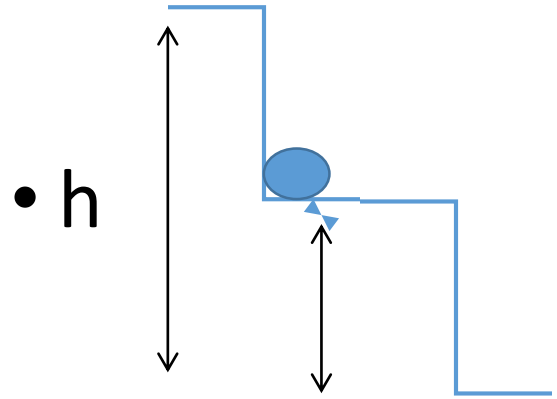
Roughening transition in crystal growth: Strong coupling limit maps into the weak coupling limit of the Kosterlitz-Thouless Coulomb gas

Layer by layer growth at low temperature

The steps are the nucleation center for crystal growth.

At the roughening transition, the surface become rough, the free energy of te step becomes zero.



Interface described by height h that are intergers

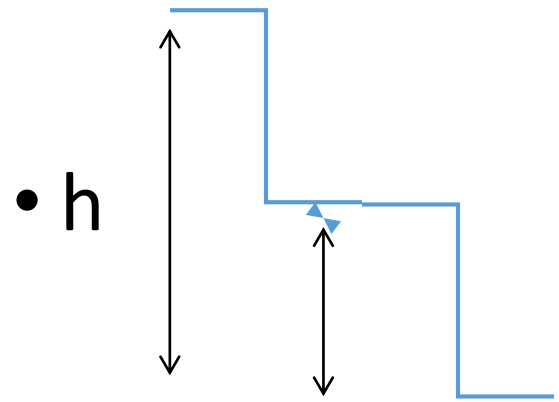
Energy

$$E = \sum_{i,\delta} -J/2(h_i - h_{i+\delta})^2$$

Partition function

$$Z = \int d\{h_i\} W(\{h_i\}) e^{-\beta E}$$

$$W(\{h_i\} = \prod_i \sum_n \delta(h_i - n)$$



At low temperatures, the surface is very smooth . It is very easy to calculate the partition function.Only small deviation from a smooth interface are important.

Energy

$$E = \sum_{i,\delta} -J/2(h_i - h_{i+\delta})^2$$

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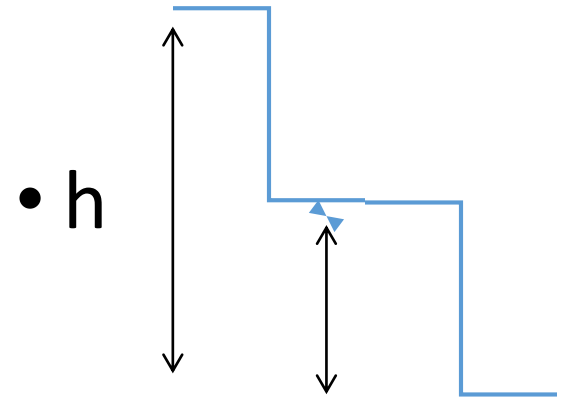

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Phase transition in the two-dimensional Coulomb gas, and the interfacial roughening transition

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The behavior of the interface in the three-dimensional (3-D) Ising and related lattice systems is modeled using a 2-D array of columns of varying heights. We show that the "roughening" transition (the transition from a localized to a delocalized interface) is directly related to the metal-insulator transition in a 2-D Coulomb gas. Implications of this relationship are discussed.

I. INTRODUCTION

In this paper we show a direct connection between the recently studied metal-insulator transition in a two-dimensional (2D) Coulomb gas[1-3] and the "roughening" phase transition for interfacial properties of the three-dimensional (3D) Ising and related lattice models.[4-6] Below the roughening temperature T_R the interface separating regions of "up" and "down" spins is localized while above T_R the interface wanders arbitrarily far from its $T=0$ location. The roughening transition has important consequences, both practically in the theory of crystal growth,[6] and more fundamentally in the basic theory of interfacial properties. The existence of the roughening transition points out difficulties in theories which associate the interface width with the bulk correlation length,[7,8] and provides a counterexample to the common supposition that the only singularities in interfacial thermodynamic properties occur at the bulk critical temperature.

