Microscopic Approach to Interfacial Structure in Ising-like Ferromagnets*

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The properties of an Ising-like ferromagnetic model are investigated in the presence of a space-dependent magnetic field which favors phase separation. The treatment is limited to very low temperatures where molecular-field theory seems to be applicable. The magnetization profile in the interface region is calculated, as well as the interface free energy, within an harmonic approximation to magnetization density modes in our model. The instability of the interface in the limit of vanishing external field, which was recently pointed out for fluid systems, is recovered and discussed in detail.

I. INTRODUCTION

IN this paper we investigate the interfacial structure of a simple ferromagnetic system. Phenomenologically one has a good picture of how a ferromagnet looks if placed in an external space-dependent magnetic field. A positive field favors a positive magnetization; and in regions where the field is negative, the magnetization also turns out to be negative. Regions of different magnetization are separated by the so-called Bloch wall whose thickness depends essentially on the mutual interaction between spins and on the temperature. As we approach the Curie temperature from below, the thickness tends to infinity, resembling the fact that the distinction between two different phases vanishes at and above the Curie point. For fluids where the situation is similar, one derives within the Maxwell-van der Waals theory[1] the following concentration profile in the transition region between liquid and vapor:

$$\rho(z)=\frac{1}{2}\left[\rho_1+\rho_2-(\rho_1-\rho_2)\tanh\frac{2z}{d}\right], \qquad (1.1)$$

where $\rho_{1,2}$ are the densities of liquid and vapor, respectively, and d is the interface thickness. We shall find exactly the same profile in our ferromagnetic model, restricting ourselves to temperatures far below the Curie temperature.

Besides the interesting problem of how the thickness d depends on temperature, another interesting feature was recently discussed by Buff, Lovett, and Stillinger.[2] They point out that the gravitational field plays a rather peculiar role in interface calculations. They calculate the thickness for a fluid system starting from a sharp interface and allowing for distortions by surface plane waves. It turns out that the thickness exhibits a logarithmic singularity in a small gravitational field, whereas the corresponding free energy suffers a bounded logarithmic anomaly in the same limit. This behavior is not at all surprising if one remembers that the gravitational field mainly serves to locate the interface between two different phases in space no matter how weak the field actually is. Switching off the field allows for macroscopic displacements of the interface; this instability is presumably reflected by the divergence of the interface thickness.

In the following investigation we shall deal mainly with the corresponding effect in a ferromagnetic system. As the properties of ferromagnets are far from being solved in the critical region, except for certain special cases as for example the two-dimensional Ising model without magnetic field, we have to confine our attention to temperatures far below the Curie temperature where molecular-field theory seems to be applicable. Section II is devoted to a discussion of our model and its relation to the Ising model of ferromagnetism. In Sec. III we apply molecular-field theory to our model functional integral which amounts to a saddle-point approximation in the presence of an external space-dependent magnetic field. The approximate evaluation of the functional integral, performed in Sec. IV, is equivalent to the harmonic expansion in the treatment of Buff, Lovett, and Stillinger.[2] It turns out that their results are in full agreement with our calculations. Section V contains a discussion and a possible shortcoming of our results.

II. DERIVATION OF A MODEL FUNCTIONAL INTEGRAL

The model we discuss was recently proposed by Langer.[3] For convenience we rederive it here from the functional integral representation of the Ising model.[4]

The partition function of the latter is given by

$$Z=\sum_{\{\sigma_i=\pm1\}}\exp\{\tfrac{1}{2}\sum_{ij}\beta V_{ij}\sigma_i\sigma_j+\sum_j\beta H_j\sigma_j\}, \qquad (2.1)$$

