Terrace-width distributions of touching steps: Modification of the fermion analogy with implications for measuring step-step interactions

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(Received 28 July 2009; revised manuscript received 24 September 2009; published 30 October 2009)

Using Monte Carlo simulations, we compute the terrace-width distributions (TWDs) of surfaces in which steps can touch each other, forming multiple-atomic height steps, but cannot cross (no overhangs), and so inconsistent with the standard mapping to spinless fermions. Our results show that the generalized Wigner distribution with minor modifications at small step separations, gives a very good fit for TWDs of touching steps. The interaction strength derived from the fit parameter (ϱ) indicates an effective attraction between steps. The strength of this effective attraction decreases for larger mean-step separations and decreasing step-touching energies; describable via finite-size scaling. Hence, accurate extraction of the true repulsion strength requires multiple vicinalities.

DOI: 10.1103/PhysRevB.80.153415 PACS number(s): 68.35.Md, 05.40.−a, 71.10.Pm

Steps on vicinal (misoriented from facet planes) surfaces can be used as templates for fabrication of metallic nanowires. They also can enhance the catalytic activity of a surface, especially when they have many kinks. This makes the study of step fluctuations technologically important.[1,2] Step configurations have fruitfully been related to the worldlines of spinless fermions in one dimension (1D). The fermion analogy originates in the veridical condition that the steps cannot cross. However, the analogy would also preclude steps from coinciding, to form multilayer steps, which could occur physically. This issue has largely been ignored or glossed over.

In this Brief Report, we show how this loosening of the nontouching condition alters the form of the terrace-width distribution (TWD) and, thence, the apparent strength of the step-step repulsion deduced from it. By understanding the form of the resulting finite-size corrections, we show how to analyze vicinal surfaces to take this issue into account. Such touching steps are more likely to be found on surfaces with one or more of the following properties: (i) low step stiffness, (ii) closely spaced steps (small separation ℓ), and (iii) steps with little or no energetic interaction between them.

We first recall the main ideas for steps forbidden to touch. At low-temperature the predominant thermal excitations are kinks along the step, with kink formation energy ϵ_k. Labeling the mean direction of the step edges $\hat{y}$ and the perpendicular direction $\hat{x}$ (so-called "Maryland notation"), the position of the ith step edge in this terrace-step-kink (TSK) model is $\mathbf{x}_i(\mathbf{y}_n)$, where sans serif denotes discreteness; $\mathbf{y}_n$ is defined only at the centers of step-edge atoms, with the index n changing by 1 for unit displacement along $\hat{y}$. The energy contribution from kinks is $\epsilon_k \Sigma_{i,n} |\mathbf{x}_i(\mathbf{y}_{n+1}) - \mathbf{x}_i(\mathbf{y}_n)|$. There can also be an elastic (or possibly dipolar) repulsion between steps, which decays asymptotically as $1/\ell^2$, the same behavior as the entropic repulsion due to noncrossing. The elastic repulsion is approximated by the "instantaneous" form $A\Sigma_{j>0} |\mathbf{x}_{i+j}(\mathbf{y}_n) - \mathbf{x}_i(\mathbf{y}_n)|^{-2}$. This expression is well defined for nontouching steps $[\mathbf{x}_{i+1}(\mathbf{y}_n) > \mathbf{x}_i(\mathbf{y}_n)]$. Touching at corners is allowed [e.g., $\mathbf{x}_{i+1}(\mathbf{y}_n) = \mathbf{x}_i(\mathbf{y}_{n+1})$].

For analytic modeling it is more convenient to use the step-continuum approximation,[1] which allows $x_i(y)$ to vary continuously with y. Since $x_i(y)$ is single valued, the configuration of steps in two spatial dimensions can be viewed as the worldlines of particles evolving in 1D: y becomes timelike. The nontouching condition underlies their characterization as spinless fermions (or hard bosons) in 1D, and the stiffness $\tilde{\beta}$, which can be related to ϵ_k, is their "mass." In this framework, the instantaneous step-step repulsion strength A enters only as a dimensionless combination $\tilde{A} \equiv A\tilde{\beta}\beta^2$, where β is $1/(k_B T)$. Since $\tilde{A}$ determines the morphology of and communication between steps, gauging it is crucial to understanding step behavior. To this end, we scrutinize the TWD, which is normally expressed as the probability distribution, $\hat{P}(\ell)$, of finding neighboring steps at separation ℓ. With only $1/\ell^2$ repulsions, there is just one characteristic length, the mean-step separation $\langle \ell \rangle$ in the $\hat{x}$ direction, and so the TWD essentially depends only on the dimensionless length $s = \ell/\langle \ell \rangle$

$$P(s = \ell/\langle \ell \rangle) = \langle \ell \rangle \hat{P}(\ell). \tag{1}$$

