Internal friction and elastic softening in polycrystalline Nb_3Sn

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The vibrating-reed technique was used to measure internal friction and Young's modulus of polycrystalline Nb_3Sn in the form of composite Nb-Nb_3Sn tapes from 6 to 300 K. In tapes with only small residual strain in the *A*15 layers, a dramatic increase in internal friction with decreasing temperature is observed with an abrupt onset at ~48 K. The internal friction Q^{-1} between 6 and 48 K is believed to be associated with stress-induced motion of martensitic-domain walls. In this temperature range, Q^{-1} is approximately proportional to the square of the tetragonal strain of the martensitic phase; $Q^{-1} \propto (c/a-1)^2$. With residual compressive strains of ~0.2%, the internal friction associated with domain-wall motion is considerably reduced. This is attributed to a biasing of domain-wall orientation with residual stress, which reduces wall motion induced by the (much smaller) applied stress. The transformation temperature, however, is unchanged (within ±1 K) by residual strains of up to 0.2%. Young's modulus exhibits substantial softening on cooling from 300 to 6 K. This softening is substantially reduced in the presence of small residual compressive strains, indicating a highly nonlinear stress-strain relationship as previously reported for V_3Si.

Nb_3Sn and other superconductors of the *A*15 structure undergo a cubic-to-tetragonal phase transformation at a temperature T_m above the superconducting transition temperature T_c. This lattice instability leads to anomalous temperature variations of a number of physical quantities including the vanishing of the shear modulus $\frac{1}{2}(C_{11}-C_{12})$ and hence of Young's modulus in the [100] direction at the transition.[1,2] For Nb_3Sn and V_3Si the transformation is weakly first order and a spontaneous tetragonal strain is observed at T_m, with a gradual increase in tetragonality as the temperature is decreased towards T_c.[1,3] Below T_m (~50 K for Nb_3Sn), tetragonal crystals subdivide into twinned domains with the *C* axes lying along ⟨100⟩ directions.[4,5] In the presence of compressive stress, domains tend to orient with their shorter axes in the direction of the stress.[5]

In this Communication, audio-frequency internal friction and dynamic Young's-modulus measurements are presented for polycrystalline Nb_3Sn subjected to static strains of up to 0.2%. A large rise in internal friction Q^{-1} is reported for the tetragonal phase of Nb_3Sn, indicating that such measurements are a useful tool for detecting the presence and studying the kinetics of the transformation. This is in contrast with earlier measurements by Finlayson,[6] who reported no correlation between internal friction and the transformation in V_3Si and Nb_3Sn. Internal friction is also shown to depend on the tetragonal strain associated with the transformation $(c/a-1)$ and to decrease rapidly in the presence of a steady bias compressive strain. This behavior is shown to be consistent with stress-induced wall motion in the tetragonal phase as the main source of the inelasticity. Young's modulus is shown to decrease continuously down to $T_c \sim 18$ K. This is in contrast with single-crystal ultrasonic measurements which showed a stiffening of the elastic constants below T_m.[7] In the presence of compressive strains of ~0.2%, Young's modulus at low temperatures is found to increase by ~45%, relative to that of the strain-free specimen, indicating a highly nonlinear stress-strain relationship.

