Temperature Dependence of the Spin Dynamics of EuO

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Neutron–inelastic-scattering techniques have been used to study the spin dynamics of EuO. High-quality single crystals were used so that temperature-dependent line shapes could be obtained at all momentum values. It was found that the magnetic excitations above T_c changed from Lorentzian-like peaks centered at zero energy near the zone center to rather well-defined spin-wave–like excitations at the zone boundary.

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There has been a great deal of interest in the nature of the paramagnetic state of ferromagnetic materials, with particular emphasis on the transition metals like nickel and iron.[1] Neutron-scattering results on these materials have shown that measurements of the magnetic inelastic scattering look similar below and above the Curie point as long as the momentum transfer q is not too small.[2,3] In fact, constant-energy scans taken in the region around $\frac{1}{4}$ of the way to the zone boundary do not change dramatically over a fairly large temperature range above and below the Curie point T_C. High-resolution measurements do show broadening and softening of the spin waves as the temperature is increased through T_C, but the line shapes are still remarkably well defined. There has been some controversy on whether or not the excitations above T_C are propagating modes.[1] The neutron-scattering measurements cannot tell this directly; however, the measurements on nickel and iron above T_C do not resemble the expected behavior for a Pauli paramagnet, or, for that matter, a Heisenberg magnet like EuO for a similar momentum transfer. To elucidate this point, spin-wave measurements on EuO single crystals have been made over a wide range of temperature and momentum transfers. It is found that spin-wave scattering merges to a peak centered at zero energy above T_C for momentum transfers in the vicinity of $\frac{1}{4}$ of the zone boundary in agreement with earlier measurements. However, as q is increased toward the zone boundary, discrete spin-wave peaks are found to persist well above T_C. One finds thus that the behavior of the magnetic scattering crosses over the peaks centered at zero energy at low q to behavior more like nickel and iron at the zone boundary. Since the magnetic interactions in EuO appear to be much simpler than in nickel or iron, perhaps a theoretical understanding of this behavior is easier to obtain.

Extensive neutron-scattering measurements have been made on EuO powders by Passell, Dietrich, and Als-Nielsen.[4] The new data obtained on single crystals is in very good agreement with the older powder data in all regions where the measurements overlap. Passell, Dietrich, and Als-Nielsen were able to measure spin-wave dispersion curves with the EuO powders and thus obtain the first- and second-neighbor exchange constants J_1 and J_2. Even though the single-crystal measurements are much more straightforward than the powder measurements, it was difficult to improve greatly on their earlier results. Passell, Dietrich, and Als-Nielsen obtained $J_1/k = 0.606 \pm 0.008$ K and $J_2/k = 0.119 \pm 0.015$ K, where the interaction between pairs of spins is $-2J_{nm}\vec{S}_n \cdot \vec{S}_m$ and k is Boltzmann's constant. The single-crystal measurements yield $J_1/k = 0.625 \pm 0.007$ K and $J_2/k = 0.125 \pm 0.01$ K, which are in very close agreement with the powder results. However, accurate line shapes at momentum transfers near the zone boundary cannot be obtained with powders and the present measurements show that these line shapes have a very interesting behavior as the temperature is increased through the Curie temperature.

The neutron-scattering measurements were made with use of the standard triple-axis technique with pyrolytic graphite used as monochromator, filter, and analyzer. The experiment was done with a constant incoming energy of 3.572 THz (14.7 mV). The crystal was a 1 $cm^2 \times 1$ mm slab of EuO isotopically enriched in ^{153}Eu to reduce neutron absorption. Despite the isotope, the sample was highly absorbing and most measurements were arranged so that almost symmetric reflection took place from the slab. In this geometry reasonably intense spin waves could be obtained. The sample temperature was measured with a Ge resistor attached directly to the sample holder. The Curie temperature of the sample was determined by measuring the critical scattering and is 69.8 ± 0.1 K.

The neutron scattering from a magnetic system can be written in terms of the relaxation function $R(K, \omega)$ and is given by[5]

$$\frac{d^2\sigma}{d\Omega dE} = \gamma F^2(K)\frac{k^1}{k_0}\sum_{\alpha\beta}(\delta_{\alpha\beta} - \vec{K}_\alpha \cdot \vec{K}_\beta)\omega\frac{\exp(\hbar\omega/kT)}{\exp(\hbar\omega/kT)-1}R^{\alpha\beta}(K, \omega), \qquad (1)$$

where α and β are the Cartesian coordinate directions, K is the scattering vector, $F(K)$ is the magnetic form factor, k^1 and k_0 are the magnitude of the outgoing and incoming neutron wave vectors, and γ is a constant.

