AIP Advances

RESEARCH ARTICLE | FEBRUARY 02 2024

Experimental evidence for ultrashort-lived spin polarons in EuSe

Special Collection: 68th Annual Conference on Magnetism and Magnetic Materials

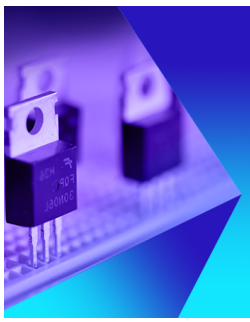
S. C. P. van Kooten ; A. B. Henriques

Check for updates

AIP Advances 14, 025006 (2024)
https://doi.org/10.1063/9.0000820

View Online
Export Citation
CrossMark

18 March 2024 16:13:53



AIP Advances
Special Topic: Field Theory Methods in Condensed Matter Physics for Future Post-Transistor Devices
Submit Today
AIP Publishing

AIP Publishing

Experimental evidence for ultrashort-lived spin polarons in EuSe

Cite as: AIP Advances 14, 025006 (2024); doi: 10.1063/9.0000820
Submitted: 4 October 2023 • Accepted: 5 December 2023 •
Published Online: 2 February 2024

View Online Export Citation CrossMark

S. C. P. van Kooten[a] and A. B. Henriques

AFFILIATIONS

Laboratório de Magneto-Óptica—LMO, Instituto de Física, Universidade de São Paulo, 05508-090 São Paulo, Brazil

Note: This paper was presented at the 68th Annual Conference on Magnetism and Magnetic Materials.
[a]**Author to whom correspondence should be addressed:** sjoerd@if.usp.br

ABSTRACT

We investigated the time evolution of the density of giant spin polarons in a magnetic semiconductor. Spin polarons (SPs) were photoexcited and observed using time-resolved Faraday rotation. We find the existence of two types of SPs, a short-lived spin polaron with a lifetime of around τ_{sl} = 0.5 ns and a long-lived spin polaron with a lifetime of τ_{ll} = 0.45 ± 0.03 μs, at T = 5 K. The stark difference of three order of magnitude between these lifetimes suggests that in the long lived SP the electron-hole pair is relaxed and its recombination is forbidden. The short-lived SP is probably associated with SP recombination before such relaxation has occured. An extraordinary finding is that the magnitude of τ_{sl}, as well as its decrease with increasing temperature, reproduces exactly the characteristic time for SP growth. This suggests that the thermal fluctuations, responsible for SP magnetic moment growth, are also responsible for increasing the recombination probability of SPs.

© 2024 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). https://doi.org/10.1063/9.0000820

18 March 2024 16:13:53

I. INTRODUCTION

Controlling the magnetic state of matter by light has emerged as one of the most promising concepts in novel magnetic devices.[1,2] Compared to traditional control through the application of a magnetic field, magneto-optic processes occur on shorter timescales, creating a straightforward mechanism for faster magnetic control. In this work, we will discuss the time-resolved dynamics involved in the photoexcitation of spin polarons (SPs), a recently discovered, energy-efficient method for optically inducing magnetization in magnetic semiconductors.[3–6]

We will discuss EuSe, in which SPs are formed due to a large increase in direct exchange interaction between a (photo)excited electron and surrounding lattice spins, compared to an electron in the ground state. This leads to spherical regions of enhanced ferromagnetic interaction centered around the excited electrons, which is sufficiently strong to overcome otherwise antiferro-, ferri- or paramagnetic ordering, or enhance already existing ferromagnetism.

The magnetic moment of such SPs can reach very large sizes, up to 6000 Bohr magnetons in EuSe[5] and 20 000 Bohr magnetons in EuS.[7] However, the exact nature of the SP state is not well-described and in this work we further investigate the SP state by investigating the formation and decay dynamics of SPs using time-resolved Faraday rotation measurements.

