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Specific heat of A15 Nb_3Sn in fields to 18 tesla

G. R. Stewart and B. Cort
University of California, 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 92093
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For the first time, the low-temperature specific heat of A15 Nb_3Sn has been measured in a magnetic field (18 T) high enough to significantly suppress the superconducting transition temperature, T_c. All previous extrapolations to $T=0$ of the normal-state specific heat, C_n, to determine γ and $N(0)$ are shown by the measured C_n data to be incorrect. The Debye temperature, Θ_D, has previously been assumed to vary smoothly with temperature from its low-temperature value [Θ_D ($T < 4$ K) = 208 K] to its higher-temperature value [$\Theta_D(T > T_c) = 270$ K]. The high-field measurement of $\Theta_D(T)$ shows that it remains constant upon cooling until 11 K, where it abruptly changes to its low-temperature value. The rather short extrapolation of the high-field normal-state data to $T=0$ from $T > T_c$ (18 T) yields a γ of 35 ± 3 mJ/mole K^2 compared to the previously accepted value of 52 mJ/mole K^2. Using this lower value for γ, and taking $\lambda = 1.75$ from tunneling results, implies $N(0) = 1.35 \pm 0.1$ states/eV atom, in excellent agreement with recent band-structure calculations of Klein *et al.* who obtained 1.46 states/eV atom for A15 Nb_3Sn. The present work calls into question all other extrapolations of C_n data in high-T_c materials where Θ_D is known to change significantly between T_c and 0 K, e.g., A15 V_3Si.

I. INTRODUCTION

The extrapolation of the normal-state specific-heat data for A15 Nb_3Sn from above the superconducting transition temperature to 0 K is the most unusual of any high-T_c superconductor. In the work of Vieland and Wicklund,[1] and in later studies in zero magnetic field, the straightforward extrapolation to 0 K of C_n/T versus T^2 was merely a straight line, since for A15 Nb_3Sn, the normal-state specific heat, C_n, above T_c obeys

$$C_n = \gamma T + \beta T^3 \ ,$$
$$C_n/T = \gamma + \beta T^2 \ . \tag{1}$$

Thus, on a C/T versus T^2 plot, the extrapolated intercept is γ, proportional to the electronic density of states at the Fermi energy, $N(0)$ and the slope is β, inversely proportional to the Debye temperature, Θ_D, of the lattice as given by the following equations:

$$N(0)(1+\lambda) = 0.1061\gamma(\text{mJ/mole K}^2) \ ,$$
$$\Theta_D = \left[\frac{1944(4)}{\beta(\text{mJ/mole K}^4}\right]^{1/3} \times 10 \ , \tag{2}$$

where $N(0)$ is in units of states/eV atom and λ, the electron-phonon coupling constant, is about[2] 1.7 to 1.8 for A15 Nb_3Sn.

Unfortunately, this straightforward extrapolation does not satisfy the following entropy constraint, based on thermodynamics:

$$S_n(T_c) \equiv \int_0^{T_c} \frac{C_n}{T} dT = \int_0^{T_c} \frac{C_s}{T} dT \equiv S_s(T_c) \ . \tag{3}$$

In the work of Vieland and Wicklund,[1] the normal-state entropy at T_c, $S_n(T_c)$, was 14% larger than the superconducting state entropy at T_c, $S_s(T_c)$. This implied that the normal-state extrapolation below T_c must in fact fall below the straight-line extrapolation of the data above T_c. Vieland and Wicklund depressed T_c to approximately 15.5 K using a 5.25 T field and found no change in slope of the extended normal state C_n/T vs T^2 data. In the recent zero-field work of Junod *et al.*[3] $S_s(T_c)$ is 2024 mJ/mole K for their sample of A15 Nb_3Sn identified as K95, while $S_n(T_c)$ is approximately 2510 mJ/mole K, i.e., $S_n(T_c)$ is 24% larger than $S_s(T_c)$. In the present work, at zero field $S_n(T_c)$ using the straightforward extrapolation is 2506 mJ/mole K versus 2032 mJ/mole K for $S_s(T_c)$, or a 23% disagreement.