II. DISCRETE GAUSSIAN MODEL

We introduce the following simplified interfacial model, which at low temperatures should closely mimic the properties of the interface in an isotropic 3D Ising model, or its anisotropic limit, the "solid-on-solid" (SOS) model.[9] Consider a square lattice made up of an $\sqrt{N}\times\sqrt{N}$ array of columns of varying heights h with interaction energy

$$E_c(\{h_i\}) = \tfrac{1}{2}J\sum_{i,\delta}(h_i - h_{i+\delta})^2 + 4JH\sum_i h_i^2 . \tag{1}$$

The first summation is over all lattice sites, denoted by the vector i, and the four nearest neighbors to site i, denoted by the vector δ. The second term introduces a dimensionless "magnetic field" H which tends to localize the interface near $h_i = 0$. We choose the lattice spacing and the coupling constant J as the units of length and energy.[10]

If the h_i vary uniformly between $\pm\infty$, Eq. (1) gives the *unweighted Gaussian* model of the interface.[11] One can show that as $H\to 0^+$, the interface width diverges logarithmically at any nonzero temperature. If, on the contrary, the h_i are restricted to be integers $0, \pm1, \pm2, \ldots$, there is a gap in the energy spectrum and the low-temperature behavior is similar to that of an interface in the 3D Ising model, which at low temperatures can be rigorously shown to have a finite interface width.[12,13] The interface in this *discrete Gaussian* (DG) model becomes delocalized at a finite nonzero T_R; the corresponding transition temperature in the unweighted Gaussian model is zero.

These examples suggest the study of the following partition function

$$Z_W = \int_{-\infty}^{\infty} d\{h_i\}\, W(\{h_i\}) \times \exp\left(-\tfrac{1}{2}\beta\sum_{i,\delta}(h_i - h_{i+\delta})^2 - 4\beta H\sum_i h_i^2\right), \tag{2}$$

where $\beta = (k_B T)^{-1}$ and W is a *weighting function* to be specified later. The integration is over continuous height variables for all the N columns. If $W(\{h_i\}) = 1$, the integration of (2) is elementary[11] and we obtain Z_U, the unweighted Gaussian model partition function, while the choice

$$W_D(\{h_i\}) = \prod_{i=1}^{N} w(h_i) = \prod_{i=1}^{N}\sum_{n_i=-\infty}^{\infty}\delta(h_i - n_i) \tag{3}$$

gives Z_D, the DG model partition function. Other choices of $W(\{h_i\})$ will be discussed below.

III. EQUIVALENCE TO A COULOMB GAS SYSTEM

We show in the Appendix

$$Z_D = Z_U Z_c , \tag{4}$$

where in the limit $H \to 0^+$,

$$Z_c \equiv \sum_{\{n_i\}=-\infty}^{\infty}{}' \exp\left(\tfrac{1}{2}\beta' \sum_{i\neq j} k_i k_j U(ij)\right) \tag{5}$$

is the partition function for a 2D lattice Coulomb gas at a dimensionless temperature $T' = (\beta')^{-1} = (k_B T/J)^{-1}$. The first summation is over all integers n_i for each site i. The "charges" $k_i \equiv 2\pi n_i$ are subject to the neutrality condition $\sum k_i = 0$. Since different values of k_i are permitted, Eq. (5) is the partition function for a mixture of charges.

The potential $U(jm)$, directly related to the 2D square lattice Green's function,[14] is

$$U(jm) = \frac{1}{8}\frac{1}{(2\pi)^2}\int_{-\pi}^{\pi} dq_x \int_{-\pi}^{\pi} dq_y \frac{1 - e^{iq(j-m)}}{1 - \frac{1}{2}(\cos q_x + \cos q_y)} . \tag{6}$$

An accurate approximation to $U(jm)$ for all $|j-m| \geq 1$ is $U(jm) = (4\pi)^{-1}(\ln|j-m| + \frac{1}{2}\ln 8 + \gamma)$, where γ is Euler's constant; thus we have a logarithmic (2D Coulomb) potential.

The same partition function Z_c occurs in the vortex model for 2D spin systems.[1,2] Assuming that only charges of unit strength are important (i.e., $|k_i| \leq 2\pi$), Kosterlitz and Thouless[1,2] presented strong evidence for a transition at T'_R from a low-T' dielectric phase with charges closely bound together in dipole pairs to a high-T' conducting plasma. They associated this transition in Z_c with a phase transition in the X-Y model.

