Resolving the genuine laser-induced ultrafast dynamics of exchange interaction in ferromagnet/antiferromagnet bilayers

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The response of exchange coupled ferromagnet/antiferromagnet metallic bilayers to laser excitation down to the femtosecond time scale is investigated. Differently from previous attempts, the ultrafast dynamics of exchange interaction is deduced by carefully analyzing precessional transients of the ferromagnetic magnetization, measured by means of time-resolved magneto-optical Kerr effect, rather than by monitoring changes in the hysteresis loop as a function of pump-probe delay. Thereby we are able to estimate the characteristic time scale of laser-induced exchange-bias quenching in a polycrystalline Co/IrMn bilayer, $\tau_{EB,0}=(0.7\pm0.5)$ ps. The fast decrease in exchange coupling upon laser heating is attributed to a spin disorder at the interface created by laser heating.

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The challenge of manipulating the magnetic properties of metallic thin films and multilayers on the subpicosecond time scale by means of femtosecond laser pulses has attracted much attention in the past decade.[1–11] This interest burst after the discovery, in 1996, that irradiation by a short laser pulse can partially quench the magnetization of a nickel thin film within a few hundred femtoseconds.[1] This observation and its confirmation[1–4,6,12] led to the discovery of a number of exciting new physical phenomena, such as laser-induced launching of magnetization precession and spin waves,[5] ultrafast buildup of magnetic moment in FeRh,[7,8] and all-optical switching in GdFeCo by circularly polarized light.[9–11]

A particularly exciting new challenge in the field is to control the spin dynamics by modifying the exchange interaction between coupled (ferro)magnetic entities, rather than the magnetic state of a single element itself. Pioneering work in this direction was performed by Ju *et al.*[13–15] in the late 1990s: the authors aimed at manipulating the magnetization of a ferromagnet (FM) exchange coupled to an antiferromagnet (AFM) by optically modifying the interlayer interaction, also known as exchange bias (EB). Exchange bias arises in FM/AFM bilayers and leads to a unidirectional anisotropy that pins the ferromagnetic spins in a certain direction.[16,17] As a consequence the hysteresis loop of such bilayers is shifted along the field axis by a quantity $H_{EB,st}$ called exchange-bias field. Ju's experiments showed that the exchange interaction J_{EB} can be perturbed by femtosecond laser heating, leading to changes in the hysteresis loop of NiFe/NiO bilayers within a picosecond and, in some cases, to a precession of the FM spins. Recently, also Seu and Reilly[18] demonstrated that the exchange-bias interaction can be influenced by ultrafast laser pulses. Studies, based on monitoring the time evolution of hysteresis loops after laser excitation, confirmed this result but never overcame the difficulty of assigning a precise time scale to the ultrafast dynamics of EB,[19,20] leaving important questions unanswered. The intrinsic limitation in this approach is the fact that *after laser excitation the direct equilibrium relation between the magnetization and the exchange-bias field is lost*. Rather, the magnetization $M[t]$ will react to a time-dependent exchange-bias field $H_{EB}[t]$ in a delayed fashion, as described by the Landau-Lifshitz-Gilbert (LLG) equation. Therefore, it is not at all trivial to get time-resolved information about the interlayer coupling since only $M[t]$, and not $H_{EB}[t]$, can be measured optically. More specifically, time-resolved measurements of hysteresis loops can lead to results which are inconclusive and difficult to interpret. For example, let us consider the hypothetical experiment depicted in Fig. 1(a), based on the results reported in Refs. 19 and 20. After laser excitation, the hysteresis loop of an exchange coupled bilayer shifts within a picosecond along the field axis, corresponding to a quenching of the exchange-bias field from $H_{EB,0}$ to $H_{EB,1}$, as indicated by the horizontal arrow. Points 1 and 2 in the figure show that the magnetization should have switched its orientation. However, such a fast subpicosecond complete switching has never been observed in time domain studies, and it is physically very unlikely since the magnetization reacts to changes in the effective field by precessing around the new equilibrium position with a frequency on the order of a few gigahertz.

