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INVESTIGATION OF THE EFFECT OF PRESSURES UP TO 30 katm ON THE CRITICAL FIELD OF TIN AND INDIUM AT TEMPERATURES OF 0.1–4° K

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The effect of pressures up to 30 katm on the critical fields of tin and indium is investigated in the temperature range between 0.1 and 4°K. It is found that to a first approximation the magnitude of the deviation of the critical-field curves from parabolas is the same throughout the indicated pressure range. This is used for calculating the pressure dependence of the density of states N_0 at the Fermi surface and of the electron-phonon interaction parameter V by employing the BCS formula. Similar calculations have been carried out for cadmium and zinc on the basis of the data in [4, 13] In tin, and apparently in cadmium and zinc, the lowering of the superconducting transition temperature T_c due to compression occurs at the expense of a decrease in N_0, the parameter V remaining the same within the accuracy of the measurements. In indium, in addition to a decrease of N_0 which slows down at pressures above 10 katm, a noticeable decrease of the parameter V is observed which is of the order of 5 percent at a pressure of about 28 katm.

INTRODUCTION

ONE of the main results of the theory of superconductivity[1] is the formula for the superconducting transition temperature:

$$T_c = 0.85\Theta \exp(-1/N_0V), \quad (1)$$

where Θ is the Debye temperature, N_0 is the density of electron states at the Fermi surface in the normal state, and V is the parameter characterizing the electron-phonon interaction.

According to (1) the dependence of the superconducting transition temperature on the pressure should be given by the pressure dependence of the parameters Θ, N_0, and V. A calculation of the pressure dependence of the Debye temperature can be carried out in accordance with the formula

$$\Theta(p) = \Theta(0)\,(v_0/v_p)^{-\xi}, \quad (2)$$

obtained by integrating the expressions $\xi = -d\ln\Theta/d\ln v$ under the assumption that the Grüneisen constant does not depend on the pressure (v is the volume of the sample). The coefficient ξ is obtained from the Grüneisen formula[2, 3] and is 2.14 for tin and 2.35 for indium. From Eq. (2) it follows that on increasing the pressure the Debye temperature should increase. In the case of tin and indium the sign of dΘ/dp is not the same as that of dT_c/dp, and consequently the basic parameters determining the pressure dependence of T_c are the parameters N_0 and V.

It is of interest to determine the pressure dependence of N_0 directly and making use of the data on the pressure dependence of T_c and Θ, obtain from Eq. (1) the change of the electron-phonon interaction parameter with pressure.

In order to determine the pressure dependence of the density of states, it is convenient to make use of the results of measurements of the critical-field curves obtained at various pressures in a sufficiently broad temperature range. Making use of the relationship for the coefficient γ in the electron part of the expression for the specific heat of metals in the normal state

$$\gamma = {}^2/_3\pi^2k^2N_0 \quad (3)$$

and of the relation

$$\gamma = (2\pi)^{-1}a_2H_{0c}^2/T_c^2, \quad (4)$$

which follows from the thermodynamics of superconductors, we obtain

$$N_0 = (3/4\pi^3k^2)\,a_2H_{0c}^2/T_c^2. \quad (5)$$

In this expression a_2 is a coefficient which characterizes the deviation of the critical-field curves from a parabola; H_{0c} is the critical field at T = 0°K.

Clearly the value of H_{0c} obtained by extrapolation from the region of temperatures above 1°K does not satisfy the requirements of the necessary accuracy. In this work we have investigated the $H_c(T)$ curves in the 0.1–4° K region of temperatures at pressures up to 30,000 atm. Tin and indium were chosen as objects of our investigation because these elements have sufficiently high values of T_c which are in a region convenient for measurements. This makes it possible, by amplifying the measurements of $H_c(T)$ at temperatures attainable by pumping helium vapor with measurements at infralow temperatures, to determine with sufficient accuracy $H_0(T)$ and the shape of the H_cT curves, and thus determine the coefficient a_2 and its charge upon compression.

