

# Charge and spin dynamics of TMTSF and TMTTF salts

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## Abstract

A comprehensive investigation of the charge and spin dynamics of the quasi one-dimensional organic compounds (TMTSF)<sub>2</sub>X and (TMTTF)<sub>2</sub>X with X = PF<sub>6</sub>, AsF<sub>6</sub>, Br, and ClO<sub>4</sub> has been performed in the temperature range from 2 K up to 500 K. By substitution of the organic donors or inorganic anions, the dimensionality of the systems can be tuned. While the resistivity strongly increases in (TMTTF)<sub>2</sub>X below 100 K, the susceptibility measured by electron spin resonance varies only slightly.

**Key words:** Transport measurements, Electron spin resonance, Organic conductors based on radical cation salts

In the well-known phase diagram which spans from (TMTTF)<sub>2</sub>PF<sub>6</sub> to (TMTSF)<sub>2</sub>ClO<sub>4</sub>, the interchain coupling of the organic systems increases and thus the dimensionality crosses over from a strictly one-dimensional to a more two or three-dimensional system. Besides ordered states like Mott insulator, spin Peierls, antiferromagnetic insulator, spin-density-wave (SDW), and superconductivity also the metallic state changes its behavior going from a Tomonaga-Luttinger liquid to a Fermi liquid. This can be nicely seen in electronic properties like the out-of-plane resistivity [1] or the optical conductivity [2]; the spin dynamics on the other hand does not show any fundamental change going from TMTTF to TMTSF except the spins are less localized [3].

As displayed in Fig. 1 the temperature dependence of the resistivity of (TMTTF)<sub>2</sub>AsF<sub>6</sub>, (TMTTF)<sub>2</sub>PF<sub>6</sub>, (TMTTF)<sub>2</sub>ClO<sub>4</sub>, (TMTTF)<sub>2</sub>Br, (TMTSF)<sub>2</sub>PF<sub>6</sub>, and (TMTSF)<sub>2</sub>ClO<sub>4</sub> was measured along the chain direction in the range 1 K < T < 400 K. Around 250 K we find a broad minimum in resistivity of (TMTTF)<sub>2</sub>PF<sub>6</sub> while in (TMTTF)<sub>2</sub>AsF<sub>6</sub> the behavior is basically temperature independent from 200 K up to 400 K. In both compounds we may identify an activated behavior  $\rho(T) \propto \exp\{\Delta_\rho/T\}$  with  $\Delta_\rho \approx 820$  K

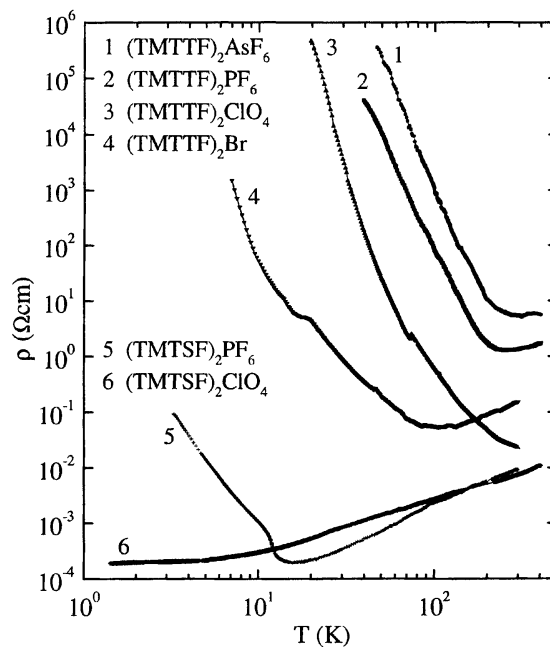


Fig. 1. Temperature dependence of the resistivity  $\rho(T)$  of different salts of the (TMTSF)<sub>2</sub>X and (TMTTF)<sub>2</sub>X family measured along the chain direction.

for 100 K < T < 200 K in accordance to earlier reports [4]. For (TMTTF)<sub>2</sub>ClO<sub>4</sub> we do not see a minimum in  $\rho(T)$  and also the activation energy is only about 510 K above the first order phase transition (due to anion ordering) at 74 K and about

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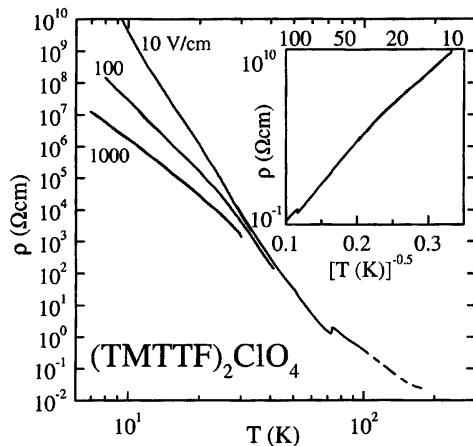


Fig. 2. Temperature dependence of the resistivity of  $(\text{TMTTF})_2\text{ClO}_4$ . Deviations for higher electric fields show the importance of non-linear effects at low temperatures. In the inset the data are plotted as a function of  $T^{-1/2}$ .