In this paper we show that it is possible to get around the

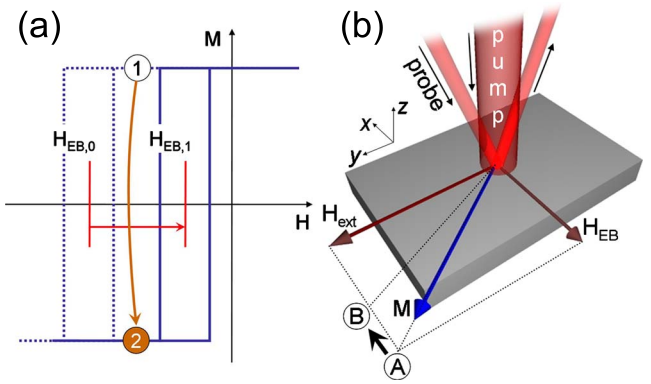

FIG. 1. (Color online) (a) A rigid shift of the hysteresis loop would cause the magnetization to instantaneously switch from position 1 to position 2: this situation is physically not very likely. (b) Sketch of the experimental configuration, as described in the text.

intrinsic limitation described above, and we resolve the genuine subpicosecond dynamics of exchange interaction after laser heating. The key element in our approach is to deduce the EB dynamics quantitatively from the magnetization precession, rather than by studying the temporal evolution of the hysteresis loops, like it was done in the past. This way we are able to address a number of outstanding issues: what is the time scale at which J_{EB} is perturbed?; does the quenching of the exchange interaction lead to a quenching of the exchange-bias field (scenario I) or to its realignment toward the direction of the FM magnetization (scenario II)?; are we observing the effect of a uniform lattice heating or does the presence of nonequilibrium (hot) electrons in the first picosecond play a role?

We chose for our study polycrystalline bilayers consisting of ferromagnetic Co and antiferromagnetic IrMn. The relative thickness of the layers was varied by growing wedges, such as those described in Ref. 21. While here we mainly report on one particular bilayer consisting of Co (10 nm) and IrMn (15 nm), the results are generally valid for a whole thicknesses range. The bilayers are sputtered on top of Cu (10 nm) and capped with Ta (3 nm) to prevent oxidation. The whole stack is deposited on a silicon substrate with a Ta (5 nm) buffer layer. The experiments are carried out using $\sim$100 fs pulses from a Ti:Sa oscillator with a pump fluence of $\sim$2 mJ/cm^2 and a pump-probe power ratio of 20:1. The probe pulses impinge on the sample at almost perpendicular incidence, giving rise to a Kerr signal $\Delta\theta$ which is predominantly polar, with a small longitudinal component along the y direction.[22]The experimental geometry is sketched in Fig. 1(b): the sample lies in the x-y plane, the exchange-bias field H_{EB}=4.7 kA/m acts along the negative x direction, and an external field H_{ext} is applied in the sample plane along the y axis. The effective field H_{eff} acting on the FM magnetization is given by the vectorial sum of H_{EB} and H_{ext}. When the laser hits the sample the exchange interaction is perturbed, leading to a change in the orientation and magnitude of the effective field (from point A to point B in the figure) and therefore a torque acts on the magnetization and a precession is triggered. Since the initial displacement of H_{eff} is, independently of scenario I or II, in the x-y plane, the initial torque tends to pull the magnetization out of the plane, along the z axis. The subsequent precession is determined at each time by the value and orientation of the effective field acting on the magnetization at that particular time, and, in turn, these depend on the value of the exchange-bias field. The idea is to retrieve the *field pulse* $\Delta H_{EB}[t]$ by carefully analyzing the precessional transients; this way we can resolve the dynamics of the exchange interaction without having to go through a measurement of hysteresis loops (and thus avoiding any interpretation problem).

