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Negative magnetoresistance in some dimethyltrimethylene-tetraselenafulvalenium salts: A signature of weak-localization effects

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The transverse magnetoresistance of the organic series $(\mathrm{DMtTSF})_2X$ (with $X = \mathrm{AsF_6}$, $\mathrm{PF_6}$, $\mathrm{BF_4}$, $\mathrm{ClO_4}$, and $\mathrm{ReO_4}$) has been investigated in a wide range of temperature and magnetic field, down to 2 K and up to 40 T. The angular dependence of the magnetoresistance on the field orientation has also been systematically studied. A negative, low-field and low-temperature magnetoresistance appears, roughly under 10 T and 20 K, only for the salts whose anion X has a tetrahedral geometry ($X = \mathrm{BF_4}$, $\mathrm{ClO_4}$, and $\mathrm{ReO_4}$). This magnetoresistance vanishes when the field or the temperature is increased. This new phenomenon is interpreted in terms of two-dimensional weak localization possibly due to some disorder in the anion lattice.

INTRODUCTION

In a previous paper[1] we have presented an exhaustive study of the magnetoresistance of different radical-ion salts belonging to the dimethyltrimethylene-tetraselenafulvalenium salt series $(\mathrm{DMtTSF})_2X$, with $X = \mathrm{AsF_6}$, $\mathrm{PF_6}$, $\mathrm{BF_4}$, and $\mathrm{ClO_4}$. Let us recall that these compounds are isomorphous to the Bechgaard salts as they crystallize in the triclinic structure (space group $P\bar{1}$).[2]

At room temperature the cell parameters of the two series $(\mathrm{DMtTSF})_2X$ and tetramethyltetraselenafulvalenium salts $(\mathrm{TMTSF})_2X$ are very similar.[3,4] According to the different transfer integrals given by Abderrabba[5] it is possible to evaluate t_a and t_b:[6]

$$220 < t_a < 245 \text{ meV},$$

$$20 < t_b < 30 \text{ meV}.$$

As an evidence these values are very close to those generally accepted for the Bechgaard salts.

The four studied compounds have metalliclike resistivities. Almost the same at room temperature ($\rho \approx 10^{-5}\ \Omega\,\mathrm{m}$), they decrease from 300 K to about 10–5 K, then saturate with sometimes a slight increase at lower temperatures.[3] Even at very low temperature (under 1 K), no superconducting transition is observed.[3]

To come back to our magnetotransport experiments, we discovered that the monotonic character of the magnetoresistance, even under extreme conditions of temperature (2 K) and magnetic field (40 T), is a good indication for the stability of the metallic state.[1] On the contrary, in the same experimental range, the TMTSF salts exhibit temperature-, magnetic-field-, and pressure-dependent instabilities.[7] However, the most striking and unexpected result in DMtTSF salts is the appearance of a negative magnetoresistance, roughly under 10 T and 20 K, only for the salts whose counterions have a tetrahedral geometry ($X = \mathrm{BF_4}$, $\mathrm{ClO_4}$, and $\mathrm{ReO_4}$). The theory of two-dimensional (2D) weak-localization provides a very good quantitative agreement with the experimental results. Here, again, the difference between the two families of salts is remarkable as such an effect has never been observed in TMTSF salts. At last we shall try to analyze the possible sources of disorder involved in this weak-localization process very unusual in organic conductors.

EXPERIMENTAL RESULTS

Transverse magnetoresistance measurements were carried out from a very high magnetic field (long-decrease pulsed field) reaching 40 T. The samples could be rotated continuously by an angle of 360° around an axis parallel to the current direction, corresponding to the needle axis. The temperature was swept from 300 down to 2 K. Some supplementary details concerning the experimental techniques have been given elsewhere.[8,9]

All the results concerning the salts with $X = \mathrm{AsF_6}$, $\mathrm{PF_6}$, $\mathrm{BF_4}$, and $\mathrm{ClO_4}$ are displayed in detail in the paper already mentioned,[1] but before focusing our attention on the negative magnetoresistance it is necessary to summarize the main features and conclusions from these experiments.

High-field region ($B > 10$ T)

The four salts exhibit some common properties at high field.

(i) The transverse magnetoresistance is positive monotonic, increasing with the field, for any orientation of this one. Even at $T = 2$ K and $B = 40$ T, no sudden change in the slope of the magnetoresistance occurs, which constitutes a good indication for a stability of the metallic state.

(ii) The magnetoresistance generally follows a B^{α} law, but α may vary with the salt and with the orientation.

