Influence of the Superconducting Energy Gap on Phonon Linewidths in Nb$_3$Sn†

J. D. Axe and G. Shirane
Brookhaven National Laboratory, Upton, New York 11973
(Received 7 December 1972)

Inelastic neutron scattering, used to study acoustic phonon linewidths in Nb$_3$Sn, has revealed abrupt changes in certain phonon lifetimes in the vicinity of the superconducting transformation temperature, $T_c = 18.3$°K. This behavior arises because phonons with energy less than that of the temperature-dependent superconducting gap energy, $2\Delta(T)$, are energetically incapable of decaying by excitation of electron-hole quasiparticle pairs. These measurements give $2\Delta(0) = 4.4k_BT_c$ and reveal a strong anisotropy in the electron-transverse phonon interaction.

This Letter describes inelastic neutron scattering measurements which have detected the influence of the superconducting energy gap upon phonon lifetimes in Nb$_3$Sn. The results yield estimates of the superconducting gap energy and the magnitude and anisotropy of the electron-phonon coupling. In addition, the results establish a new and potentially useful method of utilizing inelastic neutron scattering in the study of low-lying electron excitations in superconductors.

The experiments described were carried out on a triple-axis spectrometer at the Brookhaven high flux beam reactor. The Nb$_3$Sn crystal, grown by Hanak and Berman,[1] was that used in our previous studies.[2,3] Although of excellent quality (mosiac spread $< 0.1°$ in the cubic phase) the sample volume (~ 0.05 cm^3) is quite small for inelastic neutron scattering, even with the use of highly efficient vertically focusing pyrolytic graphite monochromator crystals.[4] Figure 1 shows an example of the changes which occur in a $[\zeta\zeta 0]$ T_1 phonon line shape for small changes of temperature in the vicinity of the superconducting transition temperature T_c. $\{T_1$ designates a transverse acoustic phonon with particle displacements in the $[\zeta\bar{\zeta}0]$ direction.$\}$ The observed low-temperature linewidth corresponds very closely to the instrumental resolution but establishes an upper limit (≤ 0.3 meV full width at half-maximum) on the intrinsic linewidth. At 26°K the observed width of this same phonon is considerably greater than the instrumental width, and the intrinsic linewidth has increased at least fourfold (to ~ 1.4 meV) between 6 and 26°K.

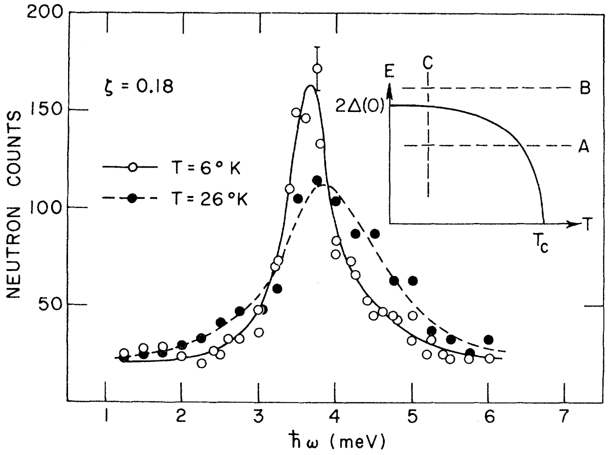

FIG. 1. The linewidth of low-energy $[\zeta\zeta 0]$ T_1 acoustic phonons broadens appreciably with increasing temperature. The inset shows graphically the effect of the energy conversion condition $\hbar\omega_p \geq 2\Delta(T)$ upon phonon damping with increasing temperature (scans A and B) and with phonon energy at a fixed temperature (scan C).

Figure 2 summarizes the temperature dependence of the observed linewidths of various transverse acoustic phonons at low temperatures. (Because of the small sample size it proved impossible to study either longitudinal phonons or higher-energy transverse phonons with sufficient resolution for a meaningful study of linewidths.[5]) The linewidth of the $[\zeta\zeta 0]$ T_1 phonon with $\zeta = 0.18$, which from Fig. 1 has an energy $\hbar\omega_p \approx 4$ meV, is seen to increase abruptly near T_c and remain nearly constant at higher temperatures. The behavior is distinctly different for a higher-energy phonon of the same type $\{[\zeta\zeta 0]$ T_1 with $\zeta = 0.3$, $\hbar\omega_p \approx 8$ meV$\}$ which has a large, measurable linewidth which persists to the lowest temperatures. Separate experiments carried out at 6°K show that the transition between these two types of behavior (i.e., from narrow to wide low temperature linewidths) occurs rather abruptly at a phonon energy ~ 7 meV. Different still is the behavior of transverse phonons propagating along $[\zeta 00]$ which are sharp (resolution-limited linewidth) at all temperatures.

