

# Monte Carlo study of first-order transition in Heisenberg fcc antiferromagnet

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Nearest-neighbor Heisenberg antiferromagnet on a face-centered cubic lattice is studied by extensive Monte Carlo simulations in zero magnetic field. The parallel tempering algorithm is utilized, which allows to overcome a slow relaxation of the magnetic order parameter and fully equilibrate moderate size clusters with up to  $N \simeq 7 \cdot 10^3$  spins. By collecting energy and order parameter histograms on clusters with up to  $N \simeq 2 \cdot 10^4$  sites we accurately locate the first-order transition point at  $T_c = 0.4459(1)J$ .

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Geometric frustration generally denotes inability of a magnetic system to find unique classical ground state. It arises due to a competition between exchange interactions for certain types of magnetic sublattices. The well-known examples are triangular and kagomé lattices in two dimensions and pyrochlore and face-centered cubic (fcc) lattices in three dimensions. Intriguing behavior of geometrically frustrated magnetic materials have attracted a lot of experimental and theoretical attention in the past decade [1]. Frustrated properties of Ising antiferromagnet on an fcc lattice (fig.1) have been recognized a long-time ago [2]. The case of vector (Heisenberg)

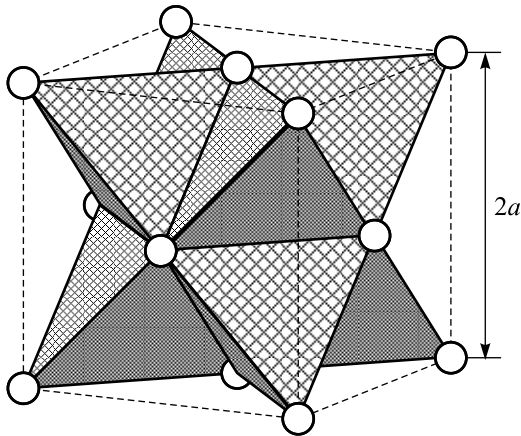


Fig.1. Face-centered cubic lattice as a network of edge-sharing tetrahedra

spins has been investigated to a lesser extent. Experimental realizations of fcc magnets include type-I antifer-

romagnet  $\text{UO}_2$  [3, 4] and type-II antiferromagnet  $\text{MnO}$  [5].

In the present work we investigate a nearest-neighbor Heisenberg antiferromagnet on an fcc lattice described by the Hamiltonian

$$\mathcal{H} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

where  $\mathbf{S}_i$  is a classical vector spin of unit length and  $J$  is the exchange constant. Every spin interacts with 12 nearest neighbors separated by  $(\pm a, \pm a, 0)$ ,  $(0, \pm a, \pm a)$  and  $(\pm a, 0, \pm a)$ . Frustrated properties of the model (1) become apparent if one calculates the mean-field transition temperature  $T_c^{\text{MF}} = \frac{1}{3} |\min\{J_{\mathbf{q}}\}|$ . The Fourier transform of exchange interaction is

$$J_{\mathbf{q}} = 4J [\cos(q_x a) \cos(q_y a) + \cos(q_y a) \cos(q_z a) + \cos(q_z a) \cos(q_x a)]. \quad (2)$$

The minimum is reached at  $\mathbf{Q} = (\pi/a)(1, q, 0)$  with arbitrary  $q$  and all equivalent wave-vectors in the cubic Brillouin zone. This set includes, in particular, the type-I antiferromagnetic structure with  $\mathbf{Q}_1 = (\pi/a)(1, 0, 0)$  and the type-III ordering with  $\mathbf{Q}_3 = (\pi/a)(1, \frac{1}{2}, 0)$ . The type-I (type-III) structure becomes the only absolute minimum of  $J_{\mathbf{q}}$  if a weak second-neighbor exchange is added to Eq. (1) with ferromagnetic (antiferromagnetic) sign, see, *e.g.*, [6].

