Metal-nonmetal transition and the superconductivity in $TaSe_3$

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Abstract:

The superconducting transition curves measured at various current densities in $TaSe_3$ were found to show the strong current density, J, dependence. In the high J regime the superconducting transition curve was consistent with published ones, but in the low J regime the resistance increases rapidly up to a value larger than the normal one from the onset temperature of the T_c transition obtained in the high J regime and had a peak, then at low temperature than the T_c the sharp superconducting transition was observed. It is suggested that these results observed first may be a new phenomenon which is related to a superconductivity itself rather than the existance of other phases such as the charge-density-waves and the spin-density-waves phase or the localization effects.

Among the transition-metal trichalcogenides, MX_3, $TaSe_3$ is an unique compound because no anomalies associated with low dimensional phase transition have been observed. In spite of the evidences of the charge-density-waves (CDW's) formation in $NbSe_3$[1)] and the Peierls transition in TaS_3[2)], $TaSe_3$ is a superconductor whose transition temperature, T_c, has been established to be 2.3 K with resistivity measurements[3, 4)]. The superconducting transition curve above T_c has not been explained by the mean-field fluctuation theory. As the simple interpretation for these facts, it would be proposed that $TaSe_3$ is a three-dimensional conductor with a great anisotropy rather than a quasi one-dimensional one.

Recent studies on the diamagnetic susceptibility, χ_{dia}, have shown that χ_{dia} is extremely small (10^{-3} of the perfect diamagnetic susceptibility, $-1/4\pi$) near T_c and increases rapidly near 0.3 K to be about 80% of $-1/4\pi$. From this temperature dependence of χ_{dia}, Bastuscheck proposed that $TaSe_3$ is a filamentary superconductor in which near T_c $TaSe_3$ behaves like a quasi-one-dimensional superconductor and at lower temperatures becomes a three-dimensional superconductors with filaments coupled by Josephson effect[5). Therefore, it needs to re-examine the superconducting transition with resistivity measurements whether a quasi-one-dimensional behavior is exhibited in the transition curves above T_c in $TaSe_3$. In order to clear the superconducting fluctuation effect, measurements of the superconducting transition have been made precisely at extremely low current densities, J, compared with those of published ones[3, 4)]. In contrast to the resistivity decrease due to the fluctuation effect, the resistivity rise from the onset temperature of the T_c transition and at a lower temperature the sharp superconducting transition have been observed in the low J regime. In the high J regime, on the other hand, the same superconducting transition as the published ones has been observed. The transition curve above T_c for both low and high J regimes has not been explained by the mean-field fluctuation theory. It is suggested that the metal-nonmetal-superconductor transition first observed here is a new phenomenon related to a superconductivity.

$TaSe_3$ was synthesized by the direct reaction of Ta(99.99%) and Se(99.9999 %). The mixture was sealed in quartz tube in vacuum of $\sim 10^{-5}$ Torr and heated to 650°C for 4 weeks. After heat treatment, the quartz tube was cooled in air. The samples were ribbonlike with typical dimensions being 10 mm long, 0.05 mm wide and 0.01 mm thick. Residual resistance ratio, RRR = R(300K)/R(4.2K), of crystals was 100 $\sim$ 150, which were the highest value in the published ones.

Resistance of cystal was measured by the usual four probe dc technique with the current parallel to the b-axis (chain-axis). The electrical contacts were made with silver paint to Te and Au pads preriously evaporated on the crystals. The samples were mounted on a holder which immersed in liquid helium. In this way the temperature could be controlled from 1.2 K to 4.2 K by pumping helium bath. The temperature stability was kept less than 5mK/min. Temperature was measured by germanium resistance thermometer. The measured current was supplied by the constant current sources in the range from 0.1 μA to 1.0 mA with 0.3% accuracy. The sample voltage was measured by a digital voltmeter (YEW model 2501) which has 0.1 μV resolution. Measurements were done by using computer controlled system in which the data was sampled four times and averaged. Thermoelectoric voltage across the measurement circuit was cancelled by reversing the current direction.