II. METHODS

We used two-color pump-probe spectroscopy to measure the photoinduced time-resolved Faraday rotation (TRFR) in EuSe. Light pulses with energy of 3.1 eV, which is above the 2.0 eV bandgap of EuSe,[5,8] were used as pump pulses to photoexcite SPs. A probe pulse with energy of 1.5 eV, within the EuSe bandgap, was then used to obtain the magnetization through Faraday rotation,[9] where the rotation angle was measured using an autobalanced photodetector.[10] Repeating the measurement and varying the time in-between pump and probe pulse then allows us to probe the magnetic state at different instants after excitation, with a resolution of around 100 fs, which is the pulse duration at the sample. Furthermore, the pump beam was modulated by an optical chopper, and the probe signal was passed through a lock-in amplifier referenced at the chopper frequency to extract only the photoinduced magnetization associated with SPs. All measurements were performed above the Néel temperature in the paramagnetic (PM) phase of EuSe. In this phase, the SPs

© Author(s) 2024

will initially be oriented randomly, and a magnetic field was applied in the Faraday geometry to align the SPs and obtain a macroscopic magnetization. Throughout this work, we used very low fluence excitations of 1 μJ/cm^2, several orders of magnitude less than what is common for other ultrafast magneto-optic techniques.[1,11–14] Experimental conditions were equivalent to those in Ref. 6, which we refer to for additional details.

Figure 1 shows a delay scan of the photoinduced magnetization ΔM, where a magnetic field of 60 mT was used to align the SPs. As discussed into detail in Ref. 6, this single curve comprises the evolution of the SP magnetic moment, the SP density and the spin temperature. To measure the evolution of these parameters individually, instead of measuring a delay scan at a fixed field, we perform a magnetic field scan at various fixed delays. An example of such a scan, at $t = 0.9$ ns, is shown in Figure 2, and from this scan we can individually extract the SP magnetic moment and SP density as follows. We consider that the SPs are free to point in any direction, and as such their magnetization as a function of magnetic field is described by the paramagnetic Brillouin function, which due to the large SP magnetic moment is very well approximated by a Langevin function. In addition, a linear term is added due to heating, as described in Ref. 15. The photoinduced magnetization as a function of magnetic field is therefore written as

$$\Delta M = \mu_{pol} n_{pol} L\left(\frac{\mu_{pol} B}{k_B T}\right) + \text{constant} \times B, \quad (1)$$

where μ_{pol} and n_{pol} are the respective SP magnetic moment and density at the specified time delay, $L(x) = \coth(x) - 1/x$ is the Langevin function, B is the internal magnetic field, k_B is Boltzmann's constant and T the temperature. A fit of Eq. 1 to the magnetic field scan is indicated by the solid line in Figure 2, whose fit parameters give us μ_{pol} and n_{pol} separately. Repeating this measurement for various delays therefore allows us to independently track μ_{pol} and n_{pol} as function of time, uncovering the dynamics behind the delay scan from Figure 1.

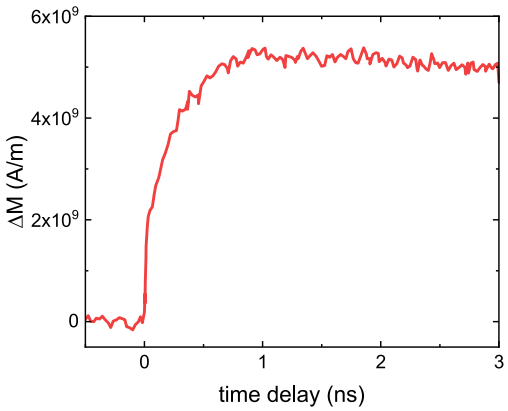


FIG. 1. Photoinduced magnetization due to SPs as a function of time, measured at a temperature of 5 K and applied magnetic field of 60 mT.

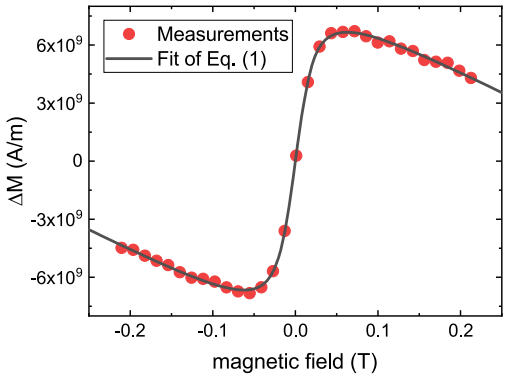


FIG. 2. Photoinduced magnetization due to SPs as a function of magnetic field, where the time delay was fixed at 0.9 ns, measured at a temperature of 5 K.

