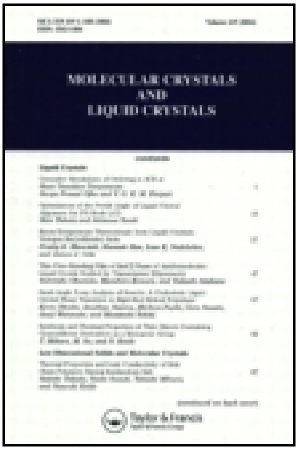

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Coulomb Repulsion in $(TMTSF)_2X$ and $(Tmttf)_2X$

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COULOMB REPULSION IN $(TMTSF)_2X$ AND $(TMTTF)_2X^*$

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Abstract On the basis of studies of transport properties of $(TMTSF)_2X$, $(TMTTF)_2X$ and their binary alloys we discuss the role of on-site Coulomb repulsion relative to the transfer integrals. In TMTTF-salts U/t_a are believed to be large, resulting in a Hubbard gap, whereas U/t_a in TMTSF-salts are weak or of intermediate size. We suggest that the difference in U/t_a is a result of difference in U rather than difference in t_a and moreover that U is closely related to the interchain banding (t_b).

INTRODUCTION

It is interesting to contrast the physical properties of the two closely related families of synthetic conductors, $(TMTSF)_2X$ and $(TMTTF)_2X$.[1] At ambient pressure the two classes of materials exhibit significant differencies in physical properties. The TMTSF salts are generally quite good conductors with metallic characteristics over wide T-ranges[1]. The metallic state is eventually destroyed by opening of a gap at E_F as caused either by SDW formation or by creation of a $2\times\underline{a}$ potential due to anion ordering. The $(TMTTF)_2X$ salts, on the opposite, show semiconducting behaviour.[2] The shallow maximum in $\sigma(T)$, typically seen at $T \approx 200$ K is not reflecting a MI transition, but due to competing T-dependencies of mobility and number of charge carriers. The thermopower shows accordingly linear 1/T relationship. In the TMTTF salts the formation of both SDW state and $2\times\underline{a}$ anion potential have only minor influence on the transport properties.

In the present paper we show that the difference in the rela-

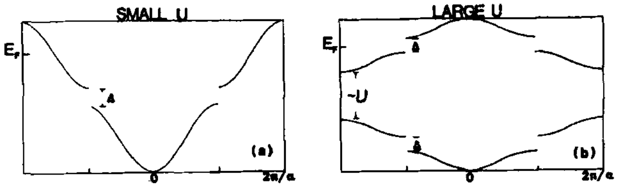

FIGURE 1 Band structure of $(TMTSF)_2X$ (a) and $(TMTTF)_2X$ (b).

tive Coulomb repulsion can account for these diverging ambient pressure properties. On the basis of a study of alloys of the form $(TMTSF_{1-x}TMTTF_x)_2PF_6$ (Figs. 2 and 3) we discuss the individual roles of on-site Coulomb repulsion, U, intra chain transfer integral, t_a, and transverse transfer integral, t_b.

HUBBARD GAP IN TMTTF

The metallic character of TMTSF salts imply that U/t_a is so small that the electrons near E_F are not markedly affected by the dimerization gap (Δ) (Fig. 1a). In salts of TMTTF, on the contrary, U/t_a is sufficiently large to effectively separate the band into the two (dimerized) Hubbard subbands (Fig. 1b). This brings E_F into the dimerization gap. The $T \rightarrow \infty$ value of S is, though, only 35 μV/K, which is significant less than the $k/e \cdot \ln 2 \approx 60$ μv/K value expected in the $U/4t_a \gg 1$ limit of symmetric subbands. This may indicate that U is not very much larger than $4t_a$. At approximately 120 K, the $(TMTTF)_2PF_6$ thermopower changes character. A new linear S vs. 1/T relationship appear with $S(T \rightarrow \infty) \approx 60$ μv/K. Similar changes are observed in other MF_6 salts[2,3]. This may reflect some kind of transition, which causes an enhanced effective $U/4t_a$ ratio.