In our analysis we will first focus on the field dependence of the precessional phase and amplitude, and then on retrieving the exact shape of the field pulse. We start by considering a precessional transient obtained for H_{ext}=16 kA/m, presented in Fig. 2(a). On the long time scale ($t>40$ ps), the data can be fitted by the function,

$$\Delta\theta = A_0 + A_1 e^{-t/\tau_{LLG}} \sin(2\pi f t + \Phi). \quad (1)$$

The fit yields the values of the precessional frequency f, amplitude A_1, and phase Φ; from the time constant τ_{LLG} the Gilbert damping parameter α can be calculated.

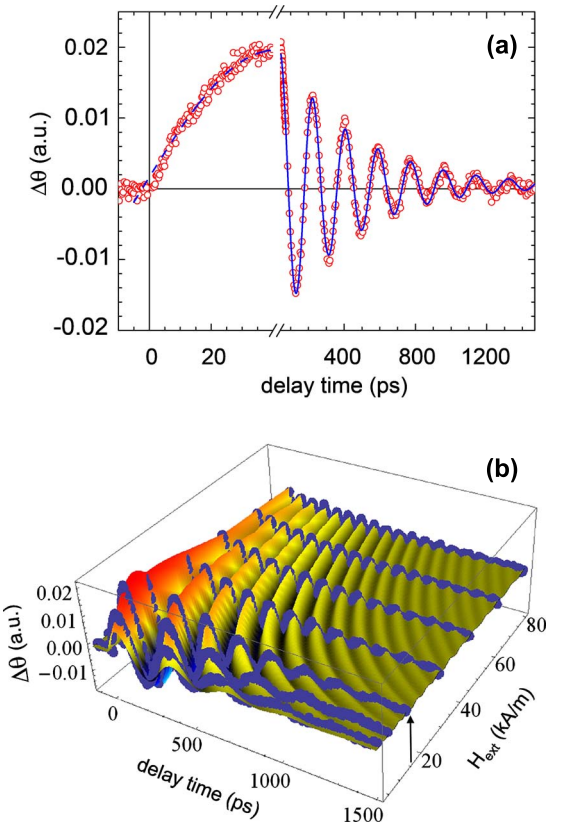

FIG. 2. (Color online) (a) Precessional transient obtained at H_{ext}=16 kA/m, the changing slope during the first 5 ps is highlighted by the dashed line, the solid line is a fit according to Eq. (1). (b) Precessional transients measured for different applied fields (dots) interpolated by simulated transients according to the LLG equation (surface). In the simulations the experimental geometry is taken into account and a field pulse according to Eq. (3) is given as an input to trigger the oscillations. The value of the damping parameter is taken from the experimental transients. The arrow highlights the data set in (a).

Beside the familiar damped oscillations, the data contain some peculiar features on the short time scale ($t<40$ ps) that provide a first qualitative glimpse on the temporal profile of $H_{EB}[t]$. In particular, the measurement shows a higher derivative during the first few picoseconds; this indicates that there is a fast buildup of torque during the first picoseconds after laser excitation, that quickly relaxes to a smaller torque. This could suggest that the exchange-bias field is rapidly quenched to a minimum and then relaxes to an intermediate value within a few picoseconds before finally decaying to its original value as excess heat is dissipated. Similar precessional transients have been measured for different values of the applied field, ranging from 4 to 80 kA/m (corresponding to starting with H_{eff} almost aligned with H_{EB} and H_{ext} respectively); these are shown in Fig. 2(b), interpolated by simulated transients according to the LLG equation. All the measured data show similar anomalies at short time delays, while the oscillatory part can be fitted with Eq. (1), with proper scaling of the parameters.

