Magnetic field-dependent interplay between incoherent and Fermi liquid transport mechanisms in low-dimensional \( \tau \) phase organic conductors

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Abstract

We present an electrical transport study of the 2-dimensional (2D) organic conductor \( \tau \)-\((P-(S,S)-DMEDT-TTF)_2(AuBr)_2(AuBr_2)_y \) (where \( y \sim 0.75 \)) at low temperatures and high magnetic fields. The inter-plane resistivity \( \rho_{zz} \) increases with decreasing temperature, with the exception of a slight anomaly at 12 K. Under a magnetic field \( B \), both \( \rho_{zz} \) and the in-plane resistivity plane \( \rho_{xx} \) show a pronounced negative and hysteretic magnetoresistance. In spite of a negative residual resistivity ratio in zero field, Shubnikov de Haas (SdH) oscillations are observed in some (high quality) samples above 15 T. Furthermore, contrary to the single closed orbit Fermi surface predicted from band structure calculations (where a single star-shaped FS sheet with an area of \( \sim 12.5\% \) of \( A_{FBZ} \) is expected), two fundamental frequencies \( F_l \) and \( F_h \) are detected in the SdH signal. These orbits correspond to 2.4\% and 6.8\% of the area of the first Brillouin zone \( (A_{BZ}) \), with effective masses \( \mu_l = 4.0 \pm 0.5 \) and \( \mu_h = 7.3 \pm 0.1 \) respectively. The angular dependence, in tilted magnetic fields, of \( F_l \) and \( F_h \) reveals a 2D character of the FS, but no evidence for warping along the \( k_z \) direction (e.g., the absence of a beating effect in the SdH signal) is observed. Angular dependent magnetoresistance (AMRO) fur-
ther suggests a FS which is strictly 2-D where the inter-plane hopping $t_c$ is virtually absent or incoherent. The Hall constant $R_{xy}$ is field independent, and the Hall mobility $\mu_H$ increases by a factor of $\sim 3$ under moderate magnetic fields. Hence the field does not alter the carrier concentration, even in the presence of a large negative magnetoresistance, but only increases the lifetime $\tau_s$. Our observations suggest a unique physical situation where a stable 2D Fermi liquid state in the molecular layers, are incoherently coupled along the least conducting direction. The magnetic field not only reduces the inelastic scattering between the 2D metallic layers, as seen in the large negative magnetoresistance and SdH effect, but it also reveals the incoherent nature of the interplane transport in the AMRO spectrum. Finally, the observed Fermi surface is at odds with band structure calculations. However, the very flat bands in the electronic structure, combined with the variable charge transfer, may be the origin of these effects. The apparent ferro-magnetic character of the hysteresis in the magnetoresistance, remains an unsolved problem.
I. INTRODUCTION

In the last two decades, the field of anisotropic low-dimensional organic conductors has become synonymous with the observation of unusual and exotic electronic properties. Examples range from the possibility of unconventional, anisotropic superconductivity [1,2], to the observation of a variety of other ground states like charge-density waves (CDW) [3], spin-density waves (SDW) [4], field-induced spin-density waves (FISDW) (associated to the observation of quantum Hall effect [5]), and the spin-Peierls state (SP) [6]. A considerable amount of effort has also been devoted to Fermiology [7] and the properties of the metallic states of these compounds. Non-Fermi liquid like behavior has been reported in photoemission spectra [8], there are indications for spin-charge separation [9] in some materials, and unconventional electrical transport properties in the presence of magnet-field induced incoherent hopping has been proposed [10,11].

More recently, new degrees of freedom are being added to these already physically rich systems, by the incorporation of magnetic anions into the structure of organic compounds. Here, due to the physical separation of the molecular orbital (cation) layers, and the inorganic anion layers, there is a corresponding separation of the localized magnetic anion moments (for example the $d$ electrons) and the itinerant low dimensional organic molecular electrons gas ($\pi$ electrons). Typical examples are the series $\lambda$-(BETS)$_2$Fe$_x$Ga$_{1-x}$Cl$_4$ compounds [12] and TPP[Fe(Pc)(CN)$_2$]$_2$ [13]. In the BETS series, the progressive substitution of Ga by Fe suppresses the superconducting state and stabilizes an insulating anti-ferromagnetic (AF) state [12]. While in TPP[Fe(Pc)(CN)$_2$]$_2$, the ground state is also insulating and presents an anisotropic magnetic susceptibility.

One of the main characteristics of magnetic organic systems as mentioned above, is the observation of a pronounced or giant negative magnetoresistance under field. This effect has been explained in terms of field alignment of the local magnetic moments. On one hand, it is expected to destroy an eventual AF ground state, i. e., to close related gaps at the Fermi level (spin-flop transition) [14], and on the other, to decrease the spin scattering of itinerant
electrons by these local moments. In any case, the necessary ingredients for explaining the magnetic-field induced enhancement of the conductivity in these compounds, seems to be the presence of localized magnetic moments, their interaction with itinerant electrons and the effects of the magnetic field on this coupled system.

