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Transient ferromagnetic state induced by ultrafast suppression of interlayer exchange coupling in synthetic antiferromagnets

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ABSTRACT

The increasing demand for faster, denser, and low-power memory technologies has intensified interest in antiferromagnetic materials for ultrafast spintronic applications. Synthetic antiferromagnets, with tunable interlayer exchange coupling, provide a versatile platform for high-speed device engineering. Here, we investigate the ultrafast spin dynamics in a synthetic antiferromagnet using time-resolved magneto-optical Kerr effect measurements. We find that laser excitation leads to a significant, transient suppression of interlayer exchange coupling, with recovery occurring on a sub-nanosecond timescale. By combining density functional theory calculations with experimental analysis, we identify this suppression as arising from the demagnetization of adjacent ferromagnetic layers. The magnitude and orientation of atomic magnetic moments on either side of the spacer layer are shown to play a critical role in determining interlayer exchange coupling strength. These findings offer a new pathway for optically controlling the behavior of synthetic antiferromagnets and underscore the potential of interlayer exchange coupling engineering for next-generation memory and ultrafast spintronic devices.

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I. INTRODUCTION

Antiferromagnetic materials have become a key focus for ultrafast and ultra-high density information storage due to their advantages, such as the absence of stray fields, high intrinsic working frequency, and strong resistance to external magnetic interference. Synthetic antiferromagnet (SAF) structures couple two or more ferromagnetic layers in an antiparallel configuration through interlayer exchange coupling mediated by an intermediate layer. The net magnetic moment of SAF structures is zero, which makes it difficult to sense and manipulate them. Therefore, controlling and detecting the antiferromagnetic moment has become a prominent research topic in the field of spintronics.¹ Several novel methods have been proposed, such as accumulating surface charge,² achieving tri-phase

transitions by modulating oxygen vacancies in oxide thin films,³ and regulating magnetic properties by changing the electron density at the Fermi level.⁴ Zhou *et al.* used time-resolved magneto-optical Kerr effect (TR-MOKE) to investigate the precessional dynamics in laser-induced antiferromagnetically coupled ferromagnetic thin films, revealing the magnetization precession behaviors in different modes, including acoustic, optical, and transient modes, which are determined by the interlayer coupling strength and external magnetic field.⁵ Malinowski *et al.* demonstrated that controlling the intermediate layer material could regulate the spin angular momentum transfer efficiency, thereby influencing the demagnetization speed in SAF structures.⁶ Igarashi *et al.* also showed the influence of interlayer exchange coupling on laser-induced magnetization reversal.⁷ Wu *et al.* explored the effect of annealing on the magnetic

dynamic behavior of Co/Ni-based SAF structures and found that annealing temperature significantly affected the interlayer coupling field and perpendicular magnetic anisotropy field, thus altering the magnetization demagnetization and precession processes.⁸

Although these studies provide valuable insights into the ultrafast spin dynamics within SAF structures, they mainly focus on the influence of interlayer antiferromagnetic coupling on ultrafast demagnetization speed. The temporal evolution of interlayer antiferromagnetic coupling itself after laser excitation is still lacking. In this study, $[\text{Pt}/\text{Co}]_2/\text{Ru}/[\text{Co}/\text{Pt}]_2$ multilayers with their antiferromagnetic behavior originating from the classical Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction⁹ are used. TR-MOKE is employed to measure the dynamic response of the interlayer exchange coupling on an ultrafast time scale, revealing the temporal evolution of RKKY in SAF structures after laser excitation. First-principles density functional theory (DFT) calculations and atomistic spin dynamics simulations are employed and show that the thermal effects from the laser excitation significantly suppress the recovery of RKKY, and the full re-establishment of RKKY can take up to or even over one nanosecond, much longer than the recovery time of direct exchange coupling between atomic spins. Compared to direct antiferromagnetic exchange coupling between magnetic atoms in conventional antiferromagnetic materials, modulating the interlayer exchange coupling requires a lower energy and has a longer retention time. These insights provide a new approach for ultrafast information storage and serve as a theoretical foundation for the design and optimization of spintronic devices.

II. RESULTS

This study employs a SAF with perpendicular magnetic anisotropy (PMA) as the research system, having a multilayer structure of $[\text{Pt}/\text{Co}]_2/\text{Ru}/[\text{Co}/\text{Pt}]_2$, as illustrated in Fig. 1(a). In this structure, the Ru spacer layer provides interlayer antiferromagnetic coupling through RKKY interaction, while the periodic Pt/Co superlattice stacks on both sides are designed to enhance the PMA. Vibrating sample magnetometer (VSM) measurements reveal that the sample exhibits a typical rectangular hysteresis loop (Fig. S1)

with a magnetic field applied perpendicular to the sample plane, indicating excellent PMA. Furthermore, the magnetic moment compensation between the top and bottom magnetic layers exceeds 99% (with remanent magnetization ratio $M_{\text{rem}}/M_s < 1\%$), and the interlayer exchange coupling field is measured to be $\mu_0 H_{\text{RKKY}} = 2.7$ kOe, confirming strong antiferromagnetic coupling characteristics. Prior to ultrafast laser excitation, two distinct magnetic states were initialized by applying desired external magnetic fields: (i) a parallel (P) state, in which both Co sublayers were magnetized along the +z direction, and (ii) an antiparallel (AP) state, characterized by fully compensated magnetic moments.

