Critical spin fluctuations in EuO

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(Received 14 January 1980)

Zero-field electron-spin resonance was used to measure the spin-relaxation time above T_c in the isotropic ferromagnet EuO. These measurements show an asymptotic behavior in the reduced temperature region $t \leq 0.02$. The dynamic exponent z was determined for this asymptotic region to be $z = 2.04(7)$. Longitudinal relaxation measurements of the kinetic coefficient in EuO of Kötzler *et al.* [Solid State Commun. 26, 641 (1978)] have been reanalyzed and provide an exponent $z = 1.93(10)$ in the asymptotic region. Both values of z agree with theoretical predictions for a dipolar dominated region in κq space.

I. INTRODUCTION

Recently theoretical calculations by Teitelbaum[1] and Raghavan and Huber[2] on isotropic ferromagnets have predicted a crossover in dynamic properties as a function of temperature. Far above T_c they predict a region in which spin nonconserving dipolar interactions may be considered as perturbations to the exchange Hamiltonian, and close to T_c they predict a region dominated by dipolar interactions. Hyperfine interactions experiments [NMR, perturbed angular correlation (PAC), and Mössbauer effect] have measured the dynamic exponent $z \simeq 2.0$, consistent with this predicted spin nonconserving behavior close to T_c.[3-6] Neutron scattering experiments, on the other hand, have typically measured z near 2.5.[7,8] The exception to this is the measurement in EuO which yielded $z = 2.29(3)$.[9]

A possible explanation for this apparent discrepancy has been given by Suter and Hohenemser[10] who suggest that the effective value of z depends on the values of the wave vector q, sampled. They note that neutron scattering experiments are limited to measuring linewidths for $q \gtrsim 0.05$ Å^{-1} while hyperfine interaction methods determine an integral over all wave vectors which is weighted toward small q for small values of the reduced temperature. Thus neutron scattering may detect Heisenberg-like behavior typical of large q while hyperfine interactions detect spin nonconserving behavior typical of small q.

In order to confirm the hypothesis of crossing in q it is important to have a measurement of the exponent z at a small, definite value of q. Zero-field electron-spin resonance (ESR) measures only fluctuations in the $q = 0$ mode and is thus an ideal method for observing the small-q behavior.

Recently we reported on zero-field ESR measurements in EuO and deduced from this values of the Onsager kinetic coefficient.[11] In the present paper we reconsider our data and calculate a value for the dynamic exponent z explicitly from the measured spin-relaxation times. We also consider the longitudinal relaxation measurements of Kötzler *et al.*[12] and obtain a value of z from these. Both our work and that of Kötzler are consistent with $z \simeq 2.0$. These results are compared with the neutron scattering measurements of Dietrich *et al.*[9] in EuO and are discussed in terms of a crossover in κq space.

II. THEORY OF CRITICAL SPIN FLUCTUATIONS

To define the critical exponent z in terms of the spin autocorrelation time we use the dynamic scaling theory described by Hohenberg and Halperin.[13] the relaxation rate of the ith spin component depends on the wave vector q and the inverse correlation length κ via

$$\omega_{ii}(q) = q^z \Omega_{ii}(\kappa/q) \quad , \tag{1}$$

where $\Omega_{ii}(\kappa/q)$ is the dynamic scaling function. For an isotropic system, there is only a single relaxation $\omega(q) = \omega_{ii}(q)$ and a single scaling function $\Omega(\kappa/q) = \Omega_{ii}(\kappa/q)$. The relaxation rate $\omega(q)$ is a homogeneous function of κ and q; Eq. (1) may thus be alternately expressed[14]

$$\omega(q) = \kappa^z \Omega'(\kappa/q) \quad , \tag{2}$$

where

$$\Omega'(\kappa/q) = (q/\kappa)^z \Omega(\kappa/q) \quad .$$

ESR determines a relaxation rate which is the inverse spin autocorrelation time[15] at $q = 0$:

