

# The temperature dependences of the upper critical field and the electrical resistivity of halogen-substituted molybdenum chalcogenides

N. E. Aleksevskii, A. V. Mitin, and G. Wolf

*Institute of Physics Problems, Academy of Sciences of the USSR, Moscow*

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The temperature dependences of the upper critical field  $H_{c2}(T)$  and electrical resistivity  $\rho(T)$  of Chevrel-phase compounds  $\text{Mo}_6\text{X}_6\text{X}'_2$  ( $\text{X} = \text{S, Se}$ ;  $\text{X}' = \text{I, Br}$ ) have been studied. It was found that as the temperature is reduced from 300 to 15 K, the resistivity increases by 30 to 40% and the  $H_{c2}(T)$  dependence is almost linear over the whole temperature range investigated. The relation between the temperature dependences of  $H_{c2}$  and  $\rho$  and features of the crystal structure of halogen-substituted molybdenum chalcogenides were analyzed.

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## INTRODUCTION

Molybdenum chalcogenides  $\text{A}_x\text{Mo}_6\text{X}_8$  ( $\text{A} = \text{Pb, Sn, Cu, \dots}$ ;  $\text{X} = \text{S, Se, Te}$ ) belong to a group of compounds which have been very extensively studied, both because of the unusually wide range of their properties and because many of these properties are unique.<sup>1,2</sup> An appreciable fraction of these compounds are superconductors,<sup>3</sup> and some of them, for example  $\text{Pb}_x\text{Mo}_6\text{S}_8$ , have fairly high critical temperatures  $T_c = 14$  to 15 K and record values of the upper critical field  $H_{c2}(0) \approx 600$  kOe.<sup>4-6</sup>

The element A can be absent ( $x = 0$ ) in a number of cases and then binary  $\text{Mo}_6\text{X}_8$  (Ref. 7) or pseudo-binary  $\text{Mo}_6\text{X}_{8-y}\text{X}'_y$  ( $\text{X}' = \text{S, Se, Te, I, Br}$ ) compounds are formed, among which the halogen-substituted molybdenum sulfides with composition  $\text{Mo}_6\text{S}_6\text{Br}_2$  and  $\text{Mo}_6\text{S}_6\text{I}_2$  are the highest-temperature superconductors ( $T_c = 13.5$  to 14 K).<sup>8</sup>

All the compounds mentioned above (Chevrel-phases) are characterized by a rhombohedral structure (space-group  $C_{3i}^2 - R\bar{3}$ ), which is formed from a periodic spatial repetition of  $\text{Mo}_6\text{X}_8$  groups. Each group includes within itself an  $\text{Mo}_6$  octahedron (a cluster) whose vertices protrude slightly beyond the face centers of a distorted  $\text{X}_8$  cube.

The  $\text{Mo}_6\text{X}_8$  groups are disposed in space in such a way that open zig-zag channels are formed along all three rhombohedral axes, within which the A atoms are located.

It has been pointed out before that the temperature dependences of the specific heat  $C_p(T)$ ,<sup>2,9</sup> upper critical field  $H_{c2}(T)$ ,<sup>1,10</sup> critical current density  $J_c(T)$ ,<sup>11,12</sup> and electrical resistivity  $\rho(T)$ ,<sup>2,13</sup> of ternary molybdenum chalcogenides (TMCh) are to a considerable extent unusual and have many features which are to a considerable extent determined by the element A. For example, a linear increase of  $\rho(T)$  is observed in the temperature range from  $T = 1.1 T_c$  to 40–50 K for the compounds  $\text{SnMo}_6\text{S}_8$  and  $\text{PbMo}_6\text{S}_8$ , which seems to be produced by scattering of conduction electrons by low-frequency optic phonons connected with oscillations of atoms A. As a rule, the experimental  $H_{c2}(T)$  dependences<sup>6,10</sup> at low temperatures for a number of TMCh's go higher than predicted by the WHH theory<sup>14</sup> for isotropic superconductors with small mean free path  $l \ll \xi_0$  ( $\xi_0$  is the correlation length).

It was of interest to elucidate how the absence of element A is reflected in the  $H_{c2}(T)$  and  $\rho(T)$  dependences for a number of halogen-substituted molybdenum chalcogenides such as  $\text{Mo}_6\text{S}_6\text{I}_2$ ,  $\text{Mo}_6\text{Se}_6\text{I}_2$ ,  $\text{Mo}_6\text{Se}_6\text{Br}_2$  and  $\text{Mo}_6\text{S}_6\text{Br}_2$ .

