# Ordering of the Face-Centered-Cubic Lattice with Nearest-Neighbor Interaction

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The phase diagram of an Ising fcc antiferromagnet in a field is investigated by a study of the ground state and Monte Carlo simulation of a lattice with 16 384 sites. At the critical field between the two "ordered" phases, the disordered phase is stable down to zero temperature due to "frustration" effects. The corresponding alloy phase diagram disagrees with all previous calculations. Order parameters and internal energy are briefly compared to experimental data on the Cu-Au system.

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Understanding the phase transitions of face-centered-cubic Ising lattices with antiferromagnetic nearest-neighbor exchange $J$ (Hamiltonian $\mathcal{H}=-J\sum S_i S_j - H\sum S_i$, $S_i=\pm 1$) has been a long-standing problem.[1-14] At zero magnetic field, the "ordered" phase is very degenerate[1] and has only two-dimensional long-range order,[2] vanishing at a first-order transition.[3] Nonzero fields have been studied in the context of the equivalent Ising models for binary $AB$ alloys.[4-12] Many ordered phases (or their mixtures) are stable in the ground state.[4,5] Phase-diagram calculations[6-11] ignored the degeneracy treating only three ordered phases ($A_3B$,$AB$,$AB_3$), as occur in Cu-Au.[15] The results heavily depend on the approximations (Fig. 1): while in molecular-field (MF) theory[6] all phases extend to a multicritical point, Bethe's method[10] does not predict any ordering at all. The quasi-chemical method[7] and Kikuchi's cluster variation[9-11] again yield very different answers, and it seems crucial to establish the accuracy of these approximations. There are attempts to make the model more realistic by including four-body[8,12] or next-nearest-neighbor interactions.[13] This approach is seriously hampered by the uncertainty about its accuracy. In addition, theories for Au-Cu including electronic effects[14] were compared to the Ising theories[8-11]—again the comparison is doubtful because it is unclear which features are due to the Ising model and which are due to shortcomings of the approximations.

Here I try to fill in this gap by Monte Carlo simulations.[16,17] Since there is long-range order in two-dimensional planes only, the number of sites in a plane must be large, not only the total system. I hence use lattices of 16 384 or 13 500 sites, about eight times more than the most recent related work,[3] and periodic boundary conditions. *As initial conditions we take fully aligned states of the various antiferromagnetic structures (or the ferromagnetic state).* Times of 180 to 900 Monte Carlo steps/spin are in most cases sufficient to reach equilibrium. Close to the phase transitions careful analysis of time-dependent behavior is needed to eliminate hysteresis.

The resulting Monte Carlo phase diagram is shown in Fig. 2 (the error is given by the size of

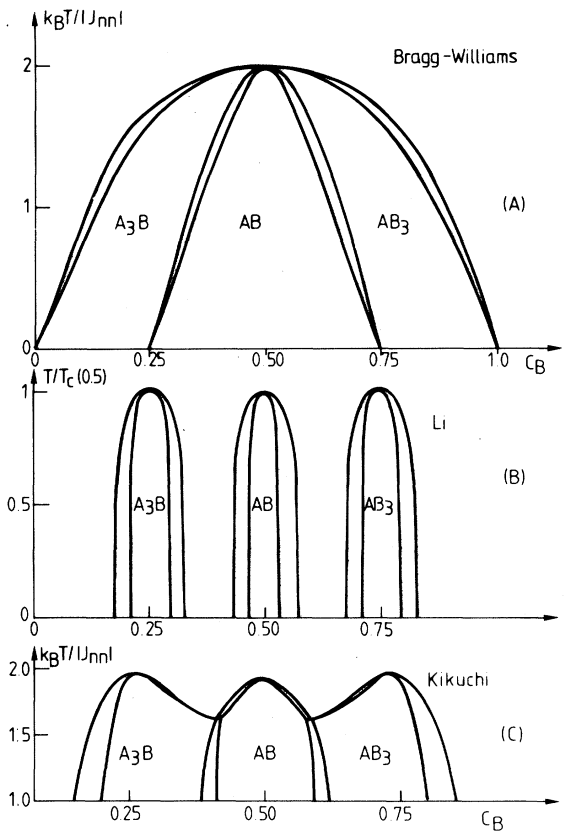


FIG. 1. Temperature-concentration phase diagram of a binary alloy $AB$ at the fcc lattice according to the Bragg-Williams approximation [(a), Ref. 6], the quasi-chemical approximation [(b), Ref. 7], and the cluster-variation (CV) method [(c), Ref. 11]. Three ordered phases ($A_3B$, $AB$, $AB_3$) are indicated.