Nevertheless, there are other families of organic conductors, the compounds of the $\tau$ crystallographic phase \[13\], whose magnetoresistivity presents remarkable similarities to what is observed, for example, in $\lambda$-(BETS)$_2$FeCl$_4$ and TPP[Fe(Pc)(CN)$_2$]$_2$, although their structure is not composed by any magnetic element, see Fig 1 a). Here we report on the electrical transport properties of the $\tau$-(P-($S,S$)-DMEDT-TTF)$_2$(AuBr$_2$)$_1$(AuBr$_2$)$_y$ compound (where $y \sim 0.75$ and P-($S,S$)-DMEDT-TTF stands for pyrazino-($S,S$)-dimethyl-ethylenedithiotetrathiafulvane), at high magnetic fields $B$ and low temperatures $T$. In this compound, an in-plane as well as inter-plane magnetoresistivity is found to decrease by a factor $\geq 75\%$ when a field $B \leq 10$ T is applied. A significant hysteresis is also observed \[16\], which points towards the formation of field induced domains and has been interpreted as an indication of the magnetic nature of these compounds \[17\]. However, magnetic susceptibility measurements revealed an almost temperature independent paramagnetic term. This term is comparable to those measured in other 2D non-magnetic organic systems which are characterized by strong electronic correlations \[18\].

The crystallographic structure of $\tau$-(P-($S,S$)-DMEDT-TTF)$_2$(AuBr$_2$)$_1$(AuBr$_2$)$_y$ is tetragonal with unit cell dimensions $a = b = 7.3546$ Å and $c = 67.977$ Å \[18\]. Inorganic anion layers alternate with mixed organic-inorganic layers, which has both ordered and disordered AuBr$_2$ anions, along with a disordered ethylene group \[18\]. The ratio of donor molecules to acceptor anions is $2 : (1 + y)$, where $y$ has been estimated to be $\sim 0.75$. The value of $y$ determines the area of the Fermi surface, which decreases with increasing $y$ \[14\]. Figure 1 b) shows the calculated Fermi surface of $\tau$-(P-($S,S$)-DMEDT-TTF)$_2$(AuBr$_2$)$_1$(AuBr$_2$)$_y$, $y \sim 0.75$, which was calculated using the extended Hückel tight binding method \[20\]. The star shaped Fermi surface, results from the four-fold symmetry of the molecules packing. While the a-b plane is metallic (conducting) the inter-plane electrical transport displays
an unusual non-metallic behavior in the whole temperature range. This behavior contrasts with what is observed in most quasi-two-dimensional (Q2D)organic compounds, where a $T^2$ behavior at low $T$, is followed by a non-metallic behavior at higher temperatures. A smooth crossover from coherent Fermi liquid excitations at low temperatures, to incoherent excitations at high temperatures, has been suggested to occur in these compounds [21].

In this paper, we report the first observation of Shubnikov de Haas (SdH) oscillations in a $\tau$ phase organic conductor; the $\tau$-$(P-(S,S)$-DMEDT-TTF)$_2$(AuBr)$_2$$_{(y)}$ compound. Two fundamental frequencies $F_l$ and $F_h$ were detected in the fast Fourier transform of the SdH signal, corresponding respectively to 2.4% and 6.8% of the area of the first Brillouin zone ($A_{FBZ}$), which is at odds with band structure calculations. High effective masses, $\mu_l = 4.0 \pm 0.5$ and $\mu_h = 7.3 \pm 0.1$ were obtained for $F_l$ and $F_h$, respectively. The angular dependence of $F_l$ and $F_h$ reveals the 2D character of the FS and the absence of frequency beatings, indicates that the FS is not warped along the $k_z$ direction. The angle dependent magnetoresistance (AMRO), suggests a strictly 2D FS, where the inter-plane hopping $t_c$ is virtually absent or is incoherent. We find the Hall constant $R_{xy}$ to be field independent, and the Hall mobility $\mu_H$ to increase by a factor of $\sim 3$, under moderate magnetic fields. This indicates that $B$ does not introduce additional carriers into the system, instead, it decreases the carriers scattering rate $\tau_s^{-1}$. As neither the inter-plane nor in-plane resistivity displays a $T^2$ dependence at zero field, we conclude, that the magnetic field induces a crossover from a “non-Fermi” liquid like behavior at moderate fields, towards a Fermi-liquid type behavior at higher fields, whose signature is the observation of quantum oscillations (QO’s).

II. EXPERIMENTAL RESULTS

Single crystals of $\tau$-$(P-(S,S)$-DMEDT-TTF)$_2$(AuBr)$_2$$_{(y)}$ ($y \sim 0.75$), synthesized by electrochemical methods [22], of which three different morphologies, were used. Gold wires of 12.5 $\mu$m, were attached with graphite paint, in a conventional 4 lead configuration for inter-layer electrical transport measurements, see Fig 1 c), while a 6 lead configuration
was used for the Hall effect measurements. Standard low frequency (20 Hz) ac lock-in
techniques, with currents of order 10 µA were employed in the measurements. Samples were
mounted in a variety of fixed as well as rotating sample holder probes, immersed in both 3He
cryostats and dilution refrigerators. Magnetic fields were provided by the resistive magnets
available at the National High Magnetic Laboratory’s DC field facility in Tallahassee Florida.