In Fig. 3 the precessional (a) amplitude A_1 and (b) phase

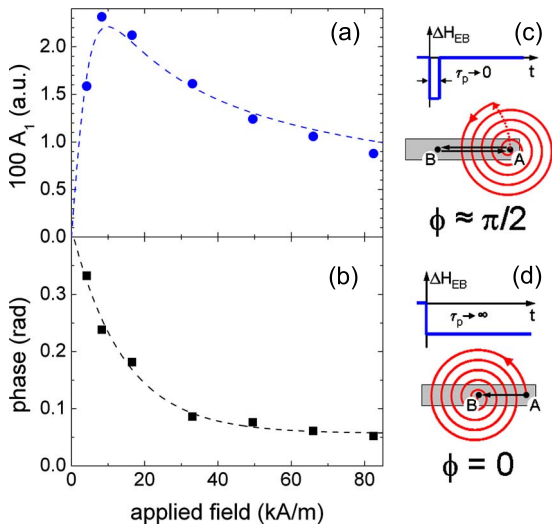

FIG. 3. (Color online) (a) Precessional amplitude vs external field; the dashed line is a fit according to the model described in Ref. 21. (b) Precessional phase as a function of the external applied field; the dashed line is a guide to the eye. (c) Sketch of the response of the bilayer to a rectangle functionlike and (d) a Heaviside functionlike field pulse, as described in the text.

Φ are plotted against the external field. The amplitude starts from zero, goes through a maximum at $H_{\mathrm{ext}} \sim H_{\mathrm{EB}}$ and then slowly decays. The trend can be fitted with a simple model based on scenario I [dashed line in Fig. 3(a)], described elsewhere.[21] The model is based on the observation that in the case of a laser-induced quenching of H_{EB} one would indeed expect that for $H_{\mathrm{ext}}=0$ and $H_{\mathrm{ext}}=\infty$ only the magnitude of the effective field is altered, but not its direction, and thereby no precession is triggered; in between, a precession is always observed and hence the amplitude must go through a maximum.

As it can be seen in Fig. 3(b), the precessional phase decreases with increasing applied field. In order to understand this trend, let us consider the cartoons in Figs. 3(c) and 3(d) where we sketched the response of the bilayer to (c) a rectangle functionlike and (d) a Heaviside functionlike field pulse, corresponding to a recovery time of the field pulse $\tau_p \to 0$ and $\tau_p \to \infty$, respectively. In the drawings we see a cross section of the sample, points A and B represent the tip of the effective field vector [and correspond to the directions A and B of Fig. 1(b) respectively], and the spiral follows the trajectory of the tip of the magnetization vector. In the case of a rectangle functionlike pulse, the tip of the effective field instantaneously goes from A to B and the magnetization feels a sudden torque that will pull it out of the sample plane along the dashed arrow. After a short time τ_p, as the field pulse goes back to zero, the magnetization will start to precess around A. Therefore the phase of the precession will be $\sim \pi/2$. In the case of a Heaviside functionlike pulse the effective field will rotate from A to B and the magnetization will precess around B with phase 0. The real field pulse will have finite decay and recovery times, and thereby we can expect a phase up to 90°.[24] More interestingly we can expect the phase to vary with the external applied field. As it can be seen in Fig. 2(b), the period of the precession τ_H decreases with the external field. For high fields, such that $\tau_H \ll \tau_p$, the field pulse approximates a step function ($\tau_p=\infty$) during the first oscillations; lowering the field so that $\tau_H \gg \tau_p$, a response corresponding to a field pulse close to a rectangle function with $\tau_p \approx 0$ can be expected; therefore a decrease in the phase with increasing applied field should be observed, as indeed confirmed in Fig. 3(b).

After having shown how the features of the magnetization precession can provide important qualitative information on the temporal evolution of the exchange-bias field after laser excitation, we can now proceed with a quantitative analysis, backcalculating the field pulse $\Delta H_{\mathrm{EB}}[t]$ from the precessional transients. Following the procedure described in Ref. 25, we write the LLG equation in the approximation of a small perturbation and invert it, expressing $\Delta H_{\mathrm{EB}}[t]$ as a function of $M_z[t]$, its derivative $M_z'[t]$ and its integral $\int_{-\infty}^{t} M_z[\xi]d\xi$,

$$\Delta H_{\mathrm{EB}}[t] = \frac{H_{\mathrm{eff}}}{M_s H_{\mathrm{ext}}}\left(-\alpha(M_s + 2H_{\mathrm{eff}})M_z[t] - \frac{\alpha^2+1}{\gamma}M_z'[t] - \gamma H_{\mathrm{eff}}(M_s + H_{\mathrm{eff}})\int_{-\infty}^{t} M_z[\xi]d\xi \right), \tag{2}$$

where M_s is the saturation magnetization of Co, γ is the gyromagnetic ratio, and $M_z[t]$ can be easily extracted from $\Delta\theta[t]$.[23]

