

# NMR Measurements of Power-Law Behavior in the Spin-Wave and Critical Regions of Ferromagnetic EuO

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Precision continuous-wave NMR measurements have been carried out over the entire magnetization curve of EuO and are presented in tabular form. Two very closely spaced resonances are observed and are attributed to domain and domain-wall signals. Both of the signals are useful for analysis in the spin-wave region. Only the domain signal is measurable above  $\sim 50$ K. The latter is used for fitting  $T_c$  and the critical exponent  $\beta$ . The critical-region fits agree with previous measurements, within experimental error. The low-temperature data exhibit a clear-cut  $T^2$  behavior, at variance with the expectations of conventional spin-wave theory. This result is discussed in relation to two semi-empirical spin-wave schemes, one formulated by N. Bykovetz, and one by U. Koebler. The NMR signal at 4.2K gives no indication of a quadrupole splitting, in contradiction to the interpretation of several previous spin-echo NMR spectra observed in EuO. This issue remains unresolved.

## I. INTRODUCTION

Precision continuous-wave (CW) nuclear magnetic resonance (NMR) measurements over the entire magnetization curve of EuO have been carried out and the behavior of these data is analyzed both in the spin-wave and in the critical regions. The low-temperature data exhibit a clear-cut  $T^2$  behavior, at variance with the expectations of conventional spin-wave theory. The critical region power-law fit, however, agrees with other previous measurements, within experimental error.

Currently, there is still great interest in the study of EuO because of the varied and multifaceted characteristics it displays. Among others, oxygen-rich or Gd-doped EuO shows colossal magnetoresistance, while Eu-rich EuO exhibits a change in conductivity of 13 orders of magnitude in its insulator to metal transition<sup>1</sup>. Some current studies of EuO have a practical angle (spintronics), while others focus on testing theories (such as the Kondo-lattice model). While much is not yet clear in this broader area, the fundamental behavior of the localized spin (pure Heisenberg) model in EuO is also being put into question by recent measurements<sup>2</sup>. The results of the current low-temperature data will hopefully shed additional light on this latter issue. The principal focus of this paper is the unconventional (spin-wave) behavior of EuO at low temperatures in the context of a pattern of such behavior in other simple magnetic systems. In addition, the critical-region parameters are examined.

While a strict  $T^2$  low-temperature behavior in pure EuO was reported<sup>3</sup> early on, those measurements were not backed up with numerical evidence. The current measurements are presented in tabular form (see Table I)

and show a linear-fit to  $T^2$  with an excellent  $R^2 = 0.9999$ . In recent years, two alternative semi-empirical spin-wave schemes have been proposed to explain the simple power-law behaviors displayed by magnetic systems (in many cases not recognized by the original authors), beginning with the very first NMR measurement of a magnetically ordered system,  $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ <sup>4</sup>. One scheme was developed by U. Koebler<sup>5</sup>, and one by N. Bykovetz<sup>6,7,8</sup>. The  $T^2$  behavior found in the current EuO NMR data is consistent with both schemes.

TABLE I. NMR Frequency (MHz) vs.  $T(K)$  for <sup>153</sup>EuO

$T_H$	Freq	$T_L$	Freq	$T_L$	Freq	$T_L$
4.200	138.500		69.703	62.519	41.715	67.570
13.981	136.080		68.562	62.807	40.151	67.741
17.990	134.190	17.830	66.654	63.286	38.681	67.882
21.269	132.240	21.127	65.730	63.507	38.654	67.888
25.850	129.050	25.701	63.776	63.973	37.674	67.974
26.991	128.090	26.838	60.773	64.608	37.230	68.015
28.637	126.715	28.491	58.989	64.976	36.833	68.055
32.188	123.523	32.014	57.067	65.347	36.471	68.082
34.360	121.370	34.189	53.371	65.993	35.736	68.144
38.075	117.352	37.945	52.340	66.160	34.782	68.221
38.926	116.350	38.773	51.135	66.357	34.442	68.250
42.402	112.020	42.277	51.019	66.369	34.121	68.271
43.180	111.000	43.023	49.248	66.643	33.663	68.312
47.488	104.720	47.357	47.544	66.852	33.086	68.357
	98.822	50.873	46.182	67.044	32.585	68.397
	92.680	54.130	46.088	67.060	31.903	68.439
	78.507	59.914	44.385	67.266	31.812	68.445
	73.502	61.444	43.945	67.322	31.748	68.451
	71.795	61.933	43.275	67.399	31.600	68.460
	70.723	62.223	43.154	67.411		

