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Roughening transition in mean-field and pair approximation of Ising models

J. P. van der Eerden
Laboratory of Physical Chemistry, University of Technology, Delft, The Netherlands

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It is shown that the macroscopic phenomena of the roughening transition in an Ising interface model can be connected with a property, corresponding to a second-order phase transition, of the local thermodynamic potential for interfaces with an even symmetry. Using this connection a method is developed to evaluate the roughening temperature in first- and second-order mean-field approximations of a simple-cubic crystal. These values are compared with Monte Carlo results and with values found from the fundamental assumption that thermodynamic quantities such as the specific heat should have singularities at the transition temperature. It turns out that the second-order results are very accurate. Finally then, the method is generalized to give results for fcc, bcc, and hexagonal structures.

I. INTRODUCTION

Below its critical temperature an Ising system has two possible bulk phases. An interface model describes the system if the two bulk phases are simultaneously present. At very low temperatures such a two-phase system will contain both phases separated by an almost flat surface. At higher temperatures this surface will become more and more rough until at the critical temperature the two phases become identical and the surface is no longer present.

At a certain temperature T_R below the critical temperature the surface becomes so rough that there is no longer any tendency to form surfaces that are oriented along crystallographic directions. We shall call this temperature the roughening temperature and the corresponding phenomenon the roughening transition of the Ising model.

In the following we select from the possible applications of the Ising model the solid-fluid lattice model, i.e., each cell of the lattice is either solid or fluid. This interpretation is very useful for the science of the growth and structure of crystal surfaces because it describes various systems such as crystal-melt, crystal-vapor, or crystal-solution. Also, the roughening temperature is probably best visualized with the morphology of a crystal: Below the roughening temperature a crystal is bounded by low-index areas that are almost flat on atomic scale, above T_R the whole surface becomes rough and the edges between the originally flat regions become rounded off. The growth mechanism at low supersaturation also changes at T_R. Below T_R the crystal grows by attachment of new atoms to step edges (i.e., nucleation or dislocation growth), above T_R these steps are hardly recognizable because small and large clusters are present all over the surface. From this reasoning it is clear that different roughening temperatures belong to each surface orientation. The roughening temperature of the crystal is the highest of these temperatures.

The idea, however, of a roughening transition is general and applies also to other Ising-type models such as the magnetic Ising model and the binary systems used in metallurgy.

Although these macroscopic phenomena are quite clear and generally accepted it turns out to be rather difficult to relate them to quantities in the microscopic statistical-mechanical theory.

Burton, Cabrera, and Frank[1] pointed out that at the transition some thermodynamic quantities exhibit singularities. They found in 1951 that the roughness versus temperature curve has a point of inflection and the specific heat, therefore, has a maximum. Temkin[2] found the same in his model in 1966. They associated this point with the roughening transition but neither of them proves directly that it is really related to the morphological change mentioned above.

Leamy, Gilmer, and Jackson[3] relate the roughening temperature with Monte Carlo data by looking for a temperature where the excess edge energy of a step vanishes. In this way they determine the critical temperature for phase separation in a layer. On the other hand they associate the roughening transition with what they call a divergence of the interface: Above the roughening temperature the surface is free to move up and down, therefore the interface width will become infinite at T_R. This divergence, however, is of long-range type and therefore it is not surprising that such a behavior is not found in static-mean-field-type models, as was proved recently by Weeks and Gilmer.[4,5] Their conclusion, however, that these models possess no roughening transition is premature.

To overcome these difficulties we make a new start. We shall define a local potential function depending on the concentration of the central layer between the two phases only (by minimizing the model grand potential function with respect to all other independent variables). Below T_R this function will have certain maxima and minima, indicating that some concentration profiles (namely, those who lead to flat surfaces) are favorable while others are not, and that, therefore, flat surfaces probably occur in the interface. At and above T_R the function will be constant and therefore the interface is free to move.

Using this criterion we give a method for computing the roughening temperatures in mean-field approximations of Ising models. In such a way we have succeeded in constructing a logical connection between the macroscopic definition and a theoretically and numerically accessible method for computing T_R.

