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Is EuSe a pseudo two-dimensional magnetic system?

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We present ⁷⁷Se NMR measurements in EuSe and review previous ¹⁵³Eu NMR measurements that show that EuSe displays 2D characteristics at both ends of the magnetization curve. The low-temperature behavior, in agreement with previous measurements, follows a simple 2D T² falloff. At the higher temperatures, a critical-exponent fit shows an apparent 2D behavior with a $\beta \sim 0.18 \pm 0.02$. EuSe has always appeared to be a complicated magnetic system, with a number of co-existing magnetic phases and an alleged first-order magnetic transition. We argue that the multiple magnetic structures are in fact compatible with 2D behavior, and that there is actually no firm evidence of a first-order transition. In the ferromagnetic state of EuSe (obtained under pressure or by an applied field), a $\beta \sim 0.33$ is found, but with a $D \sim 1$, characteristic of the behavior of the classic 2D magnets K₂CuF₄ and CrCl₃, and not of 3D critical behavior. Additional experiments to verify the 2D nature of EuSe are suggested. The nontrivial implications of the two-dimensionality of EuSe regarding the nature of magnetic interactions in general are also discussed. © 2011 American Institute of Physics. [doi:10.1063/1.3567041]

I. INTRODUCTION

In contrast to the simplicity of the other presumed prototype Heisenberg systems (EuO, EuS, and EuTe), EuSe appears to exhibit a variety of complicated behaviors in its magnetically ordered state. Two antiferromagnetic ($\uparrow\downarrow\uparrow\downarrow$ and $\uparrow\uparrow\downarrow\downarrow$) and one ferrimagnetic ($\uparrow\uparrow\downarrow$) structures have been identified.^{1,2} These exist in different temperature regions below T_c. There is evidence that these magnetic phases can coexist over certain temperature ranges, and transform from one to the other³ and that the magnetic phase composition as well as T_c can depend on the sample preparation.^{2,3} The magnetic phases consist of different 3D stackings of ferromagnetically aligned sheets of spins lying in the (111) planes (the arrow designation used above indicates the relative alignment of the ferromagnetically aligned (111) sheets of spins). In an applied magnetic field^{1,2} or under pressure,⁴ EuSe becomes ferromagnetic ($\uparrow\uparrow\uparrow$). In addition, at T_c \sim 4.6 K, EuSe is alleged to undergo a first-order magnetic phase transition.⁵ The primary cause of this complexity is believed to be that the first and second neighbor exchange interactions, ferromagnetic J₁ and antiferromagnetic J₂ nearly cancel (at least in the mean-field model), allowing weaker interactions to cause instabilities. However, no adequate explanation for the existence of a first-order transition has really been given, and measurements in 1% Sn⁺⁺-doped EuSe³ show no sudden drop in the Sn transferred hyperfine field up to at least 4.85 K.

We propose that this complexity can be understood as coming from a magnetic decoupling of the (111) planes, much as the mean-field model suggests, i.e., we argue that the various magnetic structures of EuSe consist of essentially decoupled 2D ferromagnetic sheets. If this is so, then the critical

region should display 2D behavior as well, and thus one should expect a very rapid falloff of the magnetization in the critical-region. In this paper we analyze the NMR data and show that indeed the critical-region behavior appears to be 2D. The conclusion that EuSe undergoes a first-order transition was deduced from ¹⁵³Eu Mossbauer fits of complicated spectra.⁵ In our view, this almost-certainly erroneous conclusion was made because the possibility of a 2D second-order transition, as well as the temperature variation of the composition of coexisting magnetic phases,³ was not known at that time. There has been no subsequent evidence supporting a first-order transition.

We present analyses of our, and other, NMR data using low-temperature and critical-region behaviors that support the idea that EuSe behaves as one would expect of a 2D magnetic system. In addition, we suggest two experiments that can provide definitive support or could definitely disprove the 2D nature of magnetic interactions in EuSe.

Specifically, we present spin-echo ⁷⁷Se NMR data in EuSe and previous results from ¹⁵³Eu NMR measurements that show that all structures, except the ferromagnetic one, follow a simple T² behavior in the spin-wave regime, consonant with 2D behavior. We also analyze these NMR data in the extended critical region to show, that here too, the behavior appears to be consistent with 2D critical behavior.

