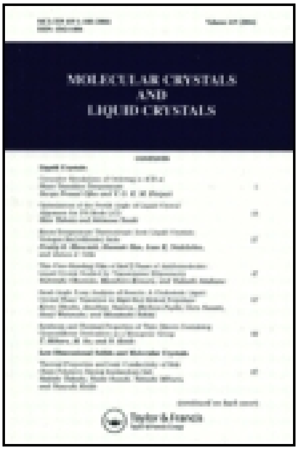

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

Low Temperature Properties of $(TMTSF)_2ClO_4$, under Pressure

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To cite this article: K. Murata , L. Brossard , R. C. Lacoe , M. Ribault , D. Jerome , K. Bechgaard & A. Moradpour (1985): Low Temperature Properties of $(TMTSF)_2ClO_4$, under Pressure, Molecular Crystals and Liquid Crystals, 119:1, 245-249

To link to this article: http://dx.doi.org/10.1080/00268948508075165

Mol. Cryst. Liq. Cryst. 1985, Vol. 119, pp. 245-249
0026-8941/85/1194-0245/$10.00/0

Printed in the United States of America

LOW TEMPERATURE PROPERTIES OF $(TMTSF)_2ClO_4$ UNDER PRESSURE

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Abstract - To examine the role of anion ordering for superconductivity we studied the low temperature properties of rapidly cooled (Q-state) and slowly cooled (R-state) states of $(TMTSF)_2ClO_4$ through resistivity measurements under pressure. In the R-state the anion ordering remained at 25 K up to 8 kb (maximum studied pressure),while the superconductivity disappeared above 3 kb. In the Q-state, below 4.2 K, we observed resistance upturn below 2 kb and metallic behaviour without superconductivity above 3 kb. The superconductivity was suppressed either by anion disorder or by pressures higher than 3 kb.

INTRODUCTION

The pressure, T_c (i.e. the superconducting (SC) transition temperature) phase diagram was studied by D. Mailly et al.[1] by means of magnetization measurements mainly on the relaxed state [2] (slowly cooled below 30 K) of $(TMTSF)_2ClO_4$ tetramethyltetraselenafulvalene-perchlorate. The T_c dropped very rapidly and SC disappered at 2-3 kbar. In addition to the effect of pressure on SC, the effect of anion ordering on SC had not been well investigated.

The purpose of the present work is to look for the role of the anion ordering through resistivity measurements by controlling the order (R-state) and the disorder (quenched state) of the anions at low temperature under pressure.

EXPERIMENT

Samples are prepared by the standard electrochemical oxidation of TMTSF. The resistivity was measured by the standard 4-terminal ac method. Two types of pressure cells were used. One is the Bridgmann cell, the pressure of which was directly regulated out-

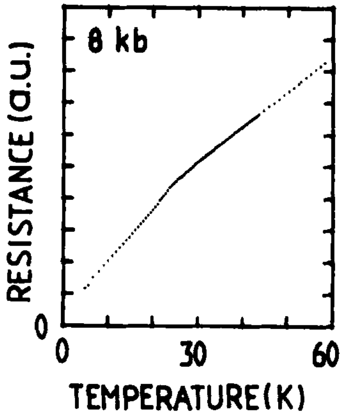


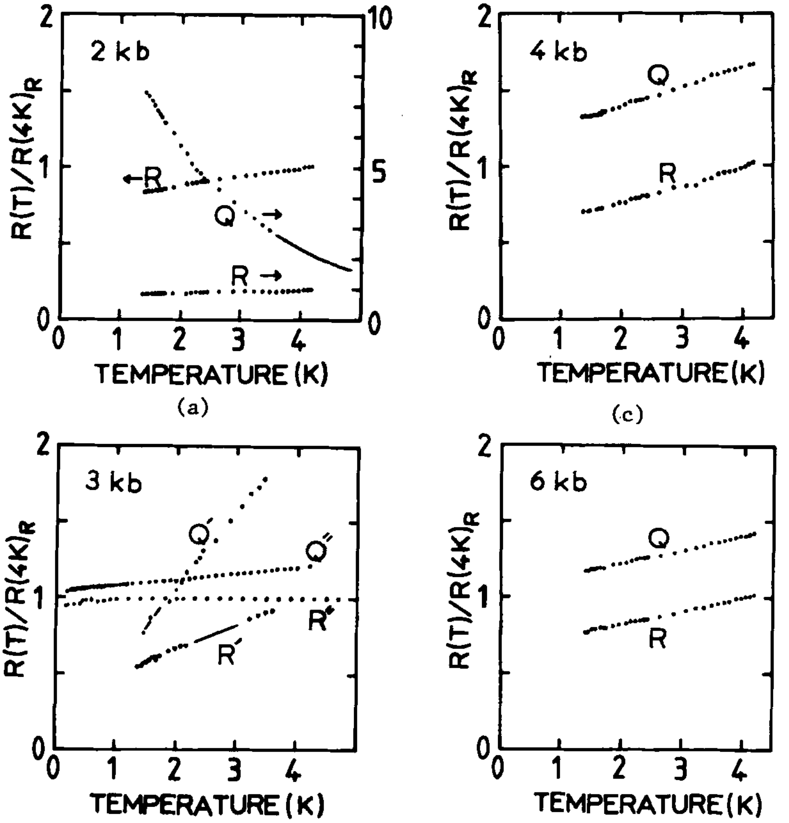
FIGURE 1 - Resistivity vs. temperature at 8 kb in the relaxed state, showing an anomaly around 25 K.