METHOD OF MEASUREMENT

The measurements on spectrally pure samples of tin and indium were carried out in a device described in detail in [4, 5] which permitted us to carry out investigations of the effect of pressure on the superconductivity at helium and infralow temperatures. The pressure was produced by means of a booster cooled by adiabatic demagnetization. At pressures up to 16 katm the pistons used in the booster were of purified beryllium bronze. Pressures exceeding 16 katm were produced with the aid of pistons of VK-3 alloy. These pistons exhibit weak ferromagnetism which can introduce an error in the determination of the magnetic field. To reduce this error, a copper rod about 6-mm long was placed between the piston and the sample; the rod was surrounded with graphite lubricant and transmitted the pressure to the sample. The pressure was determined from the magnitude of the shift of the superconducting transition temperature of the investigated samples. The samples were in the form of cylinders 2.6–2.9 mm in diameter and 2–4 mm long.

As the basic manometric substance we chose tin, for which the formula

$$T_c = 3.733 - 4.95\cdot10^{-5}p + 5.9\cdot10^{-10}p^2, \qquad (6)$$

which has been established by Swenson on the basis of measurements at pressures up to 10 katm,[6] can apparently be used for pressures up to 30–40 katm.[7] In order to check the validity of the formula

$$T_c = 3.407 - 4.36\cdot10^{-5}p + 5.2\cdot10^{-10}p^2, \qquad (7)$$

obtained for indium at pressures up to 10 katm,[6] special measurements were carried out in the region of higher pressures; in these measurements two samples were simultaneously placed in the booster: a tin and an indium sample. Satisfactory agreement between formulas (6) and (7) was observed at pressures up to 30 katm.

The terrestrial field and the residual field of the magnet at the location of the sample were compensated by a system of Helmholtz coils to within 0.1 Oe. The magnetic field used to destroy the superconductivity was produced by Helmholtz coils with an Armco-type iron core which made it possible to obtain with a current of 1 amp a field of 254 Oe. The Helmholtz system was supplied by a special electronic device with constant internal resistance. The transition of the samples into the superconducting state and from it was recorded by an electronic technique with an amplitude of the alternating field of 0.05 Oe and at a frequency of 22 cps.

The measurements were carried out in the following way. Lowering the temperature by pumping off helium vapor, we recorded the superconducting transition curve of the sample with zero magnetic field, determining thereby the transition temperature T_c. Then, at a fixed vapor pressure of the helium, we recorded the curves of the destruction of the superconductivity by a magnetic field. Simultaneously a ballistic method was used to record the calibration of the salt. The minimum temperature attainable by pumping of the helium vapor was 1.56° K. At this temperature adiabatic demagnetization was carried out starting from a field of the order of 13 kOe. In the region of ultralow temperatures, measurements of the curves of the destruction of the superconductivity by the magnetic field were carried out with slow warming of the device 30–40 minutes after the adiabatic demagnetization, a time essential for the establishment of thermal equilibrium. Heating the instrument from a temperature of ~0.1 to ~0.6°K took 6–8 hours, a time which ensured a sufficient stability of the temperature when recording each curve.

In determining the thermodynamic temperature of the salt allowance was made for corrections for the demagnetization factor of the salt and for the deviation of the temperature dependence of the magnetic susceptibility of the iron ammonium alum from the Curie law at low temperatures. For the salt pellet with the 4:1 ratio of the axes used in this work both corrections practically compensate for each other. Therefore at temperatures about 0.1° K the temperature of the salt obtained directly from the Curie law coincided with the thermodynamic temperature with an accuracy up to 5 percent. This discrepancy decreased with increasing temperature.

