Direct optical probing of ultrafast spin dynamics in a magnetic semiconductor

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We uncovered the spin dynamics involved in the birth and growth of giant spin polarons in a magnetic semiconductor. For this purpose, we developed a new measurement technique, which provides direct access to the spin dynamics, irrespective of phonons and carriers involved in the process. Moreover, we solved the Landau-Lifshitz equation in the specific scenario of spin polarons, which fits our data excellently, and demonstrates that the spin polaron growth slows down dramatically when the sample is cooled in the paramagnetic phase. Finally, temperature dependent Monte Carlo simulations were performed, which are in excellent agreement with the observed slowdown, which demonstrates that fluctuations in the Weiss field play a decisive role in spin coherence generation induced by light in magnetic materials. These results offer a new tool and new insight for spin dynamics investigations.

The control of the magnetic state of matter with light is a topic of vast current technological and scientific interest [1–4]. Understanding the physics behind light-induced magnetization is essential to develop more efficient magneto-optical devices. Previous magnetization dynamics investigations focused mainly on demagnetization driven by the heat generated by the pump pulse [5–8], as well as on some limited non-thermal mechanisms based on e.g. photoinduced changes in magnetic anisotropy [9–11] and coherent stimulated Raman scattering [12].

Recently, we reported the discovery of efficient non-thermal light-induced magnetization generation in intrinsic magnetic semiconductors, based on the photoexcitation of very large spin polarons (SPs), i.e. bound electron-hole pairs forming excitons, within which thousands of magnetic atoms are forced into ferromagnetic alignment due to the band-lattice exchange interaction [13–15]. Here we extract for the first time the spin dynamics involved in the birth and growth giant SPs. For this purpose, we developed a new technique, which extracts spin dynamics directly from measurements, irrespective of the phonon and carrier subsystems. Until now, extracting spin dynamics from transient optical measurements demanded a model involving all three interacting subsystems perturbed by the incident light: carriers, phonons, and spins. Because our technique provides direct access to spin dynamics, irrespective of carriers and phonons, it uncovers the spin dynamics unequivocally, and opens a new avenue for the investigation of light-induced magnetism. Moreover, the experimental results were analyzed in the frame of the Landau-Lifshitz equation, which was solved in the specific scenario of SP generation, yielding the SP growth rate and fully-grown state. Additionally, we demonstrate the crucial role played by Weiss field fluctuations in the spin dynamics. Due to the high interest in the control of spin phenomena using light, our results will attract the attention of a broad audience.

The EuSe samples were grown by molecular beam epitaxy onto (111) BaF2 substrates at the Johannes Kepler Universität Linz. Because of the almost perfect lattice constant matching (a = 6.191 Å and a = 6.196 Å for EuSe and BaF2, respectively), nearly unstrained bulklike EuSe reference layers with µm thickness were obtained by direct growth on BaF2. Results presented here were obtained on an EuSe epilayer of thickness 0.34 µm. The time-resolved photoinduced Faraday rotation (TRFR) was measured at the LMO of the University of São Paulo, using a two-color pump-probe technique. We used 100 fs light pulses, tuned to a repetition rate of 25 kHz, produced by a cavity-dumped Ti:Sapphire mode-locked laser coupled to a second harmonic generator. The pump photon energy was 3.1 eV, which is above the EuSe band gap of 2.0 eV, therefore exciting electron-hole pairs that form supergiant SPs [13, 17], whereas the probe photon energy was 1.55 eV, which is in the transparency range of EuSe, but sufficiently close for efficient Faraday rotation detection of magnetization [18]. The absorption coefficient for EuSe at the 3.1 eV excitation energy is greater than 20 µm−1 [19], hence the penetration depth of the excitation light is only 100 nm or less. The delay between the pump and probe pulses was controlled by a 0.6 m delay line. The Faraday rotation angle of the probe pulse was measured using autobalanced detection with a resolution better than 0.1 µrad. The image of the excitation spot on the sample had a diameter of 150 µm, about twice the diameter of the probe spot. All measurements were performed using an optical cryostat containing a superconducting coil, applied in the Faraday geometry. Measurements were taken above the Néel temperature, when EuSe is in the paramagnetic phase and the equilibrium magnetization is zero. A remarkable feature of the magnetization generated by SPs is that it is triggered by very low intensity light, which offers the advantage of investigating spin dynamics under minimal thermal perturbation. Our investigation was performed using extremely low fluence light pulses, of about 1–10 µJ/cm², which is several orders of magnitude less than employed in the investigations by other authors.