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where H_j is a space-dependent magnetic field. Without loss of generality, the interaction matrix is assumed to be positive definite. Summations are extended over all lattice sites with spacing δ. As shown by many authors,[4] the partition function (2.1) may be converted into a functional integral by introducing a set of random variables ϕ_i to linearize the quadratic argument of the exponential in (2.1). The result is

$$Z=A\int \prod_i d\phi_i\, e^{-S\{\phi\}}, \tag{2.2}$$

where

$$S\{\phi\}=\frac{1}{2\beta}\sum_{ij}(V^{-1})_{ij}\phi_i\phi_j-\sum_i \ln 2\cosh(\beta H_i+\phi_i). \tag{2.3}$$

The normalization factor A in (2.2) is determined by the condition $Z(\beta=0)=1$. We first transform to new variables by defining

$$\mu_i=\phi_i+\beta H_i, \tag{2.4}$$

such that $S\{\phi\}$ contains no uneven terms in μ except a linear term:

$$S\{\mu\}=\frac{1}{2\beta}\sum_{ij}(V^{-1})_{ij}\mu_i\mu_j-\sum_i \lambda_i\mu_i-\sum_i \ln\cosh\mu_i, \tag{2.5}$$

where henceforth we consider λ_i, $\lambda_i=\sum_j(V^{-1})_{ij}H_j$, as the external field, and where we have dropped an unimportant constant. The model we are interested in differs from (2.5) in two aspects. First we replace summations by integrations over the lattice volume L^3, where L is measured in multiples of the lattice spacing δ, thus introducing a continuum model. Furthermore we approximate the Fourier components of V^{-1} by its first two terms in an expansion with respect to the wave vector $\mathbf{k}$:

$$1/v(\mathbf{k})=\beta_0(1+R^2\mathbf{k}^2+\cdots), \tag{2.6}$$

where β_0 is the Curie temperature and R the range of the interaction forces.[5] Finally we approximate the last term in (2.5) by its first two terms in a Taylor expansion. With the abbreviations.

$$a^2=\frac{\beta_0}{\beta}R^2,\quad \epsilon=\frac{1}{2}\left(1-\frac{\beta_0}{\beta}\right)=\frac{1}{2}\frac{T_0-T}{T_0},\quad \alpha=\frac{1}{12}, \tag{2.7}$$

we end up with

$$S\{\mu\}=\int d\mathbf{r}\left[\tfrac{1}{2}a^2(\nabla\mu)^2-\epsilon\mu^2+\alpha\mu^4-\lambda(\mathbf{r})\mu\right], \tag{2.8}$$

which constitutes the basic definition of our model functional integral. As we consider only temperatures far below the true and approximate Curie temperature T_0, the parameter ϵ may be taken positive in accord with (2.7). In that case it is absolutely necessary to retain the quartic term $\alpha\cdot\mu^4$ in the expansion of ln coshμ, as it guarantees the existence of a finite lower bound to $S\{\mu\}$ under all variations of μ, and therefore the existence of the functional integral

$$Z=A\int \delta\mu(\mathbf{r})\exp(-S\{\mu\}). \tag{2.9}$$

The derivation given before should be regarded as a justification for studying the model described by (2.8) and (2.9).[6] As we shall no longer refer to the Ising model as basic, we also free the coupling parameter α from its specific value in (2.7); henceforth we consider α as small and positive.

III. SADDLE-POINT APPROXIMATION IN AN EXTERNAL FIELD

Under the conditions described above, we expect a large contribution to the functional integral (2.9) from the immediate neighborhood of the absolute minimum of (2.8) under variations with respect to μ. The stationary value $\bar{\mu}$ is determined in the usual way by the Euler-Lagrange equation $\delta S/\delta\mu|_{\mu=\bar{\mu}}=0$, which leads to the differential equation

$$-a^2\Delta\bar{\mu}-2\epsilon\bar{\mu}+4\alpha\bar{\mu}^3=\lambda(\mathbf{r}). \tag{3.1}$$

