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Neutron scattering measurements of phonons in iron above and below T_c

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We report detailed measurements of transverse phonons in Fe propagating along the [110] direction with [1$\bar{1}$0] polarization, both above and below T_c. The low-q measurements give the elastic constant C', which decreases anomalously as T_c is approached from below, in agreement with the ultrasonic measurement. Phonon-frequency measurements throughout the Brillouin zone show a kink in the vicinity of T_c, thus indicating their magnetoelastic origin.

I. INTRODUCTION

The elastic constants of many magnetic materials display anomalies near the magnetic-ordering temperature suggesting that this anomalous behavior is magnetic in origin. The anomalous behavior is especially large in Invar alloys whose elastic constants in fact decrease with decreasing temperature.[1] In Fe and Ni the elastic-constant anomalies are quite small near T_c; however, they have been measured by ultrasonic velocity measurements.[2,3] In Ni, for example, Alers *et al.* found that the elastic constants were higher in the ferromagnetic region by about 1–3% compared with the values extrapolated linearly from the paramagnetic region.

In the case of Fe, the magnitude of elastic-constant anomaly is quite dependent on what elastic constant is being measured. For example, there is a marked decrease of C' $[=(C_{11}-C_{12})/2]$ as the temperature is increased. In fact, at $T=897\,°\mathrm{C}$, this elastic constant decreases to a value of 0.13×10^{12} dyn/cm^2, which is only 28% of its value at room temperature. Whereas, in the same temperature range, the elastic constants C_{11} and C_{44} decrease to 65 and 85% of their room-temperature value, respectively. All of the elastic-constant curves show a kink in the region of the Curie temperature, which points to the magnetic origin of the peculiarities found.

Neutron scattering experiments on Fe and Fe(Si) crystals[4] show that not only is there an anomalous decrease in the C' elastic constant, but that the entire branch of $[110]T_2$ phonons shows anomalous decrease with temperature. Neutron scattering data of De Vallera[4] also indicate the existence of a kink in the phonon frequencies near T_c for practically all the phonon branches measured. The data, however, were taken at only four temperatures and include little information on the low-q region.

In this paper we report on neutron scattering experiments of phonon measurements in the Fe both above and below T_c. Our focus has been on a $[110]T_2$ transverse branch, since for this branch the decrease in phonon frequencies with temperature is the largest. We have also measured the phonon frequencies very close to T_c, to establish the existence of a kink near T_c over the entire branch of $[110]T_2$ phonons.

II. EXPERIMENT

The experiments were performed on a cylindrical single crystal of Fe, 4 cm long, and 1 cm in diameter. The crystal was oriented with the [100] axis vertical. The data were collected in the vicinity of a [110] reciprocal-lattice point and so there is a large body of data for the $[110]T_2$ mode, the low-q part of which corresponds to the elastic constant C'. Limited amounts of data were also taken with another crystal oriented in the (hhl) zone. The crystal was heated in a vacuum furnace which consisted of hollow copper spherical caps. Temperature was measured by a Chromel-Alumel thermocouple at the bottom of the crystal. The furnace temperatures were stable to 1.0 K.

Pyrolytic graphite crystals served as a monochromator and energy analyzer with a fixed final energy of 14.7 meV. A pyrolytic graphite filter was also in front of the counter. For most of the data reported, the collimation used was 20′-20′-40′-40′; however, for a limited number of scans near T_c, we used collimation of 10′-20′-20′-20′ to check the effect of magnetic ordering on the phonon linewidths. The temperature of the sample was callibrated by utilizing the known value of the Curie temperature ($T_c=768\,°\mathrm{C}$). The intensity of critical diffuse magnetic scattering was observed at $q=0.04$ at a series of temperatures above and below T_c. The Curie temperature could be determined to within one degree. Neutron scattering profiles at low q were corrected for resolution effects by fitting the data to a Lorentzian line shape convoluted with the instrumental resolution function.

III. RESULTS

In Fig. 1 we show the comparison of elastic constant C' measured with neutrons in the present work and the ultrasonic values of Dever.[2] Over the whole temperature range, the neutron values are within 8% of the values deter-

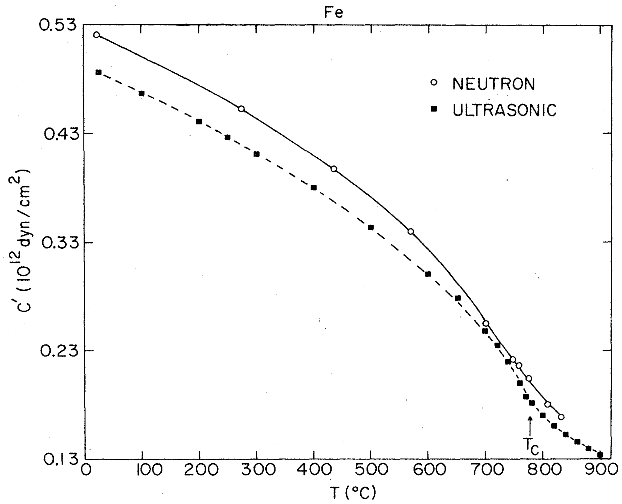

FIG. 1. The elastic constant C' $[=\frac{1}{2}(C_{11}-C_{12})]$ for Fe as a function of temperature. The ultrasonic values are from Dever, Ref. 2.