Figure 1 shows a delay scan of the TRFR for fluences of 1 and 10 µJ/cm². As demonstrated in Ref. [13] the Faraday rotation signal is due to photoexcited supergiant spin polarons (SPs). The time interval between pulses, 40 µs, is much longer than the SP lifetime of about 1 µs [13], therefore al-
photoinduced Faraday rotation (mrad) at very high densities to completely fill the illuminated volume, opening the perspective of achieving a large magnetization with low intensity light. The population of SPs generated at $T = 5$ K and a magnetic field of 60 mT.

For 1 $\mu$J/cm$^2$ pulses, the shape of the TRFR displays exponential-like growth after the arrival of the pump pulse at $t = 0$. As pulse fluence increases, a cusp begins to form immediately after $t = 0$, becoming very prominent for 10 $\mu$J/cm$^2$ pulses. The emergence of a cusp when the pump energy increases can be qualitatively explained by the fact that the pump pulse perturbs three subsystems - charge carriers, phonons, and lattice spins. The pump light generates electrons in excited conduction band states; as the electrons relax their energy and form SPs, heat is transferred to the phonon and spin baths, increasing the lattice temperature, which reduces the magnetic susceptibility, and consequently the photoinduced magnetization signal decreases, producing the cusp. As the excitation fluence is increased, more heat is generated, leading to a more prominent cusp in the delay scan. This demonstrates that the delay scan shown in Fig. 1 reflects simultaneous changes in the interacting subsystems, hence extraction of spin dynamics from such a delay scan mandates the assumption of an underlying model involving all three subsystems, which is the approach used by many researchers [5][7][20].

In this report, we circumvent the requirement of such a three-subsystem model, and extract the time evolution of the spin subsystem independently of the carrier and phonon subsystems. This is achieved by measuring the TRFR as a function of magnetic field at constant time delays, from which we then extract spin dynamics associated with SP generation. At each time delay the state of the carrier, phonon and spin subsystems remain frozen, and TRFR magnetic field scans reveal the state of the spin subsystem at that particular instant of time. The magnetic field scans at fixed delays covered only a fraction of a Tesla, which is sufficient to polarize the SPs, in view of their very large magnetic moment, but has virtually no direct effect on individual Eu spin alignment.

To minimize heating effects, we used a pump fluence of 1 $\mu$J/cm$^2$ for the remainder of this work. Figure 2 shows typical magnetic field scans, taken for various time delays at $T = 8$ K.

For the low excitation levels used in this work, the photoinduced SPs are non-interacting, forming a superparamagnetic phase [21]. The absolute value of the slope of the linear term also grows with delay, indicating warming up of the SP volume, which occurs as the extra energy of excited carriers is transferred to the lattice. Here we shall focus on

\begin{equation}
\Delta \theta_F = \Delta \theta_F^{\text{SAT}} L \left( \frac{\mu_\text{SP} B_{\text{int}}}{k_b T} \right) + \text{linear term}
\end{equation}

where $\Delta \theta_F^{\text{SAT}}$ is the Faraday rotation due to a fully polarized SP gas, $\mu_\text{SP}$ is the SP magnetic moment, $B_{\text{int}}$ is the internal magnetic field, and $T$ is the temperature of the lattice within the SP. The linear term in (1) describes the heating caused by the pump pulse, as described in detail in Ref. [21]. Solid lines in Fig. 2 represent the theoretical fitting of the experimental data, using equation (1). It can be seen that formula (1) fits perfectly to the experimental data for all delays. From the fitted curves we extract, at each delay, the amplitude of the magnetization curve, $\Delta \theta_F^{\text{SAT}}$, the magnitude of the SP magnetic moment, $\mu_\text{SP}$, and the slope of the linear term. The linear term has a negative slope, exactly as expected when the semiconductor is in the paramagnetic phase [21]. The absolute value of the slope of the linear term also grows with delay, indicating warming up of the SP volume, which occurs as the extra energy of excited carriers is transferred to the lattice. Here we shall focus on