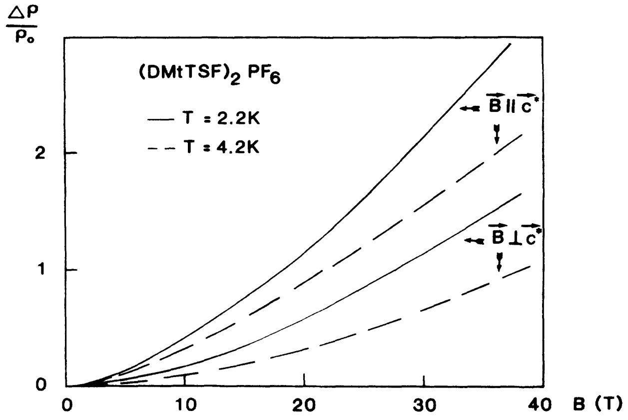

FIG. 1. Transverse magnetoresistance of (DMtTSF)$_2$PF$_6$ vs B for $\mathbf{B}\|\mathbf{c}^*$ and $\mathbf{B}\perp\mathbf{c}^*$ ($\mathbf{B}\|\mathbf{b}'$). $T=2.2$ and 4.2 K.

For example, we have in (DMtTSF)$_2$PF$_6$, $\alpha \simeq 1.7$ for $\mathbf{B}\|\mathbf{b}'$, and $\alpha \simeq 1.5$ for $\mathbf{B}\|\mathbf{c}^*$. In (DMtTSF)$_2$AsF$_6$, $\alpha \simeq 1.4$ for $\mathbf{B}\|\mathbf{b}'$.

Sometimes it is impossible to find a constant value for α in the whole high-field range (10–40 T). This is particularly true in (DMtTSF)$_2$AsF$_6$ for $\mathbf{B}\|\mathbf{c}^*$ where a beginning of saturation seems to happen towards the high-field region.

(iii) The anisotropy of the magnetoresistance $r=(\Delta\rho/\rho_0)_{\max}/(\Delta\rho/\rho_0)_{\min}$ always keeps a rather small value of a few units: for instance in (DMtTSF)$_2$PF$_6$ at 5 T and 4.2 K we measure $r \simeq 5$ [instead of $r \simeq 30$ in (TMTSF)$_2$PF$_6$ under the same experimental conditions[9]]. Compared to the TMTSF salts the DMtTSF ones are weakly anisotropic.

As an example of a typical magnetoresistance, see in Fig. 1 the records corresponding to (DMtTSF)$_2$PF$_6$.

Low-field region ($B < 10$ T)

In the low-field range the behavior of the salts is very much anion dependent.

Salts with *octahedral* anions (AsF$_6$ and PF$_6$) have a positive magnetoresistance following a B^2 field dependence whatever the orientation of the field is. The angular variation $\Delta\rho/\rho_0(\theta)$ is very close to $\sin^2\theta$ (θ being the angle between the field and b' directions) which means that only the component of B perpendicular to the conducting plane (a,b) is efficient, thus confirming the 2D character of the Fermi surface.

On the contrary, the salts containing *tetrahedral* anions (BF$_4$ and ClO$_4$) exhibit an unusual and very surprising anisotropic negative magnetoresistance. This magnetoresistance, having its greatest amplitude when B is parallel to the c^* direction, vanishes very quickly as B approaches b'. At higher fields we get again a positive magnetoresistance as in the octahedral anion salts.

This negative magnetoresistance is also temperature dependent as it disappears when the temperature rises beyond about 20 K. A typical display of these different features is given in Fig. 2 for (DMtTSF)$_2$ClO$_4$ and in Fig. 3 it is given for the salt (DMtTSF)$_2$BF$_4$ which provides