BAND-TERM DOMINANCE OF TMTSF-THERMOPOWER

When dilute concentrations (x) of TMTTF are induced into the stacks of TMTSF molecules, dramatic effects are observed in a number of

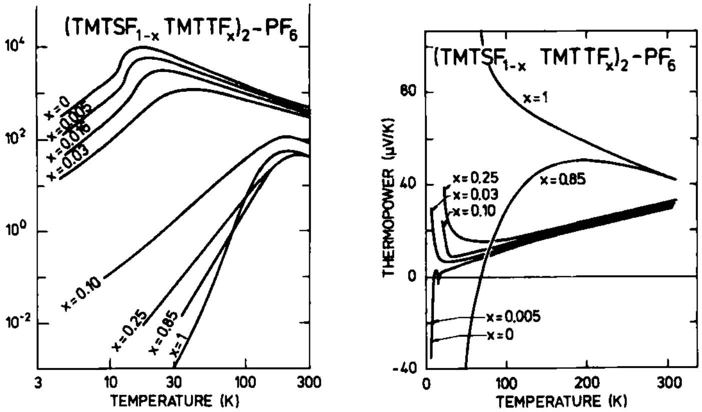

FIGURE 2 Conductivity and thermopower of $(TMTSF_{1-x}TMTTF_x)_2PF_6$

physical properties (Fig. 3)^5. For example the MI-transition temperature increases dramatically and the EPR susceptibility and g-value are changed disproportionally towards the TMTTF-values. The conductivity, as shown in Figs. 2 and 3, is markedly suppressed, indicating changes in dominating scattering processes, whereas the thermopower remains unaffected except for the lowest T range.

Conventionally the thermopower is separated into two terms: One reflecting the band properties, $S_1 \propto 1/t\cdot \cos ka/\sin^2 ka$, and one reflecting the scattering mechanism, $S_2 \propto 2d\ell n\tau/dE$. The absence of any S vs. x dependence simultaneously with marked σ vs. x, and thus τ vs. x, dependences show that the termopower is dominated by the band term.

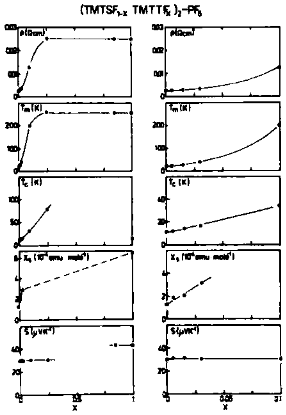

FIGURE 3 Review of some $(TMTSF_{1-x}TMTTF_x)_2PF_6$ characteristics as a function of x.

HUBBARD U AND DIMENSIONALITY

The $(TMTSF_{1-x}TMTTF_x)_2PF_6$ alloys of x=0.1 and x=0.25 exhibit unusual behaviour. The termopower displays metallic characteristics similar to the pristine TMTSF salt, whereas the conductivity appears to be basically activated. The metallic thermopower excludes the excistence of a gap at the Fermi level.

The band properties of stacks of 10% and 25% TMTTF must be determined by both the MO of TMTSF and the MO of TMTTF. Since the thermopower, as shown above, is given solely by the band-term, the absence of any x-dependence for x up to 0.25 suggests that the stacking axis transfer integral, t_a, associated with TMTSF and TMTTF are approximately identical. Therefore, the difference in U/t_a is related to difference in U rather than difference in t_a.

The large U localized on the TMTTF sites prevent the charge carriers to move freely through the chains. Therefore, the mobility is activated. Moreover, the large U of TMTTF favour single occupation of these sites. The TMTTF molecules contribute accordingly disproportionally to the EPR parameters.

The enhanced U of TMTTF relative to U of TMTSF may be related to the degree of one-dimensionality. The smaller transverse transfer integral associated with TMTTF causes the electrons to be localized on a relative small spatial size. This causes a large Coulomb repulsion for a second electron on an already ocupied site.

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