AIP | Advances

An exploration of some magnetic fundamentals in EuSe using μSR

I. Terry, P. W. Adams, N. Bykovetz, S. R. Giblin, Z. Guguchi, R. Khasanov, J. Klein, C. L. Lin, and T. J. Liu

Citation: AIP Advances **6**, 055705 (2016); doi: 10.1063/1.4943235
View online: http://dx.doi.org/10.1063/1.4943235
View Table of Contents: http://scitation.aip.org/content/aip/journal/adva/6/5?ver=pdfcov
Published by the AIP Publishing

An exploration of some magnetic fundamentals in EuSe using μSR

I. Terry,[1] P. W. Adams,[2] N. Bykovetz,[3,a] S. R. Giblin,[4] Z. Guguchi,[5] R. Khasanov,[5] J. Klein,[6] C. L. Lin,[3] and T. J. Liu[2]
[1]*Physics, Durham University, Durham, DH1 3LE, United Kingdom*
[2]*Physics and Astronomy, Louisiana State University, Baton Rouge, LA,70803-4001, USA*
[3]*Department of Physics, Temple University, Philadelphia, PA, 19122-6082, USA*
[4]*Physics and Astronomy, Cardiff University, Cardiff,Cf24-6457, United Kingdom*
[5]*Laboratory for Muon-Spin Spectroscopy, Paul Scherrer Institute, Villigen-PSI, Switzerland*
[6]*Physics and Astronomy, University of Pennsylvania, Philadelphia, PA 19104-6396, USA*

(Presented 13 January 2016; received 6 November 2015; accepted 14 December 2015; published online 1 March 2016)

EuSe is a simple magnetic system that appears to show many complicated features. Under applied pressure it undergoes a transition from an antiferromagnet (AF) to a ferromagnet (FM). This transition provides a means of testing certain basic fundamentals of magnetic theory and an opportunity to explore the complexities of EuSe. Using the muon-spin rotation and relaxation technique (μSR), EuSe was measured at pressures ranging from ambient to 11 kbar. In ambient-pressure EuSe, muon data reveal two local fields, but show only a single field in the FM state formed under pressure. The μSR measurements appear to show a continuous transition at Tc, contrary to previous Mössbauer results that were interpreted as being evidence of a first-order transition. Values determined for the critical exponent, β, in AF and FM EuSe, differ and therefore appear to be a clear counterexample to the Universality Hypothesis. The values of β also are indicative of EuSe's being a 2D magnet for pressures up to 11 kbar. The nature and values of the local fields seen by the muons is discussed and analyzed. [http://dx.doi.org/10.1063/1.4943235]

I. INTRODUCTION

The magnetic properties of EuSe offer a unique opportunity to provide answers to some fundamental questions in the theory of magnetic behavior of simple insulating magnetic systems. In this study we undertook an experimental investigation to test two things: firstly, can a three-dimensionally (3D) coupled magnetic system such as EuSe display pure 2D magnetic behavior, and secondly, is such behavior in accord with the Universality Hypothesis of critical phenomena.

At first glance, the magnetism of EuSe, unlike that of the other chalcogenides (EuCh: EuO, EuS, and EuTe), appears to be anything but simple.[1] Below the magnetic ordering temperature of 4.7 K, EuSe transforms successively into three different magnetic structures (two antiferromagnetic [AF] and one ferrimagnetic [FIM]). EuSe exhibits magnetic-phase hysteresis on thermal or applied field cycling.[2] Application of small pressures (see Fig. 1(a)) results in the formation of a ferromagnetic [FM] state.[3]

All of the magnetic structures found in EuSe can be described by different stackings of ferromagnetically aligned sheets of spins lying in the (111) planes. When cooled below $T_c \sim 4.7$ K, a single NNSS (↑↑↓↓) structure normally forms. On cooling the sample below ~2.8 K this magnetic phase changes to the FIM NNS (↑↑↓) structure, and on further cooling below ~ 1.8 K, a NSNS (↑↓↑↓)

