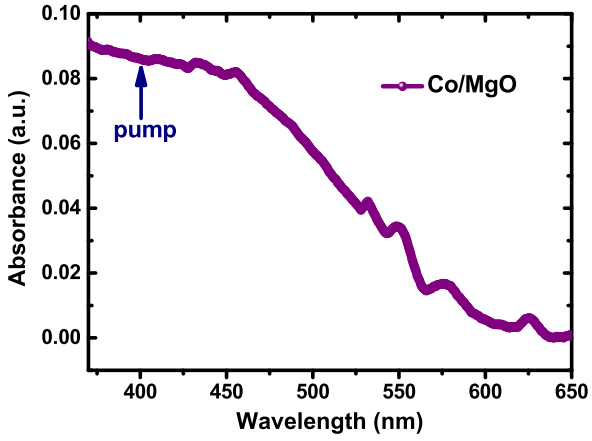

Fig. 2. Optical UV–Vis absorption spectrum of Co/MgO film.

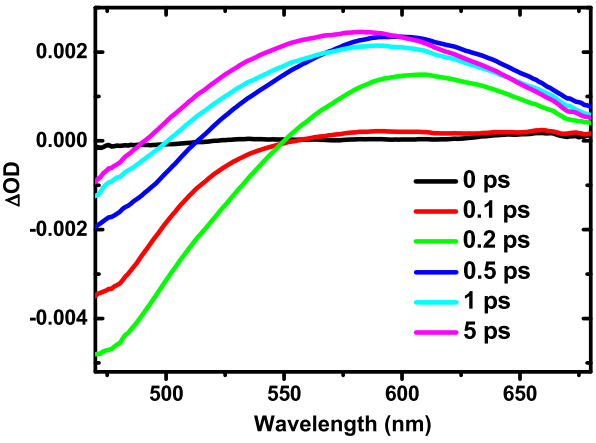

Fig. 3. Transient absorption spectra of Co/MgO at different time delay under the pumping laser energy of 6.0 J/m^2.

3. Results and discussion

The steady-state absorption spectroscopic study of Co on MgO substrate was done in the wavelength region (370~650 nm) as presented in Fig. 2. The absorption peaks observed around 549, 576 and 625 nm are explained as electronic transitions involving crystal-field split 3d levels in Co [16]. These transitions are correlated with the d–d transitions. Due to the strong absorption near 370~450 nm, we chose 400 nm for pumping. Fig. 3 shows the transient absorption spectra at different time delay with the pumping laser energy 6.0 J/m^2. As shown in the figure, the transient absorption is featured by a bleaching band centered at 460 nm and a broad absorption band around 590 nm.

For the absorption in the range below 490 nm, we try to simulate the spectra probed from the different time delay by using a two-temperature (electron and lattice) and a three-temperature (electron, spin and lattice) models. The evolutions of the electronic temperature

and the lattice temperature of Co calculated by the two-temperature model with time delay are shown in Fig. 4(a) under the irradiation of a 50 fs laser pulse of 6.0 J/m^2 onto the Co film, whereas the evolutions of the electron temperature, spin temperature and lattice temperature of Co with time delay under the same condition is shown in Fig. 4(b). In the latter the relaxation of the electron temperature is more rapid, compared with the two-temperature calculation, due to the electron–spin interaction involved, and the maximum electron temperature is also little lower in this case for the same reason. It is noted that the electron temperature begins to decay rapidly accompanied with the spin temperature rising and reaches the maximum value within 720 fs. Meanwhile, the lattice temperature rises slightly, and the variation of the spin temperature is more obvious than that of the lattice temperature. With the time delay after ten picoseconds, the electron temperature reaches a thermal equilibrium with the lattice temperature, which is more longer than that in the two-temperature description (about 2.5 ps, seeing in Fig. 4(a)).

We extracted the profiles of electron temperature evolution with time delay from the calculation results of the two-temperature model and the three-temperature model of Co, and compared them with a typical transient absorption dynamics obtained by a probe wavelength of 480 nm in a time range below 20 ps under pumping laser energy 6.0 J/m^2, as shown in Fig. 4(c), the scale of the electronic temperature in the transient absorption curve has been calibrated by its maximum and minimum values from the calculation by the three-temperature model. In the calculation of three-temperature model, we adopted three coupled differential equations to calculate the temporal evolution of the Co system [1,7,17–19]:

$$C_e(T_e)\frac{dT_e}{dt} = -G_{el}(T_e - T_l) - G_{es}(T_e - T_s) + P(t) \quad (1)$$

$$C_l(T_l)\frac{dT_l}{dt} = G_{el}(T_e - T_l) - G_{sl}(T_l - T_s) \quad (2)$$

$$C_s(T_s)\frac{dT_s}{dt} = G_{es}(T_e - T_s) - G_{sl}(T_s - T_l) \quad (3)$$

The heat propagation is negligible since the small film thickness. The laser source applies only to the electron term because the energy is initially deposited on the electron subsystem. Where T_e, T_s, and T_l represent the temperature of electron, spin and lattice systems, respectively. It is assumed that the electron, spin and lattice systems are ambient temperatures (T_0 = 300 K) before the film is heated by femtosecond laser pulse, i.e.

