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Field-induced confinement in $(TMTSF)_2ClO_4$ under accurately aligned magnetic fields

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Abstract. We present transport measurements along the least conducting c direction of the organic superconductor $(TMTSF)_2ClO_4$ performed under an accurately aligned magnetic field in the low temperature regime. The experimental results reveal a two-dimensional confinement of the carriers in the (a, b) planes which is governed by the magnetic field component along the b' direction. This 2-D confinement is accompanied by a metal-insulator transition for the c axis resistivity. These data are supported by a quantum mechanical calculation of the transverse transport taking into account in self consistent treatment the effect of the field on the interplane Green function and on the intraplane scattering time.

PACS. 74.70.Kn Organic superconductors – 73.40.-c Electronic transport in interface structures – 72.15.Gd Galvanomagnetic and other magnetotransport effects

1 Introduction

The major unsolved problem for quasi one dimensional (Q-1-D) organic conductors pertaining to the $(TMTSF)_2X$ series is the determination of the mechanism leading to superconductivity emerging in almost all of them under or without pressure. Based on the early finding of a great sensitivity of organic superconducting state to the presence of impurities and on the possibility of the critical fields being explained by an orbital limitation and consequently not destroyed by a paramagnetic mechanism, there have been a suggestion of triplet pairing superconductivity [1,2]. On the other hand measurements of the critical fields performed along the three principal axes a, b' (normal to a in the (a, b) plane *see* Fig. 1) and $c^\star$ down to 0.5K have revealed the Pauli limiting behavior and therefore the evidence of singlet pairing [3].

Recent experiments of the dependence of the superconducting T_c on the concentration of non magnetic defects in the solid solution $(TMTSF)_2ClO_{4(1-x)}ReO_{4x}$ have conclusively shown that the superconducting gap cannot keep the same sign over the whole Fermi surface [4,5]. However, these experiments are still unable to provide a clue for the spin part of the superconducting pairing, namely a singlet d-pairing or a triplet p or f-pairing. Consequently, additional experiments sensitive to the spin part of the pairing are needed to move one step further. Such an experiment has been performed measuring the spin susceptibility in the superconducting state of $(TMTTF)_2PF_6$ via the ^{77}Se Knight shift [6]. The finding of a constant susceptibility within the error bars crossing the critical temperature has enabled the authors of the reference [6] to conclude in favour of an equal spin pairing, a form of the triplet pairing [7]. Another approach has been used to probe the possibility of equal spin pairing via the measurement of the critical fields of the superconducting state of $(TMTSF)_2ClO_4$. Such data have been obtained recently using torque and resistive determinations of the upper critical field of $(TMTSF)_2ClO_4$ down to 25 mK under a precise alignment of the field along the b' direction [1]. That work has shown that $H_{c2}^{b'}$ can reach 5T at zero temperature i.e a factor about two above the Pauli limit for singlet superconductivity. These data corroborate earlier works in the same material [8] and also in the superconducting state of $(TMTSF)_2PF_6$ under pressure with the same field orientation [9]. However, the b' direction is somewhat peculiar in the $(TMTSF)_2X$ structure since it is a direction which is parallel to the (a, b) planes. Hence, as noticed by Lebed in 1986 a magnetic field-induced dimensional crossover is expected when the magnitude of the in-plane aligned field becomes of the order of the interlayer coupling so that the amplitude of the electron trajectories becomes smaller than the interlayer distance [10]. For an

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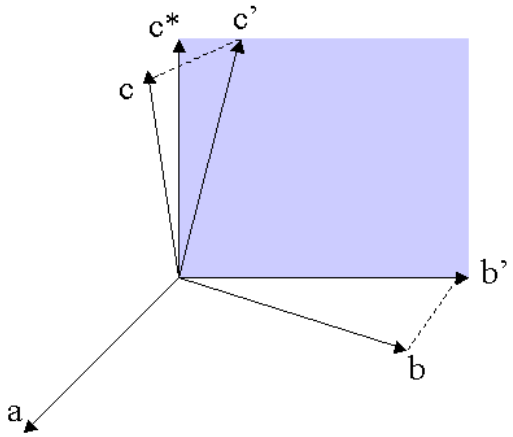

Fig. 1. Schematic representation of the different axes derived from the triclinic symmetry.

