Modulation of the Transient Magnetization of an EuO/Co Bilayer Tuned by Optical Excitation

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The magnetic proximity effect provides a promising way to increase the low Curie temperature (T_C) of europium monoxide (EuO) toward or even above room temperature, while keeping its stoichiometry and insulating properties. This work studies EuO/Co bilayers using static and time-resolved magneto-optical Kerr effect measurements, and explores the influence of magnetic proximity on T_C and on the spin dynamics in EuO. Excitation above the EuO bandgap results in an ultrafast enhancement of the EuO magnetization followed by a demagnetization within nanoseconds. This behaviour is also visible upon selectively photoexciting Co in the EuO/Co bilayer placed in an out-of-plane magnetic field, which is attributed to propagation of a superdiffusive spin current from Co into EuO. As the spin dynamics of Co shows a transient thermal demagnetization, the bilayer provides a system where the transient magneto-optical signal can be tuned in amplitude and sign by varying external parameters such as the sample temperature or pump fluence. Moreover, in a strong excitation regime it is possible to measure the magnetic hysteresis of the underlying EuO, which is present up to room temperature – giving experimental evidence for the presence of a tuneable magnetic proximity coupling between Co and EuO.

1. Introduction

In the context of spintronics,[1] a wide variety of approaches has been explored to manipulate the magnetic configuration in solids and to control spin-related phenomena.[2] In complex oxides, for example, electrons are subject to strong electron-electron interactions leading to a plethora of unconventional physical phenomena.[3,4] Among these functional oxides, ferro- and ferrimagnetic oxides are promising materials for second generation spintronics devices with advanced functionalities, e.g., the realization of spin filters,[5,6] spin transistors,[7] spin diodes and nonvolatile magnetic memories.[3,8] In particular, magnetic oxides which electronically behave as semiconductors may ideally merge spintronics and optoelectronics, for example in spin-controlled light emitting diodes or surface emitting lasers.[9]

In this study, we examine stoichiometric europium monoxide (EuO), a semiconducting ferromagnet with a Curie temperature (T_C) of 69 K. It has a half-filled 4f shell and shows an energy splitting of 0.6 eV of the conduction band. The temperature-dependent magnetization follows a Brillouin function, reaching a saturation magnetization of 7 μ_B/Eu. EuO is thus often described as an almost ideal manifestation of a Heisenberg ferromagnet. The optical spectrum is dominated by transitions from the $4f^7$ valence band orbitals to the 5d-6s conduction band across an indirect band gap of $E_g = 1.19$ eV.[10] EuO has already been successfully employed as a magnetic tunnel barrier with up to 100% spin polarized electrons.[8,11–14] It has also been recognized for its strong magneto-optical effects, with a Kerr rotation at least one order of magnitude larger than that in transition metals.[15,16] Besides its intrinsic characteristics, it exhibits intriguing transient effects on the picosecond timescale. Most strikingly, Liu et al. reported a photoinduced enhancement of magnetization, which is followed by a slower demagnetization process. The former is attributed to the formation of magnetic polarons by optical transition of 4f electrons into the 5d states.[17]

Even though EuO has the highest Curie temperature among the europium chalcogenides,[18,19] it is still too low for applications, which usually require near-room-temperature magnetic ordering. Various approaches to increase T_C have already been investigated. For example, by introducing oxygen vacancies[20–27] or doping with trivalent elements such as gadolinium,[20,23,25,28–32] it is possible to increase the magnetic ordering temperature up to

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170 K. These approaches rely on doping the rare earth oxide and thereby modifying the stoichiometry and conductivity. A promising way to increase the Curie temperature even further, while preserving the insulating properties, is to utilize the magnetic proximity effect by capping the europium chalcogenides with a high T_C transition metal. Thereby, a hybridization between the 5d and 3d states from the rare earth and transition metal, respectively, might occur, which in turn leads to an indirect exchange interaction between the 4f states from the rare earth and the 3d states from the transition metal.[33] This can lead to a significantly increased Curie temperature at the interface, as it was already successfully shown for europium sulphide (EuS), also a ferromagnetic semiconductor.[34–38] Crucially, the influence of magnetic proximity on the spin dynamics of EuO has not been uncovered yet.

In this work, we study EuO/Co as a prototypical magnetic proximity system using static and time-resolved magneto-optical Kerr effect (MOKE) measurements. In particular, we report the picosecond to nanosecond dynamics of EuO/Co bilayers at varying temperature, applied magnetic field, and laser fluence. Our results indicate that the magnetization of EuO can be transiently enhanced in the bilayer by selectively photoexciting the Co layer, a process that can be explained by the propagation of a superdiffusive spin current from Co into EuO. On the other hand, the cobalt layer contributes to the transient magneto-optical signal with the opposite sign than EuO, resulting from a transient thermal demagnetization. The different sign of the EuO and Co signals allows us to tune the transient magneto-optical response of the system in amplitude and sign by varying various external parameters: the delay between the pump and probe beams, the pump fluence, the sample temperature and the value of the applied magnetic field. Furthermore, in the regime of strong optical excitation (fluence = 20 mJ cm^{-2}) we find experimental evidence of the magnetic proximity coupling between Co and EuO, with a correspondent increase of the magnetic ordering temperature at the Co/EuO interface up to room temperature.

2. Results

2.1. Static Characterization

The static hysteresis curves recorded in polar (P-) and longitudinal (L-)MOKE configuration are presented in **Figure 1**. The background originating from the diamagnetic contribution of the substrate is removed using the procedure described in Figure S1 (Supporting Information). The EuO sample shows a clear hysteresis signal at low temperatures, see Figure 1a,b. The remanence is in both cases close to 100%. The coercive field in the P-MOKE configuration is approximately 20 times larger than in L-MOKE. Both the coercivity and the saturation magnetization decrease with increasing temperature and vanish at $T_C \cong 69$ K, the Curie temperature of bulk EuO. For thin films of EuO, a slightly reduced magnetic ordering temperature compared to the bulk was already reported in literature.[11] The slight offset between the measured T_C in the two MOKE configurations originates from different positions of the temperature sensors in the corresponding cryostats.

