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Pressure dependence of field induced SDW states of $(\text{TMTSF})_2\text{ClO}_4$ from the magnetoresistance at 1.5 K

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Résumé. — Nous présentons des mesures de magnétorésistance sur des monocristaux du conducteur organique $(\text{TMTSF})_2\text{ClO}_4$ pour des pressions jusqu'à 1,5 kbars et sous des champs magnétiques appliqués selon l'axe c^* et allant jusqu'à 11,4 T. Nous montrons qu'à la fois le champ seuil correspondant à l'apparition de l'état onde de densité de spin, ainsi que tous les autres champs caractéristiques probablement liés à des transitions entre états onde de densité de spin, augmentent rapidement avec la pression. De plus nous présentons rapidement la dépendance en pression de quelques autres grandeurs comme la température de mise en ordre des anions, la magnétorésistance dans l'état métallique et la résistivité.

Abstract. — We report magnetoresistance measurements for single crystals of the organic conductor $(\text{TMTSF})_2\text{ClO}_4$ at pressures up to 1.5 kbars, at 1.5 K in fields up to 11.4 T along the c^* -direction. It is found that both the threshold field for the onset of the spin density wave (SDW) state and the other characteristic fields, which probably represent SDW-SDW transitions, increase strongly with pressure. In addition the pressure dependence of other quantities, the anion ordering temperature, magnetoresistance and resistivity in the metallic state are briefly reported.

1. Introduction.

The first organic crystal found to become superconducting at ambient pressure [1], bis-tetramethyltetraselenafulvalenium perchlorate $(\text{TMTSF})_2\text{ClO}_4$, shows several interesting and unusual properties. Slowly cooled or relaxed (R) samples exhibit a phase transition at 25 K associated with the ordering of non-centrosymmetric ClO_4^- anions and the formation of a superlattice associated with a $(0, 1/2, 0)$ wave vector in reciprocal space [2]. R crystals exhibit bulk superconductivity with $T_c = 1.2$ K [3]. On the other hand for crystals which are rapidly cooled, quenched (Q) samples, the anions remain at least partially disordered. In this case an antiferromagnetic spin density wave develops below 6 K [4] and superconductivity is suppressed [5].

Furthermore, it has been found that application of a strong magnetic field (H) to R samples along the c^* -direction induces a metal-spin density wave transition at a certain temperature dependent threshold field H_T [6]. As H is increased above H_T a series of anomalies has been observed in several physical properties : magnetoresistance [7], Hall effect [8], specific heat [9] and magnetization [10].

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In parallel with these experimental studies a number of theoretical investigations has been made recently with a view to understanding the phase diagram and the above physical properties of $(\text{TMTSF})_2\text{ClO}_4$ as a function of field and temperature [11-14].

There have already been some indications from other work [15, 16] that H_T is strongly increased by pressure. In this paper we report the results of magnetoresistance measurements at $T = 1.5$ K on crystals of $(\text{TMTSF})_2\text{ClO}_4$ in the R-state as a function of hydrostatic (helium) pressure up to 1.5 kbar in magnetic fields up to 11.4 T. We find that as the pressure is increased the anomalies in magnetoresistance shift rapidly to higher fields. In this respect it is interesting to note that the superconducting T_c is rapidly suppressed by pressures as low as 4 [17] or even 2 [18] kbars. Also in the isostructural analog $(\text{TMTSF})_2\text{PF}_6$ the spin density wave transition is rapidly suppressed by pressure [3]. As far as we know these strong pressure dependences are not understood and may perhaps have a common origin, associated with the nesting properties of the open tight binding Fermi surface.

We also report new data regarding the pressure dependence of several other quantities : namely the residual resistivity and magnetoresistance in the metallic region, the anion ordering temperature T_{AO} and the resistivity at several fixed temperatures from 30 to 300 K.

2. Experimental.

Single crystals of $(\text{TMTSF})_2\text{ClO}_4$ were prepared in the usual way by electrocrystallization. The dimensions of the crystals used for resistivity were typically $4.5 \times 0.15 \times 0.1$ mm³ along **a**, **b'** and **c*** respectively with 1.5 mm between the voltage contacts. Four electrical contacts were made by evaporating 0.1 or 0.2 mm wide gold contact areas and attaching 17 μm gold wires with silver paint. Room temperature *a*-axis conductivities were typically $550 (\Omega\text{cm})^{-1}$. A standard 77 Hz AC method with a measuring current of 10 μA was used for resistance (*R*) measurements as a function of *T*, *p* and *H*. The crystals were oriented with **H** // **c*** by eye.

