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Crystal growth models and Ising models III. Zero-field susceptibilities and correlations

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Abstract. Analytic solutions are found for the two-site correlations of certain highly symmetric models of crystal growth disorder. These solutions include the case of the second-neighbour square lattice Ising model susceptibility at the conjectured disorder point. The general correlations satisfy a factorisation property conjectured by Welberry.

1. Introduction

In a series of recent papers a two-dimensional model of the growth of disordered mixed crystals has been investigated (Welberry and Galbraith 1973, 1975, Welberry 1977a, b). The crystal growth model was used to study the way in which crystal structure and consequent x-ray diffraction patterns depended on the underlying probabilities associated with the crystal growth. The original work was largely based on simulations of model crystals by digital computer, the simulated structures being used as masks to produce diffraction patterns by direct optical means (Welberry and Galbraith 1973). A special case of the model, the linear case described below, was solved analytically.

A new technique for obtaining further solutions for growth models arose from the observation that the probability distributions arising in these models form a subset of the probability distributions for generalised Ising models in statistical mechanics and that the Ising model formation shows explicitly any symmetry implicit in a growth model (Enting 1977a). Welberry (1977a) pointed out that such symmetry conditions give information that can be used to obtain new solutions. One such solution turned out to include a special solution, SC1, of the non-linear model previously solved by Welberry and Galbraith (1975) using other special analytic properties occurring in this case (see Welberry 1977b for further details of this model and Enting 1977b for the relation of the most symmetric case of SC1 to Ising model series expansions).

Another technique that can be used to solve a small class of growth models was introduced by Verhagen (1976) who related the probabilities to those of one-dimensional processes.

In the present paper we obtain expressions for the two-site correlations of some of the special models investigated by Welberry (1977a). There are four main reasons why such an investigation is of interest.

(i) The two-site correlations $\rho(\boldsymbol{r})$ lead to the intensity distribution of diffraction through the relation (Guinier 1963)

$$I(\boldsymbol{k})=\sum_{\boldsymbol{r}} \exp(2\pi i\boldsymbol{k}\cdot\boldsymbol{r})[(cf_1+(1-c)f_2)^2+\rho(\boldsymbol{r})c(1-c)(f_1-f_2)^2]. \qquad (1.1)$$

Here $\boldsymbol{k}$ is a vector in reciprocal space, c is the concentration of type 1 molecules and f_1 and f_2 are the two form factors.

(ii) Welberry (1977a) has suggested that for models with rectangular symmetry defined in terms of lattice vectors $\boldsymbol{a}$, $\boldsymbol{b}$ correlations have the form

$$\rho(n\boldsymbol{a}+m\boldsymbol{b})=x^{|m|}y^{|n|}. \qquad (1.2)$$

The solutions obtained below confirm this conjecture for all values of m and n in the zero-field case.

(iii) In statistical mechanics, continuous (second-order) phase transitions generally correspond to divergences in $\chi(\boldsymbol{k})$ for some appropriate $\boldsymbol{k}$ while for first-order transitions, in many of the approximate solutions divergences in $\chi(\boldsymbol{k})$ indicate the limits of regions of metastability. The studies of growth models indicate that no such models with all probabilities non-zero can correspond to an Ising model at a phase transition but that as appropriate probabilities vanish the growth models exhibit behaviour typical of the approach to a phase transition. In the growth model context phase transitions can be regarded as being cases when finite effects from the boundaries persist for arbitrarily large distances. Correlation function solutions can be used to investigate the approach to phase transitions.

(iv) Enting (1977a) pointed out that many of the simplest growth models appeared to correspond to Ising models at their disorder points. Disorder points are points at which the behaviour of correlation functions changes from monotonic to oscillatory. Approximate expressions for correlation functions were used by Stephenson (1971) and Enting (1973) to locate disorder points. The accuracy of such approximations in the disorder point region can be tested once exact solutions are available at the disorder point.

Enting (1977a) showed how growth model correlations could be obtained from the equation and obtained parametric expressions for correlations on the axes of a square-symmetric zero-field growth model. The form of the solutions indicated that the form of the decay was a sum of exponentials. The more detailed analysis below shows that even for the more general rectangular symmetry, the parametrisation is 'degenerate' so that there is only a simple exponential decay.

Other techniques of obtaining growth model correlations have been described by Pickard (1977, 1978) and, for the special case of a triangular Ising model at its disorder point, by Gibberd (1969).

