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Insulating, conducting and superconducting states of (TMTSF)$_2$AsF$_6$ under pressure and magnetic field (*)

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Résumé. — Nous avons déterminé le diagramme de phase de (TMTSF)$_2$AsF$_6$ sous pression hydrostatique (1 bar < $P$ < 12 kbar), entre 0,1 K et 300 K, par des mesures de résistivité dans la direction de haute conductivité. Nous avons montré qu'il existe un point « triple » situé aux environs de $T_c = 1,22$ K, $P_c = 10,5$ kbar et établi la réentrance de la phase supraconductrice sous le domaine de stabilité de la phase semi-conductrice.

De plus cette étude sous champ magnétique fait apparaître une forte anisotropie des champs critiques transverses $H_{c2}^t$ ainsi qu'une variation très importante de ces champs au voisinage du point « triple ». Ces résultats suggèrent que nous avons affaire à une supraconductivité de type particulier. Nous avons mis en évidence également les propriétés remarquables de la magnétorésistance transverse. Nous proposons d'interpréter celles-ci en admettant d'une part que l'instabilité supraconductrice se développe à partir de 30 K environ (du fait de la faible dimensionnalité du système électronique) et d'autre part qu'elle n'est pas éliminée complètement par l'instabilité SDW responsable de la phase semi-conductrice (du fait de couplages électron-électron dominants).

Abstract. — We report resistivity measurements along the high conductivity axis of (TMTSF)$_2$AsF$_6$ for various hydrostatic pressures up to 12 kbar, and for transverse magnetic fields of up to 70 kOe, in the temperature range 0.1 K-300 K. We derive a phase diagram which shows a « triple » point near $T_c = 1.22$ K and $P_c = 10.5$ kbar, and a reentrant behaviour of the superconducting state inside the semiconducting state. Moreover we observe a striking anisotropy in the transverse critical fields $H_{c2}^t$ and large changes in these fields near the « triple » point. These features suggest we are dealing, in these compounds, with non-usual superconducting properties. The transverse magnetoresistance also shows very interesting features including a large anisotropy and a very similar behaviour in the conducting and semiconducting phases. These peculiarities might be rationalized if one assumes (on account of the low dimensionality of the electron system) that first the superconducting instability can grow from below about 30 K and secondly that it can coexist to some extent with the competing SDW instability responsible for the semiconducting phase due to the existence of significant electron-electron couplings.

1. Introduction. — (TMTSF)$_2$AsF$_6$ is the second quasi-one-dimensional organic conductor, among the (TMTSF)$_2$X family discovered by Bechgaard et al. [1], which has been shown to undergo a superconducting transition near 1 K under a 12 kbar pressure [2].

This property was first observed in (TMTSF)$_2$PF$_6$ [3] and today we know at least 6 salts of this family which display superconductivity [4, 5]. All these salts are assumed to have the same structure characterized by slightly dimerized stacks of quasi planar TMTSF molecules along the a-axis, arranged in sheets parallel to the (a-b) plane and separated by anion sheets. The high conductivity observed in the a direction is attributed to carbon and selenium π orbital overlaps giving rise to a band 0.5 eV to 1 eV wide. The unit cell along the stacking direction contains two TMTSF molecules with a weak dimerization of the average intermolecular distance [6, 7]. Hence, resulting from the 2 to 1 stoichiometry of the salt, the conduction band is half-filled. The prototype (TMTSF)$_2$PF$_6$ which has been the most extensively studied of these

materials up to now has the following characteristic properties:

(i) the low temperature semiconducting phase is associated with a spin density wave (SDW) state \([8, 9]\) with no sign of precursor effects;
(ii) high pressure stabilizes the conducting state which in turn leads to a superconducting transition above about 9.5 kbar \([10]\);
(iii) large magnetoresistance develops below about 30 K in the conducting as well as in the insulating state \([3, 11]\).

Following preliminary studies of the P.T. phase diagram of \((TMTSF)_2PF_6\) it was noticed that the phase-boundary between the insulating and superconducting phases was ill-defined and that the possible coexistence of these states could not be ruled out \([10, 12]\).

