

Spin Dynamics of Organic Linear-Chain Compounds

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Despite two decades of research, the organic charge-transfer salts (TMTCF)₂X ($C = S$ or Se , $X = PF_6$, AsF_6 , ClO_4 , Br , ...) attract considerable interest. The reduced dimensionality leads to instabilities in the electronic system, various ground states like charge-density wave, spin-Peierls, spin-density wave, or superconductivity develop at low temperatures. By slightly changing the organic molecules or the anions, the electronic and magnetic properties can be changed significantly [1].

We carried out electron-spin-resonance experiments in a continuous wave X-band spectrometer on single crystals of (TMTCF)₂PF₆, (TMTCF)₂ClO₄, and (TMTTF)₂Br in the temperature range $1.5 \text{ K} \leq T \leq 500 \text{ K}$ [2]. Here we confine ourselves to results of the spin-chain compounds (TMTTF)₂ClO₄ and (TMTTF)₂PF₆ at $2 \text{ K} \leq T \leq 400 \text{ K}$ (Fig. 1). In the high-temperature regime the spin susceptibility χ_s of both compounds follows the predicted behavior of a $S = 1/2$ antiferromagnetic Heisenberg chain with an exchange constant $J \approx 420 \text{ K}$. The linewidth ΔH is slightly anisotropic and increases almost linear with temperature.

In (TMTTF)₂ClO₄ the ordering of the anions leads to a first-order structural phase transition at $T_{AO} = 72.5 \text{ K}$. This is accompanied by a dimerization of the spin chain leading to a strong decrease of χ_s for $T < T_{AO}$. We determined an alternation parameter $\gamma = 0.83$ and a singlet-triplet gap $\Delta \approx 82 \text{ K}$. At $T \approx 14 \text{ K}$, the ESR signal splits into five lines. It is known from ¹H-NMR experiments that the methyl groups which rotate at high temperatures slow down in this temperature range [3]. Thus the rotational narrowing might be responsible for the hyperfine splitting.

(TMTTF)₂PF₆ undergoes a spin-Peierls transition at $T_{SP} = 19 \text{ K}$. Below T_{SP} , the ESR linewidth ΔH increases and χ_s vanishes exponentially with decreasing temperature. Using mean-field theory, we determined an alternation parameter $\gamma(0) = 0.91$ and a singlet-triplet gap $\Delta(0) \approx 34.5 \text{ K}$. The visible decrease of the spin susceptibility along the b and c direction below $T_{SP}^0 \approx 62 \text{ K}$ can be attributed to 1-D

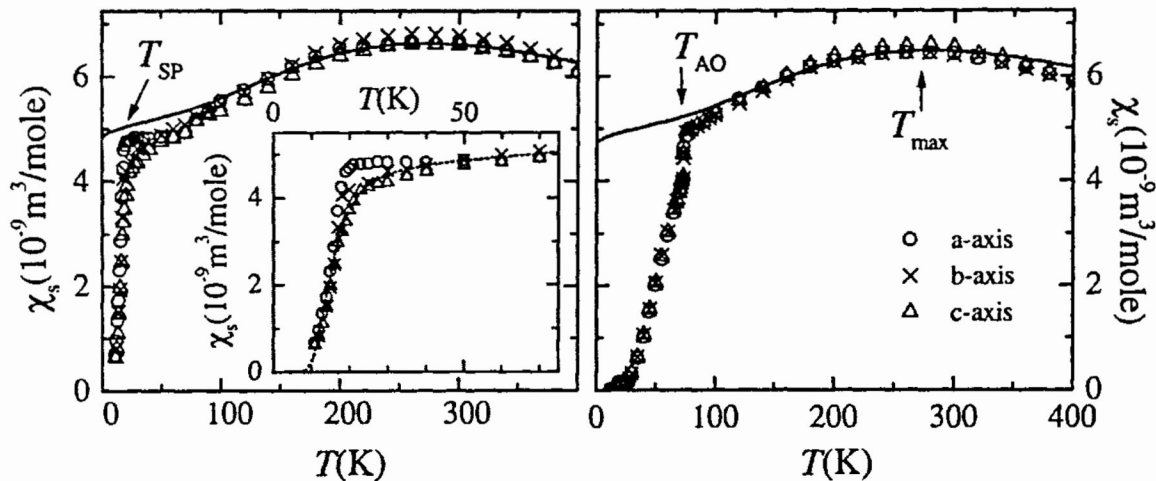


Fig. 1: Temperature dependence of the spin susceptibility χ_s of $(\text{TMTTF})_2\text{PF}_6$ (left) and $(\text{TMTTF})_2\text{ClO}_4$ (right) along the three crystal axes. The solid lines correspond to fits using the EAT-model for a $S=1/2$ AFM Heisenberg chain [4]. The absolute values of χ_s were obtained by scaling the integrated intensity at its maximum value using the relation $\chi_{\text{max}} = C/T_{\text{max}}$, $C = 1.782 \times 10^{-6} \text{ m}^3 \text{ K/mole}$. The inset indicates the fluctuations above the spin-Peierls transition in b and c direction. A good description of χ_s in the SP pseudo-gap regime is given by the dashed line [5].

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