III. RESULTS AND DISCUSSION

We will mainly consider the temporal evolution of the SP density, n_{pol}, which mirrors the excited carrier density. In Figure 3, we show n_{pol} as a function of time, extracted as described in Section II. As expected, we see an immediate rise in density upon absorption of the pump pulse at $t = 0$, which is due to the virtually instant excitation of carriers. After, we observe a decay to a constant level, indicated by the dashed line in Figure 3, at about a tenth of the initial density. We associate this constant level with earlier reported long-lived SPs in EuSe and other compounds of the europium chalcogenides.[4,5] However, a short-lived state as indicated by the fast decay has not been observed before, which we associate with short-lived SPs. By subtracting the constant level from the data and plotting the remaining density on a logarithmic scale, as shown in the inset of Figure 3, we obtain that the decay of these short-lived SPs is mono-exponential with a lifetime of around 0.5 ns.

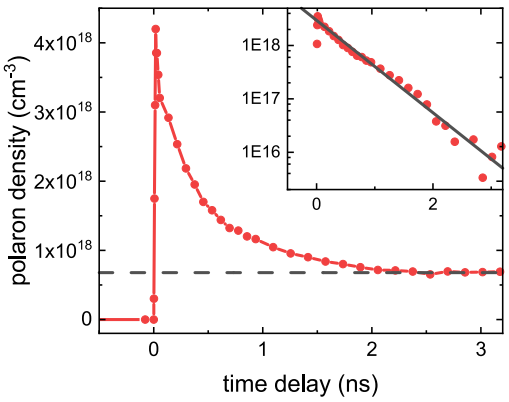


FIG. 3. The evolution of the polaron density (connected red dots) over time, after excitation at $t = 0$, measured at $T = 5$ K. The dashed line indicates the density of long-lived SPs, which appears constant on the timescale considered here. In the inset the density of short-lived SPs (red dots) is plotted on a log scale as a function of time, which is obtained by subtracting the long-lived density from the data in the main graph. The solid line in the inset indicates the fitted exponential decay of short-lived SPs.

18 March 2024 16:13:53

© Author(s) 2024

To assure ourselves that the constant level is indeed associated with the long-lived SPs observed before, we will compare its lifetime to the earlier mentioned results, which is obtained as follows. First, we consider that, as we decrease the time between pulses, we should eventually observe a non-zero SP density before $t = 0$, since insufficient time will have passed for all SPs to decay. Indeed, this is observed in Figure 4, where the (normalized) photoinduced magnetization is plotted for various pulse repetition rates, showing that as time between pulses decreases, i.e., the repetition rate increases, the photoinduced magnetization before arrival of the pump pulse increases. Then, if we assume mono-exponential decay, we can use the ratio between the final level n_f and initial level n_i to determine the lifetime of long-lived SPs through

$$\frac{n_f}{n_i} = \exp\left(\frac{1}{f_{rep}\tau_{ll}}\right), \tag{2}$$

where f_{rep} is the pulse repetition rate and τ_{ll} is the lifetime of long-lived SPs. In the inset of Figure 4 we plot this ratio for various repetition rates, and fitting Eq. 2 yields a lifetime of $\tau_{ll} = 0.45 \pm 0.03$ μs, in approximate agreement with the lifetime of 1.5 μs as reported in Ref. 5 for slightly different experimental parameters, therefore indicating that the constant level is indeed related to long-lived SPs.

Altogether, we find the existence of short-lived SPs in addition to long-lived SPs, whose lifetime difference spans three orders of magnitudes. To explain this large discrepancy, we will first consider the short-lived SP, whose lifetime is about half a nanosecond. We use that the order of magnitude of the lifetime of a dipole-allowed, spin-allowed transition is given by[16]

$$\tau \sim \frac{h}{\alpha^3 h\nu}, \tag{3}$$

where $\alpha = 1/137$ is the fine structure constant, h is Planck's constant and $h\nu$ is the energy of a photon emitted by the transition. Using $h\nu = 2$ eV, equal to the EuSe bandgap, then yields $\tau \sim 1$ ns, in good correspondence with the lifetime of the short-lived SP, indicating that this transition is indeed dipole- and spin-allowed. On the other hand, we know that the long-lived SP in the europium chalcogenides is of excitonic nature,[17,18] where electron-hole recombination is forbidden. We deduce that most SPs recombine before entering this long-lived excitonic state, leading to the existence of short-lived SPs which recombine freely as observed in Figure 3. While we leave the specific origin of the long-lived SP state open for discussion, a possible origin for this large difference in lifetime may be found in the dipole matrix elements,[19] where a rearrangement of the six remaining valence electrons of the Eu^{3+} ion prohibits the excited electron-hole pair to recombine.

