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Acoustic-Phonon Instability and Critical Scattering in Nb_3Sb†

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Neutron-scattering experiments on Nb_3Sn show the drastic softening of the [110] acoustic shear modes as well as an unusual frequency response (a central peak in addition to the phonon side bands) near the phase transition at 45°K. A model involving coupling of the bare phonon to other fluctuations with a Debye-like frequency spectrum is proposed which correctly describes the observed cross section as a function of temperature.

The structural phase transition in the high-temperature superconductor Nb_3Sn is characterized by a drastic softening[1] of the acoustic shear mode with vector $\vec{q}\parallel[110]$ and polarization vector $\vec{e}\parallel[1\bar{1}0]$. At the transition temperature $T_m = 45$°K, the crystal structure changes from a cubic to a slightly distorted tetragonal structure.[2] In a recent neutron-diffraction study,[3] we have determined the atomic displacements in the tetragonal phase and concluded that only an acoustic instability (not an optic one) is required to explain the transition.[4] This paper reports some unusual dynamical characteristics of these soft shear modes revealed by inelastic neutron-scattering techniques.

Experiments were carried out on a triple-axis spectrometer at the Brookhaven high-flux beam reactor on the same single crystal used in our recent study.[3] The crystal was grown by Hanak and Berman[5] and has a volume of 0.05 cm^3, small for inelastic neutron-scattering experiments. Bent-focusing pyrolytic graphite monochromator crystals were used with incoming neutron energies of 40, 14, and 5 meV.

The temperature dependence of the [110] transverse acoustic (TA) branch is shown in Fig. 1. First we note a relatively large decrease of phonon energies on cooling for high ζ values. Here the wave vector $\vec{q}$ is expressed as $(\zeta, \zeta, 0)2\pi/a$. This decrease, about 15%, persists up to the zone boundary, $\zeta = 0.5$. A similar decrease was also observed for phonons in the [100] transverse branch; for example, the phonon energies at the zone boundaries are 7.5 and 6.5 meV at 295 and 46°K, respectively. Thus, there seems to be a substantial softening over the entire range of wave vectors. This is unexpected and not at all understood at present.

Much more drastic softening was observed for the [110] branch for smaller ζ values. The general characteristics of the temperature depen-

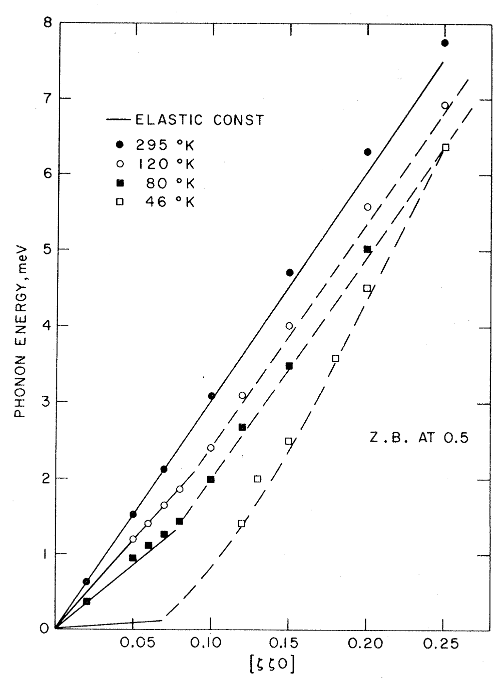

FIG. 1. Temperature dependence of TA modes along $\vec{q} = (\zeta, \zeta, 0)2\pi/a$ with polarization vector $\vec{e}\parallel[1\bar{1}0]$. $2\pi/a = 1.19$ Å^{-1} at 46°K.