In order to bring $S_n(T_c)$ into agreement with $S_s(T_c)$, Vieland and Wicklund required C_n/T for $T < 15.5$ K to fall below the straight line extrapolation of the measured normal-state data. Near 15.5 K this deviation was small, and at 0 K it amounted to

38%, i.e., the intercept γ on a C_n/T vs T^2 plot was 52 mJ/mole K^2 rather than 84 mJ/mole K^2 obtained from extrapolating the C_n/T vs T^2 data from above 15.5 K using a straight line. In the work of Junod *et al.*[3] the same arbitrary extrapolation with negative curvature was imposed from T_c to 0 K, giving a γ for their sample *K*95 of 47 mJ/mole K^2 versus a γ of 96 mJ/mole K^2 obtained from a straight line extrapolation of their normal-state data.

In addition to the thermodynamic constraint mentioned, these extrapolations were required to match Θ_D as $T \to 0$ determined from very-low-temperature zero-field data where the superconducting electronic contribution is negligible. However, the extrapolation used by Vieland and Wicklund, and also by Junod *et al.*, is totally arbitrary in assuming a monotonic decrease in Θ_D below T_c. This is emphasized by the low field-data of Vieland and Wicklund and their failure to observe any change in Θ_D down to 15.5 K.

From this early specific-heat data in a relatively low magnetic field comes the implication that Θ_D might in fact remain constant to temperatures even lower than 15.5 K. This uncertainty in Θ_D vs T between 4 and 15.5 K, with $S_n(T_c)$ fixed equal to $S_s(T_c)$, makes γ and $N(0)$ similarly uncertain. Previous workers[1,3] have assumed a monotonic function of Θ_D vs T below T_c and obtained $\gamma = 52$ mJ/mole K^2 for their best, well ordered polycrystalline *A*15 Nb_3Sn.

The present work was undertaken on a high-quality polycrystalline vapor-grown sample of *A*15 Nb_3Sn [$\rho(300\ \text{K})/\rho$ (extrapolated to 0 K) = 50] in fields of 7 and 18 T in order to accurately determine γ and $N(0)$.

II. EXPERIMENTAL

The measurement of specific heat in magnetic fields as high as 18 T has never before been accomplished. The specific technique used in this work, that of low-temperature small-sample calorimetry, is more affected by the associated electrical noise and heating problems in 18 T than the standard large-sample calorimetry technique. The measurements were performed at the Francis Bitter National Magnet Laboratory (FBNML) in Cambridge, Mass. An initial attempt was made and then a calorimeter of improved design[4] was constructed and used in the successful later experiment.

The technique used for measuring the low-temperature specific heat was the time constant method,[5] where the sample and low addenda sample platform are raised a small temperature ΔT above the copper block reference temperature and then this heat is allowed to dissipate through the supporting electrical wires. The temperature response is an exponential with a time constant τ, and the total specific heat of the sample plus platform is

$$C_{\text{tot}} = K\tau \ , \tag{4}$$

where K is the thermal conductivity of the four supporting wires (two to make electrical contact with the platform thermometer and two for the electrical heater). The temperature is measured via the platform thermometer which is one arm of a Wheatstone bridge driven and detected by a lock-in amplifier. A signal averager with 12-bit resolution was used to record the decay constant data. The time constant at each temperature was calculated using a microcomputer interfaced to the signal averager. The specific heat of the sample is then

$$C_{\text{sample}} = K\tau - C_{\text{addenda}} \ . \tag{5}$$

Some of the experimental difficulties and solutions will be discussed briefly here. A more complete discussion is given in Ref. 4.

Superimposed on the dc current needed to create 18 T in the Bitter magnet used for this experiment ($\sim$35 600 A) were significant amounts of 6- and 84-Hz noise, which raised the temperature of the reference copper block from 4.3 to 5.3 K via induced current heating. More importantly, this ac noise was present on the decay constant data collected by the signal averager. Since τ was of the order of 4 to 10 sec, the 6-Hz noise was difficult to either filter or average out. The method used relied on the stabilizer core tube present in the Bitter magnets at FBNML, which was essentially a coil of water-cooled wire in the bore of the magnet driven by a set of external electronics using negative feedback to attenuate the low-frequency noise by a factor of 100.

