Investigation of metal-semimetal phase transition in the organic metal $(TSeT)_2Cl$ in magnetic field under pressure

V. N. Laukhin and D. N. Fedutin

Division of Institute of Chemical Physics, USSR Academy of Sciences

(Submitted 24 March 1982)
Zh. Eksp. Teor. Fiz. **83**, 1212–1214 (September 1982)

The effect of pressure on the magnetoresistance and on the conductivity anisotropy of the organic metal $(TSeT)_2Cl$ is investigated at 4.2 K. It is shown that $(TSeT)_2Cl$ becomes metallized under pressure at low temperature because of the abrupt increase in the number of carriers, and not because of the increase of their mobility.

PACS numbers: 72.15.Cz, 72.15.Gd, 62.50. + p

The structure and the main properties of the organic quasi-one-dimensional metal tetraselenotetracene chloride with composition $(TSeT)_2Cl$ at normal pressure were described in Refs. 1 and 2. It was shown in Refs. 3 and 4 that when pressure is applied a low-temperature metal-semimetal phase transition is gradually suppressed, and at $P \approx 5$ kbar a transition occurs into a new metallic state that remains stable when cooled down to $T \approx 1.3$ K. We present in this paper the results of measurements of the magnetoresistance of $(TSeT)_2Cl$ at 4.2 K and of the anisotropy of its conductivity in the pressure interval 0–6 kbar. It is shown that metallization of $(TSeT)_2Cl$ in a phase transition under pressure takes place at low temperatures because of the abrupt increase of the number of carriers, and not because of an increase in their mobilities.

The measurements were performed on single crystals with characteristic transverse dimensions 20–40 μm and length 1–2 mm. Details of their synthesis are described in Ref. 5. The pressure was produced in apparatus of the "piston + cylinder" type made of the nonmagnetic alloy 40KhNYu, and was fixed at room temperature.[4] Measurements of the magnetoresistance were made in fields up to 50kOe at transverse and longitudinal orientations of the magnetic field relative to the crystal axis that has high conductivity (the "*c*" axis). The anisotropy of the conductivity of the crystals at $T = 4.2$ K and at $P = 0$–6 kbar was estimated by the Montgomery method.[6]

Figure 2 shows the field dependence of the transverse magnetoresistivity

$$\rho_\perp = [R(H_\perp) - R(0)]/R(0)$$

at atmospheric pressure and at $T = 4.2$ K for two $(TSeT)_2Cl$ crystals simultaneously synthesized immediately prior to the described measurements. It can be seen that, just as in Ref. 2, in fields $H \gtrsim 120$ kOe the magnetoresistivity varies almost linearly. Its value, however, is appreciably higher than in Ref. 2, and reaches $\sim 13\%$ for several crystals. The apparent reason is that the samples investigated by us were purer and more perfect. Evidence favoring this assumption is the fact that the conductivity maximum observed in them at atmospheric pressure is higher than that of the samples investigated in Ref. 2.

It must be noted that in the course of storage under normal conditions the quality of the crystals deteriorates gradually. The result is a decrease both in the maximum on the conductivity and in the value of the magnetoresistivity.[1] There is also a scatter (up to $\sim 30\%$) in the values of $\rho_\perp$ for crystals obtained in the same synthesis (Fig. 1). This is probably due to the differences in the qualities of samples seemingly obtained under identical conditions.

Figure 2 shows the field dependence of $\rho_\perp$ at various pressures ($T = 4.2$ K). We note that the $\rho_\perp(H)$ dependence under pressure is close to quadratic. The obtained data show that $\rho_\perp$ depends little on the pressure in the region where the semimetal phase exists ($P \lesssim 4$ kbar). On the other hand, in the region of the phase transition into the metallic state at $T = 4.2$ K($4 < P < 5$ kbar) the value of $\rho_\perp$ decreases abruptly by more than 5 times (Fig. 3).

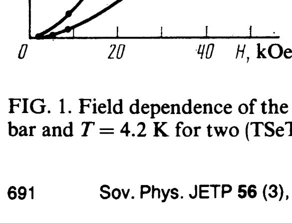

FIG. 1. Field dependence of the transverse magnetoresistance at $P = 1$ bar and $T = 4.2$ K for two $(TSeT)_2Cl$ crystals.

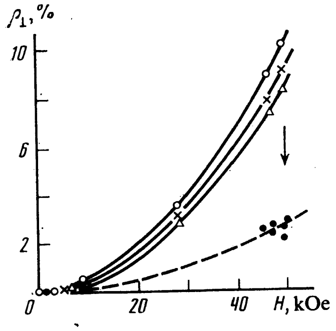

FIG. 2. Dependence of the transverse magnetoresistance ($\rho_\perp$) of the $(TSeT)_2Cl$ crystal at $T = 4.2$ K and at different pressures: × — 2,3, ○ — 3,3, Δ — 4,2, ● — 4,8 kbar. Dashed-proposed $\rho_\perp(H)$ dependenced at 4.8 kbar (see the text).

