Ultrafast Laser Modulation of Local Magnetization Orientation in Perpendicularly Exchange-Coupled Bilayer

Supplementary Information: Simulation methods

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In the simulation, we model the equilibrium magnetic state by using the Landau-Lifshitz-Gilbert (LLG) equation with Langevin dynamics, given by [S1, S2]:

$$\frac{d\boldsymbol{S}_i}{dt} = -\frac{\gamma}{(1+\lambda^2)\mu_s}[\boldsymbol{S}_i \times (\boldsymbol{H}_i + \lambda \boldsymbol{S}_i \times \boldsymbol{H}_i)], \tag{A1}$$

where $\boldsymbol{S}_i$ is an unit vector denoting the direction of the spin at site i, λ and γ are the microscopic thermal bath coupling parameter and the gyromagnetic ratio, respectively, and μ_s is the atomistic magnetic moment. The effective field acting on spin at site i can be expressed as $\boldsymbol{H}_i = -\frac{\partial \mathcal{H}}{\partial \boldsymbol{S}_i} + \zeta_i(t)$. The effect of thermal fluctuations on the magnetic dynamics can be defined by the stochastic fields $\zeta_i(t)$ through fluctuation dissipation theory as $<\zeta_i^k(t)> = 0$ and $<\zeta_i^k(t)\zeta_j^l(t')> = 2\,\delta_{i,j}\delta_{k,l}\delta(t-t')\lambda k_B T \mu_s/\gamma$, where i and j represent the lattice sites, k and l denote the Cartesian components. In the simulations, we choose the typical parameters for GdFeCo as the exchange constant $J = 2.835 \times 10^{-21}$ J per link, the anisotropy constant $A = 3.130 \times 10^{-21}$ J per spin, the gyromagnetic ratio $\gamma = 1.76 \times 10^{11}$ $T^{-1}S^{-1}$, the magnetic moment $\mu_s = 1.92\mu_B$ (μ_B is the Bohr magneton) , and $\lambda = 0.05$ [S1, S2].

To understand the laser pulse induced ultrafast magnetization dynamics, we simulate the thermodynamic behavior of macrospins by using the Landau–Lifshitz–Bloch (LLB) equation [S3-S6]:

$$\frac{d\boldsymbol{m}_i}{dt} = -\tilde{\gamma}\left(\boldsymbol{m}_i \times \boldsymbol{H}_i^{eff}\right) + \frac{\tilde{\gamma}\alpha_{\parallel}}{m_i^2}\left[\boldsymbol{m}_i \cdot \left(\boldsymbol{H}_i^{eff} + \zeta_{i,\parallel}\right)\right]\boldsymbol{m}_i$$
$$-\frac{\tilde{\gamma}\alpha_{\perp}}{m_i^2}\left\{\boldsymbol{m}_i \times \left[\boldsymbol{m}_i \times \left(\boldsymbol{H}_i^{eff} + \zeta_{i,\perp}\right)\right]\right\}, \tag{A2}$$

where $\tilde{\gamma} = \frac{\gamma}{(1+\lambda^2)}$, $\boldsymbol{H}_i^{eff} = -\frac{\partial \mathcal{H}}{M_S^0 \partial \boldsymbol{m}_i} + \boldsymbol{H}_J$ denotes the effective field acting on spin at site i, with M_S^0 the value of spontaneous magnetization at zero temperature. $\zeta_{i,\parallel}$ and $\zeta_{i,\perp}$ are the longitudinal and transverse stochastic fields, respectively. In the simulations, we consider that the magnetization evolves under temperature $T < T_c$, with the temperature-dependent longitudinal and transverse damping parameters $\alpha_{\parallel}$ and $\alpha_{\perp}$ written as

$$\alpha_{\perp} = \lambda\left(1 - \frac{T}{T_C}\right), \quad \alpha_{\parallel} = \lambda\frac{2T}{3T_C}. \tag{A3}$$

The internal exchange field $\boldsymbol{H}_J$ controlling the length of the magnetization, which is defined as

$$\boldsymbol{H}_J = \frac{1}{2\tilde{\chi}_{\parallel}}\left(1 - \frac{m_i^2}{m_e^2}\right)\boldsymbol{m}_i, \tag{A4}$$

where the zero-field-reduced equilibrium magnetization m_e, and the equilibrium longitudinal susceptibility $\tilde{\chi}_{\parallel} = \partial m/\partial H_{\parallel}$ are calculated from the fluctuations of magnetization using the Langevin

LLG stochastic equation (equation (A1)), with $\tilde{\chi}_{\parallel} = \frac{\mu_s N}{k_B T}\left(< S_{\parallel}^2 > - < S_{\parallel} >^2\right)$, and N the number of spins.

In the simulations, the atomistic spins are coupled to the temperature of the electronic spins, namely $T = T_s(t)$, with the temporal evolution function $T_s(t)$ which can be determined using the three-temperature model as [S7-S9] :

$$C_e(T_e)\frac{\mathrm{d}T_e}{\mathrm{d}t} = -G_{el}(T_e - T_l) - G_{es}(T_e - T_s) + P(t),$$
$$C_s(T_s)\frac{\mathrm{d}T_s}{\mathrm{d}t} = -G_{es}(T_s - T_e) - G_{sl}(T_s - T_l),$$
$$C_l(T_l)\frac{\mathrm{d}T_l}{\mathrm{d}t} = -G_{el}(T_l - T_e) - G_{sl}(T_l - T_s) - G_l(T_l - T_0), \tag{A5}$$

where C_e, C_s and C_l are the specific heats of electron, spin and lattice, and the parameters G_{el}, G_{es}, G_{sl} are the electron–lattice, electron–spin and spin–lattice coupling constants respectively, and G_l is the thermal dissipation factor of the lattice in the environmental temperature ($T_0 = 300$ K). $P(t)$ is the laser power density absorbed in the material with approximate Gaussian form. In our simulations, the typical parameters for the coupling constants and specific heats are taken similar to that used in references [S7-S9].

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