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Electronic density of states and T_c in Nb_3Sn under pressure

K. C. Lim and J. D. Thompson
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

G. W. Webb
Institute for Pure and Applied Physical Sciences, University of California at San Diego,
La Jolla, California 92903
and Energy Science Laboratories, San Diego, California 92121
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We have determined the effect of hydrostatic pressure on the electronic heat-capacity coefficient γ of transforming Nb_3Sn through measurements of the superconducting transition temperature T_c, the temperature derivative of the upper critical field near T_c, and the residual resistivity. We find that γ and the bare density of electronic states are suppressed by pressure. Results are discussed in terms of a pressure-dependent d-band occupancy. From an analysis of the density-of-states change under pressure, we infer the existence of a substantial electronic contribution to the Grüneisen parameter of Nb_3Sn.

I. INTRODUCTION

Compounds with the highest known superconducting transition temperatures T_c are those having the $A15$ crystal structure. A feature characteristic of this structure is the occurrence of three mutually orthogonal chains lying along cube faces. High-T_c compounds are those in which the chains are composed of the transition-metal atoms Nb or V. This chain structure led Weger[1] to propose a model and later Labbé and Friedel[2] to develop a model of $A15$ compounds in which the one-dimensionality of transition-metal atoms gives rise to a high and rapidly varying density of electronic states $N(0)$ near the Fermi energy E_F, a condition favorable to strong electron-phonon coupling and hence a high T_c. Measurements on the high-T_c $A15$ compounds Nb_3Sn and V_3Si that show strong thermal variations in quantities such as the magnetic susceptibility, elastic constants, and Knight shift have been interpreted as evidence supporting the Weger-Labbé-Friedel (WLF) model.[3] Recent self-consistent band-structure calculations,[4–6] however, now provide strong theoretical evidence suggesting that the WLF assumption of noninteracting one-dimensional chains is not justified but that the WLF model simply parametrizes a high and structured density of states which follows directly from first-principles calculations. Even though the WLF premise appears to be incorrect, these band calculations have confirmed that several high-T_c $A15$'s, including Nb_3Sn and V_3Si, do have moderately high, strongly energy-dependent densities of states near E_F, suggesting that $N(0)$ may play a prominent role in producing high T_c's in these compounds.

Another interesting aspect of Nb_3Sn is the anomalously large softening of its elastic shear modulus at low temperatures.[7] In extreme cases, this softening leads to a cubic-to-tetragonal crystallographic transformation at a temperature $T_M \sim 50$ K. Because the electron-phonon coupling parameter λ is proportional to an electronic term involving $N(0)$ divided by a mean-squared phonon energy, softening of the phonon spectrum, as evidenced by the tetragonal distortion, would be expected to enhance T_c. This has led to the proposal that soft phonons are primarily responsible for high T_c's in those compounds exhibiting a structural transformation.[7] However, Bilbro and McMillan[8] have suggested that the Martensitic transformation actually may depress T_c somewhat. In their model, the transformation is driven by a Peierls-type electronic instability in the linear-chain structure of the $A15$ lattice. The net result of the instability is a Peierls gap opening at E_F, which reduces the number of electrons available for BCS pairing and hence decreases T_c relative to the enhancement expected by lattice softening. Indeed, one commonly finds lower T_c's in transforming samples compared to nontransforming samples.

A controversy, similar to that encountered in assessing the relative importance of $N(0)$ and soft phonons on T_c at ambient pressure, is found in discussions of the effect of hydrostatic pressure on T_c.[9] In a comprehensive survey of the pressure dependence of T_c for a wide selection of $A15$ compounds, Smith[10] found no correlation between either the sign or magnitude of the pressure derivative of T_c,