Two types of lattice models are frequently used, the solid-on-solid (SOS) model, where each solid cell is situated above another solid cell (crystal science), and the unrestricted (unr) model, where each distribution of solid cells over the lattice is allowed (metallurgy, magnetism). The roughening temperatures of the first- (MFA, mean-field approximation) and second- (pair) order approximations for these two types of model are given. The assumption that the two types are equivalent up to the roughening temperature is confirmed. Possible generalizations of the method could be other crystal symmetries and longer-range interactions.

II. EXPRESSIONS FOR THE GRAND POTENTIAL

The state of a simple-cubic crystal is described by the distribution of solid and fluid cells over the cubic lattice. We number the lattice layers perpendicular to the z axis with the parameter i and use the following variables for an approximate (mean-field-type) description of the system: N_i^p: number of cells of type p in layer i; $N^{pq\alpha}$: number of cells of type p that have a cell of type q as their neighbor in the positive α direction; $p=S$ (solid) or F (fluid); $\alpha = x$, y or z.

An example of the use of the variables is given in Fig. 1. From simple geometric arguments it can be seen that it is possible to express these dependent variables in terms of the independent variables N_i^S and $N_i^{SF\alpha}$ (N is number of cells in a layer):

$$N_i^F = N - N_i^S\,, \tag{1}$$

$$N_i^{FS\alpha} = N_i^{SF\alpha}\,, \tag{2}$$

$$N_i^{FF\alpha} = N_i^F - N_i^{SF\alpha}\,, \tag{3}$$

$$N_i^{SS\alpha} = N_i^S - N_i^{SF\alpha}\,, \tag{4}$$

if $\alpha = x$ or y, and

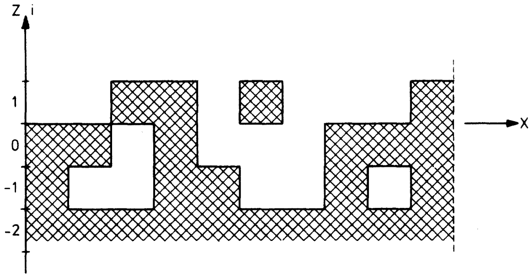

FIG. 1. Example of an unrestricted interface (only x-z plane shown) $N_1^S=4$, $N_1^{SSx}=1$, $N_0^{SFz}=4$, $N_0^{FSz}=2$, $N_{-1}^{SFx}=3$, $N_{-1}^{FSx}=3$, $N=10$.

$$N_i^{FSz} = N_i^{SFz} + N_{i+1}^S - N_i^S\,, \tag{5}$$

$$N_i^{FFz} = N_{i+1}^F - N_i^{SFz}\,, \tag{6}$$

$$N_i^{SSz} = N_i^S - N_i^{SFz}\,. \tag{7}$$

In general there are nonzero interactions $\phi^{pq\alpha}$ between cells of both types. We shall restrict ourselves to nearest-neighbor interactions dependent on the direction α and on the types p and q of cells whose interaction is considered. In such a case it is useful to define a generalized interaction parameter $\vec{\omega} = (\omega^x, \omega^y, \omega^z)$,[6] thus establishing the equivalence of a solid-fluid lattice model and a lattice gas model (each cell of the lattice is either empty or occupied), where only interactions between the occupied cells are considered:

$$\omega^\alpha = (1/2kT)(\phi^{SS\alpha} + \phi^{FF\alpha} - 2\phi^{SF\alpha})\,. \tag{8}$$

The total interaction energy E can be simplified,

$$\frac{E}{kT} = -2 \sum_{i=-\infty}^{\infty} \sum_{\alpha=x}^{z} \omega^\alpha N_i^{SS\alpha}\,, \tag{9}$$

and the equilibrium value of the chemical potential is the opposite of the interaction energy per cell of a totally solid lattice,[3]

$$\mu_e/kT = -2\omega^x - 2\omega^y - 2\omega^z\,. \tag{10}$$

From straightforward combinatorial computations the entropy S of the system is found to be[7]

$$\frac{S}{k} = \sum_{i=-\infty}^{\infty} \ln \binom{N}{N_i^S} \prod_{\alpha=x}^{z} \left[\binom{N_i^S}{N_i^{SS\alpha}} \binom{N_i^F}{N_i^{FF\alpha}} \Big/ \binom{N}{N_i^S} \right]. \tag{11}$$