In order to investigate the possibility of the existence of a temperature drop between the salt and the sample, an iron ammonium alum pellet was pressed into the booster channel. The temperatures

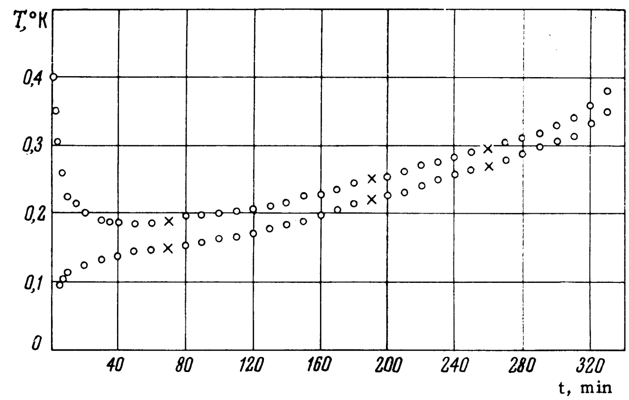

FIG. 1. Curves of the establishment of thermal equilibrium. The lower curve refers to the working salt with a ratio of axes 4 : 1 whose temperature practically coincides with the thermodynamic temperature; the upper curve refers to the salt located in the booster. x – points obtained on converting the temperature of the salt in the booster to the thermodynamic temperature.

of the upper and lower salts were measured simultaneously using one and the same method. The results of the measurements obtained in one of the control experiments are shown in Fig. 1. From a comparison of the curves of Fig. 1 it is seen that after introducing the corrections the temperatures of the upper and lower salt coincide within an accuracy of about 2 percent, in any case starting from a temperature ~ 0.15° K.

MEASUREMENT RESULTS

The width of the superconducting transitions of tin and indium in zero magnetic field changes weakly on compression, and does not exceed 0.03° K. In order to exclude any influence of a change in the transition width in compressed samples on the results of the measurements, the values of T_c were determined from the point of intersection of the straight-line portion of the transition with the horizontal line corresponding to a constant value of the signal up to the onset of the transition.

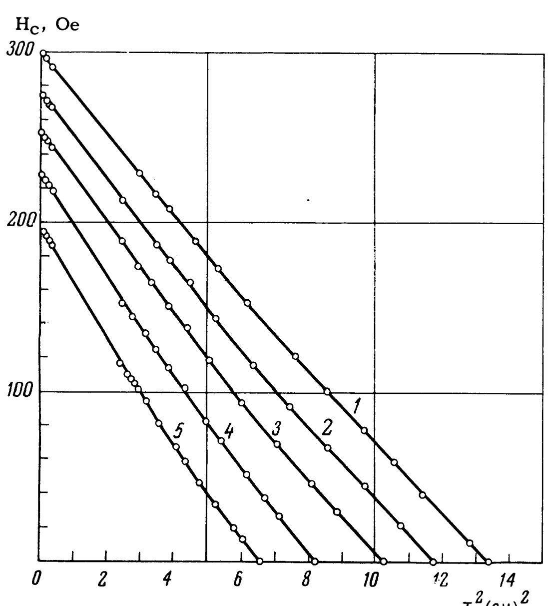

FIG. 2. Critical field curves of tin at various pressures: 1 – p = 1400 atm, 2 – p = 6600 atm, 3 – p = 11,900 atm, 4 – p = 20,000 atm, 5 – p = 31,600 atm.

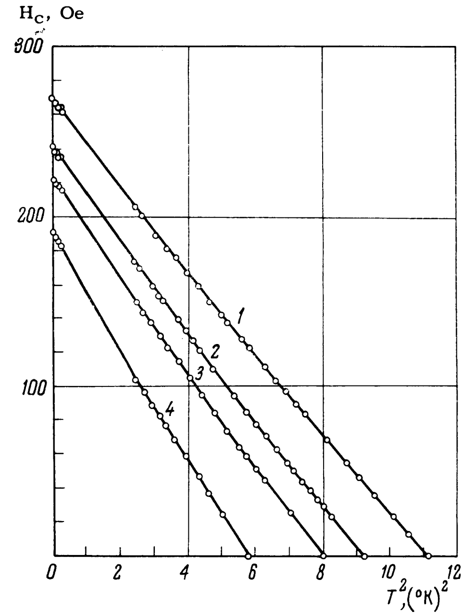

FIG. 3. Critical field curves of indium at various pressures: 1 – p = 1800, 2 – p = 9600 atm, 3 – p = 15,000 atm, 4 – p = 26,000 atm.