![FIG. 1. Time-resolved Faraday rotation in EuSe for different fluences at $T = 5$ K and a magnetic field of 60 mT.](image1)

![FIG. 2. Field scans at various fixed time delays.](image2)
The mathematical connection between spin dynamics and the growth of the magnetic moment of SPs is provided by the Landau-Lifshitz equation for the expectation value of a lattice spin vector, \( S \),

\[
\frac{dS}{dt} = \omega \times S + \frac{\alpha}{S} S \times (\omega \times S),
\]

where \( \omega = g \mu_B B \), \( g = 2 \) is the gyromagnetic factor of an europium atom in the lattice, \( B \) is the effective magnetic field acting on the Eu atom, and \( \alpha \) is a dimensionless coefficient associated with the directional relaxation of the spin vector \( S \). The first term in (2) describes the precession of \( S \) around \( B \), and follows from the Schrödinger equation with Zeeman interaction. The second term is phenomenological, corresponding to the directional relaxation of \( S \) towards \( B \), with conservation of the absolute value of \( S \) (see, for instance, Ref. [22]).

If \( B \) is constant, then the solution of (2), averaged over random initial orientations that characterize an Eu spin in the paramagnetic phase, is given by

\[
\bar{S}_z = S \tanh \left( \frac{t}{2T_1} \right), \quad \bar{S}_x = \bar{S}_y = 0,
\]

where \( T_1 = 1/\alpha \omega \) is the characteristic time for the directional relaxation of the Eu spins towards \( B \), and \( z \) is the direction of \( B \).

When an electron is photoexcited from the localized \( 4f^7 (^{9}S_7/2) \) valence state of an Eu atom by an absorbed photon, the electron is promoted to a conduction band state, which overlaps with many lattice sites, and therefore it suddenly generates an exchange field, \( B_{ex} \), acting on the surrounding Eu atoms \([15, 16, 23, 25]\). The average value within the SP sphere, \( B_{ex} \), is about 0.7 Tesla for EuSe \([15, 26]\). It is this field that polarizes the lattice spins, forming an SP. Therefore the exchange field entering (2) can be approximated by \( B_{ex} \). From (3), the growth of the SP magnetic moment is then given by

\[
\mu_{pol}(t) = \mu_{pol}^{fg} \tanh \left( \frac{t}{2T_1} \right),
\]

which indicates that \( T_1 \) is also the characteristic time for SP growth.

The solid lines in Fig. 3 indicate a fit of (4) to the experimental data. A very good fit is found, although a slight deviation is observed at \( T = 5 \) K, probably because it is too close to the EuSe Néel temperature \( (T_c=4.8 \) K \([13]\), leading to some
correlations between spins and invalidating the assumption of ideal paramagnetic order.

Figure 5 displays the extracted rise time \(T_1\) as a function of temperature, showing that the SP growth slows down dramatically as the sample is cooled towards the Néel temperature. We attribute this slowdown to fluctuations of the Weiss exchange field, as will be shown henceforward.

Directional spin relaxation towards \(B\) can be brought about by a transverse oscillating magnetic field, as in nuclear magnetic resonance, in which case the relaxation rate is proportional to the squared amplitude of the oscillating magnetic field (see, for instance, Ref. 27). In the case of a magnetic material, such as EuSe, fluctuations in Weiss magnetic field can similarly assist the directional relaxation of \(S\) towards \(B\). Since the amplitude of fluctuations decreases as temperature approaches \(T_0\), the relaxation rate assisted by Weiss field fluctuations also decreases, which explains qualitatively the slowdown of the SP growth seen in Fig. 5.