$(\partial T_c/\partial P)$, and quantities such as the e/a ratio and T_c. His results did indicate, however, that generally $\partial T_c/\partial P$ is positive for vanadium-based $A15$'s and negative for those based on niobium, although there are notable exceptions to this trend. Within the framework of an electronic interpretation of this observation, variations in these trends may be expected because of disparities in the detailed electronic structure near E_F. Since Smith's review, the effect of pressure on T_c and T_M has been studied in Nb_3Sn.[11] These experiments showed that pressure enhances T_M and suppresses T_c. That opposite signs are found for the pressure derivative of T_M and T_c is consistent with the viewpoint that the primary effect of pressure is to shift the phonon spectrum to higher energies. If soft phonons play the dominant role, then one should invariable find $\mathrm{sgn}(\partial T_c \partial P) = -\mathrm{sgn}(\partial T_M/\partial P)$. However, this is not always the case.[9] Difficulties arising from the phonon interpretation are circumvented by considering the pressure dependence of T_c and T_M to be governed not by phonons but by the d-band occupancy Q. Using the WLF model density of states, Labbé[12] found that the signs of $\partial T_c/\partial P$ and $\partial T_M/\partial P$ are determined by Q and its pressure derivative relative to Q values that maximize T_c and T_M. Therefore no correlation is necessarily expected between the effect of pressure on superconductivity and the structural instability.

Clarification of controversies surrounding $A15$ compounds is fundamental to an improved understanding of high-temperature superconductivity. To this end, we have undertaken measurements aimed specifically at establishing whether electronic changes are induced by the application of hydrostatic pressure. If pressure does produce electronic changes, these may be reflected in a systematic variation of the electronic heat-capacity coefficient γ as a function of pressure. From appropriate Ginzburg-Landau relationships, it is possible to calculate γ as a function of pressure through measurements of T_c, the temperature derivative of the upper critical field near T_c (H'_{c2}), and the residual resistivity ρ_0. Using this procedure, we have established direct evidence for pressure-induced electronic changes in both transforming and nontransforming V_3Si.[13] Our observations on V_3Si were interpreted as providing support for the idea of pressure-induced interband charge transfer. The primary purpose here is to search for the existence and nature of pressure-induced electronic changes in transforming Nb_3Sn and to determine whether the concept of charge transfer is also applicable to Nb_3Sn.

In Sec. II we give details of the sample preparation and experimental procedure used in these measurements. Section III contains a brief account of the theoretical foundation upon which our measurements are based. Results on Nb_3Sn are presented in Sec. IV and discussed in Sec. V. Conclusions are summarized in Sec. VI.

II. EXPERIMENTAL

A. Sample preparation and characterization

Nb_3Sn was prepared by closed-tube vapor transport using iodine as the transporting agent. Growth was carried out at 600 to 1000 °C over a period of about four months. From a group of crystals an irregularly shaped single crystal with approximate dimensions of $1\times1.5\times1.5$ mm^3 was selected for measurement. X-ray diffraction analysis on deposits grown at the same time showed only sharp lines characteristic of the $A15$ structure and gave a room-temperature lattice parameter of 5.290 ± 0.001 Å. Ambient-pressure resistance measurements on this sample revealed an anomaly near 50 K that we associate with a cubic-to-tetragonal lattice distortion. The resistively measured midpoint T_c was 17.87 K with a transition width ΔT_c (10–90%) of 0.08 K. Values of T_c and ΔT_c are in excellent agreement with those measured inductively on another single crystal grown at the same time and used in de Haas–van Alphen experiments.[14] The resistance ratio between room temperature and 19.4 K was 15.6. In the temperature range $T_c < T < 40$ K, the resistance followed a T^2 behavior, in agreement with previous observations.[15] Assuming that the T^2 dependence continued to $T=0$, we estimated a defect-limiting resistance ratio $R(300\text{ K})/R(0\text{ K}) = 45$. Resistance measurements on another polycrystalline part of the deposit also showed an approximate T^2 dependence and $R(300\text{ K})/R(0\text{K}) = 50$.