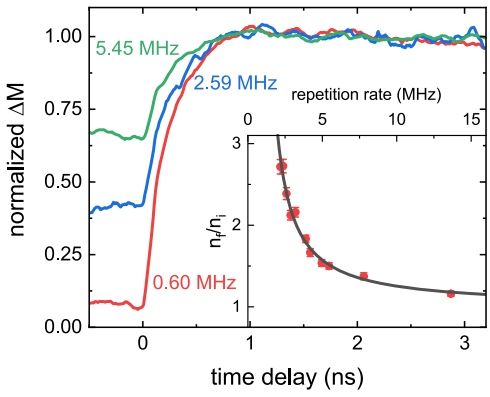


FIG. 4. The normalized photoinduced magnetization as a function of time, for various repetition rates, as indicated by the colored labels. For increasing repetition rate, the residual SP density increases. Inset: the ratio between final and initial density as a function of repetition rate (red dots), where the solid line indicates a fit of Eq. 2.

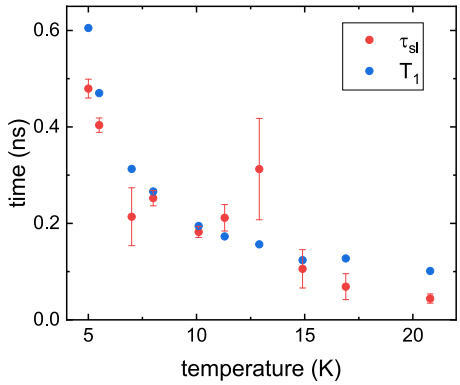


FIG. 5. The decay time of short-lived SPs (τ_{sl}) and growth time of the SP magnetic moment (T_1) as a function of temperature. The data for T_1 is reproduced from Ref. 6.

We further investigate the recombination of short-lived SPs by measuring their decay time, τ_{sl} as a function of temperature, as shown by the red dots in Figure 5. We find a remarkably exact coincidence of the recombination time of short-lived SPs with the growth time of the SP magnetic moment, as taken from Ref. 6 and indicated by the blue dots in Figure 5. Since growth is determined by thermal fluctuations,[6] Figure 5 suggests that fluctuations also favor recombination, possibly shaking the photoexcited state and adding wavefunction components that increase the recombination rate.

IV. CONCLUSION

In summary, we observed short-lived SPs in EuSe, in addition to the earlier reported long-lived SPs. We find that the difference in lifetime between the short- and long-lived SPs spans three orders of magnitude. We propose that the short-lived SP decays through a dipole- and spin-allowed transition, before entering the long-lived, excitonic SP state where recombination is forbidden. A comparison of the decay time of short-lived SPs to the growth time of the SP magnetic moment shows that they follow each other exactly, suggesting thermal fluctuations play a key role not only in SP growth, but also in recombination of short-lived SPs.

ACKNOWLEDGMENTS

The authors acknowledge support by FAPESP (Grants No. 2020/15570-0 and 2022/15791-2) and CAPES (finance code 001).

18 March 2024 16:13:53

© Author(s) 2024

We thank G. Springholz from the Johannes Kepler Universität in Linz, Austria for supplying the EuSe samples.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

S. C. P. van Kooten: Conceptualization (equal); Data curation (lead); Formal analysis (equal); Investigation (equal); Methodology (equal); Visualization (lead); Writing – original draft (lead); Writing – review & editing (supporting). **A. B. Henriques**: Conceptualization (equal); Data curation (supporting); Formal analysis (equal); Funding acquisition (lead); Investigation (equal); Methodology (equal); Project administration (lead); Resources (lead); Supervision (lead); Writing – review & editing (lead).