2. Zero-field crystal growth models

The growth models that we consider are defined on a square lattice of sites $\boldsymbol{r}=i\boldsymbol{a}+j\boldsymbol{b}$ denoted by the pairs of integers (i, j). The presence of one of the two types of molecule at each site can be denoted by any convenient two-valued variable. Following Enting (1977a) we use Ising spin variables $\sigma_{ij}=\pm1$. Welberry uses $x_{ij}=\frac{1}{2}(\sigma_{ij}+1)=0$ or 1.

Formally the growth model begins by considering the joint probability distribution $P(\{\sigma_{ij}\})$ of the set of σ_{ij} and assumes that it can be expressed as a product of factors P_{ij} which are conditional probabilities of adding a molecule of type σ_{ij} at site (i, j) given the existing crystal configuration. For completeness we need to define a set of boundary sites $\{(i, j): i+j=m\}$ along which the σ_{ij} are taken as fixed.

In mathematical terms the assumptions of the growth model state that

$$P(\{\sigma_{ij}\}) = \prod_{n>m} \prod_{i} P_{i,n-i}. \tag{2.1}$$

We restrict our investigations to P_{ij} of the form

$$P_{ij} = \tfrac{1}{2} + \sigma_{ij}(D\sigma_{i,j-1} + A\sigma_{i-1,j} + B\sigma_{i-1,j-1} + C\sigma_{i,j-1}\sigma_{i-1,j}\sigma_{i-1,j-1}). \tag{2.2}$$

The zero-field condition refers to the fact that (2.2) is invariant under changing the sign of all σ_{ij} so that the two types of molecule are treated equivalently and will each have a concentration of $\frac{1}{2}$. The linear case mentioned above corresponds to putting $C=0$ in (2.2). Enting (1977a) has shown how to relate the probability distribution $P(\{\sigma_{ij}\})$ to the probability distributions associated with Gibbs states of the Ising model in statistical mechanics. In the Ising model formalism the symmetries of the model are explicit and so the conditions required for symmetries of $P(\{\sigma_{ij}\})$ can be readily obtained. Welberry (1977a) obtains the same conditions for symmetry by considering special points in the distribution $P(\{\sigma_{ij}\})$ directly.

To obtain a rectangular lattice symmetry in the model generated by (2.2) Welberry shows that one must have

$$-2AD = (1-2C)B. \tag{2.3}$$

(The notation A, B, C, D is not that of Welberry (1977a) but is a slightly generalisation (introducing $D \neq A$) of that used by Enting (1977a).)

Square lattice symmetry is imposed by setting $A=D$ in addition to constraint (2.3).

The technique for obtaining equations connecting various expectation values is described by Welberry and Galbraith (1973). If the set of σ_{ij} considered in the probability distribution is denoted $U=\{\sigma_{ij}: i+j>m\}$ then

$$\langle f(U)\rangle = \sum_{\sigma_{ij}\in U} P(U)f(U) = \sum_{\sigma_{ij}\in U}\left(\prod_{n>m}\prod_{k} P_{k,n-k}\right) f(U). \tag{2.4}$$

The sum is over all combinations of the values ± 1 for each σ_{ij}. We now define a set $T=\{\sigma_{ij}: p+q>i+j>m\}$ and consider

$$\begin{aligned}
\langle \sigma_{pq}h(T)\rangle &= \sum_{\sigma_{ij}\in U}\left(\prod_{n>m}\prod_{k} P_{k,n-k}\right)\sigma_{pq}h(T) \\
&= \sum_{\sigma_{pq}}\sum_{\sigma_{ij}\in T}\left(\prod_{p+q>n>m}\prod_{k} P_{k,n-k}\right) P_{pq}\sigma_{pq}h(T) \\
&= 2\sum_{\sigma_{ij}\in T}\left(\prod_{p+q>n>m}\prod_{k} P_{k,n-k}\right) g(T)h(T) \\
&= 2\langle g(T)h(T)\rangle
\end{aligned} \tag{2.5}$$

where we have used the most general growth model expression for P_{pq},

$$P_{pq}=\tfrac{1}{2}+\sigma_{pq}g(T).$$

We actually use equation (2.2) for P_{pq} and with a properly chosen set of functions $h(T)$ the symmetry conditions lead to a closed set of equations for correlations.

