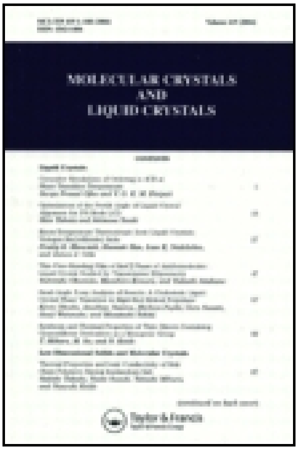

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$(TMTSF)_2ClO_4$ in High Magnetic Fields

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Abstract

We report high field magnetotransport measurements and relate them to a model which demonstrates the instability of a 2-D open orbit Fermi surface caused by the one dimensionalization of the electron motion in a magnetic field.

One of the most interesting aspects of the $(TMTSF)_2X$ salts is the low temperature magnetic field induced spin density wave (SDW) state [1-7]. The discovery of the threshold field[1] and the metal-semimetal transitions is unique to these materials. The orbital effect of a magnetic field causing a transition is almost unknown in other materials and the fact that the Fermi surface (F.S.) consists of nothing but open orbits in the metallic state makes it even more interesting.

In this paper we will discuss our experimental findings involving this unique transition. The model which we present shows that an open orbit two dimensional metal in the presence of a magnetic field become unstable against a F.S. distortion. This instablity results from the one dimensionalization of the electron motion.

The TMTSF salts are highly anisotropic conductors. A variety of experiments have yielded a general consensus for the band widths. The ratios $t_a:t_b:t_c$ are ~ 100:10:0.3[3,8]. Figure 1 shows schematically the F.S. in the a-b plane. The band width along c is smaller by a factor of 30 and is neglected in the rest of this discussion.

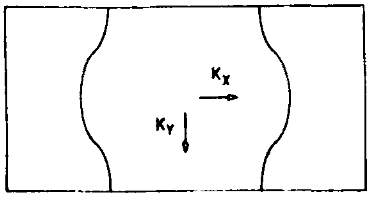

FIGURE 1 Fermi Surface schematic.

The field induced state was discovered by Kwak et al[1] in a magnetoresistance measurement of the PF_6 salt at a pressure just beyond the critical pressure which suppresses the SDW transition. They observed a threshold field followed by quantum oscillations in the magnetoresistance. Subsequently a similar effect was seen by a number of researchers independently in the CLO_4 salt at ambient pressure[2-3]. Magnetic resonance experiments indicate that the high field state is a SDW[4], and specific heat measurements show a second order transition[5].

In order to illucidate some aspects of the field induced transition we measured the sound velocity of the CL04 salt using a vibrating reed technique[9]. The sound velocity is given approximately by $v_s = v_0(1-g^2N(E_f)+a(H)n^2)$, where g is the electron-phonon coupling, $N(E_f)$ the density of states, and n the carrier density. Theoretically and experimentally it has been demonstrated that a(H) is linear for open orbits and quadratic for closed orbits. As is easily seen in Fig. 2 there is a linear dependence on magnetic field for the sound velocity until the threshold field. This indicates that the orbits remain open until the field induced transiton. After this transition there are oscillations in velocity of sound. These oscillations can be explained in terms of a reduction of n and $N(E_f)$ at threshold and another reduction at about 8 Tesla.