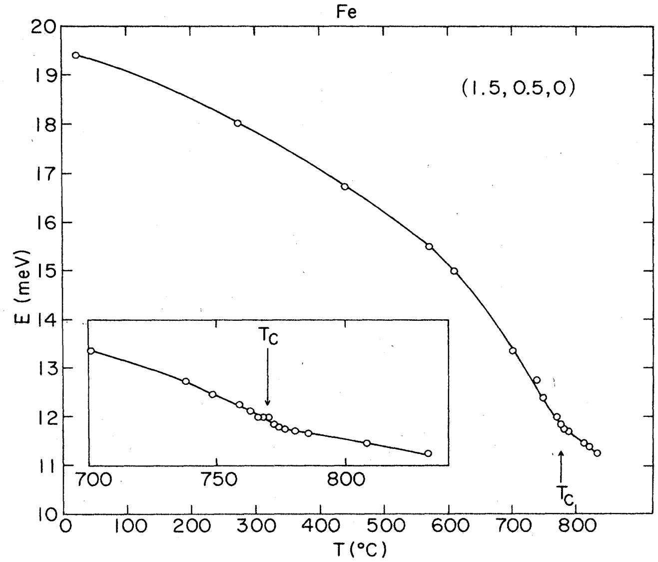

FIG. 2. Frequency of the [110]T_2 mode at the zone boundary [measured at reciprocal point (1.5,0.5,0)] as a function of temperature. The inset shows detailed measurements of the Z.B. frequency in the vicinity of T_c.

mined by ultrasonics. However, as seen from the figure, the neutron values are consistently higher (5–8%) than the ultrasonic values at all temperatures. This could be due to either systematic errors in neutron or ultrasonic results, or due to the fact that with neutrons the sampling frequency ω is larger than the inverse thermal phonon lifetime τ^{-1} and one measures the zero sound, whereas in ultrasonic measurements the sound wave propagation is in thermal equilibrium states and one measures the first sound. Neutron data are consistent with a large decrease in this elastic constant found from the ultrasonic measurements. A limited

TABLE I. Elastic constants of iron measured by neutron scattering at various temperatures.

	Elastic constants	
Temperature (°C)	C' (neutron) (10^{12} dyn/cm²)	C' (ultrasonic)[a] (10^{12} dyn/cm²)
22	0.521	0.483
274	0.452	0.418
435	0.397	0.367
569	0.338	0.318
700	0.255	0.298
748	0.223	0.210
759	0.216	0.199
766	0.210	0.193
776	0.204	0.185
808	0.180	0.167
832	0.168	0.158
	C_{44} (neutron)	C_{44} (ultrasonic)[a]
610	2.647	2.591
820	2.470	2.386
	$(C_{11}+C_{44}-C')$ (neutron)	$(C_{11}+C_{44}-C')$ (ultrasonic)
610	1.054	1.051
820	0.985	1.002

[a]Reference 1.