It should be noted that although the magnetic moments in the SAF sample measured by TR-MOKE are fully compensated in the AP state, the Kerr signals originating from the upper and lower magnetic layers remain distinguishable due to a finite skin depth. Moreover, for the same reason, there is a difference in the absorption of the 800 nm pump laser between these two layers, and the upper magnetic layer exhibits a greater degree of demagnetization than the lower one. These two factors are discussed in detail in Sec. 3 of the [supplementary material](#). Combining those two effects, the demagnetization signal contributed by the lower layer ($\Delta\theta_{k2}$) is $\sim 60\%$ of that from the upper layer ($\Delta\theta_{k1}$). Therefore, the ultrafast demagnetization dynamics of the AP state can still be effectively probed. In addition to inducing demagnetization of both upper and lower layers, laser excitation is also expected to modulate the strength of RKKY, $H_{\text{RKKY}}(t)$, in the SAF structure. In an initial AP state set by an applied external magnetic field $H_{\text{ext}} < H_{\text{iec}}(0)$, the magnetic layer experiencing a negative effective field $H_{\text{eff}} = H_{\text{ext}} - H_{\text{iec}}(0) < 0$ has its moment oriented opposite to the external magnetic field prior to the pump pulse. Upon the arrival of the ultrafast laser pulse, the interlayer exchange coupling $H_{\text{iec}}(t)$ is substantially reduced due to thermal effects, causing the effective magnetic field to reverse direction, becoming positive as $H_{\text{eff}} = H_{\text{ext}} - H_{\text{iec}}(t) > 0$. Under the influence of this reversed effective field, the magnetic layer initially aligned opposite to the external field starts to rotate toward the field direction, creating a transient ferromagnetic state. Although both ferromagnetic layers remain partially demagnetized at this stage, their originally compensated magnetic moments now

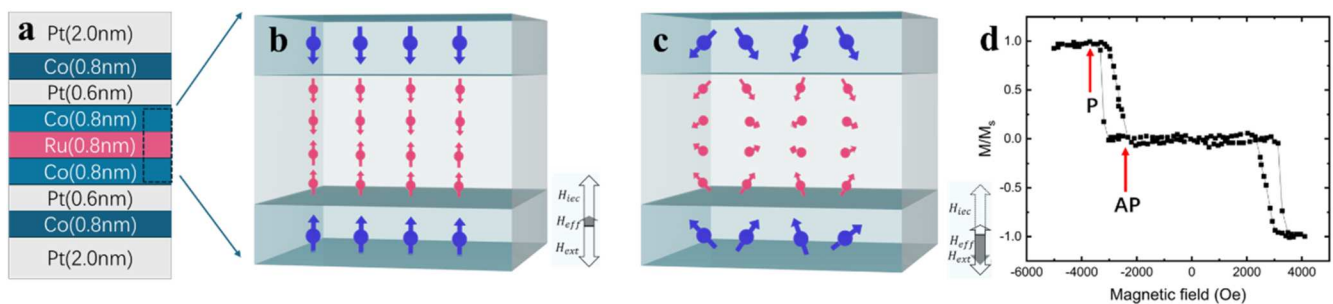


FIG. 1. Design principle and ultrafast manipulation of RKKY-coupled synthetic antiferromagnets. (a) Schematic illustration of the sample structure. (b) Before the pump pulse arrives, the magnetic moments of the two ferromagnetic layers induce a large magnetic moment in the Ru layer. Consequently, the interlayer exchange field H_{iec} exceeds the external magnetic field H_{ext} . The ferromagnetic layer-oriented opposite to H_{ext} experiences an upward effective field H_{eff} , placing the system in an antiferromagnetic state. (c) After laser-induced demagnetization of the Co layers on both sides, the Ru magnetic moment—induced by the Co layers—decreases, causing a substantial reduction in H_{iec} . As a result, H_{eff} reverses, and the ferromagnetic layer previously opposing H_{ext} is now magnetized along the external field direction, giving rise to a transient ferromagnetic state. (d) VSM measurement results of the experimental sample.

combine constructively, resulting in a measurable transient magnetization enhancement relative to the initial AP state. This scenario is illustrated in Figs. 1(b) and 1(c).

