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ELECTRICAL AND MAGNETIC PROPERTIES OF ANDERSON LOCALIZED STATE IN 1T-$TaS_{2-x}Se_x$

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The electrical resistivity, Hall coefficient and magnetoresistivity of 1T-$TaS_{2-x}Se_x$ (x=0-2) were measured from 400K to 0.1K. The well established Anderson localization of 1T-TaS_2 was proved to be much enhanced by substituting Se for S. The characteristic magnetoresistance increased remarkably with the positive sign. Alloys of $0.8<x\leq1.6$ showed almost temperature independent resistivities followed by sharp drops below ca. 1K due to the appearance of superconductivity.

The paramagnetic part of magnetic moment in 1T-TaS_2 associated with the localized state was measured down to 1.2K and was found to have a non-linear field dependence, which fits neither with the Brillouin function nor with a simple theory of localization including the correlation energy.

1. Electrical Property

The Anderson localized state is realized in 1T-TaS_2 at low temperatures under the commensurate phase of charge density wave (CDW) having the strongest amplitude among transition metal dichalcogenides. The localization is enhanced by substituting Se for S. In this section we report the electrical properties of 1T-$TaS_{2-x}Se_x$ in connection with the Anderson localization and CDW.

Low temperature electrical properties of 1T-$TaS_{2-x}Se_x$ take three characteristic ranges of composition; $0\leq x\leq0.8$ (nonmetallic), $0.8<x\leq1.6$ (poorly metallic) and $1.6<x\leq2$ (metallic). Figure(1) shows the temperature dependence of electrical resistivities for typical samples of 1T-$TaS_{2-x}Se_x$ (x=0-2). 1T-TaS_2 shows two successive first order transitions at $T_d'\simeq350$K and $T_d\simeq200$K. An incommensurate and a commensurate CDWs are stable above T_d' and below T_d, respectively. The intermediate temperature range, $T_d'<T<T_d$ corresponds to a nearly commensurate phase. The temperature hysteresis of T_d for 1T-TaS_2 is 25K, but increases with the increase of selenium doping amount, x in 1T-$TaS_{2-x}Se_x$ ($x<1.4$) and amounts to 220K in 1T-$TaS_{0.6}Se_{1.4}$(#A1). On the other hand, the resistivity rise at T_d diminishes by ca.1/5 times. When the resistivity rise at T_d is extinguished, the material becomes a similar one to 1T-$TaSe_2$ in the sense that the material has nothing of the nearly commensurate phase. However, the temperature dependence of electrical resistivity for almost all of the samples in 1T-$TaS_{0.6}Se_{1.4}$ is not similar to that of 1T-$TaSe_2$ but rather flat as exemplified as the curve of 1T-$TaS_{0.6}Se_{1.4}$(#B1) in Figure(1).

An abrupt change of sign in the Hall coefficient and a reduction of carrier concentration from 5×10^{21} to 5×10^{19} cm^{-3} are observed in

1T-TaS_2 at T_d; above T_d major carrier is n-type and below, p-type. On the other hand, the sign of Hall coefficients of 1T-$TaS_{0.6}Se_{1.4}$(#B1) and 1T-$TaSe_2$ is positive in the whole temperature range measured. The carrier concentrations are ca. 10^{22} cm^{-3}, assuming the one carrier model. Therefore, the metallic nature of 1T-$TaS_{2-x}Se_x$($x \leq 0.8$) is converted into the extrinsic p-type semiconductor below T_d, while that of 1T-$TaS_{2-x}Se_x$($x \geq 1.4$) remains metallic[1].