Figure 2 shows the typical temperature dependence at zero magnetic field of the inter-
plane resistivity $\rho_{zz}$ of a $\tau$-(P-(S,S)-DMEDT-TTF)$_2$(AuBr)$_2$ (AuBr$_2$)$_y$ ($y \sim 0.75$) single
crystal, sample# 1. Although the in-plane resistivity displays a metallic behavior [23], the
inter-plane transport, as seen in the figure, is clearly non-metallic and shows an abrupt
change in slope at $T_b \simeq 12$ K. At this temperature, a metal-insulator transition has been
suggested to occur, although specific heat measurements did not provide any evidence for
a phase transition [18] at $T_b$. Below $T_b$, the in-plane resistivity presents a logarithmic de-
pendence on temperature [23] at low $T$. This temperature dependence has been interpreted
as an indication of either weak localization [18,23] or Kondo effect arising possibly from ex-
change interaction between localized magnetic moments and itinerant conduction electrons
[23]. In any case, and as clearly seen, $\rho_{zz}$ does not display the typical $T^2$ dependence seen
at low $T$ in other Q2D organic compounds, which is the signature of coherent electrical
transport [24].

Figure 3 shows the temperature dependence of $\rho_{zz}$ for $\theta \simeq 0^\circ$ ($\theta$ is the angle between $B$
and the $c$ axis) and for several values of magnetic field $B$, as indicated in the figure. Several
striking features are observed:

i) $\rho_{zz}$ recovers a metallic character, i.e., $\rho_{zz}$ increases with $T$, below a magnetic field
dependent temperature.

ii) For all values of magnetic field, $\rho_{zz}$ shows a crossover from positive to negative mag-
netoresistance behavior at a crossover temperature $T_a(> T_b) \simeq 18$ K.

iii) The kink observed at $T_a$ is suppressed by the application of a magnetic field.

The inset of Figure 3 shows the dependence of $\rho_{zz}$ on temperature $T$ for two values
of field, $B = 5$ and 25 T respectively, and for $\theta \simeq 0^\circ$. Arrows indicate increasing and
decreasing temperature sweeps. A marked $T$ dependent hysteresis is observed for $B = 25T$ which is similar to the behavior of magnetic-field induced domains. A small, but non-negligible hysteresis is still observed at 5 tesla. The fact that all the curves meet at $T_b > T_a$ suggests that $T_a$ does not correspond to a thermodynamic phase transition. Instead, it could indicate that charge transport in this system is described by two distinct mechanisms with quite different temperature dependencies. $T_a$ would correspond to the crossover temperature between them. The mechanism that dominates at low temperatures would be by some yet unknown reason and strongly magnetic field dependent.

Figure 4 displays the magnetoresistance $R_{zz}$, from sample #1, as a function of magnetic field $B$ for $\theta \simeq 0^\circ$, and for four different temperatures: 1.45, 1.0, 0.7, and 0.55 K respectively. All curves are vertically displaced for clarity with arrows indicating field-up and field-down sweeps. As previously reported \cite{17}, the resistance decreases by a factor $\geq 65\%$, followed again, by a significant temperature dependent hysteresis. Furthermore, for $T \leq 1$ K and for fields above $B \geq 17$ tesla, Shubnikov-de Haas (SdH) oscillations are observed. This is an indication of the high quality or long mean free path at high fields of these $\tau$ phase metallic single crystals. The so-called resistivity ratio, $\Delta \rho = (\rho_{xx}(300) - \rho_{xx}(4.2))/\rho_{xx}(4.2)$, where $\rho_{xx}(300)$ is the resistivity at $T = 300$ K and $\rho_{xx}(4.2)$ is the resistivity at 4.2 K, is usually used as a criteria for judging the quality of an organic metal. Typically, $\Delta \rho_{xx} \sim 100$ or greater is found for most organic conductors that display SdH oscillations. But this particular sample provides a value $\Delta \rho_{zz} \simeq -0.9$ at $B = 0$ tesla and $\Delta \rho_{zz} \simeq -0.8$ at $B = 27$ tesla. This, at first glance, could be interpreted as a clear indication of the “low quality” or short mean free path of our sample, if, SdH oscillations were not present.

