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

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Laser-induced magnetization dynamics in a perpendicularly exchange-coupled TbFeCo/GdFeCo bilayer film are studied by using pump-probe magneto-optical Kerr spectroscopy. An ultrafast modulation effect on local magnetization orientation is observed. Such ultrafast magnetization reorientation in the GdFeCo layer is revealed to be triggered by the femtosecond laser pulse and driven by the effective exchange field. These processes occur within a timescale of hundreds of picoseconds, in which the field- and fluence-dependent dynamical behaviors are demonstrated. In addition, an atomistic Heisenberg model is proposed for studying the laser-induced magnetization dynamics by using micromagnetic simulation. The simulated results agree with the experimental phenomena and further reveal the underlying mechanism. These results show an approach for ultrafast manipulation of the local magnetization orientation in perpendicularly exchange-coupled structures.

Keywords: magnetization dynamics, exchange coupling, time-resolved magneto-optical Kerr spectroscopy, magnetization reorientation, femtosecond laser, micromagnetic simulation

1 INTRODUCTION

Exchange-coupled composites (ECCs) have attracted much interest due to their designable magnetic properties from combining individual material with different properties [1, 2] and also potential applications in magnetic recording [3, 4], magnetic sensors [5, 6] and magnonic devices [7, 8]. In particular, many research works focused on the perpendicular ECCs which have high thermal stability, reduced switching field, and thus excellent applied potential for ultrahigh-density magnetic recording [9–11]. On the other hand, laser-induced magnetization dynamics in magnetic system is also an active field. Introduction of the ultrashort laser pulses can lead to infusive effects, such as further reduce coercivity and remarkably accelerate magnetization reversal for magnetic recording [12–15]. Therefore, studying laser-induced or -modulated ultrafast magnetization dynamics in perpendicular ECCs is demanded for developing related applications.

Amorphous ferrimagnetic rare earth-transition metal (RE-TM) alloy films, such as GdFeCo and TbFeCo, are one kind of the most-concerned materials for ultrafast magnetic applications [16–18]. Typically, GdFeCo and TbFeCo can be used as the free and pinning layers, respectively, in perpendicular ECCs due to their dissimilar spin-orbit coupling and perpendicular magnetic anisotropy (PMA) [2, 19]. RE-TM ECCs have attracted attentions also due to their applied

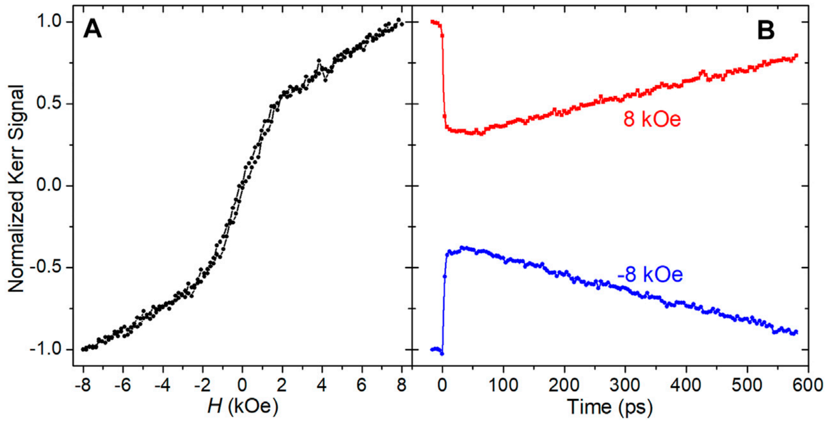

FIGURE 1 | (A) Normalized polar Kerr hysteresis loop of the GdFeCo layer. **(B)** Laser-induced magnetization dynamics measured under H = ±8 kOe and pump fluence of 9.8 mJ/cm^2. The Kerr signal is normalized to the static Kerr value at H = ±8 kOe.

potential in magnetic super resolution (MSR) for magneto-optical readout [20–22], but observation of the related dynamical process has not been reported.

In this paper, we investigate the laser-induced magnetization dynamics in a perpendicularly exchange-coupled TbFeCo/GdFeCo bilayer film by using pump-probe magneto-optical polar Kerr spectroscopy. We observe an ultrafast modulation effect on local magnetization orientation in the GdFeCo layer which is triggered by the femtosecond laser pulse and driven by the effective exchange field, and also first demonstrate the dynamics. In addition, an atomistic Heisenberg model is proposed for studying these laser-induced magnetization dynamics by using micromagnetic simulation.