Using the scheme of Bykovetz, it is argued here that the  $T^2$  behavior seen in the EuO NMR constitutes evidence against a pure localized-spin model as conceived in the conventional Dyson picture and formalism of magnetic interactions in ferromagnets.

Our NMR determination of critical parameters agrees with previous results using non-NMR methods<sup>9,10</sup>. How-

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ever, it is an open question why the two so-called ideal Heisenberg ferromagnets EuO and EuS do not have more nearly identical critical exponents,  $\beta$  ( $\beta = 0.368$  in EuO and  $0.33$  in EuS). EuS appears to exhibit the same  $\beta$  as does  $\text{MnF}_2$ <sup>11</sup>, at variance with most isotropic 3D magnets.

Lastly, we note that a discrepancy still appears to exist between CW and spin-echo NMR measurements in EuO. The current NMR data, like the original CW data of Boyd<sup>12</sup>, give no indication of significant quadrupole splitting of the NMR resonance, whereas spin-echo measurements, which involve multi-layered processing of the raw data<sup>1</sup>, appear to show the presence of a substantial electric field gradient. An extensive discussion of this issue was presented in Ref. (1), but the issue still remains unresolved.

## II. EXPERIMENT

NMR measurements on polycrystalline EuO were carried out using a frequency-modulated spectrometer in zero external field. The temperature was determined by means of a platinum resistance thermometer. Because of the strong broadening of the NMR signal on approaching  $T_c$ , it was decided to carry out the measurements by controlled temperature sweeps at fixed frequencies (similar to the method used in references (11) and (13)). For consistency, all data, except the helium-bath (4.2K) data point, were determined in this way.

## III. RESULTS AND DISCUSSION

A single narrow ( $\sim 25$  kHz) NMR line was observed at 4.2K by sweeping the frequency, as was the case in the original CW measurement of Boyd<sup>12</sup>. No indication of quadrupolar splitting was evident. At temperatures above 17K, where the frequency was kept constant, and the NMR signal observed by sweeping the temperature, two closely spaced and overlapping signals were observed all the way up to  $\sim 50$ K. The signal that peaks at the lower temperatures will be designated as  $T_L$ , and at higher temperatures as  $T_H$ . Fig. 1 shows the behavior of the  $T_H$  signal relative to  $T_L$  at three representative temperatures. One can see that the  $T_L$  peak tapers off in intensity relative to the  $T_H$  peak and becomes undetectable at 4.2K, whereas the  $T_H$  peak becomes progressively smaller as 50K is approached, and is undetectable above this temperature where only the  $T_L$  signal is observable. Near 27K, the signals have equal intensities.

The  $T_H$  signal is ascribed to NMR from domain-walls, since the domain-wall enhancement of NMR signals is strongest at lowest temperatures. The  $T_L$  signal must, therefore come from the nuclei in the domains, since the domain walls become less extensive and their enhancement factor decreases as  $T_c$  is approached. Comment, et al.<sup>1</sup>, presented an argument that in EuO, the domain-wall signal enhancement should be relatively small, but our results do not seem to support this contention.

Additional support for the assignment of the two NMR signals as originating in domain and domain-wall nuclei

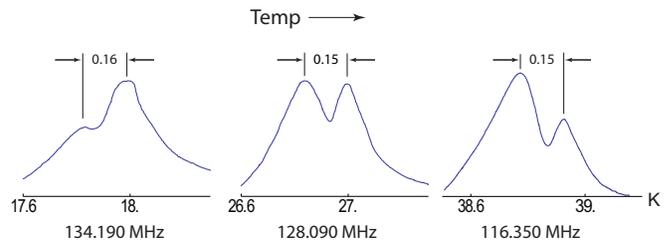


FIG. 1. NMR temperature-scans of EuO at fixed frequencies at  $\sim 17$ ,  $27$ , and  $39$ K, shown with the intensities normalized to the same height. The left peak ( $T_L$ ) increases while the right peak ( $T_H$ ) decreases with increasing temperature.