Additional noise of even lower frequency ($\sim$0.5 Hz) was observed in the time constant data in the measurements when the sample platform thermometer was unencapsulated Ge, used in the zero-field small sample calorimeter.[6,7] By changing the vibrational damping of the calorimeter and Dewar assembly, the amplitude and frequency of this low-frequency noise could be significantly altered.[4] It was postulated that the noise was the result of motion of the highly magnetoresistive sample thermometer [$\rho(18\ \text{T})/\rho\ (0\ \text{T}) \simeq 5.6$ at 4.2 K] in a nonhomogeneous magnetic field. A sample platform using an unencapsulated carbon-glass thermometer was constructed[4] and tested and used during the later experiment. The magnetoresistance of the carbon-glass thermometer was less than 2% of that of the Ge thermometer used earlier. The low-frequency noise on the time constant data was not observable using this new design sample platform.

The 57-mg Nb_3Sn polycrystalline sample was grown over a period of 4 months by closed-tube iodine vapor transport. X-ray diffraction on another part of the deposit showed only sharp *A*15 lines with a room-temperature lattice parameter of 5.290 Å. Resistivity measurements on the same sample as used in the present experiment confirm that it undergoes a martensitic transformation at 51 K.

Inductive T_c measurements and de Haas–van Alphen measurements[8] on a different crystal from the same growth tube gave an onset T_c of about 17.85 K and an upper critical field H_{c2} at 1.5 K of 21.4 T. The bulk onset of superconductivity in the present experiment is 17.9 K. Two encapsulated Ge thermometers were used in zero field in the calorimeter to determine temperature. An encapsulated capacitance thermometer kept continuously at liquid-helium temperatures for all 5 days of the experiment and calibrated in zero field was used as a transfer standard in high field in order to calibrate the platform carbon-glass thermometer. Temperatures were known accurately to better than ± 0.1 K in the full magnetic field.

III. SPECIFIC-HEAT DATA AND DISCUSSION

The specific heat of $A15$ Nb_3Sn in 0, 7.0, and 18.0 T is shown in Fig. 1. The expanded lower-temperature zero-field data are shown in Fig. 2. The zero-field data in Fig. 1 were measured on both the sample platform using Ge as the thermometer and on the sample platform using carbon glass as the thermometer. The agreement between the two sets of data is $\pm 1\%$. The accuracy of the zero-field data is $\pm 3\%$.[6,7] The accuracy of the high-field data is approximately the same, since the ± 0.1 K temperature uncertainty in high field makes less than a $\pm 1\%$ error in the data. The two high-field data points at $T^2 = 315$ and 390 K^2 are actually at 19.0 T and were taken in the earlier work. These two points agree within error limits with the zero-field normal-state data. This, in addition to the agreement between the 7- and 18-T data in the region of overlap between $T^2 = 170$ and 225 K^2, indicates that, as expected, these fields are too low to affect the normal-state specific heat, by changing the density of states versus energy distribution. This is not surprising, since 18 T times a Bohr magneton is only about a tenth of a millirydberg in energy.

As may be seen in Fig. 1, the field data show that the slope β [Eq. (1)] of the C_n/T vs T^2 plot remains constant down to 11 K and then, by 10.2 K, has changed dramatically over to the low temperature $\beta(T < 4\text{ K})$ of the C_s data in Fig. 2, where the superconducting state data, C_s, is given by

$$C_s = C_s^{\text{electronic}} + C^{\text{lattice}} ,$$
$$C_s^{\text{electronic}} = a e^{-\Delta/kT} \rightarrow 0 , \quad (6)$$