The proper potential function for this open system at constant temperature is the grand potential, and divided by NkT it is given by

$$\Omega = \frac{E}{NkT} + \frac{S}{Nk} - \frac{\mu_e}{NkT} \sum_{i=-\infty}^{\infty} N_i^S\,. \tag{12}$$

In the following we will use concentrations $C_i^p = N_i^p/N$, $C_i^{pq\alpha} = N_i^{pq\alpha}/N$, which leads to the following expressions for the grand potential in the cases we want to consider:

The unrestricted pair formula is found by substituting (4) and (7)–(11) in (12):

$$\Omega_{\text{unr}}^{\text{pair}} = \sum_{i=-\infty}^{\infty} \left(\sum_{\alpha=x}^{z} 2\omega^\alpha C_i^{SF\alpha} - 5C_i^S \ln C_i^S - 5C_i^F \ln C_i^F + \sum_{\alpha=x}^{z} \sum_{p,q=S}^{F} C_i^{pq\alpha} \ln C_i^{pq\alpha} \right). \tag{13}$$

The SOS restriction is built in by substituting $C_i^{FSz} \to 0+$ in (5) and (13):

$$\Omega_{\text{SOS}}^{\text{pair}} = 2\omega^z + \sum_{i=-\infty}^{\infty} \left(2\omega^x C_i^{SFx} + 2\omega^y C_i^{SFy} - 4C_i^S \ln C_i^S - 4C_i^F \ln C_i^F + \sum_{\alpha=x}^{y} \sum_{p,q=S}^{F} C_i^{pq\alpha} \ln C_i^{pq\alpha} \right) + (C_i^S - C_{i+1}^S) \ln (C_i^S - C_{i+1}^S). \tag{14}$$

Substituting $C_i^{pq\alpha} = C_i^p C_i^q$ if $\alpha = x$ or y and $C_i^{pqz} = C_i^p C_{i+1}^q$ in (13) we get the mean-field expression[8]:

$$\Omega_{\text{unr}}^{\text{MFA}} = \sum_{i=-\infty}^{\infty} [2(\omega^x + \omega^y) C_i^S C_i^F + 2\omega^z C_i^S C_{i+1}^F + C_i^S \ln C_i^S + C_i^F \ln C_i^F]. \tag{15}$$

Finally the SOS-MFA expression[2] is again found from (14) by $C_i^{pq\alpha} = C_i^p C_i^q$:

$$\Omega_{\text{SOS}}^{\text{MFA}} = 2\omega^z + \sum_{i=-\infty}^{\infty} [2(\omega^x + \omega^y) C_i^S C_i^F + (C_i^S - C_{i+1}^S) \ln (C_i^S - C_{i+1}^S)]. \tag{16}$$

In the SOS-type models the first term $2\omega^z$ results from the boundary condition $C_\infty = 0$ and $C_{-\infty} = 1$. Therefore, it has no influence on the structure of the surface (therefore, neither on the roughening temperature). This is the consequence of the SOS restriction in which the bulk phases contain either only solid or only fluid cells. Such phases are only relevant for physical application at $T = 0$, and we have to realize that these models are in themselves interface models. The functions given in (14) and (16) are, therefore, surface quantities. The term, then, enables us to estimate a limiting temperature below which the model can be used: If the total expression (14) or (16) becomes negative the surface tension also becomes negative; therefore, the temperature at which this happens is referred to as the critical temperature of the model.[9]

