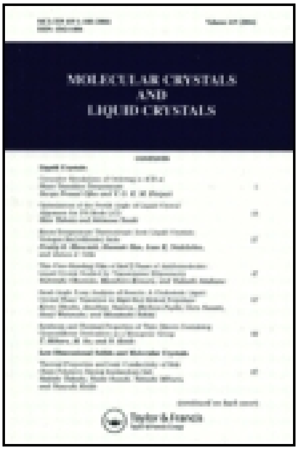

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Nonlinear Transport Properties of $(TMTSF)_2$ X Series

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NONLINEAR TRANSPORT PROPERTIES OF $(TMTSF)_2X$ SERIES

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Abstract We discuss the thermal and electrical conductivities of samples $(TMTSF)_2X$ ($X=PF_6, AsF_6, ClO_4$) in temperature range 4.2 K$<T<\theta$.

INTRODUCTION

The method we use is described in separate contribution of these Proceedings. Additional measurements of thermal conductivity of $(TMTSF)_2ClO_4$ based on the experience with six probe technique, give qualitatively the same result as the application of four and two probes reported previously.

The temperature dependence of thermal conductivity κ of low dimensional conductor $(TMTSF)_2PF_6$ is displayed on Fig.(1). An usual increase of κ with decreasing temperature starts at $\sim$100 K and exhibits the tendency to obey the well known Peierls divergence $\exp(\theta/T)$ at temperatures well below the Debye temperature θ=270 K. At $\sim$ 50 K the divergent increase is stopped and T-dependence is more flattened if the temperature decreases. Thermal conductivity anomaly due to antiferromagnetic transition appears at 14 K while electrical resistivity accompained with this transition starts to increase at $\sim$17 K. The collective magnon modes below the metal-insulator transition are discussed in separate paper of this Proceedings. The magnon part of thermal current in $(TMTSF)_2AsF_6$ is not so good contrasted as in $(TMTSF)_2PF_6$. It seems that the Peierls divergence is well developed and it is tedious job to separate magnon contribution to heat current from the contribution of the lattice.

Contrary to $(TMTSF)_2PF_6$ and $(TMTSF)_2AsF_6$ thermal conductivity of

$(TMTSF)_2ClO_4$ continously decreases with decreasing temperature. At ∿60 K there is the change of the regime in temperature dependence of κ (see Fig.2). The continous decrease is not unique below 25K where the influence of the cooling regime of the sample must be seriously respected (Fig.3).

The fast cooling regime from 40 K with cooling rate of ∿20 K/min is accompained by the rapid decrease of κ at temperatures below 25K when thermal conductivity is suppressed by the anion disorder being frozen down to the lowest temperatures. The medium cooling rate (∿1 K/min) gives rise to the formation of relaxed state and flattening of κ-dependence. This is probably due to an additional heat current locked by anion disorder at temperatures above 25 K. In the third experiment the specimen is subjected to extremely slow cooling rate (∿1 K/hour). The temperature dependent electrical resistance measured on the same sample in the same experimental set-up exhibits no discontinuities commonly attributed to the formation of microcracks within the sample and thermal conductivity turns out to be more flattened in the temperature range 25-12K. This temperature change correlates the temperature dependence of NMR relaxation rate giving rise to the possibility of collective excitations in highly correlated electron gas in the relaxed state of $(TMTSF)_2ClO_4$ below 25 K[1].

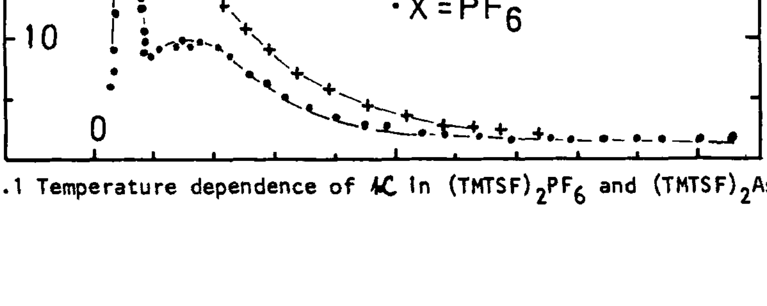

Fig.1 Temperature dependence of κ in $(TMTSF)_2PF_6$ and $(TMTSF)_2AsF_6$

DISCUSSION

In the systems of lower dimensionality there is possible the persistance of magnonlike modes at temperatures far above the transition temperature to magnetic state. In the interacting Fermi gas there exist excitations analogous to spin waves in magnetic materials and these oscillations are collective periodic variations of the macroscopic magnetization vector. Such a modes have already been observed in some 1D antiferromagnets [2].