## THE EXPERIMENTS

Powders of molybdenum, the chalcogen (S or Se) and the molybdenum dihalide ( $\text{MoI}_2$  or  $\text{MoBr}_2$ ) were taken in the appropriate proportions in order to prepare the specimens and thoroughly mixed, pressed into pellets and sealed into evacuated quartz ampoules. The ampoules were maintained in an oven at a temperature of 1050–1150 °C for 24 h. The powdered compounds obtained were again thoroughly mixed and pressed into pellets. A homogenizing anneal of the specimens was carried out in a pure helium atmosphere at 950–100 °C for 24 h.

An x-ray monitoring of the phase composition showed that the specimens obtained are practically single-phase with traces of  $\text{Mo}_2\text{S}_3$ ,  $\text{MoSe}_2$  and, apparently,  $\text{MoI}_2$ . The content of extraneous phases in the halogen-substituted specimens is appreciably less than is usual in TMCh specimens.

The transition of specimens from the superconducting to normal state was recorded by the change in resistance, using a standard four-probe potentiometric method. The specimen dimensions were usually  $1 \times 1 \times 8$  mm with the current leads affixed with a conducting silver paste.

As in earlier experiments,<sup>6,15</sup>  $H_{c2}(T)$  was measured at low temperatures in pulsed magnetic fields. The specimen was oriented perpendicular to the field and a 5 to 8 kHz alternating current of amplitude 0.1 to 1 mA was passed through it. The background signal arising at the potential contacts in the absence of a magnetic field was balanced out. The transition from the superconducting to the normal state under the action of a pulsed magnetic field of duration 0.01 sec produced an out-of-balance signal which was amplified and fed to an oscilloscope with memory. Only those values of the out-of-balance signal which corresponded to the peak values of the pulsed field were used to construct the  $\rho(H)|_{T=\text{const}}$  curves. For sufficiently good thermal contact between the specimen and the refrigerant, the influence of

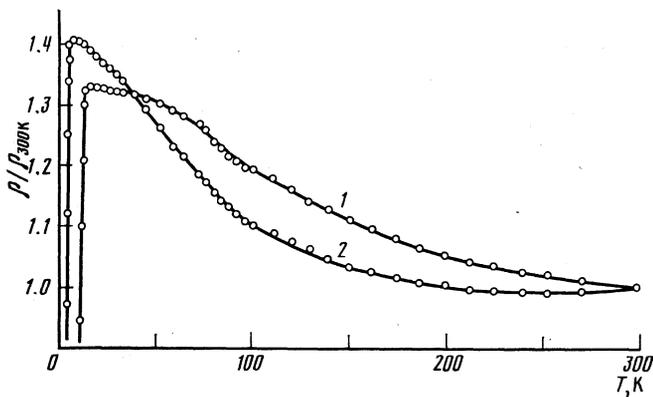


FIG. 1. Temperature dependence of the electrical resistivity of halogen-substituted molybdenum chalcogenides with Chevrel-phase structure: 1— $\text{Mo}_6\text{S}_6\text{I}_2$ , 2— $\text{Mo}_6\text{Se}_6\text{I}_2$ .

various extraneous factors could be eliminated by this procedure, in particular effects of overheating on the shape.<sup>15</sup>

## RESULTS

Compared with TMCh's such as  $\text{PbMo}_6\text{S}_8$  and  $\text{SnMo}_6\text{S}_8$ , pressed and annealed specimens of halogen-substituted compounds have appreciably less mechanical strength and considerably higher resistivity  $\rho_n = 5 \times 10^{-3}$  to  $2 \times 10^{-2} \Omega \cdot \text{cm}$ . Estimates of the critical current density at  $T = 2 \text{ K}$  and  $H = 10 \text{ kOe}$  gave for halogen-substituted specimens a value  $J_c < 10^3 \text{ A} \cdot \text{cm}^{-2}$ , which is an order of magnitude less than for ternary molybdenum sulfides containing lead.<sup>12</sup>

