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METAL-NONMETAL TRANSITION IN THE LAYER COMPOUND 1T-$Ta_{1-x}Ti_xS_2$

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Metal-nonmetal-metal transition in the pseudo-binary 1T-$Ta_{1-x}Ti_xS_2$ compounds with $0.01 \leq x \leq 0.144$ has been clarified from the low temperature specific heat above 1.5 K together with the electrical resistance down to 20 mK in the magnetic field range from 0 to 90 kOe. Especially the behaviors of the electronic component and a lower temperature anomaly in the specific heat give a strong support on the realization of the Anderson localization.

I. Introduction

Since its electrical resistance below 2.2 K was found to show the exponential dependence on temperature on the basis of the variable-range hopping in the Anderson localized states according to Di Salvo and Graebner[1], a renewed attention has been paid in the layer transition metal dicalcogenide 1T-TaS_2 system. Soon after we found[2] a very large negative magnetoresistance in the variable-range hopping regime below 0.1 K. Furthermore our study extended to the pseudobinary system discovered the metal-nonmetal transition [3].

In this paper, we present further investigations on several Ti-doped samples 1T-$Ta_{1-x}Ti_xS_2$ by measuring the temperature $\rho(T)$ and the magnetic field dependence $\rho(H)$ of the resistivity together with the specific heat at low temperatures. Emphases are laid upon the detailed behavior near the metal-nonmetal transitions and the existence of the Anderson localization.

II. Experimental

The 1T-$Ta_{1-x}Ti_xS_2$ single crystals (x=0.01, 0.03, 0.04, 0.05, 0.06, 0.07, 0.09 and 0.144) were prepared by an iodine vapour transport method. The $\rho(T)$ and $\rho(H)$ were measured down to 20 mK and up to 90 kOe. The electrical resistance was measured by a d.c. four probe method above 1.5 K and by an a.c. four probe method in a dilution refrigerator. The current parallel to the layer plane was flowed so that the power dissipated in the sample is less than 10^{-13} Watt at the lowest temperature studied. Furthermore, the low temperature specific heat on the several samples was measured above 1.5 K by use of a thermal relaxation method.

III. Results and Discussions

Figure 1(a) shows the $\rho(T)$ as a function of $T^{-1/2}$. In Ti-doped samples, the resistivity below 1 K increases with x and, after

taking the maximum value at x=0.09, the ρ value for x=0.144 returns to almost the same value as ones for the low concentration samples ($x \leq 0.04$). Here it should be remembered in such low temperatures that all Ti-doped samples are in the nearly commensurate charge density wave (NCDW) state, while the "pure" sample (x=0) is in the commensurate CDW state.

In lower ($x \leq 0.04$) and the highest (x=0.144) concentration samples, the ρ(T) increases with decreasing temperature from room temperature to a certain temperature which decreases with increasing x. Then the ρ(T) decreases down to about 1 K and becomes constant below 1 K. These constant values depend a little on concentration but do not exceed $\rho \simeq 10^{-2}$ Ωcm for every samples. Such a sample is metallic in that the ρ(T) remains to be finite.

The value of 10^{-2} Ωcm corresponds to the resistance of 2×10^{5} Ω per square per layer, which is larger than the maximum metallic seet resistance $R_{2D}=3\times10^{4}$ Ω/□ in two-dimensions[4]. This fact can be understood by taking into account an energy dependence of conductivity just above a mobility edge E_c, as pointed out by Yosida and Fukuyama[5]. However, if all electronic states in the two-dimensional system are localized due to scattering by impurities as indicated by Abrahams et al.[6], our result showing the existence of metallic states should be explained as the three-dimensional property due to interlayer correlations, where a universal maximum metallic resistance does not exist.