Generally neutron-scattering results are given in terms of the generalized susceptibility $\chi(K, \omega)$, where $\chi(K, \omega)$ is simply related to the relaxation function by

$$R^{\alpha\beta}(K, \omega) + R^{\beta\alpha}(K, \omega) = (c/\omega)\,\mathrm{Im}[\chi^{\alpha\beta}(K, \omega) + \chi^{\beta\alpha}(K, \omega)], \qquad (2)$$

where c is a constant. However, when working at low energies it is more convenient to use $R(K, \omega)$ since the cross section behaves smoothly near $\omega = 0$ for this quantity while for a constant $\chi(K, \omega)$ the cross section must diverge at $\omega = 0$. Nevertheless, $R(K, \omega)$ looks similar to $\chi(K, \omega)$ for peaks separated from $\omega = 0$, and narrow spin waves will produce similar peaks in both quantities. The cross section is usually divided into longitudinal and transverse parts where we expect the longitudinal part to be small at low temperatures and the longitudinal and transverse parts to be equal above T_C. Since we are examining the behavior of the cross section near T_C and we cannot separate the longitudinal and transverse parts, the best we can do is to combine them so that $R(K, \omega)$ can be derived from the cross section by

$$\frac{d^2\sigma}{dE\,d\Omega} = \gamma^1(K)\omega\frac{\exp(\hbar\omega/kT)}{\exp(\hbar\omega/kT)-1}R(K, \omega), \qquad (3)$$

where $F^2(K)$ has been included in $\gamma^1(K)$. Sometimes data above T_C are analyzed in terms of the spectra weight function $F(K, \omega)$, which at constant K is simply related to $R(K, \omega)$ by a scale factor.

There is a technical difficulty in obtaining scans of $R(K, \omega)$, in that as ω approaches zero the background from incoherent scattering in the sample and scattering from the cryostat peaks up and must be subtracted from the magnetic scattering. A great deal of effort was spent in measuring the temperature and q dependence of this background. It was found that this background subtraction was easiest if high resolution was used so that the incoherent scattering was confined to a small region near $\omega = 0$. Fortunately, this scattering is almost temperature independent since it is of nuclear origin only, and the background subtraction can be made accurately as long as the temperature is not too high.

There is a scarcity of information on what $R(K, \omega)$ should look like as a function of temperature even for the near-neighbor Heisenberg case. There are a number of approaches available for the hydrodynamic regime; however, for general values of the momentum transfer and temperature above T_C the best information available is that given by Hubbard.[6] Hubbard gives a number of line shapes for various temperatures for the case of the simple-cubic Heisenberg ferromagnet with nearest-neighbor interactions. I have performed neutron-scattering scans at some of the same q values and temperatures shown by Hubbard so that the results can be compared.

Calculations are shown for q values of $(\frac{1}{8}, \frac{1}{8}, \frac{1}{8})$, $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})$, and $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, which correspond to $\frac{1}{4}$, $\frac{1}{2}$, and 1 times the zone boundary for the [111] direction. For $\frac{1}{4}$ of the way to the zone boundary the line shapes above T_C are nearly a Lorentzian centered around $\omega = 0$. Line-shape measurements have been made for this value of q by Passell, Dietrich, and Als-Nielsen, and the single-crystal results are in good agreement with them. For q halfway to the zone boundary the shape definitely becomes non-Lorentzian. This is sufficiently far into the zone that line shapes obtained by powders become less reliable and Passell, Dietrich, and Als-Nielsen did not show line-shape data above T_C for this q value.

Figure 1 shows the line shapes of the magnetic scattering $R(K, \omega)$ for the single-crystal measurements for q halfway to the (111) zone boundary. The data have been corrected for the change in volume of the resolution function ellipsoid with energy and for the analyzer and detector efficiency. The spectrometer resolution was sufficiently sharp that the data are affected by it very little. For $T = T_C - 10$ K, a well-defined but broad-spin wave is observed centered around 0.43 THz. As the temperature is increased to T_C the well-defined spin wave disappears but $R(K, \omega)$ still peaks at a nonzero energy. As T is increased $R(K, \omega)$ peaks only at the origin and appears to broaden some at the highest temperature. A number of

