Pressure effect on the magnetism and structure of a spin-Peierls substance:MEM-[TCNQ]$_2$

K. Ejima[1], T. Tajiri[1], H. Deguchi[1*], M. Mito[1], S. Takagi[1], K. Ohwada[2], H. Nakao[3], Y.Murakami[3]
[1]Faculty of Engineering, Kyushu Institute of Technology, Kitakyushu 804-8550, Japan
[2]SPring-8, Koto Mikazuki-cho Sayo-gun Hyogo 679-5148, Japan
[3]Faculty of Science, Tohoku University, Sendai 980-8578 Japan

Introduction

An organic ion-radical salt, N-Methyl-N-ethyl-morphorinium- [7,7',8,8'-tetracyanoquinodimethane]$_2$ (MEM-[TCNQ]$_2$), is a one-dimensional(1D) spin system and shows a spin-Peierls(SP) transition at the SP transition temperature(T_{SP}) of 18K.

We report the significant pressure effects on the magnetic properties of MEM-[TCNQ]$_2$ [1], though they are rather different from the results reported by Bloch *et al* [2]. In order to clarify such anomalous pressure effects on the magnetism, we have carried out the crystal structure analysis of the substance under pressure by using synchrotron radiation(SR) X-rays.

Experiment

The salt, MEM-[TCNQ]$_2$, was synthesized by the standard method and crystallized by slow cooling of an acetonitrile solution. The susceptibility was measured by using a SQUID susceptometer (Quantum Design MPMS-5) with a hand-made pressure cell in the temperature range of 2-300K and at the pressure up to 8kbar.

The SR X-ray diffraction measurements of the single or powder crystals of the salt under ambient and high pressures have been carried out at the beamline 1B.

Results and Discussion

The paramagnetic susceptibility of the salt at various pressures is shown in Fig. 1 as a function of temperature. The susceptibility at ambient pressure (1bar) clearly shows the spin-Peierls transition at 18K below which the susceptibility suddenly decreases with decreasing temperature. The slight increase of the susceptibility below 7K is attributable to the small amount of paramagnetic impurities. Above 18K, the susceptibility is quantitatively explained by the Bonner-Fisher curve(dashed-line in the Fig. 1) with J/k = -52K.

As the pressure is increased up to 8kbar, the susceptibility at low temperatures becomes large. The susceptibility at above 0.84kbar monotonically increases with decreasing temperature and does no longer show a minimum. The susceptibility at low temperatures and higher pressures obeys the Curie-Weiss law with a very small Weiss temperature. Therefore, the increase of the susceptibility at low temperatures and higher pressures may result from the free spins, which must be produced by separating spin-pairs.

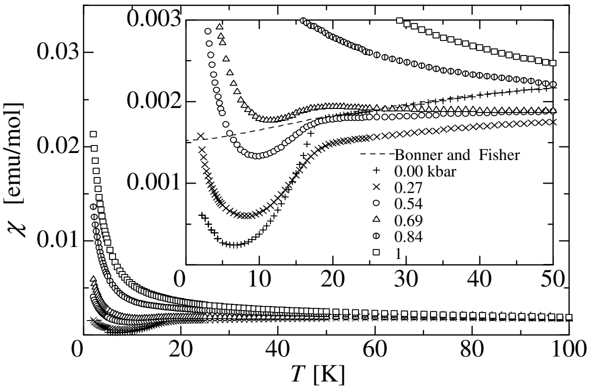

Fig.1. Temperature dependence of the magnetic susceptibility at various pressures.

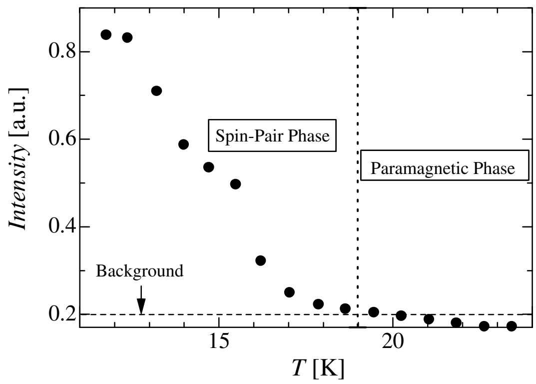

Fig.2. Temperature dependence of the reflection intensity of superlattice in the SP state.

The X-ray reflection resulting from the superlattice in the spin-Peierls state below 18K was clearly observed for a single crystal at ambient pressure. The temperature dependence of the intensity of the reflection is shown in Fig.2. However, the reflection was no longer observed under high pressures. It suggests that the spin-Peierls state is destroyed under pressure, which is consistent with the results of the magnetic measurements. The crystal structure of the salt is rather affected by the pressure even at room temperature.

References

[1] S.Takagi *et al.*, Mole.Crys. Liq. Cryst. **376** (2002) 377.
[2] D.Bloch *et al.*, Physica **119B** (1983) 43.

* deguchi@tobata.isc.kyutech.ac.jp