In this paper we present a detailed investigation of the \((TMTSF)_2AsF_6\) phase diagram, using resistive, magneto-resistive and low temperature techniques.

2. Experimental. — The single crystals used were prepared by an electrochemical technique as described by Bechgaard \emph{et al.} \([1]\). Typical sizes are 3 mm for the \(a\) direction and 0.2 mm for the perpendicular directions. The resistivity measurements along the high conductivity axis have been achieved by the usual method. Silver paste is used for the electrical contacts to the sample; its ends are covered completely with paste for the current leads and voltage is picked up all around the sample in order to reduce the unnested/nested voltage ratio (typically in the range 0.01-0.03 and always less than 0.1). The current is sufficiently low to insure ohmic behaviour and to prevent heating effects at the contacts. In all cases the current density is less than 0.5 A/cm\(^2\). The current frequency is 88 Hz and the in-phase voltage is measured with a phase sensitive detector.

Two samples are placed together within a teflon cell containing isopentane as pressure transmitting medium. The experiments have been performed either with a \(^4\)He cryostat or with a \(^3\)He-\(^4\)He dilution refrigerator. In the first case the sample holder is placed within a two stage Cu-Be pressure bomb and the pressure is kept constant on cooling down through the solidification of the isopentane; in the second case we used a clamp-typed device with an arrangement of springs to compensate for loss of pressure on cooling. The pressure on the samples was deduced from a preliminary calibration with a manganin gauge put in the teflon cell at ambient temperature; the loss of pressure associated with the solidification of isopentane in the clamp-typed bomb was estimated from the resistance increment of a TTF-TCNQ reference sample.

The main uncertainty in the final pressure determination comes from the imperfect reproducibility of the friction and amounts to about 250 bar.

Among the results we report below, the experiments at 1 bar, 1 kbar, 5 kbar, 8 kbar, 9.75 kbar, 10.25 and 10.65 kbar were made in the \(^4\)He cryostat down to 1.2-1.3 K; the temperature was measured with a copper-constantan thermocouple placed within the teflon cell and below 4.2 K directly from the vapour pressure above the helium bath. The other runs at 9.5 kbar, 10 kbar, 11 and 12 kbar were performed in the \(^3\)He-\(^4\)He dilution refrigerator down to 0.1 K; the temperature was measured with a platinum and two carbon resistance thermometers in close contact with the Cu-Be bomb.

3. Results. — 3.1 Description of the Phase Diagram. — The stability ranges of the conducting, semiconducting and superconducting phases of \((TMTSF)_2AsF_6\) as they can be deduced from our resistivity measurements are shown in figure 1. The characteristic temperature for the conducting-semiconducting transition \(T_{MI}\) has been defined at the maximum of \(-d \ln R/dT\) whereas the superconducting transition has been located at the intersection of the extrapolated normal resistivity curve with the tangent drawn through the inflexion point.

For pressure above 8 kbar the transition from the conducting to the semiconducting behaviour takes place without any discontinuity and therefore we cannot derive any characteristic temperature using the above definition. The phase boundaries near the « triple » point, as they are displayed in figure 1, will be discussed in 3.3 according to the resistivity behaviour in this region. Above 12 kbar the transition line is schematically drawn according to the pressure

![Fig. 1. — The phase diagram of \((TMTSF)_2AsF_6\) under pressure. The conducting-semiconducting transition temperature is defined either by the maximum of \(-d \ln R/dT\) \((P \leq 8\) kbar open dots) or by the criterium \(R(T_{M-I})/R_{\text{min}} = 2\), \((P > 8\) kbar open triangles).]
coefficient determined for (TMTSF)$_2$PF$_6$ [10]. Finally let us point out that no hysteresis effect has ever been detected in this study, but this cannot rule out the conductor to insulator transition from having possibly a small first order character.

