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Pressure dependence of the charge-density wave of sodium molybdenum purple bronze

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Abstract. The electrical resistivities of the low-dimensional conductors $(\text{Na}_{1-x}\text{Li}_x)_{0.9}\text{Mo}_6\text{O}_{17}$ ($x = 0, 0.2$) have been measured between 4.2 and 300 K under hydrostatic pressure. The magnitude of the resistivity anomaly associated with the charge-density wave (CDW) transition became smaller with increasing pressure, while the transition temperature itself was found to be enhanced by the pressure in contrast to the simple cases of the inorganic low-dimensional conductors.

1. Introduction

In this paper we report the pressure effect on the charge-density wave (CDW) state of sodium molybdenum purple bronze studied by electrical resistivity measurements. Sodium molybdenum oxide ($\text{Na}_{0.9}\text{Mo}_6\text{O}_{17}$) is one of the compound series of the so-called molybdenum purple bronzes described by the general formula $\text{A}_{0.9}\text{Mo}_6\text{O}_{17}$ ($\text{A}=\text{Li}, \text{Na}$ and K). The crystal structure of the purple bronzes has slabs with the ReO_3 -type cubic arrangement of MoO_6 octahedra, which are connected with the MoO_4 tetrahedra. The A atoms sit in the large space formed by the arrangement of the tetrahedra between the slabs (Vincent *et al* 1983, Onoda *et al* 1987a, b). The crystal structure explains the low dimensionality of the transport properties. As a result of this low dimensionality, $\text{K}_{0.9}\text{Mo}_6\text{O}_{17}$ has been found to undergo the CDW transition at a temperature slightly above 100 K (Buder *et al* 1982), and $\text{Na}_{0.9}\text{Mo}_6\text{O}_{17}$ at about 80 K (Schlenker *et al* 1985). On the other hand, the behaviour of the electrical resistivity of $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$ is quite different from those of $\text{K}_{0.9}\text{Mo}_6\text{O}_{17}$ and $\text{Na}_{0.9}\text{Mo}_6\text{O}_{17}$. It shows a metallic temperature dependence at high temperatures ($T > 25$ K) and a very rapid increase with decreasing temperature, followed by the superconducting transition at about 2 K (Greenblatt *et al* 1984). The increase in the resistivity was thought, at the first stage of the studies, to be caused by the formation of the CDW. It has been pointed out recently, however, that the increase is not due to the formation of the CDW but to an electron localization effect (Matsuda *et al* 1986, Sato *et al* 1987).

Attempts have been made to observe the competition between the CDW state and the superconducting state using the solid solutions of the purple bronzes (Matsuda *et al*

1986). These have been unsuccessful because the crystal structure changes discontinuously as a function of the concentration. The CDW transition temperature or the superconducting temperature is almost independent of the concentration in each phase. This means that the crystal structure of $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$ is fairly different in detail from those of other purple bronzes as confirmed by structure analyses (Onoda *et al* 1987a, b).

In the present study, the pressure dependence of the CDW of sodium purple bronze ($(\text{Na}_{1-x}\text{Li}_x)_{0.9}\text{Mo}_6\text{O}_{17}$ ($x = 0, 0.2$)) was found to be similar to that of KCP, as for the transition temperature T_c . The magnitude of the resistivity anomaly below T_c , on the other hand, had a dependence similar to that of NbSe_3 .

2. Experiment

Single crystals of $(\text{Na}_{1-x}\text{Li}_x)_{0.9}\text{Mo}_6\text{O}_{17}$ ($x = 0, 0.2$) were prepared by electrolysis as described by Wold *et al* (1964). The molar ratio of Na_2MoO_4 to Li_2MoO_4 in the starting mixture is used as the nominal value of x . The crystals are purple and have platelet shapes along the monoclinic b axis.

The electrical resistivities R were measured between 4.2 and 300 K by the standard four-terminal method under hydrostatic pressure. The electrodes were attached to the specimens with silver paint. The current direction was almost parallel to the b axis. The hydrostatic pressure was generated in a clamped piston-cylinder apparatus with a fluid pressure medium of a 1:1 mixture of n -pentane and isoamylalcohol, as described by Chu *et al* (1978). The pressure was determined with a superconducting Pb manometer in the cell. The temperature was measured with a calibrated Ge thermometer (CryoCal) and a Pt resistance thermometer.

3. Results and discussion

Figures 1 and 2 illustrate the temperature dependence of the normalized electrical resistivities of $(\text{Na}_{0.8}\text{Li}_{0.2})_{0.9}\text{Mo}_6\text{O}_{17}$ and $\text{Na}_{0.9}\text{Mo}_6\text{O}_{17}$, respectively, under various hydrostatic pressures. The resistivities are normalized by the values at 300 K. In both cases, the resistivities show a metallic temperature dependence at high temperatures under the pressures measured. They show anomalies caused by the formation of the CDW below T_c . The values of T_c were determined as the temperatures where the resistivity becomes a minimum. They are indicated by the arrows in the figures. The resistivities at ambient pressure show no exponential temperature dependence below T_c . This means that the nesting of the Fermi surface is insufficient and a part of the Fermi surface survives below T_c . A reduction in the magnitude of the resistivity anomaly can be seen with increasing pressure as in usual inorganic low-dimensional conductors. In the case of $\text{Na}_{0.9}\text{Mo}_6\text{O}_{17}$ the resistivity restores metallic behaviour at low temperatures under higher pressures. The behaviour is similar to that of NbSe_3 at ambient pressure (Briggs *et al* 1980). The phenomenon is explained by the fact that the Fermi surface area lost by the CDW gap formation decreases with increasing pressure. In NbSe_3 , the superconducting transition takes place at a higher pressure. In the case of $\text{Na}_{0.9}\text{Mo}_6\text{O}_{17}$, however, we could not find the superconducting phase transition in the temperature range examined here.

There are four main reasons why the CDW transition temperature can vary with pressure: changes in electron-phonon interaction, fluctuation, commensurability pinning and the nesting condition of the Fermi surface (Barišić 1985). Figure 3 shows the

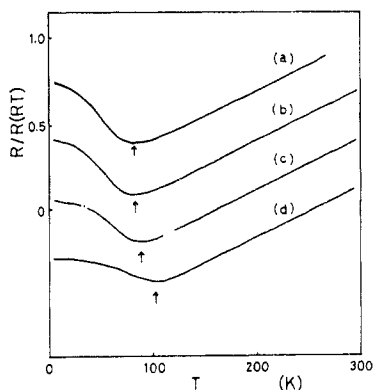


Figure 1. Temperature dependence of the electrical resistivities of $(\text{Na}_{0.8}\text{Li}_{0.2})_{0.9}\text{Mo}_6\text{O}_{17}$ under various hydrostatic pressures: curve (a), 1.5 kbar; curve (b), 5.1 kbar; curve (c), 9.2 kbar; curve (d) 12.8 kbar. The resistivities are normalized at 300 K. The arrows indicate the temperatures at the minimum resistivities. The scale on the ordinate applies to the top curve (a). Other curves are shifted on the same scale.

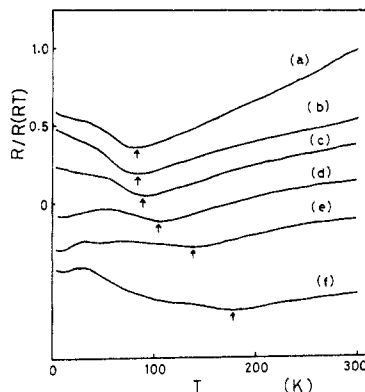


Figure 2. Temperature dependence of the electrical resistivities of $\text{Na}_{0.9}\text{Mo}_6\text{O}_{17}$ under various hydrostatic pressures: curve (a), 0 kbar; curve (b), 1.5 kbar; curve (c), 5.1 kbar; curve (d), 9.2 kbar; curve (e), 12.8 kbar; curve (f), 16.2 kbar. The resistivities are normalized at 300 K. The arrows indicate the temperatures at the minimum resistivities. The scale on the ordinate applies to the top curve (a). Other curves are shifted on the same scale.

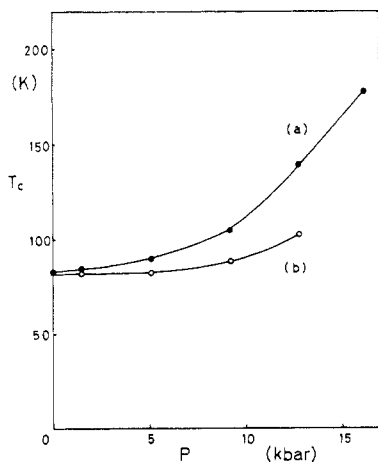


Figure 3. Pressure dependence of the CDW transition temperatures of $\text{Na}_{0.9}\text{Mo}_6\text{O}_{17}$ (curve (a)) and $(\text{Na}_{0.8}\text{Li}_{0.2})_{0.9}\text{Mo}_6\text{O}_{17}$ (curve (b)). The temperatures were determined by the minimum resistivities.

pressure dependence of T_c in $\text{Na}_{0.9}\text{Mo}_6\text{O}_{17}$. We can see that T_c increases as the pressure increases, in contrast to the simple cases of the inorganic low-dimensional conductors such as MX_2 and MX_3 (Delaplace *et al* 1976, Ido *et al* 1979, Briggs *et al* 1980). In the latter compounds the decrease in T_c is caused by a change in the nesting condition. On the other hand, an increase in T_c with increasing pressure similar to the present case has been reported in KCP, where the electronic gap below T_c decreases (Thielemans *et al* 1976). These behaviours are explained by a decrease in the electron-phonon interaction and an increase in the interchain coupling (Horovitz *et al* 1975, Thielemans *et al* 1976).

Recently, a similar pressure dependence of T_c has been reported also in γ - and η - Mo_4O_{11} (Inoue *et al* 1987, 1988). However, their resistivity behaviours indicate an increase in the CDW gap below T_c with increasing applied pressure. In the case of the $\text{Na}_{0.9}\text{Mo}_6\text{O}_{17}$ system, the increase in the interchain (or interplane) interaction seems to suppress the fluctuation effect and therefore induces an observed increase in T_c with increasing pressure, although the anisotropy is smaller than that of KCP.

It may be useful to refer to the pressure dependence of T_c of the organic low-dimensional conductor TTF-TCNQ. This dependence is complicated (Friend *et al* 1978) in that there is a change in the commensurability energy with change in $2k_F$ (Suzumura and Fukuyama 1980), where k_F is the Fermi momentum. However, the $\text{Na}_{0.9}\text{Mo}_6\text{O}_{17}$ system, because its CDW is commensurate (Pouget 1989), does not exhibit the complexity that TTF-TCNQ does.

In conclusion, we found an increase in the CDW transition temperature T_c in $\text{Na}_{0.9}\text{Mo}_6\text{O}_{17}$ with increasing applied pressure in contrast to the simple cases of inorganic low-dimensional conductors. The low-temperature resistivity of one of the present samples shows a metallic behaviour below T_c at high pressures, which is explained by the increase in three dimensionality.

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