

## Dense Two-Dimensional Classical Coulomb Gas on a Triangular Lattice

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We carry out Monte Carlo simulations of the two-dimensional neutral Coulomb gas of integer charges on a triangular lattice, as a function of temperature and chemical potential, and find rich critical behavior. We find transitions which do not seem to follow the expectations of a symmetry analysis, and nonuniversal jumps in the inverse dielectric constant.

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Systems of classical, logarithmically interacting charges have had wide application in two-dimensional statistical mechanics and related physical systems such as superfluid and superconducting films, Josephson-junction arrays, 2D melting, surface roughening, and liquid crystals.<sup>1</sup> It is thus of general interest to study the transitions of such 2D Coulomb systems. A question of particular interest is whether or not one may use the universality classes of well-known short-range interaction models to classify the transitions of Coulomb systems with the same symmetry.<sup>2</sup> That such a program might be successful was suggested by the fully frustrated  $XY$  model which maps onto a neutral Coulomb gas of half-integer charges. Here the ground state has a discrete double degeneracy, and the model was found to have an Ising transition.<sup>3</sup>

Recently we studied the dense neutral-integer Coulomb gas (CG) on a square lattice.<sup>4</sup> This has the same symmetry as an antiferromagnetic Blume-Emery-Griffiths model. As expected by symmetry, the second-order line in this Coulomb model is Ising. However, we found that the tricritical point, where the Ising and first-order lines meet, did not have the behavior of the usual Ising tricritical point. In this paper we consider the dense neutral-integer Coulomb gas on a triangular lattice. Here we find clearer evidence of a transition which does not appear to follow the critical behavior expected from a symmetry analysis. An explicit physical example of one of the phases we find is given by the fully frustrated Josephson-junction array on a honeycomb lattice.

The Hamiltonian we study is given by

$$\mathcal{H} = \frac{1}{2} \sum_{i,j} q_i V'(\mathbf{r}_i - \mathbf{r}_j) q_j - u \sum_i q_i^2 + \sum_i (q_i^4 - q_i^2). \quad (1)$$

The first term is the ordinary Coulomb gas; the sum is over all pairs of sites of an  $L \times L$  triangular lattice,  $q_i = 0, \pm 1, \pm 2, \dots$  is the integer charge at site  $i$ , and neutrality  $\sum_i q_i$  is imposed.  $V'(\mathbf{r}) \equiv V(\mathbf{r}) - V(0)$  is the 2D Coulomb potential with singularity removed, where  $V(\mathbf{r})$  solves  $D_{ij} V(\mathbf{r}_j - \mathbf{r}_m) = -2\pi\delta_{im}$  with  $D_{ij}$  the lattice Laplacian. With periodic boundary conditions,  $V'$  is given explicitly by Fourier transform,

$$V'(\mathbf{r}) = (1/N) \sum_{\mathbf{k}} V_{\mathbf{k}} [e^{i\mathbf{k} \cdot \mathbf{r}} - 1],$$

with

$$V_{\mathbf{k}} = \frac{3\pi}{6 - 2\cos(\mathbf{k} \cdot \hat{\mathbf{a}}_1) - 2\cos(\mathbf{k} \cdot \hat{\mathbf{a}}_2) - 2\cos(\mathbf{k} \cdot \hat{\mathbf{a}}_3)}, \quad (2)$$

$N = L^2$ ,  $\hat{\mathbf{a}}_1 = \hat{\mathbf{x}}$  and  $\hat{\mathbf{a}}_2 = \frac{1}{2}\hat{\mathbf{x}} + \frac{1}{2}\sqrt{3}\hat{\mathbf{y}}$  are the unit basis vectors of the triangular lattice,  $\hat{\mathbf{a}}_3 = \hat{\mathbf{a}}_1 - \hat{\mathbf{a}}_2$ , and the sum is over all  $\mathbf{k}$  in the first Brillouin zone (BZ), consistent with periodic boundary conditions. The second term in Eq. (1) controls the average charge density; the third term stabilizes the system in the very dense limit.