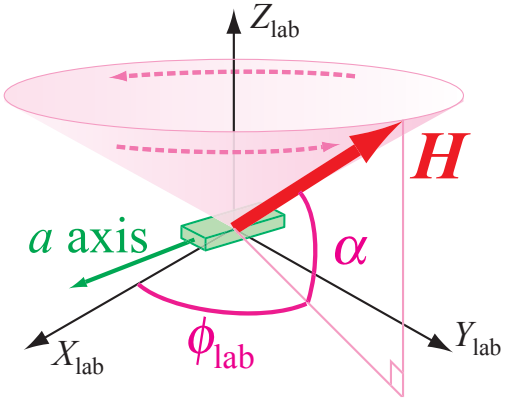

Fig. 2. Schematic representation of the sample orientation in the magnetic field.

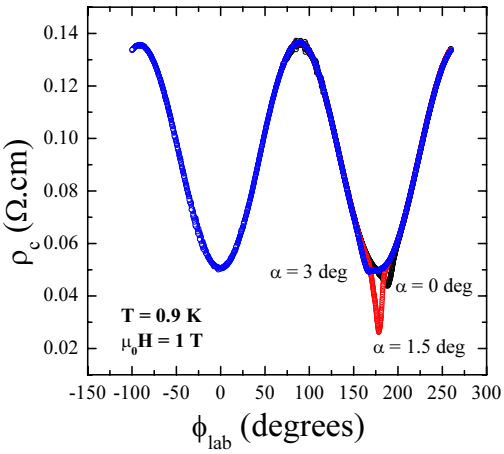

Fig. 3. Procedure of field alignment: the two fold oscillation is dominated by the intrinsic in-plane anisotropy rather than the misalignment of ±1.5 deg during the field rotation. The sharp dip at $\alpha = 1.5$ deg indicates accurate alignment of H//a axis. In this experiment, field and temperature conditions have been purposedely chosen close enough to the critical superconducting conditions in order to obtain a finite but non zero value for the resistance when the field is properly aligned.

in-plane alignment of the magnetic field H_{c2} has been predicted to exhibit an upward curvature at low temperature with a possible reentrance of the superconducting phase in high fields for both singlet and triplet pairing although the latter is more evident [10,11]. From the experimental point of view a negative temperature dependence of the resistance in the normal state has been observed for $H//b'$ in both $(TMTSF)_2ClO_4$[8] and $(TMTSF)_2PF_6$[9]. The case of $(TMTSF)_2PF_6$ is disputed since the measurements require the stabilization of the superconducting phase under pressure and there exists the possibility of a pressure regime in which superconductivity coexists with an insulating spin density wave state [12]. In such a narrow pressure regime all components of the resistivity do reveal a negative temperature behaviour prior to the superconducting transition. However no such objection can be applied to $(TMTSF)_2ClO_4$ in its very slowly cooled low temperature phase. Consequently, the insulating character of the resistivity which is observed in that compound at low temperature under field $//b'$ must be ascribed to a field-induced modification of the electron spectrum. The present situation is such that a determination of the critical fields of $(TMTSF)_2ClO_4$ down to the lowest possible temperatures with the field accurately oriented along the three respective crystal axes on the same sample is still missing.

2 Experimental

As a preliminary study, we have performed an experimental investigation of the role of the magnetic field along the principal axes in the (a, b') plane. Accurate alignment was enabled by a vector magnet system which provides high angular resolution [13] see Figure 2. Furthermore, we are also presenting a calculation explaining the electronic confinement in the (a, b) planes and the non metallic behaviour of the c axis resistivity observed at low temperature when a magnetic field is parallel to the b' direction.