Degeneracy of the nearest-neighbor model (1) is a consequence of decomposition of an fcc lattice into a network of edge-sharing tetrahedra, such that every site is shared between 8 tetrahedra. The classical ground state constraint consists, then, in a requirement of zero total spin for every tetrahedron. This yields an infinite

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number of collinear and noncollinear state with different periodicity, all of them having the same classical energy. The harmonic spin-wave analysis shows that at low temperatures thermal fluctuations select collinear states by an order by disorder effect [6]. Such a result can be most easily understood by a method suggested in Ref. [7], where the effect of short wave-length thermal fluctuations is shown to lead to an effective biquadratic exchange between neighboring spins. For the classical spins on an fcc lattice thermal fluctuations generate the following low-temperature correction to the free-energy:

$$\Delta F = -\frac{T}{32} \sum_{\langle ij \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j)^2. \quad (3)$$

Such a biquadratic interaction favors collinear spin arrangement. Two examples of collinear ground states include type-I spin structure  $\mathbf{S}_i = \hat{\mathbf{e}} \cos(\mathbf{Q}_1 \cdot \mathbf{r}_i)$  and type-III configuration  $\mathbf{S}_i = \hat{\mathbf{e}} \cos(\mathbf{Q}_3 \cdot \mathbf{r}_i + \frac{1}{4}\pi)$ . Additional collinear ground states are constructed from the above two configurations by selecting crystal planes (parallel to one of the cubic axes) with the Néel type of spin order and rotating all spins in such planes by  $180^\circ$ . All obtained collinear states have exactly the same free-energy in the harmonic approximation due to an extra gauge symmetry of a quadratic Hamiltonian [7]. Their degeneracy is lifted by anharmonicities in the spin-wave Hamiltonian—the problem, which to our knowledge has not been considered analytically.

Finite temperature transition of a type-I fcc antiferromagnet has been studied by the renormalization group approach [8, 9]. Absence of stable fixed points within the  $\epsilon$ -expansion suggests a first-order transition driven by thermal fluctuations. The above calculations have been performed for the case when the type-I structure corresponds to the absolute minimum, that is the case of the spin model (1) with a significant ferromagnetic second-neighbor exchange. In the nearest-neighbor case the anomalous contribution of thermal fluctuations is further enhanced due to extra soft modes. Therefore, the conclusion about a first-order transition should remain essentially unchanged.

Numerical Monte Carlo (MC) simulations of the finite temperature phase transition in a nearest neighbor fcc antiferromagnet have been performed by a standard single spin-flip technique [10–12]. First-order nature of the transition at  $T_c \simeq 0.45J$  was unambiguously established from the finite size scaling of the peak in the specific heat [11] and has been further supported by the energy histograms collected at the transition point [12]. As for the type of magnetic ordering at low temperatures there is no consensus among different authors. The first works have suggested the collinear type-I antiferromag-

netic structure [10, 11], though no results for the magnetic structure factor have been produced. In the subsequent more detailed study [12] the low-temperature phase of an fcc antiferromagnet was described as ‘a collinear state with glassy behavior.’ Apparent difficulty in simulation of an fcc antiferromagnet at low temperatures comes from the above mentioned degeneracy between various collinear states, which correspond to different local minima of the free-energy functional. The collinear states transform into each other under rotation of all spins in one crystal plane. The local minima of the free-energy are, therefore, separated by rather large entropy barriers  $\sim L^2$ ,  $L$  being a linear size of the system. A single spin-flip MC technique cannot produce appreciable moves in the phase space between distinct collinear configurations. As a result, the spin system hardly relaxes to the absolute minimum and the magnetic structure factor does not exhibit good averaging in a reasonable computer simulation time.

In the present work we shall apply the novel exchange MC method [13] for simulation of a Heisenberg fcc antiferromagnet. This modification of the standard MC technique, also called parallel tempering [14], has been developed for spin glasses, which is an outstanding example of hardly relaxing spin systems with numerous local minima separated by macroscopic energy barriers. In the exchange MC technique, several replicas of the spin system are simulated in parallel at a preselected set of temperatures. After a few ordinary MC steps performed on each replica, adjacent in temperature replicas are exchanged with a specially chosen probability [13]. The ensemble of parallel tempering, thus, includes two Markov processes: stochastic motion in the multidimensional phase space of the spin model and a random walk along a one-dimensional array of replicas. The second auxiliary process helps to dramatically decrease the correlation time for the main stochastic motion by repeatedly heating a given replica to high temperatures, where it quickly loses memory about the low-temperature magnetic structure. The exchange MC technique allows to equilibrate moderate size systems and has gained popularity in simulation of spin glasses [15] and frustrated Ising models [16]. This method has not been applied so far to geometrically frustrated vector spin models.

