

# The specific heat of Nb<sub>3</sub>Sn in magnetic fields up to 19 T

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The specific heat of the superconducting compound Nb<sub>3</sub>Sn has been studied experimentally in the temperature range 4.5–70 K in magnetic fields up to 19 T. The low-temperature values were determined for the coefficient of the electronic specific heat,  $\gamma = 11 \text{ J} \cdot \text{kmole}^{-1} \cdot \text{K}^{-2}$ , for the Debye temperature  $\Theta = 232 \text{ K}$  and for the parameter of the Ginzberg-Landau-Abrikosov-Gor'kov mixed state theory  $\kappa(T_c) = 24$ . It was found that  $\Theta$  and  $\kappa$  vary smoothly with temperature. The specific heat discontinuity at the superconducting transition in various magnetic fields was investigated in detail.

## INTRODUCTION

The intermetallic compound Nb<sub>3</sub>Sn has an A-15 crystal structure and is a widely used superconductor with high critical parameters. A study of the low-temperature specific heat of a superconductor in the normal state, when superconductivity is suppressed by a magnetic field, enables such important quantities as the electron density of states at the Fermi level, the Debye temperature, the difference in energy between the normal and superconducting states, the parameters  $\kappa_1$  and  $\kappa_2$  of the Ginzburg-Landau-Abrikosov-Gor'kov (GLAG) mixed state theory, etc., to be determined. As a result of the large value of the critical magnetic field  $H_{c2}(0)$ , very high (of the order of 20–25 T) magnetic fields are required to study Nb<sub>3</sub>Sn in the normal state near absolute zero. The majority of the investigations of the specific heat of Nb<sub>3</sub>Sn have been carried out in the absence of a magnetic field<sup>1–5</sup> or in insufficiently high magnetic fields.<sup>6</sup> Stewart *et al.*<sup>7,8</sup> tried to measure the specific heat of Nb<sub>3</sub>Sn in high magnetic fields (18 and 12.5 T respectively), but these measurements gave strange and contradictory results and they cannot be regarded as reliable. The low-temperature specific heat has been studied in the present work in the normal, superconducting and mixed states over the wide range of magnetic fields from 0 to 19 T.

## THE EXPERIMENTS

The polycrystalline Nb<sub>3</sub>Sn specimen was prepared by repeated sintering of powders, using the technique described by Karkin *et al.*<sup>9</sup> According to an x-ray structural analysis, the specimen was practically single-phase, the parameter of the A-15 lattice at room temperature was 5.290 Å. The temperature of the superconducting transition  $T_c$  and its width  $\Delta T_c$ , determined by a four-contact method from the disappearance of electrical resistivity, were  $T_c = 18.1 \text{ K}$  and  $\Delta T = 0.1 \text{ K}$ . The resistivity ratio  $\rho_{300\text{K}}/\rho_{19\text{K}}$  was 6. No singularity corresponding to a structural phase transition, observed by Vieland and Wickland,<sup>10</sup> was found in the temperature dependence of the specific heat. The 1.5 g Nb<sub>3</sub>Sn specimen studied was in the form of a plane disk, and the direction of the external magnetic field was perpendicular to its plane.

The specific heat was measured by an adiabatic method<sup>11</sup> over the temperature range 4.5–70 K without a magnetic field and between 4.5 and 20 K in magnetic fields. Magnetic fields up to 8 T were provided by a laboratory superconducting solenoid, and above 8 T by the KS-250 combined magnetic system.<sup>12</sup> The working space of the calorimeter was surrounded by a high-purity aluminum shield cooled to helium temperature, which ensured a considerable reduction in eddy current heating of the specimen. Temperatures were measured by a carbon resistance thermometer<sup>13</sup> with a correction applied for the effect of a magnetic field.<sup>14</sup> The working of the calorimeter was verified by measuring the specific heat of a specimen of pure copper and it appeared that the error in measuring specific heat was less than 1% in magnetic fields up to 8 T and 2% in fields up to 19 T.

For measurements in magnetic fields less than 8 T, the specimen was cooled to the minimum temperature without a magnetic field, the field was then applied and the specific heat measured. In experiments in fields greater than 8 T, the specimen was cooled in the applied field.