The behavior of the $[\zeta 00]$ phonons is normal, in the sense that in typical materials the line-

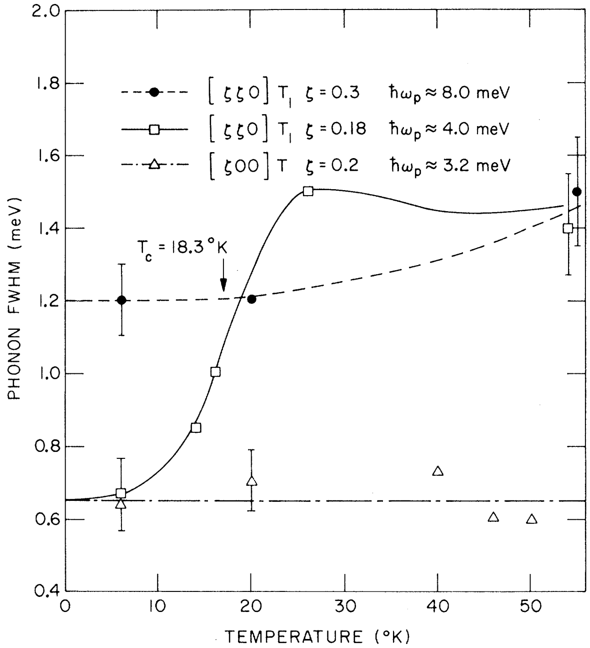

FIG. 2. Summary of transverse acoustic phonon linewidths in Nb_3Sn. The unusually large $[\zeta\zeta 0]$ T_1 phonon linewidth is suppressed if the phonon energy $\hbar\omega_p$ falls below the superconducting gap energy $2\Delta(T)$. ("Phonon FWHM" is the full width at half maximum of the observed phonon peak.)

width of low-energy acoustic phonons is below the resolution of the usual neutron scattering experiment. The first unusual aspect of the data shown in Fig. 2 therefore is the surprisingly large linewidth of the $[\zeta\zeta 0]$ T_1 phonons. Fortunately, the even more striking changes which occur in this linewidth near T_c provide the key to its understanding, for it suggests that we are dealing with that part of the phonon damping involving resonant scattering with the metallic electrons rather than with scattering with other phonons. In the superconducting phase it is useful to distinguish between two mechanisms for phonon damping involving the metallic electrons. The first is due to phonon collisions with thermally excited electron-hole quasiparticles. This process has been extensively studied in a variety of materials by the attenuation of ultrasound.[6] The other process involves phonon-induced excitation of quasiparticles across the superconducting energy gap, 2Δ. This Cooper-pair "breaking" process is energetically impossible in ultrasonic experiments since $\hbar\omega_p << 2\Delta(T)$ except when $T \approx T_c$, where the contribution is negligible. But the process should be very important for higher-frequency phonons and is worthwhile to study because it sets in abruptly at a threshold phonon energy $\hbar\omega_p = 2\Delta(T)$, and thus in principle provides a means for direct determination of the temperature dependence and the anisotropy of the superconducting gap energy. A theoretical analysis of this contribution to the attenuation of longitudinal acoustic phonons within the BCS model has been given by Bobetic.[7]

It is clear that the absorption of a phonon and the associated creation of an excited quasiparticle pair provides the basis for an understanding of the temperature-dependent $[\zeta\zeta 0]$ T_1 phonon linewidths described above. The characteristic phonon energy at which the linewidth suddenly increases at the lowest temperature (scan *C* in Fig. 1) can be identified with the superconducting gap energy, $2\Delta(0) = 7(\pm 1)$ meV. Similarly, the change in the linewidth of the lower-energy $[\zeta\zeta 0]$ T_1 phonon shown in Fig. 2 is predicted when $2\Delta(T) = \hbar\omega_p \approx 4$ meV (scan *A* in Fig. 1). This condition cannot be satisfied for the more energetic T_1 phonon (scan *B* in Fig. 1).

Previous estimates of the gap energy $2\Delta(0)$ have been obtained by tunneling[8-10] $[2\Delta(0) = (1.3\text{–}3.9)k_BT_c]$ and infrared spectroscopy[11] $(3.77k_BT_c)$; indirect estimates are available from specific-heat[12] $(4.8k_BT_c)$ and thermal-conductivity[13] $(3.56 \times k_BT_c)$ measurements. Tunneling and, to a lesser extent, infrared measurements probe the sample surface, which may not be characteristic of the bulk, while the interpretation of the specific-heat data is complicated by uncertainties in the treatment of lattice contribution. The present estimate $2\Delta(0) = (4.4 \pm 0.6)k_BT_c$, while not highly precise, is free from the above defects. However, the present value applies only to excitations with quasiparticle momentum along [110], whereas most of the previous measurements represent a different average over a possibly very anisotropic energy gap.[10]