Figure 2 presents the TR-MOKE and reflectivity measurements of the SAF structure in both P and AP states. Crucially, the pump fluence was kept identical in both configurations to isolate state-dependent spin dynamics from thermal effects induced by the ultrafast laser pulse. Although the external magnetic fields applied for stabilizing the AP and P states differ by only 300 Oe, significant differences in ultrafast dynamical behaviors were observed between these two states.

First, the magnitude of ultrafast demagnetization differs markedly between the two states. This is because in the P state, both upper and lower Co layers contribute negative demagnetization signals, while in the AP state, the demagnetization signals from these two layers have opposite signs and partially cancel each other, resulting in a significantly larger net demagnetization signal in the P state compared to the AP state, as indicated by a large peak in Fig. 2(a) and a small peak in Fig. 2(b).

Second, notable differences occur during the subsequent magnetization recovery process. In the P state, the system undergoes an ultrafast demagnetization followed by a gradual magnetic recovery, with complete restoration of magnetization ($\Delta\theta_k = 0$) occurring within ~ 600 ps, as shown in Fig. 2(a). In the AP state, in contrast, a full recovery of magnetization to $\Delta\theta_k = 0$ occurs within only about 100 ps. Moreover, after this ultrafast recovery, a pronounced ultrafast magnetization enhancement is observed in the AP state, exhibiting a transient ferromagnetic-like state analogous to that observed in single-pulse all-optical switching processes in GdFeCo systems.¹⁰ Although the change in the sign of $\Delta\theta_k$ effectively characterizes the transient ferromagnetic state, the observed transition of

$\Delta\theta_k$ from negative to positive also depends on the initial AP configuration. For detailed model discussion and atomistic spin dynamics¹¹ simulation results, see Secs. 3 and 4 in the [supplementary material](#).

Interestingly, the timescale of this transient ferromagnetic-like state varies significantly across different material systems. In GdFeCo alloys, the transient ferromagnetic-like state emerges rapidly within 0.4 ps after laser excitation and persists for less than 2 ps.¹² In contrast, in the SAF structure, this transient state appears only after ~ 100 ps but persists for a duration nearly two orders of magnitude longer. This substantial difference originates from the fundamentally different exchange mechanisms in the two systems. In GdFeCo, the transient ferromagnetic state is governed by direct exchange coupling between localized magnetic moments, enabling ultrafast angular momentum transfer and rapid exchange recovery. In contrast, the interlayer exchange coupling in SAFs originates from the indirect RKKY interaction mediated by itinerant conduction electrons across the Ru spacer layer.¹³ The recovery of this indirect exchange interaction requires reconstruction of spin-dependent electronic polarization and interfacial hybridization, which is substantially slower than local atomic exchange interactions. As a result, the transient ferromagnetic state in the SAF emerges on a delayed timescale and persists much longer than that in GdFeCo.

To elucidate this remarkable difference, it is necessary to revisit the physical origin of the RKKY interaction. IEC in metallic multi-layer systems arising from the RKKY interaction can be expressed as¹³

$$J_{\text{RKKY}}(R) = J_{\text{ex}}^2 \frac{\sin(2k_F R) - 2k_F R \cos(2k_F R)}{R^4},$$

where R represents the distance between the magnetic layers, k_F is the Fermi wavevector of the system, and J_{ex} denotes the intra-atomic