The value of the critical magnetic field was determined from the point of intersection of the straight-line portion of the curves of the destruction of the superconductivity by the magnetic field with the horizontal line corresponding to a constant value of the signal after the complete destruction of

Tin			Indium		
p, atm	T, °K	H_{0c}, Oe	p, atm	T °K,	H_{0c}, Oe
0	3.73	306	5900	3.167	258
10000	3,275	262	18500	2,705	216
14400	3.105	248	800	3.374	278
1800	3.645	295	6000	3.175	258
900	3,69	301	11500	2,973	240
5200	3,52	283	13000	2.9	232
6600	3.428	276	13000	2,9	231
11900	3.2	254	9600	3.037	243
3600	3,561	289	12400	2,911	232
15200	3,073	243	1800	3,33	272
15300	3,065	242	6600	3.145	255
20000	2,878	228	12200	2,933	233
19500	2,914	229	8300	3,083	246
1400	3,665	300	15000	2.828	226
31600	2.550	198	26000	2.434	192
1900	3,645	296	26500	2,405	192
29000	2.625	202	27500	2,365	186
			3400	3.265	265
			25000	2.478	196

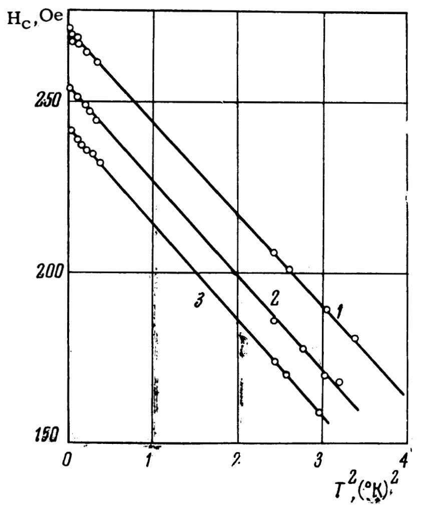

FIG. 4. Critical field curves of indium illustrating the determination of H_{0c}: 1 – p = 1800 atm, 2 – p = 6600 atm, 3 – p = 9600 atm.

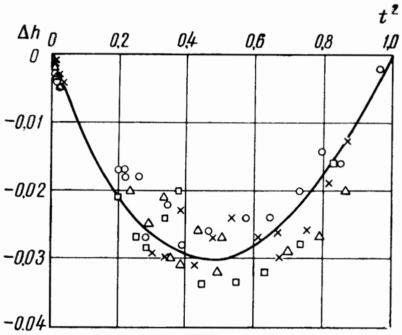

FIG. 5. The deviation $[\Delta h = h - (1 - t^2)]$ of the critical-field curves from a parabola in tin at various pressures: o – p = 1400 atm, × – p = 20,000 atm, □ – p = 6600 atm, Δ – p = 11,900 atm.

the superconductivity. The values of H_c determined by this method are in good agreement with the values of T_c determined from the onset of the transition. At the same time, effects due to variations in the transition width and changes in the demagnetization factor of the sample on compression are to a first approximation excluded.

The critical field curves measured in tin and indium under various pressures are shown in $H_c - T^2$ coordinates on Figs. 2 and 3. It is seen that all the curves deviate noticeably from parabolicity.

The values of T_c and H_{0c} obtained for various pressures in the order in which the measurements for tin and indium were carried out are given in the Table.

