

Exact Coexistence-Curve Diameter of a Lattice Gas with Pair and Triplet Interactions

In a recent Letter,¹ Goldstein *et al.* presented experimental and theoretical evidence that the pair-potential model of fluids is insufficient in the critical region. Their analysis of experimental data indicates that the critical behavior of the coexistence-curve diameter $\rho_d \equiv (\rho_l + \rho_v)/2$, where ρ_l and ρ_v are the respective coexisting liquid and vapor densities, is the same as that of the molecular polarizability a_p . They also showed that this anomalous critical behavior indeed occurs in a lattice gas when there are triplet interactions. However, their analysis of the lattice gas is based on a thermodynamic perturbative argument and assumes small triplet interactions. In this Comment we obtain the exact coexistence-curve diameter of a lattice gas with finite pair and triplet interactions. Our result, which holds quite generally for *any* interaction strengths and at *all* temperatures, corroborates the conclusion of Goldstein *et al.* that $\rho_d \sim a_p$ in the critical region.

Consider a Kagomé lattice gas of N sites with reduced nearest-neighbor interactions $-J$ and triplet interactions $-J_3$, the latter existing between three sites surrounding a triangular face of the lattice. The Kagomé lattice is the covering lattice of a honeycomb lattice whose sites reside in the triangular faces of the Kagomé lattice. Using this picture and regarding a honeycomb lattice edge as being covered by a bond if, and only if, the Kagomé lattice site residing on it is occupied by a lattice-gas molecule, the Kagomé lattice gas then describes precisely an eight-vertex model on the honeycomb lattice.² The vertex model has the vertex weights, in the notation of Ref. 2, $a=1$, $b=\sqrt{z}$, $c=xz$, $d=x^3yz^{3/2}$, where $x \equiv e^J$, $y \equiv e^{J_3}$, and z is the fugacity of the lattice gas.

It has been established^{2,3} that the eight-vertex model on the honeycomb lattice is completely equivalent to a honeycomb Ising model with nearest-neighbor interactions K and a nonzero magnetic field L . This leads to the relation

$$\Xi^{\text{Kag}}(z, J, J_3) = F^N Z^{\text{HC}}(L, K) \quad (1)$$

and a complete determination of the critical surface of the lattice gas.⁴ Here Ξ^{Kag} is the lattice-gas grand partition function, Z^{HC} is the Ising partition function, and F , L , and K , are analytic functions of a , b , c , and d whose explicit expressions have been given previously.²⁻⁴ Particularly, one finds

$$e^{4K} = 1 + x^2 z \frac{(x^2 y - 1)^2 - 4(xy - 1)(x - 1)}{[x^2 z(xy - 1) + x - 1]^2}, \quad (2)$$

and that the locus $L=0$ leads to

$$1 - x^3 y + x^6 y z^3 (x^3 y^2 - 1) + 3(1 + xz + x^4 y z^2) \times [x^2 z(1 - xy) + x - 1] = 0. \quad (3)$$

Along the coexistence curve of the lattice gas $L=0$, the Ising model possesses a spontaneous magnetization $\pm I_0$. The density of the lattice gas,

$$\rho \equiv z \frac{\partial}{\partial z} \left[\frac{1}{N} \ln \Xi^{\text{Kag}}(z, J, J_3) \right], \quad (4)$$

then splits into two branches, ρ_l and ρ_g , depending upon the sign of the spontaneous magnetization. It follows at once that

$$\rho_d = z \frac{\partial F}{\partial z} + z \frac{\partial K}{\partial z} \langle \sigma_i \sigma_j \rangle, \quad (5)$$

where z is to be eliminated using (3), and $\langle \sigma_i \sigma_j \rangle$ is the nearest-neighbor correlation of the zero-field honeycomb Ising model which is known.

The expression (5) now gives the exact coexistence-curve diameter for all temperatures and any strength of the interactions J and J_3 . Since F and K are analytic functions, (5) implies that the singular behavior of ρ_d near the critical temperature T_c of the lattice gas is the same as that of the lattice gas is the same as that of $\langle \sigma_i \sigma_j \rangle$, namely, $|T - T_c|^{1-a}$. This conclusion, which is now established quite generally for arbitrary interaction strengths, corroborates the finding of Ref. 1 for small J_3 . Furthermore, using (2) and (3), one finds $\partial K / \partial z = 0$ when $J_3 = 0$, $L = 0$ ($y = 1$, $z = x^{-2}$). As a consequence, the critical anomaly of $\rho_d \sim |T - T_c|^{1-a}$ occurs only when there is a nonzero triplet interaction.

This work was supported in part by NSF Grant No. DMR-8702596.

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Received 24 April 1989

PACS numbers: 64.60.Fr, 05.70.Jk, 61.20.Ne, 64.70.Fx

¹R. E. Goldstein, A. Parola, N. W. Ashcroft, M. W. Pestak, M. H. W. Chan, J. R. de Bruyn, and D. A. Balzarini, *Phys. Rev. Lett.* **58**, 41 (1987).

²F. Y. Wu, *J. Math. Phys.* **15**, 687 (1974).

³X. N. Wu and F. Y. Wu, *J. Stat. Phys.* **90**, 41 (1988).

⁴X. N. Wu and F. Y. Wu, *J. Phys. A* (to be published).