The first step consists in the determination of the principal axes from the magnetoresistance using the anisotropic properties of superconducting state. The sample has been glued on a thin sapphire plate for a good thermal contact with the reservoir. Keeping the sample fixed, the field of 1 Tesla (at the temperature of 0.9 K) has been rotated around the vertical axis in a plane making an angle α with respect to the plane of the sample. The field and the temperature have been chosen following the early data of reference [3] in order to keep the sample superconducting and normal for alignments along a and b' respectively. The angle α providing the accurate alignment of the field parallel to the (a, b) plane is the one which reveals the sharpest and deepest drop of the resistance in the rotation pattern for a given angle ϕ which marks therefore the position of the a axis, see Figure 3.

Next, a rotation of a 0.5 T field has been achieved in a plane perpendicular to the a direction previously determined at different temperatures between 0.8 and 0.9 K.

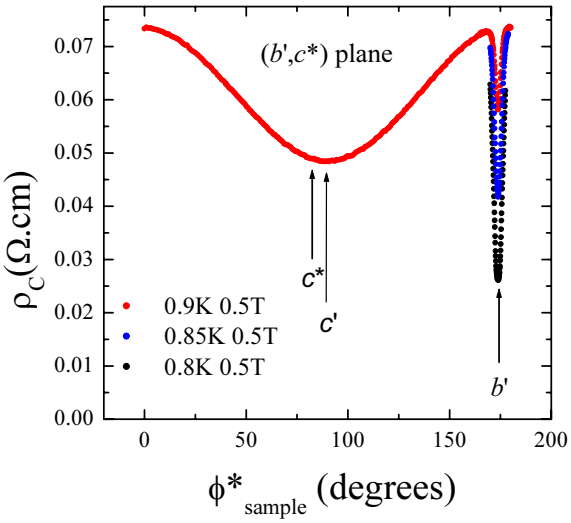

Fig. 4. Field rotation in the plane normal to the sample a axis. The a axis direction was determined by the procedure in Figure 2. The sharp dip is associated with the onset of superconductivity with the field H//b'. It also indicates that a resistivity minimum occurs for H//c' which is 7 deg away from the $c^\star$ direction. The non zero value of the resistance for the b' direction is due to the reasons explained in the caption of Figure 3.

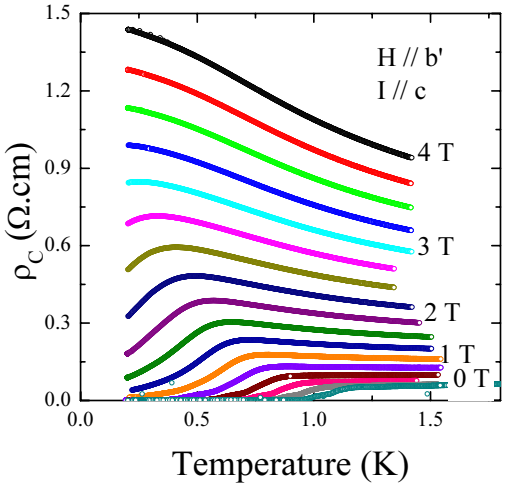

Fig. 5. Transverse resistivity versus temperature for different values of the magnetic field H//b' axis from 0 to 4 T.

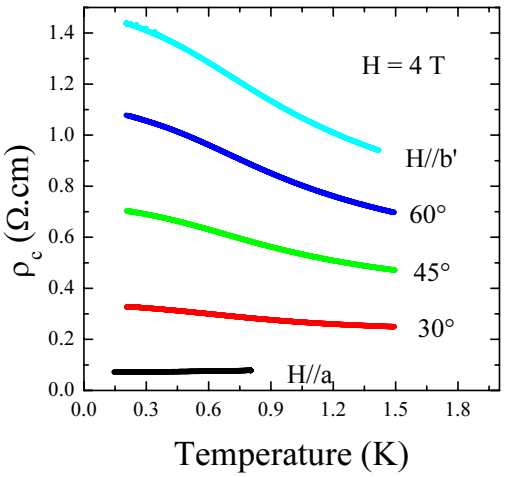

Fig. 6. Temperature dependence of the magnetoresistivity for different angles between a and b' axes under 4 T.