For a quantitative analysis, we shall use the formula connecting the relaxation rate, \(T_1\), and Weiss field fluctuations, \(\delta B_\perp\), described in the book by A. P. Guimarães (Ref. 28):

\[
T_1 = \frac{1 + \omega_0^2 \tau_0^2}{\gamma^2 \tau_0 \delta B_\perp^2},
\]

where \(\gamma = \mu_s / \hbar\), \(\mu_s = g\mu_B S\) is the magnetic moment of an Eu atom, \(g = 2\), \(S = \frac{7}{2}\), \(\omega_0 = \gamma B_\perp\), and \(\tau_0\) is the typical interval of time for which the Weiss field remains constant. We calculated the amplitude of fluctuation of the Weiss field, \(\delta B_\perp\), as a function of temperature using Monte Carlo calculations, fully described in Refs. 17, 29, and 30 and used \(\tau_0\) as an adjustable parameter in Fig. 5 to fit the experimental values of \(T_1\). As depicted in Fig. 5, a good fit of the data is found assuming a temperature-independent value \(\tau_0 = 3\) fs, which is of the same order of magnitude as the spin correlation time found in other magnetic materials 31. As temperature increases, the calculated \(T_1\) flattens compared to a slight, continued decrease of the experimental curve, which suggests that \(\tau_0\) decreases.

In conclusion, we demonstrated a novel method to extract spin dynamics, which does not require modeling of the phonon and carrier subsystems, which obviously makes the spin dynamics extracted more accurate. We combined our new method with our solution of the Landau-Lifshitz equation to describe spin dynamics, and measured the growth rate and fully-grown size of supergiant spin polarons in EuSe. We demonstrated that in the paramagnetic phase the spin dynamics depends dramatically on temperature. Using Monte Carlo simulations we demonstrate the vital role played by Weiss field fluctuations. The connection between Weiss field fluctuation and spin dynamics discovered by us offers new into the optical control of spin coherence. These results offer a new efficient tool and new insight for spin dynamics investigations in magnetic materials.

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[1] A. Kirilyuk, A. V. Kimel, and T. Rasing, Rev. Mod. Phys. 82, 2731 (2010).
[2] P. Nemec, M. Fiebig, T. Kampfrath, and A. V. Kimel, Nature Physics 14, 229 (2018).
[3] F. Siegrist, J. A. Gessner, M. Ossiander, C. Denker, Y.-P. Chang, M. C. Schröder, A. Guggenemos, Y. Cui, J. Walowski, U. Martens, J. K. Dewhurst, U. Kleineberg, M. Münzenberg, S. Sharma, and M. Schultz, Nature 571, 240 (2019).
[4] A. V. Kimel and M. Li, Writing magnetic memory with ultrashort light pulses, Nat. Rev. Mater. 4, 189 (2019).
[5] E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, Ultrafast spin dynamics in ferromagnetic nickel, Phys. Rev. Lett. 76, 4250 (1996).
[6] C. D. Stanciu, F. Hansteen, A. V. Kimel, A. Kirilyuk, A. Tsukamoto, A. Itoh, and T. Rasing, Phys. Rev. Lett. 99, 047601 (2007).
[7] B. Koopmans, G. Malinowski, F. D. Longa, D. Steiauf, M. Fähnle, T. Roth, M. Cinchetti, and M. Aeschlimann, Nature Materials 9, 259 (2010).
[8] I. A. Dolgikh, F. Formisano, K. H. Prabhapakara, M. V. Logunov, A. K. Zvezdin, P. C. M. Christianen, and A. V. Kimel, Appl. Phys. Lett. 120, 012401 (2022).
[9] N. P. Duong, T. Satoh, and M. Fiebig, Ultrafast manipulation of antiferromagnetism of NiO, Phys. Rev. Lett. 93, 117402 (2004).
[10] F. Hansteen, A. V. Kimel, A. Kirilyuk, and T. Rasing, Femtosecond photomagnetic switching of spins in ferrimagnetic garnet films, Phys. Rev. Lett. 95, 047402 (2005).
[11] A. Stupakiewicz, K. Szerenos, D. Afanasiev, A. Kirilyuk, and A. V. Kimel, Ultrafast nonthermal photo-magnetic recording in a transparent medium, Nature 542, 71 (2017).
[12] A. V. Kimel, A. Kirilyuk, P. A. Usachev, R. V. Pisarev, A. M. Balbashov, and T. Rasing, Ultrafast non-thermal control of magnetization by instantaneous photomagnetic pulses, Nature
[13] A. B. Henriques, X. Gratens, P. A. Usachev, V. A. Chitta, and G. Springholz, Phys. Rev. Lett. **120**, 217203 (2018).