2 EXPERIMENT

The sample studied here is a TbFeCo(40 nm)/GdFeCo(50 nm) coupled bilayer film prepared on a glass substrate by magnetron sputtering. The TbFeCo layer with high PMA has a strong coercivity (H_c) of ~7 kOe, while the GdFeCo layer has weak magnetocrystalline anisotropy and hence possesses in-plane magnetization.

The laser-induced magnetization dynamics are measured by using a time-resolved pump-probe magneto-optical Kerr configuration. Linearly polarized laser pulse train with a central wavelength of 800 nm, a duration of 100 fs, and a repetition rate of 1 kHz is generated from a Ti:sapphire regenerative amplifier and split into the pump and probe beams. Both the two beams are almost incident normally on the surface of GdFeCo layer with a pulse fluence ratio of pump to probe larger than 30. The focused spot diameter of the pump beam is ~150 μm, while the probe spot located at the center of the pump spot is set to be nearly a half smaller to decrease the temperature gradient within the probed area. The probe beam reflected from the sample is divided into two orthogonally polarized components by a Glan prism to measure the polar Kerr rotation by using a differential detector combined with a lock-in amplifier. A variable magnetic field generated by an electromagnet is applied perpendicularly to the sample plane. All measurements are performed at room temperature.

3 RESULTS AND DISCUSSION

Considering the light penetration depth, the polar Kerr signal in our experiment mainly comes from the out-of-plane magnetization component in the upper half part of GdFeCo layer. Note that for RE-TM materials, the Kerr signal probed at 800 nm is contributed from the magnetic moment of TM atoms [18], namely FeCo atoms here. As shown in **Figure 1A**, the out-of-plane polar Kerr hysteresis loop presents the hard-axis hysteresis of GdFeCo layer. Due to the competition between the demagnetizing field and the effective bias field from exchange coupling [2, 10], a nonuniform magnetization distribution is formed in the GdFeCo layer. The slight hysteresis under small external field just presents the different magnetization states of GdFeCo layer originated from the opposite saturation states of TbFeCo layer.

Figure 1B shows the laser-induced magnetization dynamics measured under pump fluence of 9.8 mJ/cm^2 and external field (H) of ±8 kOe which is larger than H_c of TbFeCo layer. Only ultrafast demagnetization and magnetization recovery can be observed in the dynamical process. The magnetization recovery time is ~460 ps, showing a slow heat-diffusion process.

Next, field-dependent magnetization dynamics are measured. Anomalous dynamical phenomena are observed in a range of small field. In **Figure 2A**, it seems that the trace for H = 1.6 kOe still only presents the typical dynamical processes of ultrafast demagnetization and magnetization recovery. However, the decay curves for −800 Oe, −320 Oe, and 0 Oe all cross their initial magnetization states. With decreasing H, the crossing amplitude increases, while the crossing time decreases from 462 to 176 ps. In the case of 0 Oe (without external field applied), the crossing amplitude seems even larger than the demagnetization amplitude. This anomalous behavior could not be attributed to the magnetization precession, a strong evidence is that the crossing time increases with increasing H, whereas the time period of magnetization precession should decrease with increasing H [23]. Then, what does it originate from?

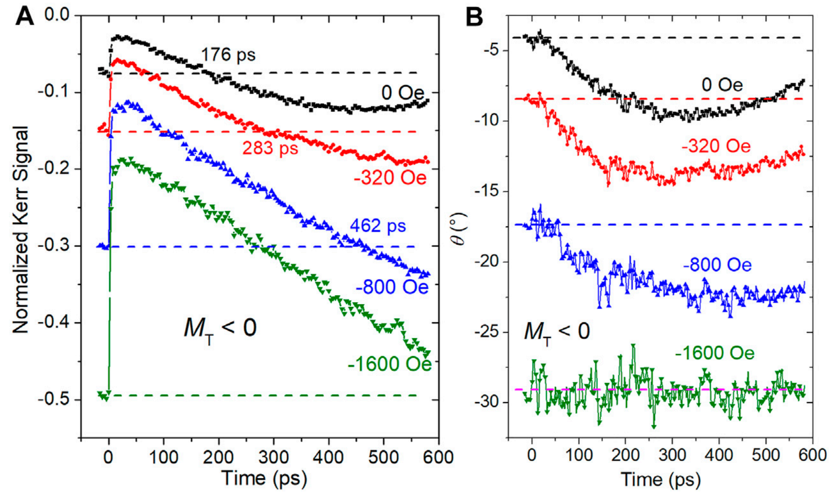

FIGURE 2 | (A) Laser-induced magnetization dynamics measured under different external field and a constant pump fluence of 9.8 mJ/cm^2. **(B)** Average magnetization orientation angle θ as functions of delay time. The dashed lines in both **(A)** and **(B)** denote the initial state before excitation. The sign of M_T shows the saturated state of the TbFeCo magnetization.