$$\omega(0) = \tau^{-1} = \kappa^z \Omega'(\infty) \quad . \tag{3}$$

The temperature dependence on the right-hand side of Eq. (3) is contained in the κ^z term. For reduced temperature $t \equiv T/T_c - 1$,

$$\kappa = \kappa_0 t^\nu \quad , \tag{4}$$

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hence we may write

$$\omega(0)=\tau^{-l}=(\text{const})t^{\nu z} \quad . \tag{5}$$

It is this expression which is used by us to determine the product νz from the ESR measurements of the relaxation time τ. To find z, we use appropriate values of the exponent ν given in the literature (ν is a static exponent and is not expected to depend on the details of the dynamics, as does z).

III. EXPERIMENTAL METHODS

In our experiment we measure a relaxation time τ_{ext} via the absorptive part of the complex susceptibility χ''_{ext}. (The subscript "ext" denotes that we are dealing with "external" or shape-dependent quantities which must be corrected for the effects of demagnetization.) The absorptive part of the susceptibility is expressed as a Lorentzian[16]

$$\chi''_{ext}=\frac{\chi_{ext}(0)\,\omega_{rf}\tau_{ext}}{1+(\omega_{rf}\tau_{ext})^2} \quad , \tag{6}$$

where ω_{rf} is the applied radio frequency and $\chi_{ext}(0)$ is the shape-dependent static susceptibility. This quantity is related to the corresponding shape-independent or external quantity by[17]

$$\chi_{ext}^{-1}(0)=\chi^{-1}(0)+N \quad , \tag{7}$$

where N is the demagnetization factor.

For EuO $\chi(0)$ has been measured by Menyuk *et al.*[18] and fitted by

$$\chi(0)=At^{-\gamma} \quad , \tag{8}$$

where $A=5.11\times10^{-3}$ emu/cm^3 and $\gamma=1.29(2)$. For our sample, a $0.208\times0.212\times0.688$ cm^3 parallelepiped, the demagnetization factor was independently measured for us by Foner with a vibrating magnetometer,[19] with the result $N=1.3(1)$. In our experiment $\omega_{rf}\tau_{ext}<0.02$; therefore Eq. (6) may be approximated by

$$\tau_{ext}=\frac{\chi''_{ext}}{\omega_{rf}\chi_{ext}(0)} \quad . \tag{9}$$

The measured external relaxation time is corrected for the effects of demagnetization by:

$$\tau=\tau_{ext}[1+N\chi(0)] \quad . \tag{10}$$

This expression is obtained from Eq. (7) and the fact that the Onsager kinetic coefficient, defined $\Gamma\equiv\chi(0)/\tau$, is a shape-independent quantity, thus

$$\chi_{ext}(0)/\tau_{ext}=\chi(0)/\tau \quad .$$

From Eqs. (7), (9), and (10) we obtain

$$\omega(0)=\tau^{-1}=\chi^{(0)}\frac{\omega_{rf}}{\chi''_{ext}}[1+N\chi(0)]^{-2} \quad . \tag{11}$$

Through N as measured by Foner, $\chi(0)$ as measured by Menyuk *et al.*,[18] and χ''_{ext} as measured by us, we arrived via Eq. (11) at experimental values of $\omega(0)=\tau^{-1}$ for various values of the reduced temperature.

IV. APPARATUS

Our ESR spectrometer is similiar to that described by Gottlieb *et al.*[20] and has a detector arrangement as described by Grambow and Weber.[21] A block diagram is shown in Fig. 1. The sample is contained in the coil of the L-C resonator and the frequency of the rf generator is locked to the resonant frequency of this L-C circuit. Locking is accomplished by using a Hewlett-Packard 8405A vector voltmeter to measure the rf phase shift introduced by the resonator. At resonance this phase shift is found to be zero. The vector voltmeter phase output gives 0 to ±0.5 V dc, the sign indicating whether the rf frequency is too high or too low. To keep the frequency at resonance, this signal is fed back to the frequency control input of the rf generator (HP 608F).