This straightforward behavior changes once the Co layer is introduced into the system. In the L-MOKE measurements, shown in Figure 1c, the hysteresis loops at low temperatures show significantly smaller coercivity and remanence. Also, the saturation magnetization approaches zero already at 50 K, i.e., at a lower temperature compared to the pure EuO sample. By further increasing the temperature, the hysteresis appears again in an inverted orientation. Since it is known from literature[39] that the Kerr rotations of EuO and Co have opposite signs for the photon energy used in our experiment, our results suggest a substitution of the dominant magneto-optical contribution upon crossing a temperature threshold, a behavior which is typically known in antiferromagnetically coupled ferrimagnets.[40] As the Kerr rotation in rare earths is described to be at least one order of magnitude larger than that in transition metals[15,16] ($\approx -0.3°$ and $\approx 2°$ at 3 eV for Co and EuO, respectively,[39] for 3 eV optical probe as used in the L-MOKE configuration) the dominant magnetic contribution at low temperatures can be attributed to EuO. With increasing temperature, the magneto-optical response of EuO diminishes and is exceeded by Co at about 55–60 K, close to the bulk Curie temperature of EuO, as indicated by the inversion of the hysteresis. The AFM coupling between transition metal and rare earth layers was already reported in the literature not only for EuO,[41] but also for EuS/Co.[34,36,38,42–44] It was attributed to the hybridization between the 3d states of the transition metal and the 5d states from the rare earth.[33,45]

The hysteresis loops of the bilayer measured in the P-MOKE geometry are shown in Figure 1d. Compared to the hysteresis of the reference EuO sample in Figure 1b, they show a vanishing remanence and a strongly increased saturation field, that varies between 2.8 T (at 5 K) and 1.8 T (at 55 K). This behavior indicates that the deposition of the Co layer induces a change in the magnetic anisotropy of the system. Above 55 K, the shape of the hysteresis does not change anymore, in agreement with our interpretation that above this temperature the signal mostly originates from the Co layer.

Recently, a magnetic proximity effect at the EuO/Co interface was reported in an XMCD study.[46] In the static hysteresis loops presented in Figure 1 it is hard to confirm this behavior besides the observed changes in magnetic anisotropy, since optical Kerr effect measurements lack element specificity. As we will see in the next section, measuring the transient magnetization after optical excitation in the P-MOKE configuration can instead provide further information about the proximity effect at the EuO/Co interface.

2.2. Transient Behavior

In order to disentangle the transient magnetic behavior of the coupled EuO/Co bilayer from the pristine EuO, we have performed time-resolved pump-probe experiments on both systems under different excitation conditions. The measurements were carried out in the P-MOKE configuration at 5 K with an applied field of $B_{ext} = 3.5$ T normal to the sample surface (z-direction), as schematically shown in **Figure 2**a. The probe wavelength was set to 800 nm, while the pump wavelength was varied from 750 nm ($\hbar\omega > E_g$) to 1700 nm ($\hbar\omega < E_g$) to excite the sample above and below the bandgap energy of EuO, respectively, as visualized in Figure 2b. By varying the pump photon energy, we can pinpoint the influence of the Co layer on the transient magnetization

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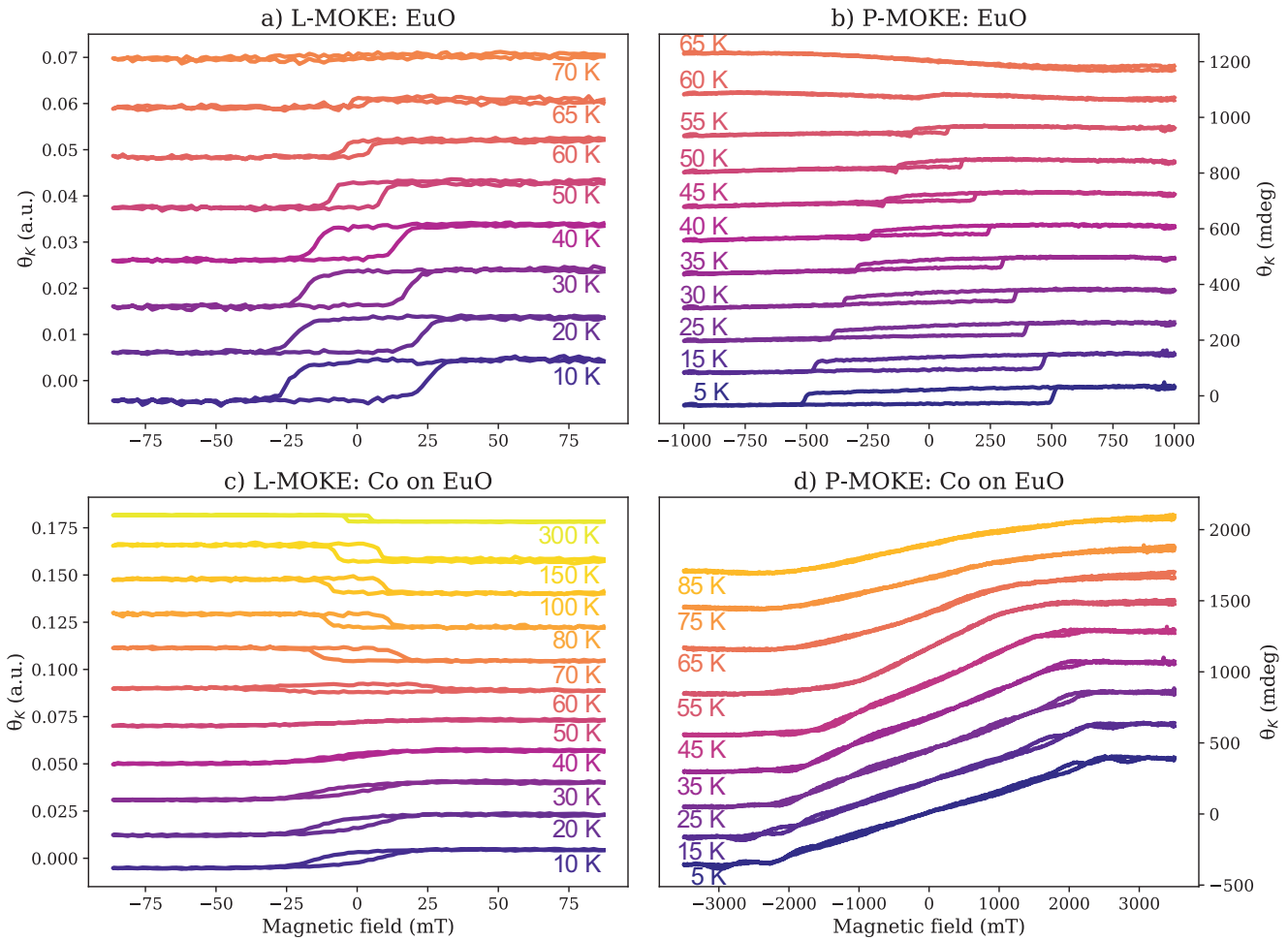