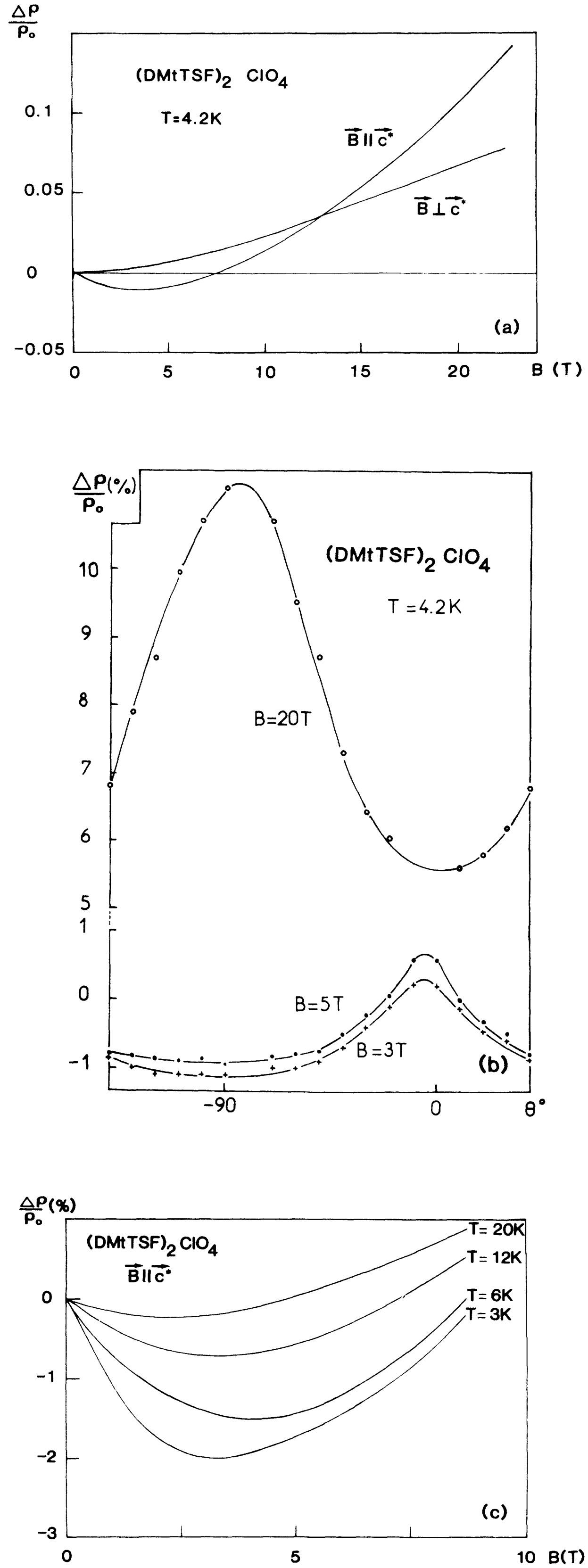

FIG. 2. Transverse magnetoresistance of (DMtTSF)$_2$ClO$_4$ (a) vs B for $\mathbf{B}\|\mathbf{c}^*$ and $\mathbf{B}\perp\mathbf{c}^*$. $T=4.2$ K, (b) vs θ for different values of B, $T=4.2$ K, and (c) vs B for $\mathbf{B}\|\mathbf{c}^*$. $T=3$, 6, 12, and 20 K.

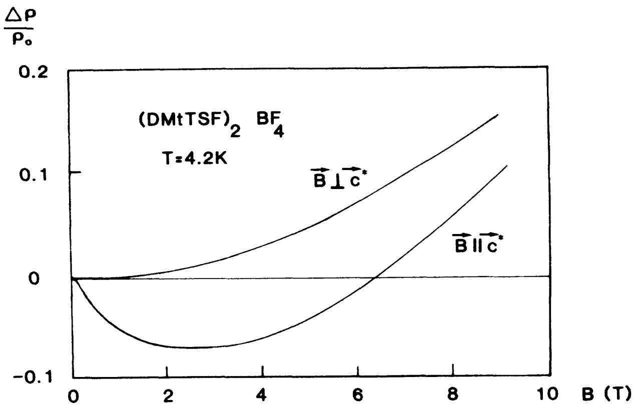

FIG. 3. Transverse magnetoresistance of (DMtTSF)$_2$BF$_4$ vs B for $\mathbf{B}\|\mathbf{c}^*$ and $\mathbf{B}\perp\mathbf{c}^*$. $T=4.2$ K.

very similar effects.

The fact that the magnetoresistance recovers a classical behavior (that is to say positive with a B^α variation) when the negative part vanishes under the influence of any of the two factors, temperature when $T>20$ K or orientation when $\mathbf{B}\|\mathbf{b}'$, is a strong argument for considering the total magnetoresistance as the sum of two contributions: one positive, "normal," almost the same for the four salts, the other, negative, due to some effect to be identified which would dominate at low field,

$$\frac{\Delta\rho}{\rho_0}=\left[\frac{\Delta\rho}{\rho_0}\right]_+ + \left[\frac{\Delta\rho}{\rho_0}\right]_- .$$

This kind of assumption has already been done by different authors for other materials.[10,11]

Negative magnetoresistances are not very rare phenomena as they have been observed in various conductors such as semiconductors,[12,13] metals containing magnetic impurities,[14] and amorphous materials.[15] For the latter the experimental results are very well explained by weak-localization effects arising from the disorder naturally present in the structure.

Coming back to DMtTSF salts, any theory involving magnetic moments to explain the negative magnetoresistance must be rejected as no magnetic atoms are present in these compounds.

The global behavior of this negative magnetoresistance (temperature, field, and orientation dependence) evokes at once localization effects, but in a 2D description. This last point is not at all surprising as we already pointed out that the normal positive magnetoresistance had itself a 2D character.