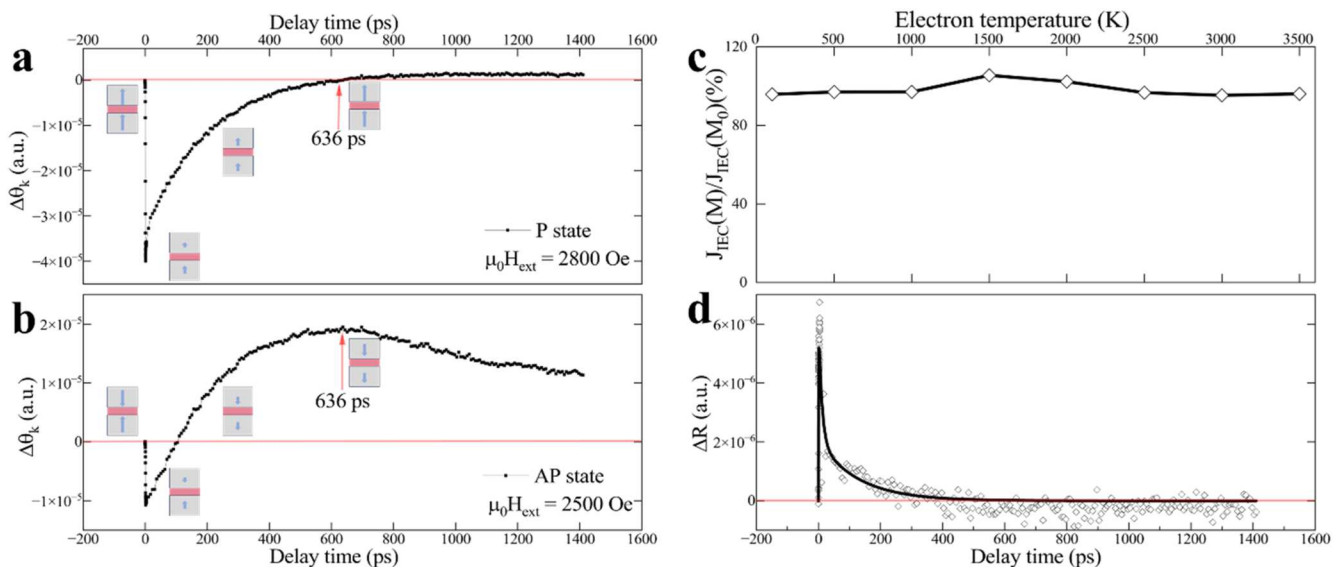


FIG. 2. TR-MOKE scans in the SAF structure $[\text{Pt}/\text{Co}]_2/\text{Ru}/[\text{Co}/\text{Pt}]_2$ under a pump power of 12 mW. Panel (a) shows the Kerr signal spectrum with the system in the P state, panel (b) shows the Kerr signal spectrum with the system in the AP state, and panel (c) shows RKKY strength as a function of electron temperature. (d) shows the reflectivity spectrum.

exchange interaction between the local magnetic moment and the conduction electrons. The laser-induced weakening of RKKY arises from its influence on R , k_F , and J_{ex} . First, the effect on k_F can be excluded since a significant increase in the electron temperature in metals does not alter the magnitude of the Fermi wavevector. Moreover, DFT calculations confirm that even when the electron temperature is raised to 3500 K, J_{RKKY} does not exhibit any appreciable change, as shown in Fig. 2(c).

The influence of the laser on R is primarily due to the lattice vibrations that arise when electrons transfer thermal energy to the lattice, leading to a change in R . Although this effect can indeed have a notable impact on J_{RKKY} , a comparison of the time-resolved reflectivity and Kerr signal spectra allows us to rule out this factor. As seen in Fig. 2(d), while ΔR recovers after 400 ps, the magnetic moment in the AP state shown in Fig. 2(b) continues to increase, indicating that even after the lattice thermal effects have subsided, J_{RKKY} remains unrecovered. Therefore, lattice heating is not the dominant factor, either.

Finally, the only remaining possibility is that the laser-induced reduction of J_{ex} is responsible. In the Co/Ru/Co system, Ru's 4d

electrons hybridize with Co's 3d electrons, thereby inducing a magnetic moment in Ru's 4d states. Although J_{ex} is intrinsically determined by the overlap of electron wave functions and does not directly depend on the magnitude of Co's magnetic moment, the pump's direct action on Co's 3d electrons increases their delocalization. This, in turn, indirectly reduces the intra-atomic exchange interaction strength between Co's local magnetic moments and the conduction electrons, leading to a reduction of J_{ex} . Comparison of the P and AP states reveals that the time at which the positive Kerr rotation $\Delta\theta_k$ in the AP state approaches maximum value essentially coincides with the time when $\Delta\theta_k$ in the P state returns to zero, as shown in Figs. 2(a) and 2(b). This observation further confirms that J_{RKKY} is indeed related to the magnetic moment in the ferromagnetic layer.

Based on the previous analysis showing that RKKY coupling is correlated with the magnitude of Co's magnetic moment, we further investigated the relationship between demagnetization levels of Co and RKKY coupling in the SAF. Figure 3(a) displays the dynamic processes in the same sample excited with different pump powers under a fixed 2500 Oe magnetic field. At lower pump powers,