3.2 $P \leq 9$ KBAR : THE CONDUCTING-SEMICONDUCTING TRANSITION. — At ambient temperature and pressure the measured conductivity along the $a$ direction varies between 1000 and 400 ($\Omega$cm)$^{-1}$ depending on the samples. It increases by a factor of about 10 on cooling down to 15 K with no adjustments for sample cracks and then drops rapidly below $T_{MI} = 12 \pm 0.2$ K. Down to 4.2 K the decrease of the conductivity by a factor of 40 can be ascribed to a semiconducting behaviour with an apparent activation energy $E_a$ of about 1.8 meV. Lowering temperature below $\sim 5$ K seems to lower $E_a$ rapidly as if there were impurity levels in the gap. However we do not have any serious experimental confirmation for this assumption.

When pressure is applied at ambient temperature, the conductivity first increases continuously at a rate of about 15% kbar$^{-1}$ up to 4 kbar, then it falls by a succession of steps probably resulting from strain induced microcracks; hence the measured conductivity we get near 10 kbar remains about 500 ($\Omega$cm)$^{-1}$. We think that a rise of conductivity could be observed up to 12 kbar or more as in (TMTSF)$_2$NO$_3$ [13, 14] provided the occurrence of these microcracks could be prevented. On cooling under pressure and especially during the solidification of isopentane more stresses are exerted on the samples and their resistance may suffer variable increases. Finally, after the solidification we get effective conductivities ranging from 100 to 1000 ($\Omega$cm)$^{-1}$ at 100 K. Below this temperature the temperature dependence of the resistivity in the metallic-like phase (see Fig. 2) seems to depend on the structural quality of the samples although it is not strictly correlated with the absolute value of the resistivity.

As pressure is increased the conducting to semiconducting transition temperature falls according to figure 1. Moreover the ratio of the activation energy upon the phase transition temperature does not stay pressure independent since under 8 kbar for $T_{MI-1} \sim 7.3$ K, $E_a$ amounts to about 0.45 meV. However, for such small transition temperatures (i.e. at $P \approx 8$ kbar) the derivation of $E_a$ may not be reliable for two reasons :

(i) $E_a$ becomes of the order of the temperature of the experiment;
(ii) the conduction at low temperature may be affected by extrinsic effects (i.e. impurities).

The maximum of $- d \ln R / dT$ is progressively smeared out under pressure and vanishes between 8 and 9 kbar. $T_{MI-1}$ cannot longer be defined with the rule of § 3.1. A low temperature upturn of the resistivity is still observed up to 10.65 kbar but its amplitude decreases at increasing pressures (see Fig. 3). In figure 1, the dash-dot phase boundary above 8 kbar has been obtained with the definition

$$R(T_{MI-1})/R_{min} = 2,$$

using the data of figure 3.

Below 30 K a magnetic field applied along a direction perpendicular to the $a$-axis gives an important magnetoresistance contribution (see Fig. 2) in all

![Fig. 2. — The temperature dependence of the resistivity normalized at 100 K for different samples and pressures: --- $P = 1$ bar; .... $P = 1$ kbar; ----- $P = 5$ kbar; --.--.-- $P = 8$ kbar; ---.-- $P = 10$ kbar. The insert shows a typical log $\rho$ versus $1/T$ plot at ambient pressure and the effect of a transverse magnetic field.](image)

![Fig. 3. — The upturn of resistivity (normalized to the value at the minimum) for pressures just below the « triple » point pressure.](image)
studied samples except if the field is strictly aligned with the $b^*$ direction. We shall describe this magneto-resistive effect in more details in the following sections.

3.3 $9 \leq P \leq 11$ kbar : THE CRITICAL REGION. — In this range of pressure the semiconducting behaviour described above gives way to a partial superconducting transition near $T_c = 1.2$ K. Figure 4 shows the resistivity measured on 3 crystals down to about 0.1 K. An other sample which has been studied at 9.75 kbar down to 1.15 K has displayed the same behaviour with $T_c = 1.2$ K.