Magnetic fields and variable temperatures (in these first experiments the minimum temperature was 1.5 ± 0.1 K) were provided by an Oxford Instruments superconducting magnet cryostat system. The copper-beryllium alloy pressure bomb was cooled from below by opening a needle valve to the main liquid helium reservoir. The helium gas pressure in the bomb was measured by a calibrated pressure transducer on the room temperature end of the pressure capillary. In most cases, for *p* between 0.5 and 1.5 kbars, there was an anomaly (~ 2 K wide) in the *R(T)* curve which started at the expected helium solidification temperature, thus giving an extra check of the pressure in the bomb.

In order to obtain the R-state the crystals were slowly cooled through the anion ordering temperature (T_{AO}), typically from 35 K to 4.2 K in 2-3 hours.

The samples were initially cooled from room temperature under pressures of 1 or 2 kbars. All subsequent changes in pressure were made at low temperatures which minimized problems associated with resistance jumps which often occur on cooling these crystals. Most of the results presented here were obtained with a sample having a resistance ratio $R(295 \text{ K})/R(1.5 \text{ K})$ of 144 at 1 bar (sample # 6). The results in figures 4 and 5 are for samples # 6 and # 7. The latter had a lower resistance ratio ($R_{293}/R_{32} = 10$ at 1 bar compared with 20 for sample # 6).

3. Results.

Figure 1 shows typical magnetoresistance curves at representative pressures for the lowest temperature obtainable in the present work, $T = 1.5 \pm 0.1$ K. The curve at atmosphere pressure closely resembles that given by Kajimura *et al.* [7], but the anomalies occur at lower fields because in our case **H** is parallel to **c***. The characteristic fields of the anomalies (H_1 , H_2 , etc...) are defined as the points where quasi linear behaviour begins or ends, as indicated by the dashed lines and the arrows in figure 1 (i.e. beginning or end of the plateaus in derivative dR/dH , insert of Fig. 1).

Fig. 1. — Representative magnetoresistance curves for three pressures, 1 bar, 0.74 kbars and 1.49 kbars (10 times expanded) at 1.5 K, for fields up to 11.4 Tesla along c^* . This sample had a room temperature resistance of 1.8 ohms corresponding to an a -axis conductivity of $550 (\Omega \text{ cm})^{-1}$. The insert shows the derivative dR/dH at ambient pressure. The arrows show how the characteristic fields are defined.

Data at all pressures are summarized in figure 2 where the characteristic fields are plotted *versus* pressure. As indicated by the various symbols, at $p = 1$ bar we can identify these anomalies in terms of the phase diagram given by other authors. In particular it is clear that the anomaly at H_5 corresponds to the highest phase transition reported by Naughton *et al.* [10] and that the anomaly at H_1 corresponds to the threshold field (H_T) reported by many groups [7, 8, 10, 15]. Thus both H_T , the threshold field at which the SDW first occurs, and H_5 , where there is a strong first order transition [7, 10] and the electron entropy is completely lost [10], increase very strongly with pressure at approximately the same rate, 25-40 % per kbar depending on the characteristic field and the pressure range considered.

The remaining results refer to the effect of pressure in the metallic state below the threshold field. Figure 3 shows the variation of magnetoresistance at 6 T with pressure and also that, for this crystal with a resistance ratio of 144 at 1 bar, pressure strongly reduces the low temperature

Fig. 2. — Pressure dependence of the characteristic fields H_1 to H_5 with same notation as in figure 1. The symbol $\circ$ refer to the present work and the lines are guides to the eye. The other symbols refer to characteristic fields determined by other authors : at ambient pressure from magnetization data ($\times$, Ref. [9]), magnetoresistance ($\blacksquare$, Ref. [6], fields multiplied by $\cos 33.5^\circ$) or composite data from Ref. [7] ($\square$), and also under pressure ($\bullet$, Ref. [14]).

Fig. 3. — Low temperature resistance $R(1.5 \text{ K})$ as a function of pressure, $\circ$ symbols and left hand scale. Ratio of resistance at 6 T and 1.5 K to that at zero field, $\times$ symbols and right hand scale. The numbers indicate the sequence in which pressure was applied.

resistivity (note that in these crystals the resistance ratio is affected both by the number of defects and by the size on any irreversible resistance jumps which often occur on cooling, at temperatures around 180 K. The above crystals showed jumps near 150 K corresponding to a total resistance increase of about 50 %. One major advantage of the helium gas technique is that the pressure was then altered at 60 K or below and no subsequent jumps occurred. There appear to be no quantitative changes in the magnetoresistance with pressure but a detailed discussion of this requires a specific picture for the magnetoresistance.