The precessional transients $\Delta H_{\mathrm{EB}}[t]$ retrieved with this method are dominated by very high noise due to the presence of the derivative $M_z'[t]$ in Eq. (2). While achieving a better signal-to-noise ratio in the original data set (and thereby less scatter in the $M_z'[t]$ transient) is, in principle, possible by increasing the power of the pump pulse, in practice there is an intrinsic limit due to the fact that permanent changes in the magnetic properties of the sample were observed to take place at higher fluences. Therefore we consider our data set a "best in class" example of what is currently achievable, and we will show in the following paragraphs that it is still possible to extract useful information from the data.

In fact, careful analysis of the retrieved $\Delta H_{\mathrm{EB}}[t]$ transients reveals that all the pulses corresponding to different external field values display common features. In order to visualize this, we present a smoothed version of the retrieved field pulses, obtained by performing an adjacent average procedure over a 7 ps interval. This procedure is only meant to obtain a visual representation of the data trend and does not affect the rigorous mathematical analysis reported below; we will use the *smoothed* data in the figures and the *nonsmoothed* data for the calculations.

The *smoothed* data are shown in Fig. 4(a) (symbols), up to 25 ps. Interestingly, all the pulses corresponding to different external field values display the same qualitative shape: they start to deviate from zero at $t \sim -4$ ps, reach a minimum at $t \sim 4$ ps and then grow back stabilizing on an intermediate value at about 13 ps. This observation leads to the important conclusion that laser heating induces a quenching of exchange bias (scenario I), and does not alter its direction, since in the latter case the retrieved pulse would vary with the applied field. The different transients can then be averaged yielding the solid line in the figure.

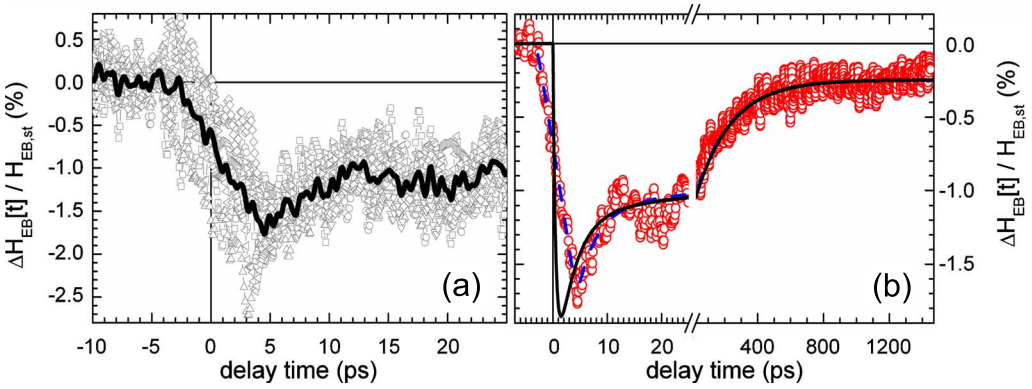

FIG. 4. (Color online) (a) Temporal profiles reconstructed using Eq. (2) for different applied fields (different symbols) and their average (solid line). (b) Average of the reconstructed pulses on the short and long time scales (symbols); the dashed lines are fits according to Eq. (3) and the solid line is the final field pulse after deconvolution as described in the text.

