where $\gamma=0$ or 1 if $\lambda_1-\lambda_2-\lambda_3$ is positive or negative, respectively, and $S(w)$ is given by

$$S(w)=\tfrac{1}{2}\Omega[-w^2+(-1)^{\gamma}2w_1w-w_4^2 +4(2\lambda_2\lambda_3+\lambda_1\lambda_2+\lambda_3\lambda_1)]. \quad (12)$$

In the third range, $w_2\leq w\leq w_3$, we have a quadrilateral for which we have

$$\xi_1(w)S(w)=(\Omega a_1\lambda_3/3\lambda_1) \times(-3w^2+6\lambda_2w+3\lambda_1^2-3\lambda_2^2-\lambda_3^2),$$
$$\xi_2(w)S(w)=(\Omega a_2\lambda_3/3\lambda_2) \times(-3w^2+6\lambda_1w-3\lambda_1^2+3\lambda_2^2-\lambda_3^2),$$
$$\xi_3(w)S(w)=\tfrac{2}{3}\Omega a_3\lambda_3^2, \quad (13)$$

and

$$S(w)=2\Omega\lambda_3(-w+\lambda_1+\lambda_2). \quad (14)$$

In the last range, $w_2\leq w\leq w_4$, there is a triangle for which

$$\xi_1(w)=(a_1/3\lambda_1)[w+(-1)^{\gamma}w_1+\lambda_1],$$
$$\xi_2(w)=(a_2/3\lambda_2)(w-w_2+\lambda_2),$$
$$\xi_3(w)=(a_3/3\lambda_3)(w-w_3+\lambda_3), \quad (15)$$

and $S(w)$ is given by

$$S(w)=\tfrac{1}{2}\Omega(w-w_4)^2. \quad (16)$$

We now have all the expressions required for obtaining $I(\omega)$ from Eq. (5). In this treatment the transition probability represented by $F_j(\vec{q},\omega)$ is calculated to the same degree of accuracy as $g(\omega)$, the density of states. The importance of this lies in the fact that the computed spectrum $I(\omega)$ reflects faithfully and sensitively the features of a theoretical model which can be compared in detail with the measured spectrum. This method has a wide range of applicability to various fields in solid-state physics where one is interested in computing one excitation transition. In its present form it is not applicable to problems where two or more transitions are involved (e.g., second-order Raman effect), and some modifications are required to adapt it for such cases. It is hoped to apply this method to some practical calculations in the near future.

We wish to express our gratitude to Dr. E. Cohen for many stimulating discussions and to Dr. L. J. Raubenheimer for his interest and useful comments.

[1]G. Gilat and G. Dolling, Phys. Letters 8, 304 (1964).
[2]G. Gilat and L. J. Raubenheimer, Phys. Rev. 144, 390 (1966), hereafter to be referred to as I.
[3]L. J. Raubenheimer and G. Gilat, Phys. Rev. 157, 586 (1967), hereafter to be referred to as II.
[4]Z. Kam and G. Gilat, Phys. Rev. 175, 1156 (1968), hereafter to be referred to as III.
[5]C. W. Higginbotham, F. H. Pollak, and M. Cardona, Solid State Commun. 8, 513 (1967).
[6]L. R. Saravia and D. Brust, Phys. Rev. 170, 683 (1968), and 171, 916 (1968).
[7]F. Herman, R. L. Kortum, C. D. Kuglin, and J. P. Van Dyke, Methods in Computational Physics (Academic Press, Inc., New York, 1968), Vol. 8, p. 193.
[8]Although in II triangular prisms are used also, it is possible by some enlargement of the IBZ of the hexagonal lattice to fill it with only rectangular prisms, as in III.
[9]In some cases resonance behavior may exist which shows up as a singularity in $F_j(\vec{q},\omega)$ for some special values of $\vec{q}$. These cases deserve special treatment.

