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Electrical resistivity of indium: Deviation from linearity at high temperatures

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An accurate measurement of the electrical resistivity ρ of polycrystalline indium from 30 to 425 K is reported. Indium has an exceptionally large value of $d(\rho/T)/dT$ at high temperatures. This feature is explained by the influence of the thermal expansion, in particular, changes in the electronic structure connected with the anisotropic expansion coefficient.

I. INTRODUCTION

This paper reports the measurement and analysis of the electrical resistivity ρ of polycrystalline indium from 30 to 425 K. Several previous measurements of ρ up to room temperature can be found in the literature,[1-5] in one case[1] with an alleged discontinuity in $d\rho/dT$ which is shown here to be nonexistent. Barisoni *et al.*[6] have reported resistivity data for polycrystalline indium up to the ice point and for single-crystal specimens up to 350 K. The ρ data of Powell *et al*,[7] extending up to 420 K, are of rather limited accuracy.

In simple models, e.g., of the Bloch-Grüneisen type, $d(\rho/T)/dT$ is zero at high temperatures. Indium, however, is the element with the largest value of $d(\rho/T)/dT$ at high temperatures known to us. This is of particular interest in comparison with the still unsettled question of resistivity saturation (small or negative $d\rho/dT$) in highly resistive metals and alloys.[8]

II EXPERIMENTAL DETAILS AND RESULTS

Wires were prepared from pure indium (Materials Research Corp., Marz Grade) with nominal purity 99.999 wt.% by cold-rolling and drawing. The wires were dipped into liquid nitrogen before each pass through a die. In this way a diameter of 0.8 mm could be reached without problems with the extraordinary softness of indium. No annealing was necessary since the resistance ratio $R(273\text{ K})/R(4.2\text{ K})$ was as high as 6000 after this treatment.

Resistances were measured in a four-probe sample holder where contact with the sample was obtained via spring-loaded knife edges. A standard dc potentiometer, corrected for small deviations from linearity as described previously,[9] was used to measure the voltage drop over the sample. Regulated temperatures, obtained either in a cryostat or in an oil bath, were read from a platinum resistance thermometer with the relative precision of 2 mK.

The data presented here were gathered from three wire samples prepared from the same lot and measured in separate experiments covering different temperature intervals (30-120, 110-295, 290-425 K). The geometric factor of the 110-295 K sample was determined to an estimated accuracy of 0.2% by measuring its mass-to-length ratio and the distance between voltage-drop edge marks, and using reported density data.[10] For the mass-to-length ratio, weighings and length measurements (under a traveling microscope, with the sample resting in a glass capillary) were repeated a few times for averaging, each time changing the length of the wire by cutting off a small portion. For the two other samples the geometric factors were adjusted to yield a smooth overall data set of resistivity versus temperature. We checked particularly that joining the resistivities smoothly also resulted in a smooth resistivity derivative. It is concluded that no kink or discontinuity in the temperature derivative of the resistivity arose from this data handling.

The results are given in Table I. ρ^* is proportional to the measured resistance and scaled so as to coincide at low temperatures with the resistivity at constant (atmospheric) pressure ρ_p. These quantities are related by

$$\rho_p(T)=\rho^*(T)\left(1+\int_0^T\alpha(T')dT'\right), \quad (1)$$

where the thermal expansion $\alpha(T)$ was obtained by fitting Eq. (13) below to directionally averaged data for the thermal expansion,[11] with a Debye model expression for C_{ph} and a linear term for C_e. Table I also lists ρ_v, the calculated resistivity at constant volume, obtained as

$$\rho_v(T)=\rho^*(T)(1+\alpha_R p), \quad (2)$$

TABLE I. Resistance and resistivity of indium from 30 to 425 K. ρ^* is resistance, scaled so as to coincide at low temperature with ρ_p, the resistivity at atmospheric pressure. ρ_v is the calculated resistivity at constant volume [Eq. (2)].