for low T. In the above equation, C^{lattice} is βT^3 from

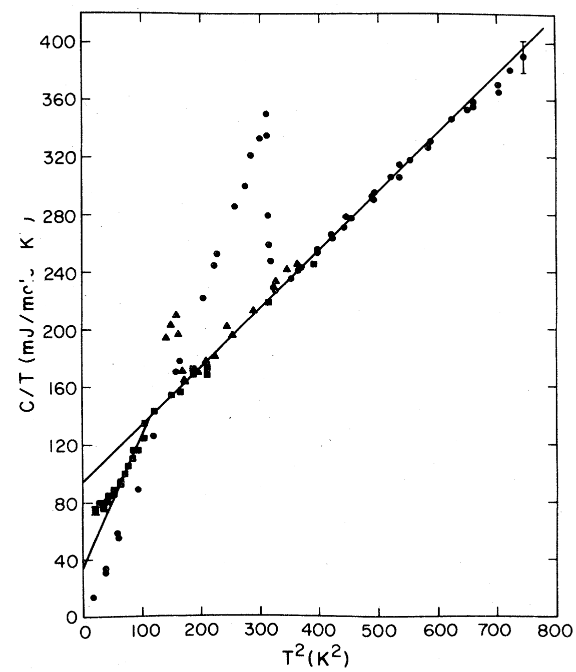

FIG. 1. Low-temperature specific heat of $A15$ Nb_3Sn in 0 (circles), 7 (triangles), and 18 T (squares) field. Note the abrupt change in slope of the high-field data at $T^2 \simeq 120\ K^2$. Also note the superconducting transition in 7 T ($T^2 \simeq 170\ K^2$) and in 18 T ($T^2 \simeq 40\ K^2$). The negative deviation of the highest-temperature data from the straight line is accounted for in the simple Debye model (Ref. 9) of the lattice specific heat. At higher temperatures, the specific-heat data of Knapp *et al.* (Ref. 10) for Nb_3Sn indicate that the Debye model becomes invalid, as discussed in Ref. 11.

Eq. (1). As discussed in the Introduction, the important test of this normal-state data in a field is the calculation of $S_n(T_c)$ in Eq. (3). Using the straight-line extrapolation shown in Fig. 1 for C_n below the superconducting anomaly at $T^2 \simeq 40\ K^2$ in 18 T, and the measured data in 18 T above this anomaly, $S_n(T_c)$ is calculated to be 2038 mJ/mole K. The agreement of this *measured* $S_n(T_c)$ in an 18 T field with the measured $S_s(T_c)$ (2032 mJ/mole K) in zero field is strong confirmation of the accuracy of the measured field data.

Before turning to the implications of these data for γ and $N(0)$, the anomalous change in the lattice specific heat, as well as the transitions in 7 and 18 T will be discussed.

This behavior of β, or Θ_D, versus temperature has never been observed before in any $A15$ material. However, the abruptness of the lattice softening may be a function of the applied field. This is because there is no similar abrupt change at 11 K in the slope of the C_s/T vs T^2 data also plotted in Fig. 1, even though C^{lattice} is more than 85% of C_s at 11 K. The common assumption for the lattice specific heat is

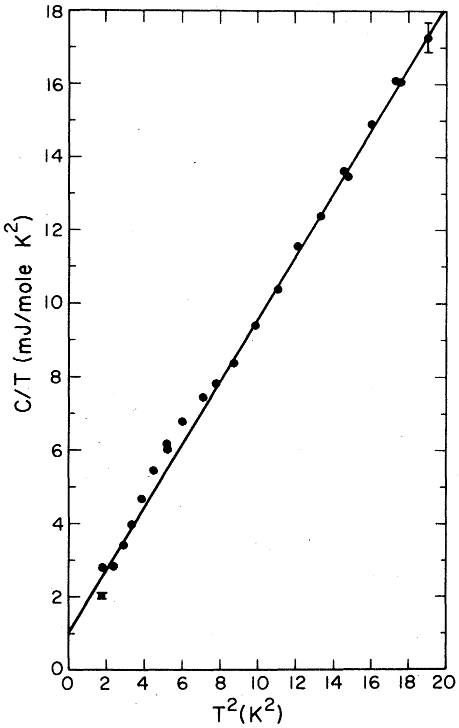

FIG. 2. Low-temperature specific heat of $A15$ Nb_3Sn in zero field for $T < 4.5$ K. The superconducting electronic specific heat is negligible [Eq. (6)] and $\Theta_D(0\text{ K})$ is found to be 208 K from these data. The hump centered at $T^2 = 6\text{ K}^2$ and the finite intercept are qualitatively similar to the results of Ref. 3 and are thought not to be due to a second phase, but perhaps to very-low-frequency phonons (Ref. 3) arising from a Kohn anomaly. Two superconducting energy gaps have also been suggested (Ref. 12) as the source of the finite intercept.

