Anisotropic Magnetoresistance of Metallic (TMTTF)$_2$Br.

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Abstract. – The metallic state of a high-quality (TMTTF)$_2$Br sample was stabilized under a pressure of 26 kbars. The magnetoresistance of the system was then studied up to 20 teslas for a field rotating in the least-conducting plane. The anisotropy of the magnetoresistance was found to be different from what had been observed in the TMTSF family. For a field along the c-axis, a small kink in the magnetoresistance at $H = 13$ T is detected which may be the first signature of the appearance of the FISDW states.

Introduction. – Over the past 15 years, the (TMTSF)$_2$X family (where TMTSF is tetramethyltetraselenafulvalene and X is an anion: PF$_6$, ClO$_4$, ReO$_4$, ...) has proved to be host to a variety of interesting phenomena. These quasi-one-dimensional conductors are subject to two competing instabilities, namely superconductivity and spin-density-wave. Pressure, by increasing the transverse coupling, favours the superconductivity and suppresses the SDW ground state[1]. A magnetic field along the least-conducting axis, on the other hand, after destroying the superconducting ground state, induces a succession of spin-density-wave transitions (known as FISDW)[2]. Besides this wealthy phase diagram, the slightly warped character of the open Fermi surface leads to other remarkable properties, notably the commensurability effect in the angular magnetoresistance known as Lebed resonance[3-6].

Recently[7], superconductivity was observed in (TMTTF)$_2$Br (where TMTTF is tetramethyltetrafluorafulvalene) which belongs to an isostructural family. The critical pressure to suppress the SDW ground state and to preserve a metallic state (which goes through a superconducting transition at about $T_c = 1$ K) was found to be about 25 kbar (to be compared with the corresponding value, 8 kbar, in (TMTSF)$_2$PF$_6$). This discovery lent support to the idea that both families belong to the same generic class of quasi-one-dimensional conductors where the properties of one compound at a given pressure are analogous to those of another.
compound under higher pressure. From such a point of view, it is natural to expect the occurrence of FISDW transitions at high fields in the pressure-induced metallic state of (TMTTF)$_2$Br. The first study of the high-field magnetoresistance of this system, however, did not reveal any signature of a phase transition up to 19 teslas. But, due to the uncertainty on the precise orientation of the magnetic field and the quality of the sample, the absence of the FISDW transitions was not considered as conclusive.

In this paper we present an experimental investigation of the magnetoconductance for a field rotating in the least-conducting (i.e. (b, c)) plane of a high-quality crystal. A small kink in the magnetoconductance for a field along the c-axis is detected. But the resistivity remains moderate and the question of the existence of the FISDW instability in this system is not set. Moreover, our study reveals that, concerning the angular dependence of the magnetoconductance, (TMTTF)$_2$Br is qualitatively different from the TMTSF family.

Experimental. – Detail of the preparation of (TMTTF)$_2$Br single crystals is described elsewhere[7]. Four-contact longitudinal resistivity measurements were performed on several single crystals. A non-magnetic pressure cell was used to obtain 26 kbars. Most of the samples showed only partial superconducting transition with $T_c's$ of the order of 1 K. One sample with a complete superconducting transition and more than two orders of magnitude of drop in the resistance (fig. 1) was selected for high-field measurements in Grenoble. This sample, with dimensions of $(1.3 \times 0.2 \times 0.1)$ mm$^3$, presented no cracks neither during the rise in pressure nor by the cooldown afterwards.

The high-field measurements were performed using a rotation device in the hybrid magnet of the CNRS-MPI joint High Magnetic Field Laboratory. The relative orientation of the crystal and the magnetic field was obtained using a Hall probe mounted on the pressure cell. Due to a leak in the He$^3$ system, high-field measurements were performed at 1.2 K.

The orientation of the sample was determined first by the crystal morphology and then confirmed by measuring the second critical field, $H_c2$, along the c-axis which yielded a value of 200 Oe at $T = 300$ mK.

![Graph](image)

Fig. 1. – Zero-field resistivity of the sample ((TMTTF)$_2$Br) as a function of temperature, at $P = 26$ kbar. Insert: The superconducting transition yields a mid-point $T_c$ of 1.1 K.
Results. – Figure 2 shows the angular variation of magnetoresistance for various fields rotating in the \((b, c)\)-plane. Surprisingly, the magnetoresistance passes through a maximum in the vicinity of the \(b\)-axis. This is in contrast with the behaviour of the angular magnetoresistance of the Bechgaard salts. Metallic longitudinal resistance, for \((\text{TMTSF})_2\text{PF}_6\)\(^{[5]}\), \((\text{TMTSF})_2\text{ClO}_4\)\(^{[3, 4]}\) as well as \((\text{TMTSF})_2\text{NO}_3\)\(^{[8]}\), is known to pass through a minimum in the vicinity of the \(b\)-axis.

A second difference between \((\text{TMTTF})_2\text{Br}\) and the TMTSF family concerns the fine structure of this angular dependence. Several studies have shown the existence of dips in the magnetoresistance of \((\text{TMTSF})_2\text{ClO}_4\)\(^{[3, 4]}\) and \((\text{TMTSF})_2\text{PF}_6\)\(^{[5, 6]}\) at the so-called Lebed's magic angles. For these angles, the trajectory of the electronic movement becomes periodic and the system's dimensionality decreases. For a triclinic system the magic-angle formula is

\[
\tan \theta = \frac{p}{q} \frac{b \sin \gamma}{c \sin \beta \sin \alpha^*} - \cot \alpha^*,
\]

where \(p\) and \(q\) are integers. In the case of \((\text{TMTTF})_2\text{Br}\), using the ambient pressure room temperature crystal data\(^{[9]}\), one would expect dips at \(\theta = 1.1^\circ (p/q = 0)\), \(\theta = -28^\circ (p/q = -1)\) and \(\theta = 26^\circ (p/q = 1)\). As seen in fig. 2, no dip is found at these angular positions. On the other hand, a clear structure appears with increasing magnetic field. It consists of a peak at the vicinity of the \(c\)-axis \((\theta = 4^\circ)\) and a weaker feature for \(\theta = 45^\circ\).