B. Measurements

Hydrostatic pressure was generated in a self-clamping beryllium-copper cell, with a 1:1 mixture of isoamyl alcohol and n-pentane as the pressure-transmitting medium. Pressure in the cell was determined at low temperatures by a lead manometer located in close proximity to the sample. Before measuring the lead transition temperature, we carefully demagnetized the cell to ensure that any remanent magnetic field in tungsten-carbide components was removed.

Liquid hydrogen was employed as the cryogen for measurements in the temperature range $14 < T < 20$ K. The temperature of the liquid-hydrogen bath could be determined, from its vapor pressure, and maintained to an accuracy of ±10 mK. The use of liquid hydrogen proved extremely valuable because it eliminated the need for tedious magnetic field corrections to our carbon-glass thermometer, which was embedded in the side of the cell, and ensured good thermal equilibrium between the cell and sample. To reach temperatures outside the liquid-hydrogen range, the cell was slowly lowered toward a pool of boiling liquid helium. Previously, we have established this as a reliable technique in zero magnetic field.[16]

The upper critical-field slope H'_{c2}, T_c, and residual resistance were measured using a four-terminal ac technique. Values we quote for T_c and H_{c2} represent resistive midpoint transitions. An indication of the degree of hydrostaticity present in our pressure cell is provided by a measure of transition-width broadening at pressure. We find that at the highest pressure (16 kbar) ΔT_c has increased by only 20 mK relative to ΔT_c of 80 mK at $P=0$. To determine H'_{c2}, H_{c2} was measured typically at 10 to 12 different temperatures in the range $14\text{ K} < T < T_c$. Magnetic fields up to 9 T were generated by a superconducting solenoid. The field was homogeneous to $\pm 0.1\%$ over the high-pressure volume.

III. THEORETICAL RELATIONS

The electronic heat-capacity coefficient γ can be calculated through the Ginzburg-Landau relationship[17]

$$-H'_{c2}=\frac{\eta_{H_{c2}}(T_c)}{R(\lambda_{tr})}\times\left[9.55\times10^{24}\gamma^2 T_c\left[\frac{n^{2/3}S}{S_F}\right]^{-2}+5.26\times10^{4}\gamma\rho_0\right]. \qquad (1)$$

In Eq. (1) S is the Fermi-surface area, S_F is the Fermi-surface area of a free-electron gas of density n, ρ_0 is the residual resistivity, and $R(\lambda_{tr})$ is a correction factor of order unit $[R(\lambda_{tr})\simeq 1.003$ and decreases slowly toward unity with increasing pressure]. The effect of strong coupling corrections to H_{c2} near T_c is contained in $\eta_{H_{c2}}(T_c)$, which depends only on T_c and a frequency characteristic of the phonon distribution. The pressure dependence of $\eta_{H_{c2}}(T_c)$ is governed primarily by $\partial T_c/\partial P$. Neglecting any reasonable pressure-induced shift in the phonon spectrum in calculating $\eta_{H_{c2}}(T_c)$ produces at most a 0.3% uncertainty in the pressure dependence of γ. Orlando *et al.*[17] have shown that $S/S_F=0.35$ is characteristic of nontransforming Nb_3Sn thin films at ambient pressure. The ratio S/S_F was selected by requiring that γ calculated from Eq. (1) agree with heat-capacity measurements by Vieland and Wicklund.[18] In light of the prevailing controversy concerning the interpretation of heat-capacity measurements on Nb_3Sn, we have chosen to adjust S/S_F to agree with the more recent heat-capacity results[19] on a transforming polycrystalline sample of Nb_3Sn (also grown at the same time as our sample) which show that γ is notably smaller than previously supposed. With this normalization, we find $S/S_F=0.25$. We note that this value is also consistent with the measurements of Orlando *et al.* when allowance is made for the revised γ. In calculating $\gamma(P)$, we assume that $n^{2/3}S/S_F$ is pressure independent. Because $\partial T_M/\partial P>0$, the Bilbro-McMillan model for the structural transformation suggests that S/S_F decreases with increasing pressure. Because this ratio enters Eq. (1) as $(\gamma S_F/S)^2$, for a given H'_{c2}, we underestimate changes in γ *provided* γ decreases with pressure.