The curves presented in figure 4a are very unusual; first because a superconducting behaviour follows at low temperature a semiconducting one, as it was found in (TMTSF)$_2$PF$_6$ [12], but also because the resistivity turns up again and recovers its semiconducting character at lower temperature. This feature can be explained if we take into account a small inhomogeneity of the samples and assume a dramatic vanishing of the superconducting phase when pressure is lowered below about 9 kbar. More precisely we have to assume that different parts of a sample are characterized by phase diagrams differing slightly from the mean phase diagram shown in figure 1. For example, under a pressure of about 9.5 kbar a part of the sample can become superconducting below 1.2 K whereas the rest of the sample remains semiconducting down to the lowest temperature. The resistivity curves show that the sample inhomogeneity results in a relatively narrow dispersion of $T_c$; this implies that the portion of the sample which undergoes a superconducting transition between 1.1 and 0.1 K is very small in any case, which means that the boundary line separating the superconducting phase from the semiconducting phase falls very rapidly towards $T = 0$ between 9.5 and 9 kbar, as we have schematically shown in figure 1. Moreover the combination of the two conductivity mechanisms in series shows that there are no uninterrupted superconducting channels between the two ends of the samples studied.

According to the above description we expect that a magnetic field will convert the superconducting regions into semiconducting ones and restore the monotonic temperature dependence of the resistivity below 4 K. It is indeed what we observe when we apply the magnetic field perpendicular to the $a$ direction (see Fig. 5). Moreover it appears that the effect of the field depends strongly on its orientation in the $(b^*\cdot c^*)$ plane : so it suppresses the superconducting transition much more easily when applied along $c^*$ than along $b^*$. This anisotropy is shown more precisely on the curves which present the temperature dependence of the critical field $H_{c2}$ (Fig. 6), defined by the same convention already used for $T_c$. In the whole temperature range it appears that the critical field parallel to $b^*$, $H'_{c2}$, is about 14 times greater than $H'_{c2}$. More-
over it may exceed the value we can ascribe to the paramagnetic limit [15]. We shall discuss these very striking properties in section 4.2.

We have also observed an anisotropy of similar amplitude for the magnetoresistance of the semiconducting state: for example at 1.4 K with a field of 40 kOe along $c^*$ we found $\Delta \rho/\rho_0 = 4.3$ whereas for a field along $b^*$ we found only $\Delta \rho/\rho_0 = 0.2$ (see Fig. 7).

3.4 $P > 11$ kbar: THE CONDUCTING-SUPERCONDUCTING TRANSITION. — When the pressure applied is greater than 10.8-11 kbar the resistivity displays a metallic behaviour down to the superconducting transition. The results shown in figure 4b have been obtained on the same two samples first measured at 9.5 kbar and presented above in figure 4a. If we compare these two figures it appears clearly that the behaviour of the two samples is similar at 11 kbar but not at 9.5 kbar; in particular the difference in $T_c$ is $\sim 0.02$ K and 0.05 K respectively. This confirms the increase of the slope of the semiconducting-superconducting transition line below about 10 kbar. Note that the sample (1) seems to be less homogeneous than the sample (2) because it displays a two stage resistive transition ending at 1.05 K with a non-zero resistance (its residual resistivity is about 1 $\mu\Omega$cm). Note also in figure 4b the sharp temperature dependence of the « normal » resistivity down to 1 K which now seems to be a typical property of all the quasi-1-D organic conductors in the neighbourhood of the superconducting state and which has been attributed to superconducting fluctuations [16]. In this pressure range the effect of a magnetic field on the transition temperature (see Fig. 8) and on the magnetoresistance (Fig. 9) is again very anisotropic. The ratio $H_{c2}/H_{c1}$ remains about 15 under 11 kbar (Fig. 10) although the critical fields have dropped by approximately a factor 2 below their values under 9.5 kbar. This step decrease of the critical fields will be discussed in section 4.2.

