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The Effect of Hydrostatic Pressure on the Transport Properties of the Organic Charge Transfer Salt $(TMDTDSF)_2PF_6$

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THE EFFECT OF HYDROSTATIC PRESSURE ON THE TRANSPORT PROPERTIES OF THE ORGANIC CHARGE TRANSFER SALT $(TMDTDSF)_2PF_6$.

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Abstract - The transport properties of $(TMDTDSF)_2PF_6$ were measured as a function of temperature and hydrostatic pressure. At ambient pressure, the conductivity of $(TMDTDSF)_2PF_6$ increases slowly below room temperature, reaching a broad maximum at $\sim$ 220 K before decreasing sharply at lower temperatures. The room temperature conductivity, $\sigma(RT) \sim 40(\Omega\text{-cm})^{-1}$ while $\sigma(max/\sigma(RT) \sim 2.0$. The ambient pressure thermoelectric power (TEP) decreases linearly with temperature over the entire temperature range measured, indicative of an ungapped Fermi surface. As pressure is increased, the temperature of the conductivity maximum is suppressed ($\sim$ 10 K/15 kbar), while the character of the TEP remains unchanged. These results are discussed with respect to the physical properties of $(TMTSF)_2PF_6$ and $(TMTTF)_2PF_6$.

The ambient and high pressure properties of $(TMTSF)_2X$, and their all sulfur (S) isostructural [1] analogues $(TMTTF)_2X$ appear to be quite different.[2,3,4] For example,[2] $(TMTSF)_2PF_6$ undergoes a SDW transition at $\sim$ 12 K at ambient pressure, while at P $\sim$ 9 kbar a superconducting ground state is stabilized at T $\sim$ 1.4 K. Alternately, $(TMTTF)_2PF_6$ is a poorer room temperature conductor than its all selenium (Se) analogue ($\sigma(RT) \sim 40(\Omega\text{-cm})^{-1}$), and the conductivity (σ) shows a broad max $\sim$ 230 K, below which σ decreases exponentially, with an apparent temperature dependent activation energy $\sim$ 600 K. At low temperature, magnetic[3,5] and X-ray data [6] indicates the material undergoes a transition to a spin-Peierls state. Application of pressure suppresses the conductivity maximum

to low temperature [4] and NMR data [7] suggests the ground state is magnetic in character.

The molecule TMDTDSF is intermediate between the all S and all Se analogues, containing one S and one Se each side of the C=C bond.[8] The salt $(TMDTDSF)_2PF_6$ (isostructural to Bechgaard salts[1,8]) is hence expected to be disordered in the arrangement of S and Se. A comparison of structural data[1,8], such as lattice parameters and shortest interstack distance (SX-SX, SX = S or Se) shows $(TMDTDSF)_2PF_6$ to be truly intermediate between $(TMTTF)_2PF_6$ and $(TMTSF)_2PF_6$.

The ambient pressure σ and TEP are shown in Figure 1. The $\sigma(RT) \sim 40\ (\Omega\text{-cm})^{-1}$, the broad maximum in dc σ at ~ 220 K (T_m) and activated σ below T_m with characteristic energy $\Delta \sim 600$ K is suggestively similar to that of $(TMTTF)_2PF_6$. The TEP is smooth over the entire temperature range measured, indicating no gap has opened over the Fermi surface.

At ~ 3 kbar, the resistivity minimum is depressed to ~ 140 K, and the shift to semiconductor-like behaviour is sharper than at ambient pressure. For higher pressures (9-15 kbar), the resistivity remains metallic down to ~ 10 K, below which it gradually increases with decreasing temperature (Figure 2). At ambient pressure, the TEP at 9 kbar was measured and shows no sign of a transition.

