# A phase transition in the organic metal $TSeTBr_{0.5}$

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Temperature dependences of the magnetic susceptibility and of the specific heat of the quasi-one-dimensional organic complex $TSeTBr_{0.5}$ were measured. The Pauli paramagnetic susceptibility decreases sharply in the range 15–30 K. At $T \approx 26$ K the specific heat changes abruptly by $(0.36 \pm 0.1)\,Nk$, which is an order of magnitude larger than the estimate obtained from the theory of the Peierls transition.

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The organic metals known as $TSeTCl_{0.5}$ and $TSeTBr_{0.5}$,[1,2] together with HMTSEF–TCNQ,[3] have a special place among the quasi-one-dimensional organic conductors. They are the only compounds of this type whose conductivity, after peaking as the temperature drops, instead of disappearing remains quite large (approximately at its room-temperature value) down to the lowest temperatures.[4]

Analysis of the electric and magnetic properties of these complexes suggests that they have a Peierls-type phase transition connected with partial dielectrization of their electronic systems.[1–3] However, a direct observation of such a transition using the x-ray or calorimetric method has not been reported in the literature. This letter describes a study of the specific heat of the $TSeTBr_{0.5}$ complex in the temperature range of 15–30 K.

This temperature range for measuring the specific heat was chosen from data on the conductivity of the complex[2] and from the results of measurements of the magnetic susceptibility, which are shown in Fig. 1. The magnetic susceptibility was measured in the temperature range 1.3–300 K on a Faraday-type magnetic balance.[5] The results of the measurements, corrected for ferromagnetic impurities and the diamagnetism of the lattice according to the Pascal law, are represented by curve 1 in Fig. 1. The spike at $T < 15$ K is well described by the Curie–Weiss law with a constant corresponding to $1.12 \times 10^{21}$ free spins per mole. The susceptibility obtained by subtracting the contribution of these impurities is represented by curve 2 in Fig. 1. We see that $\chi$ depends weakly on temperature over a wide interval; it then decreases significantly at 15–30 K, approaching a constant value at low temperatures. Such behavior, which is consistent with the temperature dependence of the conductivity, indicates that phase transition might occur in the neighborhood of 15–30 K.

The specific heat of $TSeTBr_{0.5}$ was measured in this temperature interval. An adiabatic calorimeter[6] whose design was modified extensively was used for the measurements. The following modifications were introduced: a) the copper conductor against which the sample was pressed was eliminated; b) the thermometer (a platinum resistance thermometer was used in the temperature interval measured) was brought out to the adiabatic shield; c) the calorimeter was lightened as much as possible and

  

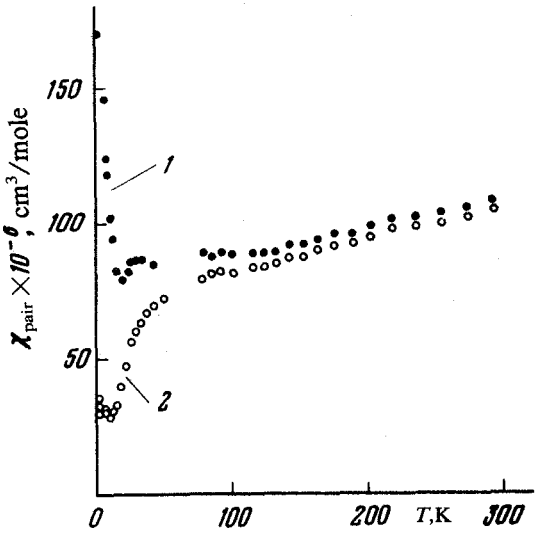


FIG. 1. Temperature dependence of the magnetic susceptibility of the $TSeTBr_{0.5}$ complex. 1) Measurements corrected for ferromagnetic impurities and lattice diamagnetism; 2) susceptibility after subtraction of the contribution of paramagnetic impurities.