However, one must not forget that weak localization cannot occur without a certain amount of disorder. Furthermore, we will discuss the possible sources for such a disorder.

WEAK-LOCALIZATION ANALYSIS

Brief theoretical background

The expressions for the 2D conductivity and magnetoresistance due to the weak localization have been established by several authors:[15-17]

$$S_0=S-\frac{e^2}{2\pi^2\hbar}\ln\left[\frac{\tau_i}{\tau}\right]$$

for the zero-field conductivity (in fact a conductance) and

$$S(B)-S_0=\frac{e^2}{2\pi^2\hbar}\left[\ln\left[\frac{B}{B_i}\right]+\psi(\tfrac{1}{2}+B/B_i)\right]$$

for the variation of conductivity under a magnetic field.

The constants involved in these expressions have the following meaning: S denotes the conductivity without localization, τ_i denotes the inelastic electronic relaxation time, τ denotes the elastic electronic relaxation time, $B_i=(\hbar/4eD)\tau_i^{-1}$, $D=v_F^2\tau/2$ (diffusion constant), v_F is the Fermi velocity, and ψ is the digamma function.

For small variations of S and R we have

$$S(B)-S_0=\Delta S\simeq-\frac{\Delta R}{R_0^2} ,$$

with of course $R_0=1/S_0$.

Actually, we consider that the sample is made of a set of conducting sheets having a thickness d, parallel to the highly conducting planes. The sheet resistance R_0 is thus related to the experimental three-dimensional (3D) resistivity $\rho_{0\,\mathrm{expt}}$:

$$R_0=\frac{\rho_{0\,\mathrm{expt}}}{d} .$$

Thus if we follow the hypothesis that the negative component of the magnetoresistance $(\Delta\rho/\rho_0)_-$ is due to the localization, we get

$$\frac{\Delta R}{R_0}=\left[\frac{\Delta\rho}{\rho_0}\right]_- =-R_0\frac{e^2}{2\pi^2\hbar}\left[\ln\left[\frac{B}{B_i}\right]+\psi(\tfrac{1}{2}+B/B_i)\right] .$$

When $B/B_i=x>1$ the digamma function can be approximated by an easy-to-handle form:[18]

$$\psi(\tfrac{1}{2}+x)\simeq\psi(\tfrac{1}{2})+4x\left[\frac{1}{1+2x}+\frac{1}{9+6x}+\frac{85+32x}{150(5+2x)^2}\right]+\ln\left[1+\frac{2x}{5}\right] ,$$

with $\psi(\frac{1}{2})=-\gamma-2\ln 2\simeq-1.964$ (where γ is the Euler's constant).

Calculations and results

Now we can try to fit a theoretical magnetoresistance of the form

$$\left[\frac{\Delta\rho}{\rho_0}\right]_{\mathrm{theor}}=\left[\frac{\Delta\rho}{\rho_0}\right]_+ + \left[\frac{\Delta\rho}{\rho_0}\right]_-$$

to the experimental one. If we restrict ourselves to the low-field region we can take a quadratic positive magnetoresistance as is found at higher temperature ($T>20$ K),

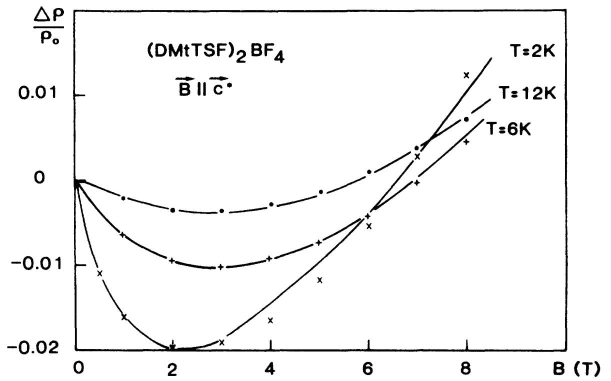

FIG. 4. Best fits for the transverse magnetoresistance of (DMtTSF)$_2$BF$_4$ for $\mathbf{B} \parallel \mathbf{c}^*$. Calculated points are represented by × at $T=2$ K, + at $T=6$ K, and ● at $T=12$ K.

or in the salts with octahedral anions:

$$\left[\frac{\Delta\rho}{\rho_0}\right]_+ = AB^2 .$$

So, to fit to the experimental magnetoresistance we need to determine the three parameters A, B_i, and R_0 at a given temperature.

In practice we fix B_i, then calculate A and R_0 (or d). We retain, for each temperature, the three parameters which allow the best overlapping of the two magnetoresistances (experimental and theoretical).

Figure 4 shows the quality of the fits obtained for a sample of (DMtTSF)$_2$BF$_4$ at several temperatures. All the numerical values are summarized in Table I.