comes from the temperature independence of the separation of the peaks in the doublet. It can be seen from the data in Table I that the separation  $\Delta T = T_H - T_L$  between the observed double peaks (cf., Fig 1) is constant, with  $\Delta T = 0.149 \pm 0.0124$ K (on the average), over the temperature range where both signals could be measured. As will be seen, both the  $T_H$  and the  $T_L$  curves give an excellent fit to a  $T^2$  power-law. Consequently, it can be shown that a constant temperature separation translates into a curve separation,  $\Delta\nu \propto T$ . A similar  $\Delta\nu = \nu_H - \nu_L \propto T$  separation was observed in  $\text{CrBr}_3$ ,<sup>14</sup> where the identification of which resonance came from domain walls and which from within the domains was more clear-cut. It is to be noted that, as a consequence, if one of the curves ( $\nu$  vs.  $T_L$  or  $T_H$ ) goes as  $BT^2$ , the other cannot be a pure  $T^2$  curve, but must have an additional small corrective term,  $(2B\Delta T)T$ . It is conjectured that this term should be part of the  $T_L$  curve, although the data are not precise enough to make a definitive determination using fitting techniques.

Focusing on data in the critical region, the following can be seen. Fits<sup>15</sup> of the data (to the standard equation  $D(1 - T/T_c)^\beta$ , re-cast in the form of a power law) give a  $\beta$  of  $0.366$  for  $T$  data points within 10% of  $T_c$ , and a  $T_c$  of  $69.23$ K. The parameter  $D = 1.18$ . Fitting only points closer to  $T_c$ , results in a small, but not a significant increase in  $\beta$  as well as  $T_c$ . Thus, for the temperature interval within 3% of  $T_c$ ,  $\beta$  becomes  $0.378$  and  $T_c$   $69.28$ K. The determinations of  $\beta$  agree with previous macroscopic<sup>9</sup> and neutron-scattering<sup>10</sup> results, but are at odds with the Mossbauer<sup>16</sup> determination. A check for frequency-pulling effects near  $T_c$  was made by measuring the temperature dependence of the ratio of NMR frequencies in  $^{151}\text{EuO}$  to that in  $^{153}\text{EuO}$  (see Table II). The lack of a systematic dependence of the ratio on temperature proves that any such effect is negligible. The value of the ratio of  $^{151}\text{Eu}$  to  $^{153}\text{Eu}$  frequencies is, however, lower than in  $\text{EuS}$ <sup>13</sup>, but agrees with the EuO measurement at 4.2K in Ref. 1 (see footnote 40).

In the low-temperature (spin-wave) regime, both the  $T_H$  and the  $T_L$  data can be fitted with a simple power-law of the form  $\nu = A + BT^c$ . For the domain-wall,  $T_H$ , data, one obtains a power law with an exponent of

TABLE II.  $T$  vs.  $\nu$  for  $^{153}\text{Eu}$  and  $^{151}\text{Eu}$ 

$T(K)$	$\nu_{153}(MHz)$	$\nu_{151}(MHz)$	$\nu_{151}/\nu_{153}$
67.971	37.718	84.875	2.250
65.811	54.451	122.665	2.252
65.036	58.690	132.312	2.254
64.566	60.991	136.741	2.242

$2.04 \pm 0.01$ , by including all data points, except the one at 47.488K, which is already showing deviation to below the spin-wave  $T^2$  curve. Fig 2 shows a plot of the  $\nu$  vs.  $T_H^2$ , displaying the excellent fit. When this data set is fit to a  $T^2$  power law, one gets  $\nu_0 = 138.93\text{MHz}$ , and an  $R^2$  of 0.99992. For the  $T_L$  data below  $\sim 43\text{K}$ , the best-fit exponent is  $2.035 \pm 0.011$ . When fitted to a straight line as a function of  $T^2$ , one obtains  $\nu_0 = 138.96\text{MHz}$  and an  $R^2$  of 0.99994.