To obtain $P(s)$ we pursue the analogy to the Calogero-Sutherland models[3] of spinless fermions along a chain [or on a 1D ring]. The key parameter ϱ in these models is[4]

$$\varrho = 1 + \sqrt{1 + 4\tilde{A}}; \quad \tilde{A} = \frac{\varrho}{2}\left(\frac{\varrho}{2} - 1\right). \tag{2}$$

For $\varrho = 1, 2, 4$, the ground-state probability density, which corresponds to $P(s)$, reduces to the distribution of eigenvalues for random matrices with orthogonal, unitary and symplectic symmetry, respectively.[5] Accordingly, they are excellently approximated by the Wigner surmise[6]

$$P_\varrho(s) = a_\varrho s^\varrho e^{-b_\varrho s^2}, \quad b_\varrho = \left[\frac{\Gamma\left(\frac{\varrho+2}{2}\right)}{\Gamma\left(\frac{\varrho+1}{2}\right)}\right]^2, \quad a_\varrho = \frac{2b_\varrho^{(\varrho+1)/2}}{\Gamma\left(\frac{\varrho+1}{2}\right)}. \tag{3}$$

For stepped surfaces there is no reason for $\tilde{A}$ to have the special values 0 or 2 ($\varrho = 2$ or 4), and $-1/4$ ($\varrho = 1$) is unphysi-

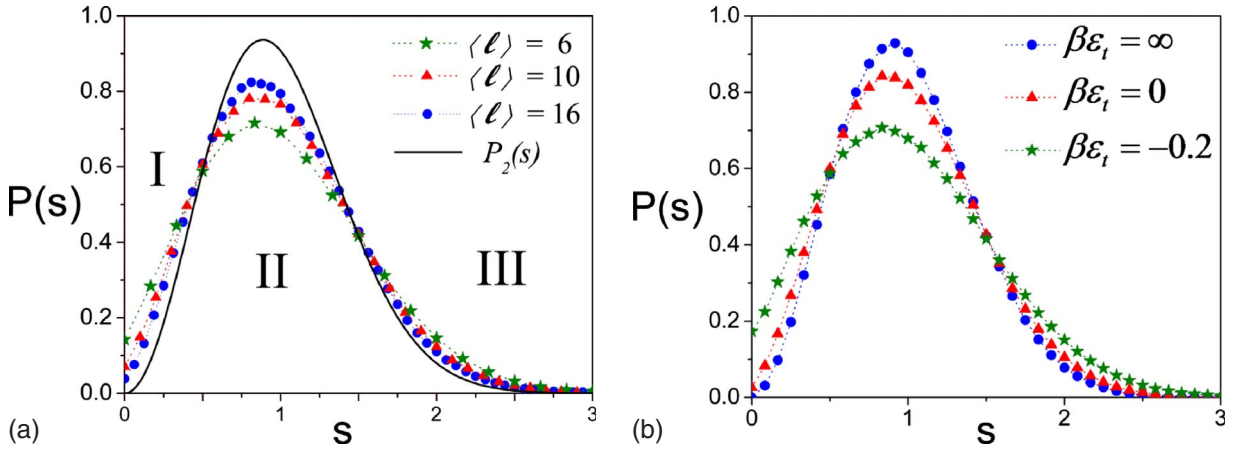

FIG. 1. (Color online) TWDs of touching steps computed (a) for different $\langle \ell \rangle$ values with $\beta\epsilon_t=-0.1$. The solid curve is $P_2(s)$, the GWD for nontouching steps with no energetic interactions. (b) TWDs for different $\beta\epsilon_t$ values with $\langle \ell \rangle=12$.

cal. Thus, we have taken Eq. (3) to apply for arbitrary $\varrho \geq 2$ or $\tilde{A} \geq 0$ and call it the generalized Wigner distribution (GWD). The GWD gives a better description of TWDs measured from both experiments and numerical simulations than any of the pre-existing models[7] and has many other virtues, discussed elsewhere.[8,9] Richards *et al.*[10] emphasized that differences between Eq. (3) and its discrete analog become nonnegligible for $\langle \ell \rangle \lesssim 4$.