Figure 5 shows the SdH signal as a function of inverse field $B^{-1}$ for $\theta \simeq 0^\circ$ and for several values of $T$, as indicated in the figure. The SdH signal is given by $(\sigma - \sigma_b)/\sigma_b$, where $\sigma$ is the conductance or the inverse of the actual resistivity of our sample (always valid if the Hall component is small, which is the case for a metal), and $\sigma_b$ is the background conductance, obtained by inverting the background resistance. $\sigma_b$ is obtained by fitting the actual sample resistance to a third degree polynomial. The dotted line is a guide to the eye, showing
the envelope of the SdH signal. As clearly seen, it is necessary to postulate more than one frequency for describing the SdH envelope through the Lifshitz-Kosevich formalism. The inset of Fig. 5 shows the amplitude of the fast Fourier transform (FFT) of the SdH signal corresponding to $T = 0.55$ K as a function of frequency $F$. The FFT spectrum displays two peaks at $F_l = 186$ tesla and $F_h = 516$ tesla, respectively. The observation of two frequencies, i.e., two Fermi surface extreme cross-sectional areas is surprising, since, according to band structure calculations [18], the FS of this compound is composed only of a closed star-shaped sheet (see Fig. 1b). Furthermore, from published crystallographic data [18], the area of the first Brillouin (FBZ) zone is given by $A_{FBZ} = 72.986$ nm$^2$. Using the Onsager relation, $F = A(h/4\pi^2e)$, where $F$ is the SdH frequency, $A$ the respective FS cross sectional area, $e$ the electron charge and $h$ Planck’s constant, we obtained 2.4% and 6.8% of the $A_{FBZ}$ for $F_l$ and $F_h$, respectively. Nevertheless, the ratio of the area of the calculated closed Fermi surface in Fig. 1b) to $A_{FBZ}$ is estimated to be 1:8, corresponding to a frequency of $F_{FS} = 955.8$ T. In other words, the estimated $F_{FS}$ is considerably higher than either value determined in the present work. The fraction of “disordered” anions $y$, which determines the area of the FS has been found to be time dependent in the $\tau$-(EDO-$(S,S)$-DMEDT-TTF)$_2$($I_3$)$_{1+y}$ compound [19]. If $y$ in our sample were to differ from $\sim 0.75$, we would expect the geometry of the FS to be quite different from what is shown in Fig. 1a), perhaps this would explain the disagreement. The best refinement of the liquid crystal solution of the compound $\tau$-(P-$(S,S)$-DMEDT-TTF)$_2$(AuBr)$_2$ (AuBr$_2$)$_1+y$ was found for $y = 0.6$ [15]. An alternative explanation could be that the “kink” observed at $T_a$ is the onset of an eventual AF transition; as an AF transitions would open partial gaps at the Fermi level and also affect the original geometry of the FS. However, to date no indications of a phase transition at $T_a$ have not been found in specific heat or magnetic susceptibility measurements [16,18].

Figure 6 shows the logarithm of the FFT amplitude of SdH signal shown in Fig. 4 normalized with respect to $T$, as a function of temperature for both frequencies $F_l$ (solid squares) and $F_h$ (opened circles). The solid line is a fit to the expression $X/\sinh X$ where $X = \alpha\mu_cT/B$, $\alpha = 14.69$ T/K and $\mu_c$ is the effective cyclotron mass in relative units of
the free electron mass $m_e$. The slope yields the effective cyclotron masses, $\mu_l = 4.0 \pm 0.5$ and $\mu_h = 7.3 \pm 0.1$ for $F_l$ and $F_h$, respectively. These are relatively high effective masses for an organic metal, may not be surprising, since the curvature of the proposed star-shaped Fermi Surface presents singularities at its vertices. However, it could also suggest the presence of localized magnetic moments: The exchange interaction between carriers and localized moments are known to modify dramatically the transport of carriers \[23\], especially near a metal-insulator transition. In general, complex magnetoresistive behavior (combinations of positive and negative magnetoresistivities as, for example, in manganites) has led to theories for the formation of magnetic polarons \[26\], i.e., ferromagnetic regions of local moments aligned with the spin of the carrier, via the exchange interaction. Magnetic polarons increase the carrier effective mass and hence tend to localize the carrier, since it is dressed with a polarization “cloud”. At the moment, there is no experimental evidence for this scenario.

Additional information can be obtained by the Lifshitz-Kosevich formalism \[7\] by plotting the amplitude of the SdH oscillations, normalized with respect to $B^{1/2}$. From this, we obtain the Dingle damping factor: $R_D = \exp(-\alpha \mu_c T_D / B)$ where $T_D = h/(4\pi^2 k_B \tau)$ ($k_B$ is the Boltzmann constant, $\mu_c$ is the carriers effective mass in electronic mass units, and $\tau$ is the relaxation time). We calculated $T_D = 1.32 \pm 0.15$ K, which is a small value typical of organic metals \[7\], and indicates the high quality of this $\tau$ phase single crystal despite the negative value of $\Delta \rho_{zz}$. Large effective masses make it difficult to observe SdH oscillations for temperatures above 1 K, due to the Lifshitz-Kosevich damping factors.

Figure 7 displays the inter-plane magnetoresistance $R_{zz}$ as a function of magnetic field $B$ at $T \simeq 0.55$ K and for several values of the angle $\theta$ (between $B$ and the inter-plane $c$ axis) as indicated in the figure. Dotted arrows indicate field up and down sweeps. As indicated, the negative magnetoresistance, as well as the hysteresis between field up and down sweeps, is markedly angle dependent. Apparently the higher the angle, the smaller the hysteresis. Moreover, for $\theta > 50^\circ$ the quantum oscillations become virtually undetectable.

In figure 8 the angular dependence of both $F_l$ and $F_h$ is plotted. Solid lines are fits to the
expression $F(\theta) = F(\theta = 0^\circ) / \cos \theta$. The fit is excellent and provides values of $184 \pm 3$ and $520 \pm 6$ tesla for $F_l$ and $F_h$, respectively. It also clearly indicates that the FS of the $\tau$-(P-$(S,S)$-DMEDT-TTF)$_2$(AuBr)$_2$ (AuBr)$_2$$_y$ ($y \sim 0.75$) compound is 2-dimensional as expected for an anisotropic layered compound.