We have performed exchange MC simulations of the model (1) on finite clusters with periodic boundary conditions and  $N = 4L^3$  sites for  $L = 4, 6, 8, 10$ , and  $12$  ( $N = 256$ – $6912$ ). The highest temperature for the exchange MC ensemble was selected at  $T_{\text{up}} = 0.75J > T_c$ , where the system equilibrates fast with a standard MC technique. The lowest temperature in our simulations

was  $0.1J$ . The intermediate temperatures have been determined empirically starting with a geometric progression in such a way that the exchange rates for replicas swaps are roughly uniform and exceed 0.1 for the largest clusters. In particular, in the vicinity of the first-order transition point the temperature steps have to be decreased in order to avoid a bottleneck for replicas drifts across  $T_c$ . In total we have employed  $N_T = 72$  replicas for most cluster sizes. The MC procedure for simulation of individual replicas is the multiflipping Metropolis method, see, *e.g.*, [11], which includes several flipping attempts ( $m = 5$  in our case) on every spin under the same local field before moving to a next spin. This method is especially suitable for an fcc lattice with a large coordination number  $z = 12$ , when calculation of local fields takes a significant part of the CPU time. After sweeping 2 times over the whole cluster, the replica exchanges have been performed. The above procedure constitutes one exchange MC step. All replicas were set in random initial configurations and equilibrated over  $10^5$  exchange MC steps. Measurements have been performed for additional  $5 \cdot 10^5 - 4 \cdot 10^6$  exchange MC steps. Equilibration of various physical characteristics has been checked (i) by absence of time evolution over at least a half of measuring time, and (ii) by comparison to the results for a different set of initial replica configurations obtained by gradual annealing of a single replica from high temperatures.

During simulations of the exchange MC ensemble the internal energy  $E$ , the specific heat, the magnetic structure factor, and the collinear order parameter have been measured. Results for the internal energy and the specific heat are identical with the previous results obtained by the standard MC method [11, 12] and are not discussed here. The magnetic structure factor is given by a square of the antiferromagnetic order parameter:

$$S^{\alpha\alpha}(\mathbf{q}) = \frac{1}{N^2} \sum_{i,j} \langle S_i^\alpha S_j^\alpha \rangle e^{i\mathbf{q}(\mathbf{r}_i - \mathbf{r}_j)}, \quad (4)$$

where  $\langle \dots \rangle$  denotes a thermal average. For  $\mathbf{q} = \mathbf{Q}_3$  the structure factor exhibits a rather weak  $T$ -dependence and scales as  $1/N$  with increasing cluster size at all temperatures. This generally indicates an absence of the corresponding order in the thermodynamic limit at any  $T$ .

Results for the structure factor of the type-I antiferromagnetic ordering are presented in fig.2. Contributions from three nonequivalent wave-vectors of the type-I structure are added together. Symbols are used to distinguish different curves, while the lines are drawn through the actual data for  $N_T = 72$  replicas. For the largest cluster with  $N = 6912$  spins only  $N_T = 39$  repli-

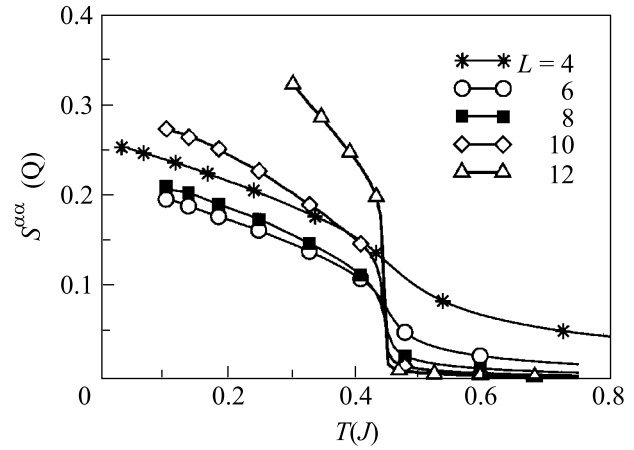


Fig.2. Temperature dependence of the structure factor corresponding to the type-I antiferromagnetic ordering