side of the cryostat. The temperature was monitored by the vapor pressure of liquid helium and by the copper-constantan thermocouple inside the pressure cell. Another type of pressure cell, which is used for the experiment at 3 kb down to 0.12 K was the clamp cell, whose pressure calibration between room and low temperature was taken before the actual experiment. A platinum and a germanium thermometer were attached to the pressure bomb which is in thermal contact with the mixing chamber of a dilution refrigerator.

For both apparatus, the pressure medium was isopentance. The cooling rate of the R-state was more than two hours from 30 K to 4.2 K, which is sufficient to observe superconductivity. For achieving the Q-state, we used a heater inside the pressure cell and warmed up to 40 K, which was monitored by the resistance of the sample itself. The time for cooling from 40 K to low temperature was several seconds after the heater was switch off.

RESULTS AND DISCUSSION

The anion ordering, viewed by the resistance anomaly around 25 K, was observed up to 8 kb (the maximum studied pressure) for samples in the relaxed, as shown in Fig. 1. The anomaly could be removed completely by quenching. Figure 2 shows the low temperature resistivity of R- and Q- states at 2, 3, 4 and 6 kbar. The larger temperature coefficient of the resistance observed in the Q-state relative to the R-state at $\sim$ 3 kbar prompted us to measure down to 0.12 K. As shown in Fig. 2(b), the SC was already very weak in



(a) (c)

(b) (d)

FIGURE 2 - Resistivity vs. temperature at 2 kb (a), 3 kb (b), 4 kb (c) and 6 kb (d). Figures are normalized by the resistivity of the R-state at 4 K. Figures of (a), (c) and (d) are of the same sample. Figure (b) is of two different samples distinguished by R' and R".

the R-state. The subtle drop in resistivity in Q-state (Q" in this figure) below 0.3 K might be caused by an imperfect quenching. Combined with the results of D. Mailly et al.[1], we can state that SC in $(TMTSF)_2ClO_4$ requires both anion ordering and a pressure lower than 3 kb.

The fact that the Q-state is unstable to SDW formation while the R-state is unstable to SC at ambient pressure (where in the general phase diagram for $(TMTSF)_2X$ the SDW state occurs at lower pressures than SC), and the preliminary X-ray data[3] suggest the lattice constants in the Q-state are larger than in the R-state. Another explanation for the difference between Q-state and R-state properties is the existence of the disorder itself and the lack of 4 k_F (0,1/2,0) ordering. The results that the region of stability for SC in the R-state is the same as that of the SDW in the Q-state suggests that the difference in the effective pressure is small. This is consistent with the thermal expansion study[4] on both states showing that the difference in a-axis between the two states corresponds to about 0.1 kbar. The difference along other axes is expected to be much less according to the study on the PF_6 salt.[5]

From the point of view of increased scattering centers in the Q-state, it is worth comparing the coherence length of the anion ordering (ξ_a = 200 Å, ξ_b = 35 Å, ξ_c = 60 Å) in the Q-state[4] and the critical alloying concentration necessary to suppress SC in $|(TMTSF)_{1-x}(TMTTF)_x|_2ClO_4$[6] and $(TMTSF)_2((ClO_4)_{1-x}(ReO_4))$[7], where the mean distance between the impurities are 730-180 Å and 360-75 Å, respectively. The stronger "metallic"-like behaviour (enhanced scattering mechanism) exhibited in the Q-state ∿ 3 kbar (larger slope in low temperature resistivity) to that of the R-state indicates the Q-state is not simply the R-state with the introduction of more scattering centers.

CONCLUSION

The existence of superconductivity in $(TMTSF)_2ClO_4$ requires both anion ordering and a low pressure (< 3 kbar). The effect of low pressure on the electronic structure of the Q-state was small.

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