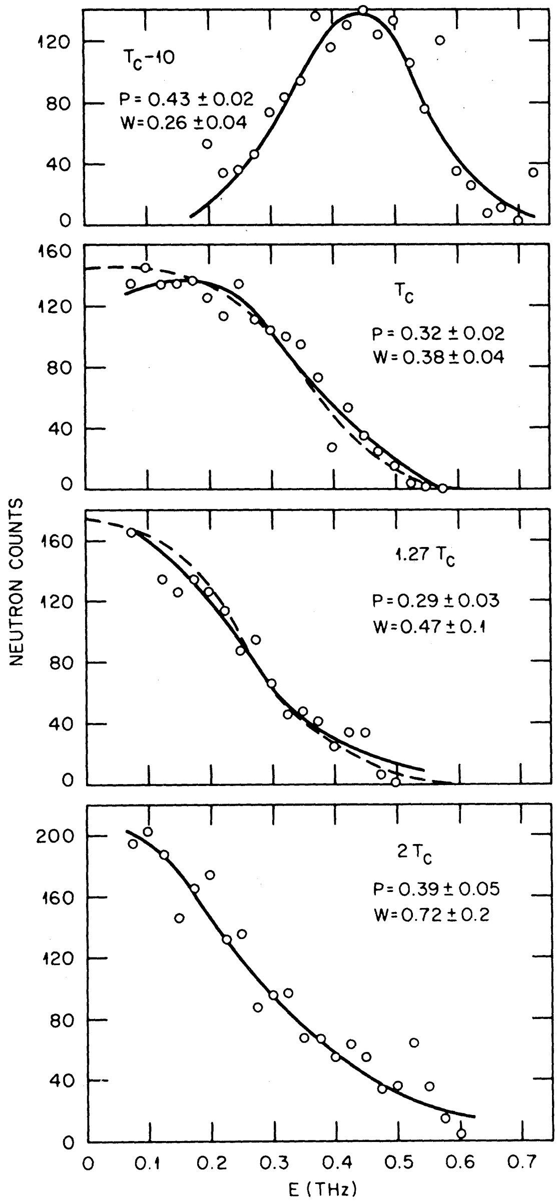

FIG. 1. The magnetic scattering $R(K, \omega)$ measured half the way to the zone boundary for the [111] direction for temperatures below and above T_C. The solid lines show least-squares fits to the data of a Gaussian below T_C and a damped harmonic oscillator above T_C. The peak position parameter p and width parameter w are given for each of the least-squares fits. The broken line shows the results calculated by Hubbard for a near-neighbor ferromagnet.

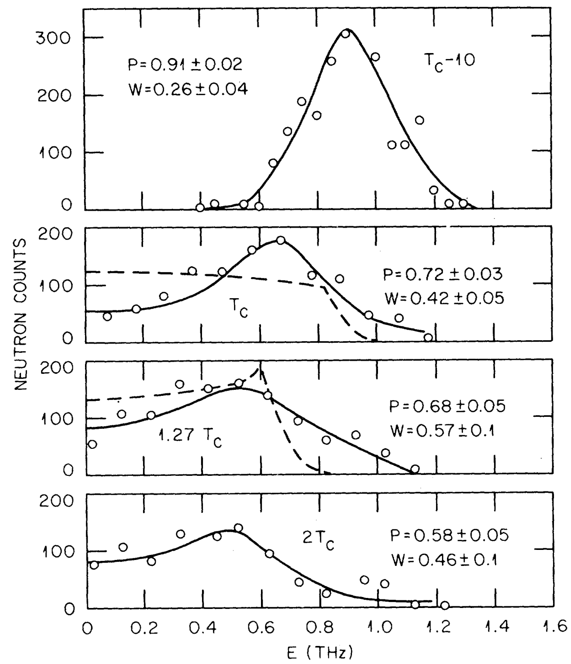

FIG. 2. The magnetic scattering $R(K, \omega)$ measured at the zone boundary. The solid and broken lines, and the fitting parameters, have the same meaning as in Fig. 1.

analytical peak shapes were least-squares fit to the data with varying degrees of success. A Lorentzian centered at $\omega = 0$ gave an unsatisfactory fit at all temperatures. Other peak shapes that were tried are Lorentzians centered at arbitrary position symmetric to $\omega = 0$, Gaussians symmetric to $\omega = 0$, and damped-harmonic-oscillator functions of the form $F(\omega) = \gamma^2\omega_0^2[(\omega^2 - \omega_0^2)^2 + \gamma^2\omega^2]^{-1}$. Of these the Gaussian gives the best fit below T_C, and the damped harmonic oscillator was best above T_C. The solid lines in Fig. 1 are given by these functions, and the peak position and width parameters are shown for each plot. The width parameter is the full width at half maximum for the Gaussian below T_C and γ of the damped-harmonic-oscillator function above T_C. The dashed line shown on the graph for T_C and $1.27T_C$ is taken directly from Hubbard's calculations normalized so that the curves have equal areas. Hubbard gave results for $2.65T_C$ so that the $2T_C$ measurement cannot be compared with the calculated results. It was felt that the background subtraction is not sufficiently accurate much above $2T_C$, and so this was the highest temperature used. The exchange constant used to determine the energy scale for Hubbard's curves was taken to be the sum of J_1 and J_2 since only near-neighbor exchange is considered in the calculations. As can be seen in Fig. 1, the agreement is remarkably good.