that it is unaffected by the superconducting state. However, the Debye temperature, Θ_D, at low temperatures is a measure only of a very limited part of the total phonon spectrum $F(\omega)$ — at 11 K perhaps only of the first 5 or 6 meV in energy, where $F(\omega)$ extends[13] to 30 meV. It has been shown by Axe and Shirane[14] using inelastic neutron scattering in Nb_3Sn that in fact the superconducting state has a pronounced effect on certain low-frequency phonons of approximately equal energy to those sampled by the low-temperature specific heat. The onset of superconductivity radically decreases the linewidth of some low-frequency phonons, since phonons with energies less than the superconducting gap energy cannot decay by exciting electron-hole quasiparticle pairs. A similar lattice-superconductivity interaction is observed in V_3Si, where the lattice stiffness parameter $(c_{11} - c_{12})/2$ of nontransforming samples normally softens as temperature is lowered until T_c is reached, when the softening is halted by the onset of superconductivity. The application of a magnetic field to lower T_c causes the softening to continue.[15]

Explanation of this anomalous behavior at 11 K in $A15$ Nb_3Sn in 18 T would be aided by other experiments in a field, both electrical and elastic measurments. It is interesting to note that the Θ_D derived at low temperatures in the superconducting state from the data in Fig. 2 is 208 K, nearly identical to the low temperature Θ_D of 204 K derived from the field data below 11 K in Fig. 1.

Another obvious feature of the field data in Fig. 1 is the presence of two superconducting transitions, one at 13.0 K in 7 T and a quite broad transition in 18 T with an onset T_c of approximately 6 to 6.5 K. The lower field transition is still in the linear region where the Werthamer formula[16] may be used to calculate H_{c2}, the upper critical field at 0 K. Using a coefficient of 0.725 from the work of Orlando *et al.*[17] on $H_{c2}(T)$ of clean Nb_3Sn

$$H_{c2} = 0.725 \frac{dH_{c2}}{dT}\bigg|_{T=T_c} T_c \qquad (7)$$

one obtains $H_{c2} = 18.5$ T. Using the de Haas—van Alphen result[8] for $H_{c2}(T = 1.5\text{ K})$ of 21.4 T, one can normalize the curve of $H_{c2}(T)$ vs T data of Ref. 17 for their Nb_3Sn sample for the Nb_3Sn used in the present work and obtain a T_c in 18 T of about 5.7 K, versus the observed 6 to 6.5 K. These disagreements are signs that current models for H_{c2} are not totally accurate. Work is continuing to determine H_{c2} experimentally for the sample measured in the present work.

Equally as interesting as the anomalous behavior of Θ_D are its implications for γ and $N(0)$. As seen in Fig. 1, the correct γ for $A15$ Nb_3Sn is not 47 mJ/mole K^2, as measured by Junod *et al.* on their sample $K95$, or 52 mJ/mole K^2, as measured by Junod *et al.* on their $J212$ sample and by Vieland and Wicklund, but is instead 35 ± 3 mJ/mole K^2. It should be stressed that the sample of $A15$ Nb_3Sn used in the present work is of high quality, made up of several large single crystals, with a high residual-resistivity ratio, a sharp superconducting transition, a lattice parameter which corresponds to stoichiometric Nb_3Sn, and is in general fully intercomparable with the best quality samples measured by previous workers. The agreement betwen the zero-field data for the sample used in the present work [$S_s(T_c) = 2032$ mJ/mole K, $\gamma^{\text{extrapolated}} = 96$ mJ/mole K^2, $\Theta_D(T > T_c) = 270$ K] with that for the $K95$ sample of Junod *et al.* [$S_s(T_c) = 2024$ mJ/mole K, $\gamma^{\text{extrapolated}} \simeq 96$ mJ/mole K^2, $\Theta_D(T > T_c) \simeq 273$ K] further emphasizes this. The sole reason why previous workers arrived at $\gamma \simeq 52$ mJ/mole K^2 instead of 35 ± 3 mJ/mole K^2 was the incorrect assumption that Θ_D decreased smoothly from T_c down to lower temperatures.

