A study of a coarse-grained free energy functional for the three-dimensional Ising model

1983 J. Phys. A: Math. Gen. 16 L623

(http://iopscience.iop.org/0305-4470/16/16/007)

J. Phys. A: Math. Gen. **16** (1983) L623–L627. Printed in Great Britain

LETTER TO THE EDITOR

A study of a coarse-grained free energy functional for the three-dimensional Ising model

K Kaski†, K Binder and J D Gunton†

Institut fur Festkorperforschung, Kernforschungsanlage Julich, Postfach 1913, D-5170 Julich 1, West Germany

Received 30 August 1983

Abstract. The joint distribution function of the nearest-neighbour cell magnetisation of the three-dimensional Ising model has been studied by Monte Carlo methods. This function has a double-peaked structure for $T < T_c$ and can be represented by a coarse-grained functional of two cell variables. We have determined the dependence of this functional on the coarse-graining cell size for several temperatures below T_c. In particular we have calculated the dependence of the 'spinodal curve' on the (scaled) coarse-graining size.

There are several important problems in critical phenomena and two-phase coexistence in which the 'coarse-grained' Helmholtz free energy functional (Ginzburg–Landau Hamiltonian) is of fundamental importance (Kawasaki *et al* 1981, Wilson and Kogut 1974, Bhatia and March 1978, Ohta and Kawasaki 1977, Rudnick and Jasnow 1978). In addition to the well known problems in critical phenomena, one class of problems involves the nonlinear dynamics of metastable and unstable states, in which changes in the free energy provide the driving force for phase separation via nucleation and spinodal decomposition processes (Langer *et al* 1975, Gunton *et al* 1983). It would be of considerable importance to calculate this free energy functional, starting from some initial model Hamiltonian, for a coarse-graining cell edge size L which is of the order of the correlation length ξ. To do this would require dividing the system into hypercubical cells of edge size L and calculating the coarse-grained distribution function (for different values of L) as a functional of all the cell variables, starting from a microscopic Hamiltonian‡. This is currently an overwhelmingly difficult task. A more modest, but still useful, project is to calculate a coarse-grained, reduced distribution function which yields the same qualitative features as the Helmholtz free energy functional§.

In this letter we report some progress in this direction based on a Monte Carlo study of the joint distribution function $P_L(S_\alpha, S_\beta)$ for adjacent cells of magnetisation S_α and S_β respectively. We have calculated this distribution function for the three-dimensional nearest-neighbour Ising model for several different values of the cell edge size L and for several values of the temperatures T both below and above T_c. From

† Permanent address: Physics Department, Temple University, Philadelphia, Pa 19122, USA.

‡ It is important to note that one is particularly interested in the results for $L \sim \xi$, rather than $L \to \infty$, as is discussed by Kawasaki *et al* (1981), Langer *et al* (1975) and Gunton *et al* (1983).

§ The next step beyond this current work would be to calculate $P_L(S_\alpha, S_\beta; \{S_\gamma\})$ where $\{S_\gamma\}$ stands for the remaining six nearest-neighbour cells of cell α (in addition to cell β). Even this next step is computationally non-trivial, but should yield results rather close to the Helmholtz free energy functional.

these results for $P_L(S_\alpha, S_\beta)$ we are able to determine a coarse-grained functional of two cell variables which is qualitatively similar to the Helmholtz free energy functional. We are thus able to see how a double-well potential changes as a function both of the coarse-graining cell edge size L and of temperature for $T < T_c$. In particular, we are able to obtain the dependence of a 'spinodal curve' on the scaled coarse-graining variable L/ξ. We will report our results for $T > T_c$ in an expanded discussion of this work.

We adopt the procedure of an earlier Monte Carlo finite size scaling study (Binder 1981). Namely, we divide the lattice sites of an $N \times N \times N$ Ising model (with $N = 24$) into cells of linear dimension L. The magnetisation per spin of cell α is $S_\alpha = L^{-d} \Sigma_{i \in \alpha} S_i$, where the sum is over all lattice sites i in the cell α and the Ising spin variables $S_i = \pm 1$. The coarse-grained Helmholtz free energy functional is then defined as $F_L = -k_B T \log P_L(\{S_\alpha\})$, where $P_L(\{S_\alpha\})$ is the probability function for finding the cells in a specified configuration $\{S_\alpha\}$, subject to the constraint that the total magnetisation per spin of the system is fixed and given by $M = n^{-1} \Sigma_\alpha S_\alpha$, where n is the number of cells. In practice it is difficult to calculate this distribution function, so that one usually assumes that F_L is of a Ginzburg–Landau form, with a double-well potential for $T < T_c$. It is, however, possible to calculate a qualitatively similar quantity, namely $P_L(S_\alpha, S_\beta)$ defined as