Changes in the bare density of electronic states under pressure are found through the relationship

$$N(0)=\gamma/[\tfrac{2}{3}\pi^2k_B^2(1+\lambda)]\ , \qquad (2)$$

where λ is the electron-phonon mass enhancement parameter. From the measured pressure dependence of T_c, we may estimate $\lambda(P)$ by numerically inverting the equation[20]

$$T_c=\left[\frac{f_1f_2\omega_{\log}}{1.2}\right]\left[\exp-\frac{1.04(1\pm\lambda)}{\lambda-(1+0.62\mu^*)\lambda}\right], \qquad (3)$$

where $\omega_{\log}$ is a logarithmically averaged phonon frequency, μ^* is the Coulomb interaction parameter, and functions f_1 and f_2 are given by Allen and Dynes.[20] To a very good approximation f_1 and f_2 are insensitive to pressure variations in the phonon spectrum. We take $\omega_{\log}=10.8$ meV (Ref. 21) and $\mu^*=0.13$. Because the pressure dependence of $\omega_{\log}$ is unknown, we assume that it increases linearly with pressure by 3% up to 16 kbar. This assumption is consistent with estimates[22,23] of the lattice contribution to the Grüneisen parameter of Nb_3Sn.

IV. RESULTS

Results of our measurements on Nb_3Sn are summarized in Fig. 1. From a linear least-squares fit to $T_c(P)$ data, we find $\partial T_c/\partial P=-2.2\times10^{-5}$ K/bar. Chu and Vieland[24] have determined the pressure dependence of T_c on a transforming single crystal of Nb_3Sn using a calorimetric technique. In contrast to our results, they found $\partial T_c/\partial P=-1.4\times10^{-5}$ K/bar. This discrepancy may be attributed to

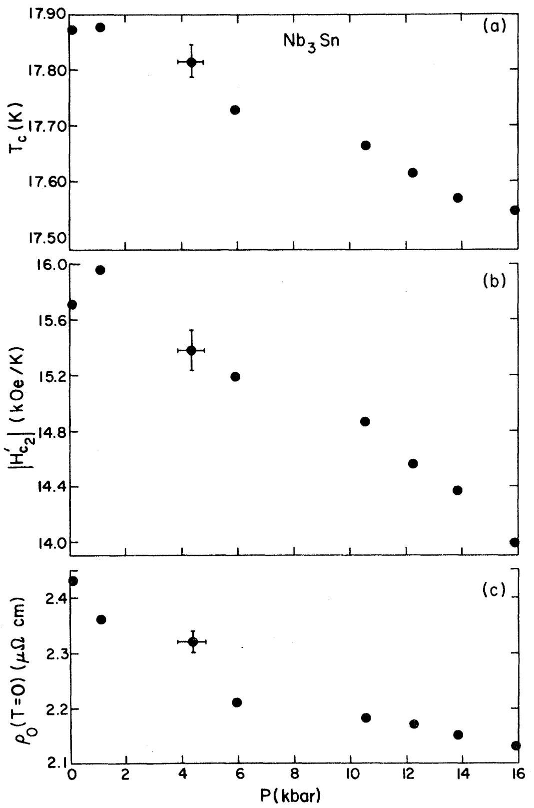

FIG. 1. (a) Midpoint transition temperature, (b) temperature derivative of the upper critical field near T_c, and (c) extrapolated $T=0$ K residual resisitivity of transforming Nb_3Sn as functions of pressure. Error bars represent estimated uncertainty in absolute values of T_c (±30 mK) and of H'_{c2} (±150 Oe/K) and relative uncertainty in ρ_0 (±1%). Error in measuring the pressure is estimated to be ±0.5 kbar.

