BNL-26196

CONF-790487--1

Fluctuations in Two-Dimensional Six-Vertex Systems

R. W. Youngblood

J. D. Axe

B. M. McCoy

MASTER

The submitted manuscript has been authored under contract EY-76-C-02-0016 with the Division of Basic Energy Sciences, U.S. Department of Energy. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

FLUCTUATIONS IN TWO-DIMENSIONAL SIX-VERTEX SYSTEMS*

R. W. Youngblood, J. D. Axe and B. M. McCoy*

Physics Department
Brookhaven National Laboratory
Upton, N.Y. 11973

*Institute of Theoretical Physics
State University of New York at Stony Brook
Stony Brook, N.Y. 11794

ABSTRACT

The character of polarization correlations in six-vertex systems will be discussed. Making use of a connection between the 1-d Heisenberg-Ising chain and the six-vertex problem, we draw upon existing results for the chain correlations to obtain information about long-wavelength polarization correlations in six-vertex models. These results are compared with a neutron scattering study of 2-d polarization correlations in the layered compound copper formate tetrahydrate. Because the six-vertex model is equivalent to a particular roughening model, these results also explicitly predict the critical behavior of that roughening model just above its roughening temperature. The results correspond to the predictions of Kosterlitz and Thouless for the phase transition in the 2-d Coulomb gas.

*Research has been performed under Contract EY-76-C-02-0016 with the Division of Basic Energy Sciences, U.S. Department of Energy.

Although quite a bit is known about the statistical mechanics of 2-d six-vertex systems [1], much remains to be said about the fluctuations in these models. Here, we will present a short discussion of those aspects of the fluctuations that seem particularly germane to the subject of this particular conference. In particular, we will discuss analytic expressions for the long wavelength polarization correlation functions for these models which are asymptotically exact at large distance [2]. We will apply these results to neutron scattering experiments on a quasi-2-d hydrogen-bonded system, copper formate tetrahydrate (CFT) [3]. We will also show the relevance of these results to models of the solid-on-solid interfacial roughening transformation [4], and comment on the relation of Kosterlitz-Thouless theory [5] to six-vertex systems.

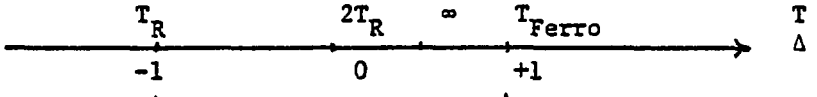
The models under discussion have been introduced by Dr. Weeks in his lectures at this conference [4]. The allowed vertex configurations are prescribed by the ice rules. The vertex weighting scheme is shown in Figure 1. (In adopting these weights, we are tacitly ruling out applied fields.) If vertices 1 and 2 are favored energetically, the ground state is ferroelectric along ± y; if 3 and 4 are favored, the ground state is ferroelectric along ± x; if 5 and 6 are favored, the ground state is antiferroelectric. The parameter Δ, defined in Fig. 1, is a measure of how close the system is to being ferro- or antiferroelectric. η, also defined in Fig. 1, measures the anisotropy in the polar configurations. Baxter [5] showed that singularities in the free energy corresponding to ferroelectric and antiferroelectric transformations occur at $\Delta = +1$ and -1, respectively. The relation between the three phases (antiferroelectric, disordered, and ferroelectric) is summarized in Fig. 1. For the weighting scheme we employ, no single physical system displays the full range of behavior shown in Fig. 1; the point $\Delta = 1/2$ corresponds to $T = \infty$. But the polarization correlations we discuss are a property of the entire disordered regime, and it is natural to discuss them as a function of Δ rather than T. We will also have occasion to mention the so-called FSOS roughening model [4] and the 1-d Heisenberg-Ising chain; certain relevant attributes which arise from equivalences within these models are summarized in Fig. 1.