cas down to  $T = 0.3J$  have been equilibrated. The remarkable feature of the presented data is an *inverse* finite-size scaling at temperatures below  $T_c \simeq 0.45J$ : the order parameter increases with the system size. The equilibrium sublattice magnetization deduced from  $S^{\alpha\alpha}(\mathbf{q})$  is still significantly smaller than 1 even at  $T = 0.1J$ . This is a consequence of thermally excited stacking faults, domain walls and other defects in an ideal type-I antiferromagnetic structure. Linear size of such defects coincides with the lattice size. Their concentration, therefore, decreases with increasing system producing a significant increase of the order parameter. Unfortunately, the lattice sizes are still too small to observe an asymptotic thermodynamic behavior for  $S^{\alpha\alpha}(\mathbf{q})$ , though they definitely point at the spin ordering with the wave-vector of the type-I antiferromagnetic structure. The data for two large clusters exhibit a clear jump at the transition temperature indicating the first-order transition.

Measurements of  $S^{\alpha\alpha}(\mathbf{q})$  cannot distinguish between presence of three domains of a single- $q$  type-I structure, which is always the case for finite clusters, and a non-collinear triple- $q$  spin state. In order to study this aspect of the magnetic ordering in an fcc antiferromagnet we define a collinear order parameter. This is a single-site characteristics, which is insensitive to periodicity (wave-vector) of the magnetic structure but describes instead breaking of a spin-rotational symmetry. The collinear order parameter is given by a traceless second-rank tensor:

$$P^{\alpha\beta} = \frac{1}{N} \sum_i \langle S_i^\alpha S_i^\beta \rangle - \frac{1}{3} \delta^{\alpha\beta}. \quad (5)$$

It vanishes for a noncollinear triple- $q$  structure and has nonzero value for a collinear state. In the present case

$P^{\alpha\beta}$  is a secondary order parameter: it couples linearly to a square of the primary antiferromagnetic parameter. For an  $XY$  antiferromagnet on a checkerboard lattice,  $P^{\alpha\beta}$  is, however, a primary order parameter and characterizes a spin-nematic state [7].

In MC simulations, a square of the order parameter (5) is measured, which is given by

$$P_{\text{col}}^2 = \frac{1}{N^2} \sum_{i,j} \langle S_i^\alpha S_i^\beta S_j^\alpha S_j^\beta \rangle - \frac{1}{3}. \quad (6)$$

The corresponding results are shown in fig.3. Since  $P_{\text{col}}^2$  is proportional to the fourth power of an antiferromag-

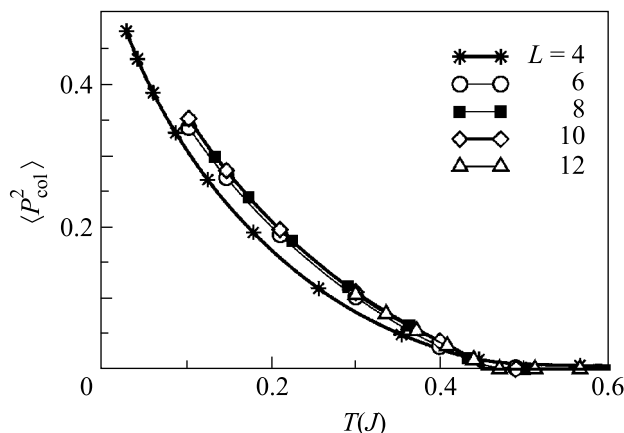


Fig.3. Temperature dependence of the collinear (nematic) order parameter

netic order parameter, it does not show an appreciable jump at the first-order transition temperature. The data for the collinear order parameter exhibit a remarkable lack of finite size scaling. This indicates that spins over the whole lattice are predominantly parallel or antiparallel to a certain direction. At  $T = 0.1J$  the aligned component of spin is  $\langle S^\alpha \rangle \approx 0.78$ . Thus, the combination of the structure factor results (fig.2) and the collinear order parameter data (fig.3) points uniquely to the collinear type-I antiferromagnetic order in a Heisenberg fcc antiferromagnet.