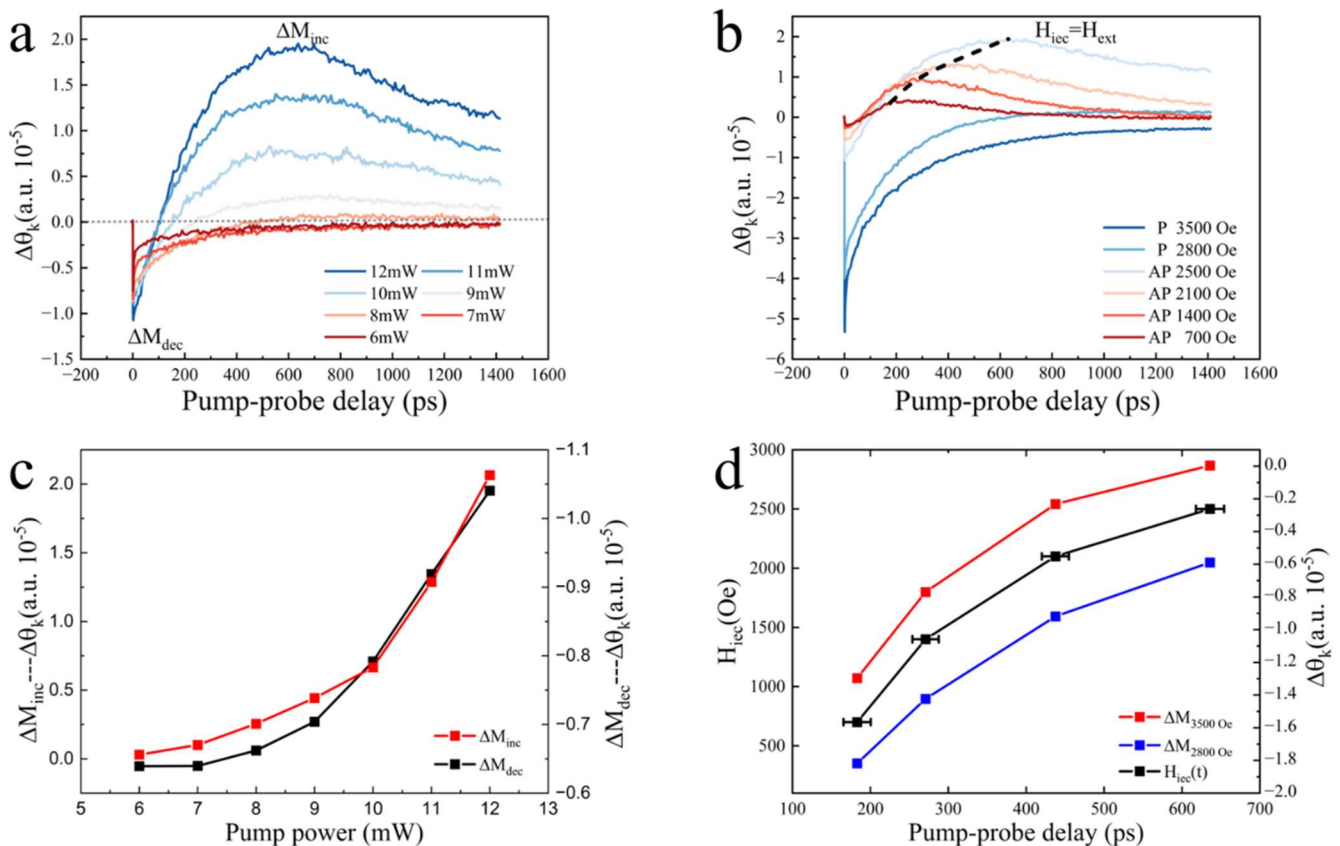


FIG. 3. TR-MOKE measurements under different pump powers and external magnetic fields. (a) TR-MOKE measurements under a constant external magnetic field of 2500 Oe with varying pump powers. (b) TR-MOKE measurements under a fixed pump power (12 mW) with varying external magnetic fields. The red arrows indicate the time points where $\Delta\theta_k$ reaches its maximum. (c) Demagnetization and magnetization levels as a function of pump power. (d) The black dots represent the points where $\Delta\theta_k$ reaches its maximum as indicated by the red arrows in panel (b), corresponding to the condition where $H_{rec}(t) = H_{ext}$. The red and blue dots represent the corresponding $\Delta\theta_k$ values at the same time point in the P state.

the Kerr rotation scans did not cross the zero line to the positive side during recovery processes. Only when the pump power reached 8 mW and over, the Kerr rotation scans started to cross the zero line and become positive, showing a magnetization enhancement ($\Delta\theta_k > 0$), with the ultrafast magnetization enhancement becoming more pronounced at higher pump powers. In conjunction with our earlier analysis, these results indicate that the degree of demagnetization is linked to the extent of the magnetization enhancement. The values of the maximum demagnetization and the maximum magnetization enhancement extracted from the Kerr rotation scans in Fig. 3(a) are plotted as a function of the pump power in Fig. 3(c). The relationship between both values and the pump power is almost identical, clearly demonstrating that the demagnetization degree is positively correlated with the maximum magnetization enhancement.

