Charge and spin dynamics of TMTSF and TMTTF salts

Martin Dressel, P. Hesse, S. Kirchner, G. Untereiner, Michael Dumm, Joachim Hemberger, Alois Loidl, L. Montgomery

Charge and spin dynamics of TMTSF and TMTTF salts

M. Dressel [a,1], P. Hesse [a], S. Kirchner [a], G. Untereiner [a], M. Dumm [b], J. Hemberger [b], A. Loidl [b], L. Montgomery [c]

[a] 1. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, D-70550 Stuttgart, Germany
[b] Experimentalphysik V, Universität Augsburg, D-86135 Augsburg, Germany
[c] Department of Chemistry, Indiana University, Bloomington, IN 47405, U.S.A.

Abstract

A comprehensive investigation of the charge and spin dynamics of the quasi one-dimensional organic compounds $(TMTSF)_2X$ and $(TMTTF)_2X$ with $X=$ PF_6, AsF_6, 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 resisitivity strongly increases in $(TMTTF)_2X$ below 100 K, the susceptibility measured by electron spin resonance varies only slightly.

In the well-known phase diagram which spans from $(TMTTF)_2PF_6$ to $(TMTSF)_2ClO_4$, 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 resistivty of $(TMTTF)_2AsF_6$, $(TMTTF)_2PF_6$, $(TMTTF)_2ClO_4$, $(TMTTF)_2Br$, $(TMTSF)_2PF_6$, and $(TMTSF)_2ClO_4$ 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 $(TMTTF)_2PF_6$ while in $(TMTTF)_2AsF_6$ 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 $\Delta_\rho \approx 820$ K
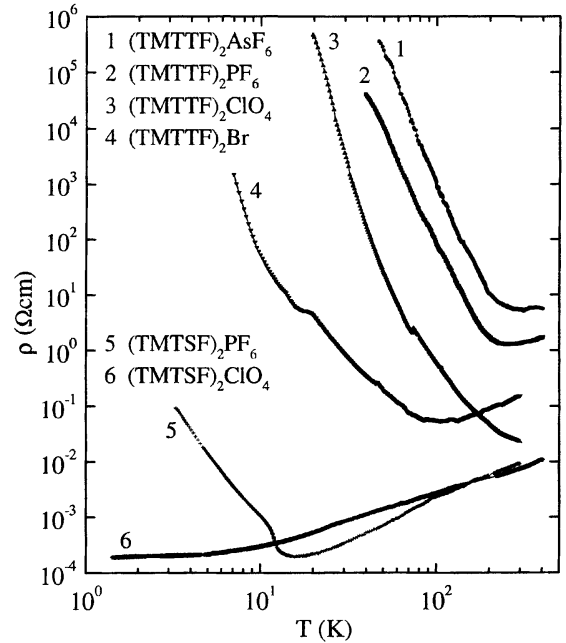
Fig. 1. Temperature dependence of the resistivity $\rho(T)$ of different salts of the $(TMTSF)_2X$ and $(TMTTF)_2X$ family measured along the chain direction.

for $100 K < T < 200$ K in accordance to earlier reports [4]. For $(TMTTF)_2ClO_4$ 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
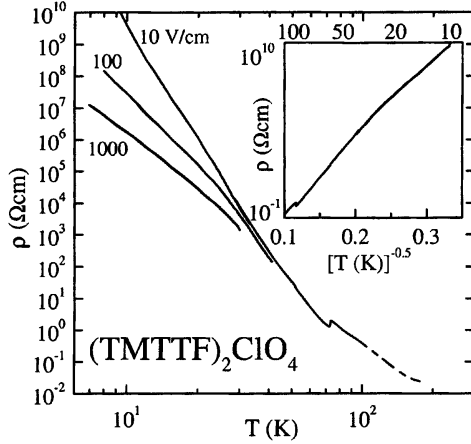
Fig. 2. Temperature dependence of the resistivity of $(TMTTF)_2ClO_4$. Deviations for higher electric fields show the importance of non-linear effects at low temperatures. In the inset the data are plotted as a function of $T^{-1/2}$.