Temperature (K)	ρ^* (Arbitrary unit)	ρ_p ($\mu\Omega$ cm)	ρ_v ($\mu\Omega$ cm)
4.2150	0.001427	0.001427	0.001427
29.9276	0.3869	0.3870	0.3862
39.9546	0.6511	0.6513	0.6484
49.9659	0.9263	0.9267	0.9199
54.9481	1.0647	1.0653	1.0558
59.9355	1.2035	1.2043	1.1915
64.9355	1.3427	1.3438	1.3271
69.9312	1.4818	1.4831	1.4621
74.9223	1.6209	1.6225	1.5966
79.9137	1.7597	1.7616	1.7302
84.9212	1.8998	1.9021	1.8646
89.9347	2.0402	2.0429	1.9987
94.9308	2.1809	2.1841	2.1326
99.9423	2.3227	2.3265	2.2670
104.9279	2.4645	2.4688	2.4008
110.2928	2.6183	2.6232	2.5454
115.2908	2.7625	2.7680	2.6803
120.2309	2.9057	2.9119	2.8138
125.2487	3.0521	3.0590	2.9498
130.1530	3.1962	3.2039	3.0831
135.3073	3.3487	3.3572	3.2236
140.2883	3.4972	3.5065	3.3598
145.2205	3.6454	3.6556	3.4953
150.1748	3.7957	3.8069	3.6322
155.1325	3.9474	3.9596	3.7698
160.0859	4.1002	4.1135	3.9078
165.0786	4.2555	4.2698	4.0475
170.0433	4.4115	4.4269	4.1873
175.0542	4.5701	4.5868	4.3290
180.0070	4.7283	4.7462	4.4697
185.0119	4.8898	4.9091	4.6128
190.0010	5.0524	5.0730	4.7563
195.0030	5.2168	5.2389	4.9008
200.0041	5.3826	5.4062	5.0461
205.0067	5.5506	5.5757	5.1927
210.0091	5.7198	5.7465	5.3397
215.0169	5.8907	5.9191	5.4877
220.0239	6.0629	6.0930	5.6361
225.0396	6.2375	6.2694	5.7862
230.1204	6.4156	6.4494	5.9386
235.0527	6.5898	6.6255	6.0871
240.0530	6.7678	6.8055	6.2382
245.0886	6.9486	6.9884	6.3911
250.0978	7.1302	7.1721	6.5441
255.1202	7.3138	7.3579	6.6982
260.1335	7.4988	7.5452	6.8529
265.1535	7.6853	7.7340	7.0082
270.1733	7.8737	7.9249	7.1645
275.1799	8.0630	8.1167	7.3209
280.2053	8.2544	8.3106	7.4784
285.2383	8.4473	8.5062	7.6365
290.2707	8.6413	8.7030	7.7949

TABLE I. (*continued*)

Temperature (K)	ρ^* (Arbitrary unit)	ρ_p ($\mu\Omega$ cm)	ρ_v ($\mu\Omega$ cm)
295.3124	8.8368	8.9013	7.9539
301.9670	9.0974	9.1658	8.1648
311.8330	9.4874	9.5617	8.4783
321.4020	9.8713	9.9517	8.7846
331.0560	10.2614	10.3482	9.0933
341.2850	10.6804	10.7744	9.4223
351.5090	11.1063	11.2078	9.7542
361.8780	11.5436	11.6532	10.0921
361.9360	11.5459	11.6555	10.0939
372.4650	11.9953	12.1134	10.4382
384.8970	12.5346	12.6634	10.8478
392.9160	12.8844	13.0203	11.1111
401.5600	13.2667	13.4107	11.3972
410.9620	13.6890	13.8419	11.7110
411.0250	13.6928	13.8459	11.7140
420.7370	14.1372	14.3001	12.0423
425.8400	14.3764	14.5446	12.2183

where p is the pressure that restores the sample to its zero-temperature volume and α_R is the pressure coefficient of resistance. Room-temperature data for the compressibility (which enters p) and α_R were used,[12] neglecting the temperature dependence of these quantities. Note that $\rho_v(T)$ from (2) is not to be compared with, e.g., a theoretical calculation with unchanged lattice parameters, since the ratio of the lattice parameters is temperature dependent even under constant-volume conditions.