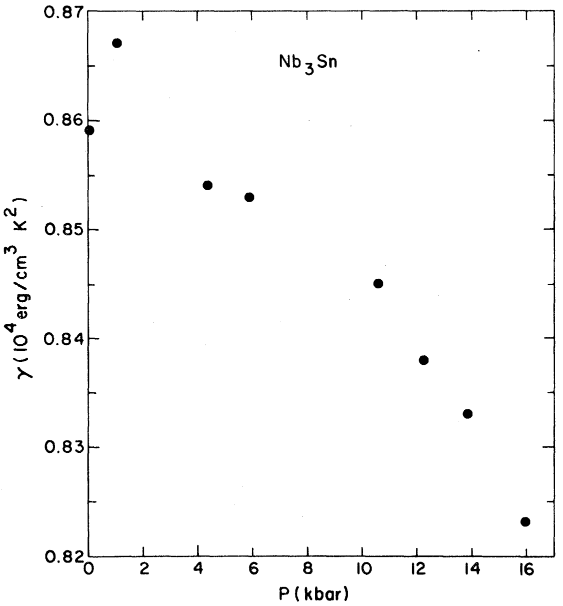

FIG. 2. Electronic coefficient of the heat capacity as a function of pressure.

differences in sample quality. Their sample had a resistance ratio of seven between 300 and 20 K, whereas ours is over 15. Determinations of $\partial T_c/\partial P$ on several V_3Si samples have shown that the pressure derivative of T_c is strongly sample dependent.[25] We expect this to be true also of Nb_3Sn. Contrary to our observations on transforming V_3Si,[13] in which $\partial H'_{c2}/\partial P > 0$, we find that H'_{c2} for Nb_3Sn is depressed by pressure. We note that the zero-pressure value of H'_{c2} agrees very well with that determined on the de Haas–van Alphen Nb_3Sn single crystal[26] which is of comparable quality. The relatively low H'_{c2} is consistent with the low residual resistivity of our sample and the fact that it transforms. We should also point out that we do not know the crystallographic orientation of the sample with respect to the applied magnetic field. However, this should not affect our conclusions since Foner and McNiff[26] have shown that the critical-field anisotropy is small and temperature independent. Finally, we see in Fig. 1(c) that the resistivity extrapolated to $T=0$ decreases with increasing pressure. The resistivity at 19.4 K follows the same pressure dependence as ρ_0 ($T=0$).

With the data of Fig. 1, we can calculate γ from Eq. (1). The pressure variation of γ is displayed in Fig. 2. The existence of pressure-induced changes in γ is obvious. At 16 kbar, γ has decreased by more than 4% from its zero-pressure value. This variation is well outside our estimated uncertainty (±1%) in the relative values of γ vs P. As indicated in Sec. III, because γ decreases with pressure, we are probably underestimating its suppression. The point at 1 kbar appears anomalous compared to the relatively small scatter in the rest of the data; however, it does fall within our experimental error. A smooth extrapolation of the high-pressure data to near the zero-pressure point is likely to be characteristic of $\gamma(P)$.

The density-of-states contribution to γ can be calculated from Eq. (2). However, the pressure dependence of the electron-phonon coupling must be determined first. Using the procedure outlined in Sec. III, we have obtained $\lambda(P)$, which is plotted in Fig. 3. Two cases have been considered for the effect of pressure on the phonon spectrum, corresponding to the two data sets shown. The upper set of points corresponds to a pressure independent $\omega_{\log}$, while the lower set reflects the behavior of λ assuming $\omega_{\log}$ increases linearly by 3% at 16 kbar. With