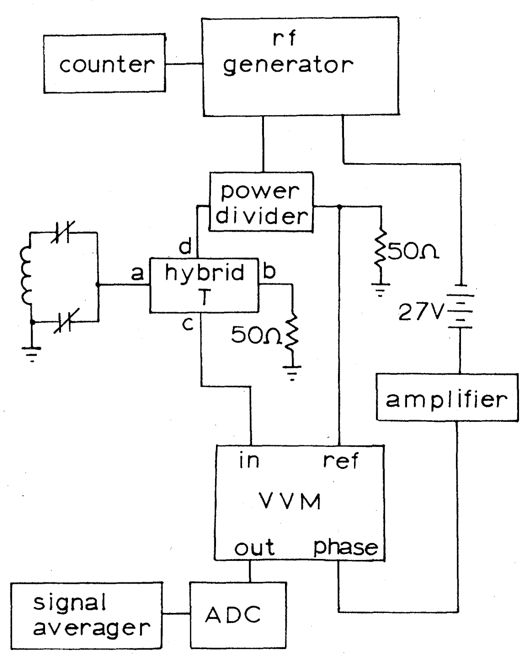

FIG. 1. Block diagram of the ESR spectrometer. VVM is a vector voltmeter, ADC an analog-to-digital converter.

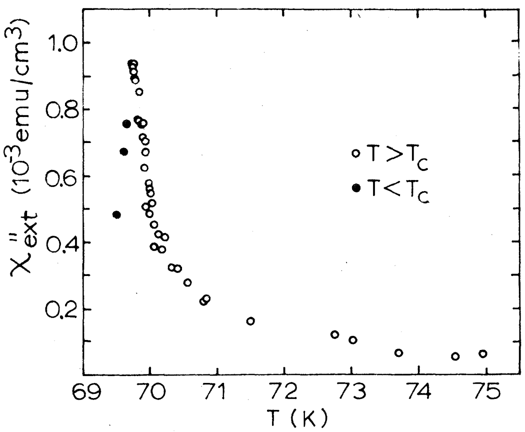

FIG. 2. Absorptive part of the complex susceptibility, χ''_{ext}, as a function of temperature. The divergence of this quantity was used to determine $T_c = 69.72(1)$ K.

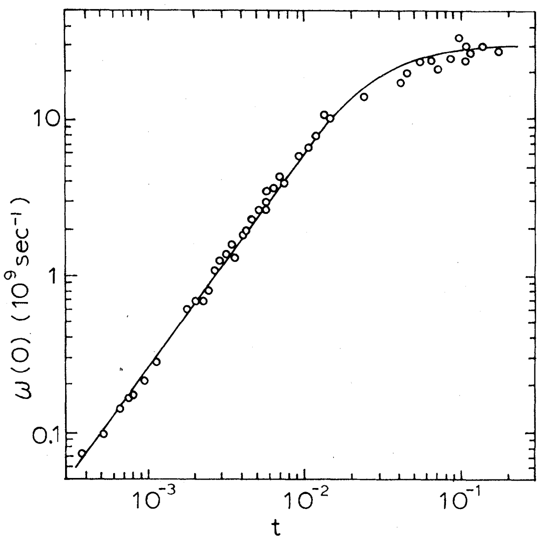

FIG. 3. Temperature dependence of the shape-independent relaxation rate $\omega(0)$, in EuO.

The absorptive part of the complex susceptibility is measured using the vector voltmeter to monitor the voltage drop across the resonator. A change in this component of the susceptibility is expressed in terms of a change in the output voltage of the vector voltmeter ΔV, as[20]

$$\Delta\chi''_{ext} = \frac{\Delta V}{\pi\eta Q a V_{rf}} , \tag{12}$$

where η is the sample filling factor, Q is the quality factor of the resonator, a is the vector voltmeter gain setting, and V_{rf} is the rms output of the rf generator. At a fixed frequency of about 20 MHz, field sweeps are made from $H = 0$ to $H \simeq 6.5$ kOe. For this work

$$\chi''_{ext}(H = 6.5 \text{ kOe})/\chi_{ext}''(H = 0) < 0.02 ,$$

so we make the approximation $\Delta\chi''_{ext} \simeq \chi''_{ext}(H = 0)$.