Since it is well known that truly 1D system cannot exhibit a long-range order we expect a rapid decay of correlation relaxation rates with increasing temperature in $(TMTSF)_2X$. A strong increase of ESR linewidths with increasing temperature in $(TMTSF)_2PF_6$ [3] and in $(TMTSF)_2ClO_4$ [4] supports this conclusion. In fact the low dimensional materials $(TMTSF)_2X$ exhibit the tendency of decreasing dimensionality with increasing temperature and according to the calculation of Fisher and Bonner [5] it should be expected that the paramagnon correlation amplitude will increase, as a result of the reduced dimensionality.Otherwise the fluctuating magnetization δM correlation function related to static susceptibility χ

$$\langle \delta M(q)\, \delta M(-q) \rangle = k_B T V \chi / g^2 \mu_B^2 \qquad 1)$$

turns out to be the increasing function of the temperature.

An increase of the correlation amplitude is compensated by the dissipation of the spin fluctuations in phonon system. In highly correlated electron gas the phonons are scattered by paramagnon excitations and we believe that the lack of the full Peierls divergence in $(TMTSF)_2PF_6$ below 50 K is due to the scattering of phonons by paramagnons. This assumption is supported by the fact that at this temperature transverse spin relaxation rate T_2^{-1} changes regime [3] and the anisotropy of electrical conductivity decreases below 40-50 K . The relaxation of short ranged correlations of fluctuating spin system is controlled by the release of energy to the lattice via the spin diffusion currents. The Fermi liquid may be described for the slow variations of variables in the phonon system by the quasi-particle density matrix $\rho_{\alpha\alpha'}(p,X)$[6]

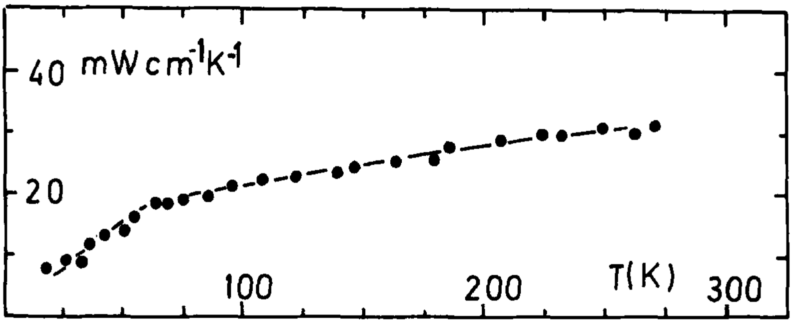

Fig.2 Thermal conductivity of $(TMTSF)_2ClO_4$

$$\rho_{\alpha\alpha'}(p,X) = n_0\delta_{\alpha\alpha'} + \vec{M}\vec{\sigma}_{\alpha\alpha'} \qquad 2)$$

In equilibrium the quasi-particle energy is simply $E_0(p) = p^2/2m^x$, n_0 is Fermi equilibrium distribution. In nonequilibrium the quasi-particle density matrix may be found by the solution of ordinary Boltzmann equation. In the absence of external fields a simple relation holds

$$\partial g/\partial t + (\vec{v}\nabla)(g + \delta\epsilon) = -\frac{1}{\tau}g \qquad M^{+} = M_x + iM_y = -\frac{\partial n_0}{\partial E(p)}g$$

The change of the quasi-particle energy due to the collisions with phonons is

$$\delta\epsilon = \frac{2}{2\pi^2}\int d^3p\langle\delta M(p)\,\delta M(-p)\rangle g(p)\,\delta[\mu - E^0(p)] \qquad 4)$$

The correlation function $\langle \ldots \rangle$ must be considered in some details.