Now the question is how exactly the interlayer exchange coupling field $H_{RKKY}(t)$ is affected after laser excitation and how fast it recovers. This crucial information can be extracted by fixing the pump power but varying the magnitude of the external magnetic field H_{ext} instead. Figure 3(b) presents TR-MOKE measurements performed at the same pump power (12 mW) but under different external magnetic fields. First, the degree of demagnetization in SAF is found to correlate positively with the strength of the external magnetic field. This is because, after excitation by the pump pulse, the sample cannot return to its initial magnetic state under low magnetic fields. As a result, the laser-irradiated region evolves into a multidomain state, reducing the Kerr signal and, therefore, leading to a smaller detected demagnetization amplitude. In addition, we observe a pronounced transient ferromagnetic enhancement with the external magnetic field ranging from 700 Oe to 2500 Oe, under which the initial magnetic state of SAF is in an AP state. When the external magnetic field is larger than 2500 Oe, resulting in an initial P state, the transient ferromagnetic enhancement disappears. As discussed earlier, the transient magnetic enhancement occurs when $H_{eff} = H_{ext} - H_{RKKY}(t) > 0$. The magnetic enhancement

continues increasing until $H_{iec}(t)$ recovers to a value equal to or greater than H_{ext} . Therefore, the peak points of the enhancement in the $\Delta\theta_k$ curves shown in Fig. 3(b) directly correspond to the moments when $H_{RKKY}(t) = H_{ext}$. By plotting these peak times, we effectively track the temporal evolution of $H_{iec}(t)$ as presented in Fig. 3(d). The corresponding degree of demagnetization of the Co layers at these specific times is also plotted. The synchronized evolution of these two quantities clearly demonstrates that the magnitude of the Co magnetic moments is the key factor determining the dynamic recovery of interlayer exchange coupling.

III. DISCUSSION

In order to further verify this point, we perform DFT calculations to examine how the demagnetization of the Co magnetic moment leads to a decrease in J_{RKKY} . Femtosecond laser-induced ultrafast demagnetization involves both a rapid reduction in atomic magnetic moments¹⁴ and a loss of spin-direction coherence.¹⁵ Accordingly, using VASP's constrained magnetic moment approach, we investigate how both the magnitude and orientation of the Co magnetic moments affect J_{RKKY} . In the random spin-orientation calculations, we construct a supercell where each Co moment in the xy-plane is randomly oriented, while maintaining either parallel or antiparallel orientations of the top and bottom Co layers along the z axis. As illustrated in Fig. 4(a), the results confirm that the coherence of Co spin orientations significantly influences J_{RKKY} . As shown in Fig. 4(b), J_{RKKY} is highly dependent on the magnetic moment of the ferromagnetic layer. As the Co magnetic moment decreases, J_{RKKY} rapidly diminishes; once the Co moment falls below $1.6 \mu_B$, the coupling shifts between antiferromagnetic and ferromagnetic. Hence, during demagnetization, both the magnitude of the Co magnetic moment and its directional consistency play critical roles in governing J_{RKKY} .

In our calculations, we observed that variations in the magnitude of the Co magnetic moments also led to changes in the Ru

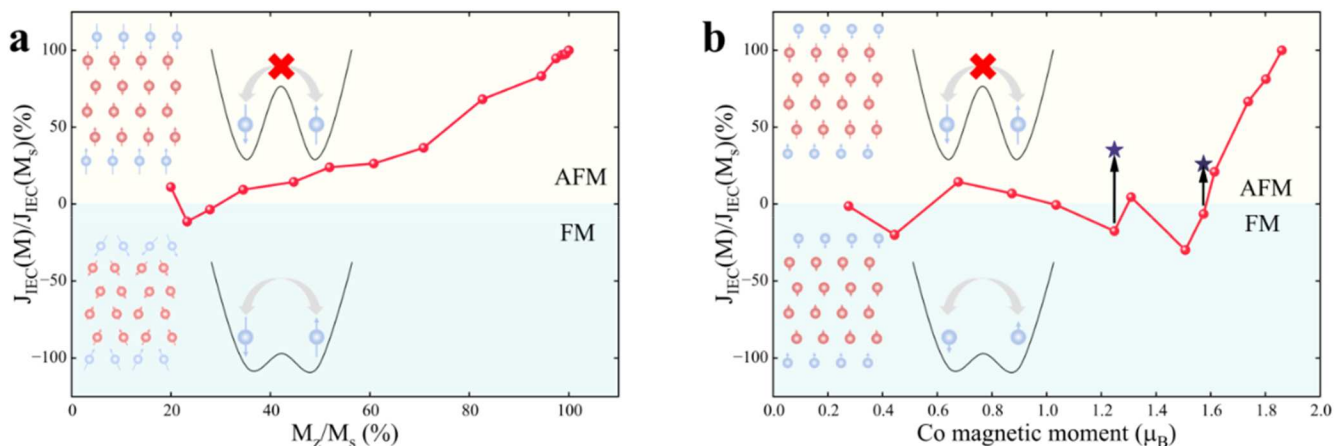


FIG. 4. DFT calculation results showing the relationship between RKKY coupling strength and the z-component of the Co layer magnetic moments (a), as well as the correlation with the magnitude of the Co atomic magnetic moments (b). The blue star data points in (b) represent calculations where the Co magnetic moments are set to the same values as the corresponding lower red dots, but with the Ru magnetic moment fixed at its AFM value. This simulates a scenario during ultrafast demagnetization in which the Co moments decrease while the Ru moment is assumed to remain temporarily unchanged to simulate a transient nonequilibrium state.