$$T_e(x,0) = T_s(x,0) = T_l(x,0) = T_0 \quad (4)$$

C_e, C_s, and C_l represent the specific heat of electron, spin and lattice systems, respectively. The specific heat of electron is proportional to the electron temperature, $C_e = \gamma_e T_e$, wherein γ_e = 704 J m^{-3} K^{-2} [20]; the specific heat of lattice was set to be a constant since its relatively small variation with the temperature $C_l = 3.5 \times 10^6$ J m^{-3} K^{-1} [17]; and the specific heat of spin is proportional to the spin temperature when the spin temperature is lower than that of the Curie temperature, i.e., $C_s = \gamma_s T_s$, where $\gamma_s = 2.0 \times 10^3$ J m^{-3} K^{-2} [20]. The Curie temperature of Co is reaches 1388 K [14], which is higher than our calculation value. Therefore, the assumption of the specific heat of spin is reasonable within the temperature range involved in our calculation. G_{es}, G_{sl}, and G_{el} are the free parameters describing the energy transfer rate of electron–spin, spin–lattice, and electron–lattice. The values of free parameters were $G_{el} = 6\times10^{17}$ W m^{-3} K^{-1}, $G_{es} = 8\times10^{17}$ W m^{-3} K^{-1} and $G_{sl} = 0.3\times10^{17}$ W m^{-3} K^{-1} [17,19,21]. P(t) is the laser heat source term. The heat source P(t) can be modeled with a Gaussian temporal profile [22,23]:

$$P(t) = \sqrt{\frac{\beta}{\pi}}\frac{1-R}{t_p\delta}J \cdot exp\left[-\frac{x}{\delta} - \beta\left(\frac{t-2t_p}{t_p}\right)^2\right] \quad (5)$$

where R is the reflection coefficient, δ is the penetration depth, t_p is the pulse width, J is the energy of the laser, β = 4ln2. In our simulation, the reflection coefficient R for a laser wavelength of 400 nm is 0.49, the penetration depth δ is 11.4 nm [20].