In all the work on analytic solutions it is assumed that the points considered are sufficiently far from the boundaries for the correlation functions to depend only on the relative positions of the sites involved and not on the absolute position in the lattice. This assumption is discussed in the final section. From (2.5),

$$\begin{aligned}\rho(\boldsymbol{r})&=\langle\sigma_{pq}\sigma_{p-i,q-j}\rangle\\&=2A\rho(\boldsymbol{r}-\boldsymbol{a})+2D\rho(\boldsymbol{r}-\boldsymbol{b})+2B\rho(\boldsymbol{r}-\boldsymbol{a}-\boldsymbol{b})+2Cv(\boldsymbol{r}-\boldsymbol{a}-\boldsymbol{b})\end{aligned}\tag{2.6}$$

$$\begin{aligned}v(\boldsymbol{r})&=\langle\sigma_{pq}\sigma_{p+1,q}\sigma_{p,q+1}\sigma_{p-i,q-j}\rangle\\&=\langle\sigma_{pq}\sigma_{p+1,q}\sigma_{p,q-1}\sigma_{p-i,q-j}\rangle\\&=2A\rho(\boldsymbol{r}+\boldsymbol{b})+2Dv(+\boldsymbol{r}'-\boldsymbol{b})+2B\rho(\boldsymbol{r})+2C\rho(\boldsymbol{r}+\boldsymbol{a}+\boldsymbol{b})\end{aligned}\tag{2.7}$$

where $\boldsymbol{r}=i\boldsymbol{a}+j\boldsymbol{b}$ and $\boldsymbol{r}'=i\boldsymbol{a}-j\boldsymbol{b}$. Rectangular lattice symmetry gives $\rho(\boldsymbol{r})=\rho(\boldsymbol{r}')$ and the equation of expectation values in (2.7). (Actually the derivation of (2.7) requires a larger set of sites in the set U than implied by (2.5). The formalism remains valid so long as the set U does not include sites for which (p, q) is a predecessor (see Enting 1977a). This condition restricts (2.7) to $\boldsymbol{r}=i\boldsymbol{a}+j\boldsymbol{b}$ such that $i\geqslant 0$.)

The derivation of correlations along the axes follows that used by Enting (1977a) for the $A=D$ case.

We put

$$a_n=\rho(n\boldsymbol{a})\tag{2.8a}$$

$$b_n=\rho(n\boldsymbol{a}+\boldsymbol{b})\tag{2.8b}$$

$$c_n=v((n-1)\boldsymbol{a}-\boldsymbol{b})\tag{2.8c}$$

$$d_n=v((n-1)\boldsymbol{a}).\tag{2.8d}$$

Equation (2.6) gives

$$a_n=2Aa_{n-1}+2Db_n+2Bb_{n-1}+2Cc_n\qquad(\text{using } \boldsymbol{r}=n\boldsymbol{a})\tag{2.9a}$$

and

$$b_n=2Ab_{n-1}+2Da_n+2Ba_{n-1}+2Cd_n\qquad(\text{using } \boldsymbol{r}=n\boldsymbol{a}+\boldsymbol{b}).\tag{2.9b}$$

Equation (2.7) gives

$$d_n=2Ab_{n-1}+2Dc_n+2Ba_{n-1}+2Cb_n\qquad(\text{using } \boldsymbol{r}=(n-1)\boldsymbol{a})\tag{2.9c}$$

and

$$c_n=2Aa_{n-1}+2Dd_n+2Bb_{n-1}+2Ca_n\qquad(\text{using } \boldsymbol{r}=(n-1)\boldsymbol{a}-\boldsymbol{b}).\tag{2.9d}$$

These equations lead to two pairs of equations

$$(1\mp 2D)(a_n\pm b_n)=(2A\pm 2B)(a_{n-1}\pm b_{n-1})+2C(c_n\pm d_n)\tag{2.10a}$$

$$(1\mp 2D)(c_n\pm d_n)=(2A\pm 2B)(a_{n-1}\pm b_{n-1})+2C(a_n\pm b_n).\tag{2.10b}$$

