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Magnetic Phase Transition in a Low-Dimensional Organic Antiferromagnet, 1,3-Bisdiphenylene-2-p-chlorophenyl Allyl, under High Pressure. Observation of a High Néel Temperature of 5.03 K

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Hydrostatic pressure dependence of the magnetic phase transition temperature in the organic free radical, 1,3-bisdiphenylene-2-p-chlorophenyl allyl, a one-dimensional Heisenberg antiferromagnet, was investigated from the heat capacity measurements in the pressure range of up to 6 kbar at the liquid helium temperature region. A high Néel temperature of 5.03 K was observed at 5.4 kbar.

The organic free radical, 1,3-bisdiphenylene-2-phenyl allyl (BDPA) has been one of the subjects of extensive study.[1-3] It has been concluded that BDPA is a one-dimensional antiferromagnet with a Heisenberg-type isotropic exchange interaction.[3] The Néel temperature is 1.78 K.[2] A derivative with a chlorine-atom substitution in the para-position of the central phenyl ring, 1,3-bisdiphenylene-2-p-chlorophenyl allyl (p-Cl-BDPA), exhibits a Néel temperature of 3.25 K,[4] which belongs to the highest category for the Néel temperature in the organic radical.[5] p-Cl-BDPA has been extensively investigated in order to disclose its magnetic interaction and spin-ordered state, using various experimental techniques.[3,4,6-9)]

In this communication we report the results of heat capacity measurements of p-Cl-BDPA under hydrostatic pressure in the liquid helium temperature region. The Néel temperature is increased by applying pressure, and comes up to 5.03 K at 5.4 kbar.

The sample was prepared by the literature procedure.[3,4)] The heat capacity measurements were carried out by the same method employed for the study of 2,2,6,6-tetramethyl-4-piperidinol-1-oxyl (TANOL).

Figure 1 shows the temperature dependence of the heat capacity at several hydrostatic pressures, atmospheric (**a**), 2.9 kbar (**b**), and 4.9 kbar (**c**). The heat capacity anomaly indicates the Néel temperature of p-Cl-BDPA.[4)] Under hydrostatic pressure this anomaly shifts to the high temperature side as is shown in Fig. 1, indicating that the Néel temperature increases with increasing pressure.

The pressure dependence of the Néel temperature, $T_N(P)$, is plotted in Fig. 2. There appears to be a good linear correlation between them. $T_N(P)$ for $P < 6$ kbar can be fitted by the following equation

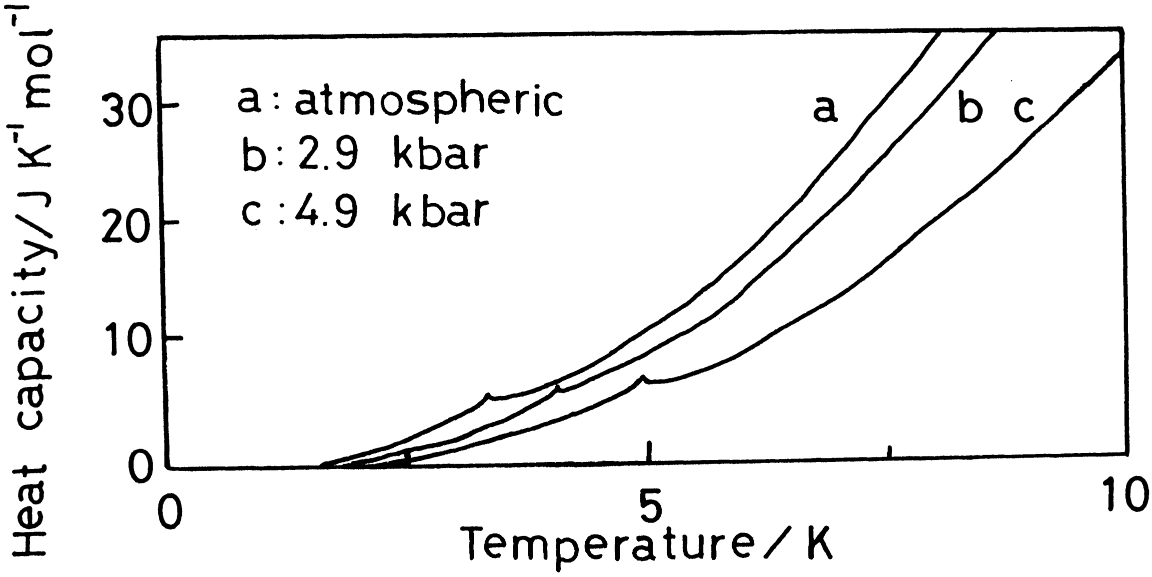

Fig. 1. Heat capacities under hydrostatic pressure.

$$T_N(P)/T_N(P_O)=1+0.083P \quad (1)$$

The linear coefficient $\gamma=0.083$ for p-Cl-BDPA is smaller than $\gamma=0.15$ for TANOL,[10] but is larger than those observed in many transition-metal compounds.[10]

The characteristic result of this study is the observation of the Néel temperature of 5.03 K at 5.4 kbar. This Néel temperature belongs to the highest category for the magnetic long-range-ordering temperature in the organic radical crystal.[5] There is a possibility of finding a magnetically long-range-ordered state with a higher Néel temperature by means of high pressure.

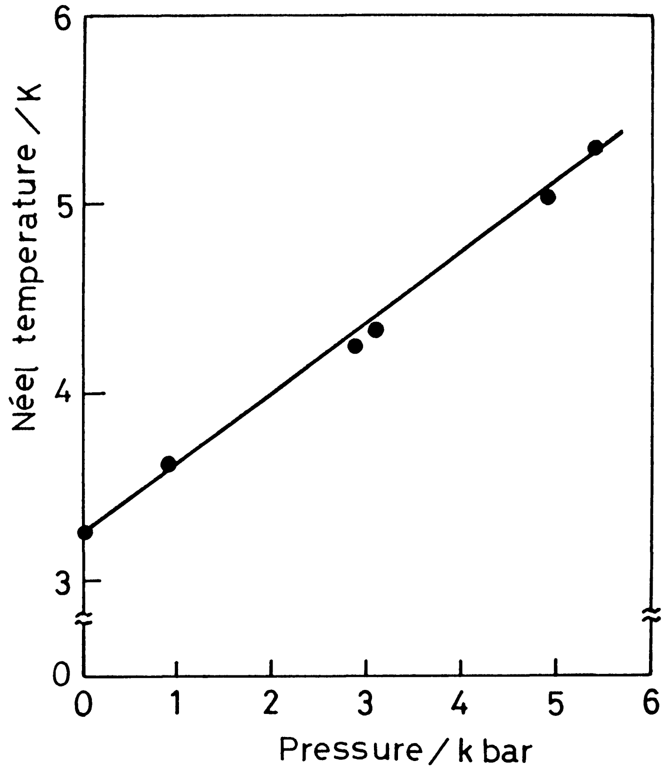

Fig. 2. Pressure dependence of the Néel temperature.

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