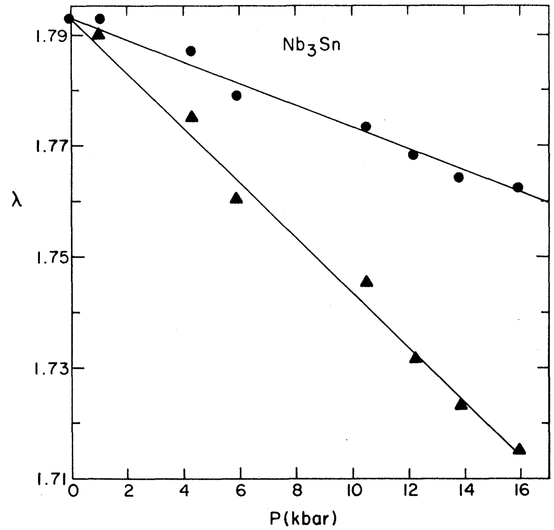

FIG. 3. Electron-phonon coupling parameter as a function of pressure. Upper data set calculated assuming $\omega_{\log}$ is pressure independent; lower set obtained assuming $\omega_{\log}$ increases linearly with pressure by 3% at 16 kbar. Lines through the data are to indicate the nearly linear dependence of λ on P.

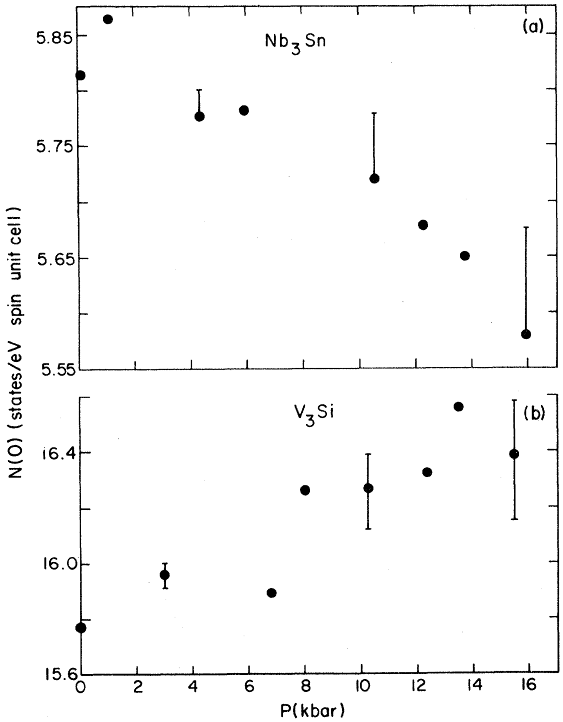

FIG. 4. (a) Bare density of electronic states at the Fermi energy as a function of pressure for Nb_3Sn. Vertical bars correspond to assumed pressure variations in $\omega_{\log}$. (b) $N(0)$ as a function of pressure for transforming V_3Si. Data taken from Ref. 13.

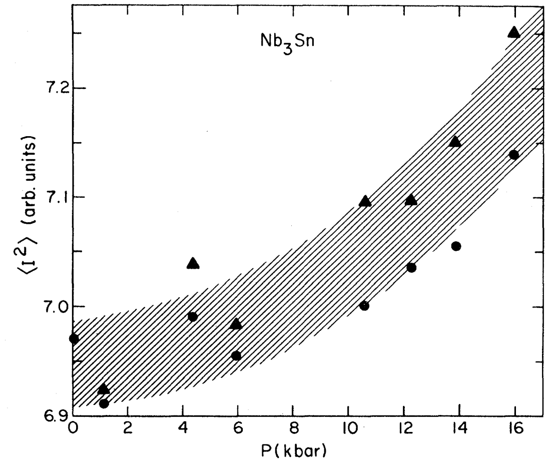

FIG. 5. Average electron-phonon matrix element squared as a function of pressure. ●—calculated assuming $\langle \omega^2 \rangle$ is pressure independent. ▲—calculated assuming $\langle \omega^2 \rangle \propto [\omega_{\log}(P)]^2$.

this information, we can calculate $N(0)$ vs P. Our results are shown in Fig. 4(a). Vertical bars on representative points are indicative of our assumed pressure variation in $\omega_{\log}$. Even with this allowance, there is still clearly a trend for $N(0)$ to decrease under pressure. For comparison, we show in Fig. 4(b) the pressure dependence of $N(0)$ for a transforming V_3Si sample. The contrast is striking and reflects the influence of $\partial N(0)/\partial P$ on determining the sign of $\partial T_c/\partial P$.