Eliminating $c_n \pm d_n$ gives

$$[(1\mp 2D)^2-4C^2](a_n\pm b_n)=(2A\pm 2B)(1\mp 2D+2C)(a_{n-1}\pm b_{n-1}) \tag{2.11}$$

whence

$$a_n\pm b_n=\frac{2A\pm 2B}{1\mp 2D-2C}(a_{n-1}\pm b_{n-1}).$$

In general this would mean, as stated by Enting (1977a), that a_n and b_n would decay as the sum of two exponentials. However, application of the rectangular symmetry constraint (2.3) shows that

$$\frac{2A+2B}{1-2D-2C}=\frac{2A-2B}{1+2D-2C}. \tag{2.12}$$

This means that

$$a_n\pm b_n=\tau^n(1\pm\theta)$$

$$a_n=\rho(n\boldsymbol{a})=\tau^n \tag{2.13a}$$

$$b_n=\rho(n\boldsymbol{a}\pm\boldsymbol{b})=\tau^n\theta \tag{2.13b}$$

$$\tau=(2A-2B)/(1+2D-2C). \tag{2.13c}$$

Interchanging axes gives

$$\rho(n\boldsymbol{b})=\theta n \tag{2.14a}$$

$$\rho(n\boldsymbol{b}\pm\boldsymbol{a})=\tau\theta^n \tag{2.14b}$$

$$\theta=(2D-2B)/(1+2A-2C). \tag{2.14c}$$

Beginning from these axial correlations we can build up longer-range correlations $\rho(m\boldsymbol{a}+n\boldsymbol{b})$ by induction on $m+n$. This is we assume $\rho(m'\boldsymbol{a}+n'\boldsymbol{b})=\tau^{m'}\theta^{n'}$ for $m'+n'<m+n$ and on the basis of this assumption show $\rho(m\boldsymbol{a}+n\boldsymbol{b})=\tau^m\theta^n$.

The equations that we use are from (2.6)

$$\rho(\boldsymbol{r}+\boldsymbol{a}+\boldsymbol{b})=2A\theta\rho(\boldsymbol{r})+2D\tau\rho(\boldsymbol{r})+2B\rho(\boldsymbol{r})+2Cv(\boldsymbol{r}); \tag{2.15a}$$

from (2.7)

$$v(\boldsymbol{r})=2A\theta\rho(\boldsymbol{r})+2Dv(\boldsymbol{r}'-\boldsymbol{b})+2B\rho(\boldsymbol{r})+2C\rho(\boldsymbol{r}+\boldsymbol{a}+\boldsymbol{b}) \tag{2.15b}$$

and

$$v(\boldsymbol{r}'-\boldsymbol{b})=2A\rho(\boldsymbol{r})+2Dv(\boldsymbol{r})+2B\theta\rho(\boldsymbol{r})+2C\tau\rho(\boldsymbol{r}). \tag{2.15c}$$

These are three linear equations in $\rho(\boldsymbol{r}+\boldsymbol{a}+\boldsymbol{b})$, $v(\boldsymbol{r})$ and $v(\boldsymbol{r}'-\boldsymbol{b})$ valid for $\boldsymbol{r}=n\boldsymbol{a}+m\boldsymbol{b}$, $m, n>0$. The equations can be simplified somewhat by using the relations

$$D\tau+B=0 \tag{2.16a}$$

$$A\theta+B=0 \tag{2.16b}$$

which are obtained from (2.13c), (2.14c) subject to constraint (2.3).

Except in special cases when probabilities take on limiting values of 0 and 1 the solution will be unique. We show that (2.15a–c) are satisfied by

$$\rho(\boldsymbol{r}+\boldsymbol{a}+\boldsymbol{b})=\tau\theta\rho(\boldsymbol{r}) \tag{2.17a}$$

$$v(\boldsymbol{r})=\tau\theta\rho(\boldsymbol{r}) \tag{2.17b}$$

$$v(\boldsymbol{r}'-\boldsymbol{b})=\tau\rho(\boldsymbol{r}). \tag{2.17c}$$

Substituting these values into (2.15a), (2.15b) leads to $\tau\theta=-2B+2C\tau\theta$ in each case, an equation that is satisfied by (2.13c), (2.14c) subject to (2.3). Equation (2.15c) reduces to $\tau=2A+2C\tau$ which is satisfied by (2.13c) subject to constraint (2.3).