The samples were small foils of approximate dimension $12\times2\times0.3$ mm^3, cut by electrical discharge from larger tapes fabricated by the solid-state diffusion process.[8] Three samples, designated as BT62 in Table I, were made at Brookhaven by initially casting Cu–13 wt % Sn around a pure Nb strip and rolling the composite to a final thickness of ~25 μm. The other sample was made at Airco Industries by sputtering Cu–13 wt % Sn on each side of a thin (~25 μm) foil of pure Nb. The bronze-Nb composites were then reacted in vacuum at 1015 or 1040 K to form Nb_3Sn layers of 3–10 μm on each side of a central unreacted Nb layer (see Table I), cut to size, and the bronze removed in dilute nitric acid. Because the thermal-expansion coefficient of the bronze substantially exceeds that of the Nb and Nb_3Sn, cooling from the reaction temperature results in internal stress. When the bronze layer is sufficiently thick compared to the Nb ($d_{\mathrm{bronze}}/d_{\mathrm{Nb}} \geq 3$) it was found[9] that an internal stress associated with plastic flow in the Nb remains in the Nb_3Sn-Nb-Nb_3Sn composite after removal of the bronze layers. For the BT62 samples ($d_{bronze}/d_{Nb}=4$), plastic yielding of $\sim 0.25\%$ occurs in the Nb, resulting in compressive strains of ~ 0.1 and 0.2%, respectively, in the Nb_3Sn layers of samples BT62/16 and BT62/17. The plastic yielding in the Nb was deduced, as described elsewhere,[9] by etching away one of the Nb_3Sn layers and measuring the radius of curvature of the remaining Nb_3Sn-Nb strip. For the Airco tape with a bronze-to-niobium ratio $=2$ no yielding of the Nb occurs. Since the residual stress associated with a mismatch between the thermal-expansion coefficients of Nb_3Sn and Nb is less than 10^{-5} (Ref. 9) the strain in the Airco tape is $\sim 10^{-5}$. For the same reason it was possible to relieve the residual strain in sample BT62/16 by annealing at 1040 K for $\frac{1}{2}$ h, resulting in sample BT62/16A (Table I). Onset critical temperatures, measured inductively lay in the range of 17.6 to 17.8 K for all specimens. Young's modulus and internal friction were determined by measuring the resonant frequency of forced vibrations and the rate of decay of free vibrations using an experimental setup described elsewhere.[10,11] Corrections due to the finite ratio of thickness to width of the reeds and to thermal contraction are estimated at less than 0.2% and were neglected.

Because of the composite nature of the samples, analysis is required to deduce Young's modulus and internal friction in the Nb_3Sn layers. Taking the flexural rigidity and areal density of the composite as the sum over each layer,[12] one obtains the following relation for the ratio of Young's modulus at a temperature T to that at 300 K in terms of the resonant frequency, f

$$\left(\frac{E_T}{E_{300}}\right)_1 = \left(\frac{f_T}{f_{300}}\right)^2 + \left(\frac{E_0}{E_1}\right)_{300}\left[\left(\frac{f_T}{f_{300}}\right)^2 - \left(\frac{E_T}{E_{300}}\right)_0\right]\left[\left(\frac{Z}{Z_0}\right)^3 - 1\right]^{-1} , \tag{1}$$

where Z is the thickness of the reed and indices 0 and 1 correspond, respectively, to the central and outer layers. The values of (E_T/E_{300}) for niobium were taken from Jones *et al.*[13] Similarly, an analysis for internal friction in the Nb_3Sn layer can be obtained by assuming that Q^{-1} for the composite is the sum of the energy loss over each layer divided by the sum of the elastic energies,[14] resulting in

$$Q^{-1} = \left\{Q_0^{-1} + Q_1^{-1}\frac{E_1}{E_0}\left[\left(\frac{Z}{Z_0}\right)^3 - 1\right]\right\} \Big/ \left\{1 + \frac{E_1}{E_0}\left[\left(\frac{Z}{Z_0}\right)^3 - 1\right]\right\} , \tag{2}$$

where Q^{-1}, Q_0^{-1}, and Q_1^{-1}, are, respectively, the internal friction in the composite, the central layer, and the outer layers.

The temperature dependences of Young's modulus in the Nb_3Sn layers of the three BT62 samples under bias compressive strains of 0, 0.1, and 0.2% are shown in Fig. 1. For all three samples Young's modulus decreases dramatically as the temperature is reduced from 300 to 6 K with ratios E_6/E_{300} of 0.42, 0.53, and 0.69, respectively, for strains of 0, 0.1, and 0.2%. Measurements on a number of other unstrained samples, including the Airco listed in Table I gave values of E_6/E_{300} in the range of 0.38 to 0.42. These results are in excellent agreement with previously published data based on static-beam-deflection measurements.[9]

The large increase of modulus with compressive strain is a clear indication of a highly nonlinear stress-strain relationship similar to the behavior observed by Patel and Batterman in V_3Si.[15] This behavior is observed over the entire range of temperature between 6 and 300 K but is more pronounced at low temperatures. As described later all three samples of Fig. 1 transformed at ~ 49 K.

TABLE I. Description of Nb_3Sn-Nb composite samples.

Sample	Total thickness Z (μm)	Thickness of central Nb Z_0 (μm)	Compressive strain in Nb_3Sn (%)	Heat treatment	Frequency[a] at 300 K f_{300} (Hz)
Airco	27.6	12.0	$<10^{-3}$	750 °C, 189 h	1877
BT62/16	23.0±0.3	15.5	0.1	725 °C, 50 h	509
BT62/16A	23.0±0.3	15.5	$<10^{-3}$	725 °C, 50 h + anneal without bronze 750 °C, $\frac{1}{2}$ h	715
BT62/17	36.8±1.5	32.8	0.2	725 °C, 50 h	707

[a]First harmonic.