The coupling parameter is commonly written as $\lambda = N(0)\langle I^2 \rangle / M \langle \omega^2 \rangle$, where $\langle I^2 \rangle$ is the average electron-phonon matrix element squared, M is the ionic mass, and $\langle \omega^2 \rangle$ is a squared characteristic phonon frequency. From the data in Figs. 3 and 4(a), $\langle I^2 \rangle$ can be calculated, assuming the pressure dependence of $\langle \omega^2 \rangle$ is given by $[\omega_{\log}(P)]^2$. $\langle I^2 \rangle$ as a function of pressure is shown in Fig. 5. Although considerable scatter in the data is evident, a tendency toward larger $\langle I^2 \rangle$ with pressure is indicated. This is understandable since pressure-induced band broadening results in reduced screening and, hence, an increase in $\langle I^2 \rangle$. Because $\langle I^2 \rangle$ has been deduced from a product of calculated quantities, the experimental error has been compounded. The maximum change in $\langle I^2 \rangle$ given by the lower bound in Fig. 5 is just outside our estimated uncertainty in the relative pressure variation of $\langle I^2 \rangle$. Therefore, if $\langle I^2 \rangle$ increases at all under pressure, the effect is small.

V. DISCUSSION

A. Charge transfer

From self-consistent atomic-sphere calculations on $4d$ transition metals, Pettifor[27] has established theoretically that pressure broadens the narrow *d* bands and shifts both the bottom of the *s* band and E_F up in energy. These calculations imply a pressure-dependent *d*-band occupancy Q. Based on a WLF density-of-states model, Labbé[12] has calculated T_c and T_M as functions of Q. For Nb_3Sn, he found that T_c and T_M reach a maximum at Q_{cM} and Q_{MM}, respectively, where $Q_{cm} < Q < Q_{MM}$. Provided charge transfer is from *s* to *d* bands, these calculations imply $\partial T_c/\partial P < 0$ and $\partial T_M/\partial P > 0$, in agreement with experiment.[11] Similar arguments applied to V_3Si successfully predict both the sign difference for the pressure derivatives of T_c and T_M (Ref. 9) and the thermal response of the shear modulus pressure derivative.[28] When considered in terms of band-structure calcuations for cubic V_3Si, a shift of E_F to higher energies due to increased *d*-band occupancy qualitatively explains pressure-induced enhancement of $N(0)$ in nontransforming V_3Si.[13] We emphasize that these conclusions are based on general arguments[27] not specific to the Labbé[12] model but in agreement with it. Interestingly, $\partial N(0)/\partial P$ is comparable in transforming V_3Si [Fig. 4(b)], which suggests that the energy dependence of the density of states near E_F in tetragonal V_3Si is similar to that in cubic V_3Si.