The magnetoresistance of the conducting phase measured at 11 kbar and 2.5 K is $\Delta \rho/\rho_0 = 3.9$ when the 40 kOe field is applied along the $c^*$ direction whereas it is less than 0.15 when $H \parallel b^*$. Moreover upon lowering temperature it increases more rapidly in the first case and thus the anisotropy increases at the same time. Note also that the huge effect of the field in the $c^*$ direction leads to a negative temperature coefficient of the resistance below about 20 K whereas this coefficient remains positive for $H \parallel b^*$ (Fig. 11).
4. Discussion. — 4.1 The phase diagram. — The phase diagram of (TMTSF)$_2$AsF$_6$ under pressure is characterized by a « triple » point situated at about $P = 10.5$ kbar, $T_c = 1.22$ K and by the reentrance of the superconducting phase inside the semiconducting phase. No hysteresis effect has been detected at the transition between these two phases; this suggests that the transition takes place without discontinuity of the free energy. In this fact is confirmed, it could indicate that the semiconducting state (known as a SDW state [17]) gives way without discontinuity to a superconducting state. Within the framework of the Landau theory, the symmetry properties of the electron-gas in its superconducting state would thus be included in the symmetry group of the SDW state. Such a situation is supported by the theoretical work of Machida and Machida and Matsubara [18] although their mean-field treatment is probably inadequate for these quasi-1-D conductors in which we expect the existence of large fluctuation effects (see for example [19]). Note however that this theory can account for the abrupt disappearance of the SDW state and the rapid drop of $T_c$ in the critical region.

4.2 Anisotropy of the critical field and magnetoresistance. — Our measurements have shown a very strong anisotropy of the critical fields $H^*_{c2}$ in the plane perpendicular to the high conductivity axis. This implies that the diamagnetic currents develop much more easily in the $(a-b)$ plane ($H \parallel c^*$) than in the $(a-c)$ plane ($H \parallel b^*$) and therefore flow more easily in the $b$ direction than in the $c$ direction. This agrees with the crystal structure of these compounds in which the TMTSF molecules are stacked along the $a$-axis and form $a$-$b$ sheets, separated by sheets of anion molecules along the $c$ direction [6, 7].

We can also anticipate a large anisotropy between the critical fields along $a^*$ and $b^*$ because of the greater overlap of the selenium $\pi$ orbitals along the $a$ direction: more precisely the ratio of the overlap integrals $t_1/t_3$, estimated from optical reflectance experiments [20] and microwave conductivity measurements [1], is about 30 at 25 K. However a reliable measurement of the critical fields along $a$ is rather difficult to achieve since it requires a very accurate alignment of the small crystals with the field. Murata et al. [21] have undertaken such a measurement in (TMTSF)$_2$ClO$_4$ at ambient pressure and they have found only a small difference between $H^*_{c2}$ and $H^*_{a2}$; however they cannot exclude it comes from a misalignment of $\pm 5$ deg. Gubser et al. [22] have found a much higher value for $dH^*_{c2}/dT$ (70 kOe/K) rather than 8 kOe/K. Moreover our results indicate that the relation between the critical fields and the overlap-integrals could be more intricate than that inferred from a Ginzburg-Landau treatment [19]. As a matter of facts we have shown first, that the critical fields $H^*_{c2}$ can exceed the paramagnetic limit $H_p$ derived within the framework of the B.C.S. theory [15], namely $H_p = 18400$ Oe ($\sim 22$ kOe). Secondly, we have pointed out that both critical fields are decreased by a factor of 2 between 9.5 and 11 kbar, whereas the $T_c$'s are almost $P$-independent. It seems unlikely that such a rate of decrease is only related to an increase of the orbital overlaps and that it can be extended up to much higher pressures. One may speculate that these peculiar features could arise from the proximity of the SDW instability.

The transverse magnetoresistance of (TMTSF)$_2$AsF$_6$ is also very unusual:

(i) It exhibits a strong anisotropy which might be even greater with properly optimized alignment; however the magnetoresistance measured for $H \parallel b^*$ is distinctly above that which could be attributed to misalignment. Note also that this anisotropy is about field independent between 0 and 50 kOe. Recently a similar anisotropy has been reported for the conducting phase [23] and the semiconducting phase [24] of (TMTSF)$_2$PF$_6$.

(ii) It does not saturate, neither for $H \parallel b^*$ nor for $H \parallel c^*$.

