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RESISTIVITY OF TERNARY CHEVREL-PHASE SUPERCONDUCTORS

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Résumé.- Nous avons mesuré la résistivité entre 4.2 K et 400 K d'une série de phases de Chevrel qui sont des composés supraconducteurs. Tous montrent une courbure très négative ($d^2\rho/dT^2$) au-dessus d'environ 60 K, semblable à ce qu'on observe dans les composés A-15. Les résistivités à la température ambiante sont élevées, de l'ordre de $10^{-3} \Omega \text{ cm}$.

Abstract. We have measured the resistivities of a series of superconducting Chevrel-phase compounds from 4.2 K to 400 K. All show strong negative curvature $d^2\rho/dT^2$ above about 60 K, similar to that observed in the A-15 compounds. Room temperature resistivities are high, in the $10^{-3} \Omega \text{ cm}$ range.

Recent interest has developed in the deviations from linearity in the resistivity $\rho(T)$ of transition-metal superconductors. In particular a number of the A-15 compounds exhibit a negative curvature $\frac{d^2\rho}{dT^2}$ at high temperatures, including Nb_3Sn , Nb_3Sb , V_3Si , and V_3Ge . The data do not appear to approach a linear temperature dependence at high T , as expected from the Bloch-Grüneisen Theory. Woodard and Cody /1/ and Williamson and Milewits /2/ have pointed out that an exponential term $e^{-T_0/T}$ can be used to fit the data; Fisk and Webb /3/ and Wiesmann et al. /4/ have suggested that $\rho(T)$ may "saturate" at high T when the electron mean free path becomes comparable to the lattice spacing. In the latter case the lattice spacing imposes a lower bound for the mean free path and a maximum for ρ .

We have measured the resistivity in a series of superconducting ternary Chevrel phase compounds $\text{M}_{1.2}\text{Mo}_6\text{X}_8$, where M is a metal and X is either S or Se , and find a similar negative curvature in $\rho(T)$ in these materials. Measurements were made on compressed, sintered pellets reacted in an inert gas atmosphere, using a standard four probe DC method. To date, single crystals of these compounds large enough for resistivity measurements have not been synthesized.

Representative results of ρ vs. T for the non-magnetic compound $\text{Pb}_{1.2}\text{Mo}_6\text{Se}_8$ are shown in figure 1. The results for $\text{Pb}_{1.2}\text{Mo}_6\text{S}_8$, $\text{Sn}_{1.2}\text{Mo}_6\text{S}_8$, $\text{Sn}_{1.2}\text{Mo}_6\text{Se}_8$, $\text{La}_{1.2}\text{Mo}_6\text{S}_8$ and $\text{La}_{1.2}\text{Mo}_6\text{Se}_8$ are qualitatively similar, as are compounds where M is one of the rare earth elements Gd , Dy , or Sm . The resistivities for all these compounds show negative cur-

vature above about 60 K. Resistivity ratios ρ_{300}/ρ_{15} are typically about 2.5, but range as high as 10. The data can be fit above 40 K within a rms deviation of 1 to 2 % by either the saturation resistivity model of Wiesman et al. or by an exponential dependence on $1/T$. The model of Wiesmann et al. assumes that the total resistivity of the system is given by the electron-phonon resistivity $\rho_{\text{ideal}}(T)$ shunted by the limiting resistivity ρ_{max} :

$$\frac{1}{\rho} = \frac{1}{\rho_{\text{ideal}}(T)} + \frac{1}{\rho_{\text{max}}}$$

Here ρ_{ideal} is assumed to have the form $\gamma + \rho_1 T$, where γ and ρ_1 are constants. For the compounds studied, $\rho_{\text{max}}/\rho_{300}$ is typically equal to about 1.5, suggesting that there is relatively little further change in resistivity above room temperature.

Fig. 1 : Resistivity of $\text{Pb}_{1.2}\text{Mo}_6\text{Se}_8$ ($\rho_{300} = 1.8 \text{ m}\Omega \text{ cm}$) representative of the compounds studied. curvature is negative above about 60 K.

Fitted to an empirical exponential relation of the form $\rho(T) : a + bT + de^{-T_0/T}$, these compounds have discussed various physical interpretations of this thermally activated resistivity.

The room temperature resistivities of all ternary compounds studied were high, typically 2-10 m Ω - cm. This value is somewhat sensitive to the method of sample preparation and grain size, but in all cases it was found to exceed 1.8 m Ω - cm. These values are roughly ten times higher than those found in A-15 superconductors /3/. It is not clear how much of the measured resistivity is due to contact resistance between grains in the compressed samples. However, AC resistivity measurements on two samples at 10 and 100 kHz at 300 K and 77 K were identical to the DC results, suggesting that the impedance between grains is resistive in character, and probably not large in comparison with the bulk resistivity. Residual resistivity below T_c was unobservably small. At low temperatures above T_c , the measured resistivities were in good agreement with estimates obtained from electron mean free paths derived from critical field measurements /5/.

C.Y. Fong /6/ has suggested that the large values of ρ_{300} are an intrinsic bulk property of the Chevrel phase, due to the channeling effect of the Mo-Mo bonds on the conduction electron motion. Because the conduction d orbitals of adjacent Mo atoms are canted with respect to each other, the electrons must scatter through an angle of about 70° to move from one atom to the next. This results in a factor of $\cos(70^\circ) \approx \frac{1}{3}$ in the matrix element used to calculate the collision term in the transport equation. Aside from this, the situation is similar to that found in A-15 compounds, where the conduction is along the d-orbitals of the transition metal ions, which are aligned in linear chains. In this model the Chevrel phase conductivity would be lower than A-15 compound conductivity by a factor of $\cos^2(70^\circ) \approx \frac{1}{10}$, in agreement with our observations.

We are indebted to Dr. R.N. Shelton for supplying us with the Chevrel-phase compounds.

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