## RESULTS

The specific heat of Nb<sub>3</sub>Sn for different values of the magnetic field is shown in Fig. 1, plotted as  $C/T$  vs  $T^2$ . For every value of the magnetic field, the temperature dependence of the specific heat shows sharp jumps, corresponding to a phase transition from the superconducting or mixed state to the normal state. In the case of a type II superconductor, such a transition is a second-order phase transition both in the absence of a magnetic field and in a magnetic field. The application of a magnetic field lowers the temperature of this transition, so that the specific heat of Nb<sub>3</sub>Sn can be studied in the normal state at fairly low temperatures (down to 9 K in a field of 19 T).

A departure is observed in the temperature dependence of the specific heat of Nb<sub>3</sub>Sn in the normal state at low temperatures from the relation usual for metals,  $C = \gamma T + \beta T^3$ . If the specific heat followed this relation, then it would be represented in  $C/T$  vs  $T^2$  coordinates by a straight line, and the intersection of this line on the  $y$  axis would give the value of the coefficient of the electronic specific heat  $\gamma$ . The rela-

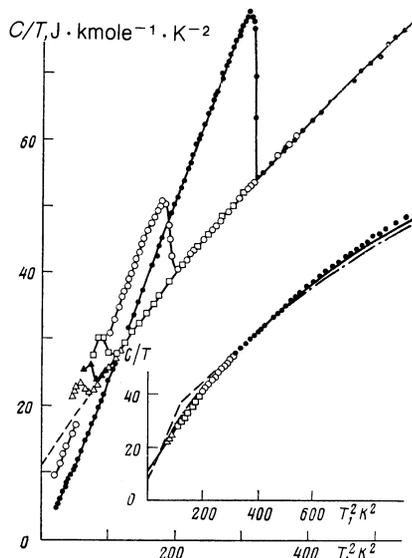


FIG. 1. The specific heat of  $\text{Nb}_3\text{Sn}$  in different magnetic fields:  $\bullet$ ) 0 T;  $\circ$ ) 8 T;  $\square$ ) 16 T;  $\blacktriangle$ ) 18 T;  $\triangle$ ) 19 T. Not all the experimental points below 18 K are shown. The inset shows that specific heat of  $\text{Nb}_3\text{Sn}$  in the normal state: points, the present work; dashed line, Stewart *et al.*;<sup>7</sup> solid line, calculation of the specific heat according to the phonon spectrum without taking anharmonicity into account or the temperature dependences of the coefficient  $\gamma$  and of the phonon spectrum; the dashed-dot curve takes these effects into account.

tion  $C = \gamma T + \beta T^3$  is only fulfilled for  $T < 12$  K, so that the specific heat must be measured directly in the normal state in this temperature range for a correct determination of the coefficient  $\gamma$ . Extrapolation of the temperature variation of the specific heat from higher temperature can lead to appreciable errors (100% in the present case) in the determination of the coefficient  $\gamma$ .

The temperature dependence of the specific heat of the specimen studied in the normal state in the temperature range 9–18 K is satisfactorily described by the polynomial

$$C [\text{J} \cdot \text{kmole}^{-1} \cdot \text{K}] = 11.06T + 0.155T^3 - 0.000065T^5 \quad (1)$$

with a mean square deviation of 1.2% and a maximum devi-

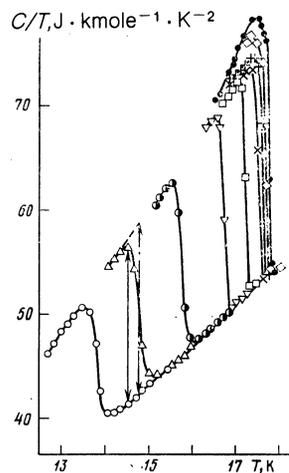


FIG. 2. The specific heat of  $\text{Nb}_3\text{Sn}$  in magnetic fields up to 8 T:  $\bullet$ ) 0 T;  $\diamond$ ) 0.1 T;  $+$ ) 0.2 T;  $\times$ ) 0.5 T;  $\square$ ) 1 T;  $\nabla$ ) 2 T;  $\circ$ ) 4 T;  $\triangle$ ) 6 T;  $\circ$ ) 8 T. The arrows show how the magnitude of the specific heat jump at the superconducting transition was determined: dashed-dot curve, by extrapolating to the limitingly narrow transition; solid line, without extrapolation.