III. DERIVATION OF THE MICROSCOPIC CRITERION

The equilibrium values of the variables we defined are those for which the grand potential is minimal. Taking, therefore, the first-order partial derivatives equal to zero gives for each model a set of equations for the independent variables which have to be solved:

pair-unr:

$$5 \ln \frac{C_i^F}{C_i^S} + \ln \frac{C_i^{SSx}}{C_i^{FFx}} \frac{C_i^{SSy}}{C_i^{FFy}} + \ln \frac{C_i^{SSz}}{C_{i-1}^{FFz}} \frac{C_{i-1}^{FSz}}{C_i^{FSz}} = 0, \tag{17a}$$

$$C_i^{SF\alpha} C_i^{FS\alpha} = e^{-2\omega^\alpha} C_i^{SS\alpha} C_i^{FF\alpha}; \tag{17b}$$

pair-SOS:

$$4 \ln \frac{C_i^F}{C_i^S} + \ln \frac{C_i^{SSx}}{C_i^{FFx}} \frac{C_i^{SSy}}{C_i^{FFy}} + \ln \frac{C_i^S - C_{i+1}^S}{C_{i-1}^S - C_i^S} = 0, \tag{17c}$$

$$C_i^{SF\alpha^2} = e^{-2\omega^\alpha} C_i^{SS\alpha} C_i^{FF\alpha}; \tag{17d}$$

MFA-unr:

$$2(\omega^x + \omega^y)(1 - 2C_i) + 2\omega^z (1 - C_{i+1} - C_{i-1}) - \ln (C_i^F / C_i^S); \tag{17e}$$

MFA-SOS:

$$2(\omega^x + \omega^y)(1 - 2C_i) + \ln \frac{C_i^S - C_{i+1}^S}{C_{i-1}^S - C_i^S}. \tag{17f}$$

From these equations it can be seen that two types of solution are possible ($\bar{S} = F$, $\bar{F} = S$):

odd solution:

$$C_i^S = C_{-i}^F, C_i^{pq\alpha} = C_{-i}^{\bar{q}\bar{p}\alpha} \text{ if } \alpha = x \text{ or } y, C_i^{pqz} = C_{1-i}^{\bar{q}\bar{p}z}, \tag{18a}$$

even solution:

$$C_i^S = C_{1-i}^F, C_i^{pq\alpha} = C_{1-i}^{\bar{\bar{q}}\bar{\bar{p}}\alpha} \text{ if } \alpha = x \text{ or } y, C_i^{pqz} = C_{-i}^{\bar{\bar{q}}\bar{\bar{p}}z}. \tag{18b}$$

Thus the solutions possess a mirror symmetry leading to either an atom-center boundary (odd solution) or an interstitial-center boundary (even solution) as they were named by Kikuchi and Cabin.[10]

In the atom-center boundary the central layer is half filled, therefore, the roughness of the interface will be high, whereas the interstitial-center boundary corresponds to a more flat surface. So the crystal will have flat surfaces if the even solution has a lower potential than the odd solution. At low temperatures this condition will be fulfilled (at $T=0$ the interface is given by $C_{-i}=1, C_{i+1}=0, i \geqslant 0$, which is an example of an interstitial-center boundary). The roughening temperature is thus the temperature where the grand potential of the interstitial boundary becomes equal to the grand potential of the atom-center boundary. At temperatures below T_R the even solution corresponds to a minimum in the potential, the odd solution to a saddle point.

Upon minimizing Ω with respect to all independent variables except the concentration C_0 of the central layer [i.e., solving all the equations (17) except the one derived from $\partial\Omega/\partial C_0=0$] these other variables are expressed in terms of C_0. Substituting these values back in the expressions for the grand potential (13)–(16) one defines a probability function for C_0:

$$\Omega_T^*(C_0) \equiv \Omega(T, C_0, C_i(C_0), C_i^{SF\alpha}(C_0)) - \Omega_{\text{even}}(T), \tag{19}$$

where Ω_{even} is the grand potential of the even solution. The shape of this probability function below T_R is given schematically in Fig. 2. When C_0 is equal to any concentration of the even solution all the other concentrations will be such that an interstitial-center boundary is formed, so these values lead to minima in Ω_T^*. On the other hand when C_0 equals one of the concentrations of the even solution an atom-center boundary will result and these values correspond to maxima in Ω_T^*. When T rises towards T_R the difference between the minimal and maximal value will shrink down until at $T \geqslant T_R$ the function is constant:

$$\Omega_T^*(C_0)=0 \text{ if } T \geqslant T_R. \tag{20}$$

To get a criterion which is more accessible for numerical calculation of the roughening temperature we go one step further. Again we minimize Ω with respect to all variables except C_0 but now subject to the constraint that the boundary should be of the atom-center type for all concentrations except for C_0:

$$\begin{gathered} \tilde{\Omega}_T(C_0)=\Omega(T, C_0, C_i(C_0), C_i^{pq\alpha}(C_0))-\Omega_{\text{even}}(T), \\ C_i+C_{-i}=1 \text{ if } i \neq 0. \end{gathered} \tag{21}$$