 

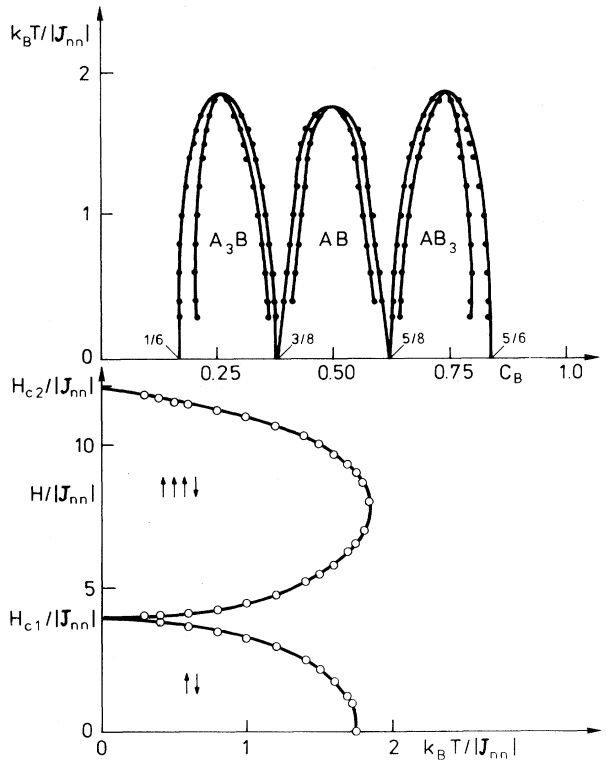


FIG. 2. Monte-Carlo phase diagram of the fcc antiferromagnet in a field (lower part) and of the corresponding alloy model (upper part).

the points or smaller). It disagrees with all previous approaches (Fig. 1) as it predicts that triple points occur (at about $c_B = \frac{3}{8}, \frac{5}{8}$) at zero temperature. The predicted critical temperatures of the first-order transitions at stoichiometric composition[18] ($A_3B, k_BT_c/|J| = 1.86$; $AB, k_BT_c/|J| = 1.76$) are a few percent less than those of the cluster variation[11] ($AB, k_BT_c/|J| = 1.89$) but distinctly higher than that of Ref. 7 ($AB, k_BT_c/|J| = 1.46$). For Cu-Au the phase diagram has the topology of Fig. 1(c), but is not symmetric around $c_B = 0.5$. Such an asymmetry is expected, since due to lattice expansion the effective interaction $J$ must depend both on $T$ and on $c_B$.[14,19] Hence from Fig. 1(c) one could conclude that the effective interaction responsible for the ordering is nearest-neighbor, and this work was used to compare with theories including electronic effects.[14] From the more accurate Fig. 2, however, I conclude that a $J$ depending on $c_B$ and $T$ can only distort the diagram but cannot change its topology, and hence interactions to more distant neighbors are needed to reproduce the Au-Cu phase diagram. This conclusion agrees with the MF analysis of diffuse scattering in the disordered phase.[20] Deducing interaction constants by using MF theory is inaccurate also, since the MF phase diagram fails [Fig. 1(a)]. Hence a comparison of the experiments to more accurate Monte Carlo results on diffuse scattering is needed and will be given elsewhere, for more realistic interactions.[21]