[a]email:nbykovetz@temple.edu

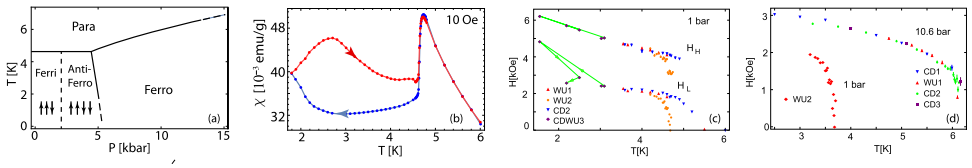

FIG. 1. (a) Schematic diagram of EuSe magnetic ordering temperature T_c vs. applied pressure based on combined results of previous susceptibility and NMR measurements (references in text). (b) Our SQUID magnetometer susceptibility measurements as a function of temperature made on cooling, then warming (arrows). Curves indicate a hysteresis in magnetic-phase composition, but a fixed T_c at 4.7K. (c) The μSR measurements at ambient pressure showing two distinct fields or two inequivalent muon sites in EuSe. Temperature dependences were measured for various cool-downs (CD) and warm-ups (WU), with the first CD from well above T_c down to 3.5K. (d) Critical-region fall-off of the magnetization (as measured by the lower muon field) in ambient-pressure EuSe, and critical and intermediate region fall-off in the ferromagnetic state at 10.6 kbar (on CDs and WUs).

structure forms. When EuSe is heated from lower temperatures, the above transformations are not repeated. Instead, coexisting mixtures of these phases[2] are formed and can persist up to T_c. Fig. 1(b) illustrates SQUID magnetization measurements of our sample, showing a very precise $T_c = 4.7$ K, and the hysteresis on cooling to just below 2 K and then warming the sample back up to 6 K.[4]

This seeming complexity can be understood in a simple way by hypothesizing that the (111) sheets of spins are strongly exchange-coupled by two-neighbor-exchange interactions, J_1 and J_2, but become de-coupled from each other when $J_2 \sim -J_1$, forming a 2D magnetic system, in which the much weaker interactions between these sheets determine the stacking arrangements. A recent review of this perspective, and evidence for this explanation is discussed by Bykovetz *et al.*,[1] and motivated the current study.

An early Mössbauer study[5] was interpreted as showing that EuSe undergoes a first-order magnetic phase transition at T_c. Several subsequent papers repeated this claim without providing any further direct evidence. It is very difficult to differentiate the fast fall-off of a 2D second-order transition from a 3D first-order transition. A stong motivation for undertaking these studies and using μSR was to obtain clearer evidence of whether EuSe undergoes a first-order transition and if not, to measure its β.

II. EXPERIMENTAL ISSUES AND OUTCOMES

Our experiments were carried out at the PSI General Purpose spectrometer (GPD) using a high-energy μ^+ beam incident on a sample of EuSe immersed in Daphne oil to ensure uniform hydrostatic pressure and encased in a BeCu pressure cell. Because we intended to follow the ambient-pressure measurements with measurements at high pressure on the same sample, we used the BeCu cell throughout the meaurements despite larger backgrounds at 1 bar. Below T_c, the raw data were fitted using the *Musrfit* program with an oscillatory function plus a Kubo-Toyabe term[6] to account for the BeCu pressure cell. The values of the local muon magnetic fields were determined from the oscillation frequencies. The μSR data are similar to those for EuO[7] and EuS,[8] showing a single oscillation (single field) for the FM EuSe, but two oscillations (two fields), at presumed two different muon sites at ambient-pressure. Our initial interest was to study the critical region of ambient-pressure EuSe, and thus we restricted T to $\gtrsim$ 3.5K, to avoid mixed magnetic phases. Fig. 1(c) shows the results. The sample was cooled from higher temperatures to 3.5K, and then warmed (Warm-up1 [WU1]) to 6K for a quick overview scan. Data acquisition was terminated at each temperature after $2 - 10 \times 10^6$ events (~1hrs or less per point). Two distinct fields are clearly evident in the $p \sim 0$ data, suggesting that muons are localized at two inequivalent sites. The sample was then cooled back down to 3.5K (Cool-down 2 [CD2]) and careful, fine-temperature-mesh data acquired during a warm-up (WU2). The lower-field, H_L, data collected in WU2 is shown in Fig. 1(d). After the critical region scans were completed, a decision was made to do a quick exploratory run to observe the behavior below 3.5K. Five points were taken, and are shown as CD-WU3. The higher field, H_H, behaved consistently here, but H_L showed an unexpected excursion.

The pressure in the cell was then raised to 10.6 kbar, to insure a complete FM state, per Ref. 3. Fig. 1(d) shows the resulting curves that include WU and CD cycles. Again, the focus was on the critical region, so only the region $T \gtrsim 3.5$K was investigated. As in the case of previous NMR data[9] no discernible hysteresis was observed in the FM state on cycling the temperature, but we obtained a T_c higher by more than one degree from that obtained by macroscopic measurements.[3]

Based on these preliminary results, we obtained more beam time to test the hypothesis that EuSe behaves as a 2D magnet and may not be in accord with the Universality Hypothesis. The goals were to explore the pressure region shown in Fig. 1(a) where EuSe undergoes a transformation from a pure NNSS phase (existing between ~2 and ~5 kbar) to a FM phase (starting at ≥ 5 kbar).[9,3] μSR measurements in this region could give a more direct determination of any change(s) in the critical exponent β. The low-temperature behavior of the magnetization curves, would also be useful in distinguishing 2D from 3D behaviors.

