Critical Acoustic Relaxation in EuO

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Acoustic damping and dispersion in EuO have been measured at low kilohertz frequencies near the ferromagnetic critical point. The experimental observations are discussed in terms of thermoelastic and critical dissipative processes. Bounds are established for the critical relaxation rate for sound, $2\times 10^5<\tau_c^{-1}<2\times 10^7$ sec^{-1}, in order-of-magnitude agreement with the EuO spin-lattice relaxation rate calculated by Huber.

Near a magnetic phase transition, the coupling of lattice vibrations and critical fluctuations leads to a damping and velocity shift of phonons. EuO, although a particularly simple ferromagnet possessing reasonably large spin-phonon coupling, has been considered anomalous in that it has not shown any obvious critical attenuation or velocity shifts near T_c for sound waves of frequency $\omega/2\pi \gtrsim 30$ MHz.[1,2] On the basis of low-frequency ($\omega/2\pi \lesssim 3$ kHz) measurements of sound velocity and damping given here, we present a description of sound relaxation processes in EuO which is consistent with both high- and low-frequency experiments near T_c. It is estimated that the relevant critical relaxation rate τ_c^{-1} for sound, presumably due to the decay of spin energy density fluctuations, has the bounds $2\times 10^5<\tau_c^{-1}<2\times 10^7$ sec^{-1}, in order-of-magnitude agreement with the calculation of Huber[3] of the EuO spin-lattice relaxation rate.

We have measured the damping Γ and resonant frequency ω_0 of thin bars (reeds) of single-crystal EuO vibrating in flexural modes between 0.4 and 2.8 kHz. For the vibrating reed, there exists a dissipative process in addition to those critical processes which become measurable only near a phase transition. This relaxation mechanism arises from the interaction of bounded flexural modes with transverse thermal modes, i.e., thermoelastic relaxation, a process which has not been studied previously near a magnetic critical point. In order to understand this mechanism we have utilized data from precise thermodynamic and transport measurements near T_c to calculate the damping and dispersion expected from thermoelastic relaxation. We find that the thermoelastic process exhausts the observed low-frequency damping. After removing the influence of the thermoelastic process from the data, we interpret the results in terms of *critical* absorption and dispersion of sound. The $\omega\approx 0$ adiabatic velocity shift near T_c yields the relaxation strength for the interaction of sound with critical fluctuations. An upper limit for critical attenuation at our frequencies is estimated, thus allowing the establishment of limits for the critical relaxation rate τ_c^{-1}.

The theory of the damping of thin reeds under transverse vibration was first given by Zener.[4] The bending of the reed causes dilations of opposite signs to exist on the upper and lower halves. Thus, in the presence of finite thermal expansion, a transverse temperature gradient is produced. If the temperature across the reed can equalize in a time τ_R much less than a flexural period, $\tau_R \ll \omega^{-1}$, the vibration is isothermal; on the other hand, for $\tau_R \gg \omega^{-1}$, adiabatic conditions prevail. Zener solved the coupled mechanical and heat-flow equations for this system and obtained for the decay rate or damping coefficient

$$\Gamma(\omega, T) = A(T)\mathcal{R}(\omega, \tau_R) \tag{1}$$

and, for the Young's modulus dispersion,

$$[E(\omega) - E_s]/E_s = A(T)\tau_R(T)\mathcal{R}(\omega, \tau_R). \tag{2}$$

Here, $A(T)$ is a frequency-independent amplitude $A(T) = \alpha^2 T E_T/C_p$, with α and C_p the isobaric linear thermal expansivity and specific heat and E_T and E_s the isothermal and adiabatic Young's moduli, respectively. The relaxation function and relaxation time are given explicity by $\mathcal{R}(\omega, \tau_R) = \omega^2\tau_R/[1+(\omega\tau_R)^2]$ and $\tau_R(T) = (d/\pi)^2 D^{-1}(T)$. Here, d is the transverse dimension of the reed and D ($\equiv\lambda/C_p$) is the thermal diffusivity, with λ the total thermal conductivity. Thus, near a phase

transition, the thermoelastic damping and dispersion should be enhanced since α and C_p increase as T approaches T_c.