350 K below. However, as shown in Fig. 2 for the example of  $(\text{TMTTF})_2\text{ClO}_4$ , one-dimensional hopping transport with  $\rho \propto \exp\{T_0/T^{0.5}\}$  is also a possible explanation. At low temperature non-linear and heating effects make the evaluation of the transport mechanism difficult.  $(\text{TMTTF})_2\text{Br}$  stays metallic above 100 K, in the range down to 20 K we can identify an activation energy of  $\Delta_\rho \approx 100$  K. In the temperature range  $15 \text{ K} < T < 20 \text{ K}$  where the antiferromagnetic ordering occurs ( $T_N = 17.6 \text{ K}$ ),  $\rho(T)$  increases only slightly, but for  $T < 10 \text{ K}$  the same activation energy of 100 K is found. This agrees with the fact that the antiferromagnetic ordering is not a SDW transition with the opening of an energy gap at  $E_F$  but it is due to localized spins.  $(\text{TMTSF})_2\text{PF}_6$  and  $(\text{TMTSF})_2\text{ClO}_4$  show the well-known metallic behavior in the resistivity with a SDW transition of  $(\text{TMTSF})_2\text{PF}_6$  at 12 K.

At high temperatures the spin susceptibility at constant volume of the  $(\text{TMTTF})_2X$ -compounds (Fig. 3) can be described by a spin 1/2 antiferromagnetic Heisenberg chain with exchange constants  $J = 420 \text{ K}$  ( $X = \text{PF}_6$ ),  $J = 430 \text{ K}$  ( $X = \text{ClO}_4$ ), and  $J = 500 \text{ K}$  ( $X = \text{Br}$ ) using the common EAT model [5]. Even though  $(\text{TMTSF})_2\text{PF}_6$ ,  $(\text{TMTSF})_2\text{AsF}_6$  and  $(\text{TMTSF})_2\text{ClO}_4$  are one-dimensional organic metals down to low temperatures, for  $T > 100 \text{ K}$  the temperature dependence of  $\chi(T)$  can be described within the framework of the Hubbard model in the limit of strong Coulomb repulsion [6] with  $J \approx 1400 \text{ K}$  and  $t_a/U = 0.2$  [3].

Going from the fully insulating  $(\text{TMTTF})_2\text{PF}_6$  to the highly metallic  $(\text{TMTSF})_2\text{ClO}_4$  there is a sudden change in the charge-transport properties when the transfer integral becomes comparable to the charge gap, while the spin dynamics changes

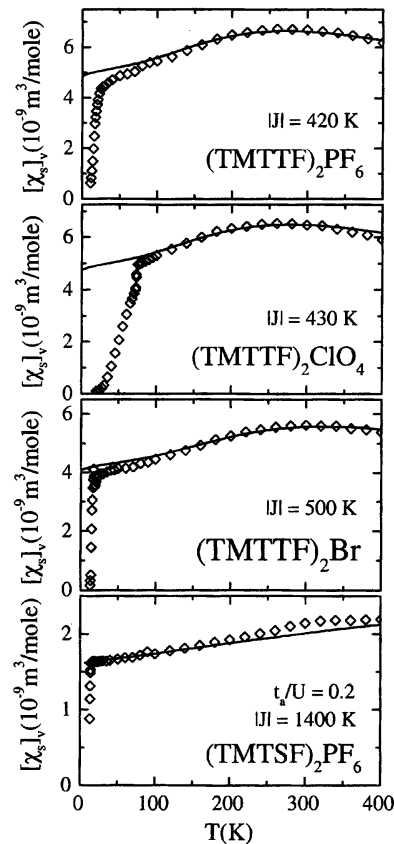


Fig. 3. Temperature dependence of the spin susceptibility  $(\chi_s)_v$  at constant volume of  $(\text{TMTSF})_2\text{PF}_6$ ,  $(\text{TMTSF})_2\text{ClO}_4$ ,  $(\text{TMTTF})_2\text{Br}$ , and  $(\text{TMTTF})_2\text{PF}_6$  as obtained by ESR intensity. The lines in (a) to (c) correspond to fits using the EAT-model for a  $S = 1/2$  AFM Heisenberg chain with  $J = 420 \text{ K}$ ,  $J = 430 \text{ K}$  and  $J = 500 \text{ K}$ , the line in the lowest frame (d) corresponds to a fit using the model of Seitz and Klein with  $t_a/U = 0.2$ .

continuously described by a steadily increasing exchange constant. The resistivity in  $(\text{TMTTF})_2X$  strongly increases below 100 K; in contrast the susceptibility and also the line width vary only slightly. This behavior indicates the separation of spin and charge degrees of freedom.

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