Our primary interest is in long-wavelength polarization fluctuations. For this purpose, it is convenient to define a coarse-grained polarization. Consider the correlation function between two parallel arrows for the isotropic case, a = b. Introduce the variable $\sigma_j(m,n)$ to denote the sense of the arrow at site (m,n) (the subscript j = 1,2 denotes the arrow direction -- right or left sloping in Fig. 1). For $\Delta = 0$, Sutherland used an equivalence to a free fermion system to show that for large r

$$\langle \sigma_j(0)\sigma_j(m,n)\rangle \sim \frac{2}{\pi^2 r^2}\left|(-1)^{m+n} - \frac{(m^2-n^2)}{(m^2+n^2)}\right| \qquad (1)$$

a a b b c c

$$\Delta = \frac{a^2 + b^2 - c^2}{2ab} \qquad \eta = \frac{b}{a}$$



ANTIFERROELECTRIC	DISORDERED	FERROELECTRIC
SMOOTH INTERFACE	ROUGH	TILTED INTERFACE
GAP IN EIGENVALUE SPECTRUM	NO GAP IN EIGENVALUE SPECTRUM	GAP IN FIGENVALUE SPECTRUM

$$\mathcal{H} = -\sum \left(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y + \Delta S_i^z S_{i+1}^z \right)$$

Figure 1.

where m,n are integer coordinates and $r = \sqrt{m^2+n^2}$. We see that there is an antiferroelectric contribution which oscillates rapidly, and a ferroelectric component which does not. We therefore form the "coarse-grained" ferroelectric polarization

$$P_j(\vec{r}) = \sigma_j(m,n) + \sigma_j(m+1,n).$$

It is easy to see that the leading contribution to $\langle P_j(0)P_j(\vec{r})\rangle$ is the smooth contribution to $\langle \sigma_j(0)\sigma_j(m,n)\rangle$; the other contributions cancel, to leading order in $1/r$. In fact, making use of a connection between this model and the dimer model [6], we have shown [2] that leading contributions to the coarse-grained polarization correlations at $\Delta = 0$ have the form

$$\langle P_y(0)P_y(\vec{r})\rangle \sim -A \frac{(x^2-y^2)}{(x^2+y^2)^2} \tag{2}$$

$$\langle P_y(0)P_x(\vec{r})\rangle \sim A \frac{2yx}{(x^2+y^2)^2} \tag{3}$$

$$\langle P_x(0)P_x(\vec{r})\rangle \sim A \frac{(x^2-y^2)}{(x^2+y^2)^2} \tag{4}$$

where $A = 2/\pi^2$. These quantities also depend on η; for simplicity, we have temporarily set $\eta = 1$. In these expressions and in the following, the symbol $\sim$ means "is asymptotically equal to in the limit of large r."

The simple form of these functions reflects an underlying simplicity in the system. The six-vertex condition (Fig. 1) is the condition that polarization be locally "divergenceless" at each lattice site. This condition is largely responsible for the form of Eq. (2-4). The ice rules (Fig. 1) state that the polarization is "divergenceless" at each lattice site, which in turn means that $\vec{P}(\vec{r})$ is a solenoidal vector field, $\nabla\cdot\vec{P}(\vec{r}) = 0$. This guarantees that $\vec{P}(\vec{r})$ is the curl of a mathematically simpler vector field, $\vec{h}(\vec{r})$. Fortunately, there is a physical as well as a mathematical motivation for introducing $\vec{h}(\vec{r})$. We know from the work of van Beijeren [8] that $\vec{h}(\vec{r})$ is simply related to the height variable of a surface roughening model.

Dr Weeks' lecture in this volume [4] discusses the relation of a particular roughening model (which he denotes FSOS) with the six-vertex model which we are discussing. In particular, Fig. 3 of his third lecture shows on a discrete lattice how to associate a spin variable with the local gradient of the column height h. With the appropriate choice or coordinate system, this is just

$$\sigma_1(m,n) = -[h(m,n+\tfrac{1}{2}) - h(m,n-\tfrac{1}{2})] \tag{5}$$

$$\sigma_2(m,n) = [h(m+\tfrac{1}{2},n) - h(m-\tfrac{1}{2},n)] \tag{6}$$

Thus, for example (Fig. 2a), an ordered polar six-vertex configuration pointing along (say) y, is equivalent to a surface with a monotonically increasing height, $\partial h/\partial x$ = constant. The local polarization conservation $\nabla\cdot\vec{P} = 0$ completely suppresses longitudinal polarization fluctuations, which in the language of the height variable translate into a discontinuous "tear" on the crystal surface, as shown in Fig. 2b. Such configurations correspond to unacceptably large step sizes in the roughening model.