Figure 9 displays $R_{zz}$ as a function of $\theta$ for two values of the in-plane angle $\phi = 0^\circ$ (solid line) and $\phi = 45^\circ$ (dotted line) at $T = 4.2$ K and $B = 14$ tesla. As indicated by both schemes in the figure, $\phi$ is defined with respect to one of the sample’s edge, consequently, $\phi = 45^\circ$ corresponds to a rotation along one of the diagonals of the square-shaped sample. There is no sign of the Yamaji effect, instead, at $\theta = 0^\circ$ there is an incoherent peak. Furthermore, at $\theta = \pm 90^\circ$, the $\phi$ dependence indicates a 4-fold symmetry which at high fields agrees with what is expected from Fig. 1b. There, the resistance peaks have a period of $\phi = 90^\circ$ for the in-plane MR. The observation of a central peak in $R_{zz}$, i.e., for $B \parallel I \parallel c$, is quite surprising, since magnetoresistance is not expected under these conditions according to classical transport theory. In fact, in most 2D organic compounds, peaks are observed at $\theta = 90^\circ$ (as in our data for $\phi = 45^\circ$) and also with a well defined periodicity in $\tan \theta$ [1,7]. This periodicity, according to a semi-classical approach [27], results from the warping of FS along the $k_z$ reciprocal lattice direction. More recently, in the frame of an incoherent interlayer transport model, McKenzie and Moses [28] demonstrated that the existence of a three-dimensional (3D) FS is not a necessary ingredient to explain the above mentioned angle dependent structures which are observed in the magnetoresistance of most organic conductors. In any case, the beating between two close frequencies corresponding to the two FS extremal cross sectional areas is a clear signature of a 3-D closed orbit which is warped along the $k_z$ direction. This beating is absent in our data. An indication of a finite interlayer transfer integral is the observation of a sharp and pronounced peak at $\theta = 90^\circ$ [29,30]. In our data, a broad peak showing a maximum at $\theta = 90^\circ$ is observed for $\phi = 45^\circ$ but is absent for $\phi = 0^\circ$, which may suggest a strictly 2-D FS for these $\tau$ phase organic compounds. Furthermore, our preliminary angular studies indicate that the four-fold symmetry observed for $\phi = 45^\circ$, decreases to a two-fold one at lower fields. A detailed angular study will be the
subject of a future report [31].

The in-plane resistivity, \( \rho_{xx} \), as a function of \( B \) from sample \#2 and for four different temperatures is presented in figure 10 a). The respective temperatures are indicated in the figure. In order to measure the Hall effect, a conventional 6 lead configuration was used. The general behavior of \( \rho_{xx} \) is essentially similar to what is observed in \( \rho_{zz} \) under field: A remarkable resistivity drop, which in this case, is observed for \( B \leq 2 \) T instead of \( B \leq 6 \) T as for \( \rho_{zz} \). Nevertheless, for this particular sample no quantum oscillations are observed, which is surprising since, this single crystal originally is from the same electrocrystallization cell as sample \#1. This clearly indicates that single crystals of different qualities and/or physical properties are produced during the synthesis process [32] and may also suggest that the content of acceptor anions, \( 1 + y \), may vary from sample to sample in a single batch. Also, the morphology of this sample differs from what is shown in Fig. 1 c), in this case we have chosen a thin rectangular platelet. Figure 10 b) shows the Hall resistance \( R_H \) as a function of \( B \) and for the four values of \( T \) in figure 10 a). \( R_H \) is obtained by anti-symetrization of the Hall voltage \( V_H \): \( R_H \equiv [V_H(+B)−V_H(−B)]/2I_x \) where \( I_x = 50 \) \( \mu \)A is the injected current. As seen, \( R_H \) is linear in field, as expected for a metal characterized by only one type of carrier, whose sign indicates that electrons are the charge carriers, in agreement with previous results [33]. Moreover, \( R_H \) is temperature independent below 4.2 K (solid line in this figure is a guide to the eye). The Hall constant \( R_{xy} = E_y/j_x \equiv (R_H \cdot t/B) \), where \( E_y \) is the transverse electric field and \( j_x \) is the in-plane density of current, is shown as a function of \( B \) in figure 11 a), from the traces in Fig. 10 b). Except at low fields, where the Hall signal is too small for an accurate determination, \( R_{xy} \) is essentially constant in magnetic field for \( B \) up to 30 T. In other words, there is no clear evidence which could indicate that \( B \) introduces carriers into the system, hence decreasing its resistivity. An estimation of the density of carriers in our system is provided by the standard expression for the Hall coefficient in a isotropic system: \( n = (R_{xy} \cdot e)^{-1} \) where \( e \) is the electron charge. \( n \) is presented in Fig. 11 b) and is basically constant for \( B > 4 \) T saturating to a value \( n \simeq 3.75 \times 10^{26} \) m\(^{-3}\). By multiplying \( n \) by the unit cell volume \( \Omega = 3676.9 \) \( \text{Å}^3 \) we obtain a value of \( \simeq 1.4 \) carriers per unit cell. This value
is remarkably close to the number of acceptor anions \( 1 + (y \simeq 0.75) = 1.75 \, \Omega^{-1} \), considering the usual uncertainty associated with the sample and contacts geometrical factors as well as the inadequacy of the above expression for describing a temperature dependent Hall effect in an anisotropic 2-D system. Consequently, and at least at low temperatures, the number of carriers seems to be given by the number of acceptor anions in this \( \tau \) phase system.