If steps can touch (two or more step edges can be at the same position), the analogy with 1D fermions is not strictly valid. The issue is much more significant for discrete models $[\mathbf{x}_i(\mathbf{y}_n)=\mathbf{x}_j(\mathbf{y}_n)]$. Since touching is a contact interaction, its effect on TWDs should be insignificant only for $\langle \ell \rangle \gg 6$ or $s \gg 1$; TWDs of touching steps should then converge to the GWD. With Monte Carlo simulations we investigate how touching does alter the TWD and how one then should measure $\tilde{A}$.

Since we are interested in equilibrium TWDs, we used the Metropolis method in our simulations. We modeled the vicinal surface using the TSK model with steps along the $\langle 100 \rangle$ direction (straight steps). The underlying lattice was taken to be simple cubic,[11] with (screw-)periodic boundary conditions along $(\hat{x})\hat{y}$; $L_y=5-600$ and $L_x=N\langle \ell \rangle$, for $N=28-40$ steps. Steps were allowed to fluctuate via the attachment-detachment process. Step-edge diffusion was suppressed in our simulations. To focus clearly on the problem of concern, we considered only the case of "free fermions:" $A=\tilde{A}=0 \Rightarrow \varrho=2$. This eliminated divergences of the elastic repulsion term when the steps touched and also avoided the issue of whether all steps or just neighboring experienced this repulsion. There are two energy parameters in this model: the kink formation energy (ϵ_k) and the energy (ϵ_t) of two adjacent steps touching to form a double-height unit segment. For simplicity, we assumed the energy to form a p-layer high step is $(p-1)\epsilon_t$. We set the temperature of the simulations such that $\beta\epsilon_k=2$, as in our group's previous simulation studies.[7,12] We simulated the TWDs for values of ϵ_t ranging from $\beta\epsilon_t=\infty$ (nontouching case) down to $\beta\epsilon_t<0$. In the rest of this Brief Report, we refer to steps with an energy cost for doubling $(\beta\epsilon_t \geq 0)$ as repulsively touching (RT) steps and steps for which doubling is energetically favored $(\beta\epsilon_t<0)$ as attractively touching (AT) steps.

Allowing touching alters the resulting TWD $P(s)$ from that of nontouching steps $P_2(s)$ in two major ways: (i) $P(0)>0$ and (ii) $P(s)$ is broader than $P_2(s)$. For terraces with the same step touching energy (ϵ_t), the deviation is greater for surfaces with smaller $\langle \ell \rangle$ values [cf. Fig. 1(a)] and for surfaces with the same $\langle \ell \rangle$, the deviation is greater for surfaces with smaller ϵ_t values [cf. Fig. 1(b)]. The deviation can also be quantified as follows: $P_2(s)$ divides the TWDs of touching steps into three regions as marked in Fig. 1(a). In regions I $(0 \leq s \lesssim 0.5)$ and III $(s \gtrsim 1.5)$, TWDs of touching steps have higher values compared to $P_2(s)$, while in region II, they have lower values. The TWDs of both kinds of steps rise as power laws for small s and decay as Gaussians for large s. For the former, we make the ansatz

$$P_{\gamma,\varrho} = P^t(s) + a_{\gamma,\varrho} s^{\varrho} e^{-b_{\gamma,\varrho} s^2}; \quad P^t(s) \equiv P(0) e^{-\gamma s}, \tag{4}$$

where $P^t(s)$ describes the distribution for small values of s, capturing the effect of touching. The second term is the GWD modified to accommodate the first term; γ and ϱ are the fit parameters and $P(0)$ is the value of the distribution at $s=0$ measured experimentally or through simulations. Via Eq. (1) $\langle \ell \rangle^{-1} P(0)$ gives the ratio of double- or multiple-atomic height step segments to the total length of steps. Equation (4) can be rewritten more conveniently in terms of P_ϱ [cf. Eq. (3)]

$$P_{\gamma,\varrho}(s) = P(0) e^{-\gamma s} + \Theta P_\varrho(s/\lambda), \tag{5}$$