[14] A. B. Henriques, G. D. Galgano, P. H. O. Rapp, and E. Abramof, Phys. Rev. B **93**, 201201(R) (2016).

[15] A. B. Henriques, F. C. D. Moraes, G. D. Galgano, A. J. Meaney, P. C. M. Christianen, J. C. Maan, E. Abramof, and P. H. O. Rapp, Phys. Rev. B **90**, 165202 (2014).

[16] A. B. Henriques, P. H. O. Rapp, and E. Abramof, Phys. Rev. B **80**, 245206 (2009).

[17] A. B. Henriques, S. C. P. van Kooten, E. Abramof, P. H. O. Rapp, and G. D. Galgano, J. Appl. Phys. **131**, 043903 (2022).

[18] S. C. P. van Kooten, P. A. Usachev, X. Gratens, A. R. Naupa, V. A. Chitta, G. Springholz, and A. B. Henriques, J. Appl. Phys. **126**, 095701 (2019).

[19] A. B. Henriques, A. Wierts, M. A. Manfrini, G. Springholz, P. H. O. Rapp, E. Abramof, and A. Y. Ueta, Phys. Rev. B **72**, 155337 (2005).

[20] M. Battiato, K. Carva, and P. M. Oppeneer, Superdiffusive spin transport as a mechanism of ultrafast demagnetization, Phys. Rev. Lett. **105**, 027203 (2010).

[21] A. B. Henriques, A. R. Naupa, P. A. Usachev, V. V. Pavlov, P. H. O. Rapp, and E. Abramof, Phys. Rev. B **95**, 045205 (2017).

[22] A. G. Gurevich and G. A. Melkov, *Magnetization Oscillations and Waves* (CRC Press Inc, New York, 1996) Chap. 7.

[23] A. B. Henriques, M. A. Manfrini, P. H. O. Rapp, and E. Abramof, Phys. Rev. B **77**, 035204 (2008).

[24] B. Kaminski, M. Lafrentz, R. V. Pisarev, D. R. Yakovlev, V. V. Pavlov, V. A. Lukoshkin, A. B. Henriques, G. Springholz, G. Bauer, E. Abramof, P. H. O. Rapp, and M. Bayer, Phys. Rev. Lett. **99**, 091906 (2011).

[25] A. B. Henriques, G. D. Galgano, B. L. Diaz, P. H. O. Rapp, and E. Abramof, J. Phys.: Condens. Matter **19**, 406234 (2007).

[26] A. B. Henriques, G. D. Galgano, E. Abramof, B. Diaz, and P. H. O. Rapp, Appl. Phys. Lett. **99**, 091906 (2011).

[27] C. Kittel, *Introduction to solid state physics*, seventh ed. (John Wiley & Sons, New York, 1996) p. 389, formula (22).

[28] A. P. Guimaraes, *Magnetism and Magnetic Resonance in Solids* (John Wiley, New York, 1998) Chap. 7.

[29] X. Gratens, Y. Ou, J. S. Moodera, P. H. O. Rapp, and A. B. Henriques, App. Phys. Lett. **116**, 152402 (2020).

[30] S. C. P. van Kooten, X. Gratens, and A. B. Henriques, Phys. Rev. B **103**, 035202 (2021).

[31] T. S. Seifert, S. Jaiswal, J. Barker, S. T. Weber, I. Razdolski, J. Cramer, O. Gueckstock, S. F. Maehreim, L. Nadvornik, S. Watanabe, C. Ciccarelli, A. Melnikov, G. Jakob, M. Münzenberg, S. T. B. Goennenwein, G. Woltersdorf, B. Rethfeld, P. W. Brouwer, M. Wolf, M. Kläui, and T. Kampfrath, Nat. Commun. **9**, 2899 (2018).