It was found in studies of the temperature dependences  $\rho(T)$  that the resistivity of halogen-substituted specimens (Fig. 1) increases by 30 to 40% before the superconducting transition. The temperature variation for the  $\text{Mo}_6\text{S}_6\text{I}_2$  specimen (curve 1) at high temperatures ( $100 \text{ K} < T < 300 \text{ K}$ ) can be approximated sufficiently well by a dependence of the form  $\rho_T(T) = A_1 \rho_{300\text{K}} \exp(\Delta_1/kT)$ , where  $A_1 = 0.92$  and  $\Delta_1 = 2.4 \times 10^{-3} \text{ eV}$ . The experimental results for the temperature range 60 to 200 K for  $\text{Mo}_6\text{Se}_6\text{I}_2$  (curve 2) can be approximated by a similar expression, but with slightly different coefficients  $A_2 = 0.91$  and  $\Delta_2 = 1.5 \times 10^{-3} \text{ eV}$ . The experimental values of  $\rho(T)$  depart from the exponential dependence for both specimens at low temperatures.<sup>1)</sup> The temperatures,  $T_1 \approx 90 \text{ K}$  and  $T_2 \approx 60 \text{ K}$ , at which the departure of  $\rho(T)$  from the exponential law starts to exceed 1% (Fig. 2) are in just about the same ratio as the characteristic temperatures  $\Theta_D$ , for the compounds  $\text{Mo}_6\text{S}_6\text{I}_2$  and  $\text{Mo}_6\text{Se}_6\text{I}_2$  (Ref. 16) determined from specific heat measurements.

The superconducting transition curves for pseudo-binary halogen-substituted compounds are more spread out than for ternary molybdenum chalcogenides, and in addition they broaden by an additional amount under the action of an external magnetic field. For example, in a  $\text{Mo}_6\text{Se}_6\text{I}_2$  specimen the half width of the transition  $0.5 \Delta T$ , corresponding to a change in resistivity from 0.9 to  $0.5 \rho_n$ , increases from 0.5 K (in zero field) to 2 K in a field  $H = 120 \text{ kOe}$ . Since  $\Delta T$  changes from specimen to specimen, it can be supposed that the broadening of the transition curve in halo-

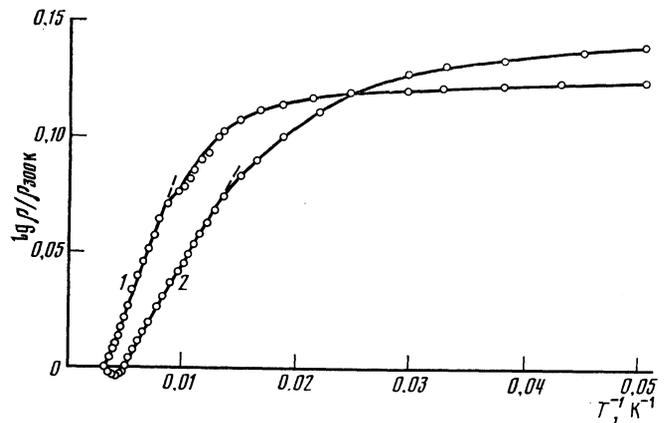


FIG. 2. Departure of the experimental values of the reduced electrical resistivity  $\rho/\rho_{300\text{K}}$  from an exponential dependence (dashed lines) for: 1— $\text{Mo}_6\text{S}_6\text{I}_2$  and 2— $\text{Mo}_6\text{Se}_6\text{I}_2$ .

gen-substituted systems can be produced by variations in composition over the specimen volume and the strong dependence of critical parameters on the halogen content. This refers mainly to halogen-substituted molybdenum sulfides which have a very narrow region of homogeneity near  $x = 2$  (Ref. 8). It is also possible that the extra spreading of the curves can be due to an anisotropy of  $H_{c2}$  in these systems as a result of preferential location of the halogen atoms in  $2C$  positions, i.e., along the three-fold  $C$  axis.<sup>17</sup>