$$\frac{b_{\gamma,\varrho}}{b_\varrho} = \frac{1}{\lambda^2}, \quad \lambda \equiv \frac{1-\dfrac{P(0)}{\gamma^2}}{1-\dfrac{P(0)}{\gamma}}; \quad \frac{a_{\gamma,\varrho}}{a_\varrho} = \frac{\Theta}{\lambda^\varrho}, \quad \Theta \equiv \frac{1-\dfrac{P(0)}{\gamma}}{\lambda} \tag{6}$$

using the normalization and unit mean of $P_{\gamma,\varrho}(s)$. The argument s/λ of P_ϱ can be reduced to s if we work in terms of an effective mean-step spacing $\langle \ell \rangle_{\text{eff}}=\lambda\langle \ell \rangle$. For the special case $\epsilon_t=0$, the TWD for the discrete case is simply that of a vicinal surface with mean spacing $\langle \ell \rangle+1$ with the form

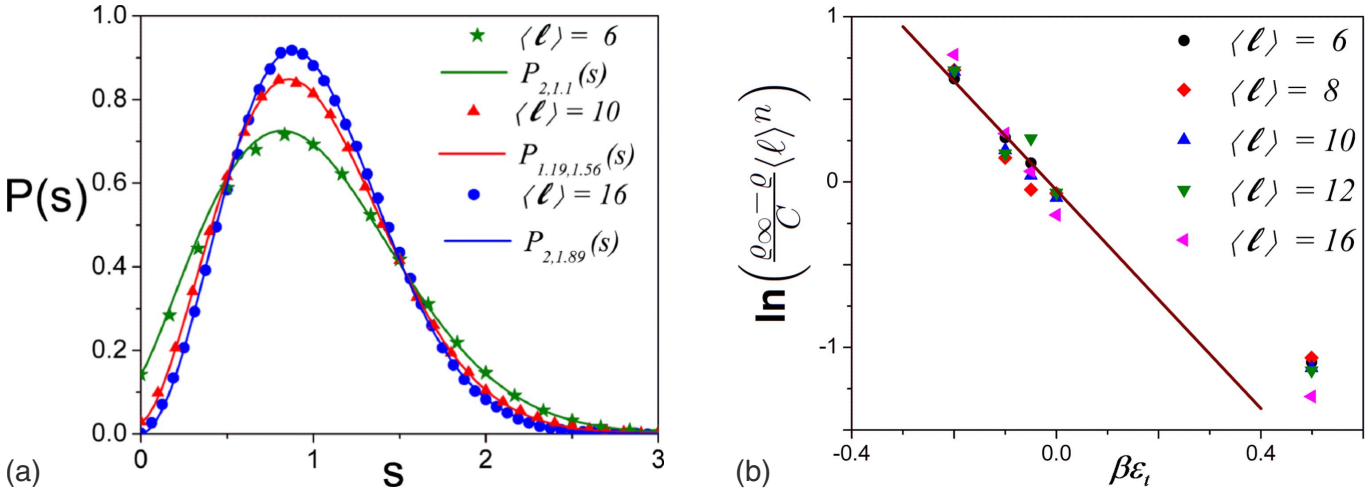

FIG. 2. (Color online) (a) MGWD fits (solid curves) to the TWDs (symbols) of both straight and fully kinked touching steps. The $\beta\epsilon_t$ value for $\langle\ell\rangle=6$ is -0.1, for $\langle\ell\rangle=10$ (fully kinked steps) is 0, and for $\langle\ell\rangle=16$ is 0.5. (b) Collapse of our simulation data (line slope $m=3.3$) onto the finite-size relation given in and after Eq. (7): $\varrho_\infty-\varrho(\langle\ell\rangle,\beta\epsilon_t)=C\langle\ell\rangle^{-n}e^{-m\beta\epsilon_t}$ (or, since $\beta\epsilon_k=2$, $C\langle\ell\rangle^{-n}e^{-2m\epsilon_t/\epsilon_k}$).