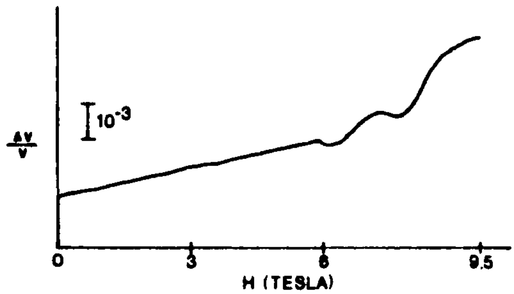

FIGURE 2 Sound velocity vs. H at 0.6K

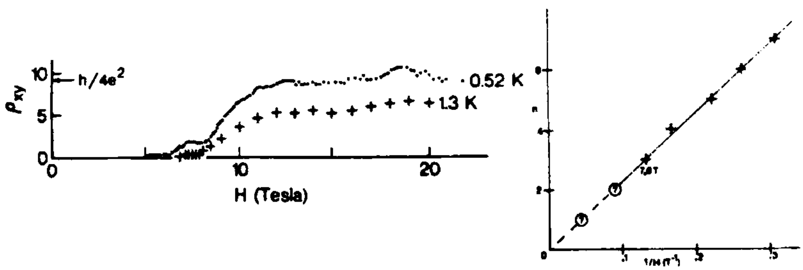

FIGURE 3 Hall resistance, position of Hall steps from ref.6

Our measurement of the hall resistance (ρ_{xy}) is shown in fig. 3. Similar independent measurements were reported in ref . 7. Recall that $\rho_{xy} = R_H H$ where the hall coefficient $R_H=1/nec$. Therefore, for a fixed density of carriers ρ_{xy} should be a straight line going through the origin. For fields larger than threshold ρ_{xy} increases sharply showing that the density of carriers has been reduced. As the field is increased further we see that ρ_{xy} has plateaus and steps until 8 Tesla above which there is one flat plateau that stretches from 12 Tesla to 22 Tesla. The general trend is a reduction in the number of carriers as the magnetic field is increased and the appareance is that of a series of phase transitions. Note that if the carrier concentration was fixed but quantum oscillations were present we would see oscillations about a straight line through the origin. Even in the extreme case of the quantum Hall effect (QHE) in a system with fixed carrier concentration the plateaus occur with their centers lying on a straight line.

The steps and plateaus are reminescent of the QHE[10] but there are large differences from the conventional QHE. Among the most striking differences are the temperature dependence of the plateaus, the fact that they are not in the ratio of consecutive integers, and that their positon does not correspond to the ratio between the steps. Also as can be seen in ref. 6 for fig. 3 or in fig. 4 there are not strong dips in the longitudinal magnetoresistance corresponding to the plateaus in ρ_{xy}. Thus interpretation in terms of the conventional QHE is not appropriate.

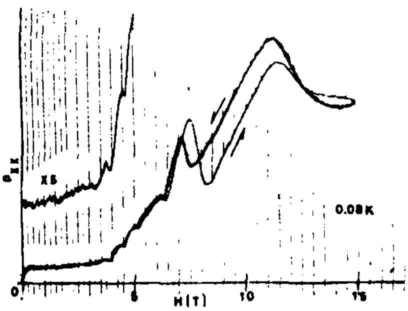

FIGURE 4 Magnetoresistance at high field

The longitudinal magnetoresistance ρ_{xx} is plotted at low temperature for another sample in Figure 4. The threshold field is apparent at 3.5 Tesla and the magnetoresistance has a general increase with magnetic field with structures which appear quite different from the smooth sinusoidal behavior usually associated with Schubnikov - deHaas oscillations. Moreover hysteresis can be seen as a difference with increasing and decreasing field. This is most pronounced for the 8 Tesla step. We should note that higher temperature data reported in ref. 6 shows more conventional oscillations at high temperature. These more rapid oscillations which can be associated with closed orbits that would be formed with the nesting vector $2k_f,0$ are not present at lower temperature.

The ratio of the plateaus in $1/\rho_{xy}$ are 0.2:1:2.66:4 and 0.2:1:2.1:3.6 for plateaus at 12, 7.2, 5.7, 4.7 Tesla respectively, for our two best samples. Grossly speaking the latter recipocal ratios are 1:2:3 which would correspond to Landau levels if the state at 7.2 T has one level located below the Fermi energy. The positions of the onset of the steps is plotted vs. 1/H in Figure 2 from data of ref. 6. Although we obtain a straight line through zero there are steps missing at 11.2 and 22 Tesla the former of which should certainly have been in our experimental range but which was not observed.

In the only other orbital field induced transition from metal to semiconductor ref.11 showed that for closed orbits in three dimensions the dispersion relation becomes one dimensional and the density of states becomes proportional to H. They predicted a F.S. instability with $T_c = T_o\ e^{-1/N(O)V} = T_o e^{-A/H}$. It was pointed out[12] that in two dimensions for open orbits a magnetic field leads also to a one dimensional dispersion relation. The question was whether the density of states also varies linearly with the magnetic field and whether this can lead to a F. S. instability characteristic of the one dimesnional behavior[13].

In the presence of a magnetic field an electron moves on a open orbit F.S. from one end of the Brillouin zone to the other and repeats this motion. There is a characteristic frequency $\omega_c'=(eH/mc)(k_x b)$ which characterizes this motion and corresponds to the periodic crossing of the zone[12]. The velocity of the electron is perpendicular to the fermi surface at all points. Thus in a magnetic field an electron in real space has a velocity which oscillates along y but always remains the same sign along x. This can be quantified by noting that the velocity in the y direction is given by $v_y=(2t_b b/\hbar)\cos(\omega_c' t)$. Thus the excursion of an electron in the y direction is $4t_b b/\hbar\omega_c'$. For very high fields the electron motion is so localized in the y direction that it remains on a single chain and is literally one dimensional. However even at very low magnetic fields the motion is limited along y and extended along x illustrating the one dimensional nature in any magnetic field.