The shape of this potential function is given in Fig. 3. For $T<T_R$ the function has two minima corresponding to configurations where the even solution is best imitated [$C_{-1}-C_0 \approx (C_0-C_1)_{\text{even}}$ or $C_0-C_1 \approx (C_0-C_1)_{\text{even}}$, these layers give the main contribution to the potential]. At $C_0=\frac{1}{2}$ a relative maximum is present and the whole boundary is of the atom-center type. At temperatures above T_R the interface is by the symmetry restriction forced towards the atom-center boundary, so then $C_0=\frac{1}{2}$ is a minimum. We now have a simple criterion for the roughening temperature:

$$\tilde{\Omega}''_{T_R}(\tfrac{1}{2})=0. \tag{22}$$

This property of $\tilde{\Omega}$ can easily be transformed into a property for the total grand potential Ω giving:

$$\begin{aligned} \tilde{\Omega}''_{T_R}(\tfrac{1}{2}) = {} & \frac{\partial^2\Omega}{\partial C_0^2}+\sum_{i\neq 0,\alpha}\frac{\partial^2\Omega}{\partial C_0\,\partial C_i^{SF\alpha}}\frac{dC_i^{SF\alpha}}{dC_0} \\ & +\sum_{i\neq 0}\frac{\partial^2\Omega}{\partial C_0\,\partial C_i}\frac{dC_i}{dC_0}=0, \end{aligned} \tag{23}$$

and the dependence of the C_i and $C_i^{SF\alpha}$ on C_0 is given by the solution of the Eqs. (17) under the symmetry constraint. The partial derivatives have to be computed for the odd solution.

The method of finding the roughening temperature is (i) derive the minimization Eqs. (17); (ii) compute $(dC_i/dC_0)_{C_0=1/2}$ and $(dC_i^{SF\alpha}/dC_0)_{C_0=1/2}$ from

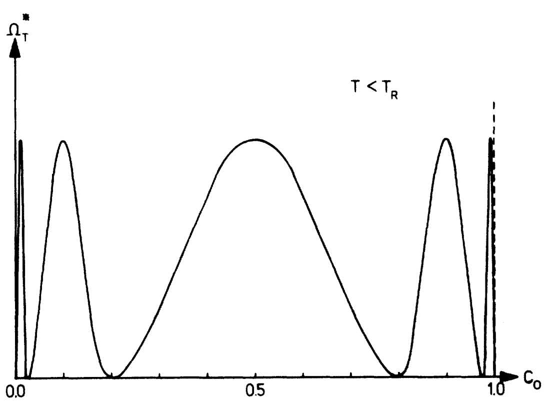

FIG. 2. Unconstrained local thermodynamic potential.

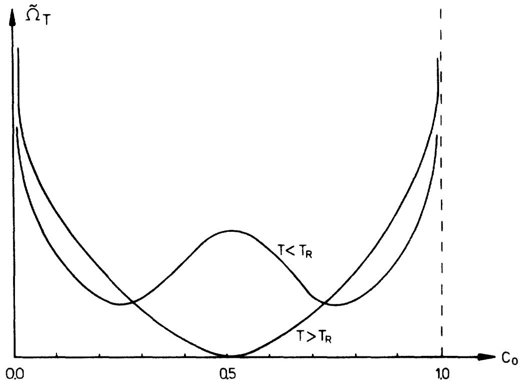

FIG. 3. Evenly constrained local thermodynamic potential.

these equations; (iii) solve from the minimization Eqs. (17) and the roughening Eq. (23) the interface profile $(\ldots, C_{-1}, C_0, C_1, \ldots)$; (iv) determine the temperature T_R for which this is a physical interface.