$\hat{P}(\ell_T+1)$, where ℓ_T for this case of (free) touching starts at 0 rather than 1. While it is straightforward to formalize this procedure, the rather complicated result does not allow inclusion of nonzero ϵ_t nor lead to an effective value of ϱ.[13,14]

The distribution, $P_{\gamma,\varrho}(s)$, gives an excellent fit to the TWDs of touching steps [see Fig. 2(a)]. We refer to $P_{\gamma,\varrho}(s)$ as the modified GWD (MGWD). Broadly speaking, the values of $P(0)$ and γ determine $P_{\gamma,\varrho}(s)$ in region I, Θ determines its peak height in region II, and λ determines the decay rate of $P_{\gamma,\varrho}(s)$ in region III. For fixed values of ϱ and γ, higher (lower) values of $P(0)$ and λ combined with a lower (higher) value of Θ implies a broader (narrower) distribution.

The values of $P(0)$ obtained from our simulations are listed in Table I along with the values of γ and ϱ determined from fits using the nonlinear fitting function (with all data points weighed equally) in MATHEMATICA®. The proportion of double- or multiple-atomic height step segments, $P(0)$, is higher for surfaces with smaller $\langle\ell\rangle$ and lower ϵ_t [cf. Table I, Figs. 1(a) and 1(b)], as expected: with smaller $\langle\ell\rangle$, step segments are more likely to meet during fluctuations, and the lower ϵ_t, the more likely such steps stay touched. In all of our cases, the parameter γ is invariably 2, regardless of $\langle\ell\rangle$ and ϵ_t, until negative ϵ_t heralds the instability of the steps to collapse. Especially for RT steps, the very small values of $P(0)$ values lead to insensitivity to γ in the quality of the fit. Due to its weak dependence on $\langle\ell\rangle$ and ϵ_t, no physically relevant information can be extracted from the value of γ.

From Table I we see that $\varrho<2$,[15] implying an effective attraction between steps [$\tilde{A}_{\rm eff}<0$, cf. Eq. (2)], even though actually $\tilde{A}=0$. This attraction is greater for surfaces with smaller $\langle\ell\rangle$ and smaller ϵ_t. As for entropic effects, this point interaction manifests as a ℓ^{-2} interaction between steps, albeit as a finite-size effect. The fact that touching leads to an attraction between steps has also been observed in the recent analytic study.[11] These two studies show that touching could impact step-step interaction strength measurements significantly. There are other problems that occur for relatively small $\langle\ell\rangle\lesssim 4$. The distinction between discrete and continuum models becomes non-negligible.[10] Higher-order corrections $[\mathcal{O}(\ell^{-3}),\mathcal{O}(\ell^{-4})]$ to the repulsive interaction are known play a role at small step separations.[16] Thus, experiments to measure $(\tilde{A})$ should involve several misorientations, at least some of which should be shallow ($\langle\ell\rangle\gg 1$).

As substantiated by Table I, $\gamma>1$ and $P(0)<1$, from which $\lambda>1$ and $\Theta<1$. Since $\lambda>1$, $\langle\ell\rangle_{\rm eff}=\lambda\langle\ell\rangle>\langle\ell\rangle$: steps see an effective mean spacing greater than the actual value because the multiheight steps reduce the step density on the remaining vicinal surface. Since $\Theta<1$, the peak of the TWD of touching steps is lower than that of $P_2(s)$. For fixed γ, Eq. (6) shows that λ increases and Θ decreases with $P(0)$, implying a broader TWD.

To investigate the effect of step stiffness $\tilde{\beta}$ on TWDs of

TABLE I. Values of $P(0)/\gamma/\varrho$ obtained from our simulations and fits using MATHEMATICA® for different values of ℓ and $\beta\epsilon_t$. The values inside the parentheses are from the case of fully kinked $\langle 11\rangle$ (zigzag) steps rather than nearly straight $\langle 10\rangle$ steps.

$\langle\ell\rangle$	$\beta\epsilon_t=\infty$	0.5	0	(0—zigzag)	−0.05	−0.1	−0.2
6	0.000/n.a./1.80	0.012/2/1.62	0.076/2/1.3	(0.058/2/1.41)	0.102/2/1.2	0.142/2/1.1	0.321/4/0.8
8	0.000/n.a./1.87	0.008/2/1.70	0.049/2/1.41	(0.036/1.5/1.50)	0.068/2/1.4	0.098/2/1.3	0.253/3/0.9
10	0.000/n.a./1.90	0.005/2/1.75	0.035/2/1.48	(0.029/1.19/1.56)	0.041/2/1.42	0.070/2/1.34	0.213/3/1.0
12	0.000/n.a./1.96	0.003/2/1.82	0.026/2/1.55	(0.017/1.7/1.641)	0.040/2/1.39	0.055/2/1.44	0.173/2/1.1
16	0.000/n.a./2.00	0.002/2/1.89	0.015/2/1.67	(0.011/1.2/1.645)	0.023/2/1.57	0.038/2/1.46	0.129/2/1.13