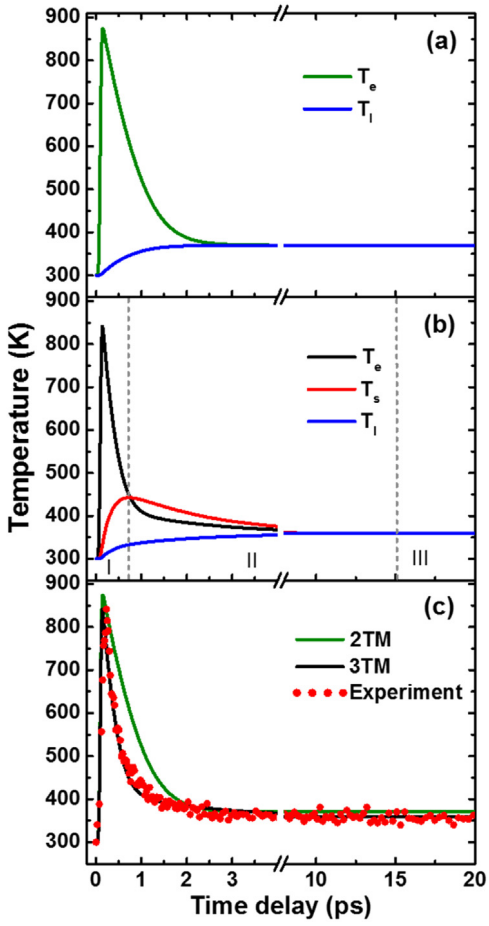

Fig. 4. The evolution of the electron, spin, and lattice temperature of Co with time delay calculated by the (a) two-temperature (2TM) and (b) three-temperature (3TM) models, (c) The comparison of the electron relaxation dynamics obtained by the two-temperature (green solid line), three-temperature (black solid line) and calibrated transient absorption dynamics obtained by a typical wavelength of 480 nm (red scatters), the pumping laser energy is 6.0 J/m^2, and the pulse width is 50 fs . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The simulation electron profile of the two-temperature model has a large deviation from the experimental measurement result in the range of several hundred femtoseconds to 2 ps (see Fig. 4(c)). The simulation result of the three-temperature model is in good agreement with the experimental ones, indicating that the spin system plays an important role in the electron relaxation process. As a ferromagnetic metal, Co has the degree of freedom (spin) in addition to two degrees of freedom (electron and lattice). Energy exchanges among electron, spin and lattice occur after the excited by femtosecond laser pulse. The hot electrons relax their energy through the interaction of electron–spin, electron–lattice and spin–lattice. Therefore, the three temperature model is more suitable to describe our experimental system. From the transient absorption curve, absorption intensity increases rapidly and reaches the maximum value at a time of 220 fs, then decays gradually till a constant value. It indicates that the electrons were excited by the femtosecond laser pulse rapidly and the whole system was heat up via electron–electron interaction in a time scale less than 220 fs. In the next step, the hot electrons relax their energy into the spin and/or lattice system due to electron-spin and/or electron–lattice interaction within a few picoseconds, which is noticed by the fact that the transient absorption signal varies exponentially after reaching the largest value and remains at a constant value.

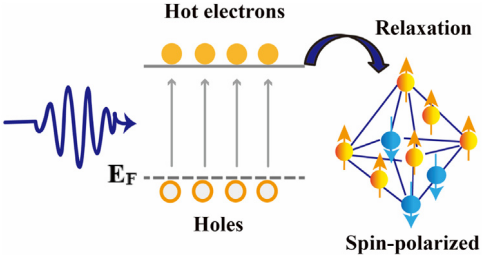

Fig. 5. Schematic of energy relaxation process of Co/MgO film.

To gain insight into the dynamic evolution of the Co film after laser excitation, a global fitting was performed to analyze the ultrafast energy relaxation process with sets of kinetic curves simultaneously. The energy relaxation process could be satisfied with three exponential fitting of lifetimes, i.e., $\tau_1 = 285 \pm 48$ fs, $\tau_2 = 15.31 \pm 1.82$ ps, and $\tau_3 = 354 \pm 30$ ps. The 285 fs time constant of the relaxation process is similar to the time scale of the electron–spin interaction observed in the previous reports [14,24], and thus can be assigned to the electron and spin relaxation process. Before, Cinchetti et al. obtained an ultrafast demagnetization time of Co through the magneto-optic Kerr effect is 300 fs [24], and Koopmans et al. obtained an ultrafast demagnetization time of 260 fs [14]. Furthermore, the second process (τ_2) should be derived from the electron–lattice interaction and the slower τ_3 characteristic time of a few hundred picoseconds should be assigned to the spin–lattice interaction, compared with the previous results [2,25,26]. Thus, to clarify energy transfers among electron, spin, and lattice in the Co film, we further illustrate the experimental data based on the three-temperature model. The spin system obtains the energy from the excited hot electrons in the time scale of 285 ± 48 fs, resulting in the increase of the spin temperature (the regime I), subsequently the energy from the electron thermal reservoir transfers into the lattice and reaches an equilibration with the time scale of 15.31 ± 1.82 ps through the electron–lattice interaction (the regime II). Thus, the three energy storage systems (electron, spin, and lattice) are in thermal equilibrium with each other and dissipate energy through the substrate (regime III). To gain further physical insight into the process of the energy relaxation of Co/MgO film clear, we give a possible energy relaxation path in Co/MgO, as shown in Fig. 5.

The influence of the electron temperature on energy transfers among electron, spin, and lattice can be discussed in Fig. 6, since the maximum electron temperature is increased with increasing the pumping laser energy irradiated onto the film. The electron temperature evolutions are calculated by the three-temperature model and show in Fig. 6 by the black solid lines under different excitation energies of 3.2 J/m^2, 4.3 J/m^2, 6.0 J/m^2 and 7.5 J/m^2, respectively. We compared these electron temperature evolutions with the typical transient absorption dynamics obtained by a probe wavelength of 480 nm under the same pumping laser energies, respectively. After the optical excitation, the energy of excited electrons delocalized in the electron reserve due to electron–electron interaction, leading to a fast increase of electron temperature which characterized by a peak in the early time of transient absorption. This electronic temperature has been calibrated by its maximum and minimum values from the calculation of the three-temperature model under the different pumping laser energy. As seen from the figure, the electronic temperature can reach 638 K, 724 K, 841 K and 935 K with the pumping laser energy of 3.2 J/m^2, 4.3 J/m^2, 6.0 J/m^2 and 7.5 J/m^2, respectively. In addition, from the global fittings, three characteristic times (as shown in Table 1) were obtained. We focus on the electron–spin interaction in energy transfers in the material after the initial optical excitation of the electron system. The electron peak temperature become larger with increasing energy of the pumping laser. It can be seen from Table 1 that the energy relaxation time of electron–spin decreases with the increase of the peak