Figure 1. Hysteresis loops measurements of the europium monoxide (EuO) samples (a) and (b) and the EuO/Co bilayers (c) and (d). The measurements were performed in the L-MOKE and P-MOKE configuration, respectively. For a better visibility, the hysteresis loops are displayed with an offset. For the data measured in the L-MOKE configuration, the *y*-scale is arbitrary. A linear background was subtracted from the curves as described in Supporting Information.

behavior of the adjacent EuO. In the experiments, the laser pump fluence was kept fixed at 0.5 mJ cm^{-2}, whereas the probe fluence was <10 μJ cm^{-2}.

The blue trace in Figure 2c shows the data for $\hbar\omega > E_g$ on the EuO sample. The transient rotation of polarization is significantly increased on a short timescale (<100 ps) agreeing with the literature,[17] where the transient increase in the photoinduced magnetization in EuO has been attributed to the formation of magnetic polarons by the optical transition of 4f electrons into the 5d states, schematically represented in Figure 2b by the green arrow. The enhancement decays for increasing time delays but remains above equilibrium magnetization ($\Delta\theta_K = 0$) even after 1000 ps. The negative peak at zero delay is also visible in the transient reflectivity (not shown here) and is therefore most likely of optical and not magnetic origin. The measurement for $\hbar\omega < E_g$ (red trace) only shows a very weak signal, which is attributed to thermal heating induced by impulsive stimulated Raman scattering process,[47,48] active also for optical excitation below the band gap. We would like to stress that at this photon energy, no transient increase in the photoinduced magnetization is observed for pure EuO, since the 4f → 5d transition is suppressed (red arrow in Figure 2b).

The measurements performed on the EuO/Co bilayer using the same parameters are shown in Figure 2d. The blue trace shows the data for $\hbar\omega > E_g$, where we observe the enhancement of the magnetization of EuO, similar to the EuO single layer. Since the pump beam was already partially absorbed by the Co layer before reaching the EuO, the enhancement of magnetization is less pronounced. In addition, the signal switches its sign after ≈750 ps, which suggests the presence of a stronger demagnetization channel in EuO/Co with respect to EuO. The transient magnetic behavior of Co after optical excitation is well documented in literature as an ultrafast demagnetization within hundreds of femtoseconds and a subsequent remagnetization within a few picoseconds.[49–52] In our experiments, the ultrafast demagnetization of the Co layer is not resolved due to a step size of the delay between the pump and probe beam of 5 ps.

The measurement for $\hbar\omega < E_g$ on the bilayer is depicted by the red trace in Figure 2d. In this case, only the Co layer is excited, while EuO is mostly transparent at this photon energy,[53] as

