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Negative Magnetoresistance in (TMTTF)$_2$Br

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Abstract. — We report the transverse magnetoresistance measurements at ambient pressure in the organic conductor (TMTTF)$_2$Br in the temperature range between 4.2 K and 40 K and in magnetic fields up to 8.5 T. We have found isotropic, negative and temperature dependent magnetoresistance which becomes negligible, close to 40 K. We interpret the observed behaviour within the picture of strongly correlated quasi-one dimensional systems.

1. Introduction

The highly anisotropic organic conductors (TMTCF)$_2$X (C = Se, S; where TMTSF is tetramethyltetraselenafulvalene, TMTTF is tetramethyltetraethiofulvalene and X is an anion: ClO$_4$, PF$_6$, ReO$_4$, Br...) are the series of isostructural compounds in which a wide variety of phenomena related to the low-dimensional nature of the electronic spectrum are found [1]. These include superconducting, antiferromagnetic (spin-density wave SDW) or spin-Peierls ground state; a number of peculiar magnetic field effects such as a large and anisotropic positive magnetoresistance, the appearance of magnetic field-induced SDW's (FISDW), i.e. a cascade of SDW phases appearing for increasing $H$, etc... It is generally accepted that exchanging Se for S and/or using a different anion X and an external pressure lead to the unified phase diagram for the (TMTCF)$_2$X series (Fig. 1), where the properties of one compound at a given pressure are analogous to those of another compound under higher pressure [2].

The selenium based (TMTSF)$_2$X salts exhibit a high conductivity at the room temperature and a metallic behaviour down to the low temperatures, where the incommensurate SDW ground state is established. The external pressure increases the transverse coupling and above 5–10 kbars the SDW ground state is suppressed in favor of superconductivity. The application of a magnetic field along the least-conducting axis destroys the superconducting ground state and induces the FISDW phases due to a progressive unidimensionalisation of electronic states under field.

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The more anisotropic, i.e. more 1D, sulfur based salts (TMTTF)$_2$X. on the other hand, have a much smaller conductivity at the room temperature, enter a charge localization regime below 200 K, and are very good insulators at low temperatures where the spin-Peierls transition occurs [2,3]. The existence of the resistivity minimum at the temperature $T_p$ is interpreted, within the 1D electron gas theory [4], as a direct consequence of a significantly stronger coupling to the anionic potential via Umklapp scattering, which induces a small dimerization along the stacks [5,6]. The repulsive interactions between electrons lead to the creation of a correlation gap $\Delta_\rho$ and to a semiconducting behaviour below $T_p$. For $T < T_p$ the development of 1D antiferromagnetic fluctuations goes along with the electron localization. The external pressure reduces the dimerization and this decreases $\Delta_\rho$ and shifts $T_p$ towards lower temperatures. The spin-Peierls ground state crosses over to the antiferromagnetic SDW state commensurate with the lattice. When higher pressure is applied, $\Delta_\rho$ is suppressed, $T_p$ is no longer visible (due to a charge delocalization), and the incommensurate SDW state emerges. The antiferromagnetic characteristics of the sulfur series become similar to those of the selenide series and the superconducting ground state can be expected at higher pressures.

Among all the known (TMTTF)$_2$X materials, (TMTTF)$_2$Br has the smallest anion and $c$ lattice parameter [7], the highest conductivity at the room temperature ($\sigma_0 = 100$ $(\Omega \text{cm})^{-1}$) and that is why it occupies a key position in the unified (TMTCF)$_2$X phase diagram. The superconductivity, with $T_C \approx 1$ K, for this compound was observed under 25 kbar [8] (for comparison, the corresponding value was 8 kbar in (TMTSF)$_2$PF$_6$ [1]), thus confirming the predictions of the unified phase diagram. (TMTTF)$_2$Br shows a metallic behaviour down to $T_p \approx 150$ K [7,9], and a semiconducting one below $T_p$. On the other hand, the magnetic susceptibility $\chi_s(T)$ does not show any anomaly near $T_p$ [3], indicating the separation between charge and spin degrees of freedom [6]. Below $T_p$ the charge degrees of freedom become frozen out without the occurrence of any additional lattice or magnetic distortions, while the spin susceptibility is left unaffected by the electron localization. A SDW phase transition

![Phase diagram](image_url)
at $T_N \approx 10 - 15$ K [9] is revealed by conductivity [7], thermopower [10] and susceptibility data [11], while the NMR measurements have shown the existence of a commensurate SDW ground state [12, 13]. The critical pressure where $\Delta_p \rightarrow 0$ for ($\text{TMTTF})_2\text{Br}$ salt is about 5 kbars [14]. At pressures higher than 10 kbars, (where $\Delta_p$ is unimportant and the ground state is the incommensurate SDW), the progressive increase of the positive magnetoresistance with increasing pressure was found [7]. However, the magnetoresistance data at lower pressures and for $T < T_p$, (where the existence of $\Delta_p$ suppresses any single-particle transverse tunneling), have not been presented up to now.