The calculated value of d, around 30 Å at $T=4.2$ K, seems to be a very good order of magnitude compared to the c parameter ($\simeq 14$ Å) in the direction perpendicular to the conducting planes.

On the other hand, to be in a 2D system from the point of view of the localization, it is necessary to have the condition

$$(D\tau_i)^{1/2} \gg d .$$

That means that the carrier free path associated with the inelastic relaxation time cannot play any role for the localization in the d direction if it is out of the conducting volume in this direction.

Taking a mean value of 0.3 for B_i we get $(D\tau_i)^{1/2} \approx 300$ Å which is about ten times larger than d.

TABLE I. Fit parameters for the transverse magnetoresistance of (DMtTSF)$_2$BF$_4$ at $T=2$, 3, 4.2, 6, 12, and 22.5 K.

T (K)	B_i (T)	$\rho_{0\,\mathrm{expt}}$ (Ω m)	A (T^{-2})	R_0 (Ω)
2	0.12	7.7×10^{-7}	7.2×10^{-4}	335
3	0.17	7.7×10^{-7}	6.1×10^{-4}	300
4.2	0.26	7.7×10^{-7}	4.4×10^{-4}	256
6	0.33	7.8×10^{-7}	3.8×10^{-4}	237
12	0.46	8.0×10^{-7}	2.4×10^{-4}	133
22.5	no more negative part	8.8×10^{-7}	1×10^{-4}	

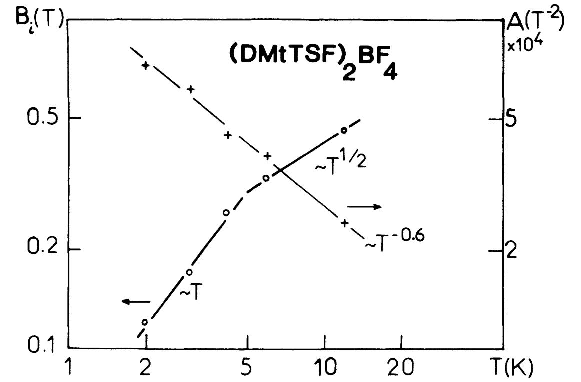

FIG. 5. ln-ln plot of B_i (○) and A (+) vs T for (DMtTSF)$_2$BF$_4$.

A good indication of the validity of calculations is also given by a logarithmic plot of B_i and A versus T (see Fig. 5). At low temperature B_i is proportional to T, then around 5 K there is a change of slope, the dependence becoming close to $T^{1/2}$. This kind of dependence in T^{β} has already been observed in localized systems.[19,20] The coefficient of the positive magnetoresistance A is nearly proportional to $T^{-0.6}$ in the range 2–12 K.

The magnetoresistances of (DMtTSF)$_2$ClO$_4$, treated in the same way, are also in good agreement with the calculated points (Fig. 6). The values of B_i and A (Table II) have the same order of magnitude as for the previous salt, however, these parameters now vary more slowly with the temperature for an unclarified reason (for instance, $A \sim T^{-0.3}$).

Thus we can conclude that the experimental results are very close to the predictions of the weak-localization theory, the agreement being better in the low-field–high-temperature region.

This effect is well known in other materials where the smaller the ratio B/T, the better the agreement between theory and experiments. At intermediate fields it is also possible that the quadratic law B^2 fails to describe the positive magnetoresistance.

An apparent discrepancy occurs for the low-

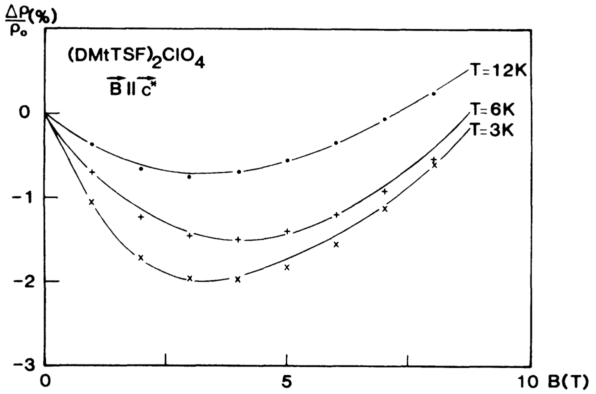

FIG. 6. Best fits for the transverse magnetoresistance of (DMtTSF)$_2$ClO$_4$ for $\mathbf{B} \parallel \mathbf{c}^*$. Calculated points: ×, $T=3$ K; +, $T=6$ K; ●, $T=12$ K.

TABLE II. Fit parameters for the transverse magnetoresistance of $(DMtTSF)_2ClO_4$ at $T = 3$, 6, 12, and 20 K.