Apart from the structures shown in Figs. 1 and 2 other stoichiometric arrangements are ground-state structures, like an $A_2B$ structure.[4,5] For the temperatures studied ($k_BT/|J| \geq 0.3$) the $A_2B$ structure is unstable, however. I suggest that this structure (as well as $A_4B$, $A_5B$, $A_5B_3$, etc.[4,5]) does not occur for $T > 0$: Consider the magnetic representation, where the ground-state energies are $U_{AF}/|J| = -2$ (independent of $H$, corresponds to $AB$), $U_5/|J| = -2H/3|J| + 2$ ($A_5B$), $U_4/|J| = -3H/5|J| + \frac{6}{5}$ ($A_4B$), $U_3/|J| = -H/2|J|$ ($A_3B$), $U_2/|J| = -H/3|J| - \frac{2}{3}$ ($A_2B$), etc. While the antiferromagnet is the ground state for $0 \leq H \leq H_{c1} = 4|J|$, the structure with one sublattice down spins, three sublattice up spins is the ground state for $H_{c1} \leq H \leq H_{c2} = 12|J|$, from where on the ground state is ferromagnetic. The structures corresponding to $A_5B$, $A_4B$ are ground states only right at $H_{c2}$, $A_2B$, $A_5B_3$ at $H_{c1}$. At these "multiphase points"[22] the degeneracy is much higher than in the $AB$ and $A_3B$ phases. In the $A_3B$ phase, antiferromagnetic and ferromagnetic square lattices alternate, with no correlations between spins in different antiferromagnetic planes. The ground-state entropy is zero, just as in the antiferromagnetic case.[2] At the critical fields, however, the $T = 0$ entropy is finite: Consider, e.g., at $H_{c1}$ two neighboring fcc cells, with down spins at the corners and up spins otherwise. There is no energy cost for overturning the spin at the center of the square joining the cells. As this spin does not interact with any spins outside the two cells, the number of "loose spins" is proportional to the system size. The antiferromagnetic planes loose their order, their average magnetization/spin is $M_{AF} = -\frac{1}{2}$, implying a total magnetization of $M = \frac{1}{4}$, i.e., $c_B = \frac{3}{8}$ as indicated by the simulation. Clearly, it is interesting to improve this crude argument by studying the spin correlations in this highly "frustrated" model. Understanding such periodic systems with compensating interactions has become a major tool for spin-glass theories.[23]

This "frustration effect" is not just academic but also shows up in the short-range order seen at finite temperatures, in Fig. 3. Already at $T > T_c(c_B)$ a shallow minimum develops in the Cowley[24] parameter $|\alpha_1|$, while $|\alpha_1|$ increases at the

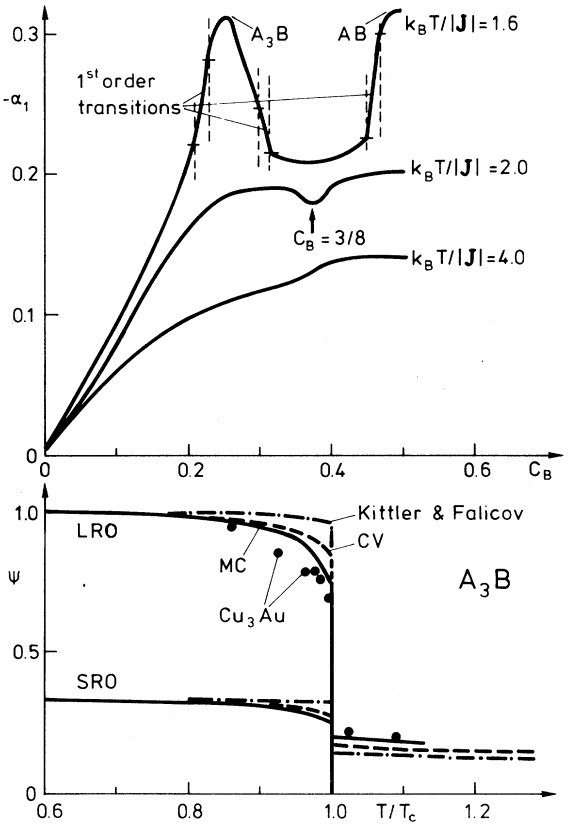


FIG. 3. Variation of the Cowley short-range order (SRO) parameter $\alpha_1$ with concentration (upper part) and plot of $\alpha_1$ and long-range order parameter (LRO) $\psi$ vs temperature at $c_B=\frac{1}{4}$ (lower part). Full curves are the Monte Carlo (MC) results; broken curves, the CV method (Ref. 9); dash-dotted curve, the Kittler-Falicov theory (Ref. 14); dots, data for $AuCu_3$ (Ref. 25).