There are few results on the negative magnetoresistance in the organic conductors. In the two-chain organic conductors the magnetic field dependence of the Peierls transition temperature was found [15, 16]. The small isotropic and negative magnetoresistance in TTF-TCNQ [15] below Peierls transition follows a $T^{-2}$ behaviour. This effect was ascribed to the small increase in the charge carrier density caused by the band splitting. The negative and anisotropic magnetoresistance has been observed in the series of (DMTTSF)$_2$X salts [17], which are isomorphous with (TMTTF)$_2$X but show metallic-like behaviour down to the low temperatures, where the resistivity saturates without the occurrence of superconductivity. The magnetoresistance has been interpreted in terms of 2D weak localization due to the disorder in the anion lattice.

In this work we present an experimental investigation of the magnetoresistance of ($\text{TMTTF})_2\text{Br}$ at ambient pressure and for $T < 40$ K. Our data reveal, for the first time, a negative magnetoresistance in one member of the (TMTTF)$_2$X family. The results will be analysed in the framework of correlated quasi-1D conductors [6].

2. Experimental Results

Three single crystals were studied. Samples were mounted in the classic four-in-line array geometry with gold wires stuck with silver paint on pre-evaporated gold contacts (the voltage contacts encircled the crystal, whereas the current contacts covered completely the ends of the samples). The resistance was measured with an on-line dc set-up, with current reversed at each field value. The dc current, along the best conducting direction (a axis) was kept low enough in order to avoid Joule heating and possible non-ohmic effects. When the sample resistance exceeded $10^6$ $\Omega$ (for temperatures below 10 K), the voltage response was measured by electrometer with input impedance $> 10^{14}$ $\Omega$. The linear $I-V$ curves obtained at 4.2 K and 8 K confirmed that all our data refer to the ohmic conductivity region. At each temperature, the magnetic field (up to 8.5 T) was aligned along the $c^*$ and $b'$ directions (i.e. perpendicular to the current).

The room temperature conductivity $\sigma_a$ values for three samples were 55, 72 and 30 ($\Omega$ cm)$^{-1}$ for samples 1, 2 and 3, respectively. Samples were cooled slowly (3 K/hour) in order to avoid irreversible resistance jumps, well known to appear in all organic conductors. We observed very few cracks which have not exceeded a few per cent of the sample resistance.

Figure 2 shows the temperature dependence of the normalized resistance $R/R_{\text{min}}$ (where $R_{\text{min}}$ is the minimum value of the resistance just above $T_p \approx 150$ K) for the three samples measured in this work. All the samples exhibited qualitatively the same behaviour. The temperature dependence of the resistance $\rho(T)$ below 150 K can be analysed using a phenomenological law for a simple semiconductor

$$\rho(T) = \rho_{\text{min}} \exp[\Delta(T)/kT]$$  \hspace{1cm} (1)

in which all the thermal evolution of $\rho(T)$ is included in the function $\Delta(T)$, defined as $T$-dependent energy gap. The prefactor is determined as $\rho_{\text{min}} = \rho(T_p)$ i.e. the resistivity value
Fig. 2. — Normalized resistance $R/R_{\text{min}}$ vs. temperature for the three samples. Inset: The temperature dependence of the activation gap $\Delta_p(T)$ deduced from the resistivity data of the sample 2 with the highest room temperature conductivity value.

just above $T_p$. As mentioned previously, this minimum is attributed to the opening of the correlation gap $\Delta_p$ in the electronic spectrum below $T_p$. Assuming that $\Delta(T) = \Delta_p(T)$ and using equation (1), $\Delta_p(T)$ for sample 2 is given in the inset of the Figure 2. As shown, $\Delta_p(T)$ starts from zero at $T_p$ and reaches the value of about 110 K at the SDW phase transition $T_N \approx 11$ K (this value was determined from our ESR data on the samples from the same batch [18]). Furthermore, there is also a change in the slope of $\Delta(T)$ below 20 K, probably related to the SDW phase transition. For $T < 10$ K the change in the slope of the resistivity towards some saturating value is sample dependent, and this can be attributed to the presence of impurity levels in the semiconductor energy gap. We can therefore assume that below 8 K the conductivity for all our samples begins being dominated by a temperature independent component, which probably results from crystal defects and impurities. This component can be expected to be relatively insensitive to an applied magnetic field, and one can anticipate a vanishing magnetoresistance at very low temperatures.

