Impurity-Induced Antiferromagnetic Order in Organic Spin-Peierls Compound *p*-CyDOV

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Abstract

The doping effect of magnetic impurity (*p*-CyDTV) on an organic radical spin-Peierls (SP) compound *p*-CyDOV ($T_{\rm SP}$ = 15.0 K) has been studied by heat capacity measurement of the doped crystals $(p\text{-CyDOV})_{1-x}(p\text{-CyDTV})_x$. The antiferromagnetic transition was observed at $T_{\rm N}$ = 0.135, 0.290 and 0.164 K for the crystals with x = 0, 0.01 and 0.07, respectively. In the low doping region of x = 0 and 0.01, the antiferromagnetic order and the spin-Peierls state coexist, and at x = 0.07 the single phase of antiferromagnetic order is realized. The doping effect showed the similarity with that for an inorganic SP compound $CuGeO_3$.

Key words: spin-Peierls state; magnetic impurity; antiferromagnetic order; *p*-CyDOV; $CuGeO_3$

1. Introduction

The spin-Peierls (SP) transition is a magnetic-to-nonmagnetic transition accompanying a structural change in the one-dimensional Heisenberg antiferromagnetic system with a spin one-half. Recently, in imuprity-doped systems for an inorganic SP compound $CuGeO_3$ (SP transition temperature $T_{\rm SP}$ = 14.2 K), the antiferromagnetic (AF) order has been observed below 5 K [1–3], and especially the coexistence of SP state and AF order in the low-doping region has attracted much attention. The appearance of the AF order has been explained by considering that the disorder caused by the impurity induces the AF magnetic moment [4].

The study of the SP transition in organic compound is older than that in inorganic one [5]. However, the example of the impurity-doped organic SP compound has not been reported until recent years, and many physical studies for intrinsic SP state has been performed for the doped $CuGeO_3$ system [1–3].

Recently, a new organic SP compound *p*-CyDOV (3-(4-cyanophenyl)-1,5-dimethyl-6-oxoverdazyl; $T_{\rm SP}$ = 15.0 K) has been reported, and the doping effects have been studied for the *p*-CyDOV systems [6,7].

In this paper, the doping effect of magnetic impurity on *p*-CyDOV is investigated by the measurement of heat capacity.

2. Experimentals

A series of *p*-CyDOV crystals doped with *p*-CyDTV ($(p\text{-CyDOV})_{1-x}(p\text{-CyDTV})_x$) was synthesized according to the method mentioned in ref.6. The molecular structures of *p*-CyDOV and *p*-CyDTV are similar to each other as shown in Fig. 1, and both the molecules have an unpaired electron in a molecule. The results of the magnetic measurements of $(p\text{-CyDOV})_{1-x}(p\text{-CyDTV})_x$ have indicated that $T_{\rm SP}$ decreases with increasing x, and the SP transition is not observed for x

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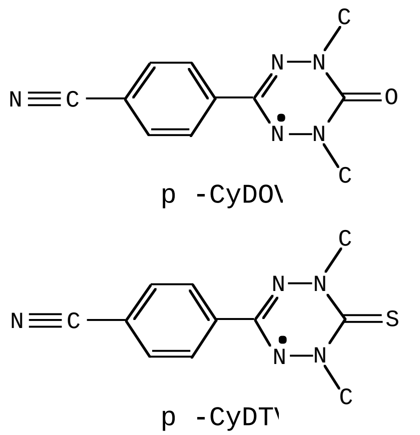

Fig. 1. The molecular structures of *p*-CyDOV and *p*-CyDTV.

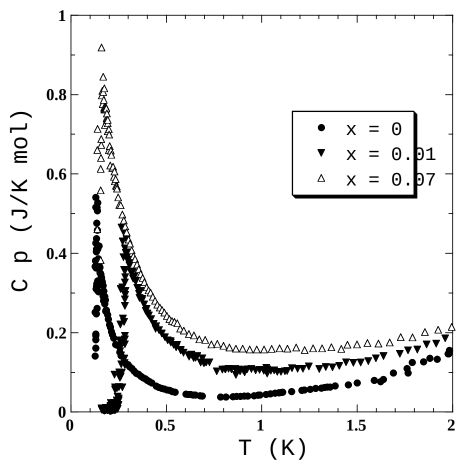

Fig. 2. Temperature dependence of heat capacity C_p for (p-CyDOV)$_{1-x}$(p-CyDTV)$_x$ for x = 0, 0.01 and 0.07.

