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LETTER TO THE EDITOR

Antiferromagnetic transitions in high-T_c materials

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Abstract. The Néel ordering transition of the 2D spin-$\frac{1}{2}$ Heisenberg antiferromagnet is studied in the small in-plane anisotropy limit through extensive quantum Monte Carlo simulations on lattices as large as 96 x 96. At an Ising-like anisotropy energy $h^A = 0.0025$, the system orders at $T_c/kT = 0.295$. This striking effect and related results agree with a wide class of experiments and give some insight into these materials.

Theoretically, two-dimensional isotropic Heisenberg quantum spins remain in a paramagnetic state at all temperatures [1,2]. However, all the crystals found in nature with strong 2D magnetic character go through phase transitions into ordered states [3,4]. These include the recently discovered high-T_c materials, La_2CuO_4 and $YBa_2Cu_3O_6$, despite the presence of large quantum fluctuations in the spin-$\frac{1}{2}$ antiferromagnets. At present, the popular explanation for the antiferromagnetic ordering transitions in these high-T_c materials emphasizes the very small coupling, J', between the 2D layers; J'/J is estimated to be about 10^{-5}. However, all these systems exhibit some kind of in-plane anisotropy which is of order 10^{-3}. An interesting case is the spin-1 crystal K_2NiF_4 discovered twenty years ago [3]. The magnetic behaviour of K_2NiF_4 exhibits very strong 2D character with an exchange coupling $J = 104$ K. It has a Néel ordering transition at $T_N = 97$ K, induced by an Ising-like anisotropy, $h^A \approx 0.002$.

In this letter, we provide clear evidence to support the picture that the in-plane anisotropy is also quite important in bringing about the observed antiferromagnetic transition in the most interesting spin-$\frac{1}{2}$ case. Adding an anisotropy energy as small as $h^A = 0.0025$ will induce an ordering transition at $T_c/kJ = 0.295$. This striking effect and related results, obtained by extensive quantum Monte Carlo simulations, agree well with a wide class of experiments and provide some insight into this type of material.

In the antiferromagnetic spin system, superexchange led to the dominant isotropic coupling. One of the high-order effects [4,5], due to crystal field, is written as $-DS_z^2$, which is a constant for these spin-$\frac{1}{2}$ high-T_c materials. Another second-order effect is the spin–orbit coupling. This effect will pick up a preferred direction and lead to a $S_i^z S_j^z$ term, which also arises due to the lattice distortion in La_2CuO_4. More complicated terms like the antisymmetric exchange [6] can also be generated. For simplicity and clarity, we focus the study on the antiferromagnetic Heisenberg model

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with an Ising-like anisotropy:

$$H = \sum_{\langle ij \rangle} J \boldsymbol{S}_i \cdot \boldsymbol{S}_j + h S_i^z S_j^z \tag{1}$$

where $\boldsymbol{S}_i$ is the spin-$\frac{1}{2}$ operator at the Cu site. The anisotropy parameter h is related to the usual reduced anisotropy energy h^{A} through $h^{\mathrm{A}} = h/4J$. In the past, the anisotropy field model [7], $\Sigma \epsilon_i H_{\mathrm{A}} S_i^z$, has also been measured [3]. However, its origin is less clear and, furthermore, the Ising symmetry is explicitly broken.

The quantum Monte Carlo simulation uses the Suzuki–Trotter transformation [8]. The rather complex algorithm is developed [2] on a state-of-the-art parallel computer, the Caltech/JPL MarkIIIfp. To minimize the finite-size effects we used spin systems of sizes up to 96 × 96. We used $1/mT \leq 0.1$ so that the systematic errors introduced in the transformation are quite small [2, 8], well within the statistical accuracy of the simulations.

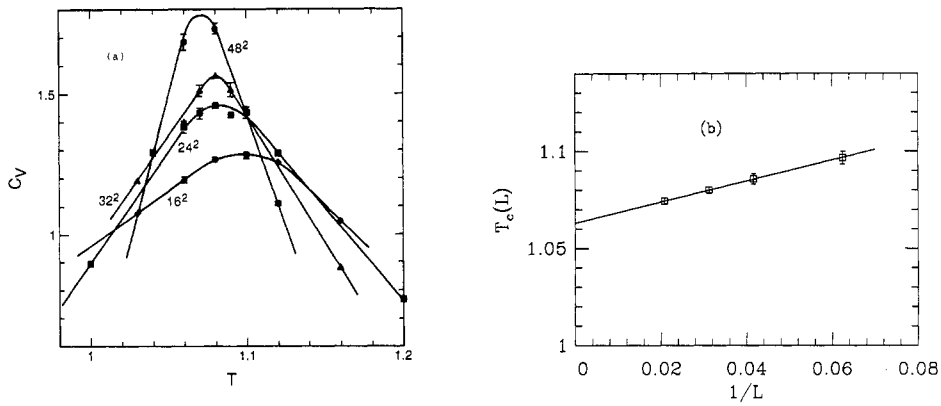

Figure 1. (*a*) The specific heat for different sized systems for $h = 1$. (*b*) Finite-size scaling for $T_c(L) - T_c \propto L^{-1}$.