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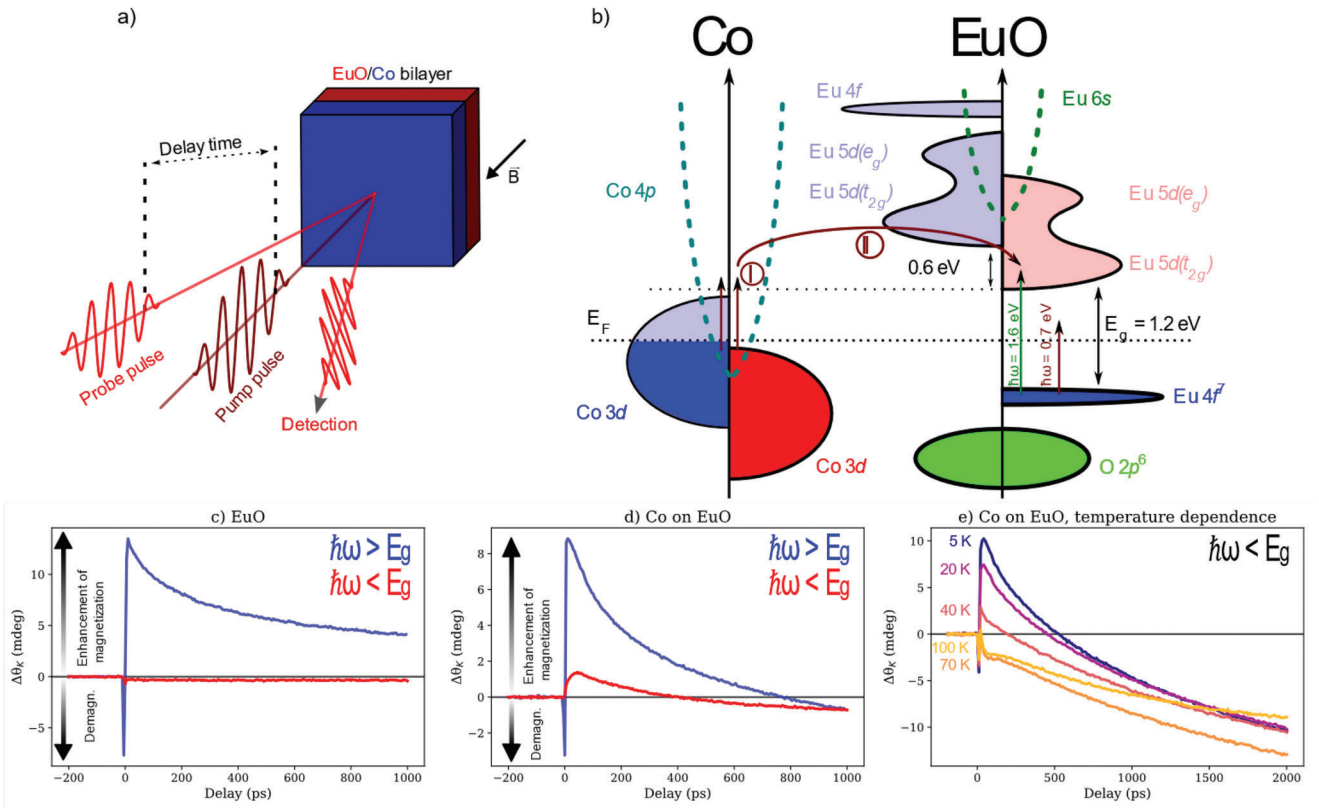

Figure 2. a) Schematic illustration of the measurement geometry, with the external field B applied along the *z*-direction. b) Schematic illustration of the density of states of europium monoxide (EuO) and Co and the excitation energies used in the described experiments. The 5d band of EuO can be populated either by photoexciting the sample above the EuO band gap (green arrow) or by photoexciting 3d electrons of the Co layer with a photon energy below the EuO band gap (dark red arrow). c,d) Pump probe measurements on c) the EuO sample and d) the EuO/Co bilayer. Measurements are performed with the pump wavelength set to $\hbar\omega > E_g$ (blue traces) and $\hbar\omega < E_g$ (red traces), respectively. The temperature was set to 5 K and a magnetic field of 3.5 T was applied normal to the sample surface. e) Pump probe measurements in temperature dependence on the EuO/Co bilayer.

confirmed by the measurement on the EuO sample in Figure 2c. In contrast to pure EuO, the EuO/Co sample shows a transient increase in the photoinduced magnetization even for excitation below the EuO band gap, which can be interpreted by assuming that there is a magnetic interaction at the EuO/Co interface. This could be either an indirect excitation of EuO (e.g., through heating) or a transfer of majority spins from Co to EuO. Since heating is expected to reduce the magnetization, our interpretation of the transient magnetization enhancement observed after exciting the EuO/Co system below the EuO band gap is that there is a transfer of optically excited majority electrons from cobalt into the (unoccupied) EuO 5d band. This process is schematically depicted in Figure 2b: the photon energy of 0.7 eV is too low to promote majority spins from the EuO 4f-states into the EuO 5d-states, but it is high enough to promote optically excited electrons in cobalt at an excited state energy corresponding to the position of the unoccupied 5d bands of EuO. Some of these electrons will travel from the cobalt into the EuO layer and populate the EuO 5d band. Although both minority and majority electrons are excited in this process, the electrons transferred to the EuO are expected to be mostly majority electrons due to their longer lifetime with respect to minority electrons.[54] This process has been described in the literature as superdiffusive spin current,[55] and was already observed in metallic bilayers.[56] Please note that superdiffusive spin currents are possible in our experiment because the magnetization vectors of the two layers are parallel to each other, since for $B_{ext} = 3.5$ T both M_{Co} and M_{EuO} are directed along the applied field in the *z*-direction. We thus argue that in our system, the majority electrons propagating as a spin current from Co into EuO have the same effect as direct optical excitation in EuO (4f → 5d), leading to a transient enhancement of the EuO magnetization.

Since the transient enhancement of magnetization in EuO/Co for excitation below the EuO band gap is the most interesting feature in our measurements, we performed additional measurements in the regime $\hbar\omega < E_g$ by keeping the pump fluence constant at 4 mJ cm^{-2} and changing the sample temperature between 5 and 100 K with $B_{ext} = 3.5$ T. The data is shown in Figure 2e. For temperatures up to 40 K, the enhancement of the EuO magnetization is still clearly detectable. For $T > T_C$ (EuO), where only the Co layer is ferromagnetically ordered, we observe a negative signal which persists well beyond the measured delay range (2000 ps). This behavior can be attributed to the heating of the thin Co layer, since it is deposited between two insulators (EuO and MgO). In other words, the cobalt layer deposited on top of EuO shows a transient demagnetization signal that persists for a timescale over 2000 ps: this behavior is crucial to understand the results in the next sections.