A summary of our results is given by the  $u$ - $T$  phase diagram shown in Fig. 1. In the insulating phase  $\mathcal{A}$ , the ground state is the vacuum and charges exist in bound neutral pairs. Crossing from  $\mathcal{A}$  to a conducting phase  $\mathcal{B}$ , there is a Kosterlitz-Thouless (KT) transition<sup>5</sup> (dashed line) determined by the vanishing of the inverse dielectric constant  $\epsilon^{-1}$ .

The phase  $\mathcal{D}$  is described by a periodic charge structure, with complex order parameter given by the Fourier component of the average charge density,  $\langle q_{\mathbf{k}_0} \rangle = (1/L) \langle \sum_i q_i \exp(i\mathbf{k}_0 \cdot \mathbf{r}_i) \rangle$ , where  $\mathbf{k}_0 = (4\pi/3)\hat{\mathbf{a}}_1$  is a wave vector which points to a vertex of the surface of the first

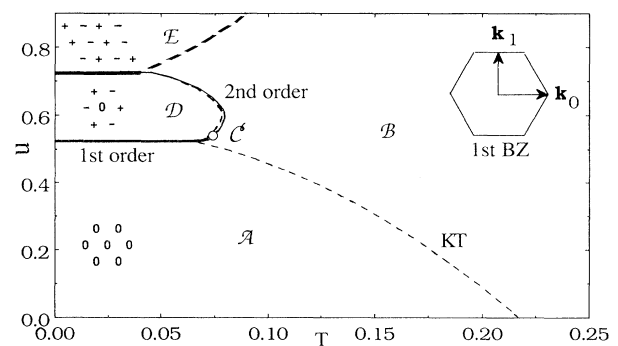


FIG. 1. Phase diagram for the classical neutral-integer-charge 2D Coulomb gas on a triangular lattice, as a function of temperature  $T$  and  $u$ . As  $u$  increases, the average charge density increases. The thin dashed lines are where the inverse dielectric function  $\epsilon^{-1}$  jumps to zero. The thick solid lines are first-order transitions. The thin solid line is a second-order transition. The thick dashed line denotes a crossover region as opposed to a true thermodynamic transition.

BZ,<sup>6</sup> and is shown in the inset to phase  $\mathcal{B}$  in Fig. 1. At  $T=0$ , the ground-state charge configuration has an average  $\langle q_i^2 \rangle = \frac{2}{3}$  and is shown as the inset to phase  $\mathcal{D}$  in Fig. 1. The ground state is sixfold degenerate corresponding to rotations in the phase of  $q_{k_0}$  by  $\pi/3$ .  $\mathcal{D}$  is separated from phases  $\mathcal{A}$  and  $\mathcal{B}$  by a first-order (heavy solid) line which meets a second-order (light solid) line at a point  $\mathcal{C}$ . The inverse dielectric function  $\epsilon^{-1}$  in phase  $\mathcal{D}$  vanishes with a nonuniversal jump (dashed line), at a temperature slightly lower than the second-order line. Both this line and the KT line between phases  $\mathcal{A}$  and  $\mathcal{B}$  meet the first-order line below the point  $\mathcal{C}$ .

Phase  $\mathcal{E}$  is described by a ground state with  $\langle q_i^2 \rangle = 1$ . It is ordered in one of the three directions  $\hat{\mathbf{a}}_i$ , with charges of alternating sign, and is random in the other directions. The degeneracy is therefore  $2^L$ . A sample ground state is shown as the inset to  $\mathcal{E}$  in Fig. 1. We argue below that this ground-state order is lost at any finite  $T$ , and that  $\mathcal{E}$  is separated from the disordered phase  $\mathcal{B}$  by a crossover region (heavy dashed line) rather than a sharp thermodynamic transition.  $\mathcal{E}$  is separated from phase  $\mathcal{D}$  by a first-order (heavy solid) line.

