High precise thermopower measurement system and its applying to organic conductors $(TMTSF)_2ClO_4$ in a direction

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We present a carefully designed apparatus for high precise measurement of the thermopower. The resolution of the system can reach to $0.01 \mu V/K$. The thermopower of organic conductors $(TMTSF)_2ClO_4$ single crystals were measured. A linear behavior at high temperature and a deviation at 140 K attributed to 1D—2D crossover were observed. Different cooling rates are realized to study the anion order-disorder transition in thermopower at $T_{ao}=24K$. There is an obvious upturn just below T_{ao} when the cooling rate reaches 0.3 K/s. A clear trace of SDW transition at 4.5 K can be spotted by the fastest cooling process.

INTRODUCTION

The thermoelectric power S (thermopower or TEP) is the voltage generated across two points on a material divided by the temperature difference between the two points [1]. Besides being an important transport property, the thermoelectric power is very sensitive to the sign of charge, the density of states at Fermi level, composition, structure, pressure and external fields. It is essential for understanding the physics of materials both theoretically and experimentally. However, the measurement of thermopower on the $(TMTCF)_2X$ (C=Se for TMTSF or S for TMTTF, and X= PF_6, AsF_6, ReO_4, ClO_4, Br etc)family of organic quasi-one-dimensional (1D) conductors is very hard to perform, due to their needle-like shapes and fragile body condition.

Due to the strong anisotropy and subtle balance between interchain and intrachain electronic coupling, these quasi-1D conductors exhibit a variety of electronic ground states, including SDWs, superconductivity, magnetic-field-induced spin-density waves (FISDWs), anion order-disorder transition and so on [2]. In the case of $(TMTSF)_2ClO_4$, which exists the anion order-disorder transition (T_{ao}= 24 K), if quenching above T_{ao}, its ground state is SDW state (T_{SDW}=6 K). In relaxed states (very slow cooling procedure), its ground state is metallic/superconductivity state ($T_C \approx 1$ K) [3].

The object of this paper is to present a carefully designed apparatus for the high precise studying of the thermopower in the temperature range from 4-300 K. The thermopower data of $(TMTSF)_2ClO_4$, at different cooling rates through T_{ao}, are reported.

DESIGN OF THE THERMOPOWER MEASUREMENT SYSTEM

The schematic diagram of the computer-interfaced system is shown in Figure 1. A sample is connected between two copper blocks. A thermal gradient across the sample is applied by heating the two blocks to different temperatures. Thermometers were used to monitor the temperatures of the blocks. In order to

control the temperature of either of the copper blocks, first, by using 7150 Digital Multimeters, computer
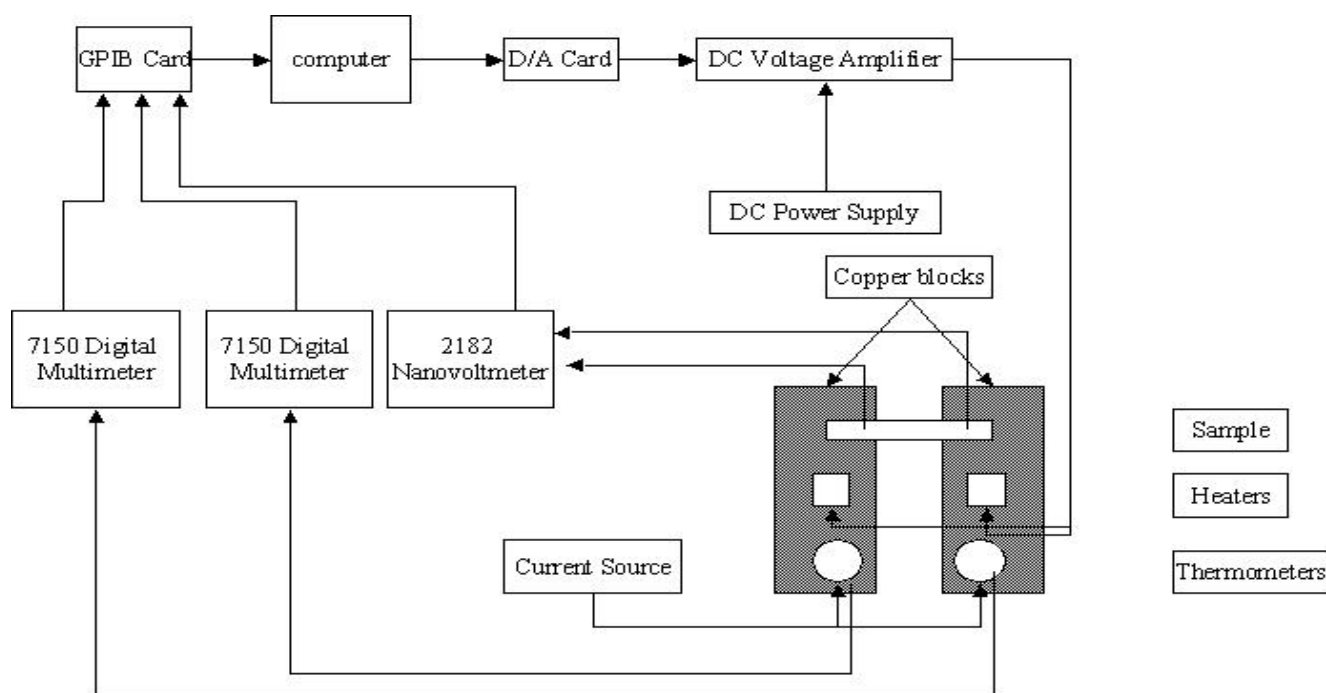