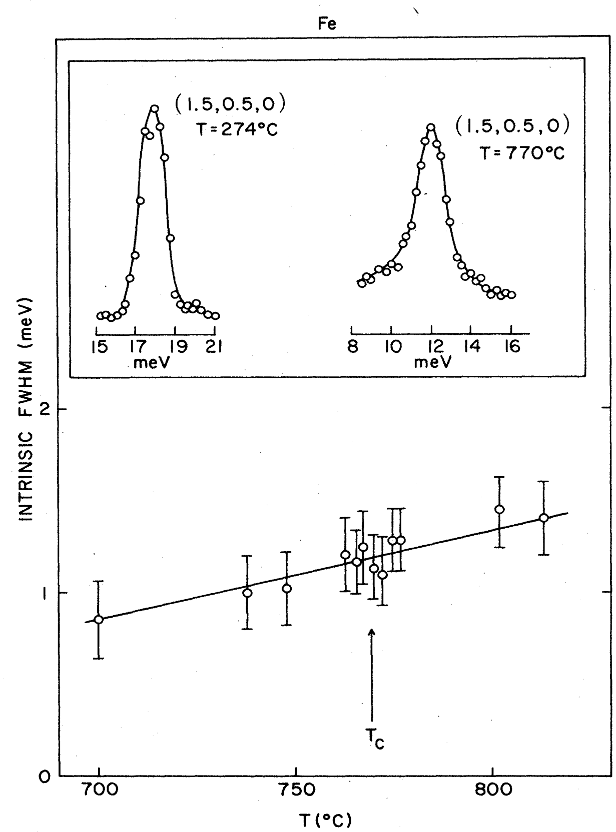

FIG. 3. Intrinsic full width at half maximum for zone-boundary [110]T_2 phonon as a function of temperature. The inset shows two of these phonon groups at $T=274$ and 770 °C (near T_c). For these phonons a final-energy configuration, $E_f=14.7$ meV, with collimation of 20′-20′-40′-40′, was used.

number of other elastic-constant measurements are within 2–3% of the ultrasonic values. These values are given in Table I.

We have measured the temperature dependence of the whole [110]T_2 transverse phonon branch up to the zone boundary. The frequency of this mode at the zone-boundary (Z.B.) position (1.5,0.5,0) is shown in Fig. 2, as a function of temperature. It is clear that this branch shows an anomalous decrease in frequency all the way up to the zone boundary. At a temperature of 828 °C the frequency of this Z.B. mode is 11.25 meV, which is only 58.1% of its value at room temperature. These results are in agreement with the earlier neutron scattering experiments. The inset in Fig. 2 shows detailed measurements of the Z.B. phonon in the vicinity of T_c. The kink in the phonon frequency within 2–3 °C of the T_c is quite evident in the data.

In order to determine whether the change in the slope of the phonon frequency near T_c is a dynamic effect, we measured the linewidth of the [110]T_2 zone-boundary phonons as a function of temperature. From the measured linewidth, an intrinsic linewidth was extracted by assuming a pure Lorentzian cross section for phonons and a Gaussian resolution function. These intrinsic linewidths are plotted in Fig. 3. Within the uncertainty of the experimental resolution we did not detect any anomaly in the phonon linewidths near T_c. Only a gradual increase in the linewidth of the phonons was observed, which is consistent with the increase in anharmonicity as a function of temperature increase. At room temperature, the phonon linewidths are close to being resolution limited.

IV. DISCUSSION

The anomalous behavior of the shear modes along the [110] direction in Fe is observed to extend over a wide wave-vector range, from $q=0$ in the ultrasonic measurements to zone boundary. The origin of such an effect has been considered by Hausch[1] and Wohlfarth.[5] Hausch has considered the effect of magnetoelastic coupling, where the anomalous behavior of the phonons is a direct consequence of the change of magnetic energy due to shear deformation.

The free energy of the magnetic system can be expanded in terms of the order parameter $\eta = I/I_0$:

$$U_m = \tfrac{1}{2}A\eta^2 + \tfrac{1}{4}\gamma\eta^4 + \cdots \quad , \tag{1}$$

where the coefficient A is considered to be strain dependent by Hausch.[1] Neglecting the strain dependence of the quartic term, one can write

$$U_m = (U_m)_0 + \frac{1}{2}\left(\frac{\partial A}{\partial E}\right)_0 E\eta^2 + \frac{1}{4}\left(\frac{\partial^2 A}{\partial E^2}\right)_0 E^2\eta^2 \quad , \tag{2}$$

where E is the appropriate strain. The third term in Eq. (2) is the one responsible for the anomalous behavior of the elastic constants observed. The second term gives rise to a forced volume magnetostriction effect, which is supposed to be much smaller. In Hausch's theory, coefficient A is related to the combination of exchange parameters for the first and second near-neighbor exchange J_1 and J_2. The magnetic contribution to the elastic constant can then be written in terms of distance derivatives J_1' and J_2'. These are, however, treated as parameters in the theory, and are to be derived from the experimental data.

No satisfactory microscopic theories exist in the literature to explain the phonon anomalies in itinerant magnets, where the exchange interaction may not be limited to only two near neighbors.

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[1] G. Hausch, Phys. Status Solidi (a) **15**, 501 (1973).

[2] D. J. Dever, J. Appl. Phys. **43**, 3293 (1972); J. L. Routbort, C. N. Reid, E. S. Fisher, and D. J. Dever, Acta Metall. **19**, 1307 (1971).

[3] G. A. Alers, J. R. Neighbors, and H. Sato, J. Phys. Chem. Solids **13**, 401 (1960).

[4] A. M. B. C. De Vallera, Ph.D. thesis, Cambridge University, 1977 (unpublished).

[5] E. F. Wohfarth, J. Phys. F **6**, L59 (1976).