T (K)	B_i (T)	$\rho_{0\,\mathrm{expt}}$ (Ω m)	A (T^{-2})	R_0 (Ω)
3	0.39	$\simeq 7\times10^{-7}$	4.5×10^{-4}	445
6	0.45		3.2×10^{-4}	350
12	0.48		2.8×10^{-4}	207
20	0.5		2.3×10^{-4}	100

temperature–zero-field resistivity as many authors report a logarithmic increase of this one with the temperature in 2D structures. It arises from $\ln(\tau_i/\tau)$ appearing in the expression of S_0 if the inelastic relaxation time is assumed to vary like T^{-p} when the elastic one is considered as weakly temperature dependent owing to the high degree of disorder in the materials.

In our case, we cannot measure directly the sheet resistance R_0 but it is obtained as a fit parameter and we see from the different tables that it actually increases when the temperature is lowered, in good agreement with the theory. The measured resistivity $\rho_{0\,\mathrm{expt}}$ keeping an almost constant value, this would lead to a sheet thickness d increasing with T. This result is not at all unreasonable if we think that the conductivity in the d (c^*) direction is rather of an activated type and that the effect of increasing T is to couple more conducting planes giving thus a greater mean value of d.

On the other hand, as we have rather "clean" compounds, certainly less disordered than amorphous materials for instance, it is possible that the elastic relaxation time τ is not truly constant. Under these conditions the resistivity could result from a combined variation of τ_i and τ. But the scattering mechanisms are not well identified in organic compounds and it is very difficult to interpret more physically the results, particularly the kind of crossover observed at 5 K in the temperature dependence of B_i.

DISCUSSION

At this point, even if there is nothing to invalidate the hypothesis of the weak localization to explain the observed negative magnetoresistance, there remains a very important question: What is the nature of the scattering centers which could induce such a localization? The studied compounds have a crystalline structure, not an amorphous one with a natural disorder. We could evoke some impurities being present, but if it was the case there is, however, no reason for the salts with BF_4 and ClO_4 to be less pure than those with AsF_6 and PF_6. But once more, we must recall the importance of the *anion geometry* as the negative magnetoresistance is only associated to tetrahedral counterions. As there was not evidence for an anion ordering we though that the nonperfect periodic potential of the tetrahedral anions randomly oriented could produce the localization. To check the validity of this assumption we studied a salt with tetrahedral but ordered anions: $(DMtTSF)_2ReO_4$.

Magnetoresistance of $(DMtTSF)_2ReO_4$

The salt $(DMtTSF)_2ReO_4$ is different from the other salts as it has a monoclinic symmetry. So the anions are ordered in an antiferromagnetic way along the a and c axes (corresponding respectively to the c and b axes of the triclinic structure).

Between 300 and 200 K the resistivity is similar to that of other salts. Then, lowering the temperature, we fall in the region of resistivity jumps where the behavior begins to change with a resistivity always increasing (instead of decreasing between jumps as observed in the other salts). Figure 7 gives a typical record of the resistivity ρ versus T for a complete temperature cycle.

The study of the magnetoresistance gives at once a striking result: As for the other salts with *tetrahedral* anions we also get a *negative anisotropic* magnetoresistance at low field. However, the maxima of the positive and negative magnetoresistance do not coincide anymore but occur at 90° from one another.

We see in Fig. 8 the low-temperature magnetoresistance of this salt.

All the different features of the negative magnetoresistance in $(DMtTSF)_2ReO_4$ are truly similar to those obtained for the anions BF_4 and ClO_4.

These experiments are very important: they show that the localization effects cannot be due to a completely random orientation of the anions as they appear in a naturally ordered material.

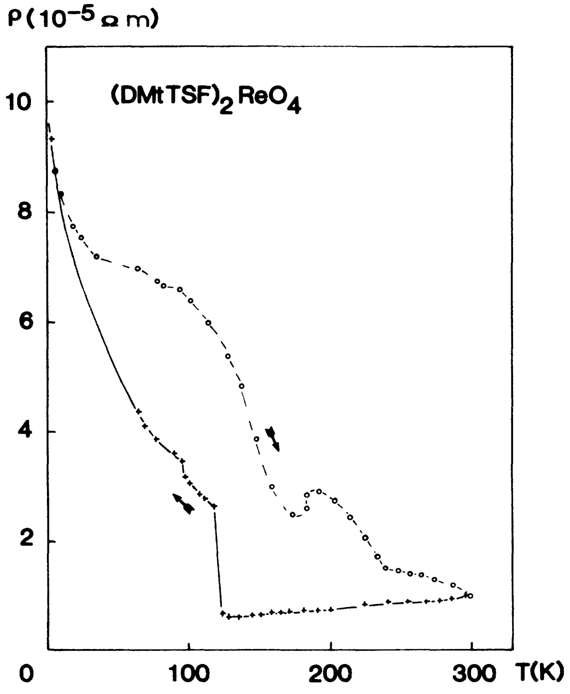

FIG. 7. Resistivity vs temperature for a sample of $(DMtTSF)_2ReO_4$. + are for the way down and ○ for the way up.