This peak brings us to the question of the existence of FISDW instability in the metallic \((\text{TMTTF})_2\text{Br}\). Figure 3 shows the longitudinal magnetoresistance for fields along the \(c\)- and \(b\)-axes. While the magnetoresistance is always larger for a field along the \(b\)-axis, there is no feature in the curve for this orientation. On the other hand, for \(H \parallel c\), a small kink is found in the magnetoresistance for \(H = 13\ \text{T}\). The sudden increase in the magnetoresistance for this orientation is thus associated to the emergence of the small angular peak of fig. 2. One can speculate on the possible attribution of this kink to the threshold field for the entry of the system in the FISDW states. Measurements at higher fields are necessary in order to clarify this question.

A last feature of magnetoresistance in TMTSF compounds absent in \((\text{TMTTF})_2\text{Br}\) is the

**Fig. 2.** Angular dependence of the sample resistance for different magnitudes of magnetic field. \(\theta\) is the angle between the field and the \(c^*\)-axis at \(T = 1.2\ \text{K}\). A structure appears at higher fields with a peak emerging in the vicinity of the \(c\)-axis.

**Fig. 3.** Magnetoresistance for two orientations of the magnetic field at 1.2 K and \(P = 26\ \text{kbar}\). Note the small kink for \(H \parallel c\) orientation.
so-called rapid oscillations. These quantum oscillations with $1/H$ periodicity have been reported in the metallic state of selenium compounds\[^{[10]}\] even with open electronic orbits and their origin remains mysterious. As seen in fig. 3, no such oscillations are detected in (TMTTF)$_2$Br.

Discussion. – The outcome of our experiment raises some questions regarding our understanding of the nature of the magnetoresistance and the FISDW transitions in the quasi-one-dimensional conductors.

The origin of the huge magnetoresistance of the metallic state of the Bechgaard salts is a subject of experimental and theoretical research. It has been recently proposed\[^{[11]}\] that the enhancement of the one-dimensional electron-electron Umklapp scattering and antiferromagnetic spin correlations by a magnetic field along the $c$-axis is at the origin of the strong magnetoresistance in (TMTSF)$_2$PF$_6$ and (TMTSF)$_2$ClO$_4$. The unusual angular dependence of magnetoresistance in (TMTTF)$_2$Br does not fit in this picture and suggests that there is some qualitative difference between this compound and members of the TMTSF family.

It is unlikely that the absence of FISDW at moderate fields in this system is related to the relative shortness of the electronic mean free path. Partially quenched (TMTSF)$_2$ClO$_4$ with comparable residual resistivity still display FISDW transitions with a slightly enhanced threshold field\[^{[12]}\]. This comparison is, however, subject to caution because in quenched (TMTSF)$_2$ClO$_4$ the length scale associated to disorder could be considerably higher.

In the so-called standard model of FISDW transitions, the magnetic field by making the system more one-dimensional favours a spin-density-wave transition. Though pressure and magnetic field play opposing roles in this picture, they are related to two different parameters. The critical pressure to kill the SDW state is determined by the deviation from the perfect nesting which is related to the quantity $t'_c = t'_c/a$, where $t_b$ and $t_a$ are band integrals along the $a$ and $b$ directions\[^{[13]}\]. Applying the pressure increases $t'_c$ and when it exceeds a critical value comparable to the energy scale of the SDW transition, the system ceases to be nested enough to go through a SDW transition. The threshold field to induce the FISDW instability, on the other hand, is related to the transverse coupling $t'_c = t'_c/a$. Recent band calculations\[^{[7]}\] showed that while $t'_c$ is roughly the same for the two compounds at ambient pressure, $t'_c$ in (TMTTF)$_2$Br is 3 to 5 times smaller than in (TMTSF)$_2$PF$_6$. Now, at ambient pressure, $T_{SDW}$ is not very different for the two compounds, which means that the same critical value of $t'_c$ is to be attained. This can explain why the critical pressure to suppress the SDW transition is several times higher in (TMTTF)$_2$Br than in (TMTSF)$_2$PF$_6$. It would mean that under a pressure of 26 kbars the value of $t'_c$ is multiplied by a factor of 5 suggesting a strong modification of band parameters by pressure.

The absence of FISDW at moderate fields is not surprising in this scheme, because $t'_c$ at 26 kbars in (TMTTF)$_2$Br would be much higher than the corresponding value in (TMTSF)$_2$PF$_6$ at 8 kbars. Experimentally, the increase in the threshold field of FISDW has been observed in (TMTSF)$_2$ClO$_4\[^{[14, 15]}\]$ and (TMTSF)$_2$PF$_6\[^{[15]}\]$. In the latter compound, the threshold field (about 4 T at 8 kbar) increases roughly as $d\ln H_T/dP = 0.2$ kbar$^{-1}$ so that one would expect a threshold field of about 15 T at 26 kbars. Thus, a large value for the threshold field of the FISDW transitions in (TMTTF)$_2$Br, as suggested by this work, would not oppose the standard model.

Conclusion. – Our experiment highlights the differences between (TMTTF)$_2$Br and other quasi-one-dimensional superconductors. The anisotropy of magnetoresistance is reversed. No Lebed commensurability effect is detected. Finally, the existence of a FISDW instability at higher fields in this system remains an open question.
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