It is clear from Figure 1, that for T > 60 K, the conductivity of $(TMDTDSF)_2PF_6$ and $(TMTTF)_2PF_6$ are nearly identical (same

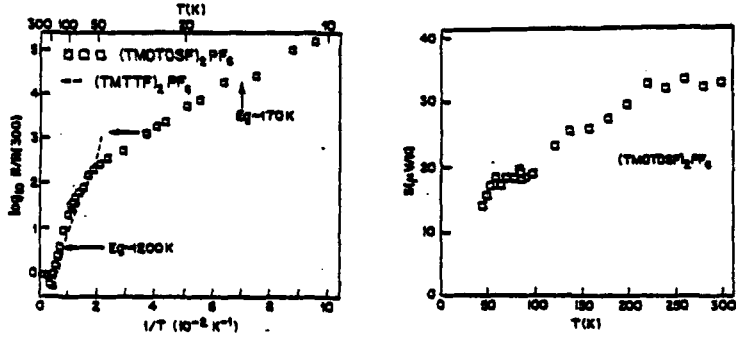

FIGURE 1 - Conductivity and TEP of $(TMDTDSF)_2PF_6$ at ambient pressure. Results for $(TMTTF)_2PF_6$ from reference 3.

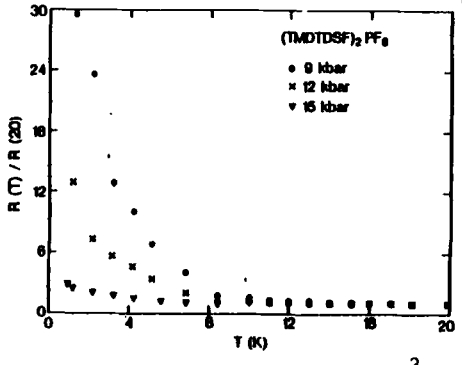

FIGURE 2

R(T)/R(20 K) versus T for $(TMDTDSF)_2PF_6$ at P = 9, 12 and 15 kbar.

value of $\sigma(RT)$, T_m and Δ). This suggests the physical mechanisms producing this similar behaviour are the same in both salts. Coulon et al.[3] suggested the lower dimensionality expected in the all S TMTTF salts relative to the all Se TMTSF salts results in localization of the electrons on a chain. Mortensen et al.[9] suggested the localization is a result of electron correlation, which results in a room temperature gap. They explain the conductance behaviour as resulting from the combination of a room temperature gap and a temperature dependent mobility. The TEP, which has the form $\beta(T)/T$, where $\beta(T)$ is a temperature dependent coefficient, is attributed to both the combined effect of electron correlations and impurities.[9] The difference in the TEP of $(TMDTDSF)_2PF_6$ (which indicates an ungapped Fermi surface) and $(TMTTF)_2PF_6$, in light of their similar conductivities, is peculiar, and suggests that a room temperature gap may not be present in either salt.

If one explains the conductivity behaviour as resulting from a mobility gap, one again encounters problems with respect to the TMTTF salt. That is, correlations between such parameters as $\sigma(RT)$, T_m, $\sigma_m/\sigma(RT)$ ($\sigma_m \equiv \sigma(T_m)$) and the shortest S-S contact were made by Coulon et al.[3] to support a picture of one-dimensional localization. The values for $\sigma_m/\sigma(RT)$ are 2.0 and 1.1 for $(TMDTDSF)_2PF_6$ and $(TMTTF)_2PF_6$, respectively. However, since the former salt may be disordered, one would not expect the valve of $\sigma_m/\sigma(RT)$ to exceed that of the latter salt. Also, it should be noted the difference in the conductivity at low temperature between the two salts could result from the disorder possible in the $(TMDTDSF)_2PF_6$,

leading to a contribution to σ from variable range hopping.

The pressure necessary to suppress to low temperature the conductivity decrease is considerably less in $(TMDTDSF)_2PF_6$ than in $(TMTTF)_2PF_6$ (ref. 4). The transition in the former is also vary broad (decrease in σ slower than $\exp-1/T^{1/4}$). Whether the transport behaviour represents a transition to a magnetic ground state as in $(TMTTF)_2PF_6$ (ref. 7), with the lower pressure required reflecting a difference in compressibility between the two salts, or some other phenomenon will require future study.

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