Phonon softening of Nb_3Sn in [ζζζ]T modes

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Neutron scattering measurements have been extended to the [ζζζ] transverse branch of Nb_3Sn. In contrast to the prediction of Gor'kov, this branch also shows a pronounced phonon softening. We conclude that all phonon branches in Nb_3Sn exhibit softening for a wide range of q towards the martensitic transition at 45 K.

I. INTRODUCTION

The A-15 compounds have attracted considerable attention in recent years[1] because of their high-temperature superconductivity and their structural instabilities. Pronounced phonon softening in Nb_3Sn above its martensitic transformation[2,3] at T_m = 45 K has been investigated by ultrasonic[4-6] and neutron[7] measurements. As seen in Fig. 1(a) and Table I, the sound velocity of the transverse $[\zeta\zeta 0]T_1$ mode, with polarization along $[\zeta\bar{\zeta}0]$, is proportional to $(C_{11}-C_{12})^{1/2}$, and this vanishes at T_m.[2,3] The $[\zeta 00]T$ mode, proportional to $C_{44}^{1/2}$, also shows a modest softening towards T_M.

The most surprising result of the neutron measurement by Axe and Shirane[7] was a large extension in q space of the $[\zeta\zeta 0]T_1$ phonon softening as shown in Fig. 1(b). The initial slope of the phonon dispersion curve closely follows the measured ultrasonic velocities. Then the curve shows a break around $\zeta \sim 0.1$; beyond this q value, a substantial softening persists up to the zone boundary. This extended softening for the entire q range was also observed for the $[\zeta 00]T$ and $[\zeta\zeta 0]T_2$ branches.[7]

Several theoretical models[8] have been proposed to explain this pronounced softening in Nb_3Sn. In particular, Gor'kov[9] proposed an interacting chain model which explains many dynamical characteristics of Nb_3Sn. Gor'kov's model makes an explicit prediction that the extended softening will be absent for the $[\zeta\zeta\zeta]$ direction. In this model, the electrons on each set of Nb chains can contribute to the softening of phonons with a relatively large wave vector q, so long as the projection of the wave vector along the chain does not exceed a critical value $q_{crit} \sim 0.1(2\pi/a)$. In the Gor'kov model, the break in the phonon dispersion at $\zeta \sim 0.1$ seen in Fig. 1(b) is explained as the "switching off" of contributions from the x and y chains as q_x and q_y exceed q_{crit}, leaving only the contributions of the z-axis chains. It follows, as an explicit prediction of the Gor'kov model, that softening should be totally absent for q along $[\zeta\zeta\zeta]$ for $\zeta \lesssim 0.1$. In order to check this prediction, we have carried out additional neutron measurements on Nb_3Sn.

We used a portion of the identical small Nb_3Sn crystal used in our previous neutron scattering experiments.[3,7] This crystal was grown by Hanak and Berman[10] in 1967. It has a narrow mosaic (less than 0.1°) in the cubic phase, and exhibits a well-defined phase transition at 45 K.[2] Because of its small[11] size (~0.035 cm^3), measurements were restricted to selected phonons in $[\zeta\zeta\zeta]T_1$ and $[\zeta 00]L$ branches.

II. PHONON SOFTENING ABOVE T_m

Phonon data were taken on a triple-axis spectrometer at the Brookhaven high flux beam reactor. Outgoing neutron energy E_F is fixed either at 14 or 24 meV with collimation of 40′. The details of the experimental setup were almost identical to those previously described.[7] The crystal was mounted with the $[1\bar{1}0]$ axis vertical. Data taken for $[\zeta\zeta\zeta]T$ modes are summarized in Fig. 2 and Table II, and typical phonon profiles are shown in Fig. 3. The limiting velocity of this branch has not been measured ultrasonically, but it is given

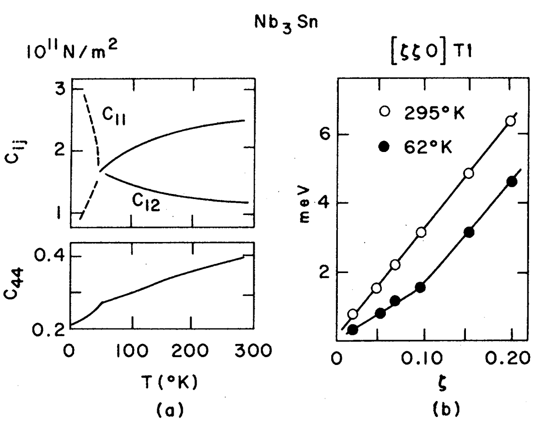

FIG. 1. Summary of previous data on Nb_3Sn. (a) Elastic constants derived from ultrasonic measurements by Rehwald *et al*. (Ref. 6). (b) Extended phonon softening observed by neutron scattering (Ref. 7).