SOUND PROPAGATION NEAR THE MAGNETIC PHASE TRANSITION IN EuO†

B. Lüthi and R. J. Pollina
Department of Physics, Rutgers, The State University, New Brunswick, New Jersey 08903
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In EuO we observe no critical attenuation of sound near T_c, but a corresponding critical change in sound velocity is noticeable. These experiments can be explained by the experimentally verified fact that the sound wave couples to the energy of the spin system.

If the volume magnetostrictive energy is proportional to the exchange energy, then the ultrasonic attenuation α is

$$\alpha\propto q^2\,\mathrm{Re}\int_0^{\infty}\langle E_qE_{-q}(t)\rangle e^{i\omega t}dt$$
$$=q^2\langle E_qE_{-q}\rangle\frac{\Lambda}{v^2}=q^2\frac{\lambda}{v^2}, \quad (1)$$

where we used the hydrodynamic form for the Fourier transform of the exchange energy $E_q(t)=E_qe^{-\Lambda q^2t}$ and for the energy diffusion constant $\Lambda=\lambda/C$ (C is the magnetic part of the specific heat and v the sound velocity), and where we used $\omega\gg\Lambda q^2$ (our q values range between 10^2 and 10^4 cm^{-1}). Scaling-law arguments[1] and factorization

approximation[2] both show the spin thermal conductivity λ to be nondivergent at T_C. Hence α would not show any singular behavior either.[3] This is in contrast to the usual case where one observes a pronounced critical attenuation of sound.[4]

In this Letter we show that EuO represents precisely such a case where the above conjecture (namely that the volume magnetostrictive energy is proportional to the exchange energy and that therefore the sound wave couples to the energy fluctuations of the spin system) holds to good approximation. This we do by giving three pieces of experimental evidence supporting this idea:

(1) Absence of critical attenuation of sound. In Fig. 1 we show ultrasonic-attenuation data for longitudinal waves of 50 and 170 Mc/sec as a function of temperature. A pulse-echo technique using a calibrated attenuator for measuring echo-height differences was used. For both frequencies one does not observe any indication of critical attenuation as observed in many other substances.[4] What one observes is a rather sharp increase in attenuation below T_C. The same behavior was reported earlier[4,5] and also in (100) oriented crystals. The volume magnetostrictive coupling constant B_v is estimated from the pressure dependence of T_C and from the anomalous thermal expansion[6]: $B_v \sim 50\times10^6$ erg/cm^3. (The linear magnetoelastic coupling constants are an order of magnitude smaller,[7] $b_1/S^2 = 2.5\times10^6$ erg/cm^3.) Comparing this coupling constant with the ones in MnF_2[8] and Ho[9] we expect a critical attenuation in EuO of about 2 dB/cm at 50 Mc/sec and about 15 dB/cm at 170 Mc/sec. We would get similar figures by comparison with other substances. The experimental data of Fig. 1 show that we could easily see an effect of >0.5 dB/cm. Hence the absence of any such critical scattering is a strong evidence for the above conjecture.

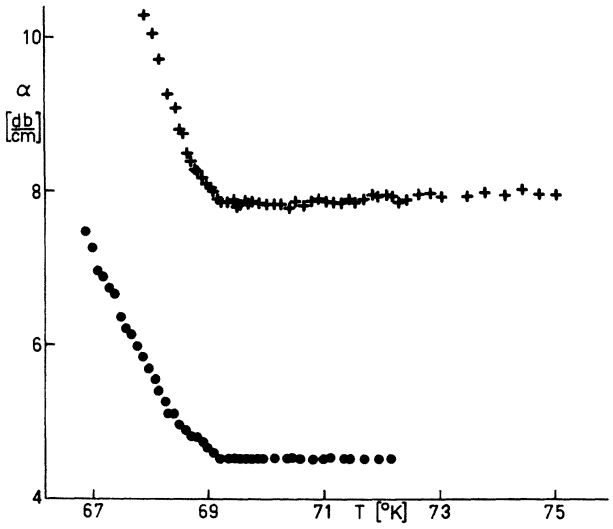

FIG. 1. Attenuation of longitudinal sound waves in EuO propagating along a (110) axis: closed circles, 50 Mc/sec; plusses, 170 Mc/sec.