According to Figure 4 a very sharp and strongly temperature dependent resistance minimum is observed at an angle corresponding to the direction of the $b^{'}$ axis while the position of the shallow minimum reveals the direction of the c' axis (the projection of c on the plane normal to a) and not of the$c^\star$ axis. In a second step the temperature dependence of the resistance has been measured along the three previously determined axes at various fields up to 4 T for a and b' and up to 0.18 T for $c^\star$. As far as the a and $c^\star$ directions are concerned the resistance always exhibits a transition between a superconducting state at low temperature and a metallic state at high temperature. Such a behaviour is no longer followed for the $b^{'}$ axis, Figure 5, as a very strong magnetoresistance is observed at high temperature with the resistance changing from a metallic to a non metallic behaviour at fields above ≈1 T. In order to better characterize the phenomenon of localization observed on the c-axis transport under field several temperature sweeps have been recorded at various angles of the field in the (a, b') plane at 4 T, *see* Figure 6.

Figure 6 clearly shows that the metallic behaviour under the magnetic field of 4T is only observed when the field is aligned along a. Actually, comparing the data in Figures 5 and 6 we can show that the localization is governed by the component of the field along the b' direction, see Figure 7. Furthermore, it is interesting to see that the longitudinal resistivity ρ_a retains its metallic character down to low temperatures even at high fields up to 25T parallel to the b' axis, see the insert in Figure 1 of reference [14]. Therefore, the electronic confinement within the (a, b') planes is accompanied by a localisation only for the conduction along the c-axis.

The coupling along the least conducting direction is not known very accurately but it is of the order of ≈5 K or less. Hence, it is likely that the band model may breakdown in the $c^\star$ direction due to thermal fluctuations when k_BT exceeds t_c,namely around liquid helium temperature. This situation is supported by the finding of a small Drude component in the low frequency optical conduction only at low temperature [15] and also by a resisitivity which is smaller than the maximum metallic resistivity (0.06 Ω cm) [16] only when the temperature is in the liquid helium range [4].

In a preliminary approach one can view the $c^\star$-axis transport as diffusive with a probability for interlayer hopping given by $1/\tau_\perp \approx t_\perp^2 \tau_{a-b}$ where τ_{a-b} is an average lifetime in the (a, b) planes, extending a model of interchain to interplane hopping [17,18]. The $c^\star$ axis transport thus reads $\sigma_{c^\star} \approx t_\perp^2 \tau_{a-b}$. Assuming an interplane coupling insensitive to the magnitude of the magnetic field along b' the magnetoresistance along $c^\star$ should in turn follow that of τ_{a-b} (or ρ_a). However, at the temperature of 1.2 K

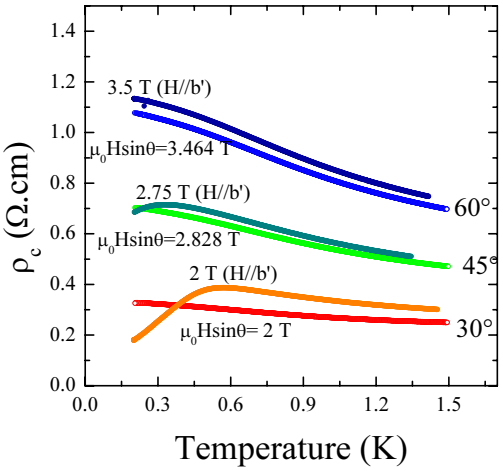

Fig. 7. Contribution to the transverse localisation of the component of the magnetic field along the b'-axis and comparison with similar values when H//b'.

for instance, the magnetoresistance for $\rho_{c^\star}$ amounts to a factor 17.5 in a field of 4 T which is *at variance* with the factor 3.5 observed for the a axis component at similar values of temperature and field [19]. Consequently, we can infer that the interlayer coupling must be affected by the magnetic field and possibly drops by a factor 5 under a field of 4 T along b' as a result of a field-induced confinement and in turn becomes two dimensional (2D) under field.