The current vs. voltage characteristic above 2.3 K showed the linear relation in all regions of the measured current density, J, that is, the ohmic resistivity was confirmed. However, below 2.3 K the strong J dependent resistivity was found. This anomalous behaviors were observed in several samples. In Fig. 1 is shown the typical temperature dependence of the electrical resistance normalized by the resistance at 4.2 K, $R_n = R(T)/R(4.2K)$ for two of samples measured. In relatively high J regime, R_n decreases sharply at near 2.3 K with decreasing temperature and becomes fully zero near 1.7 K within the experimental accuracy. T_c defined as the midpoint of the resistive transition curve is about 2.1 K. The temperature width of 80% signal change, ΔT, is about 0.35 K. The T_c and ΔT were constant for the region of J from 2 A/cm^2 to 40 A/cm^2. These results measured in high J regime are consistent with those of the superconducting transition with resistivity measurements reported already[3, 4]. Therefore, it can be concluded that the decrease of R_n obtained in the high J regime is due to the superconducting transition. In relatively low J regime, on the other hand, R_n increases rapidly with decreasing temperature from the nearly same temperature as the onset temperature of the superconducting transition, T_c^{onset}, and has a peak, then decreases sharply down to zero resistance. Thus it is found that $TaSe_3$ shows the metal-nonmetal-zero resistance transition in the low J regime.

The peculiarities of the zero resistance transition with giant resistivity can be summarized as follows. (1) The metal-nonmetal-zero resistance transition occurs within the same temperature region as the T_c transition observed in the high J regime. (2) The onset temperature of the metal-nonmetal transition, T_{MN}, is almost equal to T_c^{onset} and is independent on the magnitude of J. (3) The peak of the R_n is easily reduced by weak J, but the temperature corresponding to the peak, T^p, is independent on J. For higher J, the metal-nonmetal-zero resistance curve coincides with the superconducting transition curve obtained in the high J regime.

Considering that R_n decreases with increasing J, the anomalous temperature dependence of R_n in the low J regime is not interpreted in terms of heating effects. We consider that the J dependent resistivity should be ascribed to an intrinsic mechanism in the electronic system.

In the peculiarities of the metal-nonmetal-zero resistance transition, one of the most interesting problems is whether the state of zero resistance is a superconducting state. In order to understand the zero resistance state, the temperature dependence of R_n at several magnitudes of the magnetic field, H, was measured. The H was applied perpendicular to the ribbon surface of the crystal. The result is shown in Fig. 2. It can be seen that R_n^p is strongly suppressed by weak H and at the same time the T^p is shifted to low temperatures. The temperature corresponding to $R_n = 0$ also decreases as H increases. From these evidences it is concluded that the state of $R_n = 0$ observed in the low J regime also is a superconducting state. Therefore, we find that $TaSe_3$ shows the two different superconducting transition; one of them is the usual superconducting transition which is insensitive to J and the other is the metal-nonmetal-superconductor (M-N-S) transition which is strongly dependent on weak J. Furthermore, it is found that the resistance in the nonmetal region is strongly suppressed by weak H, that is, $TaSe_3$ shows the large negative magnetoresistance.

We replotted the resistance measured in the log R vs. reciprocal temperature as shown in Fig. 3. In the nonmetal region it is found that all experimental data points are approximately on a straight line. Thus, the activation type temperature dependence of resistance is observed in the nonmetal region. The activation energy, which is obtained as a slope of the straight line, is strongly suppressed by J. However, T_{MN} is almost independent on J.

In Fig. 4 it is shown that the resistance measured in the low J regime at several magnitudes of the magnetic field is reprotted in the log R vs. reciprocal temperature. In the nonmetal region the temperature dependence of resistance in the magnetic field also is the activation type. The activation energy decreases with increasing the magnetic field. It is found that T_{MN} decreases with increasing the magnetic field, which is different from the fact that T_{MN} is independent on J. This suggests that the role of the

J and H for the metal-nonmetal transition is independent each other.