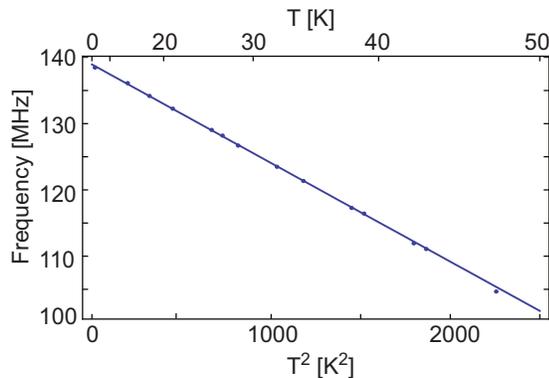


FIG. 2.  $\nu$  vs.  $T^2$  in the spin-wave region. Deviation below the  $T^2$  line begins above  $\sim 43\text{K}$ .

The  $T^2$  behavior is in agreement with the expectations of Koebler’s scheme<sup>5</sup>, wherein it is claimed that all isotropic 3D magnetic systems should exhibit a  $T^2$  behavior in the low-temperature region. We disagree with the generalization to all magnets, the prime counter-example being the  $T^{1.71}$  behavior of  $\text{CrBr}_3$ . (see Ref. 6). However, the  $T^2$  spin-wave region behavior of  $\text{EuO}$  follows the  $n = 2$  behavior described within the broader semi-empirical scheme devised by Bykovetz<sup>6</sup>, wherein both 3D and 2D ferromagnets show power-law behaviors depicted in Table III. The power-law behaviors are derivable from the formula  $\Delta M \propto (T^{\frac{3}{2}})^{f(n)}$ , where  $f(n) = 1/(1-(1/2)^n)$ , with  $n = 1, 2, 3$  for 3D and  $n = 1, 2$  for 2D magnets, where  $T^{\frac{3}{2}}$  is replaced by  $T$ . Note that other ferromagnets, e.g.,  $\text{CrI}_3$ ,<sup>17</sup> and the near BCC structured compounds,  $\text{M}_2\text{CuX}_4 \cdot 2\text{H}_2\text{O}$ <sup>18</sup>, exhibit a  $T^2$  behavior, not noticed by the original authors<sup>19</sup>.

The scheme of Bykovetz, if further validated, implies that the conventional Bloch result ( $T^{\frac{3}{2}}$  for 3D and  $T^1$  for 2D) determines the power-law behaviors, but with an absence of any lattice-dependent effects (i.e., the Dyson  $T^{\frac{5}{2}}$  term, expected in 3D ferromagnets, is missing). The gen-

TABLE III. Ferromagnetic Exponents

$n$	$D=3$	$D=2$	$D=1$
3	$T^{1.71}$		
2	$T^2$	$T^{1.33}$	
1	$T^3$	$T^2$	$T$

eral implication would be that even in localized-spin magnetic systems such as the europium chalcogenides, the exchange interactions are somehow long-ranged, and/or the spin waves propagate in a spin-density-polarized medium (band). It is well known that a model of ferromagnetism, in which the magnetization can vary continuously (as opposed to having discrete spins at localized sites) results in spin waves that display a pure  $T^{\frac{3}{2}}$  behavior with no “lattice-correction” terms. The recent optical measurements by Miyazaki, et al.<sup>2</sup>, suggest that, in fact, the Heisenberg-Dyson picture of localized exchange interactions may not be quite correct.

Last but not least, the measurements above, particularly the 4.2K domain-wall measurement, as well as those of Boyd<sup>12</sup>, give no evidence of a quadrupole splitting, whereas spin-echo NMR measurements invariably do. An extended discussion of this problem is given in Ref 1. The issue at this point is unresolved. One could conjecture that the domain-wall resonance in CW measurements somehow averages over the electric field gradients. On the spin-echo side, the source of the difficulty may lie in the uncertainties of the multi-layered analyses of the complex spin-echo measurements (see Ref 1).

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<sup>15</sup>All fits in this paper were made using *Mathematica*.

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<sup>19</sup>The low-temperature data from Ref 18 appear in tabular form in Ref 8.