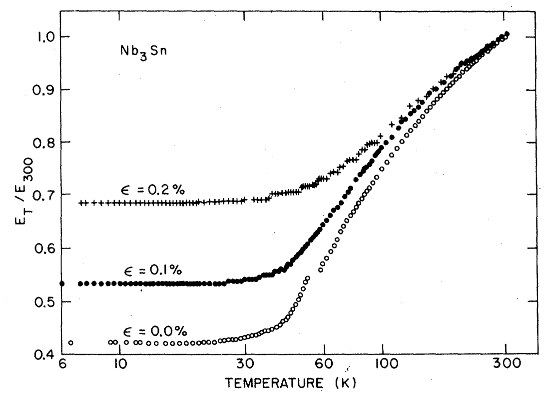

FIG. 1. Ratio of Young's modulus as a function of temperature for Nb_3Sn under compressive strains of 0.0% (BT62/16A), 0.1% (BT62/16), and 0.2% (BT62/17).

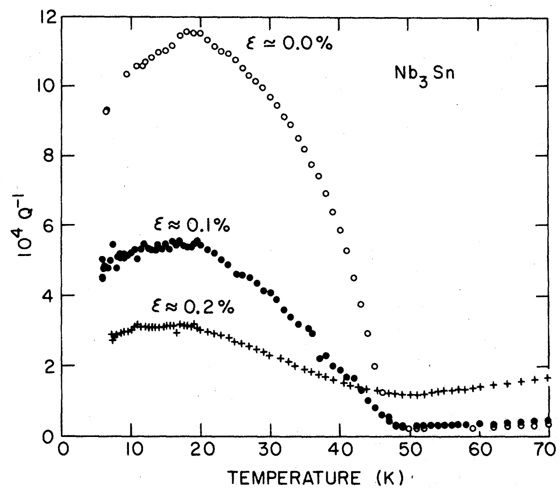

FIG. 2. Temperature dependence of internal friction for Nb_3Sn under compressive strains of 0.0% (BT62/16A), 0.1% (BT62/16), and 0.2% (BT62/17).

Young's modulus, however, is seen to decrease smoothly down to 18 K with no discontinuity in slope at 49 K. Appreciable softening is also found to occur between 49 and 18 K in contrast with the Voigt-Reuss-Hill polycrystalline average based on Rehwald *et al.*'s[7] single-crystal data, which yield a modulus increasing rapidly to nearly the room-temperature value on cooling from T_m to 4.2 K. Although we have no definite explanation for the softening below T_m, it may be associated with untransformed material which softens to T_c as observed by Keller and Hanak.[16] From x-ray diffraction measurements[17] we estimate that less than ~50% of the sample did not transform. The discrepancy may also be due to softening associated with reorientation of domains under stress; if so, the discrepancy would be frequency dependent. At the high frequencies used for ultrasonic measurements (typically 100 MHz) domain motion may be impeded by viscous drag and domain-wall inertia effects, resulting in a stiffer modulus than observed at the low frequencies employed here.

Internal-friction results below 70 K for the three BT62 samples are shown in Fig. 2. The raw data for the composite strips were corrected for the presence of the central Nb layer using Eq. (2), and assuming that $Q_0^{-1} = 0$. For the unstrained specimen, Q^{-1} increases rapidly as the temperature is decreased below ~48 K, reaching at 18 K a maximum ~40 times larger than the background level. The strained specimens show a similar behavior with the same onset temperature (~48 K) but with a smaller increase in Q^{-1}. The higher background level above 48 K for sample BT62/17 (0.2% strain) is believed associated with the strained Nb substrate. Below 18 K (T_c) all samples show a decrease in Q^{-1}, this decrease being less pronounced in the presence of strain.

Measurements on a number of other samples, including the Airco listed in Table I gave similar results with onset temperatures between 40 and 49 K. Normalized data for the Airco sample (onset temperature ~49 K) are given in Fig. 3; no hysteresis was observed for measurements taken during heating and cooling.

To confirm the occurrence of a transformation in our samples, x-ray measurements were carried out on the Airco sample and a tape processed similarly to the BT62 sample. Both samples showed splitting of the [400] lines below 50 K. Similar observations were also recently reported for bronze-processed multifilamentary wires.[18] We therefore associate the rise in internal friction below 49 K to the occurrence of the tetragonal-to-cubic phase transformation.