$$P_L(S_\alpha, S_\beta) = \sum_{\{S_\gamma\}}{}' P_L(\{S_\gamma\}) \tag{1}$$

where the sum is over all cell magnetisations $\{S_\gamma\}$ holding S_α and S_β fixed. This distribution function is symmetric under interchange of cell variables ($P_L(S_\alpha, S_\beta) = P_L(S_\beta, S_\alpha)$) and under spin inversion (i.e. $P_L(-S_\alpha, -S_\beta) = P_L(S_\alpha, S_\beta)$, at zero external magnetic field). We have evaluated $P_L(S_\alpha, S_\beta)$ by standard Monte Carlo methods for the case in which α and β are nearest-neighbour cells, exploiting the above symmetries in our sampling procedures. A typical example of our results for $P_L(S_\alpha, S_\beta)$ is shown in figure 1 for the case $L = 6$, $kT/J = 4.4$, where J is the nearest-neighbour Ising interaction constant. (We have used periodic boundary conditions for the $N \times N \times N$ system.) The two-peaked structure shown in figure 1 reflects the expected features of two-phase coexistence. As the cell size increases (from $L = 2$ to $L = 8$) this peak structure becomes sharper and the values of the maxima move closer to the equilibrium values of the magnetisation.

There is no unique way to represent our data for $P_L(S_\alpha, S_\beta)$, so we have chosen as the simplest form a discrete version of the Ginzburg–Landau type Hamiltonian for nearest-neighbour cells

$$P_L(S_\alpha, S_\beta) = Z^{-1} \exp - \mathcal{H}_L(S_\alpha, S_\beta) \tag{2}$$

with

$$\mathcal{H}_L(S_\alpha, S_\beta) = c_L(S_\alpha - S_\beta)^2 + V_L(S_\alpha) + V_L(S_\beta) \tag{3}$$

where

$$V_L(S) = -r_L S^2 + u_L S^4. \tag{4}$$

The term involving c_L is the discrete version of the gradient term in the continuum model, while r_L is positive and Z is an appropriate normalisation factor. Representing the potential $V_L(S)$ by a fourth-order polynomial is arbitrary, since higher-order terms are to be expected, but (4) does have the double-well structure which corresponds to the double-peaked structure of $P_L(S_\alpha, S_\beta)$. Equations (2), (3) and (4) provide a reasonable representation of our data for $P_L(S_\alpha, S_\beta)$ and allow us to estimate the

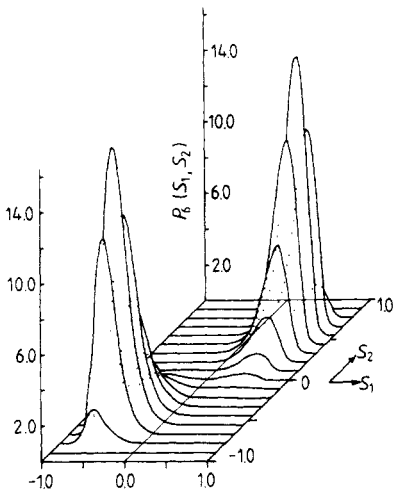
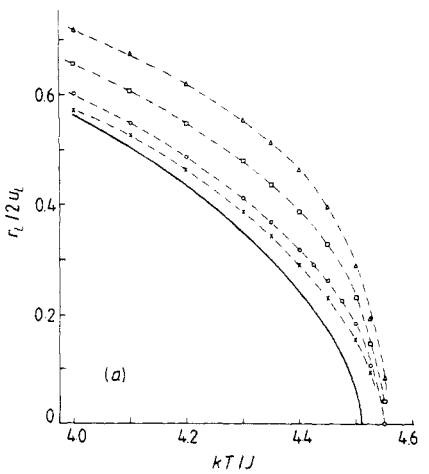


Figure 1. The joint distribution function $P_L(S_1, S_2)$ of the nearest-neighbour cell magnetisations S_1 and S_2 for $L = 6$ at $kT/J = 4.4$, where $kT_c/J = 4.51$.