The locations of the two first-order lines at  $T=0$  are easily explained. Taking the Fourier transform of the first two terms in the Hamiltonian (1) gives  $\mathcal{H} = \sum_{\mathbf{k}} (\frac{1}{2} V_{\mathbf{k}} - u) q_{\mathbf{k}} q_{-\mathbf{k}}$ . When  $u$  becomes greater than  $\min_{\mathbf{k}} [\frac{1}{2} V_{\mathbf{k}}]$ , the system will order in a state with nonzero  $\langle q_{\mathbf{k}} \rangle$ . From Eq. (2) we find that  $\min[V_{\mathbf{k}}] = \pi/3$  at  $\mathbf{k} = \mathbf{k}_0$ . There is thus a discontinuous change in ground state from the vacuum to phase  $\mathcal{D}$  at  $u_{c1} = \pi/6$ . Since the average charge density  $\langle q_i^2 \rangle = \frac{2}{3}$  in  $\mathcal{D}$ , then for  $u > u_{c1}$ , the ground-state energy density decreases as  $(\frac{1}{2} \pi/3 - u) \times \frac{2}{3}$ . As  $u$  increases, eventually the system prefers a higher-density phase with  $\langle q_i^2 \rangle = 1$ . The most symmetric such state would have periodicity given by a wave vector pointing to the center of one of the faces of the first BZ, for example,  $\mathbf{k}_1 = (2\pi/\sqrt{3})\hat{\mathbf{y}}$  (see inset in Fig. 1). However, from Eq. (2) it is seen that for *any*  $\mathbf{k}$  such that  $\mathbf{k} \cdot \hat{\mathbf{a}}_i = \pm \pi$ ,  $V_{\mathbf{k}} = 3\pi/8$  is constant. The resulting phase  $\mathcal{E}$  is thus ordered with alternating  $\pm 1$  charges in one direction  $\hat{\mathbf{a}}_i$ , and is completely random in the other directions. The energy of this state is  $(\frac{1}{2} 3\pi/8 - u) \times 1$ , and it becomes the ground state with lower energy than phase  $\mathcal{D}$  for  $u > u_{c2} = 11\pi/48$ . At  $T=0$ ,  $u = u_{c2}$ , we find numerically that there is a finite surface tension,  $\approx 0.05$ , between the ground states of  $\mathcal{D}$  and  $\mathcal{E}$ .

To map out the rest of the  $u$ - $T$  phase diagram we have used Monte Carlo (MC) simulations of the Hamiltonian [(1) and (2)]. A nearest-neighbor pair of sites is selected randomly at each step of the simulation, and a unit charge is added to one site and subtracted from the other. This change is accepted or rejected according to the standard Metropolis algorithm. We refer to  $N=L^2$  steps as 1 pass. An initial 10000 passes were discarded for equilibration, with 200000 passes used to compute averages. Error bars are estimated by making five in-

dependent runs.

We first consider the second-order transition between phases  $\mathcal{D}$  and  $\mathcal{B}$ . This transition line has been located by observing, at fixed  $u$ , the temperature at which the specific heat  $C$  and order-parameter susceptibility  $\chi$  have their peak. Domany *et al.*<sup>7</sup> have carried out a symmetry analysis of possible order-disorder transitions in 2D adsorbed monolayers. The symmetry of phase  $\mathcal{D}$ , in their classification scheme, is denoted  $(\sqrt{3} \times \sqrt{3})R30$  with particle-hole symmetry ( $q_i \leftrightarrow -q_i$ ) and is expected to be in the universality class of the  $XY$  model with sixfold anisotropy. This model has an intermediate phase with algebraic order and divergent  $\chi$ , which is separated by KT transitions from a low- $T$  ordered and high- $T$  disordered phase.<sup>8</sup> In this case, the specific heat should have only a finite cusp. In contrast, we find in our case evidence for a diverging  $C$  at a single transition.<sup>9</sup> Assuming the usual scaling relation