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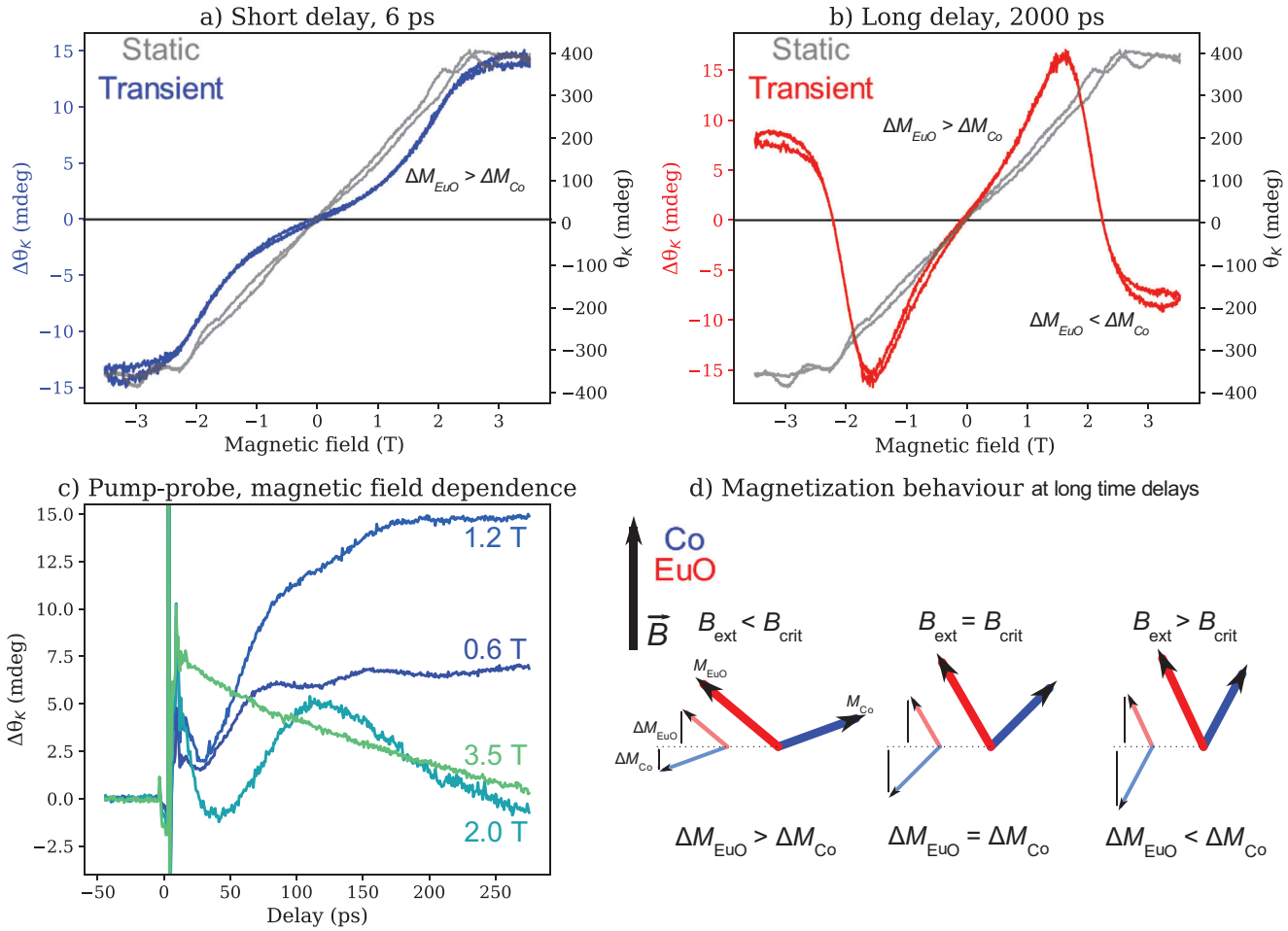

Figure 3. Transient hysteresis measurements on the europium monoxide (EuO)/Co bilayer for a short delay of 6 ps (a) and a long delay of 2000 ps (b). The pump photon energy is set to $\hbar\omega < E_g$ with a fluence of 5 mJ cm^{-2}. Measurements were performed at 5 K. The data plotted in gray depict a static hysteresis measured at the same temperature. c) Pump probe measurements performed on the EuO/Co bilayer in magnetic field dependence. The pump fluence was increased to 20 mJ cm^{-2}, the remaining parameters were kept the same. d) Schematic illustration of the behavior of the static and transient magnetization vectors of EuO (red) and Co (blue) in the EuO/Co bilayer. The static magnetization vectors (M_{Co}, M_{EuO}) rotate toward the magnetic field direction. Due to the different transient response of the EuO and the Co layers (enhancement versus demagnetization), the transient magnetization vectors (ΔM_{Co}, ΔM_{EuO}) are, respectively, parallel (ΔM_{EuO}) and antiparallel (ΔM_{Co},) to the static magnetization vectors.

2.3. Transient Hysteresis Loops

For a better understanding of the time-resolved magnetization behavior, we performed transient hysteresis measurements. We started by measuring both the transient and the static Kerr rotation, i.e.: ($\theta_K + \Delta\theta_K$) and ($\theta_K$), respectively. From these signals we extract the so-called transient hysteresis loops, which are described by the dependence of $\Delta\theta_K$ from the magnetic field, as shown schematically in Figure S2 (Supporting Information). This measured quantity is proportional to the transient magnetization ΔM, i.e., the pump induced variation of the static magnetization M along the z-direction. (We recall that the measurements are performed in the P-MOKE geometry, i.e., with the external field B_{ext} along the z-direction.) In the simplest case of a single magnetic layer, positive values of $\Delta\theta_K$ at positive magnetic fields indicate an amplified magnetization, while negative $\Delta\theta_K$ at positive fields can be attributed to a demagnetization. As the EuO/Co system consists of two layers, that are coupled antiferromagnetically in the ground state, the transient hysteresis loops will exhibit a complex behavior resulting from the different spin dynamics of the two materials and from the orientation of their magnetization in the external magnetic field. To disentangle the different contributions, we have recorded transient hysteresis loops at different time delays, where the EuO and Co layer are expected to follow intrinsically different transient behavior.