TABLE I. Relation between sound velocity v and elastic constant C_{ij} for a cubic crystal with density ρ.

Branch	ρv^2
$[\zeta\zeta\zeta]T$	$\frac{1}{3}(C_{11}-C_{12}+C_{44})$
$[\zeta\zeta 0]T_1$	$\frac{1}{2}(C_{11}-C_{12})$
$[\zeta\zeta 0]T_2$	C_{44}
$[\zeta\zeta 0]L$	$\frac{1}{2}(C_{11}+C_{12}+2C_{44})$
$[\zeta 00]T$	C_{44}
Bulk modulus	$\frac{1}{3}(C_{11}+2C_{12})$

by

$$\rho^{1/2}v_{[\zeta\zeta\zeta]}=[\tfrac{1}{3}(C_{11}-C_{12}+C_{44})]^{1/2}.$$

These elastic constants are already known by previous ultrasonic studies and are shown in Fig. 1(a). The ultrasonic velocities calculated in this way from data given by Rehwald *et al.*[6] are shown in Figs. 2 and 4 as solid lines. As we already know, $C_{11}-C_{12}$ goes to zero at T_m, while C_{44} shows a smaller temperature dependence [Fig. 1(a)]. Thus $v_{[\zeta\zeta\zeta]T}$ shows a large, but not divergent, anomaly toward T_m.

We see in Figs. 2 and 4 that for $\zeta \leq 0.1$, neutron data agree well with the sound velocities obtained by ultrasonic data. As shown in Fig. 2, for $[\zeta\zeta\zeta]_T$ there is a gentle break in slope around $\zeta = 0.10$. Note that both breaks appear at the same reduced

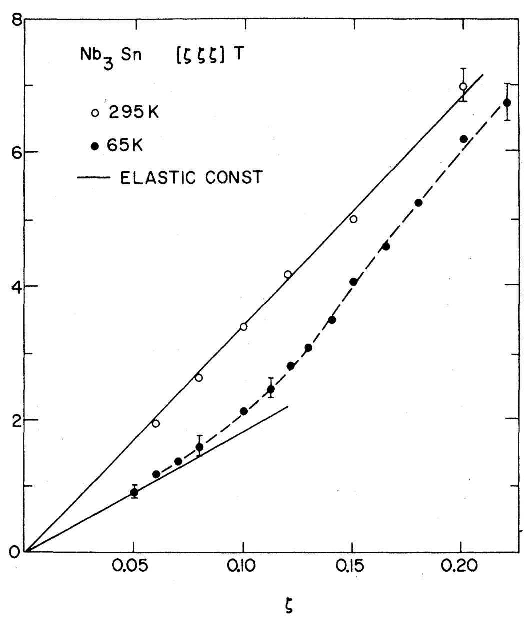

FIG. 2. Phonon-dispersion relation $\omega(q)$, Nb_3Sn as a function of temperature. Solid lines correspond to initial slopes derived from the ultrasonic data (Ref. 6).

TABLE II. Acoustical-phonon energies (meV) for Nb_3Sn. Estimated errors are ± (0.1 – 0.3) meV.

	ζ	295 K	120 K	65 K
$T[\zeta\zeta\zeta]$				
	0.05	···	1.30	0.90
	0.08	2.65	2.2	1.6
	0.10	3.4	···	2.1
	0.12	4.1	3.5	2.8
	0.15	5.0	4.5	4.1
	0.20	7.0	···	6.3
	0.25	8.6	···	7.7
$L[\zeta 00]$				
	0.15	5.7		5.0
	0.20	7.8		7.0

unit $\zeta = 0.10$; note, however, that the absolute value of q is somewhat larger along $[\zeta\zeta\zeta]$ than for $[\zeta\zeta 0]$. At higher ζ values, the softening seems to converge to a general value of 10% decrease as seen in Fig. 2 and Table II. We were unable to measure phonons beyond $\zeta = 0.25$. These features are very similar to those of the $[\zeta\zeta 0]T_1$ branch and in contradiction to the prediction of Gor'kov's model. This suggests a need for further theoretical refinement of the chain-chain interaction scheme.