A few remarks concerning the accuracy of the measurements. The error in the determination of T_c does not exceed 0.01°K, i.e., ~0.3 percent. The accuracy of the graphical determination of H_{0c} by the least-squares method amounts to about 1.5 Oe, i.e., ~0.6 percent. Examples of the determination of H_{0c} in indium from the $H_{0c}(T^2)$ curves plotted on a scale larger than in Figs. 2 and 3 are shown in Fig. 4. In the region of infralow temperatures the possible error in the temperature determination amounts to about 10 percent near 0.1°K and decreases with increasing temperature; this can introduce an additional error of the order of ~0.1 percent in the determination of H_{0c}. In order to exclude possible systematic errors, the measurements were carried out on a large number of samples of various dimensions; boosters of various constructions and various methods of pressing the salt were employed. In measurements with each sample the pressure was increased nonmonotonically, and control measurements at low pressure were always carried out after several compression cycles.

DISCUSSION OF RESULTS

A. Pressure dependence of the density of states. In order to calculate the density of states at various pressures from formula (5), one must, in addition to the corresponding values of $H_{0c}(p)$ and $T_c(p)$, know the behavior of the parameter a_2 on compression. As is well known, the parameter a_2 which characterizes the deviation of the shape of the critical-field curves from parabolicity is the first coefficient in the polynomial

$$h(t) = 1 - \sum_{n=2}^{N} a_n t^n, \qquad (8)$$

where $h = H_c/H_{0c}$ and $t = T/T_c$. The value of a_2 can be determined from the slope of the $h(t^2)$ curves in the region of small t^2, as well as from the dependences of $\Delta h = h - (1 - t^2)$ on t^2 which illustrates the deviation of the h(t) curves from the parabolas $h' = 1 - t^2$.

$\Delta h(t^2)$ curves of tin and indium plotted for various pressures are shown in Figs. 5 and 6. In these

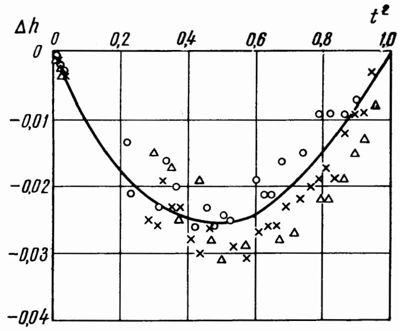

FIG. 6. The deviation of the critical-field curves from a parabola in indium at various pressures: × – p = 6600 atm, o – p = 1800 atm, Δ – p = 18,000.