ation of 3%; the equality of the entropies of the normal and superconducting states at a temperature of 18 K was then fulfilled. The low-temperature values of the coefficient of the electronic specific heat and of the Debye temperature obtained from Eq. (1) were  $\gamma = 11 \text{ J} \cdot \text{kmole}^{-1} \cdot \text{K}^{-2}$  and  $\Theta = 232 \text{ K}$ . An estimate of the electron-phonon coupling constant  $\lambda$  according to the McMillan formula,<sup>15</sup> using the values we obtained for  $\Theta$  and  $T$ , assuming that the Coulomb pseudopotential is  $\mu^* = 0.13$ , gave  $\lambda = 1.37$ . Smoothed values of the specific heat of  $\text{Nb}_3\text{Sn}$  in the superconducting and normal states are given in Table I.

The specific heat of  $\text{Nb}_3\text{Sn}$  in fields up to 8 T is given in Fig. 2; in these experiments the specimen was cooled before the magnetic field was turned on. The specific heat jumps, corresponding to the superconducting transition, are somewhat drawn out in temperature, evidently the results of some specimen nonuniformity. The width of the jump, determined as the difference between the temperature at which the departure from the normal state specific heat started and

TABLE I. The specific heat of  $\text{Nb}_3\text{Sn}$  at constant pressure  $C_p$  in the superconducting and normal states.

Superconducting state		Normal state			
T, K	$C_p, \text{J} \cdot \text{kmole}^{-1} \cdot \text{K}^{-1}$	T, K	$C_p, \text{J} \cdot \text{kmole}^{-1} \cdot \text{K}^{-1}$	T, K	$C_p, \text{J} \cdot \text{kmole}^{-1} \cdot \text{K}^{-1}$
5	28.7	5	—	18	980
6	48.7	6	—	19	1140
7	76.7	7	—	20	1249
8	115.1	8	—	25	2069
9	168	9	207	30	3068
10	236	10	260	35	4212
11	322	11	317	40	5457
12	425	12	385	45	6732
13	546	13	460	50	7958
14	689	14	548	55	9102
15	857	15	645	60	10 218
16	1051	16	750	65	11 374
17	1260	17	862	70	12 284

Note: Below 25 K the specific heat of  $\text{Nb}_3\text{Sn}$  in the normal state is described by Eq. (1) with an error of not more than 1.5% (1 kmole = 99.35 kg).

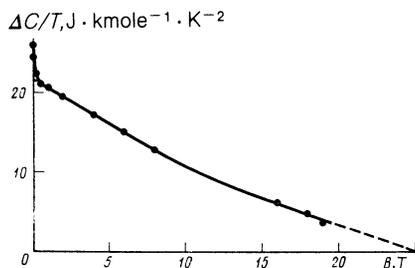


FIG. 3. The magnitude of the specific heat jump  $\Delta C/T$  at the superconducting transition (without extrapolation) as a function of the magnetic field.

the temperature at which the specific heat reached the maximum, increased with magnetic field, and in zero field was  $\sim 0.4$  K. The finite width of the jump introduced some uncertainty in the determination of its amplitude, so that the value of the jump in specific heat at the superconducting transition was determined by two means, as shown in Fig. 2: by extrapolating to the limitingly narrow transition and without extrapolation, as the difference between the specific heats at the maximum and in the normal state.

The dependence of the magnitude of the specific heat jump  $\Delta C/T_c$  at the superconducting transition on the magnitude of the magnetic field  $B$  is shown in Fig. 3. This dependence is practically linear in the range of fields 0.5–8 T, while the linearity is destroyed in fields less than 0.5 T.

Values of the temperatures corresponding to the start and middle of the superconducting transition, the specific heat peak and the magnitude of the specific heat jump  $\Delta C/T_c(B)$  at the superconducting transition are given in Table II as a function of applied magnetic field  $B$ . Values are also given here of the thermodynamic magnetic field  $B_c(T)$  and the parameters  $\kappa_1$  and  $\kappa_2$  of the GLAG mixed state theory, at temperatures corresponding to the middle of the superconducting transition in a magnetic field. The thermodynamic field  $B_c(T)$  was evaluated from the difference between the specific heats in the normal and superconducting states,  $C_n(T)$  and  $C_s(T)$ , according to the relation

$$\frac{VB_c^2(T)}{2\mu_0} = \int_{T_c}^{T_c} \frac{C_s(T) - C_n(T)}{T} d^2T, \quad (2)$$