For convenient reference, Table II summarizes reported values of the ice-point resistivity of polycrystalline and single-crystal indium. The resistivity of indium is slightly anisotropic with $\rho_\perp/\rho_\parallel = 1.034$ at 273 K.[6] Furthermore the ratio $\rho_\perp/\rho_\parallel$ has a weak temperature dependence, hence the reduced resistance $\rho^*(T)/\rho^*(273\text{ K})$ of polycrystalline indium will be affected by any anisotropy of the grain orientation in the polycrystal. On the temperature interval where comparison is possible (80-300 K) our data for $\rho^*(T)/\rho^*(273\text{ K})$ agree closely with the average for isotropically polycrystalline indium given by Ref. 6. From this we conclude that the anisotropy in grain orientation that might result from cold-working is quite small and may be neglected as far as the electrical resistivity is concerned.

TABLE II. Ice-point resistivity of indium. Samples are polycrystalline unless otherwise indicated.

ρ at 273.15 K ($\mu\Omega$ cm)
8.2[a]
8.0[b]
7.98 ±0.05[c]
8.13 ±0.05[d]
7.85 ±0.05[e]
8.04 ±0.02[f]

[a] W. Meissner, H. Franz, and H. Westerhoff, Ann. Phys. 13, 555 (1932).

[b] Reference 7.

[c] Interpolated from data in Ref. 6.

[d] Interpolated from data in Ref. 6, $\rho_\perp$ of single crystal.

[e] Interpolated from data in Ref. 6, $\rho_\parallel$ of single crystal.

[f] This work.

It is customary in the literature on accurate resistivity measurements to approximate data by polynomials. Although seldom physically transparent, such a polynomial provides a convenient summary of the data. Indeed, in the present case the second-degree polynomial

$$\rho^* = -0.1612 + 0.022075T + 2.8347\times 10^{-5}T^2 \qquad (3)$$

fits our data over the temperature range 100-425 K with a rms deviation of only 0.08%. Terms in T^3 and T^4 do not significantly improve the fit, thus the first correction to Eq. (3) is of the order T^5.

III. THEORY

A. The electrical resistivity

The electron-phonon interaction as it enters expressions for the electrical resistivity has in the past usually been given as a function of the momentum transfer in a scattering event.[13]

In the present case we are interested in the temperature dependence of $\rho(T)$ and then it is more convenient to represent the electron-phonon interaction with an effective coupling function $\alpha_{\rm tr}^2 F(\omega)$, where $\hbar\omega$ is the phonon energy involved in the scattering. The lowest-order variational solution of the Boltzmann equation yields the result[14,15]

$$\rho(T)=\frac{4\pi m}{ne^2k_BT}\int_0^{\omega_{\max}}\frac{\hbar\omega\alpha_{\rm tr}^2F(\omega)}{(e^{\hbar\omega/k_BT}-1)(1-e^{-\hbar\omega/k_BT})}d\omega\,. \tag{4}$$

We have, for simplicity, written Eq. (4) for a free-electron-like system with electron number density n and free-electron mass m. In the high-temperature limit,

$$\rho(T)=\frac{2\pi mk_BT}{ne^2\hbar}\int_0^{\omega_{\max}}\frac{2\alpha_{\rm tr}^2F(\omega)}{\omega}d\omega\equiv\frac{2\pi mk_BT}{ne^2\hbar}\lambda_{\rm tr}\,, \tag{5}$$

where the last equality defines the electron-phonon transport coupling parameter $\lambda_{\rm tr}$. This parameter is closely related to the well known electron-phonon mass enhancement parameter λ that appears, e.g., in the theory of superconductivity.[15] The only difference is that λ is defined by an integral as in Eq. (5) with $\alpha_{\rm tr}^2F(\omega)$ replaced by the Eliashberg coupling function $\alpha^2F(\omega)$. The function $\alpha_{\rm tr}^2F(\omega)$ differs from $\alpha^2F(\omega)$ essentially by the average of a geometrical factor $1-\cos\theta_{\vec{k}\vec{k}'}$, where $\theta_{\vec{k}\vec{k}'}$ is the scattering angle between two wave vectors $\vec{k}$ and $\vec{k}'$ on the Fermi surface. Following McMillan,[16] we can write

