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Despite two decades of research, the organic charge-transfer salts (TM-$TCF)_2X$ ($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 $(TMTCF)_2PF_6$, $(TMTCF)_2ClO_4$, and $(TMTTF)_2Br$ in the temperature range 1.5 K $\leq T \leq$ 500 K [2]. Here we confine ourselves to results of the spin-chain compounds $(TMTTF)_2ClO_4$ and $(TMTTF)_2PF_6$ at 2 K $\leq T \leq$ 400 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 K. The linewidth ΔH is slightly anisotropic and increases almost linear with temperature.

In $(TMTTF)_2ClO_4$ the ordering of the anions leads to a first-order structural phase transition at $T_{AO} = $ 72.5 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 K. At $T \approx$ 14 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.

$(TMTTF)_2PF_6$ undergoes a spin-Peierls transition at $T_{SP} = $ 19 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 K. The visible decrease of the spin susceptibility along the b and c direction below $T_{SP}^0 \approx$ 62 K can be attributed to 1-D
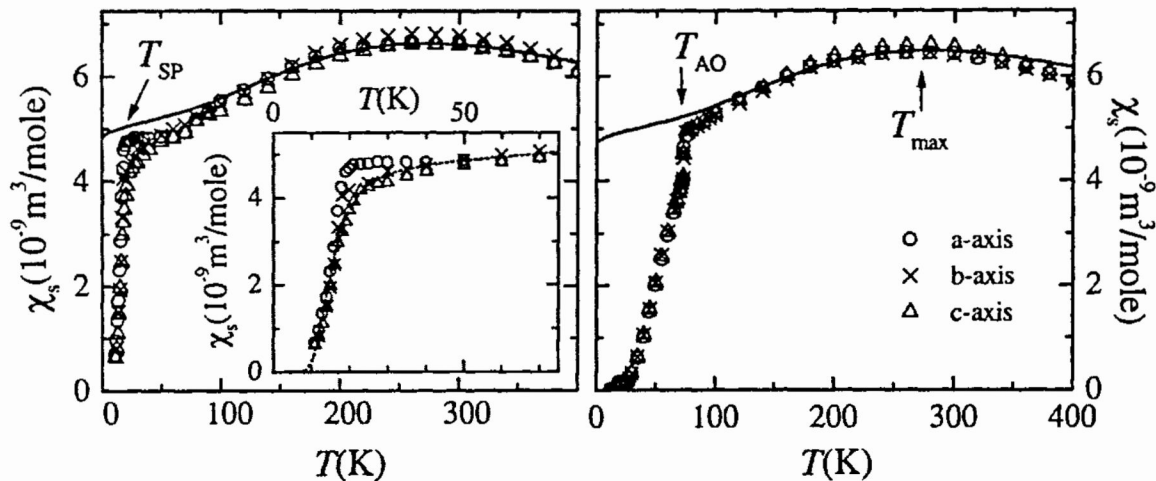
Fig. 1: Temperature dependence of the spin susceptibility χ_s of $(TMTTF)_2PF_6$ (left) and $(TMTTF)_2ClO_4$ (right) along the three crystal axes. The solid lines correspond to fits using the EAT-model for a $S = 1/2$ AFM Heisenberg chain [4]. The absolute values of χ_s were obtained by scaling the integrated intensity at its maximum value using the relation $\chi_{max} = C/T_{max}, C = 1.782 \times 10^{-6} m^3 K/mole$. 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].

lattice fluctuations above the spin-Peierls transition [5].

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