The magnetoresistance data, $\Delta \rho/\rho = [\rho(H) - \rho(0)]/\rho(0)$, are shown in Figure 3, as a function of applied magnetic field ($H||c^*$ and $H||b^*$) at several fixed temperatures. The magnetoresistance is negative, approximately linear up to 8.5 T and independent of the orientation of the applied magnetic field perpendicular to the current. Such an isotropy suggests that the observed behaviour is dominated by the magnetic field coupling to the spins only, and is not due to the anisotropic effect which would be caused by the orbital coupling.
The temperature dependence of the magnetoresistance for three samples (for $H = 8.5$ K and 4 T) is shown in Figure 4. It can be clearly seen that the magnetoresistance $\Delta \rho/\rho$ starts being observable below 40 K, and its negative value increases to about 18% at 8 K. At 4.2 K the magnetoresistance becomes negligible for all three samples. For two of them, the variation of $\Delta \rho/\rho$ is about the same, whereas for the third sample the magnitude of $\Delta \rho/\rho$ is 20% weaker and vanishes above 30 K. We believe that such a behaviour is due to the lower quality of this particular sample, since it had also the lowest room temperature conductivity value.

3. Discussion

In order to discuss the observed magnetoresistance behaviour, let us again recall the unified phase diagram for organic quasi-1D systems [2] and locate (TMTTF)$_2$Br (Fig. 1). One sees immediately that three temperature scales characterize our material. The highest is the crossover temperature $T_\rho$, at which the charge degrees of freedom start to freeze, which means that the electrons become more and more localized at the lattice sites. This behavior is quite well illustrated (see Fig. 2) by the progressive increase of the energy gap on lowering the temperature, which is deduced from our experimental data and a simple law given by equation (1). The electron localization is driven either by the $4k_F$ scattering process (which is relevant if the band is half-filled), or if there is a $4k_F$-gap $\Delta_{4k_F}$ in a non half-filled band, which is our case. The corresponding bare $4k_F$ scattering amplitude, i.e. the strength of the electron-electron Umklapp process, commonly called $g_3$ in the "g-ology" decomposition of the direct electron-electron
Fig. 4. — The temperature dependence of the magnetoresistance for three samples and for $H = 8.5$ T. For sample 2 the magnetoresistance data are also shown for $H = 4$ T. $\Delta \rho / \rho = 0$ for all three samples at 4.2 K. The full and dashed lines are fit to equation (4) (see text) for $H = 8.5$ T and 4 T, respectively.

interaction, is directly proportional to $\Delta_{4kF}$ [19]:

$$g_3 \approx g_1 \Delta_{4kF} / E_F$$

(2)

where $g_1$ is backward scattering amplitude. This relation is a crude approximation, but as far as we know this is the only one that can be applied to our case.

For temperatures below $T_\rho$ the spin degrees of freedom are decoupled from the charge ones, because of the 1D nature of all the motions [20]. The spins on different chains become correlated at the temperature $T_{x2}$. Since the spin operator consists of electron-hole (e-h) pairs like $\Psi^\dagger \sigma \Psi$, this temperature has the meaning of a two-particle cross-over from 1D to 2D (or 3D) regime. Finally, a phase transition to a 3D antiferromagnet occurs at $T_N$.

The spin-charge separation is exact as long as the electronic spectrum is only linear around Fermi level. The non-linearity can introduce small mixing, and consequently, the effects of magnetic field to $g_3$, as pointed out in [15]. Figure 5 shows typical 1D scattering process in the case of Zeeman splitted linear electronic spectrum, and it is evident that only the spin transfer coupling $g_{1\perp}$ ceases to couple the electrons exactly in Fermi points, i.e. looses its relevance. One should notice that the bosonised Hamiltonian for charge degrees of freedom [20] depends only on Fermi velocity and scattering amplitudes $g_{1||} - 2g_2$ and $g_3$, which are all independent on magnetic field.