This one dimensionalization can be seen mathematically by looking at the dispersion relation and the effective magnetic field. We take the dispersion relation as $E(k_x,k_y)=\hbar^2 k^2{}_x/2m-2t_b\cos(k_y b)$ [12]. In the presence of a magnetic field this equation becomes:

$$-(\hbar^2/2m)(\partial^2/\partial x^2)\psi-2t_b\cos(K_y b-eHbx/\hbar c)\psi=E\psi \qquad (1)$$

in the Landau gauge. ky occurs only in the argument of the cosine and can be removed by redefining the x origin. Thus the energy eigenvalues can not depend on k_y. The general form for the energy eigenvalues is $E(K_x')$. This unexpected conclusion can be understood in the following way. In the presence of any finite magnetic field the electron completely traverses the zone along y. Thus in some sense the electron averages over the value of k_y. This of course neglects temperature, the tranfer integral along c[12] and the scattering time.

The one dimensional property of interest is the F.S. instablity - Peierls transition, charge density wave or SDW caused by the large degeneracy of states at the Fermi energy coupled by the same wave vector. We define the joint density of states $N_j(E,q)$ as the density of states at energy E coupled by wave vector q. For the dispersion relation $E(k_x,k_y)$, this joint density of states is zero (as appropriate for a material which has no instability down to zero temperature). We now examine the joint density of states in the presence of a magnetic field in terms of the solution to eq. 1. For $h\omega_c'/4t_b >> 1$ eq. 1 is the nearly free electron model with plane wave eigenstates, energy $E(k_x)$, $N_j(E_f,q)=N(E_f)=1/E_f$, and $q=(2k_f,0)$ (the 1-D case). We thus expect the form $N_j(E_f)=(1/E_f)F(\hbar\omega'_c/4t_b)$ where $F(x)$ is a smooth function which is zero for small x and approaches 1 as x goes to infinity. By taking the Fourier transform of the Mathieu functions defined by eq.1 we find $F(x)=x$ for small x. An open orbit 2-D F.S. is therefore unstable as soon as a magnetic field is applied[13]. In a small magnetic field we then have $N_j\sim(1/E_f)(\hbar\omega_c/4t_b)\sim H$ and $T_c=T_0\exp(-A/H)$ which gives an excellent fit to the experiments (fig. 5).

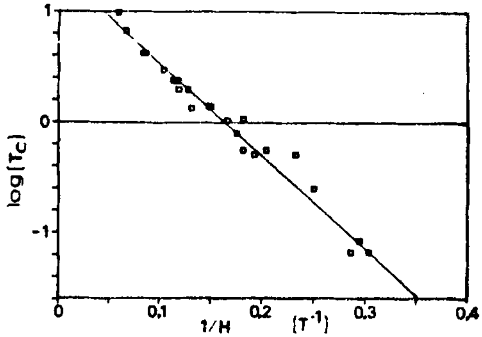

FIGURE 5 Log T_c vs. 1/H

While we have not solved for the ground state, we make the following observations[13]. The q vector in a field is in general not ($2k_f$, π/b), so that we expect a semimetal rather than the semiconductor expected for a SDW at zero field. The gap which results is field and temperature dependent - the area remaining after the distortion and hence the quantum oscillations will be H and T dependent. (To lowest order $E_{fg}=E_{f0}-G(\Delta)=(n+1/2)\hbar\omega_c=(n+1/2)rH$ or taking $G(\Delta)\sim\Delta_0+aH$, $(E_{f0}-\Delta_0)/H=(n+1/2)r+a$. This gives oscillations $\sim$ periodic in 1/H but with an apparent n not equal to the number of Landau levels below E_f. From the inset of fig.2 we thus suspect that the last Landau level has crossed E_f at 7.5T and no further oscillations will occur.) If the distortion produces electron hole pockets which are compensated[14], $\rho_{xy}=(h/ne^2)(\mu_e-\mu_h)/(\mu_e+\mu_h)$, so that the ratio of the plateaus is correct, but the value is considerably smaller. Moreover, for compensation ρ_{xx} will not have zeroes at the plateaus and will increase with H.

Just prior to this conference we became aware of the paper by Gor'kov and Lebed'[15] which independently showed the instability of the 2-D open orbit F.S. in the presence of a magnetic field due to the same mechanism.

In conclusion, the $(TMTSF)_2X$ salts show a novel and unique transition from open orbit metal to closed orbit semimetal. We have demonstrated that this transition is caused by the one dimensionalization of the electron motion in a magnetic field. Many of the unusual high field measurements can be explained if this instability produces a compensated semi-metal. The nature of the ground state and inparticular the state above 7.5 Tesla awaits further investigation.

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