It is clear from the foregoing paragraph that the proper choice of vector potential $\vec{h}(\vec{r}) = h(\vec{r})\hat{z}$, so that

$$\vec{P}(\vec{r}) = (P_x,P_y) = \left(-\frac{\partial h}{\partial y}, \frac{\partial h}{\partial x}\right) = \nabla\times\vec{h}(\vec{r}). \tag{7}$$

Then, if we define a height-height correlation function

$$\Psi(\vec{r}-\vec{r}_o) = \langle h(\vec{r})h(\vec{r}_o)\rangle \sim -A\ln(|\vec{r}-\vec{r}_o|),$$

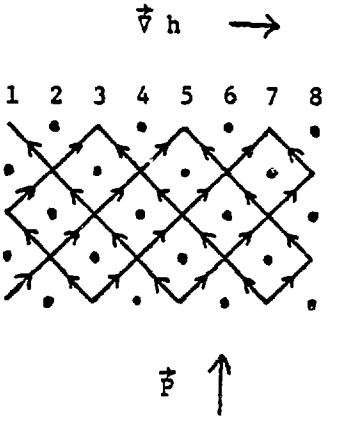


Fig. 2a. The completely polarized state of the six-vertex lattice corresponds to a monotonically sloping surface. (Compare vertex 3 of Fig. 3 of Weeks' third lecture.) The numbers give the heights of the lattice sites above which they appear.

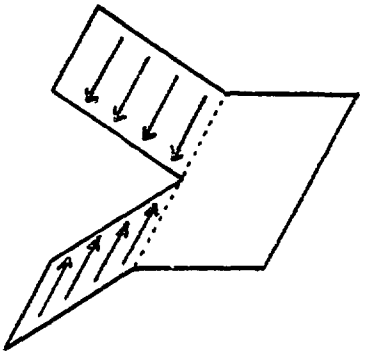


Fig. 2b. A longitudinal polarization fluctuation (violating $\nabla\cdot\vec{P} = 0$) corresponds to a tear in the surface (violating the step size constraint in the roughening model).

Eqs. (2-4) can be restated very concisely in terms of the height-height correlation function, $\Psi(\vec{r}-\vec{r}_o)$.

$$\langle P_y(\vec{r}_o)P_y(\vec{r})\rangle = \frac{\partial}{\partial x_o}\frac{\partial}{\partial x}\Psi(\vec{r}-\vec{r}_o) \tag{8}$$

$$\langle P_y(\vec{r}_o)P_x(\vec{r})\rangle = -\frac{\partial}{\partial x_o}\frac{\partial}{\partial y}\Psi(\vec{r}-\vec{r}_o) \tag{9}$$

$$\langle P_x(\vec{r}_o)P_x(\vec{r})\rangle = \frac{\partial}{\partial y_o}\frac{\partial}{\partial y}\Psi(\vec{r}-\vec{r}_o) \tag{10}$$

Thus far, all of our results have been restricted to $\Delta = 0$, where the problem is especially simple. However, we can now proceed to extend the results throughout the regime $-1<\Delta<1$, by making contact with previous work on the 1-d Heisenberg-Ising chain. McCoy and Wu [9] showed that the transfer matrix of the six-vertex problem

commutes with the Heisenberg-Ising Hamiltonian, shown in Fig. 1. Therefore, there is a connection between the correlation functions in the two problems. Recent work by Luther and Peschel [10] and Fogedby [11] gives the leading asymptotic contribution to $\langle S^z(0,0)S^z(x,t=0)\rangle$, which (by virtue of the above-mentioned commutation relation) has the same general form as $\langle \sigma_j(0,0)\sigma_j(m,n=0)\rangle$ (equation 1), including a prefactor A which is a known function of Δ. This result effectively prescribes the coarse-grained correlation function along one axis. Since $\nabla\cdot\vec{P} = 0$, there is still a generating vector potential which can be analytically continued from that axis to cover the entire x-y plane. Thus we arrive at the following result, valid for $-1<\Delta<1$.

$$\Psi(\vec{r}) \sim -A \ln r + \text{constant} \tag{11}$$

where $A = (\pi^2\theta)^{-1}$ (Luther and Peschel, [10], Fogedby [11])

and $\theta = \frac{1}{2} - \frac{1}{\pi}\sin^{-1}\Delta$ (Johnson, Krinsky and McCoy [12])

The formulation of the correlation function $\Psi(r)$ given in Eq. (11), together with Eq. (8-10) (which generate the asymptotic polarization correlation functions), constitute the principal results of this paper. We now turn to a discussion of their significance in two different areas.