Figure 4 shows the pressure dependence of the anion ordering temperature T_{AO} which was determined from the temperature of the cusp in the $R(T)$ curve on warming slowly, and sometimes on cooling through the transition. We could see no significant changes in the shape of the resistivity anomaly in the pressure range studied. As can be seen from figure 4 the initial slope dT_{AO}/dp , for $p < 200$ bars is substantially larger than at higher pressures. This is consistent with thermal expansion measurements [19].

Figure 5 shows the results obtained for the pressure dependence of the electrical conductivity ($\sigma_{||}$) at various temperatures. The main result is that at room temperature $d\ln \sigma_{||}/dp = 22 \pm 1\%$ per kbar, and is very reproducible from sample to sample. Unlike TTF-TCNQ [20] this derivative shows very little change with temperature in the region studied, namely from 32 to 295 K, $d\ln \sigma_{||}/dp$ remains in the range 22-25 % per kbar.

Fig. 4. — Pressure dependence of the anion ordering temperature T_{AO} as determined from the cusp in $R(T)$ curves ($\circ$, $\times$) results for sample # 6 warming and cooling respectively, ($\blacksquare$) results for sample # 7 cooling slowly. As in figure 3 the numbers indicate the pressure sequence.

Fig. 5. — Pressure dependence of a-axis conductivity at various temperatures, for three samples. (×, Δ) for # 6 and # 7 at 32 K, (O) for # 7 at 60 K, (■) for # 7 at 83.5 K and (●) for # 7 and # 8 at 295 K. The normalizing factor $\sigma(1\text{ bar})$ was determined by extrapolation of the low pressure data, except at 295 K where the measured value was used. Resistance ratios of # 6 and # 7 are given in the text.

4. Discussion.

As mentioned in the Introduction several theoretical treatments of the field induced spin density wave in (TMTSF)₂ClO₄ have been performed in the last year or so. The details of these approaches are different but they do have certain common features. Firstly in the field induced SDW state there are pockets of electrons (and possibly also holes) arising from imperfect nesting associated with the SDW nesting vector **Q**. Secondly the stabilization of a particular SDW state (defined by the SDW amplitude and **Q** value) is favoured by having completely full Landau levels in the high field semi-metallic state (except in Ref. [11]). However the parameter which defines the degree of imperfect nesting seems to be different in the various approaches. For example in reference [11], it is the next nearest overlap integral t'_b , while in reference [13], it is the nearest neighbour overlap integral t_b . On the other hand the authors of reference [12] consider that the nesting is made imperfect because of the superlattice associated with anion ordering. Furthermore the discussion in [12] and [13] is concerned with small pockets of electrons (or holes) while in [11] large pockets are considered. As far as we know the pressure dependence has been considered explicitly only in [13], and from this work one does expect t_b and hence the critical fields to increase with pressure, as is observed. Also from reference [21], one can see that area of pockets increases with t_b .

The present theories (except Ref. [11]) emphasize the importance of 2D behaviour, in particular in understanding the Hall effect plateaus. Thus one possible reason for the increase of H_T with pressure could be that the 2D character is progressively reduced because of increased c^* bandwidth under pressure. However we think that the concomitant increase of the other characteristic fields with pressure tends to go against this interpretation.

Independently of which model is used to interpret our data, we can say that the increased value of H_T shows that the nesting becomes more and more imperfect under pressure. The fact that H_T , H_5 and probably all the other characteristic fields increase at approximately the same rate is consistent with the picture invoking full Landau levels. Furthermore, the strong dependence of the superconducting transition temperature [17, 18] may perhaps also be connected with the pressure dependence of H_T , i.e. with the decrease of nesting under pressure, as suggested by others [16, 22]. The precise mechanism for this is not known although even in a simple band picture the degree of nesting does affect both the screening of electron interactions and the phonon frequencies.

Although we have found that, in agreement with other authors [17, 18], T_{AO} is hardly affected by pressure; nevertheless the initial slope of $T_{AO}(p)$ is significantly larger. This is consistent with recent thermal expansion measurements [19]. Furthermore for the crystals studied here R (1.5 K) is strongly decreased by pressure. This might be an indication that for the standard cooling rate used here the degree of anion ordering is improved by applying pressure, as could be expected from the fact that the disordered state has a larger specific volume [19]. However this needs to be confirmed by measuring samples with higher resistance ratios.

5. Conclusion.

We have studied the pressure dependence of the characteristic fields associated with the field induced spin density wave state in $(TMTSF)_2ClO_4$, at 1.5 K. The main result is that all these fields are strongly increased by relatively low pressures, implying that the nesting of the tight binding Fermi surface is reduced by pressure.

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