The peculiarities of the nonmetal region can be summarized as follows. (1) The conductivity in the nonmetal region is nonohmic. (2) The temperature dependence of the conductivity in the nonmetal region is the activation type. The activation energy depends on both J and H. (3) The large negative magnetoresistance is observed in the nonmetal region. (4) T_{MN} is almost independent on J, but sensitive to H. (5) The metal-nonmetal transition observed here is very sharp.

Now investigate the mechanism of the metal-nonmetal transition to explain consistently these facts observed here. As one of the possible explanations, it is natural to consider that the metal-nonmetal transition may be due to the CDW formation, since the metal-nonmetal transitions observed in $NbSe_3$[1, 6] and TaS_3[2] have been interpreted in terms of the CDW formation. The resistivity rise below CDW's transition in $NbSe_3$ and TaS_3 is reduced by dc electric field, E, above a threshold field, E_t[7, 8]. This E dependent conductivity is understood that the CDW's are weakly pinned and can released to more freely at high field. It is predicted by Frohlich that if the CDW's can move without attenuation with a constant speed, the ground state would be a superconducting state. It is known that the nonohmic conductivity in the CDW system is not affected by the application of a magnetic field, but rigid for a magnetic field[9, 10]. This is due to the highly one dimensional motion of the depinned CDW. However, in the present work it is shown that the conductivity strongly depends on H, and the large negative magnetoresistance is observed. These facts are not expected in the state of sliding CDW. In fact, no evidence due to the CDW formation has been observed yet in $TaSe_3$. It is therefore concluded that the metal-nonmetal transition near T_c in $TaSe_3$ is not caused by the CDW formation.

The nonohmic conductivity, $\sigma(E)$, has been observed also in the SDW system such as $(TMTSF)_2PF_6$[11] and $(TMTSF)_2ClO_4$[12]. On the assumption that the $\sigma(E)$ is caused by the depinning of the SDW in analogy with the phenomena found in the CDW systems, it is considered that the conductivity of the depinned SDW also should be highly E sensitive but independent on H. Furthermore, the shift of T_{MN} by H of 150 Oe is observed to be 0.15 K whose value is too large to compare with the Zeeman energy for 150 Oe (0.01 K).

Therefore it is supposedly that the H sensitive nonohmic-conductivity is not due to the sliding motion of the SDW. However, to confirm clearly whether this phenomenon is due to the SDW formation, we need more studies such as static susceptibility and ESR measurements.

As the other possible explanation of the metal-nonmetal transition, the localization by impurities of electrons may be proposed. The activation type conductivity and the large negative magnetoresistance observed in the nonmetal region are qualitatively consistent with the features of the electron transport in the localized state. However the transition from the metal to the localized nonmetal regime is known to be sluggish from the temperature dependence of the resistivity. This is contrast to the present result in which the metal-nonmetal transition occures within the narrow temperature region about 0.4 K. This very sharp transition suggests that it may be the phase transition related to the collective excitation effects. Therefore it is difficult to explain the metal-nonmetal transition in terms of the localization effects.

These experimental results in the present work are not explained sufficiently in terms of any of the CDW transition, the SDW transition and the localization effects. The origin of the new phenomenon observed here is unsetted at the present state. However, we should note the following experimental results. (1) T_{MN} is almost equal to T_c^{onset}. (2) ΔT is independent on the magnitude of J. (3) With increasing J the M-N-S transition undergoes the usual superconducting transition. (4) The M-N-S transition is easily suppressed by the magnetic field. These results suggest that the M-N-S transition observed first is a new phenomenon which is closely related to a superconductivity itself rather than the existence of other phases. Then we believe that this may be the key to the origin of the M-N-S transition in $TaSe_3$ observed in the low J regime.

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Figure Captions

Fig. 1. Normalized resistance $R_n = R(T)/R(4.2K)$ vs. temperature at several current densities for two samples.