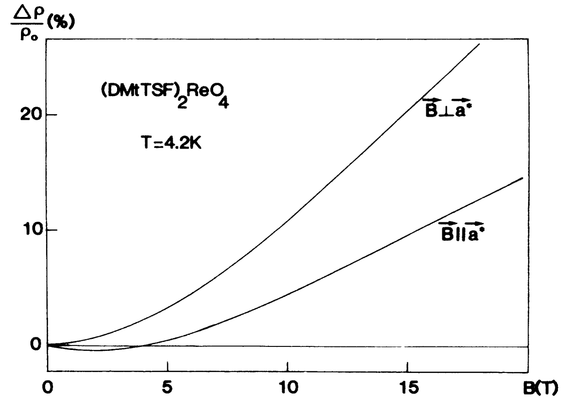

FIG. 8. Transverse magnetoresistance of $(\mathrm{DMtTSF})_2\mathrm{ReO}_4$ vs B for two orientations of the field. $T=4.2$ K.

Origin of the localization

Now, let us review the main data.

The negative magnetoresistance is intimately related to the anion geometry. It was observed in *all* the studied salts with tetrahedral anions, but *only* in these. It depends on the nature of the organic molecule. For instance, in the TMTSF salts, never such a phenomenon has been observed. It is not induced by a total disorder of the anions [a counter example is provided by $(\mathrm{DMtTSF})_2\mathrm{ReO}_4$, already ordered at room temperature]. So we considered the possibility of a certain amount of disorder in the structure, at low temperature, which would break the lattice and potential periodicity.

Trying to clarify the problem of a possible ordering of the anions we did electronic diffraction experiments on $(\mathrm{DMtTSF})_2\mathrm{BF}_4$.[21] We discovered an unexpected *superstructure* $(0,\frac{1}{2},\frac{1}{2})$ already existing at room temperature but vanishing after a few seconds, certainly under the influence of the irradiation. This high-temperature diffraction picture is given in Fig. 9(a): the extra spots corresponding to the superstructure are not visible anymore, the angle between the [001] and $[20\bar{1}]$ directions is close to 93°, in good agreement with a triclinic crystal. Lowering the temperature down to 130 K the lines of extra spots appear more clearly (superstructure $0,\frac{1}{2},\frac{1}{2}$), but at the same time there is a distortion of the lattice seen; for instance, on the previous angle which moves from 93° to 90° [Fig. 9(b)]. These results are very important as they show for the first time the existence of a superstructure that could be due to an antiferromagnetic-type anion ordering. On the other hand, down to the low temperatures the crystal seems to undergo a deformation of its lattice, maybe towards a monoclinic symmetry.

Some structures not well understood have also been observed in $(\mathrm{DMtTSF})_2\mathrm{ClO}_4$.[22]

In $(\mathrm{DMtTSF})_2\mathrm{ReO}_4$, the irreversibility of the resistivity during a temperature cycle together with its increase at low temperature could be explained by numerous cracks producing defects in the material. The DMtTSF salts truly seem to have rather unstable structures which could produce defects and disorder. The dissymmetry of the DMtTSF molecules could also lead to a worse definition of the "cages" containing the anions.[23] In the case of tetrahedral anions the distances Se-O are rather weak. Their influence can be greater than in TMTSF salts. A small amount of either disordered, badly oriented, or slightly displaced anions could make up a set of scattering centers producing the weak localization.