magnetic moment. Previous element-resolved ultrafast studies in Co/Pt multilayers have shown that induced magnetic moments in nonmagnetic heavy-metal layers can exhibit slower demagnetization dynamics than the primary Co moments.¹⁶ Therefore, during the ultrafast nonequilibrium process, it is plausible that the Co magnetic moment decreases faster than the induced Ru moment, resulting in a transient mismatch between Co and Ru magnetizations. In Fig. 4(b), the red star data points represent calculations where the Co magnetic moments are set to the same values as the corresponding lower black dots, but with the Ru magnetic moment fixed at its AFM value. This simulates a scenario during ultrafast demagnetization in which the Co moments decrease while the Ru moment remains unchanged. It can be seen that when the Ru magnetic moment remains at its AFM value, the system, which was previously in a ferromagnetic coupling state, reverts to an antiferromagnetic coupling state. Although the Ru magnetic moment is induced by Co magnetization, during the ultrafast process, there may be a situation where the Co magnetic moment decreases while the Ru moment remains unchanged, resulting in an unchanged interlayer exchange coupling. This delayed recovery of the induced Ru moment may contribute to the experimentally observed delayed emergence of the transient ferromagnetic state (~100 ps).

By combining the reflectivity data, the ultrafast dynamics measurements in the AP/P states, and the DFT results, we provide a comprehensive picture of the spin dynamics in the SAF: the laser initially interacts with Co's 3d electrons, leading to their demagnetization, which directly reduces RKKY coupling and results in a reversal of the effective field H_{eff} . This reversal induces an ultrafast magnetization enhancement in the AP state. As Co's magnetic moment recovers and Ru is magnetized again by Co, RKKY coupling is fully restored, and the system gradually reverts from the enhanced state back to the initial AP configuration. Due to the dependence of the RKKY coupling on the magnitude of Co's magnetic moment, the transient ferromagnetic state in the SAF persists for a significantly longer duration compared to the directly coupled ferrimagnetic system, such as GdFeCo.

In summary, we investigated the relaxation process of RKKY coupling in laser-induced ultrafast spin dynamics in the SAF structure [Pt/Co]₂/Ru/[Co/Pt]₂. By comparing the ultrafast processes in both the P and AP initial states, we demonstrated that the RKKY coupling is significantly weakened by the laser-induced thermal effects, which in turn leads to an ultrafast magnetization enhancement lasting over 1 ns in the AP state. By tuning the pump power to control the degree of demagnetization and performing DFT calculations, we further established that the relaxation of the RKKY coupling is positively correlated with the magnetic moment of the ferromagnetic layers, while showing no significant dependence on either the lattice or electron temperatures. We present a new method for tuning RKKY coupling by modulating the magnetization of the FM layers in SAF. Meanwhile, the ultralong relaxation time of the RKKY coupling offers a new physical picture for ultrafast information storage, such as all-optical switching.

IV. MATERIALS AND METHODS

A. Sample growth

The sample structure for testing is as follows: glass/Ta (5 nm)/Pt (2 nm)/Co (0.8 nm)/Pt (0.6 nm)/Co (0.8 nm)/Ru (t

nm)/Co (0.8 nm)/Pt (0.6 nm)/Co (0.8 nm)/Pt (2 nm), as illustrated in Fig. 1. A total of seven samples with varying Ru thicknesses were fabricated, and we selected a sample with well-defined antiferromagnetic coupling strength for testing, featuring a Ru interlayer thickness of 0.8 nm and an antiferromagnetic coupling field of 2700 Oe. Its magnetic hysteresis loops were characterized using a vibrating sample magnetometer (VSM), as shown in Fig. S1(a). All samples were deposited on 0.13-mm-thick Corning glass substrates at room temperature through DC magnetron sputtering employing separate Co, Pt, Ta, and Ru targets. The sputtering system maintained a base pressure below 4×10^{-5} Pa, with a working Ar pressure of 0.5 Pa during deposition. The deposition rates under a DC current of 40 mA were measured as 0.41, 0.84, 0.48, and 0.52 Å/s for Ta, Pt, Co, and Ru targets, respectively. A 5-nm Ta buffer layer was initially deposited to enhance substrate smoothness, optimize Pt/Co interface quality, and promote (111) crystalline orientation, thereby improving the perpendicular magnetic anisotropy (PMA) of the Co layer. The two Co layers adjacent to the spacer layer enable crucial interlayer exchange coupling, while the repeated Co/Pt multilayer structure ensures strong PMA throughout the system. Finally, a 2-nm Pt capping layer was deposited to prevent surface oxidation of the stack.