$$C(t, L) \sim L^{a/\nu} \Phi(tL^{1/\nu}), \quad (3)$$

where  $t \equiv (T - T_c)/T_c$ , we use the method of Barber and Selke<sup>10</sup> and plot the ratio  $R \equiv C(T, L)/C(T, L')$  versus  $T$  for different values of  $L$  and  $L'$ , but the same ratio  $L/L'$ . The curves should intersect at the single point  $T_c$ , with  $R(T_c) = (L/L')^{a/\nu}$  determining the critical exponent. In Fig. 2(a) we plot  $R$  vs  $T$  for sizes  $L:L' = 12:6, 18:9,$  and  $24:12$  at  $u = 0.63$ . We find scaling in agreement with Eq. (3), with  $T_c = 0.078$ ,  $R_c = 2.0 \pm 0.2$ , and hence  $a/\nu = 1.00 \pm 0.14$ . Similar plots at  $u = 0.60$  and  $0.66$  give values  $a/\nu = 0.97 \pm 0.14$  and  $1.10 \pm 0.24$ . At  $u = 0.54$  however, where the second-order line meets the first-order line at point  $\mathcal{C}$ , the ratio analysis shown in Fig. 2(b) gives a different exponent  $a/\nu = 1.51 \pm 0.15$ . One possible explanation is that  $\mathcal{C}$  is a multicritical point. At the point where the second-order line meets the first-order line which separates  $\mathcal{D}$  and  $\mathcal{E}$ , we have observed

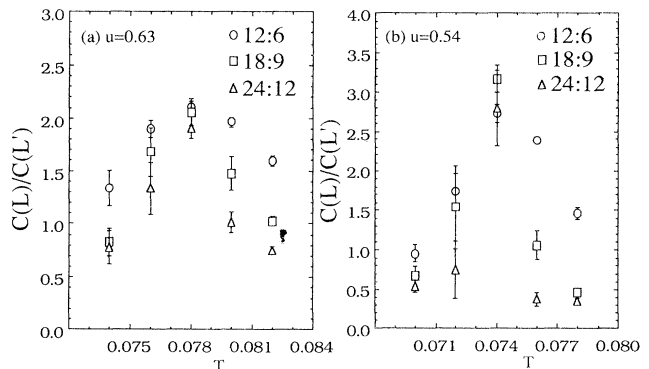


FIG. 2. Ratio of specific heats  $C(T, L)/C(T, L')$  vs  $T$  at (a)  $u = 0.63$  and (b)  $u = 0.54$ , in phase  $\mathcal{D}$ . Three different sets of lattice lengths  $L:L'$  with equal ratio 2 are shown. The common intersection of the three curves is at  $(T_c, (L/L')^{a/\nu})$ .  $10^6$  MC passes were used.

very strong fluctuations, and the situation remains unclear. We thus have evidence suggesting that the symmetry analysis fails to give the correct transition of the CG.

We next consider phase  $\mathcal{E}$ . At  $T=0$ , every site contains a  $\pm 1$  charge, and so  $\mathcal{E}$  has the symmetry of the Ising antiferromagnet, which on a triangular lattice<sup>11</sup> has  $T_c=0$  and a finite  $T=0$  entropy density. In the CG, however, the long-range interactions lift some of the frustration which characterizes the short-range Ising model, and the ground states are ordered antiferromagnetically along one of the basis directions  $\hat{\mathbf{a}}_i$ . The randomness allowed in the other basis direction gives a ground-state entropy density of  $\ln(2^L)/L^2$  which vanishes as  $L \rightarrow \infty$ . In Figs. 3(a)–3(c) we show specific heat  $C$ , inverse dielectric constant  $\epsilon^{-1}$ , and average charge density  $\langle q^2 \rangle$  versus  $T$  for  $u=0.8 > u_{c2}$ . While these numerical results suggest a possible finite  $T_c$ , we now argue that  $T_c=0$ . In Fig. 3(d) we show two types of domain excitations of a ground state. We have numerically computed the excitation energy  $\Delta E$  of such domains of length  $l$  in systems of length  $L \leq 90$ . We find that for  $l \gtrsim 10$ ,  $\Delta E$  approaches a finite constant,  $\Delta E \approx 0.79 + 2(u - u_{c2})$  and 4.9, for domains (i) and (ii), respectively, independent of which ground state is chosen as the background. Thus once a large enough domain is created, it costs no further energy to increase in size, and give a transition to a different ground state. The finite probability to excite such domains at any  $T > 0$  disorders the ground state and drives  $T_c \rightarrow 0$ , just as excitation of domain walls disorders the 1D Ising model. It is possible that other excitations exist which give lower energy barriers between the ground states than those shown in Fig. 3(d). The apparent transition seen in Figs. 3(a) and 3(c) is just a crossover region where such excitations proliferate. Figure 3(b) shows that these excitations also