Since Z_U is analytic, Eq. (4) shows that the transition in Z_c and the roughening transition in Z_D are different representations of the same transition. Our results differ in two important respects from the work of Kosterlitz and Thouless as applied to the X-Y model: (a) Eq. (4) is an *exact* relationship and (b) T in the DG system is related inversely to T' in the Coulomb gas system.

The two representations very nicely complement one another. The low-T' behavior of the Coulomb gas (non-conducting dielectric) is relatively easy to calculate since the charge density is small. Calculations in the high-T' high-density phase are more difficult.[1-3] Conversely, the low-T behavior of the DG model can be accurately determined using low-temperature series expansions and Monte Carlo simulations[15] while information above T_R is much more difficult to obtain. Previous analysis[1-6] of both systems separately using very different methods gave strong evidence for a phase transition; by combining the results and noting the internal consistency, we have hopes of determining in some detail the precise nature of the phase transition.

Kosterlitz[2] has given an estimate of the Coulomb gas transition temperature T'_R using Kondo-type renormalization-group equations. His Eq. (A18) predicts that the DG model has a roughening temperature $k_B T_R \cong 1.74$ J, which should be compared with the estimates from series expansions and Monte Carlo simulations on the SOS model of $k_B T_R \cong 1.2$ J.[4-6] These results are consistent since T_R for the DG system must be higher than for the SOS system, because equivalent excitations require equal or greater energy in the DG model than in the SOS model.[10]

IV. CORRELATION FUNCTION RELATIONS

Perhaps the most interesting results come from relating correlation functions in the DG system to those in the Coulomb gas system. This is easy to do since the partition functions are related to each other by Eq. (4). The second moment of the height difference between two columns arbitrarily labeled 1 and 2 a distance r apart is given by

$$G(12) \equiv \langle (h_1 - h_2)^2 \rangle_D = \sum_{h_1=-\infty}^{\infty}\sum_{h_2=-\infty}^{\infty} (h_1 - h_2)^2 P(h_1; h_2) , \tag{7}$$

where $P(h_1; h_2)$ is the probability of finding a column of height h_1 at site 1 and height h_2 at site 2. The limit as $r \to \infty$ of $G(12)$ gives a measure of the interface width.

There is a simple relationship between $G(12)$ and the potential of mean force $W_{MF}(12;Z)$ between two opposite "test" charges of magnitude Z on the sites 1 and 2, where

$$\exp[-\beta' W_{MF}(12;Z)] \equiv \left\langle \exp\left(\beta' Z \sum_j k_j [U(1j) - U(2j)] - \beta' Z^2 U(12)\right)\right\rangle . \tag{8}$$

The notation $\langle\ \rangle$ indicates a normalized ensemble average in the Coulomb gas system. Assuming the expansion

$$W_{MF}(12;Z) = Z^2 w^{(1)}_{MF}(12) + Z^4 w^{(2)}_{MF}(12) + \cdots , \tag{9}$$

we show in the Appendix that

$$G(12) = 2\beta' w^{(1)}_{MF}(12) . \tag{10}$$

At low T', the Coulomb gas system is a dielectric and $w^{(1)}_{MF}(r) \cong U(r)/\epsilon(T')$, where $\epsilon(T')$ is a dielectric constant which is an increasing function of T' with $\epsilon(0) = 1$. Hence $G(r)$ at high T should diverge as $A(T)\log(r)$ with $A(T)$ an increasing func-

tion of T. This is in precise agreement with recent Monte Carlo calculations of $G(r)$ for the SOS model.[16] At high T' in the conducting phase, $w_{MF}^{(1)}(r)$ reaches its asymptotic value exponentially fast because of screening; this again is in agreement with the Monte Carlo calculations.

Kosterlitz and Thouless[1,2] assumed that only unit charges ($|k_i| \leq 2\pi$) were important in the Coulomb gas system. Using Eq. (4), we can relate the average charge density at a site (i.e., the "ionic strength") to the DG system's internal energy $(\beta\Delta E/N)_D$ and correlation function $G(r)$:

$$\langle k^2\rangle = 8\beta\left[1 - 4\left(\frac{\beta\Delta E}{N}\right)_D \left\{1 - \frac{1}{4}\frac{G(\sqrt{2})}{G(1)} - \frac{1}{8}\frac{G(2)}{G(1)}\right\}\right]. \tag{11}$$