The very same average trace is reproduced on the longer time scale in Fig. 4(b) (symbols). In order to estimate the time scales of EB dynamics, we introduce the (phenomenological) function,

$$\Delta H_{\mathrm{EB}}[t] = \Theta[t] a_0 (1 - e^{-t/\tau_{\mathrm{EB},0}})(b_0 + b_1 e^{-t/\tau_{\mathrm{EB},1}} + b_2 e^{-t/\tau_{\mathrm{EB},2}}), \tag{3}$$

where $\Theta[t]$ is the Heaviside function. We then use Eq. (3) to fit the *nonsmoothed* version of the transient in Fig. 4(b). By fixing the parameters $\tau_{\mathrm{EB},1-2}$ and b_{0-2}, we fit the data up to their minimum, obtaining the characteristic decay time of exchange interaction $\tau_{\mathrm{EB},0}=(0.7\pm0.5)$ ps. Similarly, by fixing a_0 and $\tau_{\mathrm{EB},0}$, we fit the rest of the data and obtain the recovery times $\tau_{\mathrm{EB},1}=(3.4\pm0.3)$ ps and $\tau_{\mathrm{EB},2}=(215\pm4)$ ps. By plugging the results of the fit into Eq. (3), we can thus reconstruct the genuine response of H_{EB} to laser excitation, as shown in Fig. 4(b), solid line. In order to show the soundness of the fit, we simulate the effect of 7 ps adjacent averaging on the final fitting function, thus obtaining the dashed line in Fig. 4(b), that overlaps well the *smoothed* data set. Alternatively, one can also simulate the effect of 7 ps adjacent averaging on Eq. (3) and use the resulting function to fit the *smoothed* data set, obtaining exactly the same results.

Independently of the fitting and visualization strategy, the large scattering resulting from the backcalculation procedure is reflected in the large error on $\tau_{\mathrm{EB},0}$. However, we emphasize once again that this is a compromise between large signal and reproducibility of the measurement (i.e., absence of permanent effects). Moreover, we notice that the absolute value of ΔH_{EB} does not exceed 0.1 kA/m, a further indication of the high resolution of our measurements! We envision that future experiments may use the methodology proposed in this work to investigate different materials where higher laser fluences could be used, obtaining a higher signal-to-noise ratio.

After having assigned a time scale to the laser-induced quenching of EB, and having demonstrated that it is a genuine subpicosecond process, the remaining issue is to understand the microscopic origin of $\tau_{\mathrm{EB},0}$. We conjecture that the quenching of exchange interaction is caused by laser-induced disordering of the spins at the FM/AFM interface. More specifically, since the Curie temperature of Co is about 1400 K,[26] while the Neel temperature of IrMn is 690 K,[17] it is very likely that it is a loss of spin ordering in the antiferromagnet that triggers the fast decrease in H_{EB}. Therefore, we anticipate that our method could prove useful not only for investigating the dynamics of exchange interaction but also to indirectly probe the loss and recovery of magnetic ordering in antiferromagnets in the femtosecond regime.

As a first attempt to gain a more generic view on the subpicosecond dynamics of the AF spins, a few observations can be made. First, our data show that both the applied magnetic field, as well as the relative orientation of H_{EB} with respect to the magnetization of the FM layer play a minor role. Also, in the preliminary analysis of data for different thicknesses of IrMn and Co layers we did not find significant correlations. Finally we stress that, although $\tau_{\mathrm{EB},0}=0.7$ ps is extremely fast, it is significantly slower than typical demagnetization times in FM transition metals, $\tau_M \sim 100$ fs. This might place the dynamics in a different regime compared to FM transition metals, where τ_M is on the order of the electron thermalization time and a factor of $\sim$5 faster than the electron-phonon energy equilibration. However it should be noted that the latter time scales have not been systematically studied for IrMn thin films nor for Co/IrMn bilayers. Clearly further investigations are needed to shed more light on the topic.

In conclusion we were able to resolve the genuine ultrafast dynamics of exchange interaction in ferromagnet/antiferromagnet coupled bilayers upon femtosecond laser excitation using a method based on a careful analysis of precessional transients of the FM magnetization. The method was applied to the Co/IrMn system, and enabled a quantitative estimate of the time scale at which the exchange interaction is quenched: $\tau_{\mathrm{EB},0}=(0.7\pm0.5)$ ps. We envision that this result may provide insight in the microscopic mechanisms that determine the interlayer exchange coupling, and inspire possibilities for magnetic switching. Moreover, in especially engineered samples, the method we developed could be used to resolve the ultrafast laser-induced spin dynamics of antiferromagnetic metals.

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