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## RESISTIVITY OF TmSe UNDER PRESSURE AT VERY LOW TEMPERATURE

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**Résumé.**— Nous décrivons des mesures de résistivité d'un échantillon presque stoechiométrique sous des pressions allant jusqu'à 6 kbars, jusqu'à une température de 25 mK. A 6 kbars, la résistivité atteint 120 mΩ.cm à 30 mK alors que, au-dessus de  $T_N$ , elle est de 1,65 mΩ.cm à 4,2 K. La résistivité croît exponentiellement avec la pression à très basse température et linéairement au voisinage de  $T_N$ . Nous comparons ces résultats à la variation sous pression de l'aimantation d'un sous réseau mesurée par diffraction neutronique et à la résistivité d'autres composés de valence intermédiaire.

**Abstract.**— Resistivity measurements of a nearly stoichiometric sample up to 6 kbar and down to 25 mK are reported. For 6 kbar, the resistivity  $\rho$  rises up to 120 mΩ.cm at 30 mK whereas just above the ordering temperature  $T_N$  at 4.2 K its value is 1.65 mΩ.cm. The increase of  $\rho$  is exponential with the pressure at very low temperature and linear in the vicinity of  $T_N$ . Comparisons are made with the pressure dependence of the sublattice magnetization measured by neutron diffraction and with resistivity of other intermediate valence compounds.

The low temperature properties of the intermediate valent (IV) compound TmSe have been studied extensively in the last few years /1-7/. The occurrence of a magnetically ordered ground state and the main features of the magnetic phase diagram have been well documented by various experimental methods ; in particular, the magnetic structure in zero magnetic field was shown by neutron diffraction to be simple type I antiferromagnetic (AF) /2/.

As in many IV systems, the electrical transport properties are very anomalous due to the presence of a narrow 4f band at the Fermi level. The resistivity of nearly stoichiometric TmSe in the paramagnetic state increases on cooling from  $\sim 200 \mu\Omega.cm$  at room temperature to about 1500  $\mu\Omega.cm$  at 4.2 K /4-5/ with an almost logarithmic term between 6 and 40 K. This has been argued on qualitative grounds to originate from the Kondo effect. In zero applied field, the resistivity  $\rho$  jumps at  $T_N$  and increases very rapidly down to 10 mK where it may attain 14 mΩ.cm /6/. The intrinsic amplitude of this anomaly is unknown since it is dramatically reduced by small deviations

from stoichiometry. This effect is destroyed by the magnetic field ; above the critical field of the metamagnetic transition, the resistivity in the FM phase lies below the RT value /7/. There is presently no definitive explanation for this rather sharp transition to a much higher resistivity below  $T_N$  at  $H = 0$ . The few pictures which have been proposed previously fall in two categories :

i) those which consider the AF structure to be responsible for the poor conductivity below  $T_N$  ; the situation would be closed to that described by Adler /8/ for  $Ti_2O_3$  for which it was believed that  $Ti_2O_3$  presented an antiferromagnetic transition ; ii) those in which some type of correlations ("Kondo effect") tends to localize the carriers ; in the later case, the ordering would only enhance the localization or push some parameter through a critical value.

In the past, only magnetization had been measured under pressure ( $P < 10$  kbar) in the ordered phase /9/ ; the authors' interpretation was a progressive conversion of  $Tm^{2+}$  into  $Tm^{3+}$  up to the fully trivalent

state obtained by extrapolation for  $P = 20$  kbar. This explanation was later criticized by Chouteau et al./10/ as the high temperature susceptibility under pressure shows no conversion of  $Tm^{2+}$  ions into  $Tm^{3+}$  ions. Vettier et al./11/, have recently interpreted the results of neutron diffraction experiments under pressure performed up to 20 kbar by assuming a constant valence mixing, a smooth variation of the exchange parameter between the Tm ions and a strong variation of a single ion correlation energy called  $T_K$  by analogy with abnormal Ce compounds. The main results of these neutron diffraction experiments are :

- i) the persistence of the type I AF structure up to 20 kbar with an excellent scaling of the sublattice magnetization by a  $T/T_N$  law ;
- ii) the initial positive increase of the ordering temperature  $T_N$  with the  $\partial T_N / \partial P \sim 0.09$  K/kbar followed by a flat regime between 8 to 20 kbar .