II. EXPERIMENT

Table I gives our ⁷⁷Se NMR spin-echo measurements in tabular form for the region below 4 K. The ⁷⁷Se line widths (FWHM) are of the order of 0.15 MHz. The temperature was determined from the liquid helium vapor pressure. Samples were made of polycrystalline EuSe.

For our analysis we have also used the ¹⁵³Eu NMR data of Komaru *et al.*¹ Because these data were not available in

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TABLE I. ^{77}Se spin-echo NMR measurements in EuSe. (Temperature T in K, Frequency ν in MHz.)

T	ν	T	ν	T	ν
1.27	57.42	2.61	53.19	3.28	50.03
1.43	56.97	2.63	53.04	3.30	49.86
1.54	56.70	2.65	52.85	3.33	49.78
1.59	56.52	2.71	52.60	3.39	49.52
1.68	56.40	2.75	52.38	3.42	49.29
1.71	56.20	2.79	52.32	3.52	48.80
1.81	55.90	2.81	52.17	3.54	48.60
1.83	55.80	2.89	51.98	3.58	48.24
1.96	55.53	2.92	51.75	3.62	48.06
2.07	55.15	2.95	51.57	3.70	47.55
2.06	55.04	3.04	51.10	3.80	47.17
2.26	54.38	3.10	51.00	3.96	46.07
2.45	53.68	3.26	50.18	3.98	45.85

numerical form, we have digitized the data presented in their Fig. 1, specifically the H resonance, chosen because it was detectable to the highest temperature (4.6 K) and is therefore the most suitable for a critical-region analysis.

Additionally, we have digitized the ^{153}Eu NMR data of Kawakami *et al.*² for the ferromagnetic structure of EuSe (f_{F} resonance in their Fig. 2).

III. RESULTS AND DISCUSSION

The T^2 dependence of the magnetization at the low-temperatures in all three magnetic structures of EuSe ($\uparrow\downarrow\uparrow, \uparrow\uparrow\downarrow$, and $\uparrow\uparrow\downarrow$) has been noted in several publications, starting with the original observation of Kuznia *et al.*⁶ The T^2 behavior exists in these structures in the region $M_0 > M > 0.90M_0$, which translates roughly into the region $T < 3.5$ K. Our ^{77}Se data also give a good fit to a T^2 dependence ($R^2 = 0.9994$), with no deviations visible all the way up to 4 K. The conformance of our data to a T^2 dependence to such a high temperature is probably due to a higher T_c in our sample (in Ref. 2 it was shown that different sample preparations can result in a seemingly substantial variation of T_c).

The appearance of a simple T^2 behavior is difficult to understand from the perspective of conventional spin-wave theory, because the two antiferromagnetic ($\uparrow\downarrow\uparrow$ and $\uparrow\uparrow\downarrow$) structures should be affected by energy-gap effects, and thus

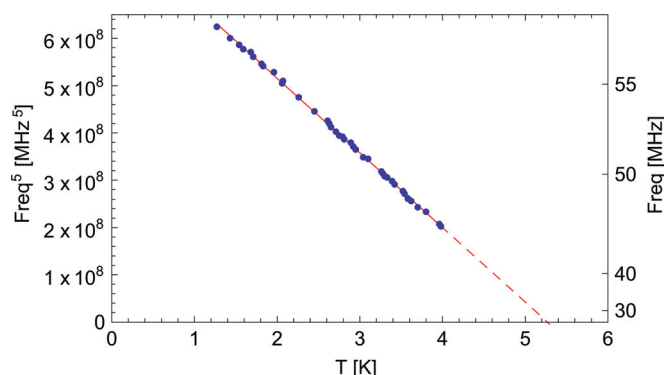


FIG. 1. (Color online) ^{77}Se spin-echo NMR data in EuSe plotted as Freq^5 vs T so as to display the 2D BCRC critical-region behavior.

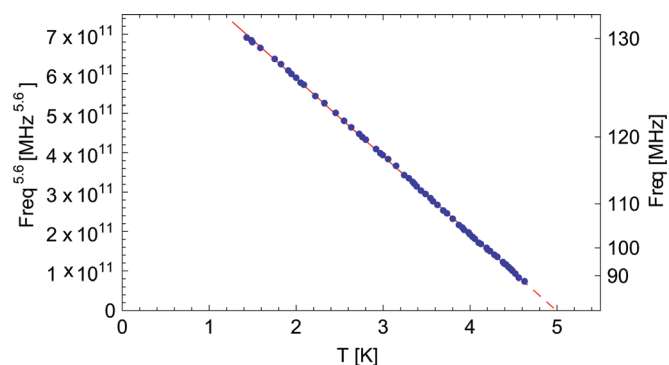


FIG. 2. (Color online) ^{153}Eu NMR data H-resonance of EuSe (Ref. 1) plotted as $\text{Freq}^{5.6}$ vs T so as to display the 2D BCRC critical-region behavior. This NMR data comes closer to T_c than any other EuSe NMR data.