Since the establishment of the superconducting energy gap selectively nullifies that part of the phonon linewidth, γ_{ep}, due to electron-phonon interaction, the present observations provide a direct measure of the magnitude of electron-phonon coupling, which is of great importance in understanding the favorable superconducting properties of this material. Just how closely γ_{ep} is related to quantities of fundamental interest to strong coupling superconductivity was recently pointed out by Allen,[14] who derived an explicit relation between γ_{ep} and the electron-phonon spectral function $\alpha^2F(\omega)$,

$$\alpha^2F(\omega)=[2/\pi N(0)\omega]\sum_q\gamma_{ep}(q)\,\delta(\omega-\omega(q)), \qquad (1)$$

where $N(0)$ is the electronic density of states at the Fermi surface. Neutron scattering experiments are thus capable, in principle, not only of determining the phonon density of states $F(\omega)$, but $\alpha^2F(\omega)$ as well [at least over the limited range $\omega\leq 2\Delta(0)$] if $\gamma_{ep}(q)$ is sampled over the whole Brillouin zone. Such a program would be prohibitively lengthy for Nb_3Sn given the present sample size; nevertheless the very limited sampling of $\gamma_{ep}(q)$ presented here is of some interest. Allen was also able to derive an expression for the average electron-phonon linewidth contribution $\langle\gamma_{ep}\rangle$ in terms of the dimensionless coupling parameter λ (see McMillan[15]):

$$\langle\gamma_{ep}\rangle=\pi N(0)\langle\omega_p^2\rangle\lambda/12N, \qquad (2)$$

where $\langle\omega_p^2\rangle$ is the mean square phonon frequency and N is the number of atoms in the crystal. Applying this relation to Nb_3Sn one estimates $\langle\gamma_{ep}\rangle=0.6$ meV.[14] Therefore the $[\zeta\zeta0]$ T_1 phonons ($\gamma_{ep}=1.4$ meV) are seen to make a nearly $2\frac{1}{2}$ times greater than average contribution to the phonon-mediated electron-electron interaction responsible for superconductivity, whereas the $[\zeta00]$ T phonons ($\gamma_{ep}\leq 0.3$ meV) contribute less than their share. In this connection note that Klose and Schuster,[16] using a one-dimensional model of the d electrons in materials with the Nb_3Sn structure, predicted a strong enhancement of T_c resulting from the coupling with transverse acoustic phonons. It is perhaps also worthwhile to point out that such simple one-dimensional models of the d electrons describe correctly the anisotropy in the electron-phonon coupling observed in the present experiments. In fact, all such models (so long as they neglect interband transitions) predict maximum coupling to transverse acoustic phonons of the $[\zeta\zeta0]$ T_1 type while the coupling to $[\zeta00]$ T phonons vanishes.

Strong electron-phonon interaction has also been invoked to explain the structural phase transformation in Nb_3Sn at 46°K, which is known to proceed by an elastic instability involving the same $[\zeta\zeta0]$ T_1 phonons. In this model,[17] the elastic softening results from a Kohn-like anomaly in the phonon spectrum which is strongly temperature dependent because of very sharp structure in the electron density of states. While the present observations in no way confirm the most controversial aspects of this model, the strong electron-phonon coupling found here is consistent with many basic features of the model. Further experiments dealing with aspects related to the structural phase transformation have been performed and will be reported separately.

We wish to thank J. J. Hanak and the RCA group for allowing us to study their Nb_3Sn crystal. We gratefully acknowledge much stimulation and encouragement through conversations with P. B. Allen, R. W. Cohen, and H. Schuster.

---

†Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] J. J. Hanak and H. S. Berman, J. Phys. Chem. Solids 28, 249 (1967).

[2] G. Shirane and J. D. Axe, Phys. Rev. B 4, 2957 (1971).

[3] G. Shirane and J. D. Axe, Phys. Rev. Lett. 27, 1803 (1971).

[4] T. Riste and K. Otnes, Nucl. Instrum. Methods 75, 197 (1969); A. C. Nunes and G. Shirane, *ibid.* 95, 445 (1971).

[5] Lower-energy $[\zeta\zeta0]$ T_1 phonons ($\zeta<0.15$) are influenced increasingly by anomalous behavior connected with the structural transformation at 46°K, and are the subject of a separate communication. (See also Refs. 2 and 3.)

[6] For a summary, see L. M. Falicov and D. H. Douglass, in *Progress in Low Temperature Physics,* edited by J. C. Gorter (North-Holland, Amsterdam, 1964), Vol. 4, pp. 97–189.

[7] V. M. Bobetic, Phys. Rev. 136, A1535 (1964).

[8] L. Y. L. Shen, Phys. Rev. Lett. 29, 1082 (1972).

[9] H. J. Levinstein and J. E. Kunzler, Phys. Lett. 20, 581 (1966).

[10] V. Hoffstein and R. W. Cohen, Phys. Lett. 29A, 603 (1969).

[11] D. R. Bosomworth and G. W. Cullen, Phys. Rev. 160, 364 (1967).

[12] L. J. Vieland and A. W. Wicklund, Phys. Rev. 166, 424 (1968).

[13] G. D. Cody and R. W. Cohen, Rev. Mod. Phys. 36, 213 (1964).

[14] P. B. Allen, Phys. Rev. B 6, 2577 (1972).

[15] W. L. McMillan, Phys. Rev. 167, 331 (1968).

[16] W. Klose and H. Schuster, Z. Phys. 241, 348 (1971).

[17] This model originates in the work of J. Labbé and J. Friedel, J. Phys. (Paris) 27, 153 (1966), and has been much elaborated. See, for example, E. Pytte, Phys. Rev. B 4, 1094 (1971).