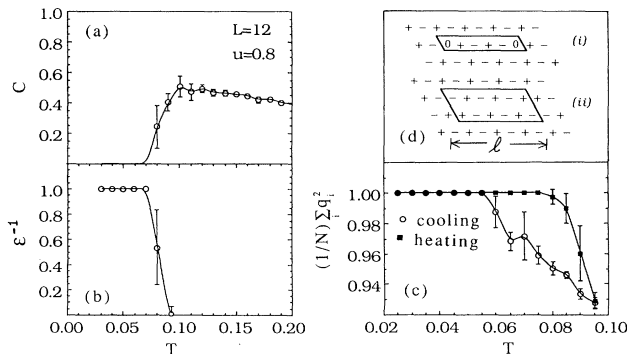


FIG. 3. (a) Specific heat  $C$ , (b) inverse dielectric function  $\epsilon^{-1}$ , and (c) average charge density  $\langle q^2 \rangle$  vs  $T$ , at  $u=0.8$  in phase  $\mathcal{E}$ . 60000 MC passes on an  $L=12$  lattice were used. (d) Two types of domain excitations which disorder the ground state at  $T > 0$ . As  $l \rightarrow \infty$ , their excitation energy remains finite.

appear to drive  $\epsilon^{-1} \rightarrow 0$ . The  $T$  where  $\epsilon^{-1} \rightarrow 0$  does show a slight downwards shift as we increase either the lattice size or number of MC steps in the simulation.

The lattice CG is a representation for vortices in the XY model on the dual lattice.<sup>5,8</sup> A physical realization close to  $\mathcal{E}$  is given by the fully frustrated ( $\frac{1}{2}$  flux quantum per unit cell) Josephson-junction array on a honeycomb lattice.<sup>3,12,13</sup> In this mapping, the only allowed charges are  $q_i = \pm \frac{1}{2}$ ; however, the ground-state order and degeneracy remain the same as in  $\mathcal{E}$ . Since vacancies  $q_i = 0$  are not permitted, we expect that in this model the ground state is disordered at any  $T > 0$  by excitations like (ii) of Fig. 3(d). Direct MC calculations by Shih and Stroud<sup>12</sup> of specific heat  $C$  and helicity modulus ( $\epsilon^{-1}$ ) in this XY model bear a striking resemblance to our Figs. 3(a) and 3(b). We have carried out independent simulations of the CG with  $q_i = \pm \frac{1}{2}$  and have found similar results.

Finally, we consider the “metal-insulating” transitions where the inverse dielectric function<sup>14</sup>

$$\epsilon^{-1} = \lim_{k \rightarrow 0} \left[ 1 - \frac{2\pi}{Tk^2} \langle q_k q_{-k} \rangle \right] \quad (4)$$

vanishes. The Kosterlitz-Thouless stability argument<sup>3,5</sup> requires that  $\epsilon^{-1}$  jumps discontinuously to zero at  $T_c$  with a jump bounded by  $\epsilon^{-1}(T_c) \geq 4T_c$ . Evaluating Eq. (4) at the smallest  $k = 4\pi/\sqrt{3}L$  in the finite lattice, and applying this bound, gives an upper bound on  $T_c$ , and locates the dashed lines in Fig. 1. In Fig. 4 we plot  $\epsilon^{-1}(T, L)$  vs  $T$  for various sizes  $L$ . Figure 4(a) is for  $u=0.5$  in phase  $\mathcal{A}$  just below the first-order line separating  $\mathcal{A}$  from  $\mathcal{D}$ ; Fig. 4(b) is for  $u=0.63$  within phase  $\mathcal{D}$ . A rough estimate of  $T_c$  is given by the  $T$  at which the curves for different sizes  $L$  intersect.<sup>4</sup> For  $u=0.5$ , this estimate is consistent with the KT bound satisfied as an