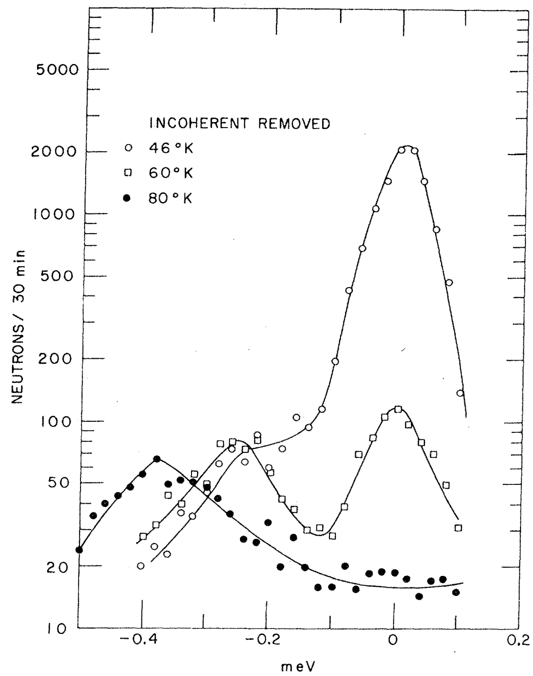

FIG. 2. Cross sections of $(\zeta, \zeta, 0)$ shear mode with $\zeta = 0.02$ measured at $(2-\zeta, 1+\zeta, 0)$. Data were taken with incoming neutron energy of 5 meV and an energy resilution of 0.1 meV. Negative energy corresponds to neutron energy gain.

dence of the [110] branch are similar to those observed recently for V_3Si.[6] In both crystals, the softening is extended to similar q values. Experimentally, the present case of Nb_3Sn is much more definitive; this is because of smaller incoherent scattering and low damping of soft phonons even at temperatures near T_m. Recently, theoretical models were proposed by Dieterich and Schuster[7] and by Cohen[8] for the q dependence of the [110] soft modes in V_3Si. Similar mechanisms may also apply to Nb_3Sn.

The principal concern of this note, however, is the unusual line shape of the soft phonons for small ζ as $T \to T_m$. An example of such data is shown in Fig. 2 for $\zeta = 0.02$. (Note the logarithmic scale.) The observed phonon energy at this ζ is 0.65 meV at 295°K, decreasing to 0.25 meV at 60°K. This decrease follows closely the change in the ultrasonic velocities[1] determined at much smaller q values. Near the phase transition, as shown for the 60°K data in Fig. 2, the response function develops a central component *in addition* to the "phonon" side bands. Upon further cooling, the intensity of this central component continues to grow (see Fig. 3) while the side bands saturate but remain as distinct shoulders on the main cen-

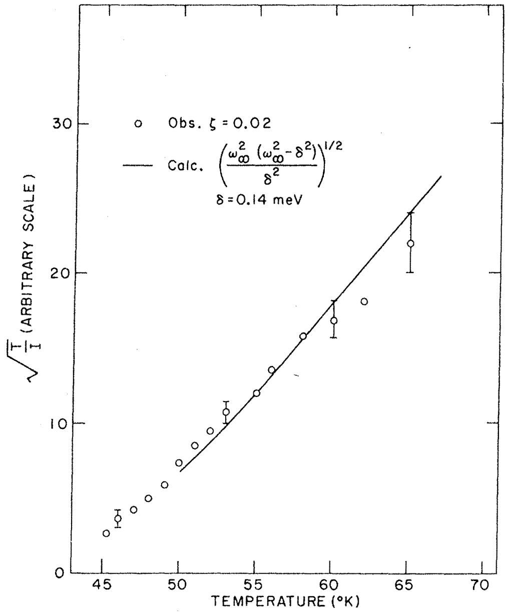

FIG. 3. The integrated intensity I of the central component of the critical scattering versus temperature. The data are plotted in such a way as to indicate the agreement with the prediction of Eq. (6).

tral peak. Additional features of the central peaks are their strong q dependence and narrow energy width (< 0.06 meV).