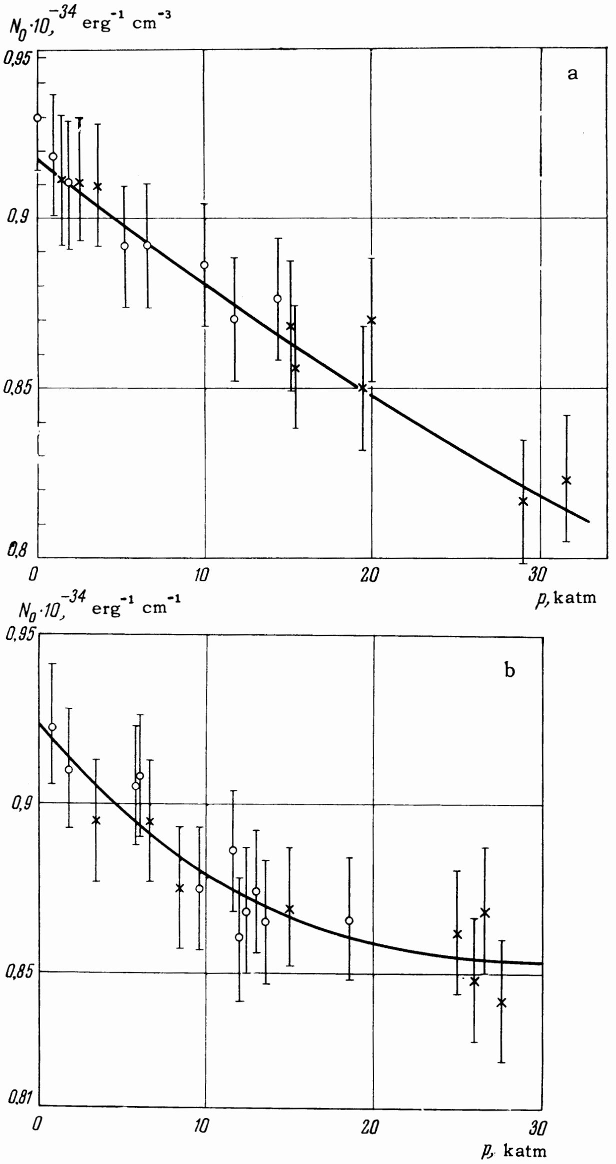

FIG. 7. Pressure dependence of the density of electron states N_0: a – in tin, b – in indium, × – points obtained with a piston of VK-3, o – points obtained with a bronze piston.

figures the continuous curves show the results of precision measurements of the critical fields of unstressed tin and indium samples.[8] It is seen that within the limits of accuracy of the measurements the coefficients a_2 retain at all pressures the value 1.07 ± 0.01 for tin and 1.06 ± 0.01 for indium.

The pressure dependences of the density of states in tin and indium calculated from (5) for these values of the coefficients are shown in Fig. 7. Values of N_0 for $p = 0$ are in satisfactory agreement with the results of calorimetric measurements: 0.81×10^{34} erg^{-1} cm^{-3} for tin and 0.77×10^{34} erg^{-1} cm^{-3} for indium.[9] The values $\nu = (\partial \ln N_0/\partial \ln v)_{p=0}$ of 2 ± 0.3 for tin and 1.5 ± 0.3 for indium are in agreement with the values $\nu = 1.7 \pm 0.3$ for tin and $\nu = 1.0 \pm 0.2$ for indium determined in [10, 11], in which ν of tin was determined by a method analogous to that used in this work at pressures of 2 katm, and ν of indium from the change in the volume of the sample on going over from the superconducting state to the normal state under the action of a magnetic field. In tin the density of states decreases on compression apparently very nearly according to a linear law. Unlike in tin, in indium the rate of change of N_0 decreases noticeably at high pressures.

It is of interest to attempt to analyze, at least qualitatively, the obtained results from the point of view of the effect of pressure on the energy spectrum of the electrons of these metals. When the crystal lattice is compressed the radius P_0 of the Fermi surface (in the approximation according to Harrison's method) increases inversely as the cube root of the volume of the sample, $P_0 \propto v^{-1/3}$. If during compression the anisotropy of the lattice does not change, then the volume of the reciprocal cell increases simultaneously with P_0. As a result the overlap of the bands and consequently the total concentration of current carriers n does not change, and the density of states calculated per unit volume $N_0 \sim n/v\epsilon_0 \sim n(v)^{-1/3}$ (ϵ_0 is the Fermi energy) increases at the expense of a decrease of the sample volume. If, on the other hand, the anisotropy of the lattice changes (usually the tendency is towards a decrease), then as a result of a change in the magnitude of the band overlap and the redistribution of the electrons over the bands the concentration of current carriers n may change. From this point of view the observed decrease in the density of states in tin and indium on compression can be related to a decrease of the concentration of current carriers. The stronger change of N_0 in tin is in agreement with the much stronger, compared to indium, change of the anisotropy of the lattice under the action of the pressure, and therefore with the possibility of a stronger change in the concentration n.

Naturally such a consideration of the problem is by no means rigorous, since no account was taken in it of a possible change of the effective mass. However, the possibility itself of an appreciable decrease in the concentration of the current carriers in the fundamental bands of such good metals as tin and indium on compression appears to us to be interesting.