The ρ(T) in four samples with $0.05 \leq x \leq 0.09$ depends on both temperature and Ti concentration. It continues to increase down to the lowest temperature studied as the temperature decreases. An exponential dependence on temperature, as

$$\rho(T) \propto \exp(T_0/T)^{1/n} \qquad (1)$$

with n=2, holds well for two samples with x=0.07 and 0.09 as well as the

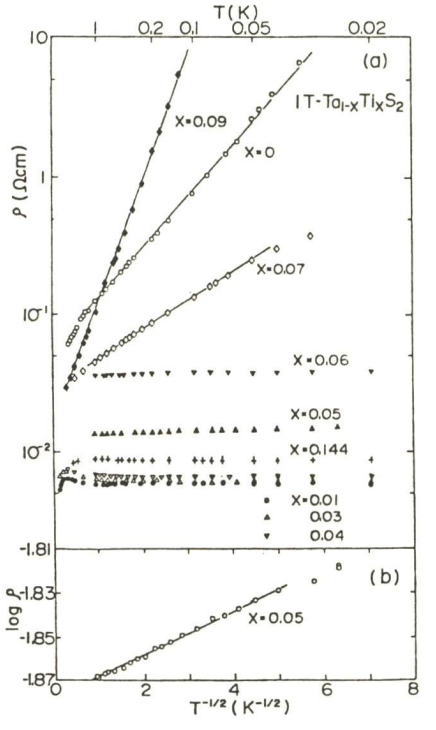

Fig. 1 Semilogarismic plot of the electrical resistivity against $T^{-1/2}$

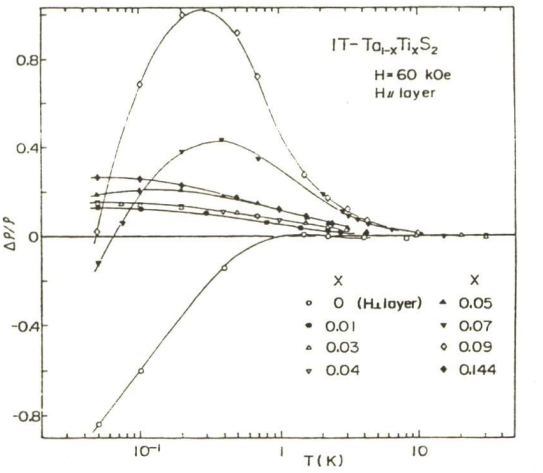

Fig. 2 Temperature dependence of the transverce magnetoresistance at H=60 kOe

pure sample. Such a temperature dependence is explained as due to the variable-range hopping in the Anderson localized states.

The rise of the resistivity for samples with x=0.05 and 0.06 is very small (e.g. $\rho(0.05\ K)-\rho(1\ K)/\rho(1\ K)\sim 0.1$ for x=0.05). However the $\rho(T)$ is well represented by eq. (1) in a wider temperature region as clearly seen in Fig. 1(b). Thus we conclude that the variable-range hopping occurs in these samples below 1 K.

In Fig. 2 shown the transverse magnetoresistance ($\Delta\rho/\rho=(\rho(H)-\rho(0))/\rho(0)$) under a magnetic field of 60 kOe as a function of temperature together with the result of the pure sample[1]. The field configuration is parallel to the layer plane of samples. In metallic samples, the $\Delta\rho/\rho$ is always positive below 10 K and it increases monotonically with the temperature decrease. It tends toward a saturation below 0.1 K.

The magnetoresistance for two samples (x=0.07, 0.09) in the localized regime rises steeply down to 0.5 K, attains to a maximum at 0.2-0.4 K. Then it decreases rapidly and becomes even negative at the lowest temperature. Also in the sample with x=0.05, a small maximum appears at about 0.1 K. It seems that the sample just enters the localized regime from the metallic one.

These anomalous behaviors of magnetoresistance in both metallic and localized states are qualitatively understood by taking into account both the Zeeman shift of the occupied states and the change of the mobility edge due to the magnetic field according to the YF model[5], which assumed the energy dependence of conductivity above E_c in order to explain the magnetoresistance in metallic states. However, we need to consider respectively the contribution from localized electrons below E_c in metallic regime and the contribution from excited electrons in localized regime. The consideration on both effects seems to become very important in such samples near transition concentration as x=0.05 and 0.06.