The DG energy $(\beta\Delta E/N)_D$ can be calculated very accurately at all T using the pair (Bethe) approximation.[17] Lower and upper bounds on $\langle k^2\rangle$ can then be easily calculated by assuming $G(r)$ increases as slowly as possible, where the term in curly brackets equals $\frac{5}{8}$, or as rapidly as possible where $G(r) = 2\beta U(r)$ and the same term equals $\frac{1}{2}$. If the approximation $w_{MF}^{(1)}(r) = U(r)/\epsilon(T')$ is good for $r = 1$, $\sqrt{2}$, and 2 for $T' < T_R'$, then the upper bound on $\langle k^2\rangle$ is in fact very close to the exact value. Assuming this, we have numerically calculated $\langle k^2\rangle$ as a function of T' and find that $\langle k^2\rangle$ is indeed very small at low T'. Here $\langle k^2\rangle \ll 8T'$. Near T_R' estimated by Kosterlitz however, $\langle k^2\rangle \cong 0.25$ and it rises rapidly thereafter to the limiting value $8T'$.

Since $\langle k^2\rangle$ is small for $T' < T_R'$, the higher k_i values in Eq. (5) implied by the discrete nature of the DG model are unimportant. Indeed *any* periodic $w(h_i)$ [see Eq. (3)] with nonzero Fourier components should give the same qualitative behavior (with a different T_R of course).

V. DIMENSIONALITY

As usual, effects of dimensionality are important. One can show that a d-dimensional DG model goes over to a d-dimensional lattice Coulomb gas. This suggests that the 2D case is the only one providing a finite nonzero T_R. For $d < 2$, the d-dimensional Coulomb potential $U_d(r) \sim r^{2-d}$ $(d \neq 2)$ binds a pair of opposite charges together so strongly that $\langle r^2\rangle$ is finite for all finite T'. This suggests that the $d < 2$ system is always an insulator. In this case $T_R = 0$. For $d > 2$ the attractive force between the dipole pair is much weaker and the dipole dissociates at any nonzero T'. This suggests that the $d > 2$ system is always a conductor and thus $T_R = \infty$. Only the marginal $d = 2$ case with $U_d(r) \sim \ln r$ seems to offer the possibility of a metal-insulator transition at finite nonzero T'. In terms of properties of the $d-1$ dimensional interface in a d-dimensional isotropic Ising model we then believe $T_R = 0$ for $d < 3$ and $T_R = T_c$ for $d > 3$, and $T_R \cong 1.2$ J for $d = 3$.[18,19]

VI. FINAL REMARKS

One might argue that by showing the equivalence of the discrete Gaussian model of the interface to a 2D Coulomb gas system, we have merely reexpressed one virtually intractable problem in terms of another. Strictly speaking, of course, this is correct, but we believe that it is precisely because of the difficulty of the problem that the equivalence is significant. In both systems, calculations are much more easily performed at low temperatures using, e.g., low-temperature expansions or Monte Carlo simulations[4-6] for the DG model and low-density expansions or an iterated mean field theory[1] in the Coulomb gas system. Since the temperature is inversely related in the two systems, a combination of the results allows an approximate but independent description of the system both above and below its transition temperature. The situation thus seems analogous to determining T_c or the critical exponents in the Ising model using only high-temperature series-expansion methods. One's confidence in the essential correctness of the results is certainly increased if low temperature series methods give a consistent picture, although one is no closer to giving an "exact" description of critical phenomena.

The methods used to predict the existence of the roughening[4-6] or the 2D Coulomb gas transition[1-3] have separately made a plausible, but by no means incontrovertible, case for a phase transition. This case seems more than doubly strengthened when the consistency of the picture from both the high- and low-temperature sides is realized. This is particularly so since the methods of approximation appropriate for the short-ranged DG system are very different from those used in the long-ranged Coulomb gas system. In a future paper we will present further results made possible by a recognition of the equivalence discussed herein.