B. <u>The effect of pressure on the electron-phonon interaction constant.</u> The result of a calculation of the parameter V according to Eq. (1) depends on the extent of the change of the Debye temperature Θ under the action of the pressure. Unfortunately

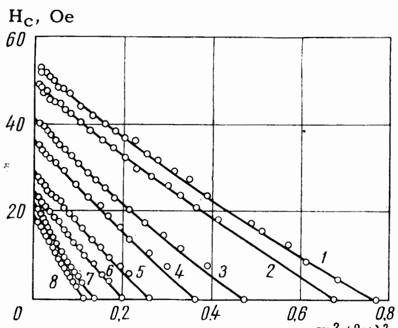

FIG. 8. Critical field curves of zinc at various pressures: 1 – p = 0, 2 – p = 3000 atm, 3 – 7300 atm, 4 – p = 11,800 atm, 5 – p = 15,600 atm, 6 – p = 19,800 atm, 7 – p = 23,900 atm, 8 – p = 26,200 atm.

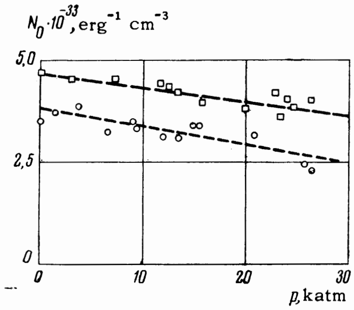

FIG. 9. Curve of the pressure dependence of the density of electron states N_0 in cadmium and zinc: o – cadmium, □ – zinc.

there exist practically no accurate experimentally obtained data on the pressure dependence of Θ. On the other hand, the calculation of Θ according to the Grüneisen formula is inaccurate, since it can only be carried out under certain assumptions. Nevertheless, the possible error in the calculation of the magnitude of the change of Θ on compression is apparently unimportant for the determination of the dependence of V on p, since the change in T_c caused by the change in Θ is much smaller than the effect of the exponential factor in Eq. (1) on T_c. As we have already stated, a calculation of Θ can be carried out with the aid of (2). It is interesting to note that for tin the value of Θ calculated from (2) at 80 katm is in good agreement with the value of Θ determined at this pressure experimentally in [12].

The values of the parameter V calculated for the values of Θ determined in such a way turn out in the case of tin to be constant within experimental accuracy in the entire region of pressures. This means that the basic reason for the decrease in the superconducting transition temperature on compressing tin is the decrease in the density of states on the Fermi surface.

A decrease of the parameter V reaching about 5 percent at p = 28,000 atm is apparently observed on compression.

It is interesting to determine how the parameters N_0 and V vary on compression in zinc and cadmium which have been investigated previously.[4, 13] Critical field curves of zinc in $H_c - T^2$ coordinates (which were not shown in [13]) are presented in Fig. 8. Notwithstanding the accuracy of the measurements which is smaller than for tin and indium, these data apparently attest to the absence of any noticeable pressure dependence of a_2 in the entire range of pressures. This permits one to use formula (5) to estimate the effect of pressure on the density of states of zinc assuming the coefficient a_2 to be constant with a value ~1.15.

Values of $N_0(p)$ for zinc calculated on this assumption are shown in Fig. 9. Values of $N_0(p)$ for cadmium in accordance with the data of [4] are plotted in the same figure.

Since the values of H_{0c} and T_c of zinc and cadmium could only be determined with an accuracy of about ± 5 percent, a rather large scatter of the values of $N_0(p)$ is observed for these elements. However, the observed decrease in $N_0(p)$ amounting to ~ 20 percent for zinc and ~ 30 percent for cadmium at a pressure of p = 30 katm exceeds several times the possible error in the determination of $N_0(p)$.

The parameter V in zinc and cadmium retains within the accuracy of the measurements a constant value in the entire range of pressures.

Thus, in nontransition metals—tin, cadmium, zinc, and to a lesser extent indium (just as in the case of transition metals[14])—one should consider the basic reason for the change of T_c under the action of pressure to be the change in the density of electron states on the Fermi surface. It is impossible to say in advance whether this situation will remain the same in the region of considerably higher pressures, and therefore at the present time the problem of the nature of the change of T_c in this region (in particular the problem of the possible disappearance of the superconductivity at some critical pressure) remains open.

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