$$\lambda=\frac{N(E_F)\langle I^2\rangle}{M\langle\omega^2\rangle}\equiv\frac{\eta}{M\langle\omega^2\rangle}\,, \tag{6}$$

where $N(E_F)$ is the electron density of states (per atom, spin) at the Fermi energy, $\langle I^2\rangle$ is a Fermi-surface average of the electron-phonon matrix element $I(\vec{k},\vec{k}')$, M is the ion mass, and $\langle\omega^2\rangle$ is an average of the phonon frequencies with weight function $\alpha^2F(\omega)$. A relation similar to (4), but valid also when the metal is not free-electron-like, has been given by Chakraborty *et al.*[17] One has

$$\rho=\frac{1}{\sigma}=\frac{2e^2}{3V}\sum_{\vec{k}}\vec{v}_{\vec{k}}^{\,2}\,\delta(\epsilon_{\vec{k}}-E_F)\tau(\vec{k})\,, \tag{7}$$

where V is the crystal volume, $\epsilon_{\vec{k}}$ electron-band energies, $\vec{v}_{\vec{k}}$ the electron velocity, and $\tau(\vec{k})$ an electron scattering time for a state $\vec{k}$. The sum is over all $\vec{k}$ on the Fermi surface, including different bands if appropriate. Chakraborty *et al.* approximate (7) by the factorization

$$\sigma=\frac{\omega_p^2}{4\pi}\langle\tau\rangle\,, \tag{8}$$

with the Drude plasma frequency

$$\omega_p^2=\frac{8\pi e^2N(E_F)}{3(V/N)}\langle v_{\vec{k}}^2\rangle\equiv\frac{8\pi e^2}{3V}\sum_{\vec{k}}\vec{v}_{\vec{k}}^{\,2}\,\delta(\epsilon_{\vec{k}}-E_F) \tag{9}$$

and the high-temperature relation

$$\langle\tau\rangle=\frac{\hbar}{2\pi k_BT\lambda_{\rm tr}}\,. \tag{10}$$

It is easy to see that we recover (5) in the free-electron limit.

B. The volume dependence of $\rho(T)$

The electrical resistivity gets an implicit temperature dependence through the thermal expansion. From (8) and (10) one obtains the high-temperature relation

$$\frac{d\ln\rho}{d\ln V}=-2\frac{d\ln\omega_p}{d\ln V}+\frac{d\ln\lambda_{\rm tr}}{d\ln V}\,. \tag{11}$$

Usually, the last term gives the dominating contribution. If we approximate $\lambda_{\rm tr}$ by λ and use (6), we can write

$$\frac{d\ln\lambda_{\rm tr}}{d\ln V}=\frac{d\ln\eta}{d\ln V}-\frac{d\ln\langle\omega^2\rangle}{d\ln V}\,. \tag{12}$$

It is often a reasonable approximation[13] to keep only the last term in (12) and replace it by $+2\gamma_{G,\rm ph}$, where $\gamma_{G,\rm ph}$ is the usual Grüneisen parameter.

C. The thermal expansion of metals

A standard expression for the coefficient of linear thermal expansion of metals is[18]

$$\alpha=\frac{1}{3B}(\gamma_{G,\rm ph}C_{\rm ph}+\gamma_{G,e}C_e)\,. \tag{13}$$

Here B is the bulk modulus, $C_{\rm ph}$ is the vibrational part and C_e the electronic part of the heat capacity, $\gamma_{G,\rm ph}$ is the usual Grüneisen parameter, and $\gamma_{G,e}$ a related parameter for the electrons. In a Debye model (Debye temperature Θ_D) one has

$$\gamma_{G,\rm ph}=-\frac{d\ln\Theta_D}{d\ln V}\,. \tag{14}$$

The low-temperature form of $\gamma_{G,e}$ is

$$\gamma_{G,e}=1+\frac{d\ln N(E_F)}{d\ln V}+\frac{d\ln(1+\lambda)}{d\ln V}\,, \tag{15}$$