3 The confinement scenario: theory

In the following, we propose a simple interpretation of the experiments reported above and those of Danner et al. [20]. In particular, we discuss the effect of a magnetic field applied parallel to the (a, b) plane, which does not modify the metallic conductivity in the a direction, but induces a metal insulator crossover along the c direction. Let us precise that we do not discuss the possibility of a superconducting order, and we discard it. We are, here, only interested in the metallic or insulating character of the phase. Our model relies on *three essential features,* which determine the physics of the system, independently of the details of the model: *(i)* the electrons can be described in a *Fermi liquid picture.* Given the experimental conditions, i.e. temperature much smaller than the interplane electron transfer integral t_c, the electronic regime is safely assumed to be three-dimensional and the Fermi liquid approach to be valid; *(ii)* the strong anisotropy of the crystal structure is reflected in the *strong anisotropy of the electronic properties.* The transfer integrals in the three directions exhibit a strong hierarchy, leading to an *open Fermi surface* both in the (k_a, k_b) plane and in the (k_a, k_c) plane; *(iii)* we propose that the effects reported here are due to *an orbital effect of the field*, since only the b component of the field is relevant. As will be discussed below, both in a semi-classical approach and in a fully quantum mechanical treatment, such an orbital effect tends to reduce the dimensionality of the electron system, therefore explaining the transport properties reported here.

To discuss this effect, we consider a simple model described by a mean field single particle Hamiltonian, with a very small interplane electron transfer and a tight binding dispersion relation:

$$\epsilon(\boldsymbol{k}) = v_F(|k_x| - k_F) - 2t_b \cos k_y b - 2t'_b \cos 2k_y b - 2t_c \cos k_z c,$$

where we have taken a linear dispersion around the Fermi level along the chains direction a corresponding to the highest conductivity and with a Fermi velocity v_F. The dispersion in the transverse directions (b, c) is described within a tight binding picture where t_b and t'_b are respectively the hopping integrals to the first and to the second nearest neighbors in the b direction whereas t_c describes the hopping along the least conducting axis c perpendicular to the conducting plane (a, b).

For the sake of simplicity, we have considered here an orthorhombic crystal structure, although the real one is triclinic, but such a simplification should not modify qualitatively the physical picture. The highly anisotropic bandwidths given by: t_a:t_b:$t_c \sim$ 3000 K:200 K:10 K, where t_a is the hopping integral in the a direction, lead to a quasi-1D character to the $(TMTSF)_2X$ compounds.

At ambient pressure and above the superconducting transition temperature $T_c \sim 1$ K, the ground state of the $(TMTSF)_2ClO_4$ is metallic. We consider a parallel magnetic field $\mathbf{H}(0, H, 0)$. Let us first discuss a *semiclassical approach*, which gives an intuitive understanding of the phenomenon. The orbital effect of a field parallel to the b direction will reduce the t_c hopping process, leading to a 3D-2D crossover for an applied field of the order of t_c. This dimensional crossover can be understood by considering the semi-classical equation of motion of the electron wave packet for H//b on the Fermi surface: $d\mathbf{k}/dt = e(d\mathbf{r}/dt) \wedge H/\hbar c_0$. The wave packet trajectory in k space is linear along k_c, with transverse periodic oscillations with an amplitude $\delta k_x = 4t_c/v_F$.This gives rise, in real space, to a linear trajectory along the a direction modulated by periodic oscillations along the c axis with an amplitude $\delta z = 4ct_c/ev_F H$ which decreases as the magnetic field increases, making the electrons confined in the (a, b) plane. For field strength of the order of t_c, the electrons get confined in a single plane perpendicular to c. In this semi-classical picture, we do not expect a strong effect of the field on the conductivity along the a direction. On the contrary, the electron confinement in the (a, b) planes should correspond to a metal-insulator crossover in the c direction. Such a *field induced confinement*, which should not be confused with a *field induced localization*, allows to understand the paradox of a qualitatively different field effect on the conductivity along the a and c directions. As a result the behavior of the c axis resistivity is expected to change from metallic to non metallic as the temperature decreases.