In **Figure 3**a,b we compare two transient hysteresis loops recorded at 6 ps (short delay, blue curve) and 2000 ps (long delay, red curve) to the static hysteresis (gray curve). The measurements were performed at 5 K with the pump fluence set to 5 mJ cm^{-2} and a photon energy of $\hbar\omega < E_g$ to only excite the Co layer. For the short delay the transient hysteresis is positive at positive magnetic fields, which is consistent with the enhanced magnetization at this timescale reported in Figure 2d. For long delays the transient rotation is also positive (for $B_{ext} > 0$), but starts to

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diminish upon passing a magnetic field of 1.6 T and switches its sign for $B_{ext} > 2.2$ T, in agreement with the measurements (for $\Delta t > 1000$ ps) shown in Figure 2d (red trace), that were performed with an applied field of 3.5 T.

We first analyze the behavior of the hysteresis recorded at short delays. At high magnetic fields ($B_{ext} > 2.8$ T), the static magnetization of both EuO and Co layers (M_{EuO}, M_{Co}) has reached its remanence value and is oriented parallel to B_{ext} along the z-axis. The transient magnetization signal is positive and maximum: it is dominated by the transient enhancement of the magnetization in EuO (ΔM_{EuO}). This signal is larger in absolute value than the transient demagnetization in the cobalt: $|\Delta M_{EuO}| > |\Delta M_{Co}|$. Since the transient magnetization follows the behavior of the static magnetization, we can conclude that it has mainly a longitudinal character (the changes in the magnetization are parallel to the magnetization itself). This behavior is consistent with a thermal demagnetization mechanism for the cobalt layer and with the already discussed optical enhancement of the EuO magnetization, i.e., the presence of a superdiffusive spin current of majority electrons from cobalt into EuO. At lower values of the external field, M_{EuO} and M_{Co} are not completely aligned along the field and optical excitation induces additional precession of the magnetization along the effective magnetic field (given by the external field and the anisotropy field).[57] This behavior is explicitly shown in Figure 3c, where it is also clear that the oscillation is suppressed when the external field is higher than the saturation threshold. Despite the oscillations, the transient magnetization signal is always positive, meaning that the longitudinal component of the transient magnetization dominates over the transverse component. We also note that the strength of the superdiffusive spin current from the Co layer into the EuO layer will also decrease with decreasing B_{ext}, and reach zero for $B_{ext} = 0$, where the magnetization vectors of the two layers are antiparallel to each other.

At long delays, the oscillations are already damped, and the signal is purely given by the longitudinal contribution. Moreover, on a timescale of 1000 ps the optical enhancement of the EuO magnetization (ΔM_{EuO}) is much weaker (Figure 2d), while the reduction of the cobalt magnetization (ΔM_{Co}) is stronger (Figure 2e): $|\Delta M_{EuO}| < |\Delta M_{Co}|$. At saturation, the negative contribution from the cobalt demagnetization dominates the transient signal, which is thus negative. At lower fields, the transient signal becomes positive, which can be easily understood by considering that the cobalt layer reaches saturation at higher fields than EuO: in other words, the angle between M_{EuO} and the external field is smaller than the one between M_{Co} and B_{ext}. Since that transient signal has mainly a longitudinal character, the same behavior can be postulated for the vectors ΔM_{EuO} and ΔM_{Co}. Depending on the value of B_{ext}, the sum between these two vectors will be positive or negative and will cross zero at a critical value of the external field $B_{ext,c}$, varying between 1 and 3 T depending on the time delay. This model is schematically depicted in Figure 3d.

2.4. Delay, Fluence, and Temperature Dependence

We now study the transient magnetic hysteresis of the EuO/Co system as a function of the delay between pump and probe, of the pump fluence and of the sample temperature. In a first set of measurements, the pump photon energy was set to $\hbar\omega < E_g$

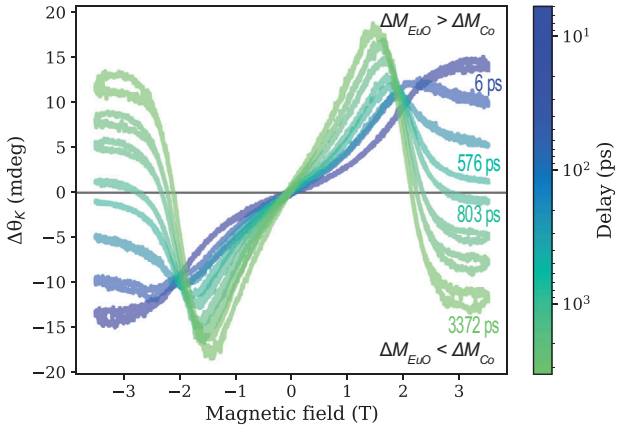

Figure 4. Transient hysteresis curves of the europium monoxide (EuO)/Co bilayer measured in pump probe delay dependence. The pump photon energy is set to $\hbar\omega < E_g$ with a fluence of 5 mJ cm^{-2}. The measurements are performed at a temperature of 5 K.

and transient hysteresis loops were recorded at various pump-probe delays (**Figure 4**), while the sample temperature was kept at 5 K and the pump fluence was set to 5 mJ cm^{-2}. We find that, at saturation, the transition between the short and long delay behavior reported in the previous section ($|\Delta M_{EuO}| > |\Delta M_{Co}|$ versus $|\Delta M_{EuO}| < |\Delta M_{Co}|$) takes place for delays between 576 and 803 ps. At such delays, the amplitude of the cobalt demagnetization can overcome the transient enhancement of the EuO.