Fig. 2. Normalized resistance $R_n = R(T)/R(4.2K)$ vs. temperature at several magnetic fields in the low current density regime.

Fig. 3. Logarithmic resistance (log R) vs. reciprocal temperature (1/T) at several current densities.

Fig. 4. Logarithmic resistance (log R) vs. reciprocal temperature (1/T) at several magnetic fields in the low current density regime.

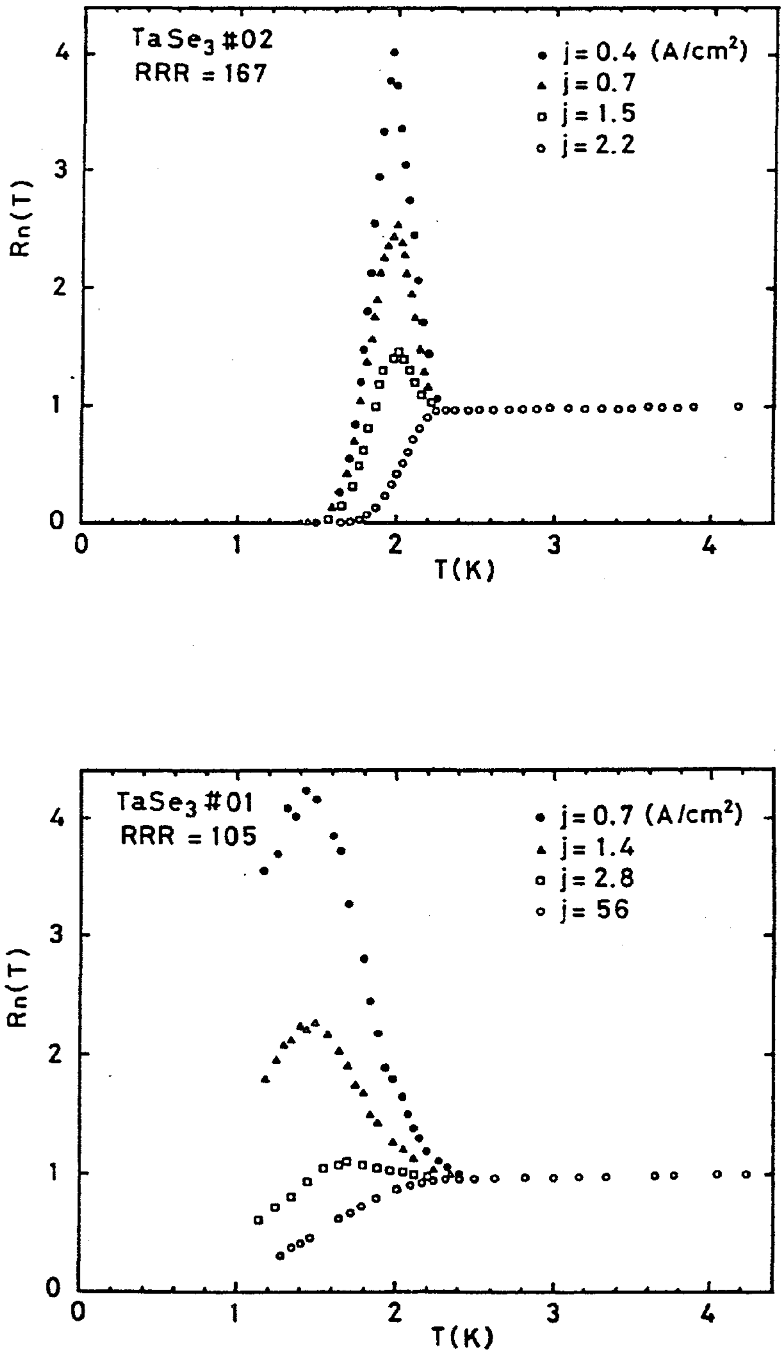
TaSe3 #02
RRR = 167
j = 0.4 (A/cm²)
j = 0.7
j = 1.5
j = 2.2
Rn(T)
T(K)
TaSe3 #01
RRR = 105
j = 0.7 (A/cm²)
j = 1.4
j = 2.8
j = 56
Rn(T)
T(K)