This behavior is a definite departure from a simple picture of soft-mode condensation with a temperature-independent damping factor. Specific predictions of the existence of a central peak were made by Cowley[9] for piezoelectric crystals. The mechanism involves coupling between a soft optic mode at the zone center and a pair of acoustic modes. Experimentally, the first example of three-peak cross sections was reported very recently by Riste *et al.*[10] in their neutron-scattering study of the 110°K phase transition in $SrTiO_3$. The soft mode in this case is the optic mode at the zone-corner R point and the crystal is centrosymmetric; the characteristics of the central peak in $SrTiO_3$ are not yet fully elucidated. The present case of Nb_3Sn involves only acoustic modes near the zone center, and this allows us to formulate a rather simple physical picture of the central component.

This picture is best understood beginning with the expression for the frequency-dependent part of the one-phonon scattering cross section. Quite generally, this may be written as

$$S(\omega) = (k_B T/\hbar\omega)\,\mathrm{Im}[\omega_0^2 - \omega^2 - i\omega\Gamma]^{-1}, \tag{1}$$

where ω_0 is a temperature-dependent quasiharmonic frequency and we assume $k_B T \gg \hbar\omega$ for the frequency range of interest. Soft-mode line shapes have previously been discussed in the "viscous damping" approximation in which Γ is taken to be a frequency-independent constant. The condition for dynamical instability, $\omega_0 \to 0$, is connected with the divergence of the integrated scattering intensity $I(\text{total}) = \int S(\omega)d\omega \propto \omega_0^{-2}$. This form leads to either a two- or one-peaked function, depending upon the ratio Γ/ω_0, and is not capable of explaining even qualitatively the profiles shown in Fig. 2.

In general, however, Γ itself has a frequency dependence which reflects the changing density of excitations with which the one-phonon state can interact. Sufficiently large changes in $\Gamma(\omega)$ in the important frequency region near ω_0 can produce more complicated spectral profiles. In what follows we demonstrate that a particularly simple form[11] of the complex self-energy function, namely

$$\Gamma(\omega) = \Gamma_0 + \delta^2(\gamma - i\omega)^{-1}, \tag{2}$$

accounts for the unusual features which we observe in Nb_3Sn. Since we are dealing with acoustic phonons, a well-established physical process for this damping may be invoked. At response frequencies much larger than the average inverse lifetime γ_T of the "bath" of thermal phonons, acoustic phonons propagate in a collision-free or zero-sound regime. In the opposite (collision-dominated or first-sound) regime, both the acoustic velocities and the attenuation are renormalized in a characteristic way.[12] Approximate expressions interpolating between these two limits have been discussed,[13] and it is possible to derive an approximate $\Gamma(\omega)$ of the form of Eq. (2) with $\gamma = \gamma_T$ and with δ proportional to third-order anharmonic coupling of the acoustic mode to pairs of thermal phonons.[14]

Inserting Eq. (2) into Eq. (1) gives

$$S(\omega) = \left(\frac{k_B T}{\hbar}\right)\Gamma_0 + \frac{\delta^2\gamma}{\omega^2+\gamma^2}\left[\left(\omega_\infty^2 - \frac{\delta^2\gamma^2}{\omega^2+\gamma^2} - \omega^2\right)^2 + \omega^2\left(\Gamma_0 + \frac{\delta^2\gamma}{\omega^2+\gamma^2}\right)^2\right]^{-1}, \tag{3}$$

where $\omega_\infty^2 = \omega_0^2 + \delta^2$. This formula[11] has the general qualitative features necessary to describe the data in Fig. 2. In the limit $\omega_\infty^2 \gg \delta^2$, it shows three distinct peaks with side bands at $\pm\omega_\infty$. In the other limit, $\omega_0 \to 0$ (i.e., $\omega_\infty \to \delta$), Eq. (3) shows a profile with shoulders, similar to the profile observed at 46°K. Moreover, we may identify ω_0 and ω_∞ with a first- and zero-sound frequency, respectively.