IV. APPLICATION TO THE MODELS

The first step of the method has already been carried out, so now we will look at the second. Because of the symmetry constraint we have

$$\frac{dC_i}{dC_0}+\frac{dC_{-i}}{dC_0}=0\,, \quad i=1,2,\ldots\,. \tag{24}$$

This property makes the last term in (23) zero, so in the MFA models only the first term survives. From the second term only the horizontal pairs in the zeroth layer and the vertical pairs in the zero and minus one layer are nonzero in the expression. For horizontal pairs the equation $\partial\Omega/\partial C_0^{SF\alpha}=0$ can be solved easily, giving in the case that $C_0=\frac{1}{2}$:

$$C_0^{SS\alpha}=C_0^{FF\alpha}=C_0^{SF\alpha}e^{\omega^\alpha}=\tfrac{1}{2}(1+e^{-\omega^\alpha})^{-1}\,, \quad \alpha=x \text{ or } y$$

$$\frac{dC_0^{SF\alpha}}{dC_0}=0\,, \quad \alpha=x \text{ or } y\,. \tag{25}$$

Finally from the equation $\partial\Omega/\partial C_i^{SFz}=0$ one derives:

$$\frac{dC_i^{SFz}}{dC_i}=-\frac{dC_i^{SFz}}{dC_{i+1}}$$

$$=\left(\frac{1}{C_i^{FSz}}+\frac{1}{C_i^{SSz}}\right)\left(\frac{1}{C_i^{FSz}}+\frac{1}{C_i^{SSz}}+\frac{1}{C_i^{SFz}}+\frac{1}{C_i^{FFz}}\right)^{-1}. \tag{26}$$

Applying this and the symmetry constraint to layers zero and minus one gives

$$\frac{dC_0^{SFz}}{dC_0}+\frac{dC_{-1}^{SFz}}{dC_0}=0\,. \tag{27}$$

The roughening equations then are

pair-unr:

$$-12+4e^{-\omega^x}+4e^{-\omega^y}+4C_1^SC_1^F(C_0^{FSz}C_0^{FFz}+C_0^{SSz}C_0^{SFz})^{-1}=0\,; \tag{28a}$$

pair-SOS:

$$-8+4e^{-\omega^x}+4e^{-\omega^y}+2(\tfrac{1}{2}-C_1)^{-1}=0\,; \tag{28b}$$

MFA-unr:

$$4-4\omega^x-4\omega^y=0\,; \tag{28c}$$

MFA-SOS:

$$-4\omega^x-4\omega^y+2(\tfrac{1}{2}-C_1)^{-1}=0\,. \tag{28d}$$

The roughening temperature for the mean-field unrestricted model is directly found from (28c):

$$T_R=(1/2k)(\phi^{SSx}+\phi^{SSy}+\phi^{FFx}+\phi^{FFy}-2\phi^{SFx}-2\phi^{SFy})$$
$$\equiv\phi/k\,, \tag{29}$$

where we defined the average interaction strength ϕ by the last equality. From now on we shall only consider the isotropic case $\omega^x=\omega^y=\omega^z$ (anisotropic results will be given in a paper about the Monte Carlo simulation of such systems[11]) and use this definition of ϕ. For the SOS models Eq. (28) can be seen as an equation for C_1. In the pair-unr model Eq. (28) together with $\partial\Omega/\partial C_0^{SFz}=0$ forms a set of two equations in the two unknowns, C_1 and C_0^{SFz}. So in all cases C_0 and C_1 are known for each temperature and the structure of the set of equations (17) is such that the whole concentration profile is found easily by a numerical procedure. This profile, however, will only be physically relevant if it meets the proper concentration boundary conditions. This will happen for only one temperature (Figs. 4 and 5), which is therefore the roughening temperature.

V. RESULTS AND DISCUSSION

Table I gives the critical temperature, the roughening temperature as computed with our method, the temperature where the roughness curve has a point of inflection, and the two-dimensional critical temperature. The first thing to mention is that the roughening temperature is predicted very accurately in the pair approximation.