Figure 5a,b, respectively, shows the fluence dependence measured at a temperature of 5 K, and the temperature dependence recorded for a pump fluence of 4 mJ cm^{-2}. The delay between pump and probe is set to 2000 ps in both measurements (long delay regime). Increasing the pump fluence increases the amplitude of the spin current diffusing from the optically excited cobalt layer into EuO. This results in an amplification of the positive transient signal in Figure 5a for an increasing pump fluence. At the same time, increasing the pump fluence induces a stronger demagnetization in the cobalt layer (due to heat accumulation in the cobalt layer). At saturation, this leads to a stronger negative signal for higher pump fluence. The temperature dependence shown in Figure 5b gives information about the influence of the EuO layer on the transient dynamics. We tune the temperature in the range from 5 to 60 K, a range that is expected to only affect EuO, as T_C of Co is significantly higher. We see that for $T > 50$ K the transient magnetization does not change sign anymore for $B_{ext} > 0$, as a result of the decrease of the EuO transient signal for increasing temperature.

The measurements presented in Figures 4 and 5 show that the sign of the transient magnetization, that results from the interplay between the cobalt demagnetization and the enhancement of the EuO magnetization, can be modified by varying various parameters: the delay between the pump and probe beam, the sample temperature and the value of the field B_{ext} applied along the z-direction. These parameters provide the possibility to specifically select which transient magnetization signal of the EuO/Co bilayer is dominant and thus to tune the magnitude as well as the sign of the signal.

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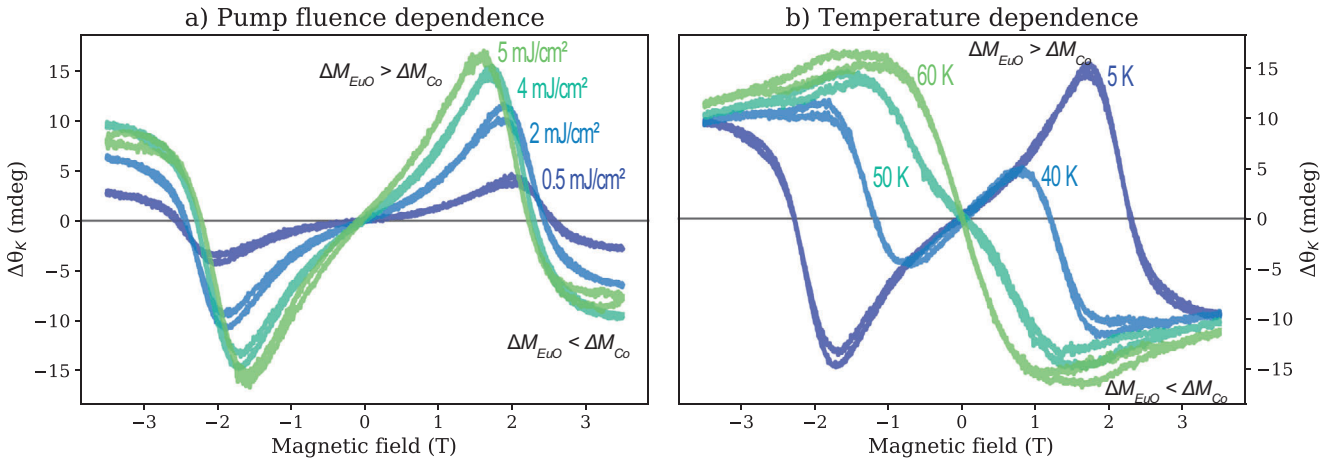

Figure 5. Transient hysteresis curves of the europium monoxide (EuO)/Co bilayer measured in a) pump fluence dependence and b) temperature dependence at a delay of 2000 ps. In both cases the pump photon energy is set to $\hbar\omega < E_g$. a) The temperature is kept at 5 K, while for b) the fluence is set to 4 mJ cm^{-2}.

2.5. Strong Excitation Regime

A final set of measurement has been performed with a pump fluence of 20 mJ cm^{-2} to explore the behavior of the system in the strong excitation regime. The data for the transient hysteresis measurement performed at 5 K with a delay of 2.1 ps is depicted in **Figure 6**a. At remanence, for positive fields, the signal is positive, similar to lower excitation fluence (Figure 3a). However, decreasing B_{ext} we observe a zero-crossing and then a negative transient magnetization. This means that for low values of B_{ext}, the transient signal is dominated by the cobalt demagnetization. In Figure 6b we subtract this contribution from the signal; the result is a hysteresis which is open around $B_{ext} = 0$ T. Crucially, this hysteresis can be attributed to the EuO layer, since the cobalt does not show any remanence in the polar configuration (Figure 1d).

The effect of the strong optical excitation seems to be twofold: it suppresses the magnetization of the cobalt, while at the same time it also reduces its influence on the anisotropy of the EuO (please recall that the presence of the cobalt strongly increases the out-of-plane saturation magnetization in the static hysteresis loops shown in Figure 1d), that shows again an open hysteresis in the P-MOKE configuration. Under these conditions it is thus possible to measure the magnetic hysteresis of the underlying EuO layer. Strikingly, the signal is present up to room temperature. Since the superdiffusive spin current signal should vanish at $B_{ext} = 0$ T (where the magnetizations of the two layers are antiparallel), this signal can be taken as experimental evidence of the presence of a magnetic proximity coupling between Co and EuO, with a concomitant increase in the Curie temperature of the EuO surface in contact with the cobalt layer. Further studies