For a more quantitative comparison with experiment, we can conveniently divide the cross section into $S(\omega)_{\text{total}} = S(\omega)_{\text{central}} + S(\omega)_{\text{side band}}$ with

$$S(\omega)_{\text{central}} = \left(\frac{k_B T}{\hbar}\right)\left(\frac{\delta^2}{\omega_0^2\omega_\infty^2}\right)\left(\frac{\gamma'}{\omega^2+\gamma'^2}\right), \tag{4}$$

where $\gamma' = (\omega_0/\omega_\infty)^2\gamma$. This formula is valid for the range of parameters of interest, $\omega_\infty \gg \gamma$ and $\Gamma_0 \ll (\delta^2/\gamma)$. As with the simpler damping, $I_{\text{total}} \propto \omega_0^{-2}$ and dynamical instability occurs as $\omega_0 \to 0$. The fractional integrated central peak intensity is simply

$$\frac{I(\text{central})}{I(\text{total})} = \frac{\delta^2}{\omega_\infty^2}. \tag{5}$$

We can use the result of Eq. (5) together with data such as those given in Fig. 2 to estimate δ. For $\zeta = 0.02$ at 60°K, we find $\delta = 0.14 \pm 0.04$ meV. We are able to estimate ω_∞ directly (essentially the maxima of the phonon side bands) for $T \geq 50$°K. Combining this with the value of δ deduced above allows us to test the dependence of $I(\text{central})$ upon ω_∞, which from Eq. (4) is predicted to be

$$I(\text{central}) \propto \frac{T\delta^2}{\omega_0^2\omega_\infty^2} = \frac{T\delta^2}{\omega_\infty^2(\omega_\infty^2 - \delta^2)}. \tag{6}$$

The result is shown in Fig. 3. The agreement is quite satisfying considering that δ, taken to be constant, may in fact be weakly temperature dependent.

We can make an independent estimate of $\delta = (\omega_\infty^2 - \omega_0^2)^{1/2}$ by using ω_∞ obtained above and ω_0 approximated by $v_0 q$, where v_0 is the measured ultrasonic (therefore first-sound) velocity.[15] At 60°K for $\zeta = 0.02$ ($q = 0.033$ Å^{-1}), this gives an estimate of 0.12 meV, in good agreement with the value of 0.14 meV deduced from the intensity of the central mode. Calculations also produce δ values of this magnitude for this mechanism.[12,14]

More generally, a cross section $S(\omega)$ of the form of Eq. (3) can result from the linear coupling of the bare acoustic phonon to any fluctuations in the solid which themselves have a Debye relaxation spectrum. A familiar example is the "Mountain" mode which results from the coupling of pressure fluctuations with internal degrees of relaxation in a nonsimple fluid.[16,17] Riste *et al.*[10] attribute the central mode in $SrTiO_3$ to higher-order differences in the isothermal and adiabatic phonon response but do not discuss the expected form of $S(\omega)$ quantitatively. A more careful characterization of the temperature and wave-vector dependence of $S(\omega)$ for both Nb_3Sn and $SrTiO_3$ is planned for the near future, in order that various possible theoretical alternatives can be assessed.

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Li-Induced Nucleon-Transfer Reactions between 1p-Shell Nuclei: Isospin and Fractional-Parentage Studies*

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Angular distributions for the one-nucleon transfer reactions $^{14}N(^6Li, ^7Li)^{13}N$ and $^{14}N(^6Li, ^7Be)^{13}C$ were investigated at $E(^6Li) = 32$ MeV and were compared with finite-range distorted wave calculations and theoretical spectroscopic factors. These one-nucleon transfer reactions induced by complex nuclei are described by the calculations just as well as those induced by light projectiles. A detailed study of the relative cross sections of the isobaric-spin analog transitions shows deviations from isospin symmetry that are due to Coulomb effects.

Among the interactions between complex ions, one of the most fundamental is single-nucleon transfer which, when studied with light projectiles, has yielded much of the nuclear-structure information currently available. Beams of heavy ions ($A \geq 10$) have also been used in a few detailed studies of single-nucleon transfer—both at relatively low energies[1] at which descriptions in terms of specialized models are applicable[2] and at higher energies to which those theoretical models have been extended.[3] The relative spectroscopic factors deduced in the reported studies are in fair agreement with results from light-ion-induced transfers. No corresponding studies with