the length of the current conductors was increased. By introducing these modifications, we were able to reduce the mass of the measured material to ~100 mg; but at the same time the contribution of the empty calorimeter rose to 50% (for a sample mass of 89 mg) and the random error of a single measurement increased to 1.5%,

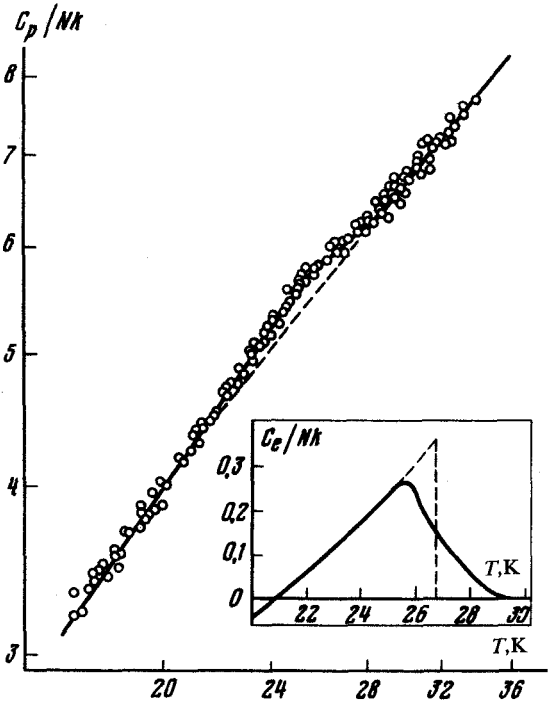


FIG. 2. Behavior of the specific heat in the neighborhood of the phase transition. The dashed line corresponds to the regular part described in the text. The specific heat near the transition after subtraction of the regular part is shown in the inset.

principally because of the direct effect of the noise from the adiabatic mode regulator on the sensitivity of the thermometer circuit.

Figure 2 shows the temperature dependence of the specific heat of $TSeTBr_{0.5}$. At a temperature of about 26 K we see a jump which comprises 4.5% of the total specific heat at this temperature; this greatly exceeds the average statistical measurement error of $\pm 0.2\%$. The regular part of the specific heat is very difficult to isolate because the behavior of the lattice specific heat in the given temperature dependence corresponding on a logarithmic scale to the straight line which describes the high-temperature part of our experimental data (dashed line in Fig. 1). The results of the subtraction are shown in the inset. Approximating the obtained curve by a triangular function, as suggested in Ref. 7, we obtain for the magnitude of the jump $\Delta C \sim (0.36 \pm 0.1)\, Nk$.

Our results indicate, then, that a phase transition occurs in the electronic system of the $TSeTBr_{0.5}$ complex at a temperature of about 26 K. Taking into account the known properties and the one-dimensional characteristics of the system, we would expect this transition to be of the Peierls type. The specific-heat jump can be estimated by using the expression given by the simple theory[7]:

$$\Delta c / Nk = 9{,}6\, k\, T_c\, \rho(\epsilon_F),$$

where $\rho(\epsilon_F)$, the density of states at the Fermi level, can be obtained from the formula for the Pauli paramagnetic susceptibility $\chi_P = 2\mu^2 \rho\ (\epsilon_F)$. Interpreting the weakly varying susceptibility in Fig. 1 as paramagnetism of the conduction electrons, we can estimate $\chi_P$ from the high-temperature value of the measured susceptibility. The estimate of $\chi_P = 10^{-4}$ cm$^3$/mole at $T = 300$ K gives $\Delta C/Nk = 0.03$, which is an order of magnitude lower than the measured value.

In conclusion, we note that the observed transition is not a pure Peierls transition because below the transition point the conductivity and susceptibility have a finite value. Since the accuracy of determining the contribution of the lattice makes it impossible quantitatively to predict the behavior of the susceptibility below the transition point, information about the low-temperature state of the complex must be obtained from measurements of other properties of this material at low temperatures.

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