Superconductivity of $Tb_2Mo_3Si_4$

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A new antiferromagnetic superconductor, the $Tb_2Mo_3Si_4$ compound, has been discovered. In this compound the transition to the superconducting state occurs at a temperature $T_c = 0.5–1.2$ K, which is considerably lower than the temperature of the magnetic transition $T_N \approx 20$ K.

1. During the past decade, progress in the synthesis of binary and ternary compounds based on rare-earth compounds and actinides has led to the discovery of "exotic" superconductors—superconducting systems with heavy fermions,[1] magnetic superconductors,[2,3] and Kondo magnetic lattices which become superconducting in a strong magnetic field.[4] The study of "exotic" superconductors is of considerable interest in determining whether superconductivity and magnetism can coexist. The superconducting transition temperature T_c of all magnetic superconductors that have so far been identified is generally slightly higher than the magnetic transition temperature. The only exception are the compounds[5] $Ho(Ir_xRh_{1-x})_4B_4$ and systems[6] Y_9Co_7 and Cr-Re (Ref. 7). The Néel temperature T_N of $Ho(Ir_xRh_{1-x})_4B_4$, however, is always higher than T_c by one degree and the magnetism of Y_9Co_7 and Cr-Re has a band nature. At the same time, among the superconductors in which the local magnetism coexists with superconductivity, there are no known compounds in which the temperature T_N is appreciably higher than the superconducting transition temperature. In this letter we report the discovery of a "superconducting magnetic material"— the $Tb_2Mo_3Si_4$ compound in which $T_N \simeq 20\ K \gg T_c \simeq 0.5–1.2$ K.

2. At temperatures 4.2–300 K we have measured the temperature dependences of the magnetic susceptibility $\chi(T)$, of the resistivity $\rho(T)$, and of the heat capacity $C(T)$ of polycrystalline samples of the compound $Tb_2Mo_3Si_4$ which belongs to a structure of the type[8] $U_2Mo_3Si_4$ (Fig. 1). The magnetic susceptibility was measured with a vibrational magnetometer in a field $H = 3$ kOe. To determine the temperature evolution of $C(T)$ and $\rho(T)$, we used an automatically controlled apparatus that was connected to a D3-28 computer.

The high-temperature parts of the $\chi(T)$ curves [Fig. 1(a)] are well described by the Curie-Weiss law with a constant $\theta = -30$ K and effective magnetic moment $\mu_{\rm eff} = 9.66\ \mu_B$. These data, in agreement with the results of Ref. 8, show that the magnetic properties of $Tb_2Mo_3Si_4$ are determined by the local moments of terbium ions. At temperatures $T = T_N \simeq 20$ K the $\chi(T)$ and $C(T)$ curves exhibit a sharp peak [Figs. 1(a) and 1(c)] which corresponds to a transition from the paramagnetic state to the antiferromagnetic state. This transition is also accompanied by a break in the $\rho(T)$ curve [Fig. 1(b)].

The magnetic susceptibility $\chi(T)$ and the resistivity $\rho(T)$ of $Tb_2Mo_3Si_4$ in the antiferromagnetic phase were studied down to $T = 0.05$ K (Figs. 2 and 3). At

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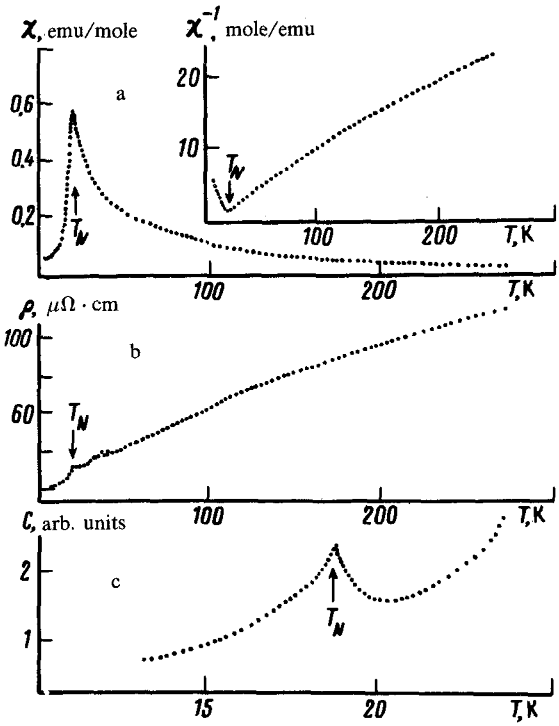

FIG. 1. Temperature dependence of the magnetic susceptibility (a), of the resistivity (b), and of the heat capacity (c) of the compound $Tb_2Mo_3Si_4$. Inset—Temperature dependence of the reciprocal of the susceptibility, $\chi^{-1}(T)$.