$\geq$ 0.03. The heat capacity C_p was measured with the adiabatic heat-pulse method using the ^{3}He-^{4}He dilution refrigerator.

3. Experimental Results

Figure 2 shows the temperature dependence of C_p of (p-CyDOV)$_{1-x}$(p-CyDTV)$_x$ for x = 0, 0.01 and 0.07. There can be the impurity of 0.1-0.5 % even in the non-doped system (x = 0). The AF transition was observed at T_N = 0.135, 0.290 and 0.164 K for the crystals with x = 0, 0.01 and 0.07, respectivey. Each magnetic entropy of the AF order for the crystals with x = 0, 0.01 and 0.07 is estimated to be 5.3%, 6.3% and 14.4% of $Nk_B ln$ 2, respectivey.

Figure 3 shows the x-dependences of T_N and T_{SP}, where the values of T_{SP} are those obtained by magnetic susceptibility measurements [6]. In the low doping re-

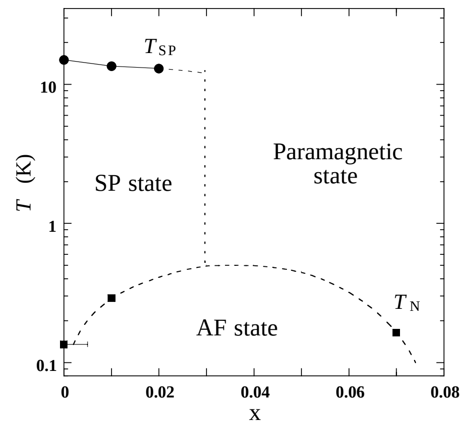

Fig. 3. T_N and T_{SP} for (p-CyDOV)$_{1-x}$(p-CyDTV)$_x$ with x = 0, 0.01 and 0.07. The value of T_{SP} is referred to the result of magnetic susceptibility in ref.6.

gion of x = 0 and 0.01, the SP state and the AF order coexist. For x = 0.07, a single phase of AF order is realized. These behaviors are consistent with the doping effect on $CuGeO_3$. However, T_N's of *p*-CyDOV are much lower than those of $CuGeO_3$. Due to the mean-field theory, the ratio between interchain and intrachain interactions is estimated to be 10^{-1} for $CuGeO_3$ and 10^{-4} for *p*-CyDOV. This indicates that *p*-CyDOV has ideal one-dimensional character.

4. Conclusion

The doping effect of magnetic impurity (*p*-CyDTV) on an organic radical SP compound *p*-CyDOV (T_{SP} = 15.0 K) has been studied by heat capacity measurement of (p-CyDOV)$_{1-x}$(p-CyDTV)$_x$. In the low doping region of x = 0 and 0.01, the antiferromagnetic order and the spin-Peierls state coexist, and at x = 0.07 the single phase of AF order is realized. The doping effect on *p*-CyDOV system is similar to that on $CuGeO_3$ system, and the disorder caused by the impurity does not only destroy the spin-Peierls state but also induces the AF order.

References

[1] S. B. Oseroff *et al*, Phys. Rev. Lett. **74** (1995) 1450.

[2] K. Manabe *et al*, Phys. Rev. B**58** (1998) R575.

[3] T. Masuda *et al*, Phys. Rev. Lett. **80** (1998) 4566.

[4] H. Fukuyama *et al*, J. Phys. Soc. Jpn. **65** (1996) 1182.

[5] S. Huizinga *et al*, Phys. Rev. B**19** (1979) 4723.

[6] K. Mukai *et al*, Chem. Phys. Lett. **311** (1999) 446.

[7] K. Mukai *et al*, J. Phys. Chem. B**103** (1999) 10876.