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Publisher: Taylor & Francis

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Molecular Crystals and Liquid Crystals

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

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

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To cite this article: C. Gaonach, G. Creuzet & A. Moradpour (1985): High-Resolution Thermal Expansion and Magnetostriction at Low Temperature in Organic Compounds, Molecular Crystals and Liquid Crystals, 119:1, 265-268

To link to this article: http://dx.doi.org/10.1080/00268948508075168

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Mol. Cryst. Liq. Cryst. 1985, Vol. 119, pp. 265-268
0026-8941/85/1194-0265/\$10.00/0

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Printed in the United States of America
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) $_2$ PF $_6$ and (TMTSF) $_2$ ClO4. Above 40 K, the thermal expansion of the two salts is very large and follows a quite linear behaviour. Within our sensitivity ($\Delta\ell/\ell\sim10^{-8}$) no anomaly is present at the metal to insulator transition in (TMTSF) $_2$ PF $_6$ (T \sim 14 K). In (TMTSF) $_2$ ClO4 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 used a capacitance technique which is rather usual for bulk samples. Details will be found eleswhere 7, but roughly it is 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) $_2$ PF $_6$ 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

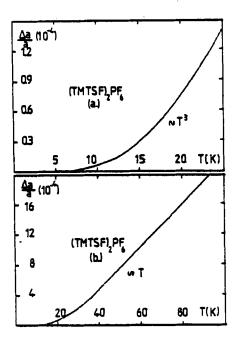


FIGURE 1 - Thermal expansion of (TMTSF) 2PF6.

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 T³ behaviour (Fig. la); at higher temperature (Fig. lb) the effect is following a Tlaw.

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

iii) At 1.4 K the magnetostric-

tion, 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 PF₆, 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.

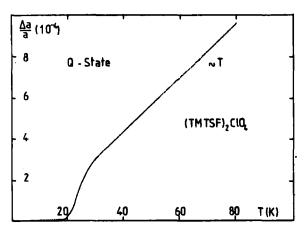


FIGURE 2
Thermal expansion of (TMTSF)₂ClO₄ in the quench state.

In fact, below the characteristic temperature of anion ordering (T \sim 24 K) the thermal expansion (Fig. 3) is very sensitive to the cooling rate, from 0.2 K/mm (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 \rightarrow 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

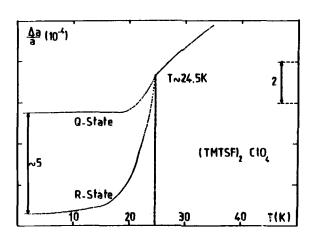


FIGURE 3

Low temperature part of the thermal expansion of (TMTSF) 2ClO4 as a function of the cooling rate.

the PF₆ 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 PF₆ salt.

<u>Acknowledgements</u> - We thank D. Jérome, F. Creuzet and 1.A. Campbell for helpful discussions.

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