Since Chui and Weeks [13] have shown that the discrete Gaussian roughening model maps onto the 2-d Coulomb gas problem, and since van Beijeren [8] has explicitly demonstrated that a similar rougening model maps exactly onto the F model, it is natural to suppose (along with Shugard et al. [14] and others [4]), that there is a close connection between the critical behavior of the Kosterlitz-Thouless transition and the critical behavior of the six-vertex transition at $\Delta = -1$. In particular, the prefactor K_∞ appearing [14] in

$$G(r) = 2\left(\langle h(0)^2\rangle - \langle h(0)h(r)\rangle\right) \sim \frac{K_\infty(T)}{\pi} \ln r + c \tag{12}$$

is to be compared with $A(\Delta)$ appearing in Eq. (11). If we can identify $K_\infty(T)$ with $2\pi A(\Delta)$, we can expand $A(\Delta)$ to obtain

$$K_\infty(T \to T_R^+) = \frac{2}{\pi} + (\text{nonuniversal constant}).\ (T-T_R)^{1/2} + \ldots \tag{13}$$

Both the value $K_\infty(T_R) = \frac{2}{\pi}$ and the leading square root behavior are predicted by Kosterlitz-Thouless theory for the unbinding of vortices. Thus, the present results for Ψ explicitly support the idea that the six-vertex model (together with its various equivalents) is in the same universality class as the 2-d Coulomb gas (together with its equivalents).

Now we turn to polarization fluctuations in CFT. A full description of this work is contained in Refs. [2] and [3]. Crystals of CFT contain 2-d layers of water molecules interleaved with 2-d layers of copper formate. Above $T = T_o = 248$ K (in the deuterated compound), there is icelike disorder in the hydrogen-bond network. Below T_o, the layers become ferroelectric, the direction of polarization alternating between +b and -b from one layer to the next. The in-plane longitudinal momentum coordinate is K; the transverse momentum coordinate is H. Figure 3 shows some results of a diffuse neutron scattering study of the long-wavelength polarization correlations. ("Polarization" here means that of the hydrogen atom positions, measured from the centers of their respective bonds.) The quantity plotted is the measured intensity, $I(\vec{Q})$.

$$I(\vec{Q}) = \overline{|F(\vec{Q})|^2 S_{yy}(\vec{q})}. \qquad (14)$$

$F(\vec{Q})$ is a geometrical structure factor, which is a rather slowly varying function of $\vec{Q} = \vec{G} + \vec{q}$, where $\vec{G}$ is a reciprocal lattice

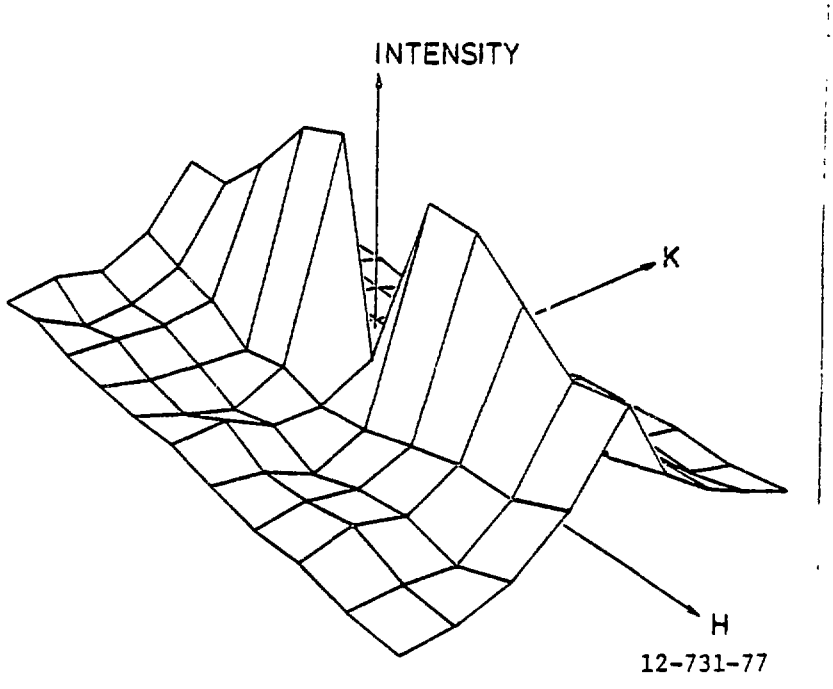


Fig. 3. Shown here are results of a neutron scattering study of polarization fluctuations in the 2-d hydrogen-bond network of copper formate tetrahydrate (Ref. [3]). The origin of the indicated coordinate system corresponds to a reduced intralayer momentum transfer $\vec{q}$ of zero. Data are shown for essentially an entire (intralayer) Brillouin zone.