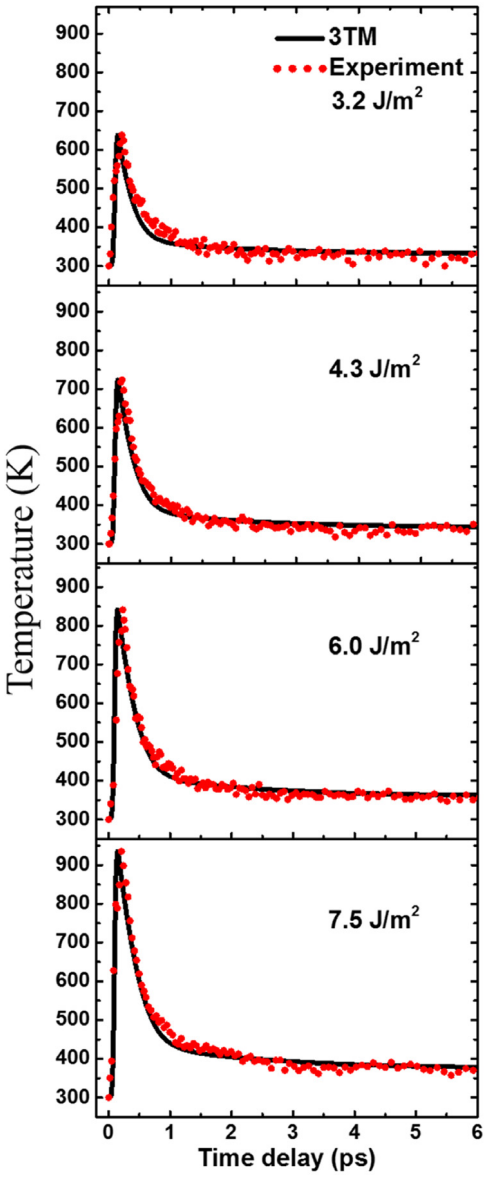

Fig. 6. The temporal evolution of electron temperature obtained from the three-temperature model (black solid line) and the transient absorption data obtained by a typical wavelength of 480 nm (red scatter) under various pumping laser energy of 3.2 J/m^2, 4.3 J/m^2, 6.0 J/m^2 and 7.5 J/m^2, respectively . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

electron temperature, which is caused by the fact that the increase of electron peak temperature makes the spin system obtain energy from the electron heat reservoir faster. After the electron–spin interaction, the electron, spin, and lattice systems trend to equilibrate toward a more moderate temperature. Through the electron–lattice and spin–lattice interactions, the electron temperature decreases further and finally the equilibration can be achieved reach, determined by the thermal diffusion of crystal lattice which in turn is with increased time when the electron peak temperature increasing.

4. Conclusions

In summary, the energy transfers among electron, spin, and lattice in Co film have been studied by femtosecond transient absorption technique. The evolution of electron temperature of Co film with time delay is calculated by a three-temperature model and compared with

Table 1
The summary of fitted transient absorption kinetic decay times of the Co ferromagnetic film by global analysis under different pumping laser energies.

Pumping laser energy (J/m^2)	Electron–spin interaction (fs)	Electron–lattice interaction (ps)	Spin–lattice interaction (ps)
3.2	341 ± 61	18.25 ± 6.04	310 ± 35
4.3	322 ± 27	14.46 ± 3.12	305 ± 45
6.0	285 ± 48	15.31 ± 1.82	354 ± 30
7.5	263 ± 47	20.79 ± 3.07	394 ± 66

the transient absorption curve measured at the same pumping laser energy. The results show that, the peak value of electronic temperature increased with the pumping laser energy increasing, and the corresponding peak values of electronic temperature were 638 K, 724 K, 841 K and 935 K with the pumping laser energy of 3.2, 4.3, 6.0 and 7.5 J/m^2, respectively. Three processes were obtained by global analyzing the femtosecond transient absorption dynamics data. They were assigned as electron–spin interaction, electron–lattice and spin–lattice interactions, respectively. Our results will provide a dynamic basis for the ultrafast control of the energy relaxation process of Co magnetic material.

Acknowledgments

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