Finally, as \( R_{xy} \ll \rho_{xx} \), the Hall mobility, which is proportional to \( \tau_s \), the inverse of the scattering rate, is approximately given by \( \mu_H \simeq R_{xy}/\rho_{xx} \) and is plotted in figure 11 c). As seen, \( \mu_H \) is rather small, on the order of \( 10^{-2} \), and slightly decreases with \( B \) indicating that \( \tau_s \) increases at higher fields. In the important low field region, where the resistance decreases considerably, it is not possible to directly extract the real behavior of \( \mu_H \) due to the uncertainties in \( R_{xy} \), as mentioned above. Nevertheless, as \( R_H \) is remarkably linear in field, we expect \( R_{xy} \) to be essentially constant in the whole field range. As the resistivity decreases by a factor of \( 3 \), \( \mu_H \) necessarily increases by the same factor. At the moment, it is not clear which mechanism is responsible for this magnetic-field induced reduction of \( \tau_s^{-1} \).

### III. DISCUSSION

It would be possible to explain most of our observations, if we assume the presence of localized spins that undergo an AF transition at \( T_b \), for example, ferromagnetic layers interacting antiferromagnetically. The negative magnetoresistance, could be ascribed to a decrement in the itinerant carriers scattering rate \( \tau_s \) (in agreement with our Hall effect measurements) due to the alignment of the localized spins, or simply to the field suppression of an insulating antiferromagnetic state as is seen in several magnetic organic compounds. The presence of ferromagnetic domains and/or hysteretic behavior would naturally be explained by this scenario. In addition, and as mentioned before, the discrepancy between the calculated Fermi surface and the SdH frequencies found by us, could be simply explained in terms of a FS reconstruction at \( T_b \). On the other hand, the heavy carrier effective masses would be the result of an indirect exchange type of interaction, through ferromagnetic po-
larons. Nevertheless, and as previously mentioned, this scenario is unrealistic, since none of the constitutive elements in $\tau$-(P-$(S,S)$-DMEDT-TTF)$_2$(AuBr)$_2$ (AuBr$_2$)$_y$ ($y \sim 0.75$) is magnetic. At this point, to explain the negative magnetoresistance seen here, it is interesting to mention the model for repulsively interacting electrons on a lattice whose band dispersion contains a flat portion, recently proposed by Arita et al. [34]. According to these authors, when the Fermi level lies in the flat part, electronic correlations cause ferromagnetic spin fluctuations and consequently an enhanced spin susceptibility. So far, the only experimental report [18] containing magnetic susceptibility measurements do not seem to sustain this scenario.

The observation of an important hysteresis in the field dependence of the resistivity and which points towards metastability in the system, might provide some insight about the nature of the ground state. Metastability is well known in density-wave (DW) type systems and is associated with the DW pinning to, for example, defects or the lattice. DW ground states are associated with the geometry of the FS [1] through the so-called FS nesting property, which is a particularly useful concept for quasi-one-dimensional systems with opened FS’s. However, in our case, we can not identify a "good" nesting wave-vector for the star-shaped FS resulting from band structure calculations (see Fig. 1 b)). Also, negative magnetoresistance is compatible with a charge-density wave (CDW) ground state which is destabilized by the Pauli effect under magnetic field [35,36]. Assuming that $T_b \sim 13$ K is the CDW transition temperature and using a simple BCS relation, we can estimate the critical field necessary to suppress a uniform CDW: $B_c = 1.765k_B/\mu_BT_c$ [36], where $k_B$ is the Boltzmann constant, $\mu_B$ is the Bohr magneton, and $T_c$ is the transition temperature to the CDW state. We obtain a critical field $B_c \sim 43$ T for suppressing an eventual CDW ground state, which is one order of magnitude higher than the magnetic fields at which a pronounced resistivity decrement is observed by us. In our opinion, a DW type ground state does not seem to be compatible with our observations.

Finally, and as already indicated [13] the resistivity at zero field, presents a $\ln T$ dependence, which suggests a weak localization type of regime. Nevertheless, it is difficult to
reconcile weak localization resulting from weak disorder, with the angular dependent hysteresis seen here. Hysteresis would be compatible with highly disordered systems, where the magnetic field is expected to affect, for example, the spin-orbit coupling, the configuration of localized charges/spins and/or the percolation path for electrical transport in the system.