The next important remark is that the roughening temperature agrees within the given accuracy of the transition temperature computed from the roughness curve. This also holds when the interface is restricted to a finite number of layers (this can be achieved by replacing the proper boundary condition $C_\infty=C_{\text{bulk}}$ by $C_i=C_{\text{bulk}}$ for i greater than a certain number n). Therefore it seems legitimate to identify the two transitions, although a rigorous proof of this statement has not yet been found. On comparing then the two ways to compute the transition temperature our method has not only the advantage of the clear correspondence between the macroscopic and microscopic phenomena but also requires less effort to compute the temperature to a great accuracy.

Often the critical temperature of the two-dimensional lattice is used as an estimate for the roughening temperature. This estimate is based on the idea that below the roughening temperature the interface mainly consists of only one layer. The roughening equations for a one layer interface (constructed by the boundary condition $C_1^S=C_{-1}^F=0$) are indeed the equations for the critical temperature of the two-dimensional lattice! From the table it is seen that this estimate leads to a 12%

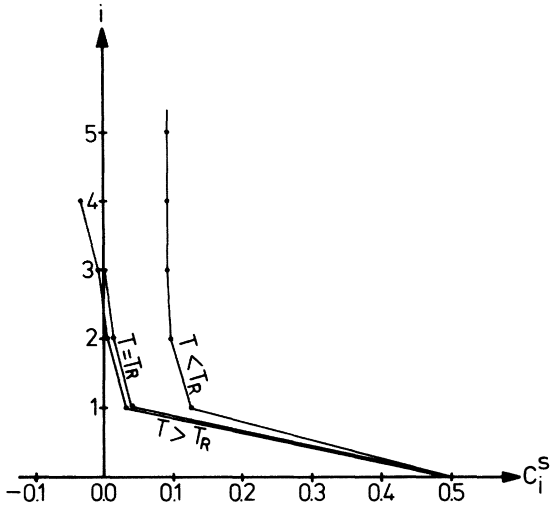

FIG. 4. SOS profiles, evaluated from Eq. (28b).

TABLE I. Transition temperature in several models.

	kT_c/ϕ	kT_R/ϕ	kT_{inf}/ϕ	$kT_c^{2\text{-dim}}/\phi$
MFA-unr	1.5	1.0		1.0
MFA-SOS	1.11 [a]	0.855	0.82± 0.05 [f]	1.0
Pair-unr	1.235 [b]	0.676		0.719 [b]
Pair-SOS		0.649	0.64± 0.02 [g]	0.719 [b]
Exact unr	1.136 [c]	~0.6 [d]		0.568 [h]
Exact SOS	1.02 [a]	0.64 ± 0.02 [e]		0.568 [h]

[a] Comment, see below Eq. (16), values from Ref. 9.

[b] Exact formula e.g. in Ref. 12.

[c] Monte Carlo result in Ref. 13.

[d] Low temperature expansion in Ref. 4.

[e] Monte Carlo result in Ref. 3, low temperature expansion: $T_R \approx 0.62$.

[f] Value given in Ref. 2.

[g] Recalculation from the Bethe method in Ref. 1 which is equivalent to the pair-SOS model.

[h] Exact formula from Onsagers method (Ref. 14).

lower transition temperature indicating that the third dimension tends to stabilize the surface. Note, however, that in the mean-field-type approximations (to the same order) this order is reversed. This effect should be mainly attributed to the fact, however, that the two-dimensional critical temperature is not very well approximated even in the pair model.

From Table II the convergence from the unr model to the SOS model can be seen in the pair approximation. This behavior corresponds of course to the well-known fact that the SOS model is equivalent to a unr model with infinite vertical interactions. The only 4% difference in T_R justifies the use of SOS models for temperatures below T_R.

The whole method can easily be generalized to more complicated bond structures. If there are h directions for the horizontal bonds and v for the vertical ones the pair equations in (17) and (28) are modified into

pair-unr:

$$(2h+2v-1)\ln\frac{C_i^F}{C_i^S}+\sum_\alpha \ln\frac{C_i^{SS\alpha}}{C_i^{FF\alpha}} + \sum_\beta \ln\frac{C_i^{SS\beta}C_{i-1}^{FS\beta}}{C_{i-1}^{FF\beta}C_i^{FS\beta}}=0\,, \quad (17a')$$

$$\left.\begin{aligned} &C_i^{SF\alpha^2}=e^{-2\omega^\alpha}C_i^{SS\alpha}C_i^{FF\alpha}\,,\\ &C_i^{SF\beta}C_i^{FS\beta}=e^{-2\omega^\beta}C_i^{SS\beta}C_i^{FF\beta}\,, \end{aligned}\right. \quad (17b')$$

