

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 $(\text{TMTSF})_2X$ and $(\text{TMTTF})_2X$ with $X = \text{PF}_6, \text{AsF}_6, \text{Br},$ and ClO_4 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 $(\text{TMTTF})_2X$ below 100 K, the susceptibility measured by electron spin resonance varies only slightly.

In the well-known phase diagram which spans from $(\text{TMTTF})_2\text{PF}_6$ to $(\text{TMTSF})_2\text{ClO}_4$, 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 $(\text{TMTTF})_2\text{AsF}_6$, $(\text{TMTTF})_2\text{PF}_6$, $(\text{TMTTF})_2\text{ClO}_4$, $(\text{TMTTF})_2\text{Br}$, $(\text{TMTSF})_2\text{PF}_6$, and $(\text{TMTSF})_2\text{ClO}_4$ was measured along the chain direction in the range $1 \text{ K} < T < 400 \text{ K}$. Around 250 K we find a broad minimum in resistivity of $(\text{TMTTF})_2\text{PF}_6$ while in $(\text{TMTTF})_2\text{AsF}_6$ 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 $\Delta_\rho \approx 820 \text{ K}$

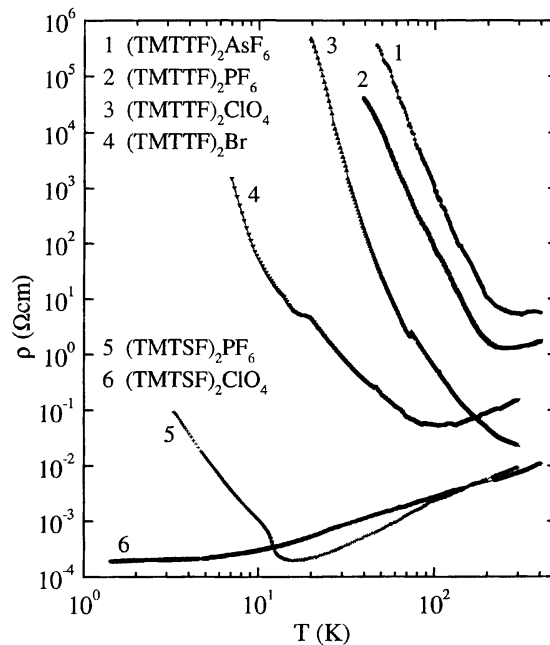


Fig. 1. Temperature dependence of the resistivity $\rho(T)$ of different salts of the $(\text{TMTSF})_2X$ and $(\text{TMTTF})_2X$ family measured along the chain direction.

for $100 \text{ K} < T < 200 \text{ K}$ in accordance to earlier reports [4]. For $(\text{TMTTF})_2\text{ClO}_4$ 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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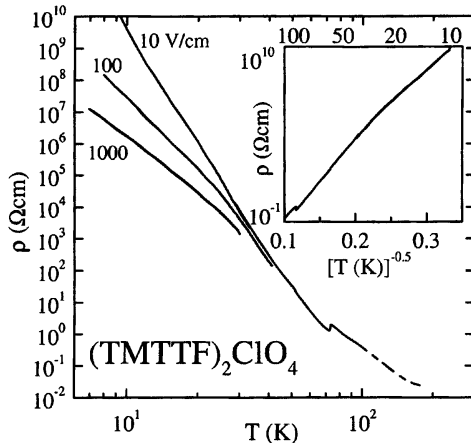


Fig. 2. Temperature dependence of the resistivity of $(\text{TMTTF})_2\text{ClO}_4$. Deviations for higher electric fields show the importance of non-linear effects at low temperatures. In the inset the data are plotted as a function of $T^{-1/2}$.

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

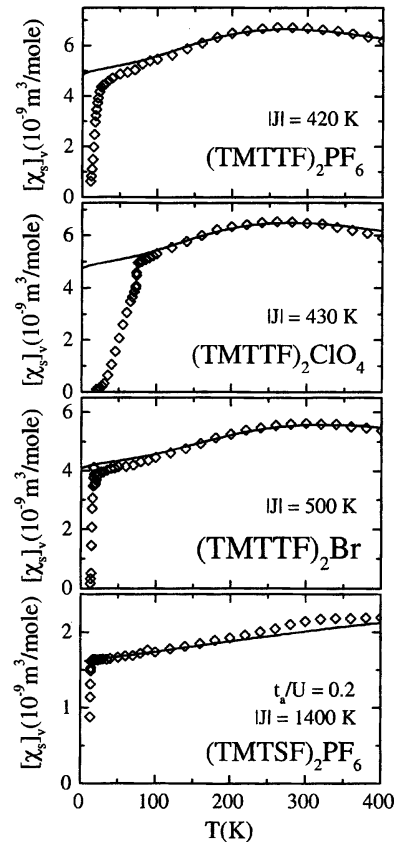


Fig. 3. Temperature dependence of the spin susceptibility $(\chi_s)_v$ at constant volume of $(\text{TMTSF})_2\text{PF}_6$, $(\text{TMTSF})_2\text{ClO}_4$, $(\text{TMTTF})_2\text{Br}$, and $(\text{TMTTF})_2\text{PF}_6$ as obtained by ESR intensity. The lines in (a) to (c) correspond to fits using the 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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