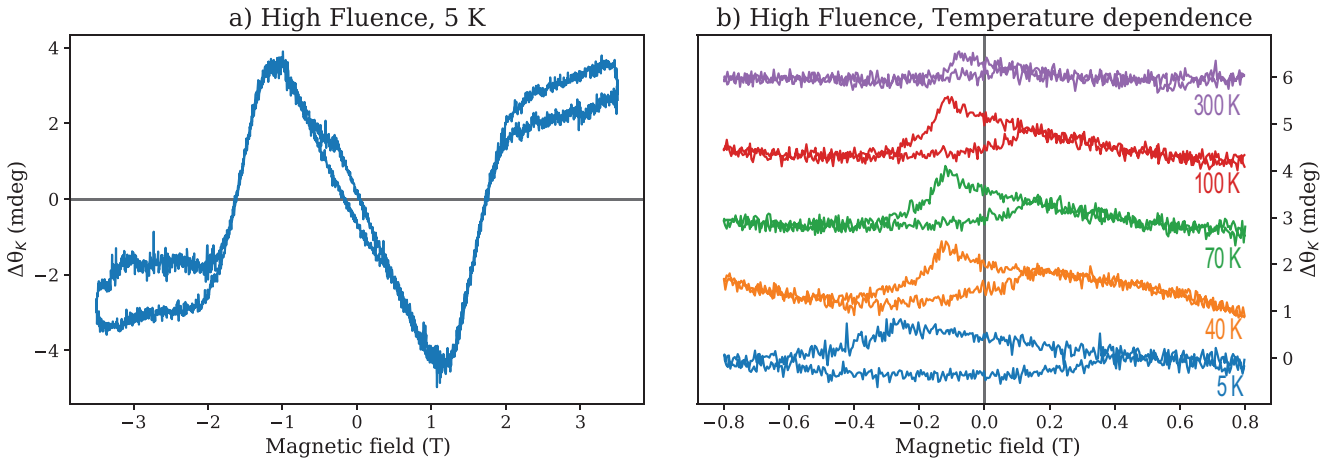

Figure 6. a) Transient hysteresis curves of the europium monoxide (EuO)/Co bilayer at 5 K with the pump photon energy set to $\hbar\omega < E_g$ with a fluence of 20 mJ cm^{-2}. The hysteresis opens up, implying a break of the coupling between the two layers which allows access to the isolated EuO magnetization. b) The same measurement was performed in temperature dependence. The opening of the hysteresis persists up to room temperature. In both figures, the delay was set to 2.1 ps.

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should be performed in order to determine if the proximity coupling affects the whole EuO layer or only the EuO/Co interface.

3. Conclusion

In summary, we examined the static and time-resolved magneto-optical response of a Co/EuO bilayer. Static in-plane hysteresis measurements show an antiferromagnetic coupling between the two layers. Pump-probe measurements performed in the P-MOKE geometry show an intriguing behavior. We found that the magnetization of EuO can be transiently excited by selectively pumping the cobalt layer, which we assign to the presence of a superdiffusive spin current of majority spins from Co into EuO. This transient magnetization enhancement is counterbalanced by a negative transient magnetization signal from the cobalt layer, that is thermally demagnetized by optical excitation. The interplay between these two signals provides a controlled way of selecting a dominant transient contribution from the two layers by varying parameters such as the pump-probe delay, temperature, magnetic field, delay, and pump fluence. Transient hysteresis measurements with fluences of 20 mJ cm^{-2} at a short delay of 2.1 ps provide the intriguing possibility to quench the Co magnetization and thereby allow access to the EuO magnetization. Temperature-dependent measurements in this regime reveal a transient contribution to the magnetic signal from EuO well above its magnetic ordering temperature — persisting even up to room temperature, which we ascribed to a magnetic proximity effect at the EuO/Co interface.

4. Experimental Section

Samples: A Co/EuO bilayer and a bare EuO reference film were deposited on double-side polished transparent yttria-stabilized zirconia (YSZ(001)) substrates (Crystec GmbH) using molecular beam epitaxy (MBE). YSZ is regarded as the best suited substrate for high-quality EuO thin film growth since its lattice constant (≈5.14 Å) almost perfectly matches that of EuO (5.142 Å).[58–60] We synthesized a 5 nm thick EuO film on YSZ applying the established MBE-based adsorption limited growth mode as described elsewhere.[61,62] The Eu deposition rate was $r = 0.13$ Å s^{-1} and the oxygen partial pressure was $p_{O2} = 1.5 \times 10^{-8}$ mbar. In situ X-ray photoelectron spectroscopy (XPS) was employed to confirm the chemical composition of EuO, a crucial procedure as EuO is metastable, highly reactive and will lose its properties if overoxidized.[62] A 4 nm thick Co overlayer was subsequently deposited onto the EuO film by e-beam evaporation at room temperature. XPS analysis of a similar sample reveals that EuO remains stoichiometric upon Co deposition (shown in Figure S3, Supporting Information). In the same way as the Co overlayer, a 24 nm thick MgO capping layer was deposited, preventing EuO (and Co) from oxidizing during ex situ handling.[63]

Setup: Static magnetic hysteresis measurements were performed in polar (P-MOKE) and longitudinal (L-MOKE) configurations to characterize the magnetic ground state. A positive magnetic field was always used as starting point for the hysteresis. The field was also applied while increasing the temperature in the temperature-dependent measurements. The magnetic field is applied parallel and perpendicular to the EuO [100] direction in the L- and P-MOKE configuration, respectively. The L-MOKE measurements were performed with 3 eV optical probe. Time-resolved measurements were carried out in P-MOKE configuration in a pump-probe setup with an ytterbium-based femtosecond laser (20 W average output power, 100 kHz pump repetition rate) driving two optical parametric amplifiers (OPAs), which allow for independent tuning of the photon energy of both pump and probe beams in the range between 0.5 and 3.5 eV, i.e., below and above the bandgap of EuO. Both static and time-resolved P-MOKE experiments were performed with a 1.55 eV probe. We used a helium flow cryostat to measure temperature dependent between 5 and 300 K with magnetic fields applied up to ±3.5 T. Further details on this setup can be found in literature.[49,64]

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The raw data that support the findings of this study are available in the ZENODO database, at https://zenodo.org/communities/interfast-fetopen.

Keywords

semiconducting ferromagnets, ultrafast spin dynamics

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