Because of the relatively larger values of  $\Delta T$  for halogen-substituted molybdenum chalcogenides, the values of  $T_c$  were determined from the start of the transition curve, in the same way as in earlier work.<sup>8,16,18</sup> If the upper critical field is treated in a similar way, the value of  $H_{c2}(T)$  can be determined as the field in which the resistivity of a specimen at a given temperature  $T < T_c$  amounts to  $0.9 \rho_n$  ( $\rho_n = \rho_{\text{max}}$ ). The values of  $H_{c2}$  determined in this way for a number of halogen-substituted molybdenum sulfides and selenides are shown in Fig. 3. For comparison, the  $H_{c2}(T)$  dependence based on the center of the transition curves ( $0.5 \rho_n$ ) for the  $\text{Mo}_6\text{S}_6\text{I}_2$  specimen is shown in the same figure by the dotted line. The linear dependence of  $H_{c2}(T)$  on  $T - T_c$  down to 1.7 K is noticeable. The linear nature of the  $H_{c2}$  dependence is preserved if the values of  $H_{c2}(T)$  are chosen at the  $0.5 \rho_n$  level.

Similar  $H_{c2}(T)$  dependences were found earlier, for example for the strongly anisotropic niobium dichalcogenide  $2\text{H-NbSe}_2$  (Ref. 19) and a number of other superconducting compounds such as  $\text{Na}_x\text{MoS}_2$  (Ref. 20) and  $\text{Nb}_3\text{Pt}$  (Ref. 21).

## DISCUSSION OF THE RESULTS

The distinguishing feature of the halogen-substituted molybdenum chalcogenides which we have studied is the temperature dependence of the resistivity, which is not quite usual for superconductors. Among molybdenum chalcogenides with the Chevrel-phase structure the semiconductor-like resistivity behavior was observed earlier in, for example, specimens of the  $\text{Mo}_2\text{Re}_4\text{X}_{8-y}\text{X}'_y$  system.<sup>22</sup> However, superconductivity was not found in specimens of this system

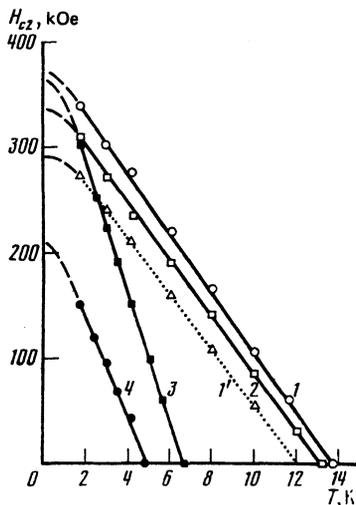


FIG. 3. Temperature dependence of the upper critical field  $H_{c2}$  for: 1— $\text{Mo}_6\text{S}_6\text{I}_2$ , 2— $\text{Mo}_6\text{S}_6\text{Br}_2$ , 3— $\text{Mo}_6\text{Se}_6\text{Br}_2$ , 4— $\text{Mo}_6\text{Se}_6\text{I}_2$ , corresponding to the establishment of a resistivity up to  $0.9\rho_n$  ( $\rho_n$  is the resistivity of the specimens in the normal state near the superconducting transition; in the present case  $\rho_n = \rho_{\text{max}}$ ). The dotted line (1') shows the  $H_{c2}(T)$  dependence derived from the middle of the transition curves ( $0.5\rho_n$ ) for  $\text{Mo}_6\text{S}_6\text{I}_2$ .

having the semiconductor-like resistivity variation, while on the other hand the resistivity of superconducting specimens ( $\text{Mo}_x\text{Re}_2\text{Te}_8$ ) decreased with decreasing temperature, i.e. the  $\rho(T)$  dependence was of the usual metallic form. The increasing resistivity<sup>22</sup> was explained on the basis of band calculations,<sup>23,24</sup> from which it followed that the energy dependence of the electronic density of states  $N(E)$  has an energy gap near the Fermi level,  $E_F$ , for compounds with the Chevrel-phase structure. The center of this gap corresponds<sup>23</sup> to 24 valence electrons per  $\text{Mo}_6$  cluster.