(2) Proportionality of $H_{\text{sp-ph}}$ and H_{ex}, the spin-phonon-interaction and exchange Hamiltonians, respectively. It has been shown experimentally[6] that in EuO there is a strict proportionality between the magnetic part of the specific heat C and the magnetic part of the differential thermal expansion β. Upon taking into account nearest-neighbor (z_1) and next nearest-neighbor (z_2) interactions this means that

$$C = N\left(z_1 J_1 \frac{\partial}{\partial T}\langle \vec{S}_0\vec{S}_1\rangle + z_2 J_2 \frac{\partial}{\partial T}\langle \vec{S}_0\vec{S}_2\rangle\right)$$

and

$$\beta = -\frac{\kappa V}{3} N\left(z_1 \frac{\partial J_1}{\partial V}\frac{\partial}{\partial T}\langle \vec{S}_0\vec{S}_1\rangle + z_2 \frac{\partial J_2}{\partial V}\frac{\partial}{\partial T}\langle \vec{S}_0\vec{S}_2\rangle\right) \quad (2)$$

are strictly proportional to each other as a function of temperature. (J_i = exchange constant, κ = compressibility.) This implies that one of the following three cases occurs: (a) J_1/J_2 is equal to $(\partial J_1/\partial V)(\partial J_2/\partial V)^{-1}$, (b) $\partial\langle \vec{S}_0\vec{S}_1\rangle/\partial T$ is proportional to $\partial\langle \vec{S}_0\vec{S}_2\rangle/\partial T$, or (c) the second term is negligible compared with the first term. Our ultrasonic-attenuation data suggest that case (c) is the likely one to occur in EuO, since we can write the spin-phonon interaction Hamiltonian in a form similar to Eq. (2) [Eq. (1) of Moran and Lüthi][10]:

$$H_{\text{sp-ph}} = A(z_1' B_1 \vec{S}_0\vec{S}_1 e^{iqR_1} + z_2' B_2 \vec{S}_0\vec{S}_2 e^{iqR_2}),$$

where $B_1 = Na\partial J/\partial a$ and where now z_1', z_2' are in general different from z_1, z_2. For the case of propagation along a (110) direction $z_1' = 2$ and $z_2' = 2$, whereas $z_1 = 12$ and $z_2 = 6$. The proportionality of C and β implies the proportionality of $H_{\text{sp-ph}}$ and H_{ex} in Eq. (2) for $z_2 = 0$. Thus, this experimental evidence gives direct support for the proportionality of $H_{\text{sp-ph}}$ and H_{ex} for EuO.

(3) Occurrence of a critical change in sound velocity. In a manner analogous to Eq. (1), the imaginary part gives an expression for the relative change in sound velocity[8]:

$$\frac{\Delta v}{v} \propto q^2 \frac{v}{\omega} \operatorname{Im} \int_0^\infty \langle E_q \vec{E}_q(t)\rangle e^{i\omega t} dt = \frac{C}{v}. \quad (3)$$

Equation (3) predicts that the relative change in

sound velocity due to critical scattering is proportional to C and hence singular. Furthermore it is frequency independent. A straightforward calculation taking the full expression for the spin-phonon interaction [Eq. (2) of Ref. 10] gives the following simple formulas for attenuation α and velocity change $\Delta v/v$:

$$\alpha = q^2\left(\frac{z'}{z}\right)^2\left(\frac{a}{J}\frac{\partial J}{\partial a}\right)^2\frac{T\lambda}{v^3M},$$

$$\frac{\Delta v}{v} = -\left(\frac{z'}{z}\right)^2\left(\frac{a}{J}\frac{\partial J}{\partial a}\right)^2\frac{TC}{v^2M}, \qquad (4)$$

where M is the molecular weight and T the temperature. With the constants mentioned above we estimate from Eq. (4) a change due to critical scattering of about 10×10^{-4} in $\Delta v/v$.