Let us first look for solutions of this equation in a vanishing field and in the one-dimensional case. It is shown in the Appendix that there is a one-parametric class of functions satisfying the differential equation with vanishing derivatives in every order at infinity, namely,

$$\bar{\mu}(z)=(\epsilon/2\alpha)^{1/2}\tanh\frac{\sqrt{\epsilon}}{a}(z-z_0). \tag{3.2}$$

The point z_0 is the location of the interface, dividing regions of positive and negative magnetization. The freedom of choosing z_0 anywhere resembles the fact that without magnetic field the position of an interface is arbitrary. If we calculate the contribution of $\bar{\mu}$ to $S\{\mu\}$, we shall find a bulk term and a positive interface energy term, as long as we choose a finite z_0. The positive interface term only vanishes if z_0 is infinite, i.e., a constant $\bar{\mu}$ resembling homogeneous magnetization. The true absolute minimum therefore corresponds to the latter solution. From this consideration we expect in three dimensions a $\bar{\mu}$ varying only in the z direction, if the field λ depends on z only, $\lambda=\lambda(z)$.

[5] The expansion in (2.6) is meaningful for an interaction which decreases exponentially at large distances. If (2.6) is taken as exact in the continuum limit, the potential is the familiar Yukawa interaction, $V(r)\cong(1/4\pi\beta_0R^2)(e^{-r/R}/r)$. Here and in (2.6) we consider r, R, and $\mathbf{k}$ as dimensionless quantities, measured in multiples of the lattice spacing δ or its inverse, respectively.

[6] As shown in Ref. 3, our model may also be interpreted as a "modified Gaussian model." Here it is derived from the Ising model. Via the usual reinterpretation of the Ising model as a lattice gas model, we can make contact to classical fluid systems. In the latter case, the spatially varying external potential may be identified with the gravitational field.

As the theory of nonlinear differential equations of the type (3.1) is not yet developed so far, there is no hope to solve Eq. (3.1) for arbitrary given field. On the other hand, it turns out that we can fit a specific form of the field to allow for an exact solution. By looking at the solution (3.2) of the zero-field equation, we guess that a field with the same functional dependence might fit. Indeed we find for

$$\lambda(z)=2\gamma\left(\frac{\epsilon+\gamma}{2\alpha}\right)^{1/2}\tanh\frac{(\epsilon+\gamma)^{1/2}}{a}z \tag{3.3}$$

a unique solution

$$\bar{\mu}(z)=\left(\frac{\epsilon+\gamma}{2\alpha}\right)^{1/2}\tanh\frac{(\epsilon+\gamma)^{1/2}}{a}z\,. \tag{3.4}$$

Fortunately the field given by (3.3) is physically quite reasonable. For positive z it favors positive magnetization ($\bar{\mu}>0$), and for negative z we have a negative magnetization ($\bar{\mu}<0$), respectively. The parameter γ allows for changing the strength of the field, and we have chosen the turning point of λ at $z=0$, therefore locating the interface at the same place. We should mention that the functional behavior of $\bar{\mu}$ is mainly determined by the left-hand part of the differential equation (3.1) and not by the specific form of the field (3.3), at least if the field is weak, i.e., γ very small. The essential effect of the field is that it *locates* the interface, no matter how weak it is. If we choose a field different from (3.3), but with only one turning-point at $z=0$, the corresponding $\bar{\mu}$ still would be given by (3.4) (with $\gamma=0$) and small corrections depending on the field strength.

The stationary value $S\{\bar{\mu}\}$ is calculated easily:

$$S\{\bar{\mu}\}=-L^3\frac{(\epsilon+\gamma)(\epsilon+3\gamma)}{4\alpha}+L^2\frac{a(\epsilon+\gamma)^{1/2}}{3\alpha}(2\epsilon+5\gamma), \tag{3.5}$$

where the first term is a contribution to the bulk free energy, and the second term contributes to the interface free energy. Remark that the second term does not depend characteristically on the strength parameter γ, as we always consider ϵ very much larger than γ.