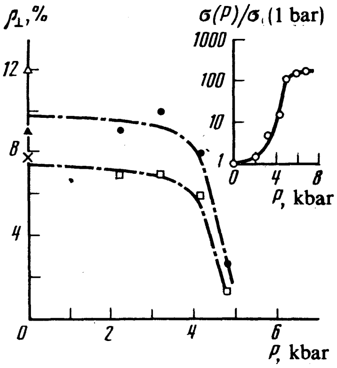

FIG. 3. Pressure dependence of transverse magnetoresistance for two $(TSeT)_2Cl$ single crystals (●, □) at $T = 4.2$ K and $H = 50$ kOe. The values of $\rho_\perp$ at $P = 1$ bar (Δ, ▲, ×) are given for three other single crystals from the same synthesis (see the text). The inset shows a typical pressure dependence of the $(TSeT)_2Cl$ single crystals at $T = 4.2$ K.

In this transition, the conductivity of the complexes increases substantially (see insets of Fig. 3 and Ref. 4). This makes measurements of $\rho_\perp(H)$ very difficult. We therefore measured $\rho_\perp$ in the metallic phase (at $P \approx 4.8$ kbar) mainly near the maximum field 50 kOe. The relative falloff of $\rho_\perp$ in the region of this phase transition is the same for different $(TSeT)_2Cl$ samples. This indicates probably that the abrupt decrease of $\rho_\perp$ on going into the metallic state depends little on the number of impurities and defects, and is due to the singularities of the considered phase transition under the influence of pressure.

The magnetoresistance in a longitudinal magnetic field ($\rho_\parallel$) at $T = 4.2$ K turned out to be close to zero for all the crystals in the field interval 0–50 kOe and at pressures 0–6 kbar. This allows us to conclude that $\rho_\perp$ is connected with the usual galvanomagnetic effects. We can therefore estimate approximately the change of the density n and of the mobility μ of the carriers as the $(TSeT)_2Cl$ crystals go from the semimetallic phase to the metallic, by starting from the very simple relation

$$\rho_\perp - \rho_\parallel = H^2 c^{-2} \mu_\parallel \mu_\perp,$$

where the longitudinal and transverse mobilities are connected with the corresponding conductivities:

$$\sigma_\parallel = ne\mu_\parallel, \quad \sigma_\perp = ne\mu_\perp.$$

For the semimetallic phase at $P = 1$ bar, taking $\sigma \approx 10^3$ $\Omega^{-1}\mathrm{cm}^{-1}$, $\sigma_\parallel/\sigma_\perp = 50$ (Ref. 2) and $\rho_\perp = 0.12$ in a field $H = 50$ kOe we obtain $n = 1.3 \times 10^{18}$ cm^{-3}, $\mu_\parallel \approx 5 \times 10^3$ cm^2/V·sec, and $\mu_\perp \approx 100$ cm^2/V·sec.

It can be seen from these estimates that the carrier density in $(TSeT)_2Cl$ is changed by the pressure from the small value $\sim 1.3 \times 10^{18}$ cm^{-3}, due apparently to impurity states, to a value $\sim 0.5 \times 10^{21}$ cm^{-3}, which is close to the initial electron density in the complex. The mobilities $\mu_\parallel$ and $\mu_\perp$, in both the metallic and semimetallic phases, remain of the same order. Thus, hydrostatic affects the carrier density more than their mobility.

In conclusion, we thank E. F. Yagubskiĭ, A. I. Kotov, and N. D. Kushch for supplying the samples, I. F. Shchegolev for helpful discussions, and Yu. S. Karimov for help with the measurements of the conductivity anisotropy of the crystals under pressure.

1) The magnetoresistance of the $(TSeT)_2Cl$ complexes synthsized more than 3 years ago and investigated in Ref. 2 is at present ~1–2% in a field 50 kOe.

---

[1] S. P. Zolothukin, V. F. Kaminskiĭ, A. I. Kotov, *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **25**, 480 (1977) [JETP Lett. **25**, 451 (1977)].
[2] S. P. Zolotukhin, Yu. S. Karimov, and I. F. Shchegolev, Zh. Eksp. Teor. Fiz. **76**, 378 (1978) [Sov. Phys. **49**, 192 (1979)].
[3] V. N. Laukin, I. A. Kotov, M. L. Khidekel', I. F. Shchegolev, and E. B. Yagubskiĭ, Pis'ma Zh. Eksp. Teor. Fiz. **28**, 284 (1978) [JETP Lett. **28**, 260 (1978)].
[4] V. N. Laukin and I. F. Shchegolev, Zh. Eksp. Teor. Fiz. **78**, 2332, (1980) [Sov. Phys, JETP **51**, 260 (1980)].
[5] O. N. Eremenko, S. P. Zolotukhin, A. I. Kotov, M. L. Khidekel', and E. B. Yagubskiĭ, Izv. AN SSSR, ser. khim. **7**, 1507 (1979).
[6] H. C. Montgomery, J. Appl. Phys. **42**, 2971 (1971).

Translated by J. G. Adashko