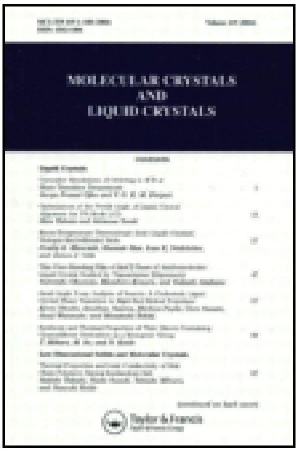

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Spin Correlations on a Site Diluted Square Lattice Heisenberg Antiferromagnet Covering the Percolation Threshold

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The reduction of the magnetic ordering temperature has been studied in the quasi-two-dimensional square lattice Heisenberg magnet, $Mn(HCOO)_2 2(NH_2)_2CO$($T_N(1)$=3.77 K) with the spin value S=5/2, randomly diluted with non-magnetic Cd^{2+} ions. It is found that, as the dilution proceeds, the Neel temperature $T_N(x)$ goes down toward 0 K around at x=0.65, higher than x_p=0.59 for the square lattice, making the contrast to the cases of the diluted Ising system and La_2CuO_4, a high Tc superconducting parent compound with S=1/2: The critical index ϕ in $T_N(x) \sim (x-x_p)^{1/\phi}$ is suggested $\phi<1$ for the Heisenberg and $\phi>1$ for the Ising systems.

Keywords; site dilution; percolation threshould; Heisenberg quantum magnet; two dimensions; heat capacity

INTRODUCTION

When the non-magnetic ions are substituted for the magnetic ions, the spin correlations on the magnetic lattice are disturbed to result in the reduction in the transition temperature. The reduction rate of $T_N(x)$ is theoretically studied as functions of the magnetic concentration x and anisotropies for various lattices[1]. The two-dimensional(2d) percolation threshold which gives $T_N(x)=0$ is expected to be $x_p=0.592746(5)$ for both the Heisenberg and Ising systems[2]. In the real case of the Heisenberg antiferromagnet with the spin value $S=1/2$, La_2CuO_4, diluted with non-magnetic and non-itinerant impurities, however, the critical concentration looks to be $x_c=0.75$ from the linear extrapolation of observed $T_N(x)$ down to zero[3], supposedly due to the strong quantum effects in the ramified 2d lattice. Moreover, the critical index ϕ in $T_N(x)\sim(x-x_p)^{1/\phi}$ for 2d quantum systems has been examined by the recent theory[4].

Here we report the site dilution effects of a quasi-2d Heisenberg antiferromagnet $Mn(HCOO)_2 2(NH_2)_2CO$(MF2U) with $S=5/2$. This is an ideal parent compound since the non-magnetic Cd^{2+} impurities can be substituted for $Mn^{2+}(3d^5)$ on the simple square magnetic lattice in the wide range of x. The pure system orders at the lower temperature $T_N(1)=3.77$ K[5] than the other compounds studied so far such as K_2CuF_4(6.25 K)[6], Rb_2CoF_4(103 K)[7], K_2MnF_4(42K)[8] and La_2CuO_4(315K)[3], and this enables us to get intrinsic magnetic ordering temperatures by the heat capacity measurement without perturbation by the thermal fluctuations at high temperatures as in other compounds.

EXPERIMENTAL

The crystal structure of $M(HCOO)_2\ 2(NH_2)_2CO$ belongs to the space group $P4_12_12$ (M;Mn, Cd) [9]. The Cd^{2+} ions are substituted for Mn^{2+} ions in the wide range of the manganese concentration x. Hereafter we express the diluted system as $Mn_xCd_{(1-x)}(HCOO)_2 2(NH_2)_2CO$.

Normally we started with preparing the slightly oversaturated solution of the calculated amount of constituent formate salts from which 1/4mol(around 90gr) of the diluted compound with a fixed value of x would be crystallized eventually. The mixture begun to grow in the solution at constant temperature (50 C) after several weeks. The quality of the diluted systems were assured by the quantitative analysis of Mn, Cd, H, N, and C atoms, besides the X-ray analysis[9]. The heat capacity was measured for about 0.3 gr of each samples by a conventional adiabatic method. The magnetic heat capacity C_m was obtained by subtracting the lattice contribution in reference of that of the Cd^{2+}(x=0) compound[5], which was approximated by the 3d Debye function with the Debye temperature θ_D=128K and 3 effective oscillators.

RESULTS AND DISCUSSION

Figure 1 shows the concentration dependence of the magnetic heat capacities for the diluted system. It is clear that the sharp peak at $T_N(x)$ sensitively shifts down to the lower temperatures as x decreases in

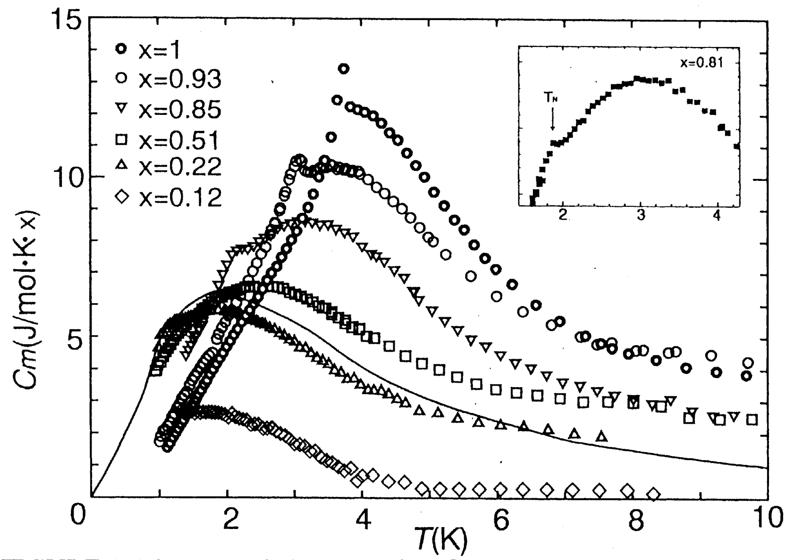

FIGURE 1. The magnetic heat capacity of $Mn_xCd_{(1x)}(HCOO)_2 2(NH_2)_2CO$. The transition temperature for the system with x=0.81 is indicated in the inset. The solid curve shows the theoretical results for the one-dimensional Heisenberg spin system for S=5/2 and J/k_B=-0.34 K.