The next step is to expand $S\{\mu\}$ around its stationary point by introducing the notation

$$\mu(\mathbf{r})=\bar{\mu}(z)+\nu(\mathbf{r}). \tag{3.6}$$

The total functional S then splits up into three parts

$$S\{\mu\}=S\{\bar{\mu}\}+S_0\{\nu\}+S'\{\nu\}, \tag{3.7}$$

where S_0 is bilinear in ν,

$$S_0\{\nu\}=\int d\mathbf{r}[\tfrac{1}{2}a^2(\nabla\nu)^2-\epsilon\nu^2+6\alpha\bar{\mu}^2\nu^2], \tag{3.8}$$

and S' is of higher order and will henceforth be considered as a small perturbation:

$$S'\{\nu\}=\alpha\int d\mathbf{r}[4\bar{\mu}\nu^3+\nu^4]. \tag{3.9}$$

In order to diagonalize the quadratic form (3.8), we have to solve the eigenvalue equation

$$[-a^2\Delta-2\epsilon+12\alpha\bar{\mu}^2]\nu(\mathbf{r})=E\nu(\mathbf{r}) \tag{3.10}$$

with periodic boundary conditions on a large box of volume L^3. The transverse part of ν (x, y direction) may be separated immediately, as the potential in (3.10) only varies in the z direction. It is convenient to use the following notation:

$$z=\frac{a}{(\epsilon+\gamma)^{1/2}}t;\quad \nu_{n\mathbf{p}}(\mathbf{r})=\frac{1}{L}e^{i(p_x x+p_y y)}f_n(t);$$

$$p_{x,y}=\frac{2\pi}{L}\times\text{integer};$$

$$E_{n\mathbf{p}}=(\epsilon+\gamma)(\omega_{\mathbf{p}}^2+K_n^2);\quad \omega_p^2=(\epsilon+\gamma)^{-1}(4\epsilon+6\gamma+a^2\mathbf{p}^2). \tag{3.11}$$

Using (3.4) we obtain

$$\left[-\frac{d^2}{dt^2}-6(1-\tanh^2 t)\right]f_n=K_n^2 f_n. \tag{3.12}$$

This equation has two bound-state solutions with K_n^2 equal to -4 and -1, respectively.[7] The normalized eigenfunctions and eigenvalues are given by

$$K_1^2=-4:\quad f_1=\left(\frac{3(\epsilon+\gamma)^{1/2}}{4a}\right)^{1/2}(1-\tanh^2 t),$$

$$E_{1\mathbf{p}}=(\epsilon+\gamma)(\omega_{\mathbf{p}}^2-4)>0 \tag{3.13a}$$

$$K_2^2=-1:\quad f_2=\left(\frac{3(\epsilon+\gamma)^{1/2}}{2a}\right)^{1/2}(\tanh t)(1-\tanh^2 t)^{1/2},$$

$$E_{2\mathbf{p}}=(\epsilon+\gamma)(\omega_{\mathbf{p}}^2-1)>0. \tag{3.13b}$$

The rest of the states ($n=3, 4, \cdots$) are scattering states with $K^2\geq 0$, the spectrum becoming dense as the volume goes to infinity. It is easy to see that the eigenfunctions are

$$f_K(t)=c_K e^{iKt}[1+K^2+3iK\tanh t-3\tanh^2 t], \tag{3.14}$$

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[7] A potential of the form $V(t)=-C(1-\tanh^2 t)$ has n bound states where n is the largest integer satisfying:

$$n<\tfrac{1}{2}[(1+4C)^{1/2}+1];$$

see L. D. Landau and E. M. Lifshitz, *Quantum Mechanics* (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts, 1958), p. 69, problem 4. A similar discussion is given in Ref. 3.

where c_K is the normalization factor. Periodic boundary conditions imply

$$f_K\left(t=\frac{C}{2}\right)=f_K\left(t=-\frac{C}{2}\right), \quad C=\frac{(\epsilon+\gamma)^{1/2}}{a}L. \tag{3.15}$$