dependence of the coupling constants c_L, r_L and u_L on L and T. The coupling constant ratios $r_L/2u_L$ and $c_L/2u_L$ are calculated from $P_L(S_\alpha, S_\beta)$ in the neighbourhood of the maximum at $S_\alpha = S_\beta = S_{\max}$. The ratio $r_L/2u_L$ is determined from $S_{\max}$, while the ratio $c_L/2u_L$ is determined from $S_{\max}$ and $S_{\beta\,\max}$. (The quantity $S_{\beta\,\max}$ denotes the maximum of $P_L(S_\alpha, S_\beta)$ for fixed S_β.) The ratios $r_L/2u_L$ and $c_L/2u_L$ are shown in figure 2. As is to be expected, $r_L(T)$ vanishes at a temperature $T_c(L)$ which is greater than the bulk critical temperature T_c, due to finite size effects. As L increases $T_c(L) \to T_c$, as it should. Also, the ratio $c_L/2u_L$ is only weakly dependent on the temperature for fixed L, as one would expect. Although these coefficients c_L, r_L and u_L do not represent the Helmholtz free energy functional F_L, they do display the same qualitative features



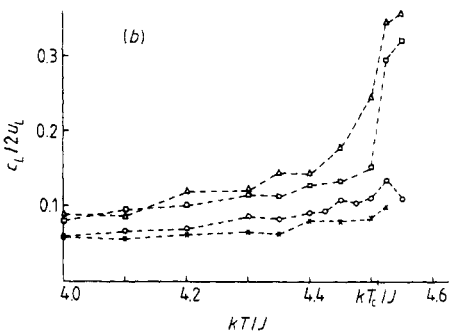


Figure 2. The coupling constant ratios (*a*) $r_L/2u_L$ and (*b*) $c_L/2u_L$ for the parametrisation given by (3) and (4), for temperatures $T \leq T_c$ and cell sizes $L = 3$, 4, 6 and 8. The broken lines are drawn as a visual aid. Also shown in (*a*) is the M^2 curve (full line) as obtained from the best Padé approximants of the series expansion (Essam and Fisher 1963). △, $L = 3$; □, $L = 4$; ○, $L = 6$; ×, $L = 8$.

one expects for the corresponding expansion coefficients of F_L. In addition, we should point out indirect evidence which suggests that our representation of $P_L(S_\alpha, S_\beta)$ by (2)–(4) is not unreasonable. First, as can be seen from figure 2 our estimate of the magnetisation (based on r_L and u_L) is within 5% of the Ising value for $kT/J \leqslant 4.0$ for $L/\xi \gg 1$. Similar agreement is found between our estimate of r_L and the low-temperature Ising susceptibility. Secondly, the vanishing of r_L yields the bulk critical temperature to within 1%, as can be seen from figure 2. Finally, we find that the critical exponents obtained from analysing (2)–(4) are within 4% of the known Ising values, as will be reported elsewhere. A plot of the double-well potential V_L as a function of the cell magnetisation S_α for different values of L (at a fixed temperature) yields results which are qualitatively very similar to the coarse-grained double-well potential for F_L as determined by a first order in ε field theoretic calculation (Kawasaki *et al* 1981) (where $\varepsilon = 4 - d$ and d denotes the dimensionality). It should be noted that our determination of the coupling constants from a microscopic Hamiltonian is one of the few calculations of its kind. Our work extends an earlier calculation (Binder 1981) of the single-cell distribution function $P_L(S)$. In this earlier work, however, no attempt was made to estimate any parameters of the coarse-grained free energy functional. The present work thus is a crucial extension of the earlier study, particularly in that it also yields information about the coefficient c_L of the gradient term.