(iii) It does not show any clear cut $H^2$ behaviour neither at low fields nor at high fields.

(iv) All the above features are observed in both phases.

Furthermore our measurements in the « triple » point region have shown that the magnetoresistance retains the same behaviour when one goes from the semiconducting state to the conducting state. Several attempts have been made to explain the magnetoresistance of these compounds. As far as the conducting state is concerned Jacobsen et al. [11] have suggested the existence of an anisotropic scattering mechanism, the existence of which is not clearly established. Chaikin et al. [24] have proposed a quasi-two-dimensional model for the Fermi surface of (TMTSF)$_2$PF$_6$ below the metal-insulator transition temperature. Admittedly, a two-dimensional semimetallic band model bearing some similarities with the 3-D Fermi surface of HMTSF-TCNQ at low
temperature \([25]\) could explain most features of the state below \(T_{m-}\). However the \(H^2\) field dependence of the magnetoresistance derived from this model does not seem to agree with the experimental data \([24]\).

In addition the large magnetoresistance of the conducting state has been attributed to the suppression of 1-D enhanced superconducting fluctuations by transverse magnetic fields \([3]\).

The existence of such fluctuations, growing below \(\approx 30\,\text{K}\) down to the establishment of the 3-D superconducting state, is now supported by much experimental and theoretical work (see \([14]\) and references therein). This fluctuating regime should result in an excess conduction (paraconduction). Admittedly the electron transfer being much easier in the \(b\) direction than in the \(c\) direction (see for example reference \([20]\) we expect that the diamagnetic currents should develop more easily in the \((a-b)\) plane than in the \((a-c)\) plane. So a transverse magnetic field applied in the \(c^*\) direction should destroy the fluctuations and consequently suppress the paraconducting contribution more easily than in the \(b^*\) direction. Furthermore when the field along \(c^*\) increases, the superconducting instability might be gradually overwhelmed, by the neighbouring SDW instability, so this mechanism could explain why a 50 kOe field gives rise, at low temperature, to a resistance exceeding the room temperature zero field value. The restoration under magnetic field of a semimetallic (or semiconducting) ground state in \((\text{TMTSF})_2\text{ClO}_4\) has now been clearly established by specific heat studies under magnetic field \([26]\).

One attractive feature of this description is that both magnetoresistance at moderate fields and critical fields are related to the transverse couplings via the diamagnetic currents. So it is not surprising to find roughly the same anisotropy for the two properties. Moreover, we have pointed out that the field dependence, the anisotropy and the order of magnitude of the magnetoresistance do not show any discontinuous change when we go from the conducting phase to the semiconducting one. It is therefore tempting to speculate that some contribution of the 1-D superconducting fluctuations may still remain in the semiconducting state. A magnetic field properly oriented could suppress the 1-D superconducting fluctuations and consequently raise the resistivity. One can notice that neutron or X-ray irradiation represents another way to suppress the paraconduction in the conducting state \([27]\). Irradiation does not restore a semiconducting state (see Fig. 11) since it destroys the transverse coherence which is required for the establishment of a 3-D ordered dielectric ground state.

5. Conclusion. — Our measurements have clearly established the reentrant nature of the superconducting phase below the semiconducting one; the large anisotropy of the transverse magnetoresistance and critical fields \(H^2\). These results suggest that the superconducting instability differs from that more commonly observed in usual metals not only because of its very anisotropic character in the 3-D ordered state but also because it looks as if it could give rise to high temperature paraconducting fluctuations and coaxist, to some extent, with the SDW instability.

The large width of the fluctuation regime is ascribed to the low dimensionality of the electron system \([16]\). The existence of the SDW instability has been attributed to the Umklapp scattering \([28, 29]\). An electron pairing differing from the BCS phonon assisted one has been put forward on account of the interplay between superconductivity and a magnetic state, and also of other experimental data \([14, 30]\). We have shown that our results are consistent with such peculiar behaviours of the 1-D electron system but further developments are still necessary to take them into account and derive quantitative comparison with the experiments.

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References