The $N(0)$ calculated from $\gamma = 52$ mJ/mole K^2 and Eq. (2), using[2] $\lambda \simeq 1.75$, is 2.01 states/eV atom. The

$N(0)$ obtained from the correct normal-state extrapolation measured in the present work ($\gamma = 35 \pm 3$ mJ/mole K^2) and using $\lambda \simeq 1.75$ is 1.35 ± 0.1 states/eV atom. The result for the $N(0)$ of $A15$ Nb_3Sn calculated in the recent definitive band structure work of Klein *et al.*[18] is $N(0) = 1.46$ states/eV atom. This had seemed in unusual disagreement with experiment, especially considering the relatively good agreement between their work and experiment for Nb_3Ge [0.98 versus 1.08 ± 0.1 (Ref. 19)], Nb_3Si [0.64 versus 0.94 ± 0.2 (Ref. 20)], Nb_3Ga [1.76 versus 1.7 ± 0.25 (Ref. 21)], and Nb_3Al [1.83 versus 1.4 ± 0.2 (Ref. 22)]. The result of the present experiment for the $N(0)$ of $A15$ Nb_3Sn is in much better agreement with the calculations of Klein *et al.* and more in line with the generally good agreement observed for the other $A15$ Nb_3X $N(0)$ values calculated in that work with experiment.

The normalized zero-field specific-heat discontinuity, $\Delta C/\gamma T_c$, using this low γ is high, 3.5 ± 0.3, compared to the largest value previously obtained for another $A15$ Nb_3X compound, 3.2 for Nb_3Al.[22] Also, this value of 3.2 for Nb_3Al was extrapolated for an ideally sharp transition and was not actually measured. Measured values reviewed in Ref. 23 for $\Delta C/\gamma T_c$ are all under 3.0, even for the non-$A15$, strong coupled superconductors like Pb (2.6) and $NbN_{0.91}$ (2.9). However, the fact that $S_n(T_c)$ agrees with $S_s(T_c)$ using the high-field measured data, the agreement in the low temperature Θ_D in a field and in zero field (204 versus 208 K), and the agreement of the $N(0)$ calculated using $\gamma = 35$ mJ/mole K^2 with the band structure calculations of Klein *et al.* substantiate the high-field data and the high $\Delta C/\gamma T_c$ value.

IV. CONCLUSIONS

The measurement in 18 T of the specific heat of $A15$ Nb_3Sn has revealed that Θ_D remains constant down to 11 K, where it rapidly softens from 270 to 204 K. These measured low-temperature, high-field normal-state specific-heat data give $\gamma = 35 \pm 3$ mJ/mole K^2, in marked contrast to the γ derived by previous workers, who assumed that Θ_D changed gradually below T_c and implied $\gamma = 52$ mJ/mole K^2. The high-field measurements in the present work above 17.8 K imply that γ is not affected by the field. This new, lower experimental value of $\gamma = 35$ mJ/mole K^2 gives much better agreement for $N(0)$ derived from specific heat (1.35 ± 0.1 states/eV atom) with that calculated by Klein *et al.* (1.46 states/eV atom). Further measurements need to be done in high field on $A15$ Nb_3Sn in order to probe the observed rapid change in Θ_D at 11 K. It would be interesting to measure the low-temperature specific heat of $A15$ V_3Si in 18 T in order to resolve the recent dispute in the literature[24,25] between two different experimental groups on the correct normal-state extrapolation of C_n/T below T_c. V_3Si also has a large increase in Θ_D between 0 K and $T > T_c$ and the assumption that Θ_D is a smooth function of T in this range is likely also to be incorrect.

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*Also at Energy Science Laboratory, San Diego, Calif. 92121.

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