vector. $S_{yy}(\vec{q})$ is the Fourier transform of $\langle P_y(0)P_y(r)\rangle$, and the bar denotes an average over instrumental resolution. The important feature in this plot is the notch at $\vec{q} = 0$. Typically (for example in Ising-type systems), pair correlations near T_c give rise to scattering which peaks at $\vec{q} = 0$; as T_c is approached, the diffuse peak associated with order parameter fluctuations is seen to grow. Here, instead of peaking, the intensity dips to near zero, in spite of the incipient transition to an ordered state in which there is a Bragg peak at $\vec{q} = 0$. (In calling the origin of Fig. 3 $\vec{q} = 0$, we have suppressed the L momentum coordinate, upon which the scattering is only weakly dependent. This is the expected quasi-2-d behavior. However, it is important to note that the data of Fig. 3 lie in a plane in which L is half-integral, in which the antiferroelectric Bragg peaks occur below T_o.)

In the CFT problem, $\eta \neq 1$, and the hitherto suppressed η dependence of the correlations must be taken into account. It is shown in Ref. [2] that this can be done by replacing r in Eq. (11) with

$$\rho(\vec{r}) = \sqrt{x^2 + \lambda^2 y^2} \tag{15}$$

For $\Delta = 0$, we have $\lambda = \eta$. Polarization correlations are still given by Eqs. (8-10). The Fourier transform of $\langle P_y(0)P_y(\vec{r})\rangle$ is given by

$$S_{yy}(\vec{q}) = \pi A\lambda \frac{h^2}{\lambda^2 h^2 + k^2} \quad \text{at small q.} \tag{16}$$

This function is plotted in Fig. 4 in units of πA, with λ chosen to correspond roughly to the experimental observations. Note that there is a strong formal resemblance between the singularity in this function and that occurring in dipolar-coupled systems (see Dr. Als-Nielsen's notes on $LiTbF_4$). However, we stress that the calculated effect is due entirely to six-vertex ineractions. The absence of longitudinal fluctuations ($S_{yy}(h,k=0)=0$) is a direct consequence of the ice rule restrictions, $\nabla\cdot\vec{P} = 0$. In real scattering experiments, the cross section one observes is somewhat smeared by finite instrumental resolution. This effect is partially taken into account in Fig. 5. There is a strong resemblance between Figs. 3 and 5; thus, the small-q regime (the notch) of Fig. 3 is evidence that the pair correlations in CFT are qualitatively obeying Eq. (2). It would be of some interest to perform further measurements with much higher resolution to test Eq. (8) quantitatively.

In summary, we see that in the disordered phase of a particular class of six-vertex systems, long-wavelength polarization correlations are governed by a logarithmic (2d-Coulomb-like) potential. The scattering cross section of hydrogen-bonded systems of this type depends on particular spatial derivatives of this potential;

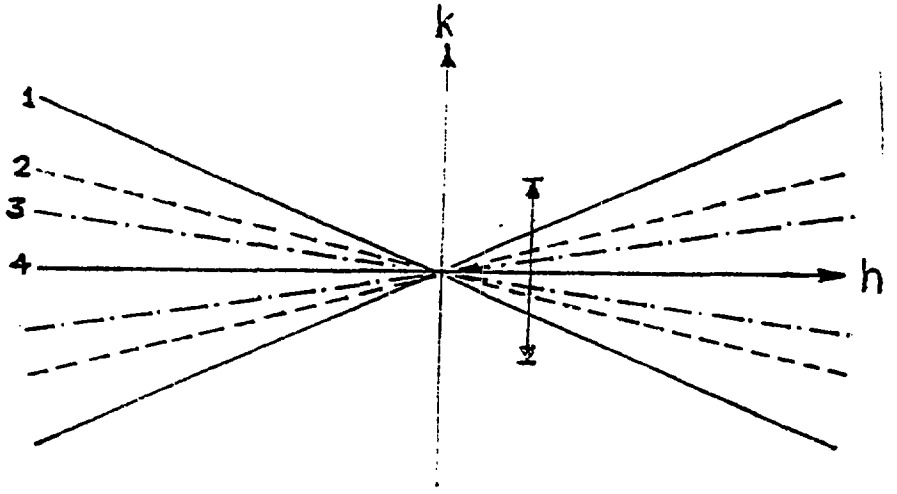


Fig. 4. This is a contour plot of $S_{yy}(q)$ (see Eq. (16)). The contours are at integer multiples of $A\pi$, for the case $\lambda = \frac{1}{4}$. The arrow indicates a longitudinal resolution width (see Fig. 5).