As mentioned above, because of the limited light penetration depth, direct laser excitation on the TbFeCo layer can be neglected. The femtosecond laser pulse mainly decreases the transient magnetization of GdFeCo layer, and simultaneously reduce the demagnetizing energy (E_d) and the exchange energy (E_A) [10]. These would change the equilibrium distribution of the magnetization orientation in GdFeCo layer, leading to the transient magnetization reorientation. Mostly in RE-TM layer, the reduction of E_d is more obvious, so the magnetization reorientation is expected to toward the orientation of TbFeCo magnetization driven by the exchange field. Note that along the vertical direction, the demagnetizing field in GdFeCo layer is nonuniform, the vertical diffusion of laser heating should increase the transient influence on demagnetizing field, and thus further singularize the role of exchange field. Especially, if the transient temperature is around the magnetization compensation point [16], a remarkable reduction of E_d should also significantly enhance this effect. Moreover, with H increased, the role of exchange field in the total effective field become minor, and thus the magnetization reorientation effect would be gradually submerged by the magnetization recovery, leading to smaller crossing amplitude and shorter crossing time.

Time dependence of the magnetization orientation can be estimated *via* the relation $M_z(t) = |\boldsymbol{M}(t)| \sin\theta(t)$, where $M_z(t)$ is the projection of magnetization on z axis (out-of-plane component) and θ is the angle between magnetization orientation and the sample plane. $|\boldsymbol{M}(t)|$ is the magnitude of magnetization which is dependent on the transient temperature and can be approximately obtained from the dynamic trace measured under $H = \pm 8$ kOe as shown in **Figure 1B** [15]. **Figure 2B** shows θ as functions of the delay time extracted from **Figure 2A**. Here we can clearly see an ultrafast modulation effect on the local magnetization orientation in GdFeCo layer. Note that those are only the average results of the nonuniform magnetization distribution in the probe depth. Such modulation effect of θ can be only observed under relatively small field ($H < \sim1$ kOe). The max change of θ is ~6.1° and occurs at ~300 ps after laser excitation without external field applied. Evidently, recovery of θ originates from the recovery of E_d and E_A.

To further demonstrate the role of exchange field in the dynamics, we compare the transient traces measured with opposite saturated magnetization states of the TbFeCo layer (denoted by the sign of M_T) and opposite directions of H, respectively, as shown in **Figure 3A**. As expected, opposite M_T results in opposite direction of magnetization reorientation, even with the same condition of external field and laser fluence. This is also another strong evidence for excluding the magnetization precession as the origin of the observed dynamical phenomena. It is clear that the initial state of magnetization in the probed area is dominated by H, while the direction of magnetization reorientation is controlled by M_T, implying that just the exchange field drives the magnetization reorientation after laser excitation. This result agrees with that of the steady measurements for MSR [20], but here we first reveal the dynamics and related timescale.

Figure 3B shows the dynamics measured under the same H and M_T but different laser fluence. The higher fluence not only leads to a more remarkable demagnetization, but also a larger crossing amplitude and a shorter crossing time, showing that increase of the laser excitation energy can accelerate the magnetization reorientation. This result further demonstrates the laser modulation effect of the local magnetization orientation.

4 MICROMAGNETIC SIMULATION

In order to further understand the mechanism of above experimental phenomena, we construct an atomistic