Detailed measurements at p = 10.8 kbar were carried out and are shown in Fig. 2(a). Measurements were then made at 5.25 kbar, down to 0.25 K. These data gave a very precise quantitative characterization of the "intermediate region"[1] of the magnetization curve, and were reasonably precise at the low-temperature end (down to 0.25 K). The 5.25 kbar data show a pure FM phase down to ~ 2.8 K, but then formation of a complex magnetic state that could not be described by the standard fitting forms available with the *Musrfit* program. Additional attempts were made at 4.33 and then 3.8 kbar to get the presumed pure NNSS curves, but no clean data could be extracted here either.

III. ANALYSIS OF DATA

The ambient-pressure measurements yielded some useful results but elicited questions that will need additional studies to resolve. Two fields were observed as expected within the three magnetic structures (as we'll outline later). Some new hysteresis issues appeared to result from the use of the muon probes. When the sample is cooled from ~ 6 K (CD2) the muons appear to respond to short-range order effects, seeing dynamical fields before magnetic ordering takes place. This has recently been reported in some μSR literature,[10] and will need further study in the case of EuSe. The quick WU1 measurements (Fig. 1(d)) show an H_L field that falls off more slowly than the very slow measurements made during WU2, in which the temperature was changed by very small steps (~ 2 mK) over a small temperature interval (4.2 - 4.6 K). Since thermal equilibrium of the sample and cell appeared to take place quickly, this discrepancy is hard to understand, and will need further investigation. The data for the gradual ΔT WU2 measurements for both H_L and H_H do appear to scale with the measured T_c (Fig. 1(b)). Despite the substantial statistical scatter of data points, we see that there is no evidence of discontinuous behavior in H_L (Fig. 1(d)), i.e., no indication of a first-order transition. Fitting the data to the critical-region equation, $H = DH_0(1 - T/T_c)^\beta$ over the "extended critical region,"[11] we obtain $\beta = 0.14 \pm 0.02$, using the procedure described in Ref. 11, and values of $D = 1.03$ (from NMR[1]) and an NMR-scaled[2] extrapolation, which gave $H_L = 2.75 \pm 0.1$ kOe at ($T = 0$). This low value of β is a clear indication of 2D behavior, and is supported by theoretical predictions that for $J_2 = -J_1$, in the FCC lattice, the magnetic dispersion in the [111] directions becomes flat, i.e., the (111) spin-planes becomes decoupled.[12] We now argue that the data of the two ~10 kbar measurements and the data in the FM part of the 5.25 kbar curve give evidence that 2D behavior persists in the EuSe system over the entire range of measured

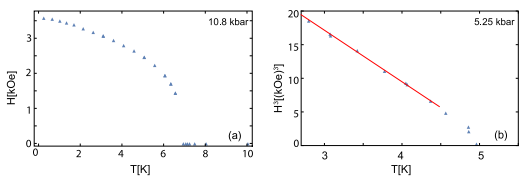

FIG. 2. (a) Precision measurements of the single muon field observed in the ferromagnetic (FM) state of EuSe at 10.8 kbar, on cooling down to 0.25K. (b) Critical and intermediate region behavior of FM EuSe at 5.25 kbar, plotted as $H^3 vs. T$ (see Ref. 1), showing a critical exponent β of ~ 0.33 in the "intermediate region" and a faster fall-off ($\beta \sim 0.25 \pm 0.05$) as T_c is approached. (A pure FM state exists only in the region $2.8K \leq T < 5K$; *c.f.*, Fig. 1(a) near ~ 5 kbar.).

pressures (from p ~ 0 up to at least 11 kbar). Our most precise data, the 10.8 kbar data shown in Fig. 2(a), show an excellent fit to $\beta = 1/3$ over an extended region of the magnetization curve. As discussed in Refs. 1 and 11, this is characteristic of 2D FMs. (3D FMs, such as EuS, show a $\beta = 1/3$ only close to T_c, and have a much larger D~1.20.[11]) Additionally, the low-temperature data show the magnetization dropping off nearly linearly, as in cases like $CrCl_3$,[11] a well studied 2D FM. The CD2 10.6 kbar data in Fig. 1(d), also show a good critical equation fit, with $\beta = 1/3$, but the 4 points closest to T_c fit to $\beta = 0.25$, $T_c = 7.14$ K. An analysis of the FM portion of the 5.25 kbar data shows similar results. Fig. 2(b) displays a plot[11] of H^3 vs. T near T_c. We see that again, the data lie on a straight line[11] (i.e., $\beta = 1/3$), but show a deviation to a smaller β close to T_c. A fit gave $\beta = 0.25 \pm 0.05$.