An increase in resistivity with decreasing temperature and a transition to the superconducting state was observed<sup>25</sup> in studying the compound  $\text{Sn}_{0.12}\text{Eu}_{1.08}\text{Mo}_6\text{S}_8$  under a pressure of 8 to 12 kbar. In the opinion of the authors, the negative derivative  $\partial\rho/\partial T$  at low temperatures could be produced by a reduction in the number of current carriers with temperature due to a small ( $\sim 50$  K) energy gap, the origin of which is connected with the instability of these systems relative to a metal-semiconductor transition. We should point out that closeness to the MS phase transition also explains the increase in resistivity with decreasing temperature in the oxygen-containing  $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$  compound.<sup>26</sup>

In considering the  $\rho(T)$  dependences found for halogen-substituted molybdenum chalcogenides, we must first look at the possible precipitation of semiconductor phases at the grain boundaries. One such phase could in principle be the compound  $\text{MoS}_2$ , the lines of which occur quite often in x-ray diffraction pictures of superconducting molybdenum chalcogenides. However, the resistivity would in that case be unlikely to go to zero as occurs for the compounds studied. It should also be noted that even the presence of 10 to 15 mol%  $\text{MoS}_2$  in  $\text{A}_x\text{Mo}_6\text{S}_8$  ( $\text{A} = \text{Cu}, \text{Sn}, \text{Pb}$ ) specimens has practically no effect on the metallic nature of the conductivity of these systems.<sup>12</sup> Although it is impossible to rule out completely the influence of semiconductor phases on the  $\rho(T)$  depen-

dence, such an explanation is unlikely, especially if the high degree of single-phase character of the specimens studied is taken into account.

In turning to consideration of other possible reasons for the semiconductor nature of the  $\rho(T)$  dependence, one must first note that replacing a chalcogen by a halogen leads to an increase in the volume,  $V_H$ , of the hexagonal cell of  $\text{Mo}_6\text{X}_6\text{X}'_2$  compounds by 20 to 24  $\text{\AA}^3$  for  $\text{X}' = \text{Br}$  and by 42–44  $\text{\AA}^3$  for  $\text{X}' = \text{I}$ . The mean distance between Mo atoms in a  $\text{Mo}_6$  cluster then decreases, while the distance between Mo atoms of neighboring clusters increases.<sup>17</sup> It is reasonable to assume that with an increase in intercluster distance, the  $\psi$ -function overlap from neighboring clusters will decrease, leading in turn to stronger localization of the conduction electrons inside the clusters. The stronger localization in the clusters can cause the so-called covalent instability to arise<sup>27</sup> especially if we take account of the number of valence electrons on a cluster  $n_{ec}$  in halogen-substituted molybdenum selenides being approximately 24, which is exactly what is required for the formation of the maximum number (12) of covalent bonds.<sup>28</sup> Although it is accompanied by an increase in the mean square of the electron-ion interaction matrix element ( $I^2$ ), the growth in the probability of formation of covalent bonds in the clusters can nevertheless represent competition for superconductivity in that instead of Cooper pairs 'condensed' in momentum space, valence pairs 'condensed' in coordinate space will form in this case.<sup>29,30</sup>

Judging by the existing data, the optimum ratio of the magnitudes of  $N(E_F)$  and  $I^2$  is reached for molybdenum chalcogenides with the Chevrel-phase structure near the metal-semiconductor phase transition, which corresponds to  $n_{ec} = 22$ –23. The absence of the third component  $A$  in halogen-substituted specimens apparently provides specially favorable conditions for MS-instability to arise in them. It is not impossible that a MS-instability can show up in a negative temperature coefficient of resistivity for  $\text{Mo}_6\text{S}_6\text{I}_2$  and  $\text{Mo}_6\text{Se}_6\text{I}_2$ . Replacing sulfur by selenium in the compound  $\text{Mo}_6\text{S}_6\text{I}_2$ , leading to an increase of 1.5 in  $n_{ec}$  (Ref. 8) is accompanied by a reduction in  $T_c$  from 13.7 to 4.8 K. (It is interesting that almost the same reduction in  $T_c$ , from 13.5 to 5 K on increasing  $n_{ec}$  by 1.2 is observed on increasing the copper content in the system  $\text{Cu}_x\text{Mo}_6\text{S}_6\text{I}_2$  ( $0 < x < 1.2$ ) (Ref. 18).

The weakening of the inter-cluster bonds as a result of replacing the chalcogen by halogen leads to a softening of their phonon spectrum, to which results of specific heat measurements bear witness.<sup>16</sup> The role of low-frequency phonons in electron scattering processes can then grow with decreasing temperature, which is reflected in turn in the derivative  $\partial\rho/\partial T$ . Nothing of this sort is observed in the Pd-H system, for example, where the contribution to  $\rho(T)$  from optic phonons has a maximum at  $T \approx 100$  K.<sup>31</sup>

The increase in  $\rho$  with decreasing temperature could also result from a reduction in the number of current carriers if it is assumed that their transfer from the valence to the conduction band is connected with, for example, inter-cluster oscillations. As follows from specific heat measurements,<sup>16</sup> the frequency of such oscillations is fairly high and they could be 'frozen-out' on reducing the temperature,

which would lead in turn to a reduction in the number of current carriers in the conduction band. This effect will, however, evidently be small.

