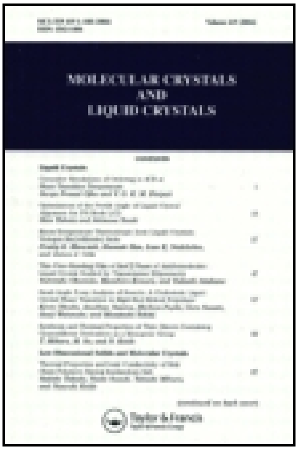

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High-Resolution Thermal Expansion and Magnetostriction at Low Temperature in Organic Compounds

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HIGH-RESOLUTION THERMAL EXPANSION AND MAGNETOSTRICTION AT LOW TEMPERATURE IN ORGANIC COMPOUNDS

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Abstract - We present the first low temperature measurements of thermal expansion and magnetostriction on $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$. Above 40 K, the thermal expansion of the two salts is very large and follows a quite linear behaviour. Within our sensitivity ($\Delta\ell/\ell \sim 10^{-8}$) no anomaly is present at the metal to insulator transition in $(TMTSF)_2PF_6$ ($T \sim 14$ K). In $(TMTSF)_2ClO_4$ we clearly observed a contraction of the lattice due to the anion ordering, the relative difference between quench and relax state at 1 K being about 5.10^{-4}.

The thermal dependance of lattice parameters in organic compounds, and especially in TMTSF salts is usually measured by X-ray techniques [1-6]. Unfortunately the sensitivity of such experiments is not sufficient to perform low temperature measurements, in a region where one expects to observe interesting correlations between the lattice behaviour and properties like the metal to insulator transition in $(TMTSF)_2PF_6$ (T = 12 K) or anion ordering in $(TMTSF)_2ClO_4$ (T = 24 K).

In order to measure the thermal expansion and the magnetostriction (in fields up to 7 T) with a high sensitivity ($\Delta\ell/\ell \sim 10^{-8}$), we useda capacitance technique which is rather usual for bulk samples. Details will be found eleswhere [7], but roughly itis similar to the dilatometer used by Schafer et al. [8] for TTF-TCNQ crystals, with a higher sensitivity. All the data presented here were obtained along a-axis.

On $(TMTSF)_2PF_6$ the main results are :
i) within our sensitivity, no sign of anomaly in the thermal expansion (Fig. 1a) has been observed at the metal to insulator transition (T $\sim$ 12 K) whose existence was checked by resistivity measurements on a sample from the same batch. This result confirms, with at least two orders of magnitude better sensitivity, the

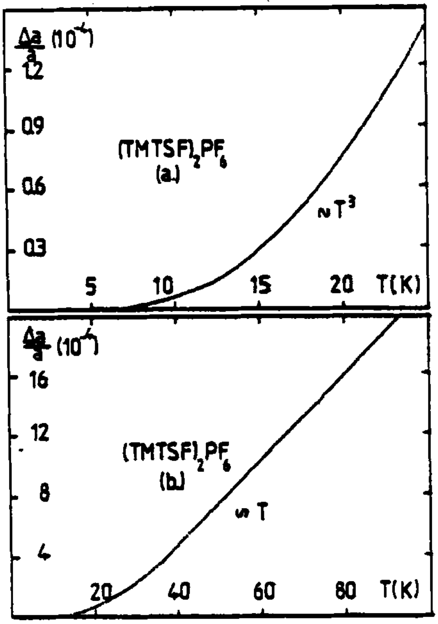

FIGURE 1 - Thermal expansion of $(TMTSF)_2PF_6$.

observation made by X-ray techniques [1-2-6]; it suggests also that the anomaly observed in sound velocity measurements by Chaikin et al. [9] is not due to thermal expansion.

ii) At low temperature, where one expects to have electronic and magnetic contributions to be dominant in the thermal expansion, we observe a quasi T^3 behaviour (Fig. 1a) ; at higher temperature (Fig. 1b) the effect is following a T law.

This behaviour is compatible with a phonon dominated thermal expansion over the whole temperature range.

iii) At 1.4 K the magnetostriction, in fields up to 7 T (parallel) and 4 T (perpendicular) is very small (less than 10^{-6}) probably due to the weakness of the staggered magnetization.

iv) Finally, the thermal expansion is of the same order of magnitude as in previous X-ray results [1-2-6] (which are not consistent together) but smaller ; the discrepancies can be ascribed to the big uncertainties in X-ray measurements at low temperature.

The thermal expansion of the perchlorate salt (Figure 2) exhibits, like in PF_6, a linear law for temperature greater than 40 K ; on the contrary, the low temperature part is very particular and clearly correlated with the problem of anion ordering.

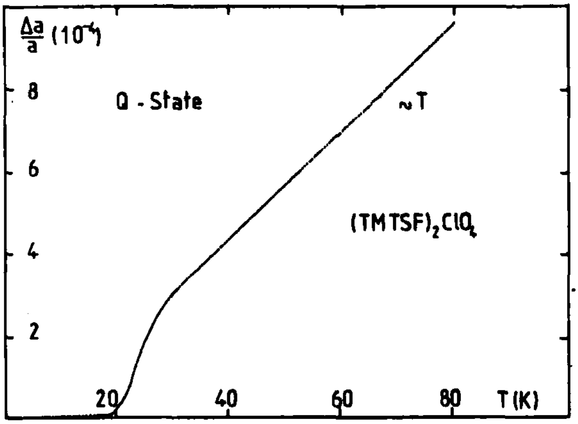

FIGURE 2

Thermal expansion of $(TMTSF)_2ClO_4$ in the quench state.

In fact, below the characteristic temperature of anion ordering (T ∿ 24 K) the thermal expansion (Fig. 3) is very sensitive to the cooling rate, from 0.2 K/mn (relax-state) to 2 K/s (quench-state) ; the general behaviour around 24 K is similar to that of the resistivity [10]. The relax curve shows that the disordering process begin at about 10 K, following by a strong increase leading to a transition which is complete by 24 K . For T → 0, the total relative gap between the two states is about 5.10^{-4}. Assuming a compressibility of 0.5 %/kbar, which had been measured on

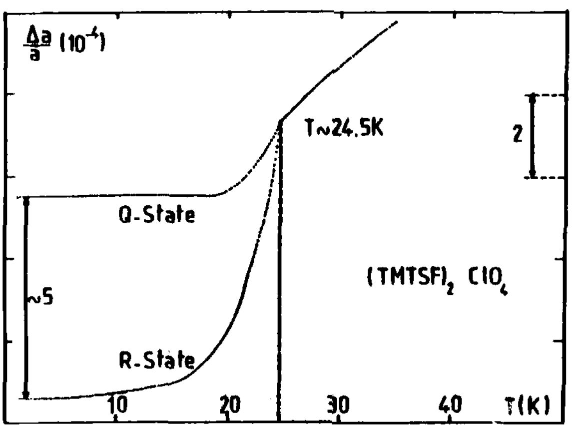

FIGURE 3

Low temperature part of the thermal expansion of $(TMTSF)_2ClO_4$ as a function of the cooling rate.

the PF_6 salt [11], the equivalent pression is about 100 bars ; this low value indicates that, rather than the change of lattice parameter as suggested by Kagoshima et al. [12], it is the order of the anions which plays the most important role in the appearance of superconductivity. Finally, for the magnitude of the thermal expansion (with respect to X-ray measurements [4]) and field effects, we can make the same remarks as in the PF_6 salt.

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