For the large anisotropy system, $h = 1$, the specific heat C_V is shown for several spin systems in figure 1(*a*). The peak becomes sharper and higher as the system size increases, indicating a divergent peak in an infinite system, similar to the 2D Ising model. Defining the transition temperature $T_c(L)$ at the peak of C_V for the finite $L \times L$ system, the finite-size scaling theory [9] predicts that $T_c(L)$ relates to T_c through the scaling law

$$T_c(L) - T_c \propto L^{-\nu}. \tag{2}$$

Setting $\nu = 1$, the Ising exponent, a good fit with $T_c = 1.063 \pm 0.003$, is shown in figure 1(*b*). A different scaling with the same exponent for the correlation length,

$$\xi \propto (T - T_c)^{-\nu} \tag{3}$$

is also satisfied quite well, resulting in $T_c = 1.05 \pm 0.01$. The staggered magnetization drops down near T_c, although the behaviours are rounded off on these finite-sized systems. All the evidence clearly indicates that an Ising-like antiferromagnetic transition occurs at $T_c = 1.06$, with a divergent specific heat. In the smaller anisotropy case,

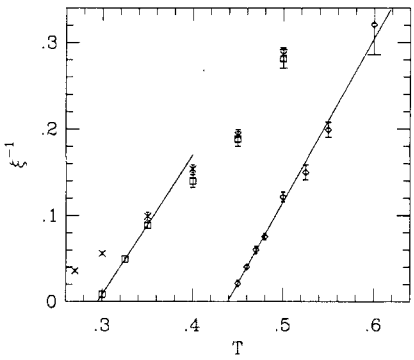

Figure 2. The inverse correlation lengths for the $h = 0.1$ system ($\Diamond$), for the $h = 0.01$ system ($\Box$), and for the $h = 0$ system ($\times$) for comparison. The straight lines are the scaling relation: $\xi^{-1} \propto T - T_c$. From it we can pin down T_c.

$h = 0.1$, similar behaviour is found. The scaling for the correlation length is shown in figure 2, indicating a transition at $T_c = 0.44$. However, the specific heat remains finite at all temperatures.

The most interesting case is $h = 0.01$ (or $h^A = 0.0025$, very close to those in K_2NiF_4 [3]). Figure 3 shows the staggered correlation function at $T = 0.3$ compared with that for the isotropic model [2]. The inverse correlation length measured, together with that for the isotropic model ($h = 0$), is shown in figure 2. Below $\xi^{-1} \approx 0.1$ the Ising behaviour is clearly shown as a straight line. Clearly, the system becomes antiferromagnetically ordered around $T = 0.3$. The best estimate is

$$T_c = 0.295 \qquad h = 0.01. \tag{4}$$

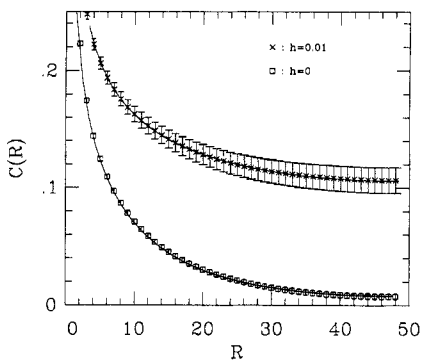

Figure 3. The correlation function on a 96 x 96 system at $T = 0.3$ for $h = 0.01$. It decays with correlation length $\xi \approx 120$. Also shown is the isotropic case $h = 0$, which has $\xi = 17.5$.

It may seem a little surprising that a very small anisotropy can lead to a substantially higher T_c. This may be explained by the following argument. At low T, the spins are highly correlated ($\xi \sim e^{2\pi\rho/T}$ [2]) in the isotropic case. Since no direction is preferred, the correlated spins fluctuate in all directions, resulting in zero

net magnetization. Adding a very small anisotropy into the system introduces a preferred direction, so that the already highly correlated spins will fluctuate around this direction, leading to a global magnetization.

More quantitatively, the crossover from the isotropic Heisenberg behaviour to the Ising behaviour occurs at T_{cr} where the correlation length is of order of some power of the inverse anisotropy. From the scaling arguments [10], $\xi \sim h^{-\nu/\phi} \approx h^{-1/2}$ where ϕ is the crossover exponent. For $h = 0.01$, this relation indicates that the Ising behaviour is valid for $\xi^{-1} \leq 0.1$, which is clearly observed in figure 2. A similar crossover around $\xi^{-1} \approx 0.3$ for $h = 0.1$ is also observed in figure 2. At low T, for the isotropic quantum model, the correlation length behaves as [2] $\xi \sim e^{2\pi\rho/T}$ where $\rho = Z_{\xi}^{(S)} S(S+1) = 0.199$ for spin-$\frac{1}{2}$. Therefore we expect $\exp(4\pi\rho/T_{cr}) \sim h^{-1}$. Since T_c is somewhat below T_{cr}, we expect

