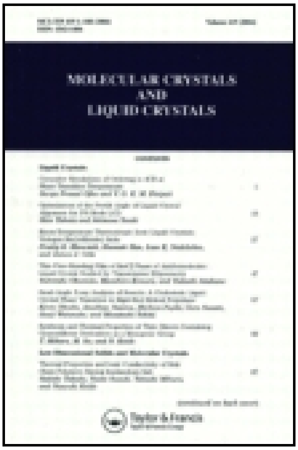

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Magnon Heat Transport in $(TMTSF)_2X(X=ClO_4, PF_6)$

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MAGNON HEAT TRANSPORT IN $(TMTSF)_2X(X=ClO_4,PF_6)$

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Abstract - We have observed a huge contribution of magnons to the heat current in $(TMTSF)_2X$ $(X=ClO_4,PF_6)$ below the antiferromagnetic transition.The results are discussed in connection with other physical properties related to these compounds.

INTRODUCTION

We report the results of an investigation of thermal transport in magnetic phases of $(TMTSF)_2PF_6$ and quenched $(TMTSF)_2ClO_4$. The method of measurements is described in additional contribution of these Proceedings and is based upon the properties of thermal wave propagated along the specimen. The typical dimensions of the samples are 4X 0.1X 0.3 mm^3. In four probe measurements of thermal conductivity (κ) we use the chromel-constantan thermocuple made of wire 12 microns in diameter. The typical distance between the thermocouple contacts is $\sim$ 3 mm. $(TMTSF)_2ClO_4$ is slowly cooled from room temperature to $\sim$ 40 K (more than 30 hours) in order to minimize the difficulties related to an appearance of microcracks. The sample was than quenched down to $\sim$4 K with the cooling rate of $\sim$20 K/min. The sample has been reheated in several cycles and deviations of data between two subsequent heatings orcoolings were within the experimental accuracy ($\sim$5 percents). The data for κ $(TMTSF)_2ClO_4$ are displayed on Fig.(1). Thermal conductivity decreases continously with decreasing temperature from 24 K until $\sim$4 K is reached. Continous decrease is again established below 2.9 K. It is evident that thermal current below 4 K and 15 K in quenched

$(TMTSF)_2ClO_4$ and $(TMTSF)_2PF_6$ respectively is not correlated to the electrical conductivity. The anomaly in κ may not therefore be attributed to the results of purely electronic effects. We believe that additional heat transport is due to magnons excited from the antiferromagnetically ordered ground state. There are additional arguments relevant for this possibility. The phonon part of thermal current in $(TMTSF)_2PF_6$ at temperatures above 15 K is remarkably separated from a huge contribution of additional excitations below the antiferromagnetic transition readily seen on Fig.(2).The lattice thermal current gradually decreases below 20 K as a result of decreasing number of phonons. In addition the bump in κ for $(TMTSF)_2ClO_4$ perfectly correlates the temperature dependent NMR relaxation rate T_1^{-1} measured by T. Takahashi and coworkers[1]. The temperature dependence of both physical quantities is shown on Fig.(3). Finally the observed structures in far infrared spectra[2] in $(TMTSF)_2PF_6$ at low wave number may be attributed to collective excitations in magnon gas.