DATA AVAILABILITY

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

REFERENCES

[1] A. Kirilyuk, A. V. Kimel, and T. Rasing, "Ultrafast optical manipulation of magnetic order," Rev. Mod. Phys. **82**, 2731–2784 (2010).
[2] F. Hellman, A. Hoffmann, Y. Tserkovnyak, G. S. D. Beach, E. E. Fullerton, C. Leighton, A. H. MacDonald, D. C. Ralph, D. A. Arena, H. A. Dürr, P. Fischer, J. Grollier, J. P. Heremans, T. Jungwirth, A. V. Kimel, B. Koopmans, I. N. Krivorotov, S. J. May, A. K. Petford-Long, J. M. Rondinelli, N. Samarth, I. K. Schuller, A. N. Slavin, M. D. Stiles, O. Tchernyshyov, A. Thiaville, and B. L. Zink, "Interface-induced phenomena in magnetism," Rev. Mod. Phys. **89**, 025006 (2017).
[3] A. B. Henriques, G. D. Galgano, E. Abramof, B. Diaz, and P. H. O. Rappl, "Zero-phonon emission and magnetic polaron parameters in EuTe," Appl. Phys. Lett. **99**, 091906 (2011).
[4] A. B. Henriques, G. D. Galgano, P. H. O. Rappl, and E. Abramof, "Light-induced polaron magnetization in EuTe at temperatures reaching 150 K," Phys. Rev. B **93**, 201201 (2016).
[5] A. B. Henriques, X. Gratens, P. A. Usachev, V. A. Chitta, and G. Springholz, "Ultrafast light switching of ferromagnetism in EuSe," Phys. Rev. Lett. **120**, 217203 (2018).
[6] S. C. P. van Kooten, G. Springholz, and A. B. Henriques, "Direct optical probing of ultrafast spin dynamics in a magnetic semiconductor," Phys. Rev. B **105**, 224427 (2022).
[7] X. Gratens, Y. Ou, J. S. Moodera, P. H. O. Rappl, and A. B. Henriques, "Hypergiant spin polarons photogenerated in ferromagnetic europium chalcogenides," Appl. Phys. Lett. **116**, 152402 (2020).
[8] A. Mauger and C. Godart, "The magnetic, optical, and transport properties of representatives of a class of magnetic semiconductors: The europium chalcogenides," Phys. Rep **141**, 51–176 (1986).
[9] S. C. P. van Kooten, P. A. Usachev, X. Gratens, A. R. Naupa, V. A. Chitta, G. Springholz, and A. B. Henriques, "Converting Faraday rotation into magnetization in europium chalcogenides," J. Appl. Phys. **126**, 095701 (2019).
[10] C. Y. Chang, L. Wang, J. T. Shy, C. E. Lin, and C. Chou, "Sensitive Faraday rotation measurement with auto-balanced photodetection," Rev. Sci. Instrum. **82**, 063112 (2011).
[11] E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, "Ultrafast spin dynamics in ferromagnetic nickel," Phys. Rev. Lett. **76**, 4250–4253 (1996).
[12] B. Koopmans, G. Malinowski, F. Dalla Longa, D. Steiauf, M. Fähnle, T. Roth *et al.*, "Explaining the paradoxical diversity of ultrafast laser-induced demagnetization," Nat. Mater **9**, 259–265 (2010).
[13] A. Stupakiewicz, K. Szerenos, D. Afanasiev, A. Kirilyuk, and A. V. Kimel, "Ultrafast nonthermal photo-magnetic recording in a transparent medium," Nature **542**, 71–74 (2017).
[14] 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 **435**, 655–657 (2005).
[15] A. B. Henriques, A. R. Naupa, P. A. Usachev, V. V. Pavlov, P. H. O. Rappl, and E. Abramof, "Photoinduced giant magnetic polarons in EuTe," Phys. Rev. B **95**, 045205 (2017).
[16] A. S. Davydov, *Quantum Mechanics*, *International Series in Natural Philosophy*, 2nd ed. (Pergamon Press, 1976), p. 405.
[17] 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. Rappl, "Luminescence properties of magnetic polarons in EuTe: Theoretical description and experiments in magnetic fields up to 28 T," Phys. Rev. B **90**, 165202 (2014).
[18] A. B. Henriques, S. C. P. van Kooten, E. Abramof, P. H. O. Rappl, and G. D. Galgano, "Bound photoinduced giant spin polaron in EuTe," J. Appl. Phys. **131**, 043903 (2022).
[19] G. D. Galgano, "Orientação óptica de spin em semicondutores magnéticos—Calcógenos de Európio," Ph.D. thesis, Instituto de Física, Universidade de São Paulo, 2012.

18 March 2024 16:13:53

© Author(s) 2024