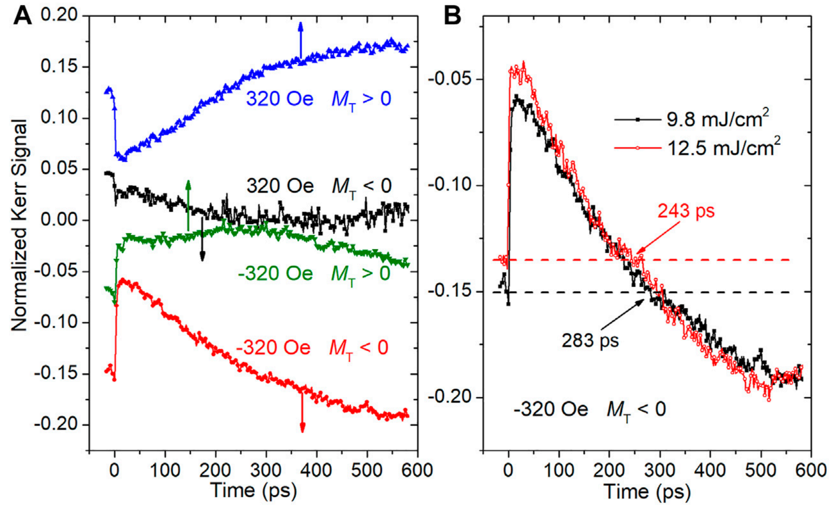

FIGURE 3 | (A) Laser-induced magnetization dynamics depended on the directions of exchange bias field and external field. The sign of M_T shows the saturated state of the TbFeCo magnetization which reflects the direction of exchange bias field. The arrows denote the direction of magnetization reorientation. **(B)** Comparison of magnetization dynamics measured under different excitation fluence.

Heisenberg model for describing the magnetic states of FeCo atoms in GdFeCo layer of the sample, which comprises of nearest ferromagnetic exchange interaction (J), anisotropy (A), and effective magnetic field (B_z) terms [24, 25]:

$$\mathcal{H} = -J\sum_{i,j} \boldsymbol{m}_i \cdot \boldsymbol{m}_j - A\sum_i \left(m_{i,x}^2 + m_{i,y}^2\right) - \boldsymbol{B}_z \cdot \sum_i \boldsymbol{m}_i, \qquad (1)$$

where the magnetic moments are imposed on a two-dimensional square lattice with periodic boundary conditions, with $\boldsymbol{m}_i = (m_{i,x}, m_{i,y}, m_{i,z})$ denoting the magnetic moment at site i in the xy-plane. We consider an easy-plane magnetic anisotropy ($A > 0$) in the GdFeCo layer as revealed by the experimental observation. In phenomenological sense, such an easy-plane anisotropy naturally arises from the strong magnetic dipolar interactions (demagnetization energy) in the system. The inter-layer exchange coupling can be parameterized by an exchange bias field acting on the FeCo atoms in the GdFeCo layer, which orients towards the same direction with that of M_{T}. For simplicity, the exchange bias field here is included into the external magnetic field as an effective magnetic field $\boldsymbol{B}_z = B_z \boldsymbol{e}_z$ normal to the xy-plane.

In the simulations, we first study the temperature dependences of equilibrium magnetic properties of GdFeCo layer using the Langevin Landau-Lifshitz-Gilbert stochastic equation [24, 25] (see *Simulation methods* in the **Supplementary Information**). The temperature-dependent behaviors are monitored by evaluating the thermal-averaged magnetization $<M>$ and magnetization components $<M_z>$ along the z-axis, which are defined as $<M> = \frac{1}{N^2}<\left(\sum_i S_{i,x}\right)^2 + \left(\sum_i S_{i,y}\right)^2 + \left(\sum_i S_{i,z}\right)^2>$ and $<M_z> = \frac{1}{N} < \sum_i S_{i,z}>$, where $\boldsymbol{S}_i = (S_{i,x}, S_{i,y}, S_{i,z})$ denotes a classical Heisenberg spin with unit length at site i, N is the number of spins, and <...> refers to the thermal average for the equilibrium states at a given temperature T. The simulated results in **Figure 4A** show that the magnetization $<M>$ decreases from 1.0 at $T = 0$ to ~0.0 at high temperature with T increases, corresponding to the transition from ferromagnetic state at low T to paramagnetic state at high T. In addition, the reduced $<M_z> - T$ curve exhibits that $|<M_z>|$ rises smoothly when $T < T_{\mathrm{c}}$ and drops down when $T > T_{\mathrm{c}}$ with T increases. This indicates that the out-of-plane reorientation of magnetization occurs with increasing temperature when $T < T_{\mathrm{c}}$ [26–28], and just agrees with the experimental results.