ACKNOWLEDGMENTS

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APPENDIX

The results in this paper make use of the well-known identity for *integer* values of m and n (Ref. 20):

$$\sum_{m=-\infty}^{\infty} \delta(x-m) = \sum_{n=-\infty}^{\infty} e^{2\pi i n x}. \tag{A1}$$

From this follows at once

$$\delta(x-m) \quad \int_{-1/2}^{1/2} dz \exp[2\pi i z(x-m)] \sum_{n=-\infty}^{\infty} e^{2\pi i n x}. \tag{A2}$$

These identities can easily be derived by writing the usual integral representation of the δ function

$$\delta(x) = \int_{-\infty}^{\infty} e^{2\pi i z x} dz \tag{A3}$$

as a sum over all integers n of integrals from $n-\frac{1}{2}$ to $n+\frac{1}{2}$. The use of Eq. (A1) and Eq. (3) in Eq. (2) gives

$$Z_D = \sum_{\{n_j\}=-\infty}^{\infty} \int_{-\infty}^{\infty} d\{h_j\} \exp\Big(i \sum_j k_j h_j - \tfrac{1}{2}\beta \sum_{j,\delta} (h_j - h_{j+\delta})^2 - 4\beta H \sum_j h_j^2 \Big). \tag{A4}$$

Here $k_j \equiv 2\pi n_j$. Assuming periodic boundary conditions, the quadratic form in Eq. (A4) can be diagonalized by going to the Fourier-transformed variables

$$h_q = N^{-1/2} \sum_j h_j e^{iqj}, \tag{A5}$$

$$k_q = N^{-1/2} \sum_j k_j e^{iqj}, \tag{A6}$$

and Eq. (A4) rewritten as

$$Z_D = \sum_{\{n_j\}=-\infty}^{\infty} \int_{-\infty}^{\infty} d\{h_q\} \exp\Big(i \sum_q h_q k_{-q} - 4\beta \sum_q h_q h_{-q} [1 - \phi(q) + H] \Big), \tag{A7}$$

where

$$\phi(q) \equiv \tfrac{1}{2}(\cos q_x + \cos q_y). \tag{A8}$$

Note that since h_j (or k_j) is real, $h_{-q} = h_q^*$ (or $k_{-q} = k_q^*$). The integration over the $\{h_q\}$ can then be performed (doing the real and imaginary parts separately) making use of the formula

$$\int_{-\infty}^{\infty} dx\, e^{-ax^2 + ibx} = (\pi/a)^{1/2} e^{-b^2/4a}. \tag{A9}$$

This gives immediately

$$Z_D = \prod_q \Big(\frac{\pi}{8\beta[1 - \phi(q) + H]} \Big)^{1/2} \times \sum_{\{n_i\}} \exp\Big(- \sum_q \frac{|k_q|^2}{16\beta[1 - \phi(q) + H]} \Big). \tag{A10}$$

The first factor on the right hand side is the unweighted Gaussian model partition function[11] Z_U, and the second, denoted Z_c, can be rewritten using Eq. (A6) as

$$Z_c = \sum_{\{n_j\}} \exp\Big[- \tfrac{1}{2}\beta' \Big(\sum_j k_j \Big)^2 V(0) + \tfrac{1}{2}\beta' \sum_{j \neq m} k_j k_m U(jm) \Big], \tag{A11}$$

where $\beta' = \beta^{-1}$,

$$V(0) = \frac{1}{8N} \sum_q \frac{1}{1 - \phi(q) + H}, \tag{A12}$$

and

$$U(jm) = \frac{1}{8N} \sum_q \frac{1 - e^{iq(j-m)}}{1 - \phi(q) + H}. \tag{A13}$$

After taking the limit $N \to \infty$ the sums can be replaced by integrals. In the limit $H \to 0^+$, Eq. (A13) gives the Coulomb potential, Eq. (6), while $V(0)$ in Eq. (A12) tends to infinity. Using Eq. (A11) we see that only those configurations with $\sum_j k_j = 0$ will contribute to the partition function Z_c, thus giving the neutrality condition. This establishes the basic Eqs. (4) and (5).

Other results in this paper are proved in a similar manner. For example, to establish Eq. (10) we first write $P(n_1; n_2)$, the probability of finding column 1 at height n_1 and column 2 at height n_2, as

$$P(n_1;n_2) = Z_D^{-1} \int_{-\infty}^{\infty} dh_1\, \delta(h_1 - n_1) \int_{-\infty}^{\infty} dh_2\, \delta(h_2 - n_2) \int_{-\infty}^{\infty} dh_3 \sum_{n_3=-\infty}^{\infty} \delta(h_3 - n_3) \cdots \int_{-\infty}^{\infty} dh_N \sum_{n_N} \delta(h_N - n_N) \times \exp\Big[- \tfrac{1}{2}\beta \sum_{i,\delta} (h_i - h_{i+\delta})^2 - 4\beta H \sum_i h_i^2 \Big]. \tag{A14}$$