As the real part of f_K is an even function in t and the imaginary part is odd [see (3.14)], this condition reduces to

$$\mathrm{Im}f_K(C/2)=0, \tag{3.16}$$

which determines the eigenvalues K_n. Using (3.14) one calculates the spacing between adjacent K values in the limit $L\to\infty$. Up to second order in L^{-1} we obtain with the ansatz

$$K_{n+1}-K_n=\frac{2\pi}{C}\left(1+\frac{s(K_n)}{C}\right), \tag{3.17}$$

$$s(K)=\frac{2}{K^2+1}+\frac{4}{K^2+4}. \tag{3.18}$$

Any summation over the scattering states may thus be replaced by

$$\sum_{n=3}^{\infty} G(K_n)=C\int_{-\infty}^{\infty}\frac{dK}{2\pi}\left(1-\frac{s(K)}{C}\right)G(K), \tag{3.19}$$

where G is arbitrary. Finally the normalization factor c_K is

$$c_K{}^2=[L(K^2+1)(K^2+4)]^{-1}\left[1-\frac{s(K)}{C}\right]^{-1}. \tag{3.20}$$

It should be remarked that the two bound states are not strictly single, but each is composed of a whole band of transverse plane waves; the same holds for the scattering states.

The diagonalization which we have performed corresponds to a transformation to normal modes. These are independent to zeroth-order and only coupled by the S' term (3.9) in higher order. The physical meaning of the first bound state is that it describes a displacement of the interface without changing the magnetization profile given by $\bar{\mu}$ (3.4). This may be seen by expanding the $\bar{\mu}$ function displaced by z_0 around $z_0=0$:

$$\bar{\mu}(z-z_0)\approx\bar{\mu}(z)-z_0(d\bar{\mu}/dz). \tag{3.21}$$

The derivative $d\bar{\mu}/dz$ is exactly the first bound-state wave function f_1 up to a constant normalization factor. In the limit of vanishing field ($\gamma\to 0$), the corresponding energy $E_{1,\mathbf{p}=0}$ goes to zero.[8] This merely expresses the fact that in zero field the location of an interface is arbitrary, therefore we need no energy for a displacement. The instability is a quite general feature and does not depend on the special choice of our model. It was first pointed out by Buff, Lovett, and Stillinger[2] in a phenomenological treatment of a liquid-gas model. We shall come back to a more detailed comparison with their results in the next section.

The second bound state and the scattering states describe not only displacements of the interface, but also a change in the magnetization profile. Accordingly, the energy eigenvalues $E_{n,\mathbf{p}}$ are always positive definite.

IV. INTERFACE FREE ENERGY, MAGNETIZATION, AND CORRELATION FUNCTIONS WITHIN THE HARMONIC APPROXIMATION

We confine ourselves to a calculation of thermodynamic quantities within the harmonic approximation where the normal modes do not interact.

The total free energy, $\beta F=-\ln Z$, may be written as

$$\beta F=S\{\bar{\mu}\}+\beta F_0+\beta F', \tag{4.1}$$

where $S\{\bar{\mu}\}$ is given by Eq. (3.5), $\beta F'$ is the contribution of the interaction part $S'\{\nu\}$, and βF_0 is the contribution of the noninteracting normal modes:

$$\beta F_0=-\ln A\int\delta\nu\exp(-S_0\{\nu\}). \tag{4.2}$$

Using the normalized eigenfunctions (3.13) and (3.14) we expand an arbitrary $\nu(\mathbf{r})$ according to

$$\nu(\mathbf{r})=\sum_{n\mathbf{p}}\xi_{n,\mathbf{p}}\nu_{n,\mathbf{p}}(\mathbf{r}), \tag{4.3}$$

where $\xi_{n,\mathbf{p}}$ are Gaussian random variables. Introducing (4.3) into (3.8), we obtain $S_0\{\nu\}$ in the diagonalized form

$$S_0\{\nu\}=\tfrac{1}{2}\sum_{n\mathbf{p}}E_{n\mathbf{p}}|\xi_{n\mathbf{p}}|^2. \tag{4.4}$$