The key solution is (2.17a) which, when combined with (2.13a), (2.14a), confirms the conjecture made by Welberry that

$$\rho(m\boldsymbol{a}+n\boldsymbol{b})=\tau^{|m|}\theta^{|n|}. \tag{2.18}$$

3. Conclusions

The solutions obtained in the previous section give useful information about each of the four points discussed in the introduction. Firstly and most obviously the conjecture of (2.18) by Welberry has been confirmed. Secondly this solution leads immediately to a simple expression for the order–disorder contribution to the diffraction expression (1.1):

$$\sum_{\boldsymbol{r}}\exp(2\pi i\boldsymbol{k}\cdot\boldsymbol{r})\rho(\boldsymbol{r})=I(\tau,2\pi k_x)I(\theta,2\pi k_y) \tag{3.1}$$

where

$$I(a,b)=\frac{1-a^2}{1-2a\cos b+a^2}. \tag{3.2}$$

With regard to phase transitions it is clear from (2.18) that long-range correlations occur in these models only when $\tau=1$ or $\theta=1$, which happens only when some of the underlying growth model properties go to 1 or 0. Thus the solutions for the class of models solved above confirm the properties that have been deduced from simulations.

The complete solutions for correlations strengthen the arguments given by Enting (1977a) for identifying various crystal growth models with Ising models at their disorder points. Not only is the axial correlation simpler than previously believed, since the two exponential factors found by Enting actually correspond to the degenerate case of a single exponential decay, but, in addition we have now seen that correlations in all directions have a simple exponential decay.

Following the results of Enting (1977a) we put

$$X=P_0^*=P_{15}^*=(1-P_8^*)(1-P_7^*)=\tfrac{1}{2}+A+B+C+D \tag{3.3a}$$

$$Y=P_2^*=P_{13}^*=(1-P_{10}^*)=(1-P_5^*)=\tfrac{1}{2}+A-D+B-C \tag{3.3b}$$

$$W=P_4^*=P_{11}^*=(1-P_{12}^*)=(1-P_3^*)=\tfrac{1}{2}-A+D+B-C \tag{3.3c}$$

$$Z=P_6^*=P_9^*=(1-P_{14}^*)=(1-P_1^*)=\tfrac{1}{2}-A+B+C-D \tag{3.3d}$$

$$\exp(-8\beta K)=YW(1-X)(1-Z)/XZ(1-Y)(1-W) \tag{3.4a}$$

$$\exp(-4\beta J_y) = (1-W)Z/X(1-Y) \tag{3.4b}$$

$$\exp(-4\beta J_x) = (1-Y)Z/X(1-W) \tag{3.4c}$$

$$\exp(-8\beta J_1) = WY(1-W)(1-Y)/XZ(1-X)(1-Z) \tag{3.4d}$$

$$\exp(-8\beta J_2) = (1-W)(1-X)(1-Y)(1-Z)/XYZW \tag{3.4e}$$

where J_1, J_2, J_x, J_y and K are the interaction strengths in the corresponding Ising model. The four-spin interaction is K, the nearest-neighbour interactions are J_x and J_y, and the second-neighbour interactions are J_1 and J_2.

The parametrisation in terms of A, B, C, D gives one constraint on the Ising interaction parameters. Applying the requirement of rectangular symmetry, $J_1 = J_2$, leads to (2.3), a second constraint on the interactions. The condition for the four-spin interaction to vanish is

$$4B^2C + 4D^2C - 8ABD - C - 4C^3 + 4A^2C = 0 \tag{3.5}$$

so that one has a second-neighbour Ising model with rectangular symmetry. Subject to constraints (2.3) and (3.5) the full square lattice symmetry is achieved by the single additional constraint $A = D$, leading to three constraints on the values A, B, C, D and a second-neighbour Ising model with one constraint. Enting (1977a) conjectured that this constraint defined the disorder point temperature. For small values of the interaction the constraints led to an approximate relation of the form

$$(\beta J_x)^2 \approx -\beta J_2. \tag{3.6}$$

The relation for a wider range of values is plotted in figure 3 of Enting (1977a).

As remarked in the introduction there are other techniques which have been used for obtaining correlations in growth models. The graphical techniques used by Gibberd (1969) for the anisotropic triangular Ising model are simple in that case but to apply the techniques directly to more complicated systems such as the second-neighbour square lattice Ising model or models in non-zero fields would require awkward parametrisations of the interactions.

One promising approach seems to be to find a graphical formulation of the techniques used in § 2 since the growth model parametrisation given in equation (2.2) (or its generalisation to non-zero field) seems to be most appropriate for expressing the solutions of these models.

For some classes of growth models Pickard's (1977a, 1978) results bypass all these approaches by demonstrating an exponential decay of the correlations by considering a specialised probabilistic characterisation of the systems, and without constructing explicit solutions. While these solutions cover a large number of systems, Pickard has pointed out that there are systems for which his techniques do not apply but for which the growth model equations can be solved using the techniques of § 2.

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