Recent band-structure calculations by Weber and Mattheiss[29] show that the tetragonal distortion in Nb_3Sn produces, on the scale of millirydbergs, significant modifications to the cubic Nb_3Sn band structure. They find that the Γ_{12} subband near E_F is split by 6—8 mRy into a pair of equally flat Γ_{1+} and Γ_{3+} subbands in the tetragonal phase. Furthermore, their calculations show that the Fermi energy is shifted up in energy, relative to E_F in the cubic phase, so that it resides on an increasing portion of the density-of-states curve instead of at a point where $N(0)$ has a negative energy derivative, as is the case for cubic Nb_3Sn. Therefore, on the scale of a few mRy about E_F, the density-of-states behavior of tetragonal Nb_3Sn is similar to that of cubic V_3Si.[5] Considering these band-structure results and assuming pressure moves E_F to higher energies, we are led to conclude that $N(0)$ vs P should be similar in cubic V_3Si and tetragonal Nb_3Sn. Figure 4 shows clearly that this is not the case. [We note again that the pressure dependence of $N(0)$ is similar for both transforming and nontransforming V_3Si.] There are at least two possible explanations for this discrepancy: (1) Either the band-structure calculations incorrectly assign the position of E_F relative to the Γ_{3+} subband, or (2) E_F moves down with pressure. Concerning the first possibility, we note that an uncertainty of approximately 2 mRy in the positioning of E_F could remove the inconsistency between our observations and the band-structure results. In spite of impressive advances in the accuracy of band calculations, uncertainty on the order of a few mRy remains a problem. Furthermore, electron lifetime broadening of sharp density-of-states features has been neglected in these calculations but many play an important role on such a fine energy scale.[30] The second possibility is unlikely since pressure-promoted band broadening moves E_F to higher energies.[27] However, under pressure, the motion of E_F relative to the Γ_{3+} state could produce an effective shift of E_F down the density-of-states peak. Confirmation of this possibility must await future band-structure calculations.

B. Grüneisen parameter

From thermal-expansion measurements, Smith *et al.*[23,31] have calculated the total Grüneisen parameter γ_G as a function of temperature for transforming Nb_3Sn. They find that as the temperature is lowered from approximately 80 K, γ_G begins to increase rapidly from its high-temperature limit of 2 to as large as 4—6 near T_c. Smith and Finlayson[31] proposed that this anomalous behavior is a consequence of significant lattice anharmonicity associated with the structural instability. However, this interpretation is inconsistent with the sign of $\partial T_M/\partial P$. That is, a large positive γ_G implies enhanced lattice stability, whereas enhanced lattice instability is suggested by a positive pressure derivative of T_M. The total Grüneisen parameter is the sum of lattice and electronic contributions. Because they were unable to separate the thermal-expansion coefficient into its lattice and electronic components, it was not possible for them to assess the relative importance of these two contributions to γ_G. From the data of Fig. 4(a), we may estimate the electronic component γ_e, which is defined through the relationship $\gamma_e \equiv \partial \ln N(0)/\partial \ln V$. This may be rewritten as $-B\partial \ln N(0)/\partial P$, where B is the bulk modulus, which we take equal to 1.65×10^3 kbar.[32] Assuming $N(0)$ decreases linearly with pressure for $P < 10$ kbar, we find $\gamma_e = 2.5\pm 1$. At higher pressures, γ_e will be larger due to the negative curvature in $N(0)$ vs P. Therefore, our results suggest that there is a significant electronic contribution to γ_G at low temperatures. Because of differences between our sample and the ones studied by Smith *et al.*, it is difficult to determine if the electronic contribution completely resolves the anomalous γ_G behavior; however, our findings do indicate that lattice anharmonicity does not play as important a role as previously supposed.

VI. CONCLUSIONS

We have established direct evidence for pressure-induced electronic changes in transforming Nb_3Sn

and argue that these changes may be due to a pressure-enhanced d-band occupancy. Density-of-state behavior is consistent with band-structure calculations for cubic Nb_3Sn, but discrepancies on the scale of a few mRy exist with calculations for tetragonal Nb_3Sn. From an analysis of $N(0)$ vs P, we are able to estimate the electronic contribution to the low-temperature Grüneisen parameter and find it to represent a significant fraction of the anomalously high γ_G deduced from thermal-expansion measurements. Finally, we note a correlation between the signs of $\partial N(0)/\partial P$ and $\partial T_c/\partial P$ for both Nb_3Sn and V_3Si, even when a reasonable allowance is made for possible pressure-induced phonon changes. Therefore electronic effects appear to be a prominent factor in determining $\partial T_c/\partial P$ in these compounds.

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