Finally, we present results for the energy and the order parameter histograms (distribution functions) at the transition point. Since an fcc antiferromagnet exhibits a weak first-order transition, we find it more advantageous to perform histogram collection with a single replica instead of setting up an exchange MC ensemble. Histograms have been collected utilizing a hybrid MC algorithm: 3 multiflipping Metropolis steps are combined with 11 overrelaxation (microcanonical) updates [17]. On average  $2 \cdot 10^6$  configurations have been generated to build one histogram. Its accuracy has been

checked by comparing the final distribution function to intermediate distributions obtained with proper rescaling from  $10^6$  and  $5 \cdot 10^5$  configurations. The quality of the data shown in fig.4 is significantly higher than in the

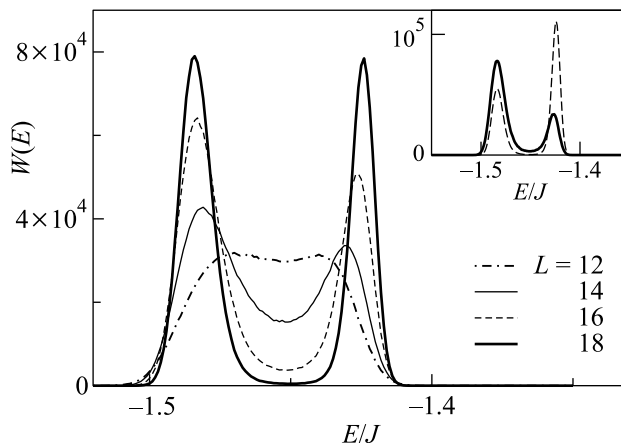


Fig.4. Distribution function (energy histogram) for several lattice sizes at the first order transition  $T_c = 0.4459J$ . Inset shows energy histograms for the largest cluster  $L = 18$  ( $N = 23328$ ) at  $T = 0.4458J$ , full line, and  $T = 0.446J$ , dashed line

previous work [12], which allows us to locate more precisely the transition point. The first-order nature of the transition is clearly observed from a double-peak structure of the energy histogram, see fig.4. Positions of the two peaks do not change significantly with growing cluster size, while probability for intermediate states rapidly drops with increasing  $L$ . At  $T_c \approx 0.4459J$ , the probability weights in the two peaks for the largest  $L = 18$  cluster ( $N = 23328$ ) differ by approximately 15%. To demonstrate a strong temperature dependence of the relative weight of two peaks we present on the inset of fig.4 the distribution functions at  $T = T_c \pm 0.0001J$ . The system clearly spends more time in the upper (lower) peak above (below) the transition temperature. We conclude, therefore, that the first-order transition in a Heisenberg fcc antiferromagnet takes place at  $T_c = 0.4459 \pm 0.0001J$ .

The distribution function for a square of the antiferromagnetic order parameter is shown in fig.5. The largest cluster also develops a double-peak structure characteristic for the first-order transition. Smaller clusters have, however, significantly wider distributions for the magnetic structure factor, which is not surprising in view of many local minima for different collinear states. It is interesting to notice that the jump of  $S^{\alpha\alpha}(Q)$  at the transition point determined from the histogram for the  $L = 18$  cluster (fig.5) is close to the jump observed for the  $L = 12$  cluster in temperature scan (fig.2), though the latter lattice does not exhibit any visible double-peak