B. Time-resolved magneto-optical Kerr effect measurement

Laser pulses with a central wavelength of 800 nm and a pulse width of 50 fs are employed to excite the SAF system. Each laser pulse is split into two pulses, pump and probe. The probe beam is frequency-doubled into a 400 nm wavelength and used to probe transient reflectivity and Kerr rotation. The pump beam is focused onto the sample with a spot diameter of ~400 μm, whereas the probe beam has a spot size of about 200 μm. The probe beam power is maintained at 30 μW, and the pump beam power is varied from 1 to 12 mW. The pump beam is s-polarized, whereas the probe beam is p-polarized. After reflecting from the sample, the probe beam is split into s and p components by a Wollaston prism, and the intensity of both components is recorded by two photodiodes. By fine-tuning the fast axis of a half-wave plate placed before the Wollaston prism, the difference between two photodiodes is set to zero in the absence of pump excitation. The sum and difference of two photodiodes are recorded by fixing the external magnetic field and continuously varying the delay between pump and probe. To eliminate optical measurement errors, signals at positive and negative fields are measured. In these measurements, the pump beam path is optically chopped and the chopping frequency is used as the lock-in reference, thus yielding $\Delta\theta_K$. All experiments are conducted at room temperature. As the sample exhibits PMA, the experimental setup is configured to be sensitive to out-of-plane magnetization, as illustrated in Fig. S2.

C. First-principles calculations

The calculations were performed using the Vienna *ab initio* simulation package (VASP), employing the projector augmented wave (PAW) method to account for electron-core interactions within the framework of the Perdew-Burke-Ernzerhof (PBE) functional.¹⁷ A plane-wave cutoff energy of 320 eV was employed, and an $18 \times 18 \times 1$ k-point grid was used to sample the Brillouin

zone of the Co/Ru/Co heterostructure. Although the experimental structure is Pt/Co/Pt/Co/Ru/Co/Pt/Co/Pt, only the Co layers adjacent to Ru significantly affect the RKKY strength, while the Pt layers primarily provide PMA. Consequently, using a simplified Co/Ru/Co model in the calculations is appropriate. We adopt a face-centered cubic (FCC) configuration, which is the most common phase for Co in Pt/Co multilayers and is also a typical structure for Ru. To investigate how the antiferromagnetic coupling strength varies with electron temperature and the Co magnetic moment, we employ a two-step approach. In the first step, structural relaxations proceed until the forces converge to less than 0.001 eV/Å. Given the lattice mismatch between Ru and Co, the initial interatomic spacing is increased, after which both the atomic positions and lattice shape are allowed to relax freely. To eliminate periodic boundary conditions along the *z* direction, a 30 Å vacuum layer is introduced.

In the second step, spin-orbit coupling was introduced, and the energy difference between the ferromagnetic and antiferromagnetic states was calculated, with the EDIFF convergence criterion set to 10^{−8} eV. In these calculations, the spin direction of the Co atoms was fixed using VASP's constrained magnetism method¹⁸ to obtain both antiferromagnetic and ferromagnetic configurations. For calculations at different electron temperatures, the Co spin direction was constrained, whereas for calculations with varying magnetic moment magnitudes, both the direction and magnitude of Co's magnetic moment were fixed. The electron temperature was simulated using a Fermi-Dirac distribution by adjusting the electronic distribution at the Fermi level.

SUPPLEMENTARY MATERIAL

See the [supplementary material](#) for additional details on the static magnetic characterization of the synthetic antiferromagnetic sample, the time-resolved magneto-optical Kerr effect measurement geometry, the analysis of TR-MOKE signal evolution in different initial SAF configurations, the atomistic spin-dynamics simulations, and the atomistic spin model of the magnetic-domain evolution after laser excitation.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Guanqi Li, Youzhi Gu, and Jiajun Li contributed equally to this study.

Guanqi Li: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Writing – original draft (equal); Writing – review & editing (equal). **Youzhi Gu:** Data curation (equal); Methodology (equal); Visualization (equal). **Jiajun Li:** Data curation (equal); Investigation (equal); Methodology (equal). **Junlin Wang:** Software (equal); Writing – review & editing (equal). **Zhendong Chen:** Formal analysis (equal); Investigation (equal); Methodology (equal); Visualization (equal). **Richard F. L. Evans:** Software (equal); Supervision (equal). **Roy Chantrell:** Software (equal); Supervision (equal); Visualization (equal); Writing – review & editing (equal). **Jing Wu:** Funding acquisition (equal); Investigation (equal); Methodology (equal); Supervision (equal); Writing – review & editing (equal). **Yongbing Xu:** Funding acquisition (equal); Supervision (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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