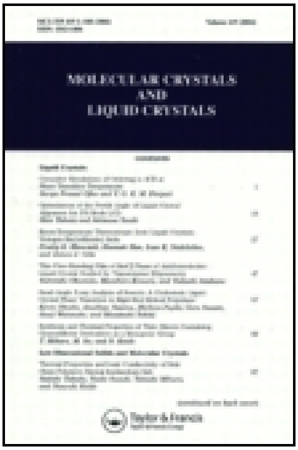

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Narrow Pressure Domain for Superconductivity in $(TMTSEF)_2ClO_4$

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NARROW PRESSURE DOMAIN FOR SUPERCONDUCTIVITY IN $(TMTSeF)_2ClO_4$

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Abstract We present resistivity data for $(TMTSeF)_2ClO_4$ as a function of pressure. We find that the superconducting transition temperature (T_c) drops rapidly as pressure is increased from 1 bar ($T_c \simeq 1.3$ K in slowly cooled samples) to below 50 mK above 4-5 kbar. This is a very rapid rate of decrease as compared to typical inorganic materials and is about three times that observed in the isostructural PF_6 salt. In addition we find evidence that the second order transition near 24 K at 1 bar associated with ordering of the ClO_4 anions is not significantly affected by pressure. We clearly observe this transition at about the same temperature for pressures up to 10 kbar.

1-INTRODUCTION

The ambient pressure phase diagram of $(TMTSeF)_2ClO_4$ is rather complicated and is very sensitive to both sample cooling rate and magnetic field[1,2] The superconducting transition originally found near 1 K[3-4] at low pressures was later shown to depend on the rate of cooling of the material for temperatures near 25 K. Recently it has been shown that at low temperatures an insulating state is stabilised in high magnetic fields[5-6]. In rapidly cooled samples a non-metallic antiferromagnetic ground state is formed[1-2]. Until now little work has been carried out to investigate the phase diagram at higher pressures. We have made resistivity versus temperature measurements on a large number of $(TMTSeF)_2ClO_4$ crystals for pressures up to 10 kbar and find some perhaps unexpected results.

2-RESULTS

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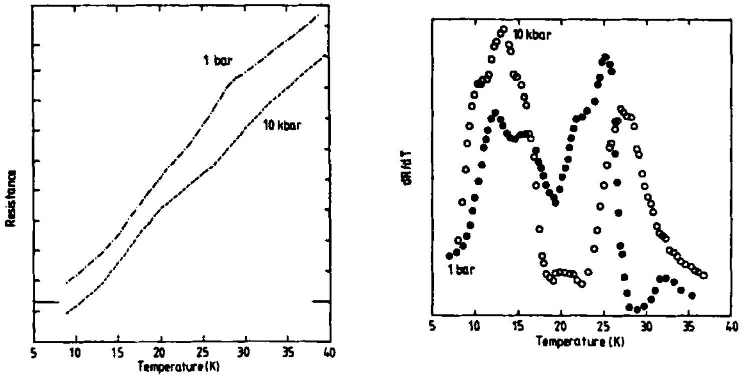

Figure 1: (a) Normalised resistance versus temperature curves for $TMTSeF_2ClO_4$ at 1 bar and 10 kbar. (b) derivative of the resistance curves in figure 1(a) with respect to temperature plotted as a function of temperature.

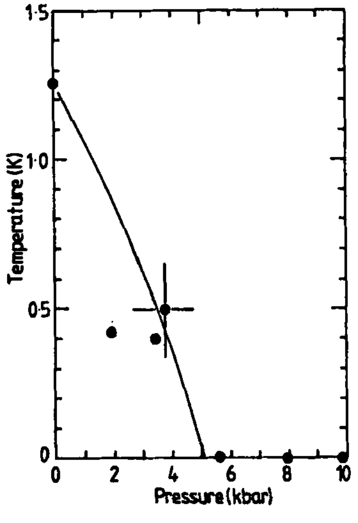

Figure 2: Pressure dependence of the superconducting transition temperature in $TMTSeF_2ClO_4$.

Resistance was measured using a standard low frequency lock-in technique with four leads attached to the crystals via silver paste contacts. Checks were made for anomalous current paths through the samples. Pressure was applied in a Be-Cu bomb with the samples contained in teflon caps filled with hexane or a mixture of organic fluids to wash out the potentially damaging effect of the freezing of the fluid on the crystals. Typical data are given in Figure 1 at 1 bar and 10 kbar for temperatures below 40 K. The low temperature data below 4 K are not shown.

At 1 bar we see a well defined superconducting transition near 1.3 K for slowly cooled samples. At 10 kbar we observe no superconducting transition and the resistivity data appear to be less sensitive to cooling rate. We find the latter to be true for all pressures we have considered. The dependence of T_c on pressure is summarised in Figure 2. Note in figure 1(a) the transition at 25 K which has been shown to be associated with ordering of the ClO_4^- anions with a wave-vector $\underline{Q}$ = (0, 1/2, 0) [11]. The transition can be more clearly identified by deriving the derivative of the resistance curve. This is shown in figure 1(b). These data suggest a second anomaly near 15 K. Similar ambient pressure resistance data by others also indicate the possible presence of a second anomaly[12]. The data at 10 kbar given in figure 1(a) and 1(b) suggest both these anomalies are present under pressure at temperatures not significantly different from those at 1 bar. We find similar data at intermediate pressures. These data suggest the 25 K anomaly at 1 bar is shifted to slightly higher temperatures under pressure.