The increasing localization of conduction electrons in a cluster on partial replacement of selenium by a halogen in the compound  $\text{Mo}_6\text{Se}_8$  should, according to calculations,<sup>32</sup> be accompanied by an increase in  $\partial H_{c2}/\partial T$ . Comparison of results for  $\text{Mo}_6\text{Se}_8$  (with  $\partial H_{c2}/\partial T = 21 - 23$  kOe/K, Ref. 33) and for halogen-substituted molybdenum selenide (Fig. 3) shows that in fact the derivative  $\partial H_{c2}/\partial T$  then increases 2 to 2.5 times. Substituting sulfur for selenium in halogen-substituted molybdenum chalcogenides should then lead to a reduction in  $\partial H_{c2}/\partial T$ , which agrees with experiment (Fig. 3).

The increase in  $T_c$  on substituting sulfur for selenium is apparently connected with an increase<sup>2</sup> in the density of states at the Fermi surface  $N(E_F)$ , which does not agree with results of calculations of the band structure of Chevrel-phases.<sup>23</sup>

As can be seen from Fig. 3 for halogen-substituted molybdenum chalcogenides, as for some other compounds,<sup>19-21</sup> a positive deviation from the  $H_{c2}(T)$  dependence is observed at temperatures  $T < 0.5T_c$  from the theoretical WHH curves<sup>14</sup> calculated on the assumption of weak binding for superconductors with small mean free path  $l \ll \xi_0$ . It follows from many theoretical papers that such a deviation can be ascribed to effects of strong binding,<sup>35</sup> anisotropy of the Fermi surface,<sup>36,37</sup> localization of conduction electrons within clusters<sup>32</sup> etc. Numerical calculations<sup>35</sup> show, however, that the effect of electron-phonon interaction on the form of theoretical WHH curves is relatively small and in most cases the corrections to the value of  $H_{c2}(T)$  do not exceed 5%.

As already noted,  $H_{c2}(T)$  dependences similar in shape to those observed in halogen-substituted niobium chalcogenides (Fig. 3) were found earlier in the layered niobium dichalcogenide  $2\text{H-NbSe}_2$  bond (Ref. 19) and in the compounds  $\text{Nb}_y\text{Pt}_{100-y}$  ( $y = 73$  to  $78$ , (Ref. 21) with the  $A$ -15 structure. Calculations<sup>36,37</sup> showed that the almost linear  $H_{c2}(T)$  dependences for  $2\text{H-NbSe}_2$  and  $\text{Nb}_y\text{Pt}_{100-y}$  can be related to the feature of their crystal structure which shows up in, for example, the Fermi surface anisotropy. Considering that the halogen atoms in halogen-substituted molybdenum sulfides lie preferentially along the  $C$ -axis,<sup>17</sup> it can be suggested that the influence of anisotropy on the shape of the  $H_{c2}(T)$  dependences is greater in them than in the selenides.

On the other hand, as noted above, molybdenum selenides have a higher degree of localization of conduction electrons in the clusters which might also affect the shape of the  $H_{c2}(T)$  curves.<sup>32</sup>

These two factors, i.e., anisotropy of the Fermi surface and localization of conduction electrons in the clusters, by reinforcing one-another evidently lead to almost linear  $H_{c2}(T)$  dependences for halogen-substituted molybdenum sulfides and selenides.

In conclusion the authors thank E. P. Khlybov for help in carrying out the x-ray measurements.

<sup>1</sup>It cannot be ruled out that a structural transformation could take place at low temperatures.

<sup>2</sup>We note that an increase in  $\partial H_{c2}/\partial T$ , accompanied by a reduction in  $T_c$  and  $N(E_F)$ , and also by a change in sign of the temperature coefficient of electrical resistivity was observed by us earlier when studying  $\text{V}_{1-x}\text{Al}_x$  solid solutions.<sup>34</sup>

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