A comparison of the critical exponents β obtained for ambient-pressure EuSe (0.14 ± 0.02), and for FM EuSe under pressure (0.25 ± 0.05), shows that the change in β so incurred is outside of experimental error, despite the limits on the statistical accuracy of our μSR measurements. The Universality Hypothesis of critical theory holds that if the dimensionality of the magnetic system, the spin dimensionality, and the range of interaction stay the same, then the critical exponents should be the same. Our measurements may constitute the first good counter-example to the Universality Hypothesis, since for EuSe under pressure, dimensionality, spin S, and the short-range exchange interaction do not change as we go from 0 to 11 kbar. We expect that higher-precision data will confirm this result.

Apart from testing certain assertions that are fundamental to magnetic theory, our measurements also give insights into the use of μSR techniques in studying the EuCh and similar magnetic compounds, where it may be important to know the number of non-equivalent muon local fields, and what gives rise to these fields. In general, the local field, H_{loc}, seen by a muon is determined by the dipole field, H_{dip}, produced by the magnetic ions (Eu), a long-range effect; and the contact hyperfine field, H_{hf}, which in insulating materials is short-range, and typically related to the electron-spin polarization of the electron cloud by *neighboring* (Eu) magnetic ions that also cause the transferred hyperfine fields at the neighboring magnetic and ligand nuclei. In simple FCC (NaCl) lattices, stopped muons are found to locate at the (1/4,1/4,1/4) positions[7] of the unit cell (see Fig. 2(a) of Ref. 7). This means that the muons locate on planes that are 1/4 and 3/4 of the distance between successive Eu ion (1,1,1) planes (and midway between (1,1,1) neighboring Eu and Se planes). Using this premise and symmetry considerations, we have calculated the dipole fields at muon locations in the four magnetic structures of EuSe, using the usual Lorentz approximation. In the latter, H_{dip} = field produced by all dipoles within a large Lorentz sphere + the Lorentz field ($4\pi M/3$).

Symmetry considerations show that in the NNN and the NSNS structures there can be only one muon field. In NNSS there can be two, and in NNS, there are three possible dipolar fields. Rounding off the results, we found that (at $p \sim 0, T = 0$) H_{FM} = +4.6 kOe and in NSNS +7.7 kOe. For NNSS we get +7.7 kOe for sites between NS, and +4.5 kOe for muon sites between NN (and likewise, SS) . For NNS the three different values from the Lorentz sphere are 9.2, 6.2, and 3.0. However, when the Lorentz field of +1.5 kOe is added appropriately, the numbers degenerate to only two values, +7.7 and +4.5 kOe. In all cases, the + sign means the dipolar fields are parallel to the direction of the moments of the closest Eu (111) planes. The upshot is that only two different dipolar field values are possible within all three non-FM structures, namely, +7.7 (at sites between NS) and +4.5 kOe (at sites between NN planes).

Consideration of the hyperfine-field contributions, H_{hf}, at the muon sites are relatively straightforward for the FM case, but not so clear-cut for the other magnetic structures. However, reasonable arguments can be made. The extrapolated (to T=0) muon fields, H_{loc}, observed in FM EuSe at 5.25 and 10.8 kbar are 3.30 and 3.60 kOe. The difference in magnitude of $|\Delta H_{loc}| = |\Delta H_{hf} + \Delta H_{dip}|$ = |0.30| kOe is too large to be accounted for by the expected increase of the (positive) dipolar field. Since the hyperfine field, H_{hf}, at the muon sites is always negative,[7] ΔH_{hf} could be either a sizeable negative increase or a small positive decrease in H_{hf}. NMR measurements[9] show that the Eu *transferred* hf fields get more negative with applied pressure, and therefore so must H_{hf} at the muon sites. Thus, the |0.30| kOe increase must be negative and so must the measured 3.30 and 3.60 kOe fields. Since $H_{dip} \sim +4.6$ kOe, the H_{hf} contributions must be ~ -7.8 kOe at 5.25 kbar and ~ -8.2 at

10.8 kbar. Since NMR also shows[9] that the magnetic curves in EuSe are identical for p between 0 and 5 kbar (to within ~ 0.1%),[13] it is reasonable to assume that H_{hf} stays at ~ −7.8 kOe down to $p = 0$.