Figure 1   The schematic diagram of the computer-interfaced measuring system

can read the voltages from GPIB port and calculate their temperatures, then, the computer calculate the heating power through PID arithmetic and control the D/A card to output certain heating current to the heaters. After reaching thermal equilibrium, ΔT can be calculated or directly measured between the blocks. The electric potential difference ΔV is measured by the Keithley 2182 nanovoltmeter with contact leads on the sample. In general, the thermopower is:

$$S=\Delta V/\Delta T$$

The thermoelectric effect of the Cu lead wires was calibrated using a type of metal as a reference. In fact, each thermopower data point is obtained by averaging over 100 ΔV and ΔT points to depress the electric random error, and then by inversing temperature gradient to eliminate contacting thermopower.

The high vacuum and two thermal shields were performed in suppressing the temperature fluctuation of sample and increasing signal-to-noise performance. The temperature of either of copper blocks, which was controlled by computer, can be stabilized to less than 0.003 K or even less depending on the time. A temperature gradient $\Delta T/T\sim0.6$ % across the samples was carefully controlled between   6 K and 300 K. Every leads coming from the computer were added filters to screen high frequent electric noise. So the resolution of thermopower is proven to be $0.01\mu V/K$ at best condition. When combined with a soft-supporting platform, our procedure leads to effectiveness and accuracy for the thermopower of small samples, with direct application to organic conductor $(TMTSF)_2ClO_4$.

EXPERIMENT

To demonstrate the techniques described here, we consider the thermopower of organic conductor $(TMTSF)_2ClO_4$ single crystal. The typical size of the sample is about $3\times0.1\times0.05$ mm^3. The sample is mounted along a direction. A small soft-supporting copper block, as shown in Figure 2, is used to prevent the needle like single crystal from cracking in a fast cooling rate. Even for the fastest cooling rate in our experiment of 1K/s, the data around the anion order-disorder transition (~24 K) of $(TMTSF)_2ClO_4$ is

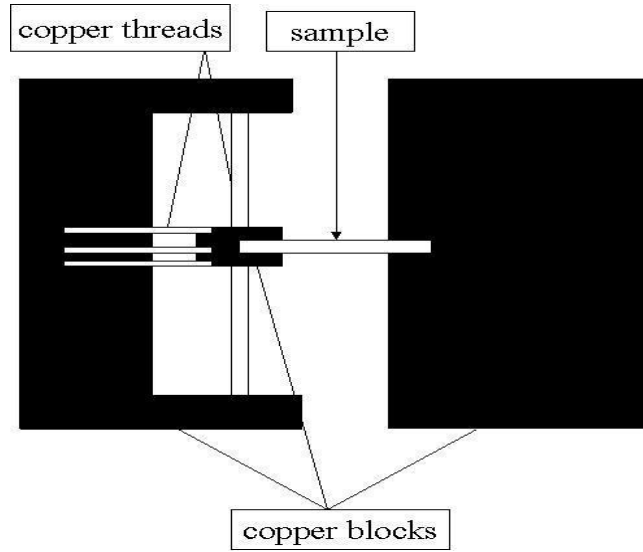credible without cracks.

