Impurity-Induced Antiferromagnetic Order in Organic Spin-Peierls Compound \( p\text{-CyDOV} \)

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Abstract

The doping effect of magnetic impurity (\( p\text{-CyDTV} \)) on an organic radical spin-Peierls (SP) compound \( p\text{-CyDOV} \) (\( T_{SP} = 15.0 \) 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 \( T_N = 0.135, 0.290 \) 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 \( \text{CuGeO}_3 \).

Key words: spin-Peierls state; magnetic impurity; antiferromagnetic order; \( p\text{-CyDOV}; \text{CuGeO}_3 \)

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 impurity-doped systems for an inorganic SP compound \( \text{CuGeO}_3 \) (SP transition temperature \( T_{SP} = 14.2 \) 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 \( \text{CuGeO}_3 \) system [1–3].

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

In this paper, the doping effect of magnetic impurity on \( p\text{-CyDOV} \) is investigated by the measurement of heat capacity.

2. Experimentals

A series of \( p\text{-CyDOV} \) crystals doped with \( p\text{-CyDTV} \) \(((p\text{-CyDOV})_{1-x}(p\text{-CyDTV})_x)\) was synthesized according to the method mentioned in ref.6. The molecular structures of \( p\text{-CyDOV} \) and \( p\text{-CyDTV} \) 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 \( T_{SP} \) decreases with increasing \( x \), and the SP transition is not observed for \( x \)
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 $0.07$. The heat capacity $C_p$ was measured with the adiabatic heat-pulse method using the $^3$He-$^4$He dilution refrigerator.

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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, respectively. 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 $Nk_B\ln 2$, respectively.

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 region 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 intra-chain 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