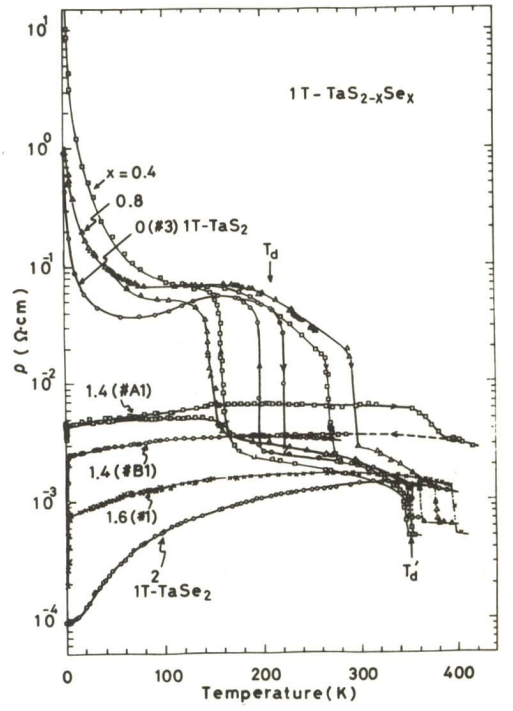

Fig.1 Temperature dependence of electrical resistivity of 1T-$TaS_{2-x}Se_x$

Now, we describe the low temperature electrical property in the first composition range of nonmetallic character(ρ(1.4K) $=10^0$-$10^3\Omega\cdot$cm). The temperature dependence of electrical resistivity is found as $ln\rho(T) \propto (T_0/T)^{1/n}$ in the temperature range of 4K to the measured lowest temperature, 0.1K, showing the variable range hopping of Anderson localized state. The n-value is nearly 2 or 3 for 1T-TaS_2, depending on the sample character[2,3] and nearly 3 for selenium doped samples. The characteristic temperature T_0 for 1T-$TaS_{2-x}Se_x$, which is a measure of randomness of the potential in the Anderson localization, increases from 1K for x=0 to 110K for x=0.001 and to 2200K for x=0.6, and ρ(T) also increases by two orders of magnitude. Namely, the addition of selenium remarkably enhances the localization. The precise data of characteristic temperature and activation energies are described in our previous paper[4].

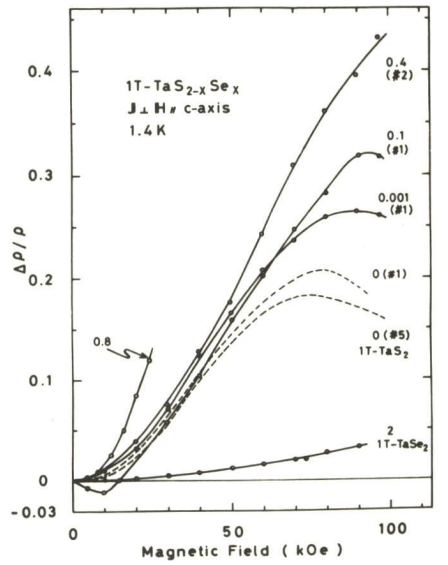

Fig.2 Transverse magnetoresistance of 1T-$TaS_{2-x}Se_x$

In this temperature range, the magnetoresistance $\Delta\rho/\rho = \{\rho(H)-\rho(0)\}/\rho(0)$ for 1T-TaS_2 takes three characteristic temperature ranges[3,5]. First, $\Delta\rho/\rho$ is negative at 4.2K, having a value of -0.015 at 20kOe, then becomes positive in the temperature range of 2K to 0.5K, having a maximum of ca. 0.28 at 40kOe followed by a rather sharp drop up to the measured highest field 100 kOe, and lastly takes a very large negative value of -0.9 at 0.1K.

By the selenium doping, the positive magnetoresistance in the second range is remarkably enhanced and the negative behavior tends to be suppressed, as shown in Fig.(2). The magnetoresistance $\Delta\rho/\rho$ at 1.4K for x of 0, 0.001, 0.1, and 0.4 are 0.20, 0.26,

0.32 and (0.47) at the maxima, respectively. Therefore, the positive magnetoresistance increases with the increase of potential disorder by alloying. These experimental results are basically explained by the recent model for the Anderson localized state by Fukuyama and Yosida[6,7], considering the change of the mobility edge as a function of magnetic field together with the Zeeman shift of the Fermi level.

Lastly, we mention the electrical property in the second composition range of poorly metallic character. As shown in Fig.(3), the steep decrease of electrical resistivity in 1T-$TaS_{0.6}Se_{1.4}$, having two steps, appears below ca.3K. Because the magnetoresistivity recovers up to the temperature independent resistivity as the extention of residual resistance above ca. 3K, the steep drop of resistivity means an occurrence of superconductivity. It was ascertained by the electron diffraction that present samples contain ca.0.1% of other polytypes such as 4H or 6R. These polytypes of $TaS_{0.6}Se_{1.4}$ become superconductive with the transition temperature at ca.3K[8]. Therefore, the resistivity step at ca.3K may be related to it, but the other step at ca.1.2K should be the intrinsic nature of the 1T-phase. The reason why the superconductivity occurs only in the poorly metallic range is under investigation.