The acoustic resonances were excited electrostatically and detected capacitively with a phase-sensitive receiver. The samples were EuO single crystals of dimensions 0.4×1.0 cm^2, with the major dimension along $\langle 110\rangle$. Two thicknesses (d) of approximately 0.0065 and 0.016 cm were used to confirm that τ_R varied as d^2. The d = 0.0065-cm (thin) sample was resonated at its fundamental frequency near 430 Hz and first overtone 2.8 kHz, whereas the 0.016-cm (thick) sample was operated primarily near its fundamental of 1.6 kHz. At each temperature the complete resonance was swept out and fitted with a nonlinear least-squares procedure by a Lorentzian line shape to determine Γ and ω_0. The thermal expansivity was measured and analyzed by techniques identical to those described previously.[5] The thermal diffusivity was measured by an ac technique reported recently.[6] Specific-heat data were obtained from measurements of Kornblit, Ahlers, and Buehler.[7] All measurements were made on samples cut from adjacent regions of a single EuO boule of high resistivity ($\rho \approx 10^7$ Ω cm and 300 K).

An example of the damping measured for the thick reed near 1.6 kHz is shown in Fig. 1 for the temperature range $T_c \pm 5$ K ($T_c \approx 69.3$ K). The decay rate calculated from Eq. (1) using the experimentally measured thermodynamic and transport properties is also shown. We emphasize that no adjustable parameters, scale factors, or background terms have been employed in this comparison of the absolute decay rates. The agreement must be considered remarkable inasmuch as absolute values as well as the temperature variations of α, C_p, and D have been used. For this example, the vibration is more adiabatic than isothermal with $\omega\tau_R = 1.0$ at 76 K and increasing to $\omega\tau_R = 1.8$ at T_c. Since $\mathcal{R}(\omega, \tau_R)$ is slowly varying near $\omega\tau_R = 1$, the increase in Γ is dominated by the amplitude factor $A(T)$, which varies near T_c as α (since α and C_p are roughly proportional here). For the thin sample (430 Hz), $\omega\tau_R$ was 0.04 at 76 K.

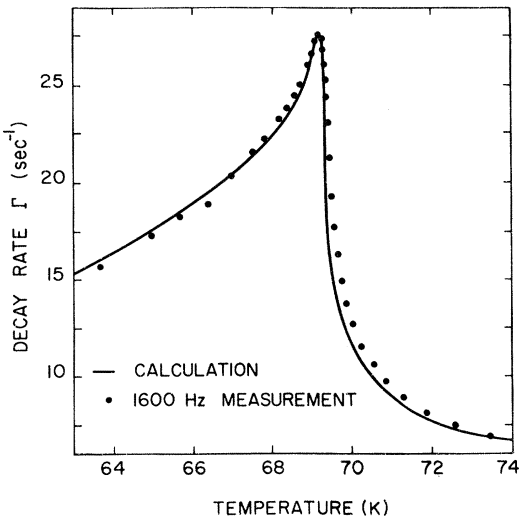

FIG. 1. Decay rate Γ of 1600-Hz flexural vibrations in EuO near T_c. The calculation is based on the theory of thermoelastic relaxation.

The fractional velocity shift relative to the measured velocity at 75 K is shown in Fig. 2 for the thick sample near 1.6 kHz, where $v = (E/\rho)^{1/2}$ (ρ is the mass density). We have used Eq. (2) and the data to compute the adiabatic (and isothermal) velocity shifts, as shown in Fig. 2. According to thermodynamics, the low-frequency adiabatic velocity should reflect the change in C_p^{-1}, as is apparent from Fig. 2 above T_c. Below T_c, other magnetic interactions not associated with critical behavior cause a reduction in the

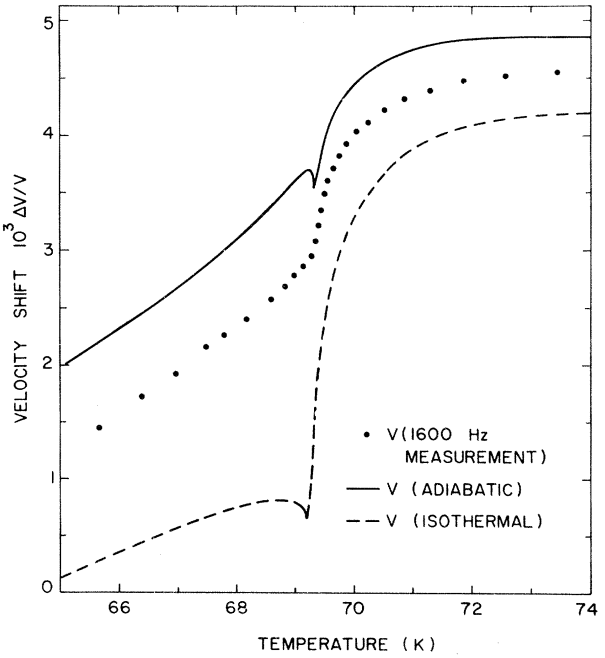

FIG. 2. Fractional shift of the $\langle 110\rangle$ Young's modulus velocity relative to its value at 75 K for 1600-Hz flexural vibrations in EuO near T_c. The solid and dashed lines are the adiabatic and isothermal velocity shifts calculated from thermoelastic relaxation theory and the experimental data. (Arbitrary zero for velocity scale.

sound velocity which are not of interest here. When the effects of dispersion are removed from the data a small minimum in $\Delta v/v$ is readily apparent in both the adiabatic and the isothermal cases.