The Curie temperature is obtained by monitoring χ''_{ext} as a function of temperature. This absorption diverges at T_c as is shown in Fig. 2. Below T_c the absorption line becomes progressively more non-Lorentzian and enables us to differentiate between spectra immediately above T_c and those immediately below. This method has provided $T_c = 69.72(1)$ K.

V. RESULTS

Measured values of χ''_{ext} and deduced values of $\omega(0)$ are given in Table I. A plot of $\omega(0)$ as a function of reduced temperature is shown in Fig. 3. These data are fitted with the function $\omega(0) \propto t^{\nu z}$. A range of fit analysis,[22] shown in Fig. 4, gives an asymptotic value of $\nu z = 1.42(5)$. This analysis shows an asymptotic region for $t \leq 0.02$ and a deviation from a single power law for larger t. Using the theoretical value,[23] $\nu = 0.70$, we obtain $z = 2.04(7)$. A similar range of fit analysis on relaxation rates calculated from the longitudinal relaxation measurements of the kinetic coefficient of Kötzler *et al.* in EuO provides an exponent $z = 1.93(10)$.[15]

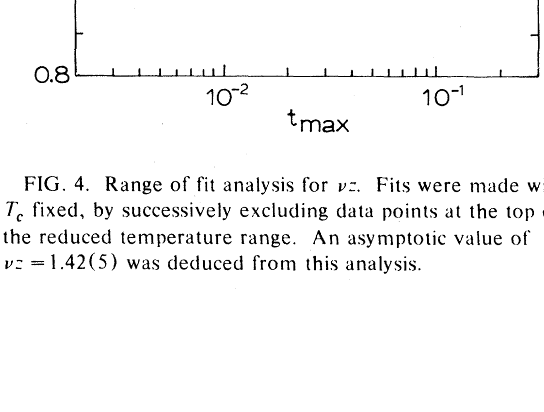

FIG. 4. Range of fit analysis for νz. Fits were made with T_c fixed, by successively excluding data points at the top of the reduced temperature range. An asymptotic value of $\nu z = 1.42(5)$ was deduced from this analysis.

TABLE I. Values of χ''_{ext} and $\omega(0)$ as a function of temperature.

T (K)	t	$\omega_{rf}/2\pi$ (MHz)	χ''_{ext} (10^{-3} emu/cm^3)	$\omega(0)$ (10^{10} sec^{-1})
82.798	0.188	21.221	0.183	2.77
79.626	0.142	22.169	0.246	3.00
78.112	0.120	20.253	0.267	2.96
77.668	0.114	20.853	0.330	2.59
77.652	0.114	20.627	0.330	2.61
77.587	0.113	20.879	0.288	3.03
77.559	0.112	20.827	0.260	3.36
76.352	0.0951	23.044	0.449	2.54
74.976	0.0754	23.496	0.695	2.09
74.516	0.0688	23.481	0.660	2.39
73.709	0.0572	23.467	0.758	2.42
73.076	0.0481	23.499	1.02	2.04
72.754	0.0435	20.817	1.22	1.77
71.554	0.0263	20.384	1.65	1.44
70.820	0.0158	20.234	2.36	1.01
70.795	0.0154	20.259	2.28	1.05
70.560	0.0121	20.207	2.80	0.791
70.484	0.0110	19.989	3.25	0.645
70.398	0.00972	20.206	3.33	0.598
70.240	0.00746	20.193	4.24	0.397
70.220	0.00717	19.935	3.74	0.431
70.218	0.00714	20.141	3.93	0.415
70.184	0.00666	20.117	4.29	0.358
70.138	0.00600	19.883	5.20	0.269
70.134	0.00594	20.093	4.54	0.309
70.134	0.00594	20.015	3.88	0.260
70.092	0.00534	20.053	4.94	0.258
70.065	0.00495	22.544	5.54	0.242
70.032	0.00448	20.176	5.61	0.195
70.031	0.00446	19.935	5.87	0.183
69.990	0.00387	19.716	7.05	0.131
69.962	0.00347	20.059	5.07	0.164
69.954	0.00336	19.875	5.84	0.138
69.930	0.00301	20.017	5.55	0.130
69.915	0.00280	20.899	6.26	0.106
69.896	0.00252	22.838	8.51	0.0797
69.886	0.00238	19.880	7.76	0.0712
69.868	0.00212	19.657	6.93	0.0692
69.854	0.00192	19.972	7.04	0.0616
69.804	0.00120	19.700	8.95	0.0273
69.791	0.00102	19.499	9.31	0.0211
69.778	0.00083	19.463	9.23	0.0166
69.776	0.00080	19.502	9.22	0.0159
69.768	0.00069	19.808	9.00	0.0136
69.757	0.00053	19.502	9.16	0.00950
69.747	0.00039	22.909	9.39	0.00730