Figure 2   Soft-supporting platform

The small soft-supporting platform mounted over two parallel copper threads to support one end of the sample softly. 30 threads were attached between both a big copper block and the end of the small platform so much as to maintain a same temperature between the big and small blocks.

Figure 3 is temperature dependent thermopower of $(TMTSF)_2ClO_4$ in a direction, from 6 K to 280 K. The cooling rate through the T_{ao} is 0.0005 K/s, which make it getting into the relaxed state.

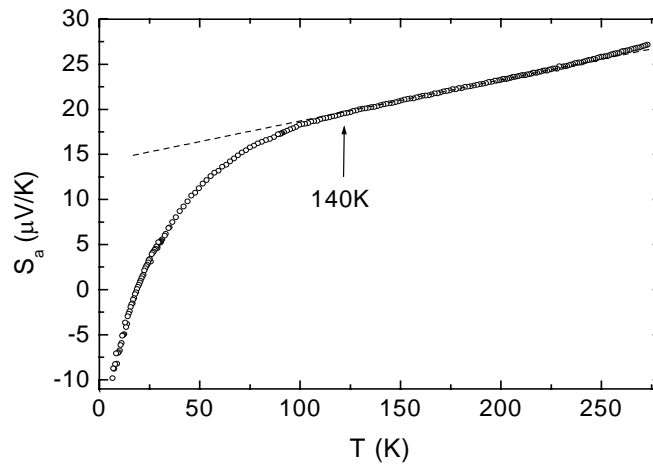Figure 3   The temperature dependent thermopower of $(TMTSF)_2ClO_4$ metallic state along the a direction

There is a linearly temperature dependent thermopower at high temperature. Below 140 K, the curve deviates from linear behavior and goes down all the way through. Such a deviation behavior can be attributed to a 1D—2D crossover, which is observed in EPR (electron paramagnetic resonance)[4]. The overall behaviors agree well with the previous results of Choi's [5]. The sign of thermopower S_a is positive at high temperature, implying the carriers are holes.

Figure 4 shows the temperature dependence of the thermopower below 30 K with different cooling rate through T_{ao}. Each cooling process below 35 K was reached by filling of He exchanging gas and the rate was controlled by the computer. The thermopower was measured by warming after bumping the gas for 12 hrs. For the fastest cooling rate of 1 K/s, it yields the SDW state at 4.5 K [3]. Comparing with the result of the slowest cooling process (relaxed state), there is an obvious upturn just below T_{ao} when the

cooling rate reaches 0.3 K/s. It is evident that random potential of the frozen anion disordering introduces
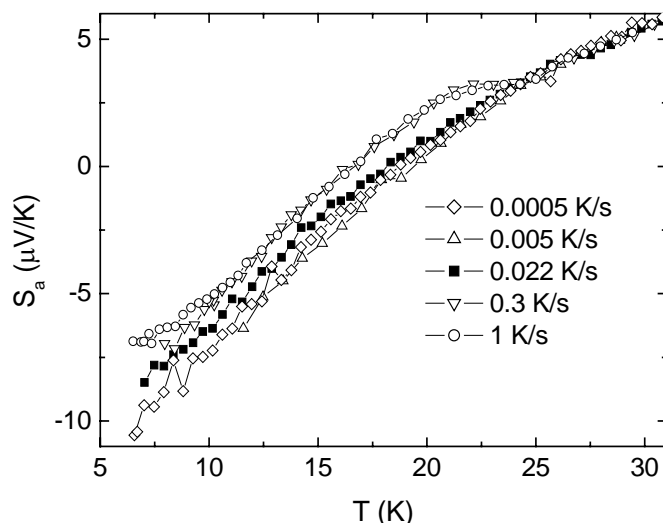

Figure 4   Temperature dependence of the thermopower with different cooling rate through the T_{ao}

the scattering. A clear trace of prelude to the SDW transition at 4.5 K can be spotted by the fastest cooling process, for there is an obvious increase in the thermopower below 10 K.

CONCLUSION

The design of a high precise measurement system of the thermopower is reported. The thermopower of organic conductors $(TMTSF)_2ClO_4$ needle single crystals were measured successfully. A linear behavior at high temperature and a deviation at 140K attributed to 1D—2D crossover were observed. There is an obvious upturn just below T_{ao} when the cooling rate reaches 0.3 K/s. A clear trace of prelude to the SDW transition at 4.5 K can be spotted by the fastest cooling process.

ACKNOWLEDGEMENT

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