touching steps, we also simulated the TWDs of surfaces with steps along the ⟨110⟩ direction (fully kinked, or zigzag, steps). The applicability of the generalized Wigner formalism for this case has not been reported. Since fully kinked steps have smaller $\tilde{\beta}$ than straight steps,[17] we consider the simplest case in which ⟨110⟩ steps fluctuate freely (without energy cost). However, attachment and detachment processes were allowed only at kink sites to maintain an equal number of kinks and antikinks. We restricted ourselves to the freely touching ($\epsilon_t=0$) case. To measure step separations, we followed the mapping method proposed by Abraham *et al.*[18] We observed these TWDs to be very similar to those for straight steps with $\epsilon_t=0$. Hence, the MGWD gives a very good fit even in the case of fully kinked steps. The fitted values of $P(0)$, γ, and ϱ are listed in Table I. The ϱ values for ⟨110⟩ steps are only slightly higher than those for ⟨100⟩ steps, while the values of $P(0)$ and γ are slightly lower. Hence, step stiffness has at most a weak effect on the TWDs of touching steps.

Returning to the idea that the effective attraction is a finite-size effect, we expect that the value ϱ_∞ when step touching is absent [and so linked to $\tilde{A}$ by Eq. (2)] is related to the values ϱ measured from fits by

$$\varrho = \varrho_\infty - f(\langle \ell \rangle, \beta\epsilon_t), \quad f(\langle \ell \rangle, \beta\epsilon_t) \underset{\langle \ell \rangle \to \infty \text{ or } \beta\epsilon_t \to \infty}{\to} 0^+. \quad (7)$$

We find that the finite-size scaling function $f(\langle \ell \rangle, \beta\epsilon_t) = C\langle \ell \rangle^{-n} e^{-m\beta\epsilon_t}$ accounts well for the simulation data,[19] leading to the nice data collapse in Fig. 2(b). Fits give $C=0.9\pm0.1$, $m=3.3\pm0.2$, and $n=0.29\pm0.07$.

Since formation of multiple-height steps is energetically favorable for AT steps, collapse (bunching[20]) occurs once $\beta\epsilon_t$ is below a particular value, weakly dependent on $\langle \ell \rangle$ and reminiscent of the extensively studied problems of step pinning/(de)wetting[21] and doubling,[22] typically as a function of T for given ϵ_t. We find no multistep bunching for ϵ_t modestly negative ($0 > \beta\epsilon_t \gtrsim -0.25$). Concerned that our time and length were too small to see such bunching, we also simulated the evolution of AT steps with longer step edges (up to $L_y=10^4$). Rather than starting with equally spaced steps (our usual procedure), we chose an initial configuration of four step bunches, each with ten steps, and allowed the steps to evolve. We saw no evidence of further coalescence; rather, the initial bunches disperse. This behavior is qualitatively different from the unstable sensitive behavior when $\beta\epsilon_t \lesssim -0.3$ reflects the presence of a finite density of steps, the entropy of which apparently stabilizes the system.

In conclusion, using Monte Carlo simulations, we have shown that step touching broadens the TWD. The MGWD was found to give an excellent fit to the TWDs of both straight and fully kinked touching steps. Step touching leads to an effective attraction between steps, amounting to a finite-size correction of the step-step interaction strength ($\tilde{A}$). In light of such results, experimental studies seeking to extract $\tilde{A}$ should consider several different misorientations, checking for occurrence of step touching.

This work was supported by the U. Maryland NSF-MRSEC, Grant No. DMR 05-20471, with ancillary support from the Center for Nanophysics and Advanced Materials (CNAM). A.B.H. thanks B. Oujia for kind support of his research at Monastir. We thank K. Kim and A. Pimpinelli for helpful interactions and collaboration. T.L.E. thanks D. B. Abraham and J. D. Weeks for edifying comments.

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