Supplementary Material

**Femtosecond Laser-heating effect on the magnetization dynamics in perpendicularly magnetized Ta/CoFeB/MgO film**

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Pump fluence dependence of spin dynamics in PMA CoFeB film

Figure S1 shows the raw data of the magnetization dynamics of PMA CoFeB film under different pump fluences. By fitting the experimental data with the equation of damped oscillations, we obtain two important parameters, i.e., precession frequency $f$ and relaxation time $\tau$. According to the equations of $f = (g/2\pi)\sqrt{H_1H_2}$ and $\alpha_0 = 1/(2\pi f \tau)$, the best-fitting parameter $H_K$ and $\alpha_0$ are obtained as a function of pump fluences, which are shown in Fig.4 of the main text.

Fitting details of the demagnetization curves

The demagnetization process as shown in Fig. 4(a) of the main text can be physically described in terms of energy distribution among electrons, phonons and spins after laser pulse excitation using the phenomenological three-temperature mode (3TM)\(^1\). The temporal Kerr signals were fitted by the analytic solution of 3TM as following,

$$ \Delta \theta = \frac{A_1}{\sqrt{t/(\tau_0 + 1)}} - \frac{(A_2 \tau_E - A_1 \tau_M)e^{-t/\tau_E}}{\tau_E - \tau_M} - \frac{\tau_E(A_1 - A_2)e^{-t/\tau_E}}{\tau_E - \tau_M} \theta(t) \ast G(t) \quad \text{(S1)} $$

where $G(t)$ is the Gaussian pulse laser, $\theta(t)$ is the step function and $\ast$ represents convolution product. The constant $A_1$ represents the magnetization at thermal equilibrium and $A_2$ is proportional to the initial electron temperature rise. The parameters $\tau_M$ and $\tau_E$ describe the time scale of magnetization loss and electron-phonon interaction, respectively. The first term in the right side represents the heat diffusion process. According to the measured transient reflectivity spectra $\Delta R/R$ as shown in Fig. S2, the amplitude of $\Delta R/R$ increases slowly in the timescale of $\sim 600\,\text{ps}$ without any decreasing trend. Based on this observation, we ignored the heat diffusion term in Eq. (S1). The pulse duration of the Gaussian pulse laser was treated as a shared global parameter with a value of $140 \pm 8\,\text{fs}$.

Performing two-temperature model on transient reflectivity $\Delta R/R$

Figure S2 (a) and (b) show the results of transient reflectivity $\Delta R/R$ of the PMA CoFeB film in short and long time-delay windows, respectively. The pump fluence is fixed at $F = 5\,\text{mJ/cm}^2$. In Figure S2 (a), the $\Delta R/R$ reaches a minimum at 0.2 ps after photo-excitation. This delayed minimum suggests that the $\Delta R/R$ is mainly sensitive to thermalized electrons, i.e. $\Delta R/R \propto \Delta T_e^2$. Thus, we can ignore the initial thermalization process of the non-thermal electrons and the energy is transferred between the electron and lattice systems via electron-phonon interaction, equilibrating the temperatures of both systems. In addition, as shown in Figure S2 (b), a slow increase rather than a decrease is seen in the $\Delta R/R$ curve and we can thus conclude that the heat diffusion process can also be ignored. The data shown in Figure S2 (a) was reproduced according to the following phenomenological model\(^3\):

$$ \Delta R/R = \{A e^{-t/\tau_{ep}} + B (1 - e^{-t/\tau_{ep}}) \} \quad \text{(S2)} $$

where $A$ and $B$ represent the electronic and lattice response, respectively. $\tau_{ep}$ is the electron-phonon relaxation time. By convolution with the Gaussian pulse laser of $140 \pm 8\,\text{fs}$, the experimental data in Figure S2 (a) was well reproduced.

Having obtained the fitted value of $\tau_{ep}$ using Eq. (S2), we can further evaluate the electron-lattice interaction constant $g_{el}$, an important parameter in the two-temperature (2T) model, via relationship\(^4\) $\tau_{ep} = C_t C_e/ g_{el} (C_t + C_e)$. The 2T model contains two differential equations as following\(^5\):

$$ C_e \frac{dT_e}{dt} = -g_{el}(T_e - T_l) + P(t) $$
Here $C_l$ and $C_e$ are the lattice and electron heat capacities, respectively. $P(t)$ describes the absorbed power density in the material determined by the Gaussian pulse laser. This term is proportional to the pump fluence. $T_e$ and $T_l$ are the electron and lattice temperature, respectively. Since the properties of Co and Fe are similar to each other, the $C_l$ and $C_e$ constants involved in the fitting procedures are replaced by the corresponding values of Co for clarity. The lattice specific heat at low pump fluence is $C_l = 25.14 J/mol \cdot K$ and the specific heat of electrons is assumed to be $C_e = \gamma T_e$ with $\gamma = 4.9 mJ/mol \cdot K^2$. The assigned values of $C_l$ and $C_e$ are from published papers. The parameter of $g_{ep}$ is obtained by fitting the $\Delta R/R$ data with Eq. (S2) as discussed above. In the practical measurements, it is difficult to exactly evaluate the absorbed heat via the optical constants due to the existence of multiple layers. However, according to the three-temperature (3T) model, the spin temperature $T_s$ as well as $T_e$ and $T_l$ reaches an equilibrium in a few picoseconds after photo-excitation. As a consequence, we tune the parameter in $P(t)$ to scale the equilibrated temperature between $T_e$ and $T_l$ to the value of $T_s$ at $\Delta t = 5 ps$. The experimental values of $T_e(t)$ are determined by the static temperature dependence of magnetization found in text books. The temperature dynamics of $T_e$ and $T_l$ at $F = 5 mJ/cm^2$ and $F = 12 mJ/cm^2$ calculated by Eq. (S3) are respectively shown in Figure S3 (a) and (b). The highest value of $T_e$ lies in the range between 1651 and 2656 K.

![Figure S1](image)

Figure S1. Time-resolved Kerr traces of CoFeB film under different pump fluence. The external magnetic field is applied along $47.5^\circ$ away from the sample normal with a constant value of 9470 Oe. The red solid lines represent the fitting results according to the equation in the main text.
Figure S2 (a) Transient reflectivity of CoFeB film with perpendicular magnetic anisotropy within a short-time window of 5 ps. The red solid line is the fitting result according to Eq. (S2). (b) The same as (a) but in a longer timescale of ~ 600 ps. The pump fluence is $F = 5 \text{ mJ/cm}^2$.

Figure S3 Temporary evolution of $T_e$ and $T_l$ at $F = 5 \text{ mJ/cm}^2$ (a) and $F = 12 \text{ mJ/cm}^2$ (b). The values of $g_{el}$ at different $F$ is different, which are determined by the fitted values of the parameter $\tau_{ep}$.

Figure S4 Magnetic hysteresis loops of the CoFeB thin film measured by TR-MOKE method. The loops were measured at a time-delay of $\Delta t = -6 \text{ ps}$ with (red) and without (black) the excitation of pump pulses.

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