In this context, it seemed of a great interest to investigate the pressure effect on the transport properties. The main questions are :

- i) how the Log T term is modified ?
- ii) are the jumps of  $\rho$  and  $T_N$  simultaneous ?
- iii) is 14 m $\Omega$ .cm a limit value for the very low temperature resistivity.

Other striking points like the comparison between the properties of stoichiometric samples under pressure and non stoichiometric samples of the same lattice parameter will not be discussed here.

We have performed resistivity measurements down to 25 mK under pressure on a sample which was previously measured at  $P = 0$  (sample 1 of the ref./6/ ; its lattice parameter is 5.712 Å. The pressure cell is described in reference /12/. For  $P = 0$ , the resistivity inside the cell is found identical to that previously reported for the sample located directly in the mixing chamber of the dilution refrigerator. At room temperature, pressures up to 10 kbar have been applied ; with this initial

value, the final pressure at low temperature is  $P = 5.7 \pm 0.3$  kbar .

Our data at room temperature are in agreement with those reported in ref./4/ ; the resistivity decreases 1.26 % per kbar. Under pressure,  $\rho(P)$ , which was initially weaker at 300 K than  $\rho(0)$ , increases more rapidly than  $\rho(0)$ . The curves  $\rho(0)$  and  $\rho(4 \text{ kbar})$  cross each other near 35 K. The proof of the coincidence between the jump of  $\rho$  and  $T_N$  has been verified ;  $T_N$  increases with  $P$  as reported in the refs./9, 11/.

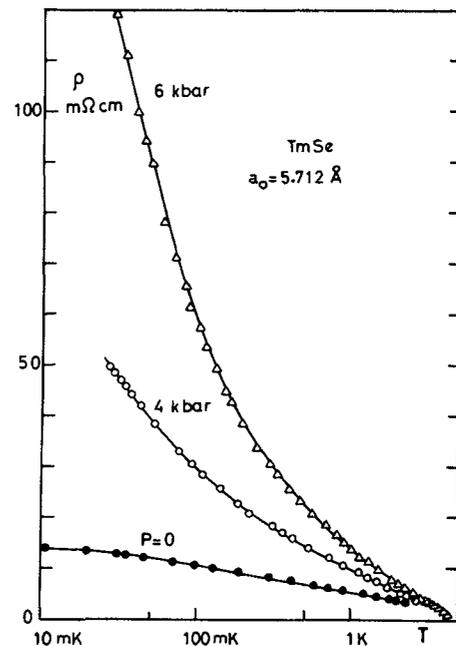


Fig. 1 : Resistivity of TmSe as a function of Log T for three pressure 0, 4, 6 kbar .

Figure 1 represents the low temperature regime of  $\rho(P)$  as a function of Log T. The high value measured at 30 mK for  $P \sim 6$  kbars shows clearly that the resistivity can rise up to a rather high value for a metallic conduction. Below  $T/T_N \sim 0.6$ , the temperature variation of  $\rho$  is strongly pressure dependent. It must be emphasized that at low temperature, for example 30 mK, our data can be represented by an exponential law :

$$\text{Log } \frac{\rho(P)}{\rho(0)} \sim 0.4 \text{ per kbar,}$$

whereas near the critical temperature, the variation is almost linear :

$$\frac{\rho(P) - \rho(0)}{\rho(0)} \sim 0.08 \text{ per kbar at } 3 \text{ K} < T_N$$

$$\sim 0.02 \text{ per kbar at } 4.2 \text{ K} > T_N$$

Since Vettier et al./11/ have reported that under pressure the sublattice magnetization is well fitted by a  $T/T_N$  law we have attempted to represent the difference  $\Delta\rho(T) = \rho(T) - \rho(T_N)$  below  $T_N$  as a function of  $T/T_N$ . No scaling law in  $T/T_N$  has been found in the resistivity data over a significant range of  $T/T_N$ .