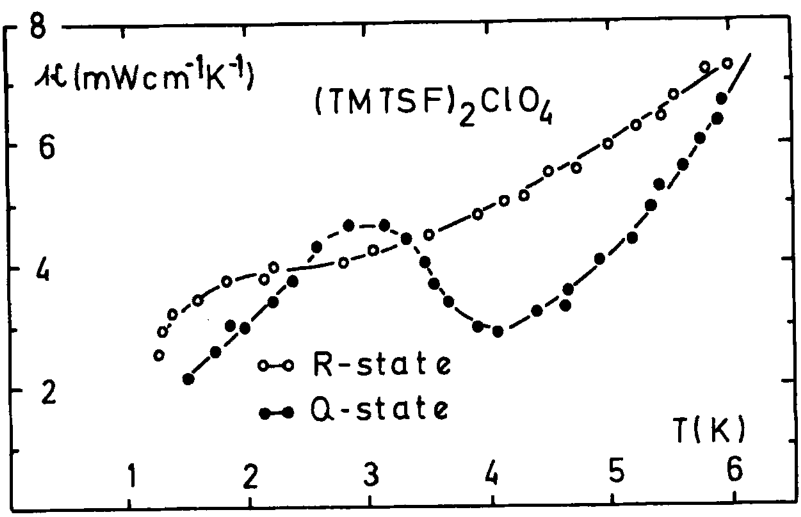

Fig.1. Thermal conductivity of $(TMTSF)_2ClO_4$

DISCUSSION

In further discussion we shall turn our attention to the calculation of the heat current in $(TMTSF)_2ClO_4$ and $(TMTSF)_2PF_6$ due to magnons. The fair correlation of T_1^{-1} and temperature dependent thermal conductivity suggests the use of the transport equations based on the linear response introduced by R.Kubo[3]. According to this theory the transport coefficients L_{ij} in the general linear relations between currents J_i and forces X_j

$$J_i = \sum L_{ij} \cdot X_j \qquad i,j = 1,2 \qquad 1)$$

are related to the correlation functions formed of currents of the conserved operators

$$L_{ij} = \int_0^{\infty} dt \int_0^{\infty} \langle I_i(t + i\lambda) I_j(0) \rangle d\lambda \qquad 2)$$

I_i and I_j are the quantum mechanical many-particle operators associated with macroscopic currents J_i and J_j. The coefficient L_{22} corresponds to thermal current $J = L_{22}\,(-\nabla T/T)$.

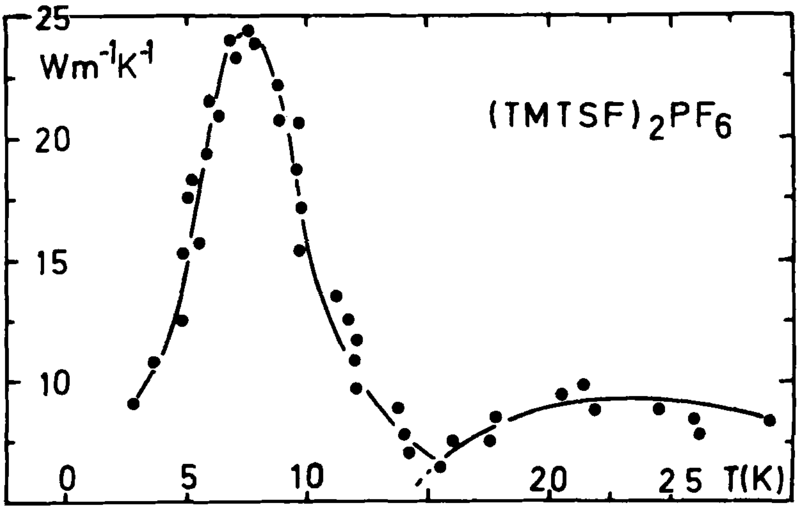

Fig.2. Thermal conductivity of $(TMTSF)_2PF_6$

∇T is the temperature gradient. The average $\langle \ldots \rangle$ means

$$\langle Q \rangle = \mathrm{Tr}\left(\frac{e^{-\beta H} Q}{Z}\right) \qquad \beta = 1/kT \qquad 3)$$

The time dependence of operators I is introduced in usual way

$$I(t) = \exp(iHt)\, I(0) \exp(-iHt) \qquad 4)$$

The standard formula for the thermal conductivity of interacting particles considers the volume heat capacity C_v, sound velocity v and the relaxation time τ.

$$\kappa = 1/3\; C_v \cdot v^2 \cdot \tau \qquad 5)$$

In fact this expression is the special case of the Eq.(2). Since the integral over currents $I(t) = n\hbar\omega_\perp v(t)$ averaged in the time interval τ between two collisions contributes to $v^2\tau$. Kubo arguments may be extended[4] to the calculation of relaxation rates in NMR experiments giving the general expression

$$T_1^{-1} = A^2/2h^2 \int dt\, \cos\omega_0 t \langle\{\delta M_q(t)\, \delta M_{-q}(0)\}\rangle \qquad 6)$$

A is hyperfine coupling constant (Fermi contact) and δM_q is Fourier component of fluctuating magnetization $\delta M = M - \langle M \rangle$. $\langle \ldots \rangle$ means symetrized product

$$\{A,B\} = 1/2\ (AB + BA) \qquad 7)$$

ω_0 is NMR frequency.