Another quantity of interest is the 'spinodal curve' for the coarse-grained Helmholtz free energy functional. By analogy with mean field theory this could be defined as the function $\hat{M}_s(L, T)$ for which $(\partial^2 F_L(M)/\partial M^2)_{T,L} = 0$. Although we have not been able to calculate F_L and hence cannot compute $\hat{M}_s(L, T)$, we can compute an analogous quantity $M_s = M_s(L, T)$, using the definition $(\partial^2 \log P_L(S, S)/\partial S^2)_{L,T} = 0$. We could estimate M_s from this definition and the representation (2)–(4). However, a more accurate determination is given by analysing the original data for $P_L(S_\alpha, S_\beta)$ to find M_s. Our results for this 'spinodal' curve are given in figure 3, in which $(1 - M_s/S_{max})$ is the abscissa. One would expect that $(1 - M_s/S_{max})$ should approach the Ginzburg–Landau (mean field) value of $(1 - 1/\sqrt{3}) \simeq 0.41$ for $L/\xi \ll 1$. As can be seen from figure 3 our results are within 5% of this limit, in gratifying agreement with theoretical expectations. An alternative scaling form can be given in which the abscissa of figure 3 is replaced by $(1 - M_s/M)$. In this case we have found heuristic arguments which predict that the asymptotic behaviour of $(1 - M_s/M)$ is $(1 - A(L/\xi)^{-\beta/\nu})$ for $L/\xi \ll 1$ and $(L/\xi)^{-(1+1/d)}$ for $L/\xi \gg 1$, where β and ν are the usual Ising critical exponents and M is the spontaneous magnetisation. These arguments will be presented elsewhere in a full description of this work. It should be noted that in contrast to a mean field theory in which there is a unique thermodynamic spinodal curve, the coarse-grained 'spinodal' curve depends on the coarse-graining size L. We would expect that a spinodal curve as determined from F_L would exhibit qualitatively the same behaviour as shown in figure 3.

Finally, we comment on the relevance of our work to the dynamical theory of first-order phase transitions. As mentioned earlier, the Helmholtz free energy functional F_L is particularly relevant to the theory of spinodal decomposition, which is the process by which a thermodynamically unstable system begins to phase separate. In the most successful nonlinear theory to date (Langer *et al* 1975), an *ad hoc* ansatz for F_L was used, in the absence of any first principles knowledge of F_L. Since our results obtained from the joint distribution function do not yield F_L itself, we cannot as yet test the validity of the above ansatz. It is interesting, however, to note that the coarse-grained spinodal curve implicit in this ansatz of Langer *et al* (1975) differs

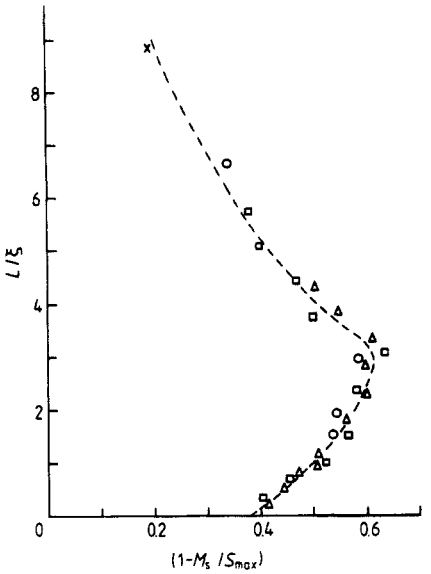


Figure 3. A scaling form of the spinodal curve which displays the consistency of our results with the Ginzburg–Landau theory for $L/\xi \ll 1$. The correlation length has been obtained from Tarko and Fisher (1975). △, $L=3$; □, $L=4$; ○, $L=6$; ×, $L=8$.

from the results shown in figure 3 by only $\leqslant 20\%$. Also, our results are qualitatively very similar to the field theoretical calculation of F_L (which is presumably quantitatively inaccurate in three dimensions). Thus our determination of a double-well potential for a coarse-grained system starting from a microscopic model seems a useful first step toward developing a continuum theory of the dynamics of phase separation.

This work was supported in part by NSF grant # DMR-8013700. Two of us (KK and JDG) wish to acknowledge the kind hospitality of the KFA, where most of this work was done.

References

Bhatia A B and March N H 1978 *J. Chem. Phys.* **68** 4651
Binder K 1981 *Z. Phys.* B **43** 119
Essam J W and Fisher M E 1963 *J. Chem. Phys.* **38** 802
Freedman B, Smolensky P and Weingarten D 1982 *Phys. Lett.* **113B** 481
Gunton J D, San Miguel M and Sahni P 1983 *Phase Transitions and Critical Phenomena* vol 8, ed C Domb and J L Lebowitz (London: Academic) to be published
Kawasaki K, Imaeda T and Gunton J D 1981 *Perspectives in Statistical Physics* ed H J Raveche (Amsterdam: North-Holland) p 203
Langer J S, Baron M and Miller H D 1975 *Phys. Rev.* A **11** 1417
Ohta T and Kawasaki K 1977 *Prog. Theor. Phys.* **58** 467
Rudnick J and Jasnow D 1978 *Phys. Rev.* B **17** 1351
Tarko H B and Fisher M E 1975 *Phys. Rev.* B **11** 1217
Wilson K and Kogut J 1974 *Phys. Rep.* **12C** 75