We now consider processes which relax the critical fluctuations near a magnetic phase transition, as opposed to the thermoelastic relaxation just discussed. In general, sound can couple to either fluctuations of the order parameter or the spin energy density.[8] In the latter case, the relevant relaxation rate for sound[9] is $\tau_c^{-1} = \tau_{sl}^{-1} + \tau_D^{-1}$, where $\tau_{sl}^{-1} = \gamma/C_s$ is the spin-lattice relaxation rate (γ is a temperature-independent parameter) and $\tau_D^{-1} = \lambda_s \omega^2/C_s v^2$ is the spin-energy diffusion rate, with λ_s and C_s the spin thermal conductivity and spin specific heat, respectively. A calculation by Huber[3] of spin-lattice relaxation rates for magnetic insulators was in reasonable agreement ($\tau_{sl}^{-1} \sim 10^9$–10^{10} sec^{-1}) with relaxation rates derived from ultrasonic experiments in $RbMnF_3$[10,11] and MnF_2,[12] where large attenuation is observed. It thus appears that $\tau_{sl}^{-1} \gg \tau_D^{-1}$ in these insulators and dominates τ_c^{-1}. For EuO, however, Huber proposed a rather small τ_{sl}^{-1} ($\sim 10^5$ sec^{-1}) and suggested that the reason for the absence of noticeable critical attenuation in previous experiments in EuO was that $\omega\tau_{sl} \gg 1$. We now present an interpretation of our data which is consistent with this conjecture.

A phenomenological description of the damping and velocity shifts due to a single relaxation process near a continuous phase transition is given by[9,12]

$$\Gamma_c = [(v_\infty^2 - v_0^2)/v_0^2]\mathcal{R}(\omega, \tau_c), \tag{3}$$

$$v^2(\omega) - v_0^2 = (v_\infty^2 - v_0^2)\tau_c \mathcal{R}(\omega, \tau_c), \tag{4}$$

where now v_0 is the thermodynamic adiabatic sound velocity and v_∞ is the adiabatic velocity to be identified with the sound velocity of the unperturbed lattice or background. If $\omega\tau_c \ll 1$, Eq. (3) yields

$$\tau_c^{-1} \approx 2\omega^2(v_\infty - v_0)/v_0\Gamma_c. \tag{5}$$

The magnitude of Γ_c at 1.6 kHz due to this critical process in unknown, but from the good agreement between the calculated thermoelastic damping and experiment, as seen in Fig. 1, it can be estimated that $\Gamma_c < 1$ sec^{-1}. Using the adiabatic sound velocity (Fig. 2) as v_0 and making an extrapolation of the background velocity to T_c to estimate v_∞, we obtain a relaxation strength $(v_\infty - v_0)/v_0 \approx 1 \times 10^{-3}$. From Eq. (5) we arrive at the lower bound, $\tau_c^{-1} > 2 \times 10^5$ sec^{-1}.[13] An upper limit to τ_c^{-1} may be obtained by the following argument. Previous velocity measurements at $\omega/2\pi = 30$ and 50 MHz[2] indicated that the dispersion between these frequencies was no greater than $\Delta v/v \approx 5 \times 10^{-6}$.[14] It is easily found from Eq. (4) that this upper limit to dispersion imposes the restriction $\omega\tau_c > 20$ at 50 MHz, or $\tau_c^{-1} < 2 \times 10^7$ sec^{-1}. This upper limit for τ_c^{-1} would predict a maximum critical attenuation near T_c of ≈ 0.3 dB/cm, although for $\omega\tau_c \gg 1$ the change in attenuation due to the temperature dependence of τ_c would be much smaller than this value. This expected attenuation change is below the sensitivity of ~ 0.5 dB/cm stated in Ref. 2.