$$T_c \approx Z^{(S)} S(S+1)/\log(h^{-1}) \tag{5}$$

where $Z^{(S)}$ is a spin-S-dependent constant of order 1. Therefore, even a very small anisotropy (h) will induce a phase transition at a substantially higher temperature ($T_c \gg h$). This crude picture, suggested a long time ago [7, 11] to explain the observed phase transitions, is now confirmed by extensive quantum Monte Carlo simulations for the first time. Note that this problem is an extreme case both because it is an antiferromagnet (more difficult for it to become ordered than the ferromagnet) and because it has the largest quantum fluctuations (spin-$\frac{1}{2}$). Since $\log(h^{-1})$ varies slowly with h, we can estimate $Z^{(S)}$ at $h = 0.01$:

$$Z^{(1/2)} \simeq 1.9. \tag{6}$$

This simple result correctly predicts T_c for a wide class of crystals found in nature, assuming the same level of anisotropy, i.e., $h^A \sim 0.002$. The high-T_c superconductor $YBa_2Cu_3O_{6.1}$ exhibits a Néel transition at $T_N = 435$ K. With $J \approx 1400$ K [12], our results give a quite close estimate: $T_c = 420$ K. Similar close predictions hold for other $S = \frac{1}{2}$ systems such as the superconductor $ErBa_2Cu_3O_7$ [13] and the insulator K_2CoF_4 [14]. For the high-T_c material La_2CuO_4, $J = 1450$ K [2]. This material undergoes a Néel transition at $T_N \simeq 220$ K [15, 16]. Our prediction of T_c=428K is in the same range of T_N (compared with the naive expectation that $T_c \sim h \sim 10$ K). In this crystal, there is some degree of frustration (see below), so the actual transition is pushed down. These examples clearly indicate that in-plane anisotropy could be quite important to bring the system to the Néel order for these high-T_c materials. For the $S = 1$ system K_2NiF_4 [3], our results predict a $T_c = 81$ K, quite close to the observed $T_N = 97$ K.

These results have direct consequences regarding the critical exponents. The onset of a transition is entirely due to the Ising-like anisotropy. Once the system becomes Néel ordered, different layers in the 3D crystals will order at the same time. Spin fluctuations in different layers are incoherent, so the critical exponents such as β, γ, ν will be the 2D Ising exponents instead of the 3D Ising exponents. $ErBa_2Cu_3O_7$ and K_2CoF_4 show clearly such behaviour. However, the interlayer coupling, albeit very small (much smaller than the in-plane anisotropy), could induce coherent correlations between the layers, so the critical exponents will be somewhere in between the 2D and the 3D Ising exponents. La_2CuO_4 and $YBa_2Cu_3O_6$ seem to belong to this category.

Whether the ground state of the spin-$\frac{1}{2}$ antiferromagnet spins has the long-range Néel order is a long-standing problem [17]. The existence of the Néel order is rigorously

proved [18] for $h \geq 0.78$. In the most interesting case ($h = 0$), numerical calculations on small lattices [20, 21] suggested the existence of long-range order. Our simulation establishes the long-range order for $h \geq 0.01$.

The fact that near T_c, the spin system is quite sensitive to the tiny anisotropy could have a number of important consequences. For example, the correlation lengths measured in La_2CuO_4 are systematically smaller than the theoretical prediction [2] near T_c. The greater weakness of the correlations probably indicates that the frustrations due to the next-nearest-neighbour interaction come into play. This is consistent with the fact that T_N is below the T_c suggested by our results.

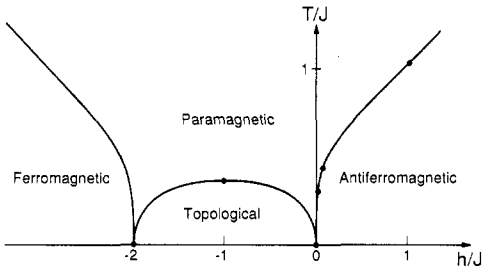

Figure 4. Phase diagram for the spin-$\frac{1}{2}$ quantum system (equation (1)). The full points are from quantum Monte Carlo simulations. For large $|h|$, the system is practically an Ising system. Near $h = 0$ or $h = -2$ the logarithmic relation (5) holds.

These results, together with those on the isotropic antiferromagnetic Heisenberg model [2] and the XY model [19] (where it is found that a Kosterlitz–Thouless-type transition occurs at $T_c = 0.350 \pm 0.004$) reveal a rich structure as shown in the phase diagram (figure 4) for these $S = \frac{1}{2}$ quantum spins. When $h \leq 0$, i.e. when the system has the XY-like anisotropy, we expect that arguments similar to those leading to (5) will also hold in this case. The antiferromagnetic ordered region and the topological ordered region are especially relevant to the high-T_c materials.

In summary, our extensive quantum Monte Carlo study of the 2D antiferromagnetic Heisenberg spins shows clearly that a very small Ising-like in-plane anisotropy could cause alignment of the spins at a relatively high temperature even in the extreme quantum case: spin-$\frac{1}{2}$. This provides an alternative interpretation of the observed Néel ordering transitions in the undoped high-T_c materials.

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