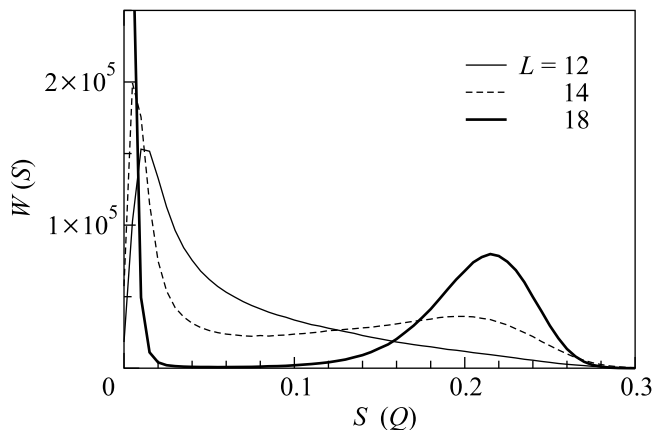


Fig.5. Histogram for the magnetic structure factor at the first order transition  $T_c = 0.4459J$

structure at  $T_c$ . This indicates that apart from a close vicinity of the transition point the  $L = 12$  cluster may be very close to the thermodynamic limit.

In conclusion, we have performed Monte Carlo simulations for a finite temperature transition in a nearest-neighbor Heisenberg fcc antiferromagnet. The obtained results clearly demonstrate a first-order transition into a collinear type-I antiferromagnetic structure due to an 'order by disorder' effect. Entropy mechanism for selection of the magnetic ordering suggests an interesting sequence of phase transitions for a weak antiferromagnetic second-neighbor exchange  $0 < J' \ll T_c$ . The higher temperature transition from a paramagnetic state is determined by thermal fluctuations and takes place into the type-I collinear antiferromagnetic structure. At low enough temperatures  $T \sim J'$  the energy selection overcomes the entropy effect and a second transition from the type-I into the type-III structure occurs.

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1. For a recent progress see Proc. of the Highly Frustrated Magnetism 2003 conference, *J. Phys.: Condens. Matter* **16**, S553–S922 (2004).
2. A. Danielian, *Phys. Rev.* **133**, A1344 (1964); K. Binder, *Phys. Rev. Lett.* **45**, 811 (1980); H. Meirovitch, *Phys. Rev.* **B30**, 2866 (1984).
3. B. C. Frazer, G. Shirane, D. E. Cox, and C. E. Olsen, *Phys. Rev.* **140**, A1448 (1965).
4. J. Faber and G. H. Lander, *Phys. Rev.* **B14**, 1151 (1976).
5. D. Bloch, R. Maury, C. Vetter, and W. B. Yelon, *Phys. Lett.* **A49**, 354 (1974).
6. C. L. Henley, *J. Appl. Phys.* **61**, 3962 (1987).
7. B. Canals and M. E. Zhitomirsky, *J. Phys.: Condens. Matter* **16**, S759 (2004).
8. S. A. Brazovskii, I. E. Dzyaloshinskii, and B. G. Kukharrenko, *Zh. Eksp. Teor. Fiz.* **70**, 2257 (1976) [*Sov. Phys. JETP* **43**, 1178 (1976)].
9. D. Mukamel and S. Krinsky, *Phys. Rev.* **B13**, 5078 (1976).
10. W. Minor and T. M. Giebultowicz, *J. de Phys. Colloq.* **49**, C8-1551 (1988).
11. H. T. Diep and H. Kawamura, *Phys. Rev.* **B40**, 7019 (1989).
12. J. L. Alonco, A. Tarançon, H. G. Ballesteros et al., *Phys. Rev.* **B53**, 2537 (1996).
13. K. Hukushima and K. Nemoto, *J. Phys. Soc. Jpn.* **65**, 1604 (1996).
14. E. Marinari, *Advances in Computer Simulations*, Eds. J. Kertész and I. Kondor, Springer, Berlin, 1998, p. 50; cond-mat/9612010.
15. H. G. Katzgraber, M. Palassini, and A. P. Young, *Phys. Rev.* **B63**, 184422 (2001); H. G. Katzgraber and A. P. Young, *Phys. Rev.* **B64**, 104426 (2001); *Phys. Rev.* **B65**, 214401 (2002).
16. L. W. Bernardi, K. Hukushima, and H. Takayama, *J. Phys.* **A32**, 1787 (1999).
17. F. R. Brown and T. J. Woch, *Phys. Rev. Lett.* **58**, 2394 (1987); M. Creutz, *Phys. Rev.* **D36**, 515 (1987).