The dimensional crossover has also been described by Strong et al. [21] as the consequence of an interlayer decoupling which happens above a critical field separating

a 3D Fermi liquid, where interlayer hopping is coherent, from a 2D non Fermi liquid where the hopping is incoherent.

Such a model is somewhat similar to another one discussed earlier [22,23] where the field is perpendicular to the most conducting planes. For such a geometry, it has been shown that the orbital effect of the field induces a one-dimensionalization of the electron motion, because it cancels the effect of t'_b and because the transfer integral in the third direction t_c can be neglected above a threshold field of a few Teslas. This model has successfully explained the overall features of the well known FISDW phases. However, the physics for a field orientation perpendicular to the $(a,\ b)$ plane is different from that of parallel field H//b. First, in the latter geometry, the transfer integral along the field direction is not t_c as in the former case but t_b which is much larger than the field, thus precluding the possibility of a SDW instability.

The semi-classical argument given above is fully confirmed by a quantum mechanical treatment. We consider a magnetic field described in the gauge $\boldsymbol{A}(0,0,-Hx)$ and we make the corresponding Peierls substitution $\boldsymbol{p} \rightarrow \boldsymbol{p} - \frac{e}{c_0}\boldsymbol{A}$, ($c_0$ is the light velocity). Because of the x dependence of A, it is more convenient to use a mixed (x, k_y) representation [22] .Our purpose is a theoretical study of the conductivity along the c direction. The latter mainly depends on two different parameters: the first is the electron transfer along the c direction, which we can study by calculating the interplane Green's function; the second is the in-plane electron scattering time. To discuss correctly the conductivity in the c direction, it is essential to take into account the fact that both parameters are strongly affected by the orbital effect of the field.

Let us first calculate the the *independant electron Green function* G_{++} near the right-hand Fermi surface sheets,the equation of motion of which is given by,

$$\left\{ i\omega_n + iv_F \frac{d}{dx} - 2t_b \cos k_y b - 2t'_b \cos 2k_y b - 2t_c \cos\left(k_z + \frac{eHx}{c_0}\right) c \right\} g_{++}(i\omega_n, k_y, k_z, x, x') = \delta(x - x'), \quad (1)$$

where $g_{++}(i\omega_n, k_y, k_z, x, x') = e^{-ik_F(x-x')} G_{++}(i\omega_n, k_y, k_z, x, x')$.

The integration of equation (1) is straightforward. We obtain for $\omega_n(x - x') > 0$:

$$G_{++}\{i\omega_n, k_y, k_z, x, x'\} = \frac{\text{sign }\omega_n}{iv_F} \exp i \left\{ \frac{i\omega_n(x-x')}{v_F} + k_F(x-x') - \frac{1}{v_F}\left[2t_b \cos k_y b + 2t'_b \cos 2k_y b\right](x-x') \right\} - \frac{4t_c}{\omega_c} \cos\left(k_z c + \frac{G_c(x+x')}{2}\right) \sin\left(\frac{G_c(x-x')}{2}\right) \Bigg\},$$

where $G_c = eHc/c_0$ is the magnetic wave vector and $\omega_c = v_F G_c$ is the magnetic energy.

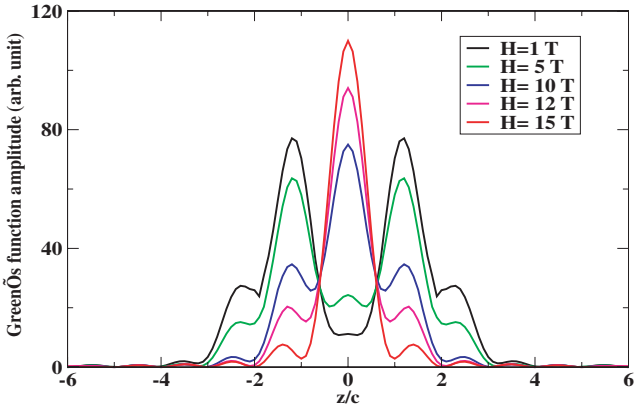

Fig. 8. z dependence of the amplitude of the Green function for different values of the magnetic field.