350 K below. However, as shown in Fig. 2 for the example of $(TMTTF)_2ClO_4$, 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. $(TMTTF)_2Br$ stays metallic above 100 K, in the range down to 20 K we can identify an activiation energy of $\Delta_\rho \approx 100$ K. In the temperature range 15 K $<$ $T < 20$ K where the antiferromagnetic ordering occurs ($T_N = 17.6$ K), $\rho(T)$ increases only slightly, but for $T < 10$ 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. $(TMTSF)_2PF_6$ and $(TMTSF)_2ClO_4$ show the well-known metallic behavior in the resistivity with a SDW transition of $(TMTSF)_2PF_6$ at 12 K.

At high temperatures the spin susceptibility at constant volume of the $(TMTTF)_2X$-compounds (Fig. 3) can be described by a spin 1/2 antiferromagnetic Heisenberg chain with exchange constants $J = 420$ K ($X = PF_6$), $J = 430$ K ($X = ClO_4$), and $J = 500$ K ($X = Br$) using the common EAT model [5]. Even though $(TMTSF)_2PF_6$, $(TMTSF)_2AsF_6$ and $(TMTSF)_2ClO_4$ are one-dimensional organic metals down to low temperatures, for $T > 100$ 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 $J \approx 1400$ K and $t_a/U = 0.2$ [3].

Going from the fully insulating $(TMTTF)_2PF_6$ to the highly metallic $(TMTSF)_2ClO_4$ 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
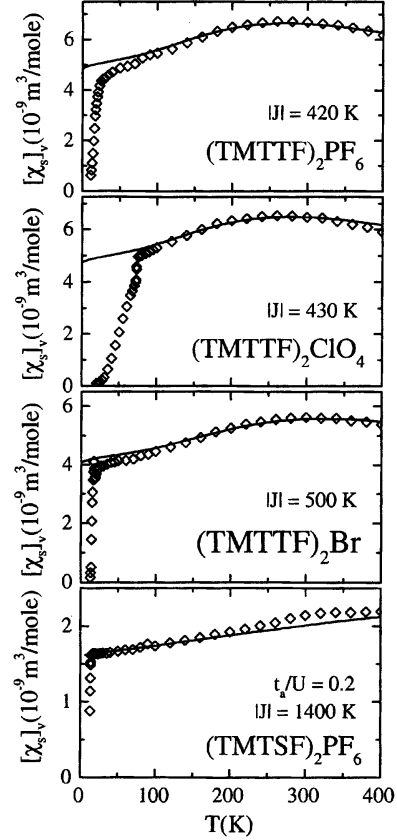

Fig. 3. Temperature dependence of the spin susceptibility $(\chi_s)_v$ at constant volume of $(TMTSF)_2PF_6$, $(TMTSF)_2ClO_4$, $(TMTTF)_2Br$, and $(TMTTF)_2PF_6$ as obtained by ESR intensity. The lines in (a) to (c) correspond to fits using the EAT-model for a $S = 1/2$ AFM Heisenberg chain with $J = 420$ K, $J = 430$ K and $J = 500$ 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 $(TMTTF)_2X$ 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.

References

[1] J. Moser, M. Gabay, P. Auban-Senzier, D.Jérome, K. Bechgaard, J.M. Fabre, Eur. Phys. J. B 1 (1998) 39.

[2] M. Dressel, A. Schwartz, G. Grüner, L. Degiorgi, Phys. Rev. Lett. 77 (1996) 398; V. Vescoli, L. Degiorgi, W. Henderson, G. Grüner, K.P. Starkey, L.K. Montgomery, Science 281 (1998) 1181.

[3] M. Dumm, A. Loidl, B.W. Fravel, K.P Starkey, M. Montgomery, M. Dressel, Phys. Rev. B 61 (2000) 511; Phys. Rev. B 62 (2000).

[4] C. Coulon, P. Delhaes, S. Flandrois, R.Lagnier, E. Bonjour, J.M. Fabre, J. Phys. (France) 43 (1982) 1059.

[5] S. Eggert, I. Affleck, M. Takahashi, Phys. Rev. Lett. 73 (1994) 332.

[6] W.A. Seitz, D.J. Klein, Phys. Rev. B 9 (1974) 2159.