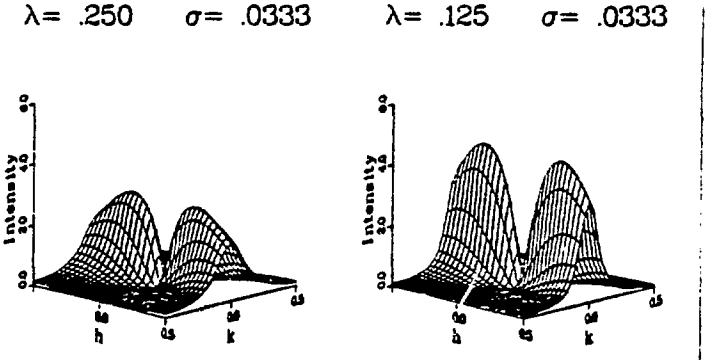


Fig. 5. These are plots of I(Q) calculated from Eq. (14). S_{yy} is given by Eq. (16), with A held constant. F(Q) corresponding to the data shown in Fig. 3 is included. σ is the halfwidth along k of a Gaussian instrumental resolution function (the resolution along h is assumed to be perfect). σ, h, and k are given in reciprocal lattice units.

the observed cross section from such correlations is quite distinctive. Given certain rigorous connections between the Heisenberg-Ising problem and the six-vertex problem, we can draw upon existing results to obtain new information about the six-vertex problem away from the free fermion limit, $\Delta = 0$. In particular, we can infer the temperature dependence of the prefactor of the logarithmic potential. In addition to making an explicit statement about a particular roughening model, the results correspond gratifyingly to predictions of Kosterlitz-Thouless theory for the vortex-unbinding transformation, thereby reinforcing the idea that roughening models are in the same universality class.

ACKNOWLEDGMENT

Two of us (RWY and JDA) have profited from conversations held at this Institute with J. D. Weeks, on the connection between roughening models and the six-vertex problem, and from several conversations with V. J. Emery.

REFERENCES

1. For a review, see the article by E. H. Lieb and F. Y. Wu in "Phase Transitions and Critical Phenomena," ed. C. Domb and M. S. Green, Academic, New York (1977).
2. R. Youngblood, J. D. Axe. and B. McCoy, submitted to Phys. Rev.
3. R. Youngblood and J. D. Axe, Phys. Rev. B17, 3639 (1978).
4. For reviews, see J. D. Weeks, this volume; J. D. Weeks and G. H. Gilmer in "Advances in Chemical Physics," ed. I. Prigogine and S. A. Rice, Vol. 40 (1979); H. Müller-Kvumbhaar in "1976 Crystal Growth and Materials," North Holland, New York (1977).
5. J. M. Kosterlitz and D. J. Thouless, J. Phys. C6, 1181 (1973); J. M. Kosterlitz, J. Phys. C7, 1046 (1974). A review is given by A. P. Young in this volume.
6. R. J. Baxter, Ann. Phys. (N.Y.) 70, 193 (1972).
7. B. Sutherland, Phys. Lett. 26A, 532 (1968).
8. H. van Beijeren, Phys. Rev. Lett. 38, 993 (1977).
9. B. M. McCoy and T. T. Wu, Nuovo Cimento 56B, 311 (1968).
10. A. Luther and I. Peschel, Phys. Rev. B12, 3908 (1975).
11. H. C.Fogedby, J. Phys. C11, 4767 (1978).
12. J. D. Johnson, S. Krinsky, and B. M. McCoy, Phys. Rev. A8, 2526 (1973).
13. S. T. Chui and J. D. Weeks, Phys. Rev. B14, 4978 (1978).
14. W. J. Shugard, J. D. Weeks and G. H. Gilmer, Phys. Rev. Lett. 41, 1399 (1978).

BNL-26196

Fluctuations in Two-Dimensional Six-Vertex Systems

R. W. Youngblood

J. D. Axe

B. M. McCoy