while at high temperatures ($T\gtrsim\Theta_D/3$) the last term in (15) involving λ is absent.[15] At high temperatures, $C_{\rm ph}$, and hence the vibrational contribution to α in (13), is independent of temperature. The electronic part in (13) is linear in T at all temperatures. Usually the vibrational term dominates except at very low temperatures. The thermodynamic relations just given have been written for a lattice with isotropic thermal expansion. They may be generalized to a nonuniform change of the lattice parameters in, e.g., a tetragonal structure such as that of indium. A general expression for the components α_{ij} of the expansion tensor is[19]

$$\alpha_{ij} = -\frac{1}{V}\frac{\partial^2 F}{\partial\sigma_{ij}\partial T}, \tag{16}$$

where σ_{ij} is the stress tensor. Just as for the isotropic case, the free energy F has a vibrational part which yields a contribution to α_{ij} that is approximately independent of the temperature when $T \gtrsim \Theta_D$, and an electronic part which increases as T^2 and hence yields a contribution to α_{ij} that is linear in T.

IV. ANALYSIS OF THE RESISTIVITY DATA FOR INDIUM

A. Absence of discontinuity in the temperature derivative

To investigate whether there is a kink in the resistivity around 210 K as claimed in Ref. 1 we analyzed the data in the region 160-260 K. Two different fits were attempted: (i) a parabola with three adjustable parameters and (ii) a piecewise straight line with a change of slope anywhere in the temperature interval (four adjustable parameters). The best fit of type (ii) was obtained by letting a change of slope occur at 208 K. This however merely reflects the fact that the broken line was fitted to data points symmetrically distributed around 210 K. The sum of the squared deviations was larger by almost 3 orders of magnitude for the broken line (ii) than for the parabola (i). We conclude that there is no kink in the electrical resistivity of indium at 210 K. Further details illustrating that an unreal effect can easily occur in this type of analysis were given previously.[20]

B. Negligible vacancy contribution to the resistivity

The resistivity contribution from thermally excited vacancies is likely to be very small in indium. Positron-trapping experiments[21] yield a vacancy-formation enthalpy of 0.48 ± 03 eV. The specific resistivity of vacancies in indium is not known but, in comparison with other metals (Ref. 22), 4 $\mu\Omega$ cm/at.% may be a reasonable value. This would yield a resistivity increase of only 60 ppm at the melting point (429 K). Our efforts to separate out a vacancy contribution did not give a stable result for the vacancy-formation enthalpy, thus confirming the small magnitude of such a contribution.

C. Nonlinear resistivity at high temperatures

Figure 1 shows our experimental data for indium, plotted as $\rho(T)/T$ vs T. Equation (5) implies that this ratio approaches a constant with increasing temperature, whereas the experimental data for $\rho(T)/T$ show an additional increase Δ which is linear in T. This feature is observed in most simple metals, but is larger in indium than in any other simple metal. We shall see that about half of Δ is caused by shifts in the phonon frequencies related to the thermal volume expansion. The remaining part is mainly caused by an unusual volume dependence of the electronic band structure, which is related to a very anisotropic thermal expansion of the tetragonal unit cell of indium.

We need the shifts of the phonon frequencies on thermal expansion as they are weighted in the electrical resistivity, i.e., $d\langle\omega^2\rangle/dV$ with $\langle\omega^2\rangle$ calculated from $\alpha^2_{tr}F(\omega)$. This quantity is not easily obtained and we must approximate it by some other measure of the frequency shifts. A good alternative is the shift derived from the lattice part of the entropy.[15] The measured entropy,[23] corrected for the electronic contribution, is set equal to the Debye model for the entropy. This gives a temperature-dependent "entropy Debye temperature" $\Theta_S(T)$. For strictly harmonic lattice vibrations, $\Theta_S(T)$ typically tends to a constant above say $T = \Theta_D/2$. In a real solid, anharmonicity gives rise to shifts in the phonon frequencies which cause $\Theta_S(T)$ to decrease approximately linearly with T when $T \gtrsim \Theta_D/2$. The $\Theta_S(T)$ thus obtained is shown in Fig. 2. We have used this $\Theta_S(T)$ in a Bloch-Grüneisen expression for $\rho(T)$ to estimate the effect of phonon frequency shifts on the temperature dependence of the electrical resistivity of indium (dashed curve in Fig. 1). Although the Bloch-Grüneisen model is too crude for an analysis of the absolute magnitude of $\rho(T)$, it is adequate for the present discussion (the prefactor of the Bloch-Grüneisen expression was adjusted to make it fit the data below 60 K).