VI. DISCUSSION

Both our data and those of Kötzler *et al.*[12] agree well with the prediction of Teitelbaum[1] of a dipolar dominated region close to T_c with $z \simeq 2.0$. This crossing to an asymptotic dipolar region has also been observed in the other low T_c isotropic ferromagnet EuS,[17] and is suggested by measurements on $CdCr_2S_4$ and $CdCr_2Se_4$.[24,25]

The crossing to a dipolar region as a function of temperature at $q=0$ as predicted by Teitelbaum[1] may be explained in terms of the crossover from Heisenberg-like to spin nonconserving behavior suggested by Suter and Hohenemser[10] in the following way. Equations (1) and (2) show an equivalence between q and κ based on the homogeneous nature of the relaxation function. Thus a crossing in q implies a crossing in κ and hence in temperature. Figure 5 illustrates the dipolar and crossover regions in κq space for EuO. The innermost region is dominated by dipolar interactions and is defined on the $q=0$ axis by the limit of the asymptotic behavior observed by us. The center of the crossover region is defined on the $q=0$ axis by the inverse correlation length corresponding to $4\pi\chi(0)=1$, and on the $\kappa=0$ axis by the dipolar wave vector q_d.[12]

In Fig. 5 we observe that the crossover region in EuO extends out into the noncritical region. This results from the relatively short wavelength of the dipolar fluctuations in EuO and the large extent of the crossover region. Thus EuO may be described by a dipolar dominated region close to T_c and a crossover region as described by Raghavan and Huber[2] for $0.02 \leq t \leq 0.1$. For $t > 0.1$ the behavior becomes noncritical.[9]

The range of q values covered by the neutron scattering measurements of Dietrich *et al.*[9] is shown on the $\kappa=0$ axis in Fig. 5. These measurements provide $z=2.29(3)$. Dietrich *et al.* have explained their failure to measure $z=2.5$, typically found via neutron scattering experiments, as resulting from the influence of dipolar interactions. They predict a deviation from the Heisenberg-like value of $z=2.5$ near $q=q_d$, which is well within the range of experimentally studied wave vectors. We see from Fig. 5 that this is certainly the case. In fact we observe that the neutron scattering measurements are spread over a large portion of the crossover region. Thus on the basis of Fig. 5 we would expect that the exponent measured by neutron scattering would not be asymptotic but would be intermediate between $z=2.0$ and 2.5.

The "tail" on the crossover region along the $q=0$ axis in Fig. 5 results from the fact that the spin-conserving exchange interactions responsible for the Heisenberg-like behavior observed in some isotropic ferromagnets have no normal $q=0$ mode of decay.