Below $T_{x2}$ the problem ceases to be one-dimensional, which implies that the spin-charge separation is not valid any more. This means that the charges start to feel the magnetic field which is coupled only to the spins. In other words, below $T_{x2}$ the spin becomes coupled to the charge, and consequently, the charge fluctuations become field dependent. The effect of the magnetic field to the critical SDW fluctuations has been analysed using RPA [21], but only in the $g_3 = 0$ limit. These calculations have shown that the spectral weight of the fluctuations for the SDW vector parallel to $\mathbf{H}$ is driven upwards (by $2\mu_B H$, $\mu_B$ is the Bohr magneton), from the fluctuations perpendicular to $\mathbf{H}$, i.e. the fluctuations parallel to $\mathbf{H}$ become less critical. (More realistic description for the $T_N < T < T_{x2}$ regime is closer to a strong coupling, and it is expected to be represented rather well by a 2D Heisenberg model). The existence of the additional gap $2\mu_B H$, for spin fluctuations parallel to the field, can be understood more
Fig. 5. — The two-electron scattering processes under Zeeman splitting. In order to preserve momenta, the spin-transferring coupling $g_{1\perp}$ is driven to irrelevance, i.e. it does not exist in the limit when the cut-off around Fermi energy tends to zero. The relevance of other processes does not change.

Fig. 6. — The creation of an opposite- (a) and a parallel-spin (b) electron-hole pair. An additional energy $2\mu_B H$ is needed for the creation of the parallel-spin electron-hole pair.

Intuitively estimating what energy we need to create a spin excitation at $q = 2k_F$ parallel ($\uparrow_+\downarrow$) and perpendicular ($\uparrow_+\downarrow$) to the magnetic field. It is evident from Figure 6, that a creation of e-h pair with opposite spins costs equal energy as for $H = 0$ (a). The creation of a parallel-spin e-h pair at $2k_F$ is not possible if both particles are to be at the Fermi surface (b). The minimum energy needed is $2\mu_B H$, which corresponds just to the field-dependent part of the gap for parallel modes. By constructing the local thermodynamics for spins around $2k_F$, we expect that the parallel-to-perpendicular spin fluctuations ratio will be given by $\exp(-2\mu_B H/k_BT)$, where $k_B$ is Boltzmann constant.

Turning now to our results, let us first emphasize that, since one-electron motion is 1D in all regimes, the only possible coupling of electrons with magnetic field is the Pauli one. The fact that there is no orbital coupling leads to the isotropic magnetoresistance, independent of field direction. This is well confirmed by our results, shown in Figure 3.
The negative magnetoresistance (cf. Fig. 4) is due to the mentioned suppression of fluctuations parallel to $\mathbf{H}$ in the critical regime. Namely, the 1D electrons have less antiferromagnetic fluctuations to scatter at, i.e. the only left are the two modes perpendicular to $\mathbf{H}$. We can calculate the magnetoresistance by using the self-energy ($\Sigma$) corrections due to the two perpendicular and one parallel spin modes. Figure 7 shows the diagrams taken into account. The bosonic lines are $2k_F$-spin fluctuation propagators, and indices $\|\)$ and $\perp$ denote their direction in magnetic field. The change of the resistance $\Delta \rho$ in a magnetic field $\mathbf{H}$ is:

$$\Delta \rho = [\rho(H) - \rho(0)] \sim \text{Im} \Sigma(\omega = 0)_H - \text{Im} \Sigma(\omega = 0)_{H=0}$$

(3)

where $\omega$ is the one-electron frequency which we put to zero, since we are interested in DC resistance.

The diagrams in Figure 7 contribute, roughly speaking, as $\exp(-\Delta_{\|}/T)$ and $\exp(-\Delta_{\perp}/T)$ where $\Delta_{\|}$ and $\Delta_{\perp}$ are the gaps for parallel and perpendicular fluctuations respectively [21]. If one assumes intuitively that $\Delta_{\|} \approx \Delta_{\perp} + \xi \mu_B H/T$ (where $\xi$ is a numerical constant of the order of 2), the magnetoresistance is:

$$\frac{\Delta \rho(H)}{\rho(0)} = \alpha \frac{\exp(-2\mu_B H/k_B T) - 1}{\rho(0) \exp(\Delta_{\perp}/T)}$$

(4)

where $\Delta_{\perp} = \Delta_0 \ln(T/T_N)$ and $\Delta_0 \approx 4\pi T \sqrt{\pi/3} \approx 4.33 \cdot T$, and $\alpha$ is a prefactor having the units of resistivity, thus providing the correct dimensionality.