The lack of a $T^2$ dependence in the resistivity and/or frequency beats in the SdH signal as well as of a peak in the angular-dependent magnetoresistance for in-plane magnetic fields, clearly suggests that the inter-plane transport is incoherent in this $\tau$ phase organic conductor. Consequently, this compound may be classified as a highly correlated low dimensional electronic system, as is the case for most transition metal oxides, like, for example, the cuprates, and for which an appropriate physical description is yet unavailable. What is really remarkable in the present case, is the observation of Fermi-liquid type of behavior at high fields. Consider, for example, the model developed in Refs. [11,37], for explaining, for example, the angular dependence of the magnetoresistance in (TMTSF)$_2$PF$_6$. According to these authors, fields applied exactly along the inter-plane axis of an anisotropic, for instance, layered material do not affect the inter-plane motion of the charge carriers, i. e., the coherence of the inter-plane transport. Consequently, if the system displays FL behavior at zero field, it should keep its physical properties at this particular orientation under field. But, for any other orientations, the magnetic field is expected to interfere the interchain coherence by “adding an effective inelasticity to the interchain hopping [11]”, and consequently, a non-FL type of behavior should emerge. Now, consider what is observed in Fig. 7 where SdH oscillations are still observed at $T \simeq 0.55$ K for angles as a pronounced as $\theta = 37.3^\circ$ and for fields as high as 30 tesla. In other words, even for an in-plane field component as high $\sim 18$ tesla this system, recovers its FL like character. A simple way to conciliate our results with this model, is to assume that inter-layer transport is incoherent at zero field and remains incoherent in the whole field range. This could explain the absence of beatings in the SdH signal, the absence of a sharp peak for $\theta = 90^\circ$ in the AMRO and the negative resistivity ratio $\Delta \rho_{zz}$, or the unusual temperature dependence of $\rho_{zz}$. Consequently, to assume that the transport is coherent within the conducting planes is the only possible way to explain
the observation of quantum oscillations at high fields. This would imply an unusual physical situation: 2-D Fermi liquid type of layers coupled incoherently between them. However, and as already mentioned, \( \rho_{xx} \) does not display a \( T^2 \) dependence at low \( T \) either. Consequently, it would still be necessary to find an explanation for the observed crossover towards FL behavior, within the conducting planes, induced by high magnetic fields. The field induced reduction of \( \tau_s^{-1} \) is a physical evidence for this crossover.

**IV. SUMMARY**

In summary we presented an electrical transport study in the 2-dimensional organic conductor \( \tau-(P-(S,S)-DMEDT-TTF)_2(AuBr)_2 \) \( \text{(AuBr)}_2 \text{y (where } y \sim 0.75) \text{ at low temperatures and high magnetic fields } B \). Both the in-plane and the inter-plane resistivities show a pronounced negative and hysteretic magnetoresistance, which in some samples are followed by the observation of Shubnikov de Haas (SdH) oscillations. Two fundamental frequencies \( F_l \) and \( F_h \) were detected in the FFT spectrum of the SdH signal, corresponding respectively to 2.4% and 6.8% of the area of the first Brillouin zone \( (A_{FBZ}) \), which disagrees with band structure calculations. High effective masses \( \mu_l = 4.0 \pm 0.5 \) and \( \mu_h = 7.3 \pm 0.1 \) were obtained for \( F_l \) and \( F_h \), respectively. The angular dependence of \( F_l \) and \( F_h \) reveals the 2-dimensional character of the FS, while the absence of frequency beatings indicates the absence of warping along the \( k_z \) direction. Furthermore, the angle dependent magnetoresistance (AMRO) suggests a FS which is strictly 2-D, i.e., the inter-plane hopping \( t_c \) is incoherent. While the Hall constant \( R_{xy} \) is field independent, the Hall mobility \( \mu_H \) increases by a factor of \( \sim 3 \), under moderate magnetic fields. This indicates that \( B \) does not introduce carriers into the system but, instead, decreases the carriers scattering rate \( \tau_s^{-1} \).

Although these observations might be explained in terms of the presence of local magnetic moments in the system, this possibility is discarded by magnetic susceptibility measurements. These observations are also difficult to conciliate with a density-wave type of ground state or with an explanation in terms of weak localization. Considering that there is
no evidence for a thermodynamic phase transition at $T_b$, we believe that we have found an example of a very anisotropic Q2D system which shows a crossover from a non-Fermi liquid type of behavior to a Fermi liquid one, induced by high magnetic fields. Considering the accumulation of evidence for incoherent charge transport between the layers, the only possible way to explain the observation of quantum oscillations at high fields, is to assume that the transport is \textit{coherent}, or FL like, within the conducting planes. Consequently, this $\tau$ phase compound seems to be described by a unique scenario: Conducting 2-D layers displaying Fermi liquid type of behavior, which are coupled incoherently. The future confirmation of this scenario will certainly be relevant for other layered materials like the transition metal oxides.

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FIGURES

FIG. 1. a) A sketch of the P-\((S,S)\)-DMEDT-TTF molecule. b) Calculated Fermi surface of \(\tau-\text{P-}(S,S)\text{-DMEDT-TTF})_2\text{(AuBr)}_2\text{(AuBr}_2\text{)}_y\) and for \(y \sim 0.75\) (solid line). The dashed line represents the calculated Fermi surface for \(y = 0\). c) Configuration of contacts for inter-plane electrical transport measurements, and morphology of the sample used.

FIG. 2. Dependence on temperature \(T\) of the inter-plane resistivity \(\rho_{zz}\) of a \(\tau-\text{P-}(S,S)\text{-DMEDT-TTF})_2\text{(AuBr)}_2\text{(AuBr}_2\text{)}_y\) \((y \sim 0.75)\) single crystal at zero magnetic field. The change in slope observed at \(T_a\) may indicate either a phase transition or a crossover between two different electrical transport regimes.