deviate substantially from a simple 3D T^2 antiferromagnetic behavior predicted by this theory. Moreover, the ferrimagnetic behavior should be quite different from that of the two antiferromagnetic structures. If, however, we can regard these structures as being composed of essentially de-coupled 2D planes, the temperature dependences of the antiferromagnetic and ferromagnets should be the same.

Bykovetz has constructed a semiempirical spin-wave scheme⁷ for low-temperature behavior of spin-only (i.e., no orbital moment) magnetic systems. Within this scheme, both 2D ferromagnets and 2D antiferromagnets should display temperature dependences of either T^2 or $T^{(4/3)}$ in the spin-wave region [$M_0 > M > 0.9M_0$].⁷ The above T^2 behaviors in nonferromagnetic EuSe are consistent with this scheme. Additionally, the 3D ferromagnetically stacked EuSe ($\uparrow\uparrow\uparrow$ structure) observed under pressure⁴ appears to exhibit the other predicted 2D behavior, i.e., a $T^{(4/3)}$ dependence, although these measurements did not extend to low-enough temperatures to ascertain such a T dependence with sufficient precision (see Fig. 3 of Ref. 4). Our ^{77}Se NMR measurements in a powdered sample of EuSe in an applied field of 8.5 kG (sufficiently high to produce 3D ferromagnetic alignment), did show an explicit $T^{(4/3)}$ behavior.

If, in fact, EuSe consists of a stacking of 2D ferromagnetic planes, and is thus a pseudo 2D system (i.e., a system in which the ferromagnetic and antiferromagnetic interactions between the (111) planes cancel out), then the various

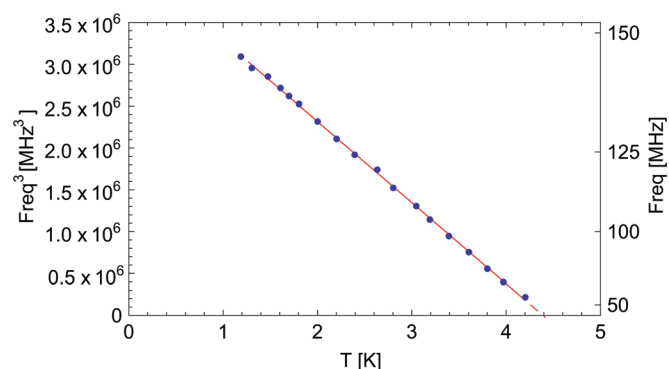


FIG. 3. (Color online) ^{153}Eu NMR data of Kawakami *et al.* (Ref. 2) for the ferromagnetic resonance of EuSe plotted as Freq^3 vs T so as to display the 2D BCRC critical-region behavior.

structures should display an appropriate 2D critical region behavior. In an article in the same issue of this journal,⁸ we presented evidence for the validity of the criterion, first observed by Birgeneau *et al.*, in K_2NiF_4 ⁹ that the critical region of 2D magnets is much wider than that for 3D systems; specifically, that the critical-region equation does not deviate from the magnetization curve up to 90% of M_0 (we refer to this characterization as the BCRC criterion⁸). Because good NMR data for EuSe exist for $M < 0.90 M_0$, a critical-region analysis of the EuSe NMR data can be performed using this perspective.

In Fig. 1 we show a plot of our ⁷⁷Se NMR data using a BCRC critical-region analysis [and displaying the result in the power-law form (see Ref. 8)] of the critical-region equation $M/M_0 = D(1-T/T_c)^\beta$. The plot shows an excellent fit of the data to a β of $1/5$ (i.e., 0.20). A least-squares analysis gives a β of 0.18 ± 0.01 , but we do not believe the data is precise enough to distinguish these two values, consistent with the experimental scatter of the data points. The parameter $D = 1.03$ is consistent with the BCRC criterion. The digitized H-resonance data of Ref. 1 gives very similar results (see Fig. 2).