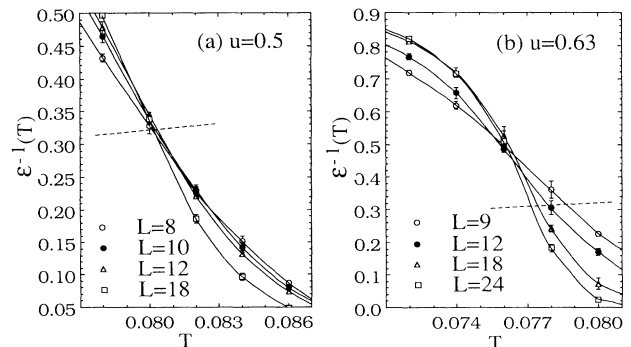


FIG. 4. Inverse dielectric function  $\epsilon^{-1}(T, L)$  vs  $T$  for (a)  $u=0.5$  (just below the first-order line) in  $\mathcal{A}$ , and (b)  $u=0.63$  in  $\mathcal{D}$ , for various lattice sizes  $L$ . The common intersection of the curves for different  $L$  gives an estimate for  $T_c$ . Intersection with the dashed line  $4T$  gives the Kosterlitz-Thouless bound on  $T_c$ .  $10^6$  MC passes were used.

equality, i.e., we find the universal jump of the ordinary dilute ( $u=0$ ) CG.<sup>14</sup> However, at  $u=0.63$  we find a larger than universal jump. Similar nonuniversal jumps are found at other  $u$  in  $\mathcal{D}$ . These conclusions are born out in more detailed finite-size scaling fits which we have performed, following the approach of Weber and Minnhagen.<sup>15</sup> Comparison of Fig. 4(b) and Fig. 2(a) suggests that in phase  $\mathcal{D}$ ,  $\epsilon^{-1}$  vanishes at a  $T'_c$  slightly lower than the  $T_c$  where the order parameter  $\langle q_{k_0} \rangle$  vanishes. While the  $\epsilon^{-1} \rightarrow 0$  transition in  $\mathcal{D}$  is presumably affected strongly by the presence of the lattice-induced ordered charge structure, in  $\mathcal{A}$  it remains driven by the pair-unbinding KT mechanism.<sup>5</sup> Recently, Minnhagen,<sup>16</sup> based on an extension of the KT recursion relations for the continuum CG, argued that larger than universal jumps in  $\epsilon^{-1}$  should occur whenever the system is dense enough that  $T_c < T^* \approx 0.14$ . For  $u=0.5$  we find  $T_c \approx 0.08$ , well below  $T^*$ ; however, we continue to find the universal jump of the dilute model.<sup>17</sup>

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<sup>1</sup>B. I. Halperin, in *Physics of Low-Dimensional Systems*, Proceedings of the Kyoto Summer Institute, edited by Y. Nagaoka and S. Hikami (Publication Office, Prog. Theor. Phys., Kyoto, 1979), p. 53; J. M. Kosterlitz and D. J. Thouless, *Prog. Low Temp. Phys. B* **7**, 371 (1978); P. Minnhagen, *Rev. Mod. Phys.* **59**, 1001 (1987).

<sup>2</sup>While power-law interactions in *ferromagnetic* spin models are known to be able to affect critical exponents [see A. Aharony, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1976), Vol. 6, p. 358], the situation is less clear for models which order at a finite wave vector  $\mathbf{k}_0$ , when the Fourier transform of the interaction is analytic about  $\mathbf{k}_0$ . Such is the situation in the Coulomb gas.

<sup>3</sup>S. Teitel and C. Jayaprakash, *Phys. Rev. B* **27**, 598 (1983); *Phys. Rev. Lett.* **51**, 1999 (1983).

<sup>4</sup>J.-R. Lee and S. Teitel, *Phys. Rev. Lett.* **64**, 1483 (1990).

<sup>5</sup>J. M. Kosterlitz and D. Thouless, *J. Phys. C* **6**, 1181 (1973); J. M. Kosterlitz, *J. Phys. C* **7**, 1046 (1974).