For most simple metals, a correction of the type just described accounts satisfactorily for Δ but in indium it only explains somewhat less than half of the measured Δ. We therefore turn to the change in ω_p^2 and η with the thermal expansion [cf. Eqs. (11) and (12)]. In a free-electron model, these terms do not vary strongly with the volume. However, indium has an unusually large and anisotropic volume dependence of the electronic band structure. This fact is most clearly seen in the experimental data for the

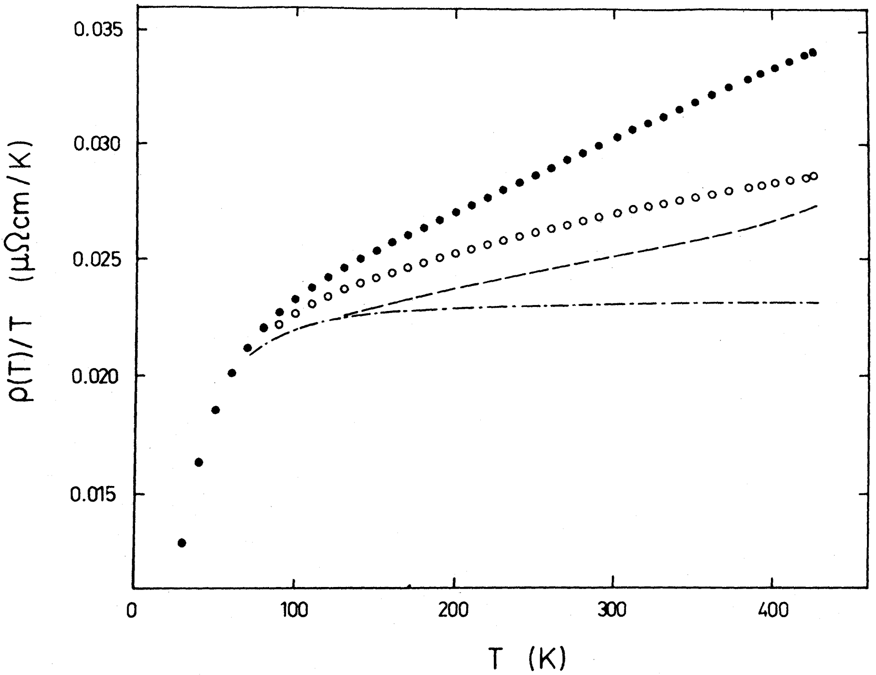

FIG. 1. Temperature dependence of $\rho(T)/T$. ●, $\rho_p(T)/T$, experimental data, at constant pressure. o, $\rho_v(T)/T$, calculated, using compressibility and pressure coefficient of resistance from Ref. 12. -•-•-•, $\rho(T)/T$, of Bloch-Grüneisen model. ——, $\rho(T)/T$, of Bloch-Grüneisen model, corrected for phonon softening.

thermal expansion. The measured coefficients[11] $\alpha_{\parallel}$ (parallel to the tetrad axis) and $\alpha_{\perp}$ can be resolved into one temperature-independent component (of vibrational origin) which is approximately the same for $\alpha_{\parallel}$ and $\alpha_{\perp}$, and one component which is linear in T (of electronic origin), increasing with T for $\alpha_{\perp}$ and decreasing for $\alpha_{\parallel}$. At room temperature, the electronic terms are approximately equal in magnitude to the vibrational terms. This fact shows that the electronic band structure changes drastically as the lattice undergoes an anisotropic thermal expansion. Lacking an accurate band-structure calculation for varying unit-cell dimensions, we refrain from giving a detailed discussion of the resistivity in terms of changes in the band structure. A similar strong variation of the band structure is also reflected in other properties, such as dT_c/dp of indium alloys which varies anomalously with the concentration.[24]