Figure 2 shows $\Delta v/v$ as a function of T for 50 and 30 Mc/sec sound waves. Depending on the strength of the ultrasonic echo pattern we are able to measure relative velocity changes of 10^{-5} to 10^{-7} using a phase-comparison method.[11] In Fig. 2 one notices for longitudinal waves a linear region for $T-T_C>8°K$, a somewhat flat region for $T-T_C<8°K$, and an abrupt change at $T=T_C$. This abrupt change occurs at the same temperature as the sudden increase in attenuation in Fig. 1. This temperature is taken as the Curie temperature $T_C=69.2°K$. It agrees fairly well with the less accurately determined T_C from magnetization measurements. Extrapolating the linear region as an estimate for the nonmagnetic background as indicated in Fig. 2 and correcting for the thermal expansion,[6] we get the net velocity change due to critical scattering, as shown in Fig. 3 for $T>T_C$. The total observed change in $\Delta v/v$ is 4×10^{-4} in fair agreement with the estimate given above. The data in Figs. 2 and 3 show no noticeable frequency dependence in agreement with Eqs. (3) and (4). Although there is a clear-cut decrease in $\Delta v/v$ as one approaches T_C (Fig. 3), the scattering of our data together with the approximate corrections for background and thermal expansion prevent us from making a sensible quantitative test of Eq. (3) or (4), e.g., the proportionality to C. Shear waves only show the thermal-expansion anomaly (Fig. 2) and no critical attenuation,[5] as expected.

In summary these three pieces of experimental evidence (absence of critical attenuation, proportionality of differential thermal expansion and specific heat, and evidence for critical scattering in the relative change in sound velocity) suggest strongly that EuO is a case where the sound wave couples to the energy of the spin system. In all other magnetic substances which exhibit critical attenuation of sound the proportionality between the spin-phonon Hamiltonian $H_{\text{sp-ph}}$ and the exchange Hamiltonian seems no longer to hold, mainly because more than the nearest-neighbor interaction is of importance. In this case one has to calculate a general four-spin correlation function which gives rise to the pronounced effects in critical attenuation and velocity change.[4]

We acknowledge gratefully useful discussions

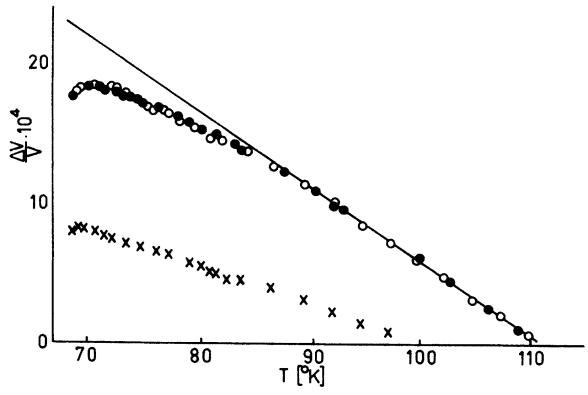

FIG. 2. Relative change in sound velocity in EuO: closed circles, 50 Mc/sec; open circles, 30 Mc/sec (longitudinal waves); crosses, 50 Mc/sec (shear waves).

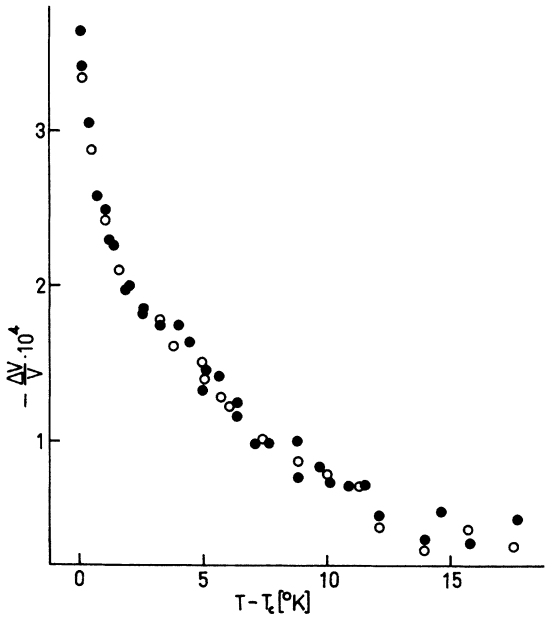

FIG. 3. Net critical change in sound velocity in EuO, from Fig. 2 after correcting for background and thermal expansion. Closed circles, 50 Mc/sec; open circles, 30 Mc/sec.