contrast to the gradual shift of the characteristic broad hump of 2d Heisenberg system. As seen in the inset of Fig.1, we can trace $T_N(x)$ as a distinct cusp on the lower temperature side of the broad hump for x=0.81. This smearing out of the heat capacity peak is not only due to the distribution of x but intrinsic dilution effects as seen even in the system with Ising type anisotropy[10].

It is noted that the reduction rate of the Neel temperature defined as

$$R = d/dx\{T_N(x)/T_N(1)\}$$

becomes 2.7 as shown in Fig.2. In the case of K_2MnF_4 diluted with Mg^{2+} ions, R=3.0 is reported[8], while for the diluted Rb_2CoF_4 system

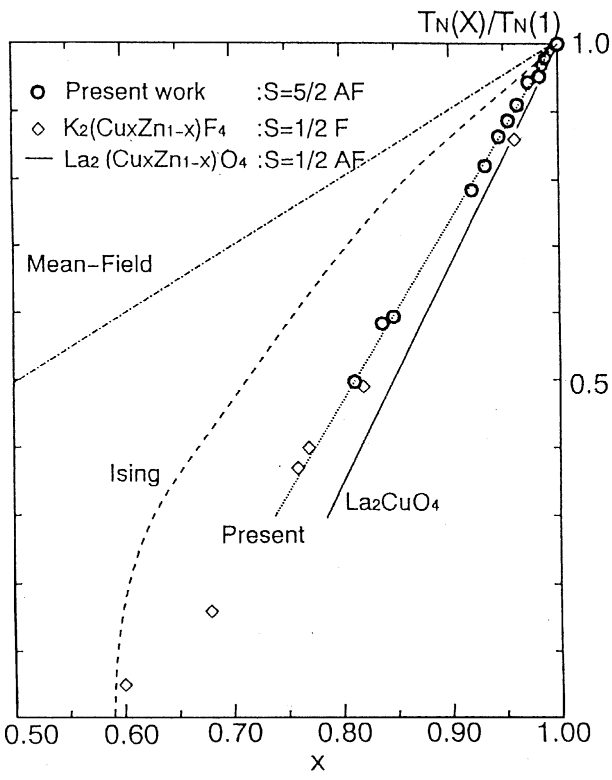

FIGURE 2. The concentration dependence of the transition temperature for the square lattice magnets. The present results are in between the cases for La_2CuO_4(S=1/2,AF)[3] and the Ising system which is estimated experimentally[7,8] and theoretically[1,11,12].

with the strong Ising type anisotropy, R=1.9 is experimentally obtained[7]. The theoretical value of R for the square lattice is estimated $R=\pi$ at the limit of Heisenberg spin symmetry by the random phase approximation[11]. In this theory, R becomes smaller for the system with the larger Ising type anisotropy, and, approaches R=1.33 at the Ising limit, comparable with 1.57 for the exact value[12]. The present experimental value R=2.7 indicates the character of the Heisenberg spin system with the small Ising anisotropy.

On the other hand , for the S=1/2 antiferromagnetic Heisenberg system, La_2CuO_4, the initial gradient is estimated around R=4.0 beyond the theoretical value. The tentative extrapolation of Eq.(1) with R=2.7 (4.0) gives an impression that T_N(x) becomes zero around at a pseudo-critical concentration $x=x_c$=0.65 (0.75). As mentioned in Introduction, however, the critical concentration for the Ising and classical Heisenberg systems are x_p=0.59, and therefore the curve of $T_N(x)$ of these Heisenberg cases must draw concavely (ϕ<1) near $T_N(x_p)$=0 at $x=x_p$, if the system would be ideally two-dimensional. When there exists infinitesimally small interlayer interactions, $T_N(x)$ behaves to come down concavely to approach the lower critical concentration for the three-dimensional systems[1]. This behavior of $T_N(x)$ with a concave curve for the Heisenberg systems makes a remarkable contrast to the convex curve (ϕ>1) for the Ising systems.

Here we comment on the variation of the magnetic heat capacity curve as x decreases. When x=1, the absolute values of both the magnetic heat capacity and the spin correlation length are well reproduced by the recent theory of the pure quantum self-consistent harmonic approximation for the two-dimensional system[13]. It is pointed out that dilution effectively reduces the magnetic lattice dimensionality[6,14-18] : At x_p=0.59, the fractal dimensionality is 1.89. The spin correlation length in the vicinity to the percolation threshold of K_2MnF_4 is studied by the neutron scattering experiment and understood in terms of the spin correlation length of the one-dimensional classical Heisenberg spin system[16]. In Fig. 2, we tentatively show the theoretical curve for the 1d Heisenberg chain for S=5/2 and J/k_B=-0.34K, the original value for MF2U. The experimental values for x=0.51 are not far from the theoretical curve as seen in other cases[6,15], giving an indication that such a ramified system may be convoluted into some low-dimensional systems by, for example, a self-avoiding walk model.

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