At low temperatures several fittings of  $\Delta\rho$  (activation or hopping) have been tried for the different pressures. No law can be found which fits the data over a significant range of temperatures. A Log  $\rho$  versus Log  $T$  plot shows a phenomenological power law dependence in  $T^{-\alpha}$  with respectively  $\alpha = 0.2, 0.4, 0.6$  for  $P = 0, 4, 6$  kbar. An attempt to fit the experimental curve in the vicinity of  $T_N$  by an activation law  $\Delta\rho = \rho_0 \exp \Delta/kT$  with a BCS like gap  $\Delta = \Delta_0 \sqrt{1 - T/T_N}$  fails for  $T/T_N < 0.9$ . It must be pointed out that the variation of  $\rho$  below  $T_N$  seems directly connected to the steep decrease of the specific heat  $C$  found below  $T_N$  which have been recently observed by Berton et al./13/ (almost a  $\lambda$  type anomaly at  $T_N$ ); the proportionality of  $\Delta\rho$  with  $C^{-1}$  has been verified down to  $T/T_N \sim 0.7$  and up  $T/T_N \sim 0.98$ .

The increase of the Log  $T$  dependence with  $P$ , the steep jump of  $\rho$  with  $T_N$  and the enormous variation of  $\rho_{30\text{mK}}$  with  $P$  support strongly the conclusions given in reference (6) and emphasize the possibility of approaching the TmSe properties by an extrapolation of a Fermi liquid Kondo phenomenon. An open question is whether the  $\rho(T)$  dependence below  $T_N$  is due: i) to an intrinsic mechanism (a metal-semiconductor transition at  $T_N$ ) plus an impurity conduction which limits the divergence of  $\rho$  at low temperature /14/ or ii) to an intrinsic mechanism (localization) in the whole temperature range. Can the necessity to achieve a constant mixing on each site by tunneling lead to a minimum of the conductivity at  $T = 0$ ?

Since there is no simple exponential law in the temperature dependence of  $\rho$  and since the  $\rho$  vs  $T/T_N$  does not fit a scaling law, it seems that two mechanisms must be taken into account, as it has been pointed out by magnetic (10) and neutrons (11) measurements. The first mechanism is related to the 4f electronic delocalization (the Kondo like parameter of f-d hybridization); the second reflects the tendency toward magnetic order (the Neel temperature). Via the first mechanism, a metal insulator transition can occur as the total number of electronic carriers (f or d) is equal to one. In this picture, the antiferromagnetic order increases the gap opening. Fruitful comparisons can be made between the low temperature resistivity of TmSe and that of bulk metallic SmS or SmB<sub>6</sub> which present IV properties and resistivity anomalies /15,16/

The problem of the temperature dependence of the resistivity in an IV compound is far from being experimentally and theoretically understood even without the occurrence of an antiferromagnetic phase. A theoretical discussion is given by Kasuya et al for SmB<sub>6</sub> /16/; the same difficulty is found here in understanding and representing resistivity at low temperature by usual models. The present result suggests that in the resistivity of TmSe hybridization may play an important role as it does for metallic SmS and SmB<sub>6</sub> (see ref./17/).

Finally, we point out: i) that at  $T \sim 4.2$  K, the resistivity of TmSe is very near that reported /18/ for fcc ytterbium at 4.2 K,  $P \sim 20$  kbar, inside the so-called metal-semiconductor transition and ii) that the very low temperature pressure dependence of  $\rho$  found here is similar to that observed /18/ for Yb at 4.2 K up to 25 kbar. This asks the question /19/ if the "metal to semiconductor" transition of Yb may not be due to an IV phenomenon

Conclusion.— The resistivity jump occurs always at  $T_N$ , the pressure dependence of  $\rho$  is linear near  $T_N$  and exponential far below. Further experiments may clarify if

the low temperature state of TmSe may be described by an AF Fermi liquid behaviour or an "ordinary" metal insulator transition due to the antiferromagnetic ordering. A main point will be the resistivity of TmSe at high pressure ( $P \gtrsim 20$  kbar) near the vicinity of the  $Tm^{3+}$  configuration.

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