The functional integral (4.2) is thereby transformed to an infinite-dimensional Riemann integral which can be solved immediately:

$$\beta F_0=-\ln A\int\prod_{n\mathbf{p}}d\xi_{n\mathbf{p}}\exp(-\tfrac{1}{2}\sum_{n\mathbf{p}}E_{n\mathbf{p}}|\xi_{n\mathbf{p}}|^2)$$

$$=\tfrac{1}{2}\sum_{n\mathbf{p}}\ln\frac{E_{n\mathbf{p}}}{2\pi}-\ln A. \tag{4.5}$$

This expression contains a bulk term ($\sim L^3$) and an interface term ($\sim L^2$) in the thermodynamic limit $L\to\infty$. The contribution to the interface free energy in which we are interested can be extracted easily by

[8] For a similar behavior of the stationary solution of a functional integral see J. Zittartz and J. S. Langer, Phys. Rev. **148**, 741 (1966).

using Eq. (3.19). We obtain

$$\beta F_0^{\text{i.f.}}=\frac{1}{2}\left(\frac{L}{2\pi}\right)^2\int d^2\mathbf{p}\times\left[\ln E_{1,\mathbf{p}}+\ln E_{2,\mathbf{p}}-\int_{-\infty}^{\infty}\frac{dK}{2\pi}s(K)\ln E_{K\mathbf{p}}\right],\quad(4.6)$$

where we have used

$$\int_{-\infty}^{\infty}\frac{dK}{2\pi}s(K)=2.$$

The quantity in brackets is calculated by residue techniques with the result

$$\beta F_0^{\text{i.f.}}=\frac{L^2}{8\pi^2}\int d^2\mathbf{p}\ln\frac{(\omega_\mathbf{p}-1)(\omega_\mathbf{p}-2)}{(\omega_\mathbf{p}+1)(\omega_\mathbf{p}+2)},\quad(4.7)$$

where $\omega_\mathbf{p}$ is defined in (3.11). Also the final integration over transverse wave vectors is elementary, if we introduce polar coordinates. It turns out that we have to use a cutoff parameter p_0 in order to avoid a short-wavelength divergence. This divergence is inherent in our continuum model and similar to the ultraviolet catastrophy in field theory. As our model is derived from a basic lattice model, it is quite natural to interpret the cutoff parameter as the maximal wave vector in the reciprocal lattice. With this in mind we get finally

$$\beta F_0^{\text{i.f.}}=-L^2\frac{\epsilon+\gamma}{8\pi a^2}\Big\{6(\bar\omega-\omega_0)+(\bar\omega^2-1)\ln\frac{\bar\omega+1}{\bar\omega-1}+(\bar\omega^2-4)\ln\frac{\bar\omega+2}{\bar\omega-2}-(\omega_0^2-1)\ln\frac{\omega_0+1}{\omega_0-1}-(\omega_0^2-4)\ln\frac{\omega_0+2}{\omega_0-2}\Big\},\quad(4.8)$$

where

$$\omega_0=\left(4+\frac{2\gamma}{\epsilon+\gamma}\right)^{1/2};\quad\bar\omega=\left(\omega_0^2+\frac{a^2p_0^2}{\epsilon+\gamma}\right)^{1/2}.\quad(4.9)$$

In the limit of vanishing external field ($\gamma\to 0$) the interface free energy per unit area reduces to

$$\frac{1}{L^2}\beta F_0^{\text{i.f.}}=\text{const.}-(4\pi a^2)^{-1}\gamma\ln\gamma.\quad(4.10)$$

This is exactly the same anomaly as found by Buff, Lovett, and Stillinger[2] in their liquid-gas model.

Other interesting thermodynamic quantities are the magnetization and correlation functions. The former one is thermodