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EFFECT OF N_2H_4 INTERCALATION ON THE TRANSITION TEMPERATURE AND ELECTRON TRANSPORT IN ANISOTROPIC SUPERCONDUCTOR: $TaSe_3$

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We have intercalated $TaSe_3$ with N_2H_4. The room temperature resistance $R_b(RT)$ along the b-axis decreases by 3[illegible] during the reaction. The ratio of $R_b(RT)/R_b(4.2\,K)$ varies from 60 in $TaSe_3$ to 5 in the intercalated samples. An anomaly in R_b is observed near 100 K. It is suggested that this anomaly is due to the formation of charge-density-wave (CDW). The superconducting transition temperature, T_c, is suppressed from 2.2 to 1.5 K. The correlation between the suppression of superconductivity and the formation of CDW is discussed as compared with the result obtained in the layered dichalcogenides.

1. Introduction

The trichalcogenides of niobium and tantalum, MX_3, have been investigated extensively because of their low-dimensional characters. Although their crystal structures are similar, their electron transport properties show significant differences [1]. $NbSe_3$ shows two charge-density-wave (CDW) transitions at 142 and 85 K [2] and TaS_3 shows Peierls transition at 220 K [3]. No anomalies associated with low-dimensional phase transition have been observed in $TaSe_3$ [1]. In fact, $TaSe_3$ is a superconductor with a T_c of (2 ± 0.5)K [1, 4]. The reasonable interpretation for the absence of the structural phase transition in $TaSe_3$ is that the interchain overlap of electron wave functions is larger than that of other MX_3 compounds.

As shown in fig. 1, $TaSe_3$ has a structure made up of infinite chains of trigonal prisms, $TaSe_6$, extending parallel to the b-axis. Ta in $TaSe_3$ is surrounded by eight Se atoms forming a bicapped trigonal prism. According to this point of view we have to consider layers made of coupled $TaSe_3$ fibers. Between them lies a Van der Waals gap with dimensions close to those of the layered dichalcogenides, MX_2. $TaSe_3$, therefore, can be considered as a two-dimensional system with a large anisotropy rather than a one-dimensional system.

MX_2 is well known to be intercalated with organic molecules and hydrogen [5, 6]. The enhancement of T_c and the suppression of the CDW formations were observed in many intercalated compounds of MX_2 [5, 6]. It is very interesting to investigate the intercalation effect on T_c and the electron transport in $TaSe_3$ and to compare them with the effect in MX_2. The measurements of X-ray reflecting power, elec-

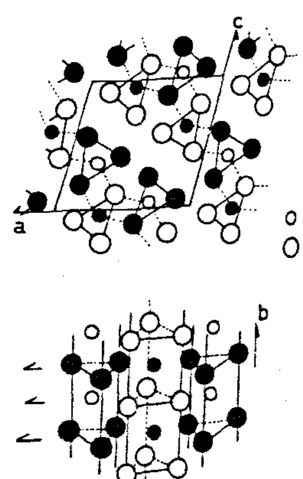

Fig. 1. The crystal structure of $TaSe_3$. The small (large) circles identify Ta (Se) atoms.

...al resistivity, and T_c in $TaSe_3$, when this is exposed to N_2H_4, are reported in this paper. T_c is found to be suppressed on intercalation of N_2H_4 and an anomaly in the resistivity is observed near ...K. The correlation between the suppression of T_c and an anomaly in resistivity is compared with that of intercalation on MX_2 and discussed.

2. Experiments

$TaSe_3$ were grown by the transport method, as reported in [4]. The resistance ratios, RRR, R(room temperature)/R(4.2 K), and T_c of the crystals were confirmed to be the same values as observed in previous works [1, 4]. The resistivity of samples was measured by a four-probe method along the b-axis with a dc current. Superconductivity was detected by the resistivity measurements. The X-ray diffraction and transport measurements of $TaSe_3$ exposed to N_2H_4 vapor in the absence of air were carried out using the method already developed for TaS_2 [7]. The measurements of the temperature dependence of the resistivity and T_c in the samples were carried out under He gas atmosphere to avoid the reaction with air.

3. Result and discussion

X-ray diffraction by the as-grown $TaSe_3$ was consistent with the reported structure [1]. The reflecting power of (400) plane in the $TaSe_3$ disappears on exposure to N_2H_4 with 12 Torr at ...°C for 90 min. This suggests that N_2H_4 must be intercalated in between the layers along the diagonal of the a- and c-axes. The stoichiometry of the complex formed $TaSe_3(N_2H_4)_x$ was determined by weight gain measurements [8]. The dependence of the rate of weight gain vs. pressure of N_2H_4 is shown in fig. 2. Different kinetics for the pressure of N_2H_4 suggests that the conditions of the intercalation in the low pressure regions are more gentle than those in the high pressure regions. Within minutes of exposure to H_2H_4, $x = 1/3$, but after 48 h under 12 Torr of N_2H_4 the value of x is equal to one. When the sample is evacuated, x is reduced to 2/3 immediately and after 48 h under 10^{-5} Torr of N_2H_4 $x = 1/3$. However, X-ray diffraction by the intercalated sample, $TaSe_3(N_2H_4)_x$, does not allow a structure determination because the lines are very broad, as shown in fig. 3. This suggests the formation of complex superstructures.

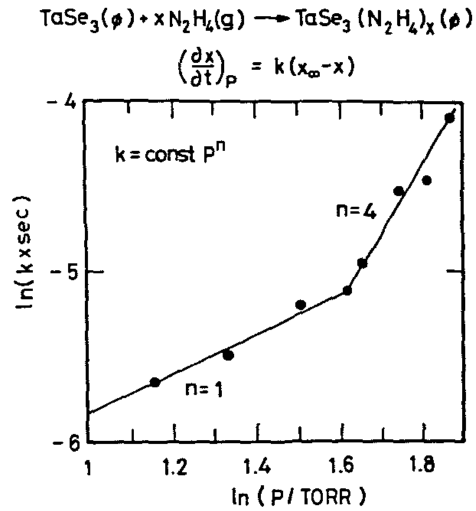

Fig. 2. Kinetics of intercalation vs. pressure of N_2H_4. x is the ratio of moles of N_2H_4 to the moles of $TaSe_3$. x_∞ is the extrapolated value as time, t, $\rightarrow \infty$. Here the same sample was intercalated and deintercalated several times for a given pressure and each point indicates an average.

On exposure to N_2H_4 vapor (12 Torr, 24°C) the resistance decreases with the time to a constant value 30% less than the initial value. This value remained when the pressure was reduced to 10^{-4} Torr. The result of the change in the resis-

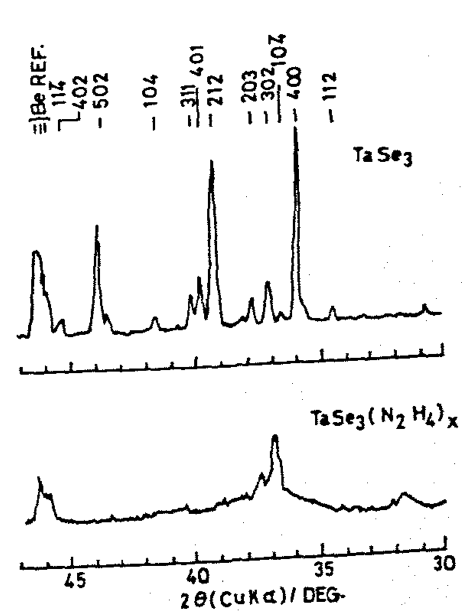

Fig. 3. X-ray reflecting power of $TaSe_3$ and its intercalated sample, $TaSe_3(N_2H_4)_x$.

tance is consistent with that in X-ray diffraction by the intercalated samples. The temperature dependence of the resistivity has been measured on four intercalated samples. Two of them (A and B) have been intercalated under a pressure of N_2H_4 at 12 Torr and other samples (E and F) have been intercalated under a pressure of N_2H_4 at 1 Torr. As shown in fig. 4, the dependence of the resistivity vs. temperature in $TaSe_3(N_2H_4)_x$–F is almost the same as that of $TaSe_3$, i.e., resistance decreases monotonically as temperature decreases and shows negative curvature. The same result is also obtained for E. Also, the onset temperature for superconducting transition is the same as that of $TaSe_3$, as shown in fig. 5. Thus, no significant changes in the resistivity and T_c are observed in the E- and F-samples exposed to N_2H_4 under low pressure. On the other hand, the temperature dependence of resistivity in $TaSe_3(N_2H_4)_x$–B intercalated under the pressure of N_2H_4 at 12 Torr, which is the same as that of the A-sample, is quite different from that obtained in $TaSe_3$ [1], as shown in fig. 6. The main differences are as follows. (1) An anomaly near 100 K is observed in the intercalated samples. (2) The positive curvature against temperature in resistivity vs. temperature is observed above 120 K, in contrast with the negative curvature obtained in $TaSe_3$ [1]. (3) The value of RRR varies from 60 to 5. Furthermore, T_c of the intercalated samples are observed to decrease from 2.2 to 1.5 K, as shown in fig. 7. The resistivity anomaly observed here is similar to that associated with the CDW formation observed in dichalcogenides and their intercalated com-

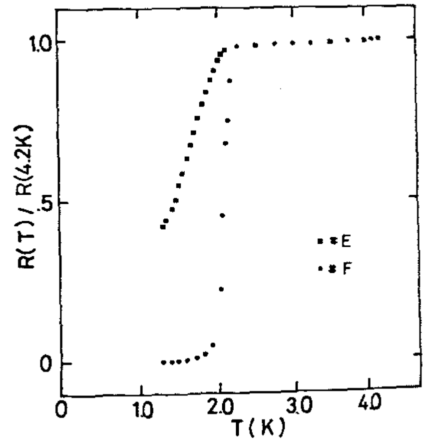

Fig. 5. The superconducting transition curves of $TaSe_3(N_2H_4)_x$–E and –F.

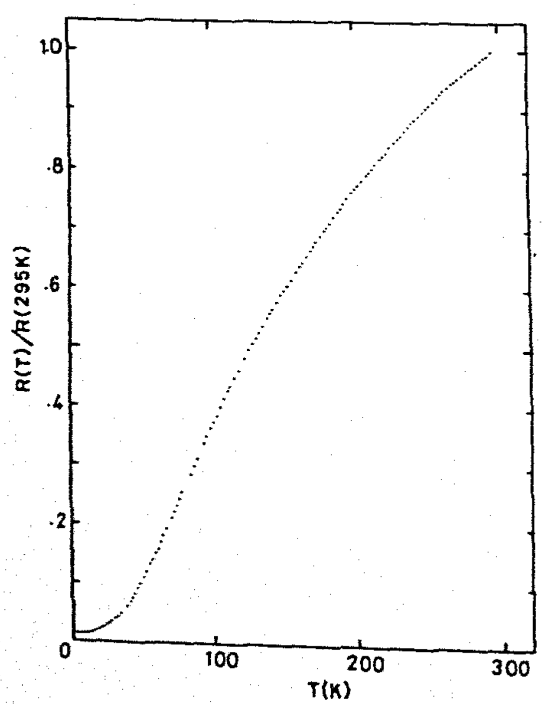

Fig. 4. The normalized resistance as a function of temperature in $TaSe_3(N_2H_4)_x$–E.

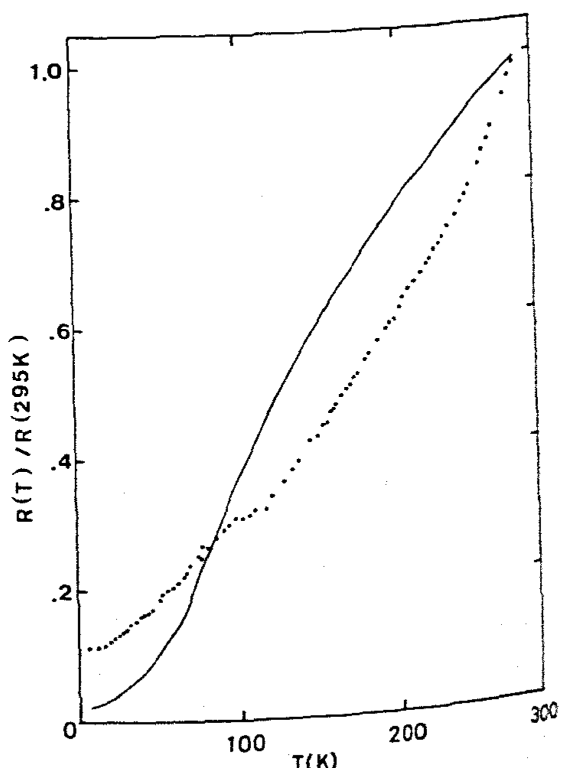

Fig. 6. The normalized resistance as a function of temperature in $TaSe_3(N_2H_4)_x$–B. Solid line shows the temperature dependence of the resistance in $TaSe_3$.

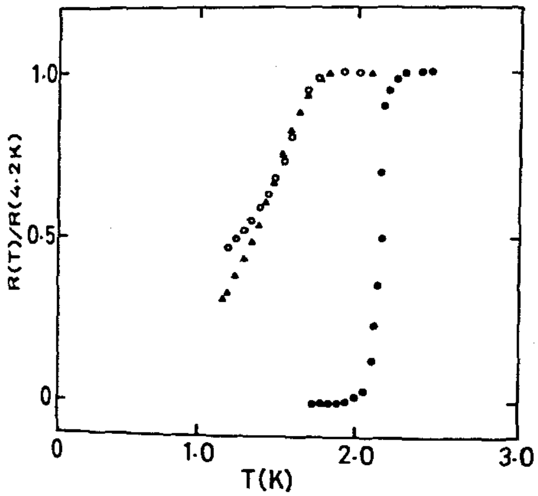

Fig. 7. The superconducting transition curves of $TaSe_3(N_2H_4)_x$–A (○); –B (▲); and $TaSe_3$ (●).

pounds [5–7]. The interlayer overlap of electron wave function in $TaSe_3$ can be expected to be reduced by the intercalation. Thus, the resistivity anomaly observed in the intercalated samples (A and B) suggests that CDW are produced and that these in turn suppress the superconductivity. This result is consistent with the fact observed in MX_2 in which the enhancement of superconductivity leads to suppression of the CDW formation. However, much more work, such as an observation of the formation of periodic lattice distortions and measurements of Hall effect and magnetic susceptibility, will be necessary to confirm the CDW formation in $TaSe_3(N_2H_4)_x$. Also, it is important to investigate if the anomaly observed here is due to the weakly pinned CDW which had been already observed in $NbSe_3$ [9].

Acknowledgements

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