Figure 2 shows similar data for the (111) zone boundary. Analysis of the data was done in the same manner as that shown in Fig. 1. For the zone boundary, wide but rather well-defined peaks are found at all temperatures. The peaks broaden and shift downward between $T = T_C - 10$ K and $T = T_C$ but change relatively little as the temperature is raised further. It appears at first glance that Hubbard's calculations do not fit my data well for the zone-boundary position, and indeed they tend to overestimate the scattering at small ω to a considerable degree. However, the peak in the scattering for the $1.27\,T_C$ data agrees well with the measurements and the overall energy scale is approximately correct.

It is surprising that the excitations above T_C at the zone boundary are such well-defined peaks. It appears that the spin correlations are more important in EuO than is found in the computed results. Of course, Hubbard's calculation may not be appropriate for EuO since second-nearest-neighbor interactions are nonnegligible. Still, it appears that EuO is one of the best materials for comparison between theory and experiment. It seems then that both the transition metals and EuO have wide but distinct excitations above T_C. The difference seems to be that above T_C the spin short-range order or correlation length is considerably longer in the transition metals since distinct excitations are found at much smaller momentum transfers for Ni and Fe than for EuO.

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[1]There are numerous references on this subject. Some of the more current ones are the following: R. E. Prange, J. Appl. Phys. 50, 7445 (1979); R. E. Prange and V. Koreman, Phys. Rev. B 19, 4691, 4698 (1979); V. Koreman, J. L. Murray, and R. E. Prange, Phys. Rev. B 16, 4032, 4048, 4058 (1977); J. Hubbard, Phys. Rev. B 19, 2626 (1979), and 20, 4584 (1979); M. V. You, V. Heine, A. J. Holden, and P. J. Lin-Chung, Phys. Rev. Lett. 44, 1282, 1479(E) (1980); T. Moriya and Y. Takahashi, J. Phys. Soc. Jpn. 45, 397 (1978); B. Sriram Shastry, D. M. Edwards, and A. P. Young, to be published; J. W. Lynn and H. A. Mook, Phys. Rev. B 23, 198 (1981).

[2]H. A. Mook, J. W. Lynn, and R. M. Nicklow, Phys. Rev. Lett. 30, 556 (1973).

[3]J. W. Lynn, Phys. Rev. B 11, 2624 (1975).

[4]L. Passell, O. W. Dietrich, and J. Als-Nielson, Phys. Rev. B 14, 4897, 4908, 4923 (1976).

[5]W. Marshall and S. W. Lovesey, *Theory of Thermal Neutron Scattering* (Clarendon, Oxford, 1971).

[6]J. Hubbard, J. Phys. C 4, 53 (1971).

Nonlinear Conductivity and Noise due to Charge-Density-Wave Depinning in $NbSe_3$

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A simple model for the depinning and dynamics of a sliding charge-density wave is presented. The model consists of viscously damped motion in a periodic potential and accounts for the main features of the nonlinear conductivity, noise, and ac-dc coupling in the charge-density–wave state of $NbSe_3$ at low temperature. Comparison with experimental data suggests that the depinning process is associated with the displacement of the charge-density wave by approximately one period λ.

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Niobium triselenide, $NbSe_3$, undergoes two Peierls transitions,[1] one at $T_1 = 142$ K and one at $T_2 = 59$ K, to charge-density–wave (CDW) states. The conductivity is strongly non-Ohmic when the electric field exceeds a critical value E_c,[2,3] and also depends strongly on the frequency.[4,5] The CDW is not destroyed by the application of an electric field,[6] showing that the depinning and subsequent motion of the CDW is responsible for the nonlinear conductivity. One of the puzzling features of the nonlinear conduction is the appearance of both broadband and narrow-band noise.[3,7] These have been suggested to arise from turbulent motion of the CDW,[8] from a moving soliton lattice,[9] or from a drifting CDW,[10] but no appropriate theory is available which relates the field- and frequency-dependent response to the noise pattern observed experimentally.