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[1]K. Kawasaki, Progr. Theoret. Phys. 40, 706 (1968).

[2]D. L. Huber, Solid State Commun. 6, 685 (1968).

[3]This has been noted by others as well: D. L. Huber, L. P. Kadanoff, private communications. A more detailed discussion of this argument can be found in K. Kawasaki, Progr. Theoret. Phys. 29, 801 (1963), and D. L. Huber, Phys. Letters 25A, 93 (1967).

[4]See, for example, B. Lüthi, P. Papon, and R. J. Pollina, in Fourteenth Annual Conference on Magnetism and Magnetic Materials, New York City, 18-21 November 1968 (to be published).

[5]Y. Shapira and T. B. Reed, in Fourteenth Annual Conference on Magnetism and Magnetic Materials, New York City, 18-21 November 1968 (to be published).

[6]B. E. Argyle, N. Miyata, and T. D. Schultz, Phys. Rev. 160, 413 (1967).

[7]B. E. Argyle and N. Miyata, Phys. Rev. 171, 555 (1968).

[8]K. Tani and H. Mori, Progr. Theoret. Phys. 39, 876 (1968).

[9]R. J. Pollina and B. Lüthi, Phys. Rev. 177, 841 (1969).

[10]B. Lüthi and R. J. Pollina, Phys. Rev. 167, 488 (1968).

[11]T. J. Moran and B. Lüthi, to be published.

NEUTRON SCATTERING FROM K_2NiF_4: A TWO-DIMENSIONAL HEISENBERG ANTIFERROMAGNET*

R. J. Birgeneau
Bell Telephone Laboratories, Murray Hill, New Jersey 07974,
and Brookhaven National Laboratory, Upton, New York 11973

and

H. J. Guggenheim
Bell Telephone Laboratories, Murray Hill, New Jersey 07974

and

G. Shirane
Brookhaven National Laboratory, Upton, New York 11973

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The quasielastic magnetic scattering from K_2NiF_4 over the temperature range from 97.2°K to at least 200°K is found to correspond to reciprocal lattice rods rather than points thus giving the first concrete evidence of the two-dimensional character of K_2NiF_4. At 97.1°K the crystal undergoes an extremely sharp phase transition to long-range order in three dimensions. From 97.0°K to 5°K the sublattice magnetization follows a $(97.1-T)^{\beta}$ law with $\beta = 0.15$.

Recently considerable theoretical attention has been directed towards two-dimensional magnetic systems.[1,2] Such systems are expected to exhibit cooperative properties, particularly in the neighborhood of the critical point, which differ appreciably from their three-dimensional counterparts.[3,4] It is therefore of considerable interest to find in nature systems which are truly two dimensional in their behavior. The most promising candidates at the present time seem to be the family of planar compounds with the K_2NiF_4 structure.[5] Early neutron scattering experiments on powdered K_2NiF_4 by Plumier[6] led him to postulate that long-range order (LRO) set in within the perovskite NiF_2 planes at 180°K but that even at 4.2°K there was no true LRO between the planes.

In this Letter we report results of quasielastic magnetic scattering from a large single crystal of K_2NiF_4. It is shown that at high temperatures Plumier's model is approximately correct although the actual details are rather more complicated. From at least 200°K down to 97.2°K the magnetic scattering indicates that there are very long-range correlations within the antiferromagnetic perovskite NiF_2 planes with no measurable correlations between the planes. At $T_N = 97.1$°K the crystal undergoes an extremely sharp phase transition to LRO in three dimensions. Below T_N the three-dimensional Bragg peaks are accompanied by "critical" scattering which is completely two dimensional in form. In