FIG. 3. Inter-plane resistivity \(\rho_{zz}\) as a function of temperature \(T\) under several values of magnetic field applied \(B\) along the inter-plane \(c\) axis. For all values of \(B\), \(\rho_{zz}\) shows a crossover from positive to negative magnetoresistance behavior at a crossover temperature \(T_a(> T_b) \simeq 18\) K. Inset: \(\rho_{zz}\) for both field up and down sweeps (indicated in the figure by arrows) as a function of \(T\) and for two values of field 5 and 25 T, respectively. A large hysteresis is observed at 25T.

FIG. 4. Inter-plane resistivity \(\rho_{zz}\) (sample \#1) as a function of \(B\) for \(\theta \simeq 0^\circ\) and for four different temperatures: 1.45, 1.0, 0.7, 0.55 K, respectively. Curves are vertically displaced for clarity. Arrows indicate field-up and field-down sweeps.

FIG. 5. SdH signal as a function of inverse field \(B^{-1}\) for \(\theta \simeq 0^\circ\) and for several values of \(T\). Dotted line is a guide to the eye. Inset: The amplitude of the fast Fourier transform (FFT) of the SdH signal corresponding to \(T = 0.55\) K as a function of frequency \(F\). Two peaks are observed at \(F_l = 186\) T and \(F_h = 516\) T, respectively.

FIG. 6. The logarithm of the FFT amplitude of the SdH signal previously shown in Fig. 5 normalized respect to \(T\), as a function of temperature for both frequencies \(F_l\) (solid squares) and \(F_h\) (opened circles). Solid lines are fits to the Lifshitz-Kosevich formalims that provide the effective masses \(\mu_l = 4.0 \pm 0.5\) for \(F_l\) and \(\mu_h = 7.3 \pm 0.1\) for \(F_h\), respectively.
FIG. 7. Inter-plane magnetoresistance $R_{zz}$ as a function of $B$ at $T \simeq 0.55$ K and for several values of the angle $\theta$ between $B$ and the inter-plane $c$ axis. Dotted arrows indicate field up and down sweeps.

FIG. 8. Angular dependence of both $F_l$ and $F_h$. Solid lines are fits to the expression $F(\theta) = F(\theta = 0^\circ)/\cos \theta$.

FIG. 9. $R_{zz}$ as a function of $\theta$ for two values of the in-plane angle $\phi = 0^\circ$ (solid line) and $\phi = 45^\circ$ (dotted line) at $T = 4.2$ K and $B = 14$ tesla.

FIG. 10. a) The in-plane resistivity $\rho_{xx}$ as a function of $B$ from sample #2 and for four different temperatures. b) The Hall resistance $R_H$ as a function of $B$ for the same four values of $T$ as in a). The solid line is a guide to the eye.

FIG. 11. a) The Hall constant $R_{xy}$ (see text) as a function of $B$ calculated from the traces shown in Fig. 10 b). b) The density of carriers $n$ as a function of $B$ obtained from the traces in Fig. a). c) The Hall mobility $\mu_H \simeq R_{xy}/\rho_{xx}$ as a function of field $B$. 
a) 

\[
P-S,S-\text{DMEDT-TTF}
\]

b) 

diagram showing the electron band structure in the \( \Gamma \) point.

c) 

diagram showing the magnetic field \( B \) and angle \( \theta \) with respect to the crystal axes \( a \), \( b \), and \( c \).
$\rho_{zz} (\Omega \text{ cm})$ vs $T \text{(K)}$

$\tau-(P-(S,S)-\text{DMEDT-TTF})_2(AuBr_2)_1(AuBr_2)_{0.75}$

$T_b$

sample #1

$B = 0 \text{ T}$
Sample #1

$B \parallel c$

$\rho_{zz} (\Omega \text{ cm})$

$T (K)$

$T_a$

$T_b$

$5T$

$25T$

$2.5T$

$15T$

$27T$

$0T$
This figure "taufig4.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0011529v1
SdH signal (A. U.)

FFT Amplitude (A. U.)

$F (T)$

$T = 0.9 \text{ K}$
$T = 0.75 \text{ K}$
$T = 0.55 \text{ K}$
\[ \ln\left( \frac{\text{Amplitude}_{\text{fit}}}{T} \right) \]

\[ \mu_h = 7.3 \pm 0.1 \]

\[ \mu_1 = 4.0 \pm 0.5 \]
This figure "taufig7.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0011529v1
\[ F_h(\theta) = \frac{520}{\cos \theta} \]

\[ F_1(\theta) = \frac{184}{\cos \theta} \]
\[ R_{zz} (\Omega) \]

-100 \( \phi = 0^0, B = 14 \, \text{T} \)
-100 \( \phi = 450^0, B = 14 \, \text{T} \)

\[ \theta (\text{degrees}) \]

-100 \( T = 4.2 \, \text{K} \)
Sample #2

\( \rho_{xx} \) (m\(\Omega\)-cm)

- 0.8 K
- 1.35 K
- 3.0 K
- 3.7 K

\( -R_H \) (m\(\Omega\))

Sample #2

\( B \) (tesla)