Now, as we've seen, at p ~ 0 two-muon fields were actually observed. When extrapolated to T = 0, they become $H_H = 7.3$ kOe (using a linear extrapolation of the points in Fig. 1(c)) and $H_L = 2.7$ kOe (as discussed previously). The higher field $H_H = 7.3$ kOe must correspond to muon sites between NS planes, since H_{hf} and H_{dip} have opposite signs and therefore cannot add up to such a large-magnitude number between the NN planes. Thus, $H_L = 2.7$ kOe must represent muon sites between the NN planes. If we assume $H_{hf} \sim -7.8$ kOe between NN planes (as it is in the FM structure at 5.25 kbar), we would conclude that H_L should equal $\sim -7.8 + 4.5 = -3.3$ kOe. Since we are ignoring zero-point spin deviations, this is in ballpark agreement with the measured $H_L = 2.7$ kOe, and thus the latter value must be negative. Similarly, $H_H = 7.3$ kOe, must be positive and is roughly explainable by the H_{dip} value of 7.7 kOe. The hyperfine field between the NS planes must to a large degree cancel, and may possibly be completely negligible, since the Eu moments at neighboring planes induce electron-spin polarizations in opposite directions (and at the Se nuclei cancel exactly by symmetry). It may be possible in the future to check these arguments by doing applied-field measurements.

IV. CONCLUSIONS

We have conducted μSR measurements in ambient and under-pressure EuSe and found solid evidence that EuSe behaves as a 2D magnet throughout the range of pressures $0 < p < 11$ kbar. We found no evidence supporting the first-order magnetic transition previously claimed in Mössbauer measurements. Our measurements across the AF to FM transition support the claim that the Universality Hypothesis of critical exponents may not hold up in this system. The various muon fields measured were analyzed, and a plausible and consistent picture developed.

ACKNOWLEDGMENTS

This project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under the NMI3-II Grant number 283883. We thank the Paul Scherer Institute (PSI) and their wonderfully helpful staff for supporting our work under proposal number SμS-20130825. Synthesis and characterization of the EuSe samples (PWA and TJL) was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, under Award No.DE-FG02-07ER46420.

[1] For a recent review, see, N Bykovetz, J Klein, CL Lin, and K Raj, JAP **109**, 07E165 (2011).
[2] T Komaru, T Hihara, and Y Kōi, JPSJ **31**, 1391 (1971).
[3] H Fujiwara, H Kadomatsu, M Kurisu, T Hihara, K Kojima, and T Kamigaichi, SSC **42**, 509 (1982); H Bach, A Eiling, and K Westerholt, JMMM **28**, 214 (1982).
[4] These measurements were made on Quantum Design MPMS SQUIDs at LSU (RSO option) and at Temple University (DC SQUID).Similar results were obtained at ISIS, where SQUID measurements were also made at 2.5, 5 and 10 kbar.
[5] G Petrich and T Kasuya, SSC **8**, 1625 (1970).
[6] A Schenck and FN. Gygax, in *Handbook of Magnetic Materials*, edited by KHJ Buschow (North Holland, Amsterdam, 1995), Vol. 9, p. 57.
[7] SJ Blundell, T Lancaster, PL Pratt, PJ Baker, W Hayes, J-P Ansermet, and A Comment, PR B **81**, 092407 (2010).
[8] DG Eshchenko, VG Storchak, E Morenzoni, and DA Andrecia, Physica B **404**, 903 (2009).
[9] K Hiraoka, T Hihara, K Kojima, and T Kino, I SSC **52**, 479 (1984).
[10] An example is, T Lancaster, SR Giblin, G Allodi, S Bordignon, M Mazzani, R De Renzi, PG Freeman, PJ Baker, FL Pratt, P Babkevich, SJ Blundell, AT Boothroyd, JS Möller, and D Prabhakaran, Phys. Rev. B **89**, 020405(R) (2014).
[11] N Bykovetz, J Klein, and CL Lin, JAP **109**, 07E119 (2011).
[12] RA Tahir-Kheli, HB Callen, and H Jarrett, J Phys Chem Solids **27**, 23 (1966).
[13] We remark that the constancy of T_c over the pressure range of $0 \lesssim p \lesssim 5$ kbar (see Fig 1(a)), substantiated by our ISIS SQUID measurements,[4] and by NMR measurements[9] is very mysterious. The pressure changes should result in changes in the exchange constants, and in the dipolar and hyperfine fields, but do not appear to do so in EuSe over this range of pressure.