where  $V$  is the volume of 1 kmole ( $V = 0.0111 \text{ m}^3 \cdot \text{kmole}^{-1}$

for  $\text{Nb}_3\text{Sn}$ ),  $\mu_0$  is the magnetic permittivity of the vacuum,  $T_c$  is the superconducting transition temperature in the absence of a field). The values of the parameters  $\kappa_1$  and  $\kappa_2$  of the mixed state were determined from the relations<sup>16</sup>

$$B_{c2}(T) = 2^{1/2} \kappa_1 B_c(T), \quad (3)$$

$$\frac{\Delta C}{T_c(B)} = \frac{V(\partial B_{c2}/\partial T)^2}{4.16\mu_0(2\kappa_2^2 - 1)},$$

where  $\Delta C/T_c(B)$  is the value of the specific heat jump at the superconducting transition in a magnetic field, extrapolated to the limitingly narrow transition, and  $B_{c2}(T)$  is the upper critical magnetic field. The values of  $\kappa_1$  and  $\kappa_2$  agree in zero field and increase smoothly as the temperature decreases,  $\kappa_2$  growing faster than  $\kappa_1$ .

## DISCUSSION

The specific heat of  $\text{Nb}_3\text{Sn}$  which we measured in the absence of a magnetic field agrees satisfactorily with earlier results.<sup>2–8</sup> The small differences can be due to the different quality of the specimens. Our values of the parameters  $\Theta$  and  $\gamma$  agree well with Junod *et al.*<sup>4</sup> and Shamrai *et al.*,<sup>5</sup> but differ appreciably from others.<sup>1,3,7,8</sup> The results of our measurements in high magnetic fields depart considerably from the results of Stewart *et al.*<sup>7</sup> and of Stewart and Brandt,<sup>8</sup> carried out on  $\text{Nb}_3\text{Sn}$  specimens in magnetic fields up to 18 and 12.5 T respectively. In those experiments, a sharp change in slope was observed in the  $C/T$  vs  $T^2$  dependence at temperatures of 11 and 15 K respectively. Our results indicate that the change in slope of the  $C/T$  vs  $T^2$  curve takes place smoothly, without a break. In our view, the sharp change observed<sup>7,8</sup> is illusory and is due to the error in the measurements,<sup>7,8</sup> which is appreciably greater than in our measurements.

The possible reasons for the departure of the temperature dependence of the specific heat of  $\text{Nb}_3\text{Sn}$  in the normal state from the usual relation for metals,  $C = \gamma T + \beta T^3$  has been analyzed theoretically.<sup>17</sup> It was shown that neither the electron spectrum fine structure nor the temperature dependence of the renormalization of the electron spectrum by the electron-phonon interaction can explain the anomalies observed<sup>7,8</sup> in the temperature dependence of the specific heat, at the same time that an anomaly in the phonon spectrum (for example, a low-frequency Einstein peak in the phonon density of states) can describe the smooth departure, agree-

TABLE II. The parameters of the superconducting transition of  $\text{Nb}_3\text{Sn}$  in different magnetic fields  $B$ .

B, T	$T_c(B)$ , K			$\frac{\Delta C}{T_c(B)}$ , J kmole $\cdot$ K $^{-1}$		$B_c$ , T	$\kappa_1$	$\kappa_2$
	peak	centre	start	without extrapolation	extrapolation			
0	17.6	17.8	18.0	26.0	28	—	24	24
1	17.1	17.3	17.5	20.8	22	—	—	26
2	16.6	16.8	17.0	19.6	21	0.056	25.4	27
4	15.6	15.8	16.1	17.4	18	0.104	27.3	29
6	14.6	14.85	15.2	15.2	16.2	0.151	28.1	31
8	13.5	13.85	14.2	13.0	14.2	0.195	29.1	33
16	9.4	9.9	10.3	6.2	6.5	0.334	33.9	43
18	8.5	8.8	9.1	5.0	5.3	0.364	35.0	45
19	7.7	8.1	8.6	3.8	4.0	0.381	35.4	46
—	—	0	—	—	—	0.473	—	—

ing in order of magnitude with that observed.