3-DISCUSSION

The pressure dependence of T_c in $(TMTSeF)_2ClO_4$ is unusually large and is about three times greater than that observed in the isostructural PF_6 salt (where $dT_c/P \simeq -0.09$ K $kbar^{-1}$ [7-8]). Moreover the superconductivity is essentially gone above 4 kbar or at least T_c is less than 50 mK, the lower limit of our measurements. It may be possible that the resistive superconducting transition is merely so smeared out in temperature that we could not detect it. Certainly the complete suppression of superconductivity by such modest pressure is quite anomalous unless some crystal phase change occurs with pressure. There is no evidence from our measurements that such a phase change takes place.

The pressure variation of T_c can be discussed using the approximate relation $T_c = <\omega>\exp[-(1+\lambda)/\lambda]$. Here $\lambda = N(0)<I^2>/M<\omega^2>$ is the dimensionless electron-phonon coupling constant, $<I^2>$ is the Fermi surface average of the electron- phonon matrix element and $<\omega>$ is the average phonon energy. Two mechanisms can be considered which would lead to a decreased λ, and hence decreased T_c under pressure. Either $<\omega^2>$ is enhanced or $N(0)<I^2>$ is decreased. Without other measurements we do not know exactly how $N(0)<I^2>$ will change with pressure but for the PF_6 salt it has been postulated that this changes very little[7-8]. Then the decrease in T_c would require a large change in $<\omega>$ with pressure such that $\Delta<\omega>/<\omega>\geq 20\%$. It is known that large changes in $<\omega>$ do occur in charge transfer compounds[9], however whether or not their magnitude is large enough to account for the observed values of dT_c/dP in $(TMTSeF)_2ClO_4$ will require further measurements of the compressibility of this compound. It has been suggested that the known changes in $<\omega>$ for compounds like TTF-TCNQ are about right to explain dT_c/dP for the PF_6 salt[8].

If T_c is really suppressed to T=0 as indicated in figure 2 then this may indicate some new phenomenon ocurring in the ClO_4' salt that is quite different from that found in other known (organic and inorganic) superconductors. On the other hand the pressure dependence of other properties is very large in the ClO_4 salt so it may be that we are just very close to some critical parameter at ambient pressure. For example the threshold field for the onset of oscillations in the magnetotransport properties of the ClO_4 salt is dramatically increased with only a few kilobars of pressure[10]. Also we have found that the magnetoresistance in the ClO_4 salt is greatly reduced at low pressures compared to comparable temperature and field regimes at 1 bar. Note that these small values of magnetoresistance are comparably small to those we have measured in the $(TMTSeF)_2BF_4$ at pressures just sufficient to suppress the anion ordering transition ($P_c^A \simeq 4.5$ kbar). Our measurements show that the BF_4 salt is also not superconducting (down to 20 mK) for pressures above P_c^A.

One of the most interesting questions raised by these studies is the effect of the order or disorder of the ClO_4^- anions on the superconducting state. It has been conjectured that these materials will not become superconducting if the anions are disordered at low temperatures [1-2] because of the strong anion disorder potential that the electrons on the organic molecular stacks would be subject to. One then supposes that rapidly cooling the ClO_4 salt through the anion order-disorder temperature destroys the superconducting state in favour of the antiferromagnetic state. However our measurements show that the superconducting state must depend very sensitively on some other parameter perhaps in addition to anion order since T_c has such a strong dependence on pressure whilst the anion ordering transition temperature has a very weak dependence. This parameter may itself depend on the anion arrangement, perhaps through some structural dependence, which in turn may account for the anomalous dependence of $(TMTSeF)_2ClO_4$ on the low temperature cooling rate at 1 bar. Note that we find no such dependence at low pressures.

REFERENCES

1 T. Takahashi, D. Jerome and K. Bechgaard, J. de Physique 43, L565 (1982).
2 P. Garoche, R. Brusetti and K. Bechgaard Phys. Rev. Lett. 49, 1346 (1982).
3 K. Bechgaard *et al* Phys. Rev. Lett. 46, 852 (1981).
4 S.S.P. Parkin, M. Ribault, D. Jerome and K. Bechgaard, J. Phys. C14, 5305 (1981).
5 M. Ribault *et al* (these proceedings).
6 P.M. Chaikin *et al* (these proceedings).
7 H.J. Schulz, D. Jerome, M. Ribault, A. Mazuad and K. Bechgaard, J. de Phys. 42, L51 (1981).
8 R.L. Greene and E.M. Engler, Phys. Rev. Lett. 45, 1587 (1980).
9 B. Horovitz, H. Gutfreund and M. Weger, Mol. Cryst. Liq. Cryst. 79, 595 (1982).
10 J.F. Kwak (unpublished).
11 J.P. Pouget, R. Moret, R. Comes, G. Shirane, K. Bechgaard and J.M. Fabre, J. de Physique 44, C3-969 (1983).
12 D.U. Gubser *et al*, Mol. Cryst. Liq. Cryst. 79, 225 (1982).