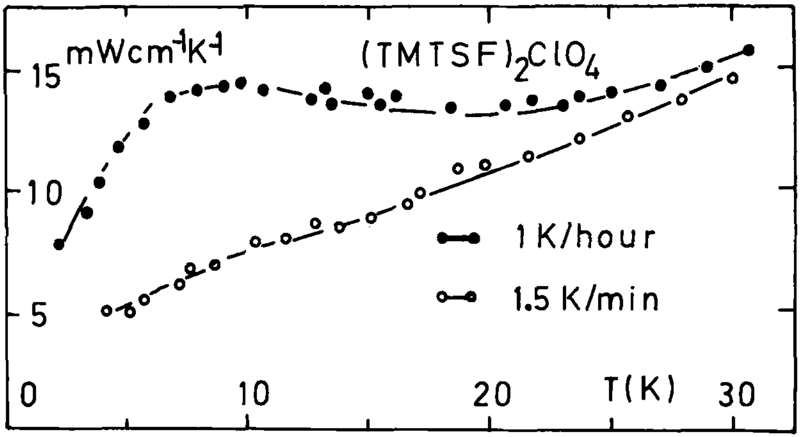

Fig.3. Thermal conductivity dependent upon cooling rate

The energy transferred to the phonon gas is mainly controlled by the heat capacity of the lattice and the rate of the transfer measured by relaxation time τ at temperatures $T_{M-I} < T \ll \theta$ should be proportional to the specific heat of the lattice i.e.

$\frac{1}{\tau} \sim C_p \sim T^3.$

This temperature dependence of the linewidth was actually been found in $(TMTSF)_2PF_6$ in the temperature range between 20 K and 40-50 K [3]. At temperatures above $\sim$50 K the paramagnon collective mode in $(TMTSF)_2PF_6$ is suppressed and the properties of the system are more governed by the pure electron-electron interaction.

In $(TMTSF)_2ClO_4$ the complet dissapearance of the collective periodic variations of macroscopic magnetization may be expected at about 60 K temperature at which the change of the regime in thermal conductivity is observed.In addition, in far infrared scattering the structure observed to develop below 60 K may be attributed to appearance of such a mode 7.

The second point related to the problem of spin fluctuations in $(TMTSF)_2PF_6$ is the lack of the correlation of electrical resistance and ESR linewidth (LW) if temperature is rised from 20 K to 40 K [3]. The electrical resistance obeys T^2 dependence and it is not excluded the possibility of dominance of the electrical resistance by the interaction of paramagnons and electrons. Such an interaction is reviewed in the work of Izuyama and Kubo [8]. The paramagnon created in itinerant magnetic system may dominate the electron scattering and the current is

$$j \sim e \langle \nabla_k \epsilon_{k+q} - \nabla \epsilon_k \rangle \qquad 5)$$

while paramagnon-electron interaction energy is of the form

$$\epsilon_{pM-e} \sim f(k) T^2 \qquad 6)$$

The T^2 dependence of electrical resistivity in $(TMTSF)_2PF_6$ is extended far above 50 K where we believe the paramagnon collective motion sets in and may be attributed to the more classical effects of electron-electron scattering. In fact Monecke [9] concluded that there is no contribution of such a scattering to electrical resistance if the translational invariance of wave functions is conserved. Impurities and phonons can destroy such an invariance for the electron subsystem giving rise to the nonvanishing contribution of resistance coming from electron-electron interaction.

The main source of nonlinearity in $(TMTSF)_2ClO_4$ in relaxed state results from the fact that the NMR relaxation rate T_1^{-1} does not obey the well known Korringa law i.e. is not linearly dependent on temperature.

According to Moriya calculation [10] the relaxation rate may generally be expressed

$$T_1^{-1} \sim T \sum_q \langle \delta M^+(q) \delta M^-(-q) \rangle / D_q \qquad 7)$$

The exponential time decay of correlation function in nonlinear regime below 25 K is governed by the relaxation rate D_q being dependent on correlation function itself.

At high temperatures ($T\sim 24$ K) D_q is proportional to the correlation function[11] since the anion disorder smears the electron spin fluctuations and the spin diffusion currents are locked. The Korringa law is than well obeyed. At temperatures 10 K$<T<$24 K spin susceptibility of collective state saturates keeping correlation function nearly constant in amplitude but relaxation time of correlations is longer with tendency to doverge at T = 0. At temperatures below 10 K number of paramagnons decreases with decreasing temperature and T_1^{-1}, as well of thermal conductivity, decrease again. More detailed calculations of collective properties are in course [12].

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