The phonon spectrum of Nb<sub>3</sub>Sn has been studied<sup>18</sup> by the inelastic neutron scattering method and a maximum with energy of the order of 9 meV was observed in the low-frequency part of the spectrum. We calculated the specific heat of Nb<sub>3</sub>Sn using the spectrum obtained<sup>18</sup> at a temperature of 5 K. The result of the calculation is shown in the inset to Fig. 1. It turned out that the calculated phonon specific heat is only proportional to  $T^3$  up to a temperature of the order of 10 K, and smoothly departs from the cubic law at higher temperatures. In the temperature range up to 45 K, the total calculated phonon specific heat and the linear term in the temperature with the coefficient  $\gamma = 11 \text{ J} \cdot \text{kmole}^{-1} \cdot \text{K}^{-2}$  describes our experimental results with an error of not more than 2%. A more detailed calculation, taking account of anharmonicity and the temperature dependences of both the coefficient  $\gamma$  and of the phonon spectrum, carried out by the method described by Panova *et al.*,<sup>19</sup> gave practically the same result at temperatures up to 30 K.

The normalized value of the specific heat jump at the superconducting transition  $\Delta C / \gamma T_c$  was 2.5, which is considerably higher than the value 1.43 given by the Bardeen-Cooper-Schrieffer theory. The large value of the specific heat jump could be produced both by the energy dependence of the electron density of states<sup>20</sup> and by the frequency dependence of the energy gap,<sup>21</sup> and estimates have shown that the contribution from the latter mechanism dominates.

The temperature dependence of the upper critical magnetic field in fields up to 19 T differs noticeably from a parabolic dependence and is well described by the theoretical dependence of Helfand and Werthamer,<sup>22</sup> while an estimate of the magnitude of  $B_{c2}(0)$  according to them<sup>22</sup> was 25 T. The size and the nature of the temperature variation of the parameters  $\kappa_1$  and  $\kappa_2$  which we determined (see Table II) are in qualitative agreement with Eilenberger's theory.<sup>23</sup>

The value of the parameter  $\kappa$  of the GLAG mixed state theory consists, according to Lynton,<sup>24</sup> of two contributions,  $\kappa_p$  appropriate to the pure material and  $\kappa_i$ , due to scattering of electrons by impurities, with the value of  $\kappa_i$  related to the residual resistivity  $\rho_0$  ( $\Omega \cdot \text{m}$ ) and the coefficient of the electronic thermal capacity per unit volume,  $\gamma_v$  ( $\text{J} \cdot \text{m}^{-3} \cdot \text{K}^{-2}$ ) by the relation

$$\kappa = \kappa_p + \kappa_i = \kappa_p + 7.5 \cdot 10^{13} \cdot 10^5 \rho_0 \gamma_v^{1/2}. \quad (4)$$

Evaluation of  $\kappa_p$  from this relation gave  $\kappa_p = 14$ , which is evidence that the pure intermetalloid Nb<sub>3</sub>Sn should be a type II superconductor with a large value of the parameter  $\kappa$ .

Since Stewart and Brandt<sup>8</sup> reported on the unusual, inadequate present understanding of the behavior of the specific heat of Nb<sub>3</sub>Sn in the mixed state, it was of interest to compare the specific heat in the mixed state measured by us with the existing theory.<sup>16</sup> We note immediately that near the transition from the mixed to the normal state, neither the shape of the specimen nor the magnetic flux pinning has an effect on the specific heat of a type II superconductor with a high value of the parameter  $\kappa$ , since both the magnetization and the magnetic flux pinning are close to zero near the transition.

The theory<sup>16</sup> predicted that as the magnetic field  $B$  tends to zero, the limiting value  $\Delta C_m / T$  of the specific heat discontinuity at the transition from the mixed to the normal state is related to the value of the discontinuity  $\Delta C_s / T$  at the superconducting transition in zero field by the equation

$$\lim_{B \rightarrow 0} \Delta C_m / T = \Delta C_s [1.16 T (1 - 1/2 \kappa^2)]^{-1}. \quad (5)$$

As can be seen from Fig. 3, the magnetic field dependence of the magnitude of the specific heat discontinuity at the superconducting transition is linear in the magnetic field range 0.5–8 T, with its extrapolation to zero field giving a value which is 85% of the value of the discontinuity in zero field, in extremely good agreement with Eq. (5). This linear relation is destroyed in fields less than 0.5 T, evidently as a result of the appearance of a specific heat anomaly corresponding to the lower critical field  $H_{c1}$ . A type II superconductor is automatically not in the mixed (Shubnikov) but in the completely superconducting (Meissner) state in fields below  $H_{c1}$ , to which Eq. (5) is not applicable.