The specific heat data for several samples are shown in Fig. 3 as C/T vs T^2 in the temperature range $1.5\leq T\leq 5$ K. The specific heat of all the samples can be fitted by a usual form

$$C=\gamma T+\beta T^3 \qquad (2)$$

above 3.5 K. The dashed line gives a least square fit for each sample. However, it should be noted that this fitting procedure cannot avoid ambiguity because of an excess specific heat at low temperature. The value of β gives a Debye temperature of 252±8 K except 266 K for x=0. The γ value at x=0 is small but not zero. This suggests that the most part of the density of states disappears due to the formation of the CCDW induced gap. Observed γ values for samples with $0.01\leq x\leq 0.05$ vary little, with about 2.5 mJ/mole·K^2. It decreases to 1.0 and 0.7 mJ/mole·K^2 at x=0.07 and 0.09, re-

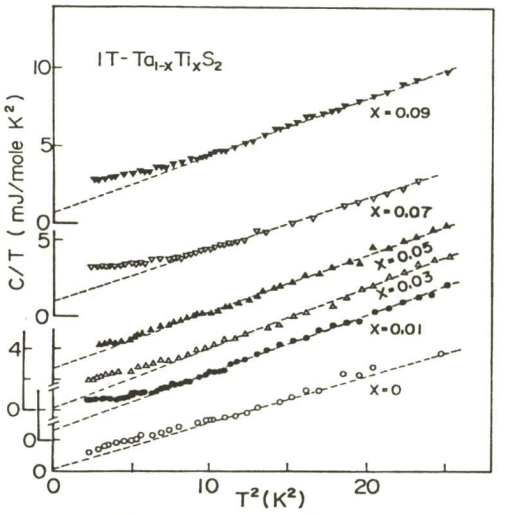

Fig. 3 Specific heat plotted as C/T vs T^2

spectively. The existence of small γ values supports that the metal-nonmetal transition in this system is understood by the Anderson picture.

In all samples, an upward departure from eq. (2) is observed below 3.5 K. It depends on the Ti concentrations and is the largest at x=0.07. This concentration dependence seems to suggest that the departure does not related with impurities but with the Anderson localization. The magnetic field dependence was further examined for x=0.07 and a shift of the departure to a higher temperature is observed under the magnetic field of 60 kOe. Such a specific heat anomaly was reported on the heavily P-doped Si according to Kobayashi et al.[7].

If we assume the anomaly as due to the single occupied electrons in the Anderson localized states, we expect a Schottky-type behavior with spin S=1/2. Then we get the numbers of spin of $N_s \sim 10^{19}$ cm^{-3}, effective fields H_{eff}=21 kOe without external field and H_{eff}=39 kOe with H_{ext}=60 kOe. The value of N_s is very smaller than the carrier numbers $n \sim 3\times10^{21}$ cm^{-3} determined from the Hall coefficient at 78 K.

IV. Conclusions

The electrical resistance as functions of both temperature and magnetic field was measured on several 1T-$Ta_{1-x}Ti_xS_2$ samples with $0.01 \leq x \leq 0.144$ and the specific heat on some of them. These samples were classified into metallic and nonmetallic groups, where the resistivity is devided into below and above a critical value of $\rho = 10^{-2}$ Ωcm. The resistivity of nonmetallic samples shows the characteristic temperature dependence due to the variable-range hopping in the Anderson localized states.

The metallic and nonmetallic samples are also quite different with each other in the behavior of the magnetoresistance. In the formers, the magnetoresistance is positive and increases monotonically with decreasing temperature, while in the latters it decreases at very low temperatures and particulaly becomes negative for x=0.07 and 0.09. These behaviors are qualitatively explained according to the YF model.

The specific heat results also support the existence of the Anderson localization mechanism in the nonmetallic samples. Thus metal-nonmetal transitions are twice observed in the concentration range $0.04<x<0.05$ and $0.09<x<0.144$. These transitions are understood on the basis of the Anderson picture.

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