In Fig. 3 we present the BCRC critical-region behavior of the ferromagnetic resonance of Kawakami *et al.*² Here, a $\beta = 0.32 \pm 0.02$ is observed, with $D = 1.03$. We have shown⁸ that the classic 2D ferromagnets K_2CuF_4 and $CrCl_3$ both display a similar 2D BCRC critical behavior, as well as the similar low-temperature $T^{(4/3)}$ magnetization falloff. Note also that the magnetization of ferromagnetic EuSe under pressure (Fig. 3 of Ref. 4) follows exactly the same “intermediate region” behavior ($T^{1.5}$) as do the two classic 2D ferromagnets K_2CuF_4 and $CrCl_3$. We showed in Ref. 8 that an intermediate region behavior of $T^{1.5}$ in these two magnets translates into a critical region fit with a β of $1/3$. Additionally, 2D ferromagnets of the type $(C_nH_{2n+1}NH_3)_2CuCl_4$ display extremely similar behavior. Kubo *et al.*¹⁰ depict the $T^{1.5}$ behavior. De Jongh *et al.* showed that $\beta = 0.33 \pm 0.02$, with a $D = 1.08 \pm 0.08$ (see Eq. 11 and Fig. 13 of Ref. 11).

Additional evidence that EuSe behaves like a 2D magnet comes from the following. So far, except for EuSe (see Ref. 3), the only simple magnets that have been found to have coexisting magnetic phases with the exact same T_c , whose proportions can change with sample preparation and with temperature, have been 2D magnets. A prime case in point is Rb_2MnF_4 .^{9,12}

In conventional spin-wave theory, 2D behavior in a magnet possessing cubic symmetry, such as EuSe, is impossible, because the magnon dispersion at small wave-vector k is spherically symmetric. This means that the coupling between (111) planes of spins can never become negligible, which would be required to give uncoupled 2D layers. The 2D behavior would be expected in EuSe if the magnetism was determined by mean-field interactions and the nearest and next-nearest exchange interactions canceled (near the point where $J_1 \approx -J_2$). However, we do know that mean-field behavior is never observed in any magnetic system at low temperatures, and is certainly not compatible with a critical-exponent β of ~ 0.18 . Because the model of short-range Heisenberg exchange interactions does not permit a decoupling of neighboring spin planes even when $J_1 = -J_2$, the inescapable conclusion is that if empirically EuSe exhibits 2D behavior

(both in the critical and spin-wave regimes), the exchange interactions in EuSe *cannot* be short-range. It will be a challenge to theory to find a model of long-range exchange interactions in which the ferromagnetic and antiferromagnetic couplings can result in inter-plane cancellations in a cubic crystal. To our knowledge, there is no theoretical model as yet (apart from mean field theory) that would predict such behavior.

While the evidence we have presented as to whether EuSe constitutes a 2D magnetic system is certainly not flimsy, more direct experimental evidence would surely be desirable. We propose two critical experiments that could help decide the issue even more definitively.

One experiment would be to measure the entire magnetization curve of EuSe using μ SR, exactly as was done recently with EuO.¹³ It should be possible to come closer to T_c and to get an independent value of D . Additionally, by applying a small external magnetic field or better yet, hydrostatic pressure (as in Ref. 4), a direct change in the β values from ~ 0.20 to 0.33 should be observable when going from the antiferromagnetic to the ferromagnetic state. This could pose a significant challenge to the currently held universality hypothesis in critical phenomena.

The most unequivocal determination of whether EuSe is or is not a 2D magnetic system would be through a neutron scattering measurement. The measurement would consist of a determination of the dispersion curves in EuSe, exactly analogous to those made in EuS.¹⁴ For EuSe only the dispersion in the (111) direction may need to be measured (and thus could possibly be done with natural-Eu samples). A flat dispersion curve, such as was obtained in the case of K_2MnF_4 (see Fig. 5 of Ref. 12) would unequivocally show the two-dimensionality of EuSe.

In summary, we have carried out ⁷⁷Se NMR in EuSe and on the basis of analyses of these as well as other data, presented empirical support for the idea that EuSe is, contrary to conventional expectations, a 2D magnetic system. Inelastic neutron scattering experiments are needed to obtain a definitive result. A positive outcome could be of great import because it would seriously put into question the long-held belief that magnetic interactions in the prototype Heisenberg systems, the europium chalcogenides, take place primarily via short-range Heisenberg exchange interactions (and by implication perhaps in most other simple insulating magnetic systems).

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