Our data (Fig. 4) show that the negative magnetoresistance is observed for $8 \text{ K} \leq T < 40 \text{ K}$, and $\Delta \rho/\rho = 0$ at $4.2 \text{ K}$. The upper limiting temperature is field independent (i.e. the magnetoresistance becomes negligible for $H = 4 \text{ T}$ and $8.5 \text{ T}$ at about the same temperature) and can be taken as the high-temperature cut-off value for equation (4). Taking $T_N = 11 \text{ K}$, our zero-field resistivity data $\rho(0)$ and fixed field value ($8.5 \text{ T}$) we have calculated $\Delta \rho/\rho(0)$ values from equation (4) for different $\alpha$ (where different $\alpha$ correspond to products of the same $\rho_{\text{min}}$ value and different numerical factors). Once the best agreement was obtained for the data at $8.5 \text{ T}$ (full line, Fig. 4), the same $\alpha$ value was taken for fitting the $4 \text{ T}$ data (dashed line, Fig. 4). One sees that qualitatively a rather good agreement between our experimental points and equation (4) is obtained (for $4 \text{ T}$ and $8.5 \text{ T}$) in the temperature range $18 \text{ K} < T < 40 \text{ K}$, but only with $\Delta_0/T \approx 7$ (i.e. 1.6 times larger value). On the other hand, it has been beyond this approach to calculate the exact form of $\alpha$ (although we believe that it is related to the resistivity due to the spin fluctuations) and thus any further quantitative comparison of the used fitting parameter $\alpha$ would be too presumptuous at this point.

Both calculated curves show a minimum at $\approx 15 \text{ K}$. The field dependence of equation (4) is monotonous and has no extreme, and this minimum is simply due to the fact that in this regime there is a clear change of the slope in the temperature variation of our zero-field resistivity (cf. Fig. 2). Had it been not the case, the calculated curves would continue decreasing as temperature approaches $T_N$. Apart from that, and bearing in mind the crude theoretical assumptions, equation (4) provides a satisfactory qualitative fit for both temperature and field.
variation of the magnetoresistance of (TMTTF)$_2$Br in the region 18 K < $T$ < 40 K, i.e. for temperatures where the antiferromagnetic fluctuations are important.

Equation (4) cannot be applied for $T$ < $T_N$, where the SDW ground state is well defined. It has been predicted theoretically [21] that the opening of the small gap in one of three Goldstone modes below $T_N$ is expected to have direct consequences on the measurable properties like magnetoresistance. The quantitative analysis of these effects is a rather complex problem and remains open for future investigations. Within the presented picture we cannot say what is the magnetoresistance value expected at 4.2 K. However, as already mentioned, the vanishing magnetoresistance at 4.2 K may be simply the consequence of the fact that the magnetic field has no influence on defects and non-magnetic impurities. In that case, when the temperature is increased and approaches the regime with strong fluctuations, the effects of the applied field on the resistance start being visible. Above 8 K (and up to 40 K) the negative magnetoresistance qualitatively agrees with the dependence given by equation (4). Finally, for $T$ > 40 K, the magnetoresistance becomes negligible again. In accordance with our previous discussion, we suggest that this upper (predicted field independent [21]) cut-off temperature is just $T_{x2}$. Our results would thus be the first to show its existence.

4. Conclusion

In conclusion, we report the isotropic and negative magnetoresistance in the organic conductor (TMTTF)$_2$Br at ambient pressure. The magnetoresistance is the largest at about 8 K (18%), decreases with increasing temperature and vanishes above 40 K. We interpret our results as the consequence of the Pauli coupling of electrons with magnetic field that yields to the isotropic magnetoresistance. The negative magnetoresistance is due to the reduced scattering of electrons on the antiferromagnetic fluctuations which develop below the crossover temperature $T_{x2}$. Our calculations reproduce qualitatively the experimental findings which estimate $T_{x2}$ to about 40 K.

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References


[9] It is shown (Ref. [7]) that significant differences are found between samples grown from different chemical solvents. In our case (so called Montpellier samples) $T_p \approx 150$ K and $T_N \approx 11$ K (Ref. [18]).


[18] The vanishing ESR susceptibility and concomitant divergence of the ESR linewidth define the critical temperature $T_N$ of the SD~V phase transition of this samples at 11 K.