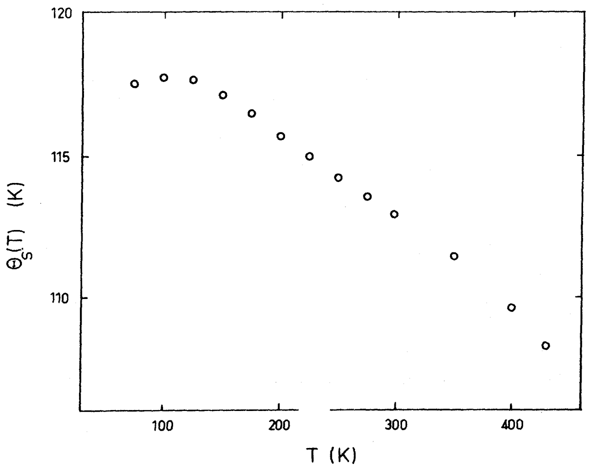

FIG. 2. "Entropy Debye temperature" Θ_S as a function of temperature.

It is interesting to compare the effect of thermal expansion on ρ (T) with $d\rho(T)/dp$ measured under hydrostatic pressure. In the latter case, the changes of the lattice unit-cell dimensions are more isotropic[25] than in the case of thermal expansion. One therefore expects that $d\rho(T)/dp$ from high-pressure experiments is reasonably accounted for by a term $2\gamma_{G,\mathrm{ph}}$ in Eq. (12). Indeed, the measured values are (Ref. 12) $d\ln\rho/d\ln V = 5.9$ and (Ref. 11) $2\gamma_{G,ph} = 4.6$. An analogous conclusion is reached if one considers the change of the superconducting transition temperature T_c under hydrostatic pressure and makes use of well known relations between T_c and λ, as was done in an analysis of $d\rho(T)/dp$ for aluminum.[26] Finally, from Fig. 1 we also note that the change in the Bloch-Grüneisen $\rho(T)/T$, when the correction for phonon softening is included, is approximately equal in magnitude to the difference between $\rho_p(T)/T$ and $\rho_v(T)/T$. This is not surprising since by prohibiting expansion (i.e., ρ_v) one has largely suppressed the frequency shifts.

V. DISCUSSION

Recently there has been much interest in the deviation of $\rho(T)/T$ from a constant at high temperatures. In particular, most highly resistive metals show a " saturation effect" in that $d\rho(T)/dT$ tends to zero or is even negative. A large number of explanations for this saturation have been proposed,[8] but no definite answer has been reached. Some of these models assume a breakdown of the Boltzmann equation as the mean free path of the electrons becomes very short. Others assume that the Boltzmann equation is adequate and that the saturation can be explained, e.g., by some features in the electronic band structure near the Fermi level. It has been argued against this latter suggestion that one would then also expect many metals to show strongly increasing $\rho(T)/T$ with T, and not only a decrease as in the saturation effect. Indium now poses an interesting problem since it is a metal with a very large $d(\rho/T)/dT$ at high temperatures. We have found that this large value may be explained as arising from two different effects. The first is the softening of the phonon frequencies on thermal expansion that is present in almost all metals. The second effect is an unusually strong volume dependence in the electronic band structure, which is reflected not only in $\rho(T)$ but also most clearly in the temperature dependence of the thermal expansion coefficient. This anomaly in the expansion is not present for the metals that show resistivity saturation. Thus, there is no connection between the resistivity saturation and the abnormal temperature dependence of $\rho(T)$ for indium

For completeness, we mention that there should be terms added to (4) which arise from multiphonon scattering and Debye-Waller factors. They also give a Δ linear in T. These terms are usually found to be small[15] and there is no reason why they should be large in indium.

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