We have thus shown that the critical relaxation rate in EuO has the bounds $2 \times 10^5 < \tau_c^{-1} < 2 \times 10^7$ sec^{-1}. Huber[3] estimated that $\tau_{sl}^{-1} \sim 10^5$ sec^{-1} and $\tau_D^{-1} \lesssim 10^3$ sec^{-1} for $\omega/2\pi \lesssim 50$ MHz. Thus, we associate our experimentally inferred relaxation rate with τ_{sl}^{-1} and see that τ_D^{-1} plays an insignificant role in critical attenuation of sound in EuO.[15] The "anomalous" behavior of EuO *vis-à-vis* $RbMnF_3$ and MnF_2 can be seen, in retrospect, to arise from the fact that conventional plane-wave ultrasonic experiments on small crystals are generally carried out at frequencies above a few megahertz. Since $\tau_{sl} \sim C_s \sim \ln|T - T_c|$, τ_c is effectively a constant. In contrast, the relaxation times in magnetic metals[16] are strongly temperature dependent near T_c and a wide range of $\omega\tau_c$ can be explored simply by changing $|T - T_c|$. In EuO, it is necessary that $\omega \approx \tau_c^{-1}$ for significant absorption to occur, and the present analysis suggests that further experiments in the frequency range 10^4–10^6 Hz would be valuable in obtaining a more precise value for τ_c and its temperature dependence.

Finally, we suggest that the technique of thermoelastic relaxation, previously unexploited near phase transitions, offers a useful means by which critical properties of solids may be measured. As has been shown here and elsewhere,[17] Zener's description of thermoelastic damping [Eqs. (1) and (2)] appears correct to at least a precision of a few percent, making it one of very few processes whose damping and dispersion magnitudes can be calculated to this accuracy. As an example, for cases in which sufficient thermodynamic data exist, it should be possible to use thermoelastic relaxation to measure a solid's thermal diffusivity.

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Note added.—We have directly observed critical attenuation in EuO above T_c at a frequency of 235 kHz. The critical contribution to Γ is approximately 500 $\sec^{-1}$ at T_c. Using the measured critical attenuation and dispersion at $\epsilon = 10^{-3}$ and Eqs. (3) and (4), we estimate $\tau_c^{-1} \approx 1.5 \times 10^6$ $\sec^{-1}$, within a factor of 3. This result for τ_c^{-1} is consistent with high- and low-frequency acoustic measurements in EuO and reduces further the uncertainty in the magnitude of the critical relaxation rate.

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[8]It has been pointed out [K. Kawasaki and A. Ikushima, Phys. Rev. B 1, 3143 (1970)] that in EuO there should actually be coupling of sound partially to energy fluctuations and partially to order-parameter fluctuations. No conclusive evidence for coupling to order-parameter fluctuations in magnetic insulators has been observed and the apparent absence of this interaction remains an unresolved problem in understanding critical attenuation of sound.

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[13]Although the present measurements obtain the ⟨110⟩ Young's modulus velocity, and in Ref. 2 the ⟨110⟩ longitudinal plane-wave velocity was measured, the thermodynamic velocity changes associated with either mode near T_c differ by no more than a factor of 2, which is not significant for our estimate of τ_c^{-1}.

[14]This dispersion limit should not be confused with the $\Delta v/v$ quoted by Lüthi and Pollina (Ref. 2). In their data analysis, Fig. 3 of Ref. 2, they apparently take $\Delta v/v \equiv [v(\omega) - v_\infty]/v$, whereas in their Eq. (4) [which follows from our Eq. (4)], $\Delta v/v \equiv [v(\omega) - v_0]/v$.

[15]In an earlier interpretation of the high-frequency measurements (Ref. 2), τ_D^{-1} was assumed to dominate the relaxation rate τ_c^{-1}.

[16]See, for example, B. Golding and M. Barmatz, Phys. Rev. Lett. 23, 223 (1969).

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Plasmon Observation Using X Rays*

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We have investigated the problem of plasmon observation using high-energy photons within the context of the random phase approximation. It is shown that the experimental results from a study of Be by Miliotis can be readily understood from this point of view. It is further shown that these results include both collective and single-particle effects and that the collective effects persist well beyond the "critical wave vector." We discuss the concept of a critical wave vector.

Observations of plasmons in experiments which can, in analogy with electron energy loss, be described as photon energy loss have now been reported by a number of workers.[1,2] Perhaps the most detailed study to date has been that of Miliotis[2] on Be. He observed that the height of the plasmon peak increased sharply with increasing q ($\hbar q$ is here the momentum transfer) at low q, reached a maximum, and then decreased slowly at higher q values, while the width of the plasmon line was roughly proportional to q^2. In addition, the plasmon dispersion curve, while satisfying the well-known relation $\omega - \omega_p \propto q^2$ for small q, bent over for larger values of q and showed little dependence of ω on q. This region of little dispersion occurred for q values well beyond q_c, the critical value of q, leading Miliotis to conclude that "plasmons" appear where they are unexpected. We will show that all of the results of the experiment of Miliotis can be un-