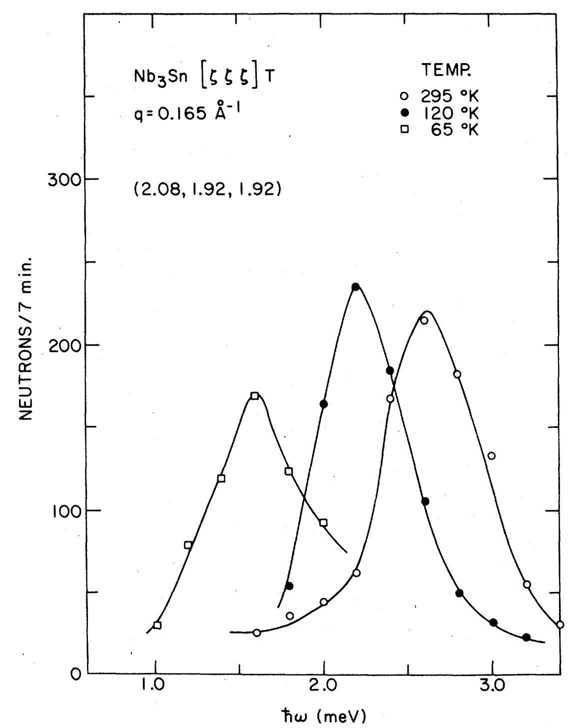

FIG. 3. Typical phonon profiles for $[\zeta\zeta\zeta]$ transverse phonon as a function of temperature. The cubic reciprocal-lattice vector $a^* = 2\pi/a = 1.189$ Å^{-1} at 46 K.

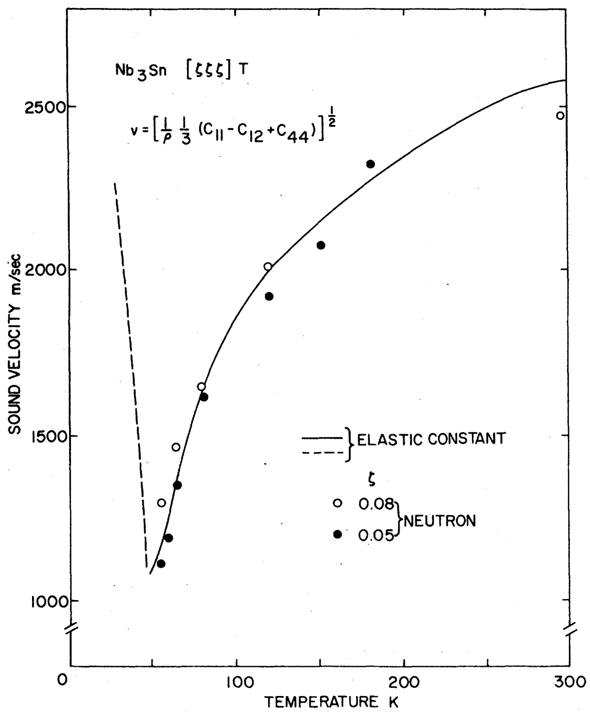

FIG. 4. Comparison of $[\zeta\zeta\zeta]T$ sound velocities derived from the ultrasonic data (solid line, Ref. 6) and the neutron measurements at small ζ values.