$T = 0.05$–2 K the magnetic susceptibility $\chi(T)$ was measured in a field $H = 0.5$ Oe with the help of the SQUID. A lowering of the temperature causes a diamagnetic anomaly in $\chi(T)$ (Fig. 2) and causes $\rho(T)$ to vanish (Fig. 3). These features of $\chi(T)$

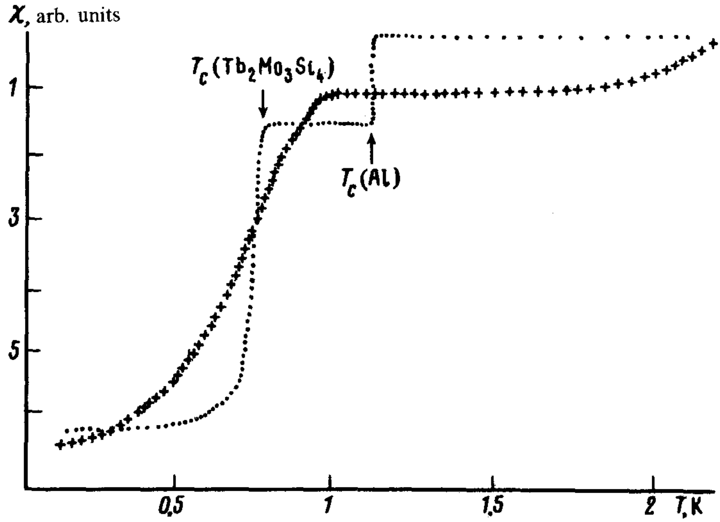

FIG. 2. Temperature dependence of the magnetic susceptibility $\chi(T)$ of ($Tb_2Mo_3Si_4$ + Al) (●) and of the annealed $Tb_2Mo_3Si_4$ sample (+).

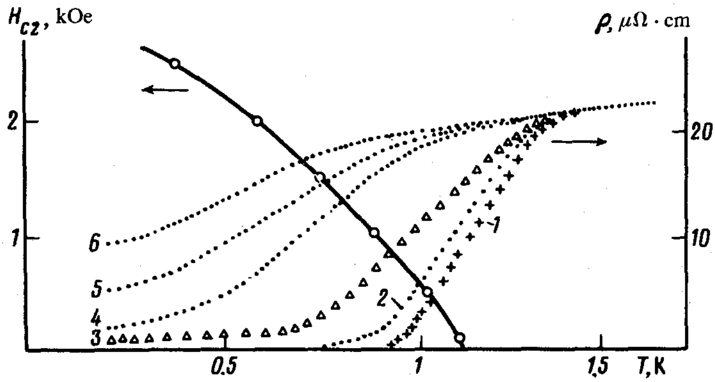

FIG. 3. Temperature dependence of the resistivity $\rho(T)$ at extremely low temperatures. 1—$H = 0$ kOe; 2—0.25 kOe; 3—0.6 kOe; 4—1.5 kOe; 5—2 kOe; 6—2.5 kOe. The curve $H_{c2}(T_c)$(O) corresponds to the midpoints of the superconducting transitions.

and $\rho(T)$, which correspond to a transition to the superconducting state, have been observed in various $Tb_2Mo_3Si_4$ samples in the temperature interval $T = T_c \simeq 0.5$–1.2 K.

3. Let us determine first of all whether the superconductivity of $Tb_2Mo_3Si_4$ is an intrinsic or extrinsic superconductivity. X-ray phase analysis shows that the superconducting impurities Mo ($T_c \simeq 0.9$ K) (Ref. 9), Mo_3Si ($T_c \simeq 1.4$ K) (Ref. 9) and $MoSi_2$ ($T_c < 0.3$ K) (Ref. 9) are absent within 1–2% in all six $Tb_2Mo_3Si_4$ samples studied. To determine the magnitude of the Meissner effect, we placed next to the $Tb_2Mo_3Si_4$ sample a 99.99% pure aluminum sample approximately one-third of the size of the sample studied. We found (Fig. 2) that the diamagnetic jump in χ due to the superconducting transition in $Tb_2Mo_3Si_4$ is roughly three times as large as that in aluminum; i.e., at $T \ll T_c$ virtually all of the magnetic field is eliminated from the $Tb_2Mo_3Si_4$ sample. This experimental result and also the data on the x-ray phase analysis show that the superconductivity of a $Tb_2Mo_3Si_4$ compound is an intrinsic superconductivity. The superconducting transition temperature varies from one sample to the next in the range $T_c \simeq 0.5$–1.2 K. The value of T_c depends on the heat treatment of the samples. For example, annealing of samples at a temperature of 800 °C for 700 h increases T_c from 0.7 K to 1.2 K. We find that the higher T_c correspond to a more diffuse superconducting transition.

Figure 3 shows the suppression of superconductivity of $Tb_2Mo_3Si_4$ in a magnetic field. The upper critical field of $Tb_2Mo_3Si_4$ is $H_{c2}(0) \simeq 3$ kOe.

We can thus conclude that at $T = T_c \simeq 0.5$–1.2 K the compound $Tb_2Mo_3Si_4$ undergoes a transition from the antiferromagnetic state to the superconducting state. A distinguishing feature of $Tb_2Mo_3Si_4$ is that, in contrast with all heretofore known magnetic superconductors, the superconducting transition temperature of this compound is considerably lower than the Néel temperature. For superconductors with $T_c \ll T_N$, which may be called "superconducting magnetic materials," Buzdin and Bulaevskiĭ[10] predicted several unusual effects, in particular, a very strong dependence of T_c on the mean free path. Further study of the superconducting magnetic compound $Tb_2Mo_3Si_4$ is clearly of considerable interest.

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