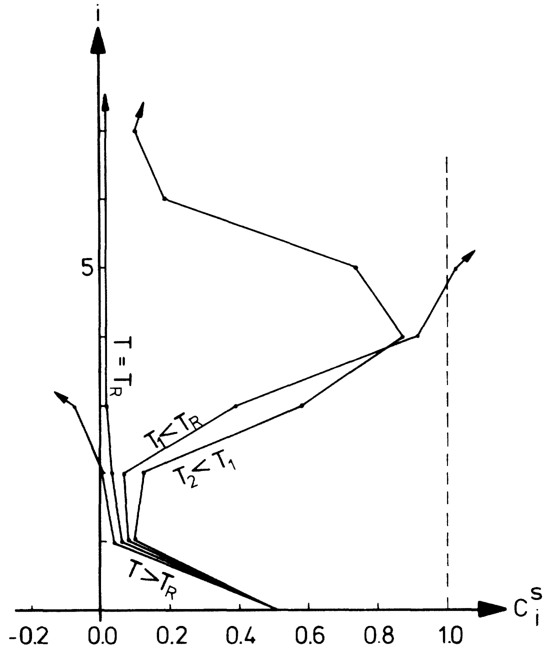

FIG. 5. unr profiles, evaluated from Eq. (28a).

TABLE II. Dependence of T_R on the bond strength in the vertical direction.

ϕ^z/ϕ^x	kT_R/ϕ^x
1.0	0.676
2.0	0.655
3.0	0.651
4.0	0.650
5.0	0.649
∞	0.649

$$-4(2h+v-1)+4\sum_{\alpha} e^{-\omega^{\alpha}}$$

$$+4C_1^S C_1^F \sum_{\beta} (C_0^{FS\beta} C_0^{FF\beta} + C_0^{SS\beta} C_0^{SF\beta})^{-1} = 0 ; \quad (28a')$$

pair-SOS:

$$(2h+v-1)\ln\frac{C_i^F}{C_i^S} + \sum_{\alpha} \ln\frac{C_i^{SS\alpha}}{C_i^{FF\alpha}} + v\ln\frac{C_i^S - C_{i+1}^S}{C_{i-1}^S - C_i^S} = 0 , \quad (17c')$$

$$C_i^{SF\alpha^2} = e^{-2\omega^{\alpha}} C_i^{SS\alpha} C_i^{FF\alpha} , \quad (17d')$$

$$-4(h+v-1)+4\sum_{\alpha} e^{-\omega^{\alpha}} + 2v(\tfrac{1}{2} - C_1^S)^{-1} = 0 , \quad (28b')$$

where β runs over all vertical pairs and α over all horizontal ones. With these equations we evaluated the results of Table III, again assuming all ω to be equal. From Table III we see that in general the horizontal structure determines T_R mainly and that the stabilization from the third dimension is the largest at the addition of the first vertical bond. In view of this result (and also on intuitive grounds) we assume that growth below the roughening transition (nucleation and spiral growth) will also depend mainly on horizontal bond structures.

TABLE III. Roughening temperature in several structures.

Type	h	v	kT_R^{unr}/ϕ	kT_R^{SOS}/ϕ
Square	2	0	0.568	
(100) sc	2	1	0.676	0.649
(110) bcc	2	2	0.679	0.664
(100) fcc	2	4	0.694	0.692
Triangular	3	0	0.910	
(100) hex [a]	3	1	1.178	1.095
(100) hex [a]	3	2	1.179	1.122
(111) fcc [a]	3	3	1.182	1.148

[a] Triangular layers can be packed in different ways to form one, two, or three nearest-neighbor bonds per atom between layers.

Another tendency which is shown in the table is the closer correspondence between unrestricted and solid on solid models if the number of vertical bonds increases. This is another consequence of the fact that the SOS model is a limiting case of the unr model, as we mentioned above.

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