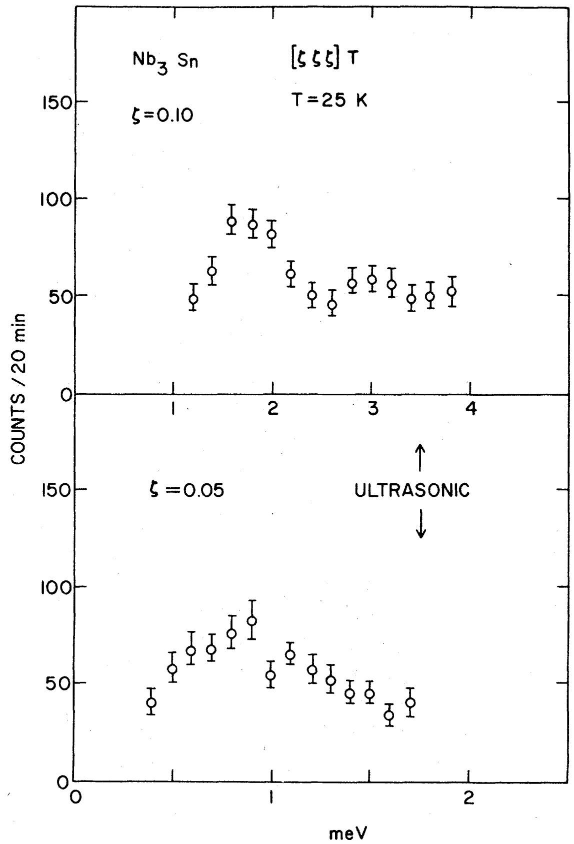

FIG. 5. Phonon profiles of $[\zeta\zeta\zeta]T$ branch below T_M. Observed peaks do not correspond to frequencies derived from the ultrasonic data. (See text for discussion.)

We have also collected data on a very limited number of $[\zeta 00]L$ phonons, as shown in Table II. Again, softening of about 10% is observed. This softening for the $[\zeta 00]L$ branch was predicted by Gor'kov.[9] Combined with previous neutron measurements, we now have data on $[\zeta 00]L$, $[\zeta 00]T$, $[\zeta\zeta 0]T_1$, and $[\zeta\zeta\zeta]T$. All of these phonons exhibit at least 10% softening from room temperature to $T_m = 45$ K. The most pronounced is, of course, the $[\zeta\zeta 0]T_1$ branch followed by the $[\zeta\zeta\zeta]T$ branch. This general softening is consistent with recent neutron powder measurements of the density of states of Nb_3Sn.[12]

III. PHONON HARDENING BELOW T_m

In the temperature range above T_m, our neutron data give the initial slopes of dispersion curves in good agreement with the ultrasonic measurements by Rehwald *et al.*[6] There appears, however, an indication of disagreement between the two measurements below T_m at 45 K. Figure 1(a) indicates that the $[\zeta\zeta 0]T_1$ mode hardens drastically below T_m. This was not what was observed by our previous neutron scattering studies (see Fig. 5 of Ref. 7). Because of heavy damping below T_m, well-resolved phonons could be observed only for $\zeta > 0.1$ in this branch. Nevertheless, the neutron peaks appear at considerably lower energy than extrapolation of ultrasonic velocities.

In the present measurements of the $[\zeta\zeta\zeta]$ phonons, we have managed to observe broad peaks at 25 K, as shown in Fig. 5. These measurements were done at small ζ below the break around $\zeta = 0.1$. Again, these peaks do not substantiate the drastic hardening shown by the broken line in Fig. 4, calculated from the ultrasonic data in Fig. 1(a). (Below T_m, the crystal becomes tetragonal and multidomain. However, all [111] directions remain equivalent.)

The broken lines in Fig. 1(a) for C_{11} and C_{12} below T_m were reported as "observed" ultrasonic measurements.[6] Actually, however, they were deduced from the measured values of $[\zeta\zeta 0]L$ and $[\zeta 00]T$ with the additional *assumption* that the bulk modulus

$$B = \tfrac{1}{3}(C_{11} + 2C_{12})$$

remains constant at its 49-K value. Since C_{44} shows a slight decrease below T_m, $C_{11} + C_{12}$ must increase by about 20% to account for the observed[6] increase of $[\zeta\zeta 0]L$, which is proportional to $C_{11}+C_{12}+2C_{44}$. One can easily see that this results in a highly leveraged change for $C_{11}-C_{12}$. In order to account for the observed neutron data at 25 K, one requires an increase in B of ~10% between 46 and 25 K.

The disagreement between the neutron data and ultrasonic prediction of the renormalization of $C_{11}-C_{12}$ below T_m calls into question the constant B assumption. The assumption appears plausible in that B changes by <5% between room temperature and T_m, with most of the variation occurring above ~150 K.[6] Also, the bulk modulus is not coupled to lowest order to the tetragonal distortion, so that one anticipates no strong changes in its behavior below T_m. Nevertheless, this latter argument applies to C_{44} as well, but dC_{44}/dT does, in fact, show a substantial discontinuity at T_m. Gor'kov and Dorokhov[13] conclude that B and C_{44} should have roughly similar temperature variation. Further studies are needed to clarify the behavior of C_{11} and C_{12} below T_m.

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