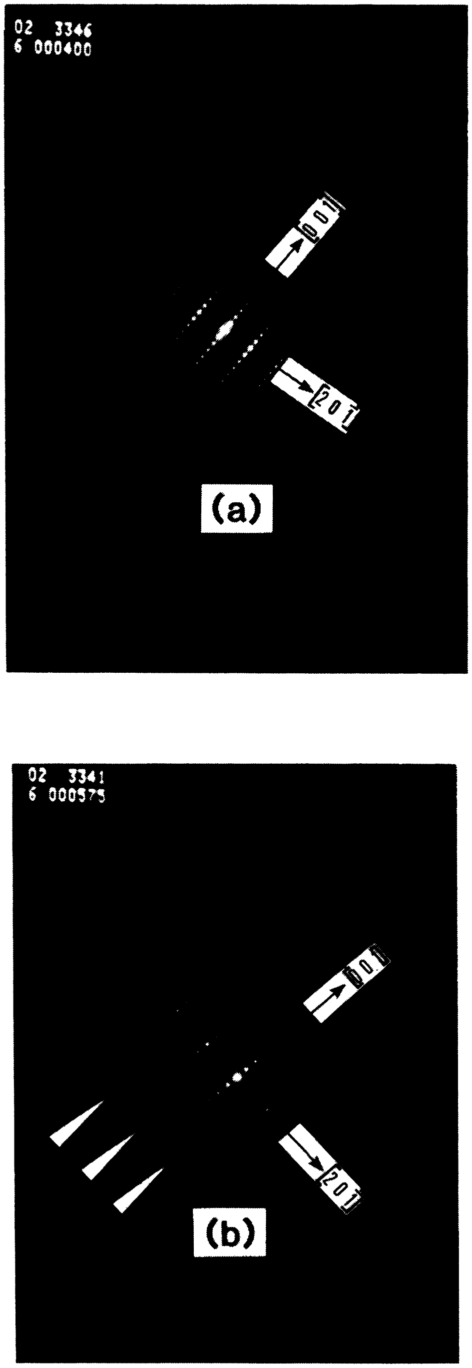

FIG. 9. Diffraction diagrams of $(\mathrm{DMtTSF})_2\mathrm{BF}_4$. (a) T is room temperature, $([001],[20\bar{1}])\simeq 93°$. (b) $T=130$ K, $([001],[20\bar{1}])=90°$. White arrows indicate extra lines related to the superstructure $(0,\frac{1}{2},\frac{1}{2})$.

CONCLUSION

The DMtTSF salts, isomorphous to the Bechgaard salts but so different in their behavior, are a source of numerous questions. Why are they so different and why do they stay so firmly in the metallic state? We think that the weakness of one-dimensional (1D) electronic correlations leading to a 2D character of these salts can partly explain this stability.[1]

The negative magnetoresistance is a very interesting new phenomenon, as it is, to our knowledge, the first observation of weak-localization effects in organic conductors. Its close relation with the presence of tetrahedral anions poses again the problem of the actual structure of these salts (including the problem of the anion ordering and possible defects). The properties of DMtTSF salts seem to result from a competition between weak-localized carriers and free ones. Obviously these compounds would deserve other types of experiments, particularly crystallographic studies, to be more clearly understood.

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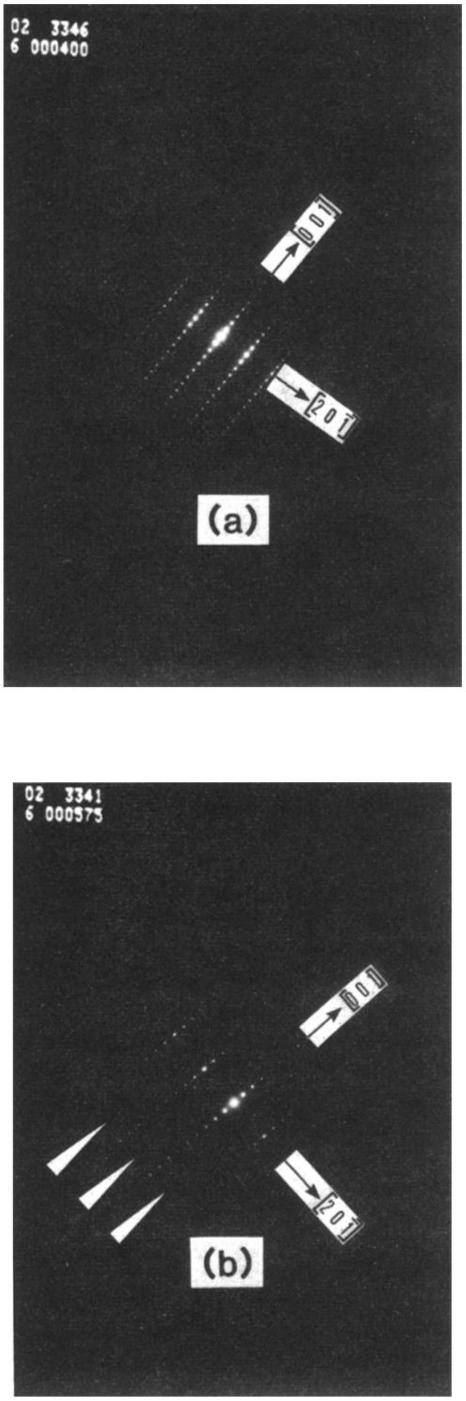

FIG. 9. Diffraction diagrams of $(DMtTSF)_2BF_4$. (a) T is room temperature, $([001],[20\bar{1}]) \simeq 93°$. (b) $T = 130$ K, $([001], [20\bar{1}]) = 90°$. White arrows indicate extra lines related to the superstructure $(0,\frac{1}{2},\frac{1}{2})$.