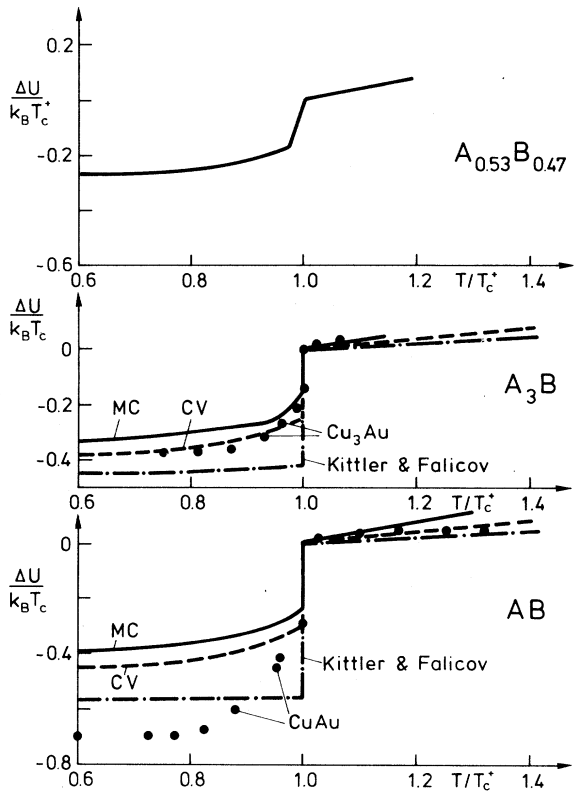


FIG. 4. Internal energy plotted vs temperature for three compositions. In the stoichiometric cases, previous theories (Refs. 9, 14) and Cu-Au data (Ref. 26) are included (notation as in Fig. 3).

ordering compositions. It is also seen that for $c_B=\frac{1}{4}$ the Monte Carlo data agree slightly better with $Cu_3Au$ data[25] than previous work.[9,14] In view of the very different phase diagram (Fig. 2) this agreement anyhow is surprising. Figure 4 shows that the ordering energy $\Delta U$ deviates distinctly from the data,[26] as expected, since a nearest-neighbor model should not be sufficient. Figures 2 and 3 suggest that a study of the ordering at off-stoichiometric compositions should be a more stringent tool to check the effects of interaction parameters, as well as approximative theories.

In conclusion, I found serious deficiencies in previous studies of ordering on fcc lattices like the cluster-variation method, which did not properly take care of "frustration" effects; forces of longer range than just nearest neighbor are needed to describe Cu-Au alloys.

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# Roughening Transition in the $^4$He Solid-Superfluid Interface

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Principal planes of the $^4$He solid-superfluid interface are expected to undergo roughening transitions at temperatures of about 1 K. An experiment is described in which two such transitions were observed in the hcp-superfluid interface: first for the basal plane at 1.08 K and second for an orthogonal face at 0.85 K.

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The roughening transition (RT) is a theoretical concept, known to apply to certain Ising models,[1] and to computer simulations of crystal growth.[2,3] In this Letter we discuss the applicability of the RT to a real physical system: the $^4$He solid-superfluid interface. We report the observation of morphological transitions in $^4$He crystals at 1.08 and 0.85 K which, we argue, are RT's. Parshin and his co-workers[4,5] have argued that the interface in this system is always rough, but their view is contrary to evidence presented here (and by Landau *et al*[6]). Balibar[7] has independently proposed the existence of a RT in the system.

The roughening temperature, $T_R$, is characterized by the vanishing of the step energy.[1,2] Above $T_R$ the interface fluctuates macroscopically, and in the thermodynamic limit, translation invariance is restored.[8] Unfortunately, this aspect of the RT is not expected to be observable in practice. Fluctuations in ordinary macroscopic systems are microscopic; for the two-dimensional interface, the mean square amplitude fluctuation is of order $\ln N$, where $N$ is the number of lattice points in the interface.

The interface is modeled by a two-dimensional (2D) lattice Hamiltonian with discrete, unbounded Ising spin $Z$: the vertical coordinate of the interface. A capillary-wave Hamiltonian[9] for the interface is

$$H = \sum_{\vec{l}} \{1 + [\nabla Z(\vec{l})]^2\}^{1/2} [\sigma_0 \Omega + (\rho_0 \hbar^2 / M^2)(\nabla \psi)^2]. \quad (1)$$

Here, $\psi$ is the superfluid field, $\rho_0$ its density, $\nabla$ the discrete gradient, $\Omega$ the area of the 2D unit cell, and $\vec{l}$ a lattice bond. The term proportional to $\sigma_0$ (a coupling constant) represents the energy density of the boundary of the solid, and the term proportional to $\hbar^2$ is the energy density of the superfluid layer near the interface.[10,11] In the lowest approximation the $\psi$ field integrates out. Similar conclusions hold for the Villain model[12] and for other models[13] of the interface. The surface tension is the free energy of the Hamiltonian $H$.

The specific choice of the interface Hamiltonian is of little importance here since the Kosterlitz-Thouless roughening transition[14] is a general feature of 2D models whose Hamiltonians are invari-

 