Using Eqs. (A1) and (A2) in Eq. (A14), and retracing the algebra which led to Eq. (A11), we find in the limit $H \to 0^+$ that $P(n_1;n_2)$ is a function only of the height difference $\Delta n \equiv n_1 - n_2$, with

$$P(\Delta n) = \int_{-\pi}^{\pi} dZ\, e^{iZ\Delta n} \exp[-\beta' W_{\mathrm{MF}}(12;Z)]. \quad \text{(A15)}$$

$W_{\mathrm{MF}}(12;Z)$ is defined in Eq. (8). Then, using Eq. (7),

$$G(12) = \sum_{\Delta n=-\infty}^{\infty} (\Delta n)^2 P(\Delta n) = \sum_{\Delta n=-\infty}^{\infty} \int_{-\pi}^{\pi} dZ (\Delta n)^2 e^{iZ\Delta n} \exp[-\beta' W_{\mathrm{MF}}(12;Z)]. \quad \text{(A16)}$$

Equation (10) follows at once, using Eq. (9) and Eq. (A1) differentiated twice.

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[8]B. Widom, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1972), Vol. 2, p. 79.

[9]The SOS model occurs when the vertical coupling constant J_z between spins on adjacent lattice planes approaches infinity. See Refs. 4 and 17 for further discussion. The model we introduce below has the same first terms in a low-temperature expansion of the interfacial properties as the SOS model or the isotropic Ising model.

[10]Note that $J = 2J^I$ where J^I is the coupling constant in the associated Ising model. The SOS model differs from the DG model only in using $|h_i - h_{i+\delta}|$ rather than $(h_i - h_{i+\delta})^2$ in Eq. (1).

[11]Although the physical interpretation of the unweighted Gaussian model is different, the mathematics is the same as the Gaussian model discussed by T. H. Berlin and M. Kac [Phys. Rev. 86, 821 (1952)]. A continuum version of the interface model was discussed by F. P. Buff, R. A. Lovett, and F. H. Stillinger [Phys. Rev. Lett. 15, 621 (1965)].

[12]R. L. Dobrushin, Theor. Prob. Appl. (USSR) 17, 619 (1972).

[13]H. van Beijeren, Commun. Math. Phys. 40, 1 (1975).

[14]F. Spitzer, *Principles of Random Walk* (Van Nostrand, Princeton, N.J., 1964), pp. 148–151. Equation (6) results after taking the limit $N \to \infty$.

[15]The methods used in Refs. 4–6 for the SOS model can be immediately extended to the DG model.

[16]See Ref. 6, p. 176; and G. H. Gilmer (unpublished work).

[17]The pair approximation energy for the SOS model gave quantitative agreement with Monte Carlo calculations. See J. D. Weeks and G. H. Gilmer, J. Cryst. Growth 33, 21 (1976). See also J. D. Weeks and G. H. Gilmer, J. Chem. Phys. 63, 3136 (1975).

[18]The fact that $T_R = 0$ for the 2D Ising-model interface was proved by G. Gallavotti [Commun. Math. Phys. 27, 103 (1972)]. Clearly $T_R = 0$ for the 1D Ising model. As far as we know, no work has been done on the $d > 3$ case.

[19]As discussed before, $T_R^{\mathrm{DG}} \geq T_R^{\mathrm{SOS}}$. A simple mean field argument suggests that for an anisotropic Ising model with coupling $J_\perp$ between different layers and $J_\parallel$ in the same layer, T_R should be an increasing function of $J_\perp$. Thus $T_R^{\mathrm{DG}} \geq T_R^{\mathrm{SOS}} \geq T_R^I$, the isotropic Ising model's roughening temperature. Actually, series-expansion methods and Monte Carlo results (Refs. 4–6) suggest that T_R depends very weakly on $J_\perp$ and $T_R^{\mathrm{SOS}} \cong T_R^I$. Note that Kosterlitz's estimate for T_R^{DG} is less than $T_c^{3\mathrm{D}}$, the 3D isotropic Ising model's critical temperature. A lower bound $T_R^I \geq T_c^{2\mathrm{D}}$ was proven in Ref. 13.

[20]See, e.g., I. M. Gel'fand and G. E. Shilov, *Generalized Functions* (Academic, New York, 1964), Vol. 1, p. 332.