<sup>6</sup>Since the  $\mathbf{k}'$  which point to the other vertices of the first BZ

are all related to  $\mathbf{k}_0$  by inversion symmetry or translation by a reciprocal-lattice vector, the Fourier components  $q_{\mathbf{k}'}$  are all equal either to  $q_{\mathbf{k}_0}$  or to  $q_{\mathbf{k}_0}^*$ . This ensures that the order parameter has only two components.

<sup>7</sup>E. Domany, M. Schick, J. S. Walker, and R. B. Griffiths, *Phys. Rev. B* **18**, 2209 (1978).

<sup>8</sup>J. V. José, L. P. Kadanoff, S. Kirkpatrick, and D. R. Nelson, *Phys. Rev. B* **16**, 1217 (1977).

<sup>9</sup>A general symmetric six-state clock model,  $\mathcal{H} = -\sum_{\langle i,j \rangle} V(|\theta_i - \theta_j|)$  with angles  $\theta_i = (\pi/3)m$ ,  $m=0, \dots, 5$ , is characterized by three coupling constants  $V(\pi/3)$ ,  $V(2\pi/3)$ , and  $V(\pi)$ . When  $V(\Delta\theta)$  is either the Villain form or a cosine, the model has an intermediate phase with algebraic order, as in Ref. 8 [see S. Elitzur, R. B. Pearson, and J. Shigemitsu, *Phys. Rev. D* **19**, 3698 (1979); A. Yamagata and I. Ono, *J. Phys. A* **24**, 265 (1991)]. For other choices of  $V$ , a single first-order transition is possible [see R. J. Baxter, *J. Phys. C* **6**, L455 (1973); also E. Domany and E. K. Riedel, *Phys. Rev. B* **19**, 5817 (1979)]. To estimate  $V(\Delta\theta)$  in our CG, we have computed the  $T=0$  surface tension between the six ground states of  $\mathcal{D}$ . If  $\theta$  is the phase of the order parameter  $\langle q_{\mathbf{k}_0} \rangle$ , we find  $V(\Delta\theta) = 0.103[1 - \cos(\Delta\theta)]$ . The algebraic phase should therefore be expected. If finite- $T$  excitations significantly perturb this  $T=0$  result for  $V$ , a first-order transition might still be possible. We have checked for a first-order transition by looking for hysteresis and for a bimodal energy distribution at  $T_c$ , and we find negative results. Of course, a sufficiently weak first-order transition, which would be detected only at larger lattice sizes, cannot be ruled out.

<sup>10</sup>M. N. Barber and W. Selke, *J. Phys. A* **15**, L617 (1982).

<sup>11</sup>R. M. F. Houtappel, *Physica (Utrecht)* **16**, 425 (1950).

<sup>12</sup>W. Y. Shih and D. Stroud, *Phys. Rev. B* **30**, 6774 (1984); **32**, 158 (1985).

<sup>13</sup>S. E. Korshunov, *J. Stat. Phys.* **43**, 17 (1986).

<sup>14</sup>D. R. Nelson and J. M. Kosterlitz, *Phys. Rev. Lett.* **39**, 1201 (1977); T. Ohta and D. Jasnow, *Phys. Rev. B* **20**, 130 (1979); P. Minnhagen and G. G. Warren, *Phys. Rev. B* **24**, 2526 (1981); G. S. Grest, *Phys. Rev. B* **39**, 9267 (1989).

<sup>15</sup>H. Weber and P. Minnhagen, *Phys. Rev. B* **37**, 5986 (1988).

<sup>16</sup>P. Minnhagen, *Phys. Rev. Lett.* **54**, 2351 (1985); *Phys. Rev. B* **32**, 3088 (1985).

<sup>17</sup>As the value  $T^* \approx 0.14$  of Minnhagen was obtained from a *continuum* CG model, it might not be appropriate for direct comparison with the triangular lattice. However, we find it suggestive that nowhere along the phase boundary between  $\mathcal{A}$  and  $\mathcal{B}$  do we find nonuniversal jumps.