In the vicinity of a martensitic transformation, several physical processes can contribute to the internal friction. The relative contribution of these

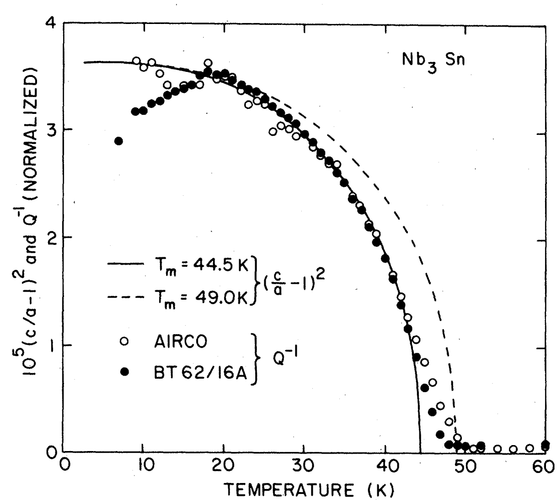

FIG. 3. Comparison between the temperature dependences of $(c/a-1)^2$ and Q^{-1} for two polycrystalline samples of Nb_3Sn. Values of $(c/a-1)^2$ are shown for transformation temperatures, T_m, of 44.5 and 49 K.

processes can be inferred by studying internal friction as a function of temperature, frequency, and amplitude.[19] The high level of Q^{-1} well below T_m and the absence of a peak in Q^{-1} near T_m imply a high mobility of tetragonal domain walls and stress-induced wall motion between tetragonal domains as the main source of internal friction.[19] This interpretation is also consistent with the slight increase in Q^{-1} with frequency recently observed in similar material,[20] the absence of hysteresis on cooling and heating, and the reduction of Q^{-1} in the presence of a bias stress. The absence of hysteresis is an indication that the quantity of martensite does not change appreciably with temperature (making negligible any contribution due to stress-induced nucleation of martensite below T_m), in agreement with the nearly second-order nature of the transformation.[3] The reduction of Q^{-1} with bias strain is consistent with a saturation of wall motion as the domains are aligned by the bias stress.

Internal friction associated with tetragonal domain-wall motion may be expected on general grounds to depend on tetragonality. Assuming that the mobility of domains is independent of tetragonality and temperature, one expects that Q^{-1} will be proportional to $(c/a-1)^2$. This arises because the strain produced when a domain wall moves is proportional to $(c/a-1)$ and the force on the domain wall produced by the stress leading to the motion is also proportional to $(c/a-1)$. The product of these two factors enters into Q^{-1} and therefore $Q^{-1} \propto (c/a-1)^2$. As shown in Fig. 3, this is in reasonable agreement with experimental observation; i.e., the temperature dependence of Q^{-1} for Nb_3Sn is largely a result of the temperature dependence of $(c/a-1)$ between T_m and T_c. The values of c and a in Fig. 3 were taken from Mailfert *et al.*[21] and the temperature scale normalized assuming that c/a depends on T/T_m only.

Note that a better fit to the data is obtained with $T_m = 44.5$ K (the transformation temperature observed by Vieland *et al.*[3] than with $T_m = 49$ K (temperature of onset of rise in Q^{-1}). This may be due to a distribution of transformation temperatures in the polycrystalline samples, to a different dependence of $(c/a-1)$ on temperature, or to the oversimplifying assumption which led to $Q^{-1} \propto (c/a-1)^2$.

In conclusion, we have shown that internal friction can be more than an order of magnitude higher in the tetragonal phase of polycrystalline Nb_3Sn than in the cubic phase, and that the temperature dependence of Q^{-1} follows approximately $(c/a-1)^2$. This behavior was interpreted as due to stress-induced motion of highly mobile domain walls below T_m. These observations provide a new tool for studying the occurrence of the phase transformation and the kinetics of domain-wall motion. Simultaneous measurements of dynamic modulus indicated the presence of considerable softening on cooling to the superconducting transition temperature T_c. Substantial softening was also observed below T_m, in contrast with the large increase in modulus expected from polycrystalline averages of the single-crystal constants of transforming Nb_3Sn. This behavior may be associated with reorientation of domains under stress, an effect not normally taken into account in polycrystalline averages. Young's modulus and internal friction were also shown to depend strongly on compressive strain although the transformation temperature was unaffected by these strains.

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