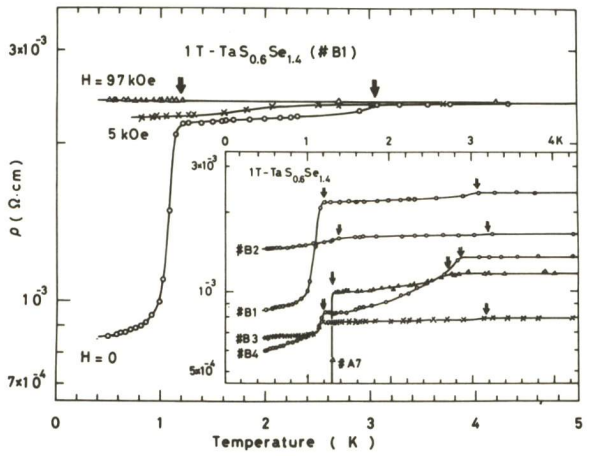

Fig.3 Temperature dependence of electrical resistivity of 1T-$TaS_{0.6}Se_{1.4}$

2. Magnetic Property

The magnetic susceptibilities of 1T-$TaS_{2-x}Se_x$ were measured by the Faraday method from 300K to 1.2K, as shown in Fig.(4). The susceptibility consists of a diamagnetic part, χ_d and a paramagnetic one, χ_p. The former χ_d is almost temperature independent, while the latter χ_p appears below ca. 100K and shows a Curie like temperature dependence. These behaviors are almost the same with the data by DiSalvo et. al[9] which was taken above 4.2K.

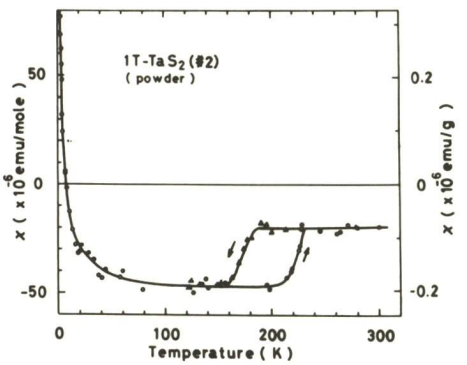

Fig.4 Magnetic susceptibility of 1T-TaS_2 powder

Below 4.2K, the paramagnetic moment shows a non-linear field dependence. Figure(5) shows the paramagnetic moment M_p, difined as $M(T) - M_d$, versus H/T. Here M_d is the temperature independent diamagnetic moment above 100K and corresponds to $\chi_d H$. In the figure, the magnetic moment M_p is proportional to H/T up to an appropriate value of the abscissa, for example ca. 6kOe/K for 1T-TaS_2(#2), but deviates downwards gradually at higher

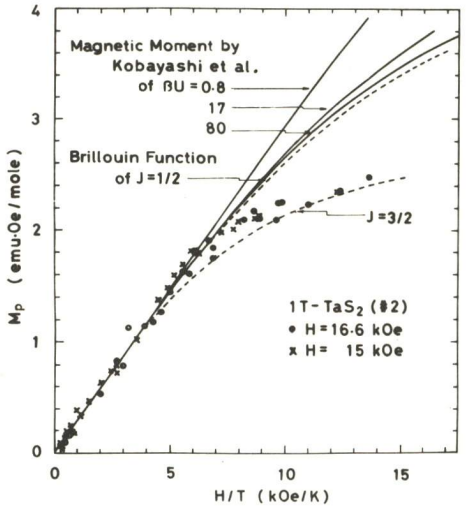

Fig.5 Paramagnetic moment vs. H/T relation of 1T-TaS_2 powder. The solid and dashed curves correspond to the formula by Kobayashi et al. and the Brillouin function for J=1/2 and J=3/2 , respectively.

values of H/T. We tried a numerical fitting of this data to the Brillouin function by choosing the angular momentum number J, as shown by the dashed lines in Fig.(5). The line of J=3/2 has a better fit than the J=1/2 one, but J=3/2 is hard to be applied in the present electronic system. We also tried with the theoretical formula by Kobayashi et al.[10] for the magnetic moment in the Anderson localized state for which the electron correlation energy U is taken into account. The data however do not fit with the curves of various values of βU.

If this paramagnetic moment is ascribed to the paramagnetic impurities of J=1/2, the amount is estimated as 1040ppm(#1), 820ppm(#2) and 1280ppm(#3) for 1T-TaS_2, and 850ppm for 1T-$TaS_{1.9}Se_{0.1}$ from the linear parts of paramagnetic moments. These values are converted into 1/5 times in the case of J=3/2. Our samples were made of Ta(99.9%) and S(99.9999%). The original Ta contains 15ppm Fe, 10ppm Nb and Ni less than 10ppm as the major metallic impurities. Among them, Fe^{2+} atoms are known to be divalent, Fe, and carry no magnetic moments from the measurements of the magnetic susceptibility and Mössbauer effect[11]. The amount of ca. 1000ppm for J=1/2 or ca. 200ppm for J=3/2 is too much large to be considered as the amount of impurities. Therefore, this paramagnetism should be related to the electronic state of Anderson localization, although the experimental data do not fit with the simple formulae.

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