To bring out the z dependence of the Green function, we have taken the Fourier transform of $G_{++}(i\omega_n, k_y, k_z, x, x')$ with respect to k_z. We substitute $i\omega_n$ by $\omega + i\eta$, where the imaginary part η is related to the quasi-particle lifetime τ by $\eta = \hbar/\tau$. we obtain:

$$G_{++}(\omega, k_y, z, x, x') = \frac{1}{2\pi} \frac{\text{sign }\omega}{iv_F} \exp\left\{\frac{-\eta(x-x')}{v_F}\right\} \exp i\left\{\frac{\omega(x-x')}{v_F} + k_F(x-x') - \frac{1}{v_F}\left[2t_b \cos k_y b + 2t'_b \cos 2k_y b\right](x-x')\right\} \times \int_{-\frac{\pi}{c}}^{\frac{\pi}{c}} \exp i\left\{k_z z - \frac{4t_c}{\omega_c} \times \cos\left(k_z c + \frac{G_c(x+x')}{2}\right) \sin\left(\frac{G_c(x-x')}{2}\right)\right\} dk_z. \quad (2)$$

In the following we give the numerical results obtained from the method explained here. In Figure 8 we have depicted the amplitude of $G_{++}(\omega, k_y, z, x, x')$ as a function of z. As the magnetic field increases, the electron Green's function gets more and more confined by the orbital effect. At a field of 1 Tesla, its amplitude is restricted in a volume between the planes $z = -2$ and $z = +2$, which is, already, a strong confinement. However, because of the strongly oscillating character of the Bessel functions entering the expression of the Green's function, an interference effect occurs at this moderate field, leading to strong peaks at $z = +2$ and $z = -2$, but to a decrease of the amplitude in between, around $z = 0$, compared to that at $z = 2$. For larger fields, namely of the order of 10 Tesla, this interference effect disappears: the amplitude peak is centered at $z = 0$, as expected and, at 15 Tesla, the Green's function is almost entirely confined in the plane $z = 0$.

However, the electrical resistivity also depends on a second factor, the *in-plane electron relaxation time* τ, which is, of course, related to the imaginary part of the frequency $\eta = \hbar\ /\ \tau$ It is worth stressing, that in equation (2), we have taken into account the dependence of the η factor on both temperature and magnetic field. This dependence is derived using the well known temperature dependence of a Fermi liquid resistivity at low T, which is governed by the electron-electron interactions. In the case

of an isotropic Fermi liquid, the quasi-particle lifetime τ obeys the law:

$$\frac{1}{\tau} \propto T^2 n(E_F).$$

Here $n(E_F)$ is the density of states at the Fermi level. For layered conductors, the Fermi surface is anisotropic and the in-plane density of states is k_z dependent for a fixed k_x ($k_x = k_F$). The η factor is then given by:

$$\eta(k_z) = -2\mathrm{Im}G_{++}\left(\omega = E_F, k_x = k_F, k_z\right) T^2.$$

Therefore the field effects on the electron transfer along the c direction and on the in-plane electron relaxation time τ are intimately coupled together. First, the field, by confining the electrons in the planes perpendicular to c, strongly reduces the amplitude of the electron transfer from plane to plane in the c direction. But, at the same time, the field, by inducing a strong electron confinement, localizes the electron density in a narrower distance along c. Straightforwardly, such a strong increase of the electron density in the planes perpendicular to c induces a subsequent increase of the electron scattering rate $\hbar/\tau$. These two effects should, therefore, be discussed together by a self consistent procedure, which, as far as we know, has not been addressed so far. Such a self consistency is essential for the soundness of the method, since the electron confinement as well as the increase of the scattering rate are induced by the same orbital effect of the field. The electron scattering rate, indeed, strongly depends on the in-plane electron density. The latter is calculated by the electron Green's function, in which the imaginary part of the frequency argument is precisely the electron scattering rate. As we shall see in the following, such calculations describe successfully the experimental data discussed in this paper and in reference [20]. We have indeed calculated the transverse component σ_{zz} of the electronic conductivity given by the Kubo formula, in which the scattering rate η plays an essential role [24,25],