We note that far from the transition to the normal state, the specific heat of a superconductor in a magnetic field does not have a clear thermodynamic meaning, because of the possibility of the capture and dissipative motion of magnetic flux and the nonequilibrium nature of the states and the irreversible thermodynamic processes associated with this. As is well known, the entropy change for irreversible processes is greater than the heat supplied divided by the temperature, and the measured effective specific heat is related to the change in entropy due to the lack of equilibrium, the sign of which depends on the sign of the temperature change in the irreversible process, namely  $C > TdS/dT$  on cooling and  $C < TdS/dT$  on heating. The equality  $C = TdS/dT$  only applies for reversible processes. The specific heat of Nb<sub>3</sub>Sn in the normal ( $C_n$ ) and mixed ( $C_m$ ) states which we measured in a field of 8 T satisfied the equality

$$\int_0^{T_c} \frac{C_n}{T} dT = \int_0^{T_c} \frac{C_m}{T} dT$$

with an error of less than 1%, which is evidence of the satisfactory equilibrium and reversibility of the thermodynamic processes in our measurements. We did not carry out such a check for other values of the field.

The specific heat of Nb<sub>3</sub>Sn in magnetic fields of 11 and 12.5 T, measured<sup>8</sup> by the relaxation method on cooling, is larger than the specific heat in the normal state  $C_n$  over the whole temperature range below  $T_c$ , so that the following inequality holds for the corresponding integrals

$$\int_0^{T_c} \frac{C_m}{T} dT > \int_0^{T_c} \frac{C_n}{T} dT. \quad (6)$$

A similar inequality is also satisfied<sup>25</sup> for the specific heat of Nb<sub>3</sub>Sn in the normal and mixed state in a field of 7.1 T. The satisfaction of such inequalities is evidence of the existence of irreversibility of the thermodynamic processes in measurements of specific heat,<sup>8,25</sup> evidently as a result of jumps of magnetic flux, and therefore the specific heat measured in the mixed state,<sup>8,25</sup> at least far from the transition to the

normal state, does not have a clear thermodynamic meaning.

## CONCLUSIONS

The experimental investigations carried out of the specific heat of  $\text{Nb}_3\text{Sn}$  in magnetic fields up to 19 T made possible reliable determinations of the low-temperature values of the Debye temperature,  $\Theta = 232$  K, of the coefficient of the electronic specific heat  $\gamma = 11 \text{ J} \cdot \text{kmole}^{-1} \cdot \text{K}^{-2}$ , of the temperature dependences of the upper critical field  $B_{c2}(T)$  and of the parameters  $\kappa_1$  and  $\kappa_2$  of the GLAG mixed state theory.

The results of other work<sup>7,8</sup> reporting a sharp change in slope of the  $C/T$  vs  $T^2$  curve in  $\text{Nb}_3\text{Sn}$  at temperatures of 11–15 K were not confirmed. The smooth departure of the  $C/T$  vs  $T^2$  relation from linearity which we observed is due to anomalies in the  $\text{Nb}_3\text{Sn}$  phonon spectrum and is well described quantitatively by using the spectrum obtained by the neutron inelastic scattering method.

The magnetic field dependence of the value of the specific heat discontinuity in  $\text{Nb}_3\text{Sn}$  at the transition from the mixed to the normal state is well described by the relations of the GLAG theory.<sup>16</sup>

The specific heat of a superconductor in a magnetic field, satisfying the inequality in Eq. (6) hardly has a clear thermodynamic meaning, especially far from the transition to the normal state. The unusual behavior of the specific heat of  $\text{Nb}_3\text{Sn}$  in the mixed state, observed by Stewart and Brandt,<sup>8</sup> could be due to dissipative processes on the motion of magnetic flux.

In conclusion, the author expresses his thanks to V. E. Arkhipov for providing the  $\text{Nb}_3\text{Sn}$  specimen, to G. V. Laskova for carrying out the x-ray structural analysis, to A. N. Kulyamzin and V. V. Stepanov for help in carrying out the experiments in high magnetic fields, and to G. Kh. Panov, N. A. Chernoplekov and P. A. Cheremnykh for their interest in the work and for valuable discussions.

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