Fig. 1

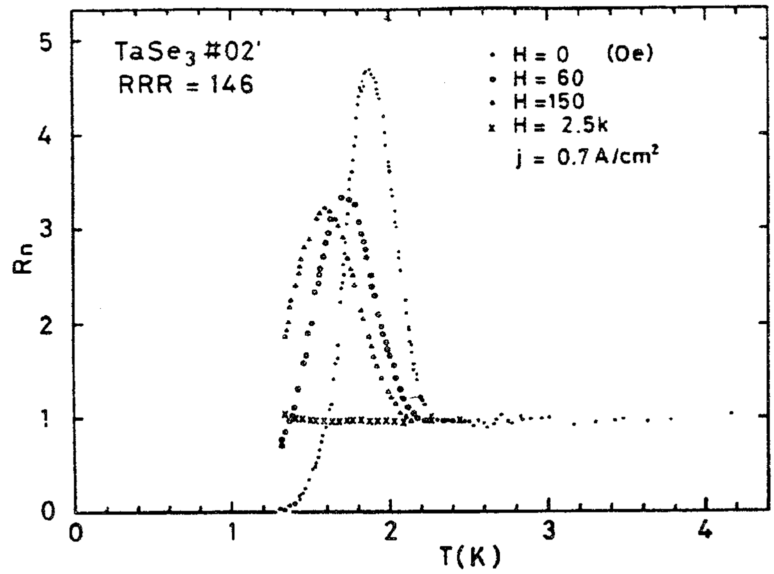

Fig. 2

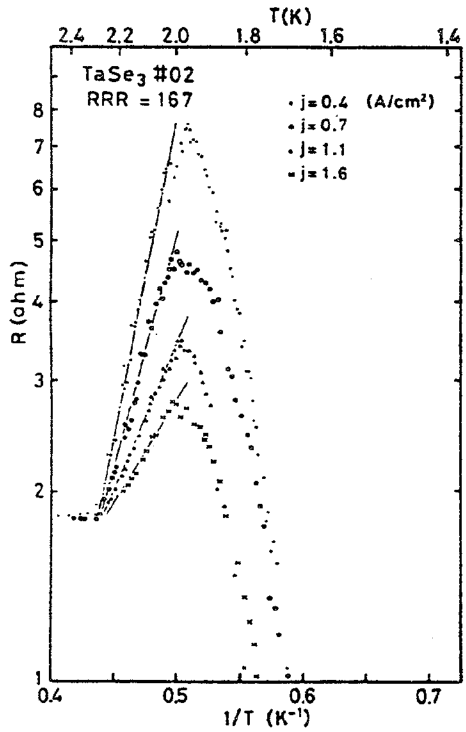

Fig. 3

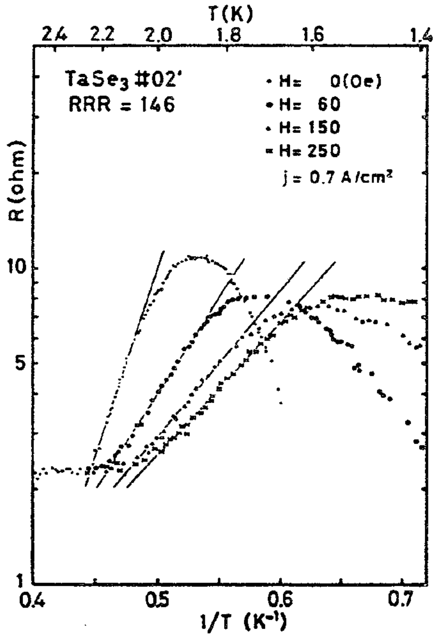

Fig. 4