$$\sigma_{zz} = \frac{e^2\hbar}{\Omega} \int dk_y \int dk_z v_z^2 \int dE \times \int \frac{d\epsilon}{2\pi} \left[2\mathrm{Im}G_R(k_y, k_z, E, \epsilon)\right]^2 \times \left[-n'_F(\epsilon)\right]$$

where $E = v_F/(x - x')$, Ω is a normalization factor, $v_z = \partial\epsilon(k_y, k_z, \omega_c)/\hbar\partial k_z$ is the electron velocity in the z direction and $n'_F(\epsilon)$ is the derivative of the Fermi distribution function. $G_R(k_y, k_z, E, \epsilon)$ is the retarded Green function.

In Figure 9 we plot the temperature dependence of the resistivity $\rho_c = 1/\sigma_{zz}$ for different values of the magnetic field. In the low temperature range, ρ_c changes from the metallic regime for H below a threshold value $H_{co} \sim 1$ T to an insulator state for higher fields. Hence, the resistivity exhibits a minimum in temperature. The larger the magnetic field, the larger the temperature at which the resistivity is minimal. These features are quantitatively consistent with the experimental data of Figure 9 and with the results of reference [20]. However, it should be stressed that for $\omega_c < T$ (i.e. above 30 K for fields of the order of 10 T), the effective dimensionality of the system is 2D. In this regime, the third direction (governed by t_c) is erased by thermal fluctuations. In this limit, the effect of the field on the conductivity should vanish. Moreover, the in-plane quasiparticle life time should not depend anymore on the magnetic field, since the electron motion, in this temperature range, is actually confined in a single (a, b) plane.

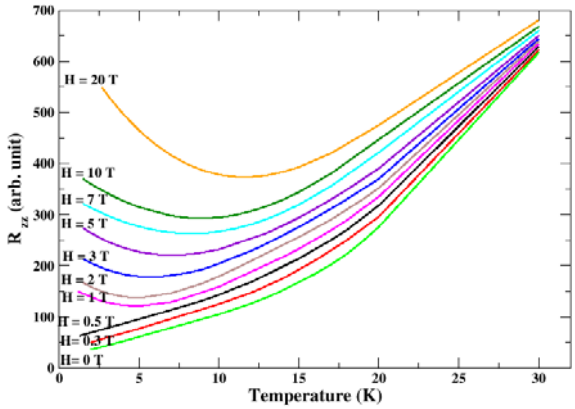

Fig. 9. Resistivity versus temperature for different values of the magnetic field.

This confinement scenario cannot explain the tendency towards saturation for the resistance measured at low temperature under high fields in Figure 5 which is possibly due to incipient superconducting fluctuations effects or impurities in a narrow gap insulator.

It is worth stressing that we did not address in our model the effect of the superconducting fluctuations which are enhanced as the temperature decreases towards the superconducting transition temperature $T_c \sim 1.2$ K. This study goes beyond the scope of this work.

4 Conclusion

A quantum calculation of the conduction along the c axis of $(TMTSF)_2ClO_4$has shown that the insulating character of the resistance which is observed at low temperature under a magnetic field larger than ≈1 T aligned along the b' direction can be explained by a confinement of the carriers within the (a, b) planes [26]. Consequently, it is now clear that the resistive signature attributed to the onset of superconductivity with a diverging critical field (H//b') below 0.2 K [1] must be reexamined in the context of superconductivity arising in a metallic multilayered compound. Long range ordered superconductivity might then be replaced by strong fluctuations at very low temperature.

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