The kinetic coefficient, $\Gamma = \chi(0)\omega(0)$, is plotted in cgs units in Fig. 6. Our results agree well with the longitudinal relaxation measurements of Kötzler *et al.*[12] The prediction of Raghavan and Huber[2] that $\Gamma \propto \chi^{7/4}$ for the crossover region is shown by the solid line in the figure and the value of the dipolar kinetic coefficient calculated by Finger[26] is indicated by the broken line.

In conclusion we have found that the dynamics of

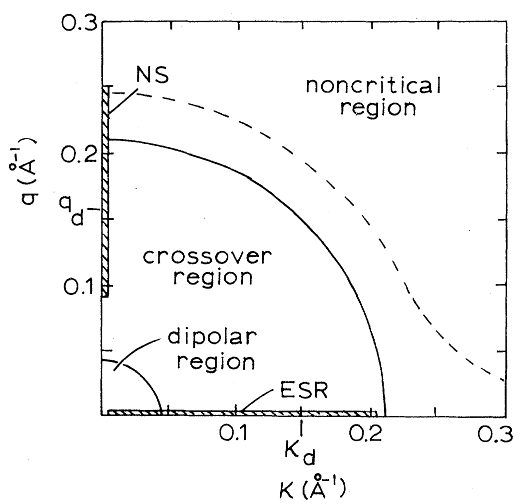

FIG. 5. Dipolar region in κq space for EuO. The dipolar dominated region is shown by the innermost region in the diagram. Regions along the axes sampled by neutron scattering (NS) (Ref. 9) and ESR (Ref. 11) are indicated. The expected outer limit of the crossover region, indicated by the broken line, lies outside of the critical region.

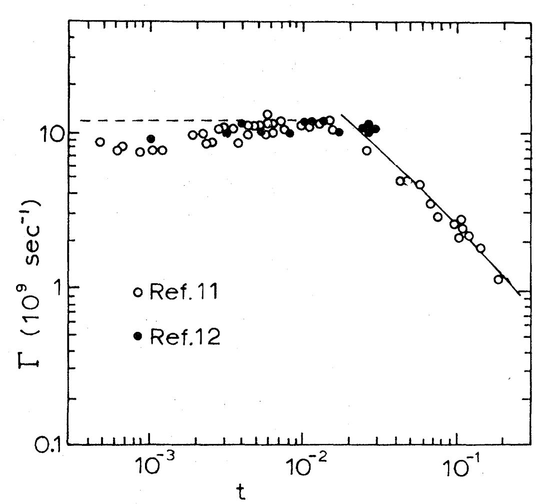

FIG. 6. Temperature dependence of the kinetic coefficient. The solid line represents the calculations of Raghavan and Huber (Ref. 2) for the crossover region, and the prediction of Finger (Ref. 26) is indicated by the broken line.

EuO agree well with the theory that dipolar interactions are dominant close to T_c and are responsible for the $z \simeq 2.0$ behavior. In particular we have confirmed that the discrepancy between neutron scattering and $q = 0$ methods may be explained in terms of a crossover in κq space. We have also explained that $q = 0$ experiments in EuO have not observed asymptotic Heisenberg-like behavior for two reasons. (i) The exchange interactions responsible for this behavior have no $q = 0$ mode of decay and are thus not detectable with these techniques. (ii) There is no asymptotic Heisenberg-like region in EuO because of the location and extent of the crossover region.

ACKNOWLEDGMENTS

The authors are most grateful to Dr. S. Foner of the Francis Bitter National Magnet Laboratory for providing demagnetization measurements of our sample. We would also like to thank Dr. C. Hohenemser, Dr. R. M. Suter, Dr. H. Gould, and Mr. Lee Chow for their comments on this manuscript and Dr. J. Kötzler for his helpful suggestions and for supplying numerical data for his relaxation measurements. This research was supported in part by National Science Foundation Grant No. DMR 7711307 and a grant from the Research Corporation.

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