To proceed, we simulate the laser-induced ultrafast magnetization dynamics by employing the Landau–Lifshitz–Bloch (LLB) equation [29–32], in which the temperature of atomic spins are determined by using the three-temperature model [33–35] (see *Simulation methods* in the **Supplementary Information**). In this scheme, the system is first relaxed by solving the LLB equation under the effective magnetic field B_z for reaching the equilibrium state. Then we carefully tracked the dynamics of out-of-plane components of magnetization ($M_z = \frac{1}{N}\sum_i m_{i,z}$) from the initial equilibrium state after the laser excitation.

Figure 4B presents the evolutions of M_z as functions of time, and one can see that a sharp demagnetization occurs after excitation of laser-pulse heating. Subsequently, M_z gradually recovers to its equilibrium state. In this process, the intriguing behavior of magnetization reorientation appears and consists with our experimental observation.

In our experiments, M_z shows a slow recovery with complete recovery time larger than 600 ps. In this sense, one may understand the M_z dynamic behaviors from the equilibrated temperature-dependence of $<M_z>$ in

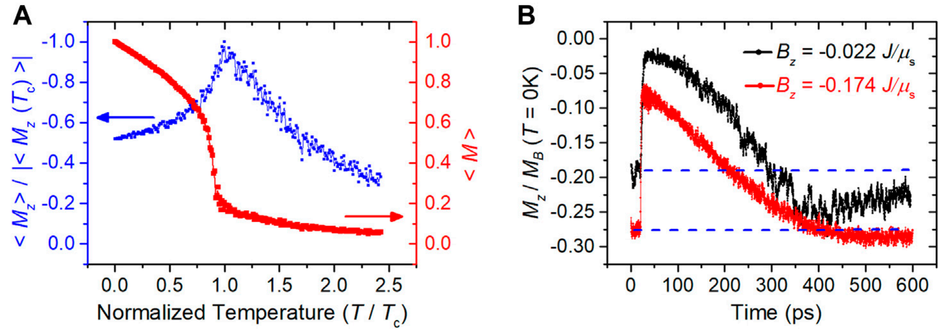

FIGURE 4 | (A) Simulated thermal-averaged out-of-plane magnetization component $<M_z>$ and magnetization $<M>$ of equilibrium states as functions of temperature T, under $B_z = -0.174\, J/\mu_s$ (μ_s is the atomistic magnetic moment). Here $<M_z>$ is normalized in the unit of $|<M_z (T_c)>|$, with $<M_z (T_c)>$ representing the out-of-plane magnetization at Curie temperature T_c. **(B)** Simulated laser-induced dynamics of out-of-plane magnetization component M_z under different effective magnetic field B_z. Here M_z is scaled in the reduced unit of M_B (T = 0 K), which denotes the magnetization under magnetic field B_z and temperature T = 0 K. The initial values of M_z before excitation are marked by the blue dashed lines.

Figure 4A, considering that the system becomes quasi-equilibrium states in the process of magnetization recovery. Thus, the orientation of spins gradually tends to the quasi-equilibrium direction depended on transient temperature, leading to M_z crossing its initial state. With the subsequent recovery of transient temperature, M_z gradually recovers to the equilibrium state. Note that the crossing amplitude decreases and the crossing time increases with increasing $|B_z|$, agreeing well with the experimental observations in **Figure 2A**.

5 CONCLUSION

In summary, the laser-induced magnetization dynamics and the ultrafast modulation effect on local magnetization orientation in a perpendicularly coupled TbFeCo/GdFeCo film are studied by using time-resolved magneto-optical Kerr spectroscopy. The magnetization reorientation in the GdFeCo layer is triggered by the femtosecond laser pulse and driven by the effective exchange field. These processes occur within a timescale of hundreds of ps. We discuss the field- and fluence-dependent dynamical behaviors, and propose an atomistic Heisenberg model to study the dynamics by using micromagnetic simulation. The simulation agrees with the experimental phenomena and further reveals the underlying mechanism. These results show an approach for ultrafast manipulation to the local magnetization orientation in perpendicularly exchange-coupled structures *via* changing exchange bias state and laser fluence.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/**Supplementary Material**, further inquiries can be directed to the corresponding authors.

AUTHOR CONTRIBUTIONS

ZX and ZC designed the experiment, set up the configuration and carried out the measurement. JZ, YC, and JP contributed to the experiment. JZ and ZC finished the data analysis with supports from WZ. JC constructed the model and performed the computations. ZX, JC, and ZC prepared the manuscript. All authors commented on the manuscript.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fphy.2021.755081/full#supplementary-material

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