The thermal conductivity, implicitly introduced in Eq.(2), turns out to be expressed in terms of NMR relaxation rate T_1^{-1}. The spin diffusion current $J_M(r,t)$ must be replaced by corresponding magnetization fluctuation δM_q. The total spin is assumed to be a constant of motion i.e. the conserved quantity, and it must be satisfied the continuity equation for the magnetization

$$(d/dt)\left[\delta M(r,t)\right] + \mathrm{div}\, j_M(r,t) = 0 \qquad 8)$$

In terms of quantum mechanical operators we consider the connection between the magnetization and magnetization current

$$M(r,t) = \sum_i s_i(t)\delta[\vec{r} - \vec{r}_i(t)] \qquad 9)$$

$$\vec{j}_M(r,t) = \sum_i s_i(t) \{\vec{p}_i(t)/2m\delta[\vec{r} - \vec{r}_i(t)]\} \qquad 10)$$

s_i is the spin density. Finally, by the use of 9) and 10) currents in 2) may be replaced by the fluctuating magnetization and thermal conductivity is readily obtained

$$\mathcal{K} = (1/4kT^2)\, v_M^2 \int_0^\infty \langle\{\delta M_q(t), \delta M_{-q}(0)\}\rangle\, dt \qquad 11)$$

v_M is magnon sound velocity connected with temperature[5] $v_M^2 \sim kT$. The NMR relaxation rate is than

$$T_1^{-1} \sim T\mathcal{K}(T)$$

The plot of $T_1^{-1}T$ is given on Fig.(3) together with thermal conductivity data displayed after the subtraction of background. The thermal conductivity dependent upon the temperature reduces to a complicated calculation of correlation function (11). We shall pay an attention to the result of planar rotator model defined by the Hamiltonian

$$H = -\sum_{ij} S_i S_j - \sum_{ij} J_{ij} \cos(\varphi_i - \varphi_j) \qquad 12)$$

Wegner[6] and Berezinsky[7] used a harmonic approximation and replaced $\cos\varphi$ by $1 - 1/2\varphi^2$. Their result is summarized in temperature dependence of the correlation function S_iS_j

$$\langle S_iS_j\rangle \sim (R_i - R_j)^{-\frac{T}{4\pi J}} \qquad 13)$$

$(R_i - R_j)$ is the first-neighbour distance. In order to obtain the temperature dependence of $\mathcal{K}$ we have to multiply this correlation function by the number of magnons exhibiting the temperature dependence of the form T^n.

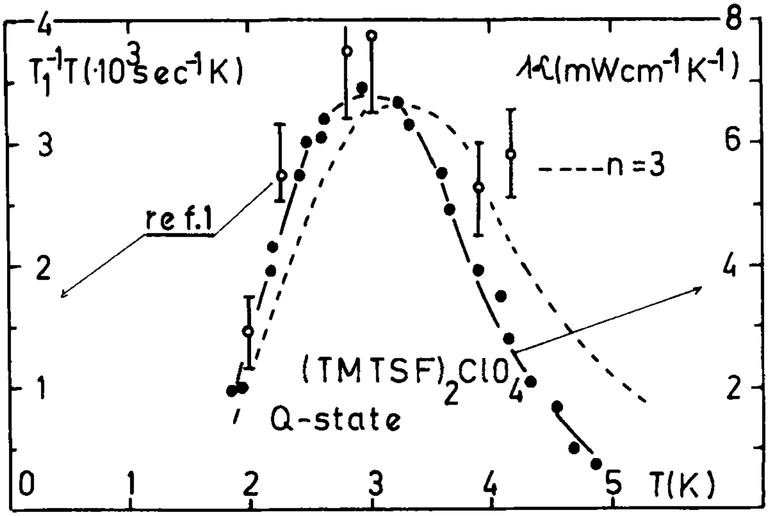

Fig.3. Magnon contribution to $\mathcal{K}$ and T_1^{-1}

Than

$$\mathcal{K}(T) \sim T^{n-1}(r_i - R_j)^{-\frac{T}{4\pi J}} \qquad 14)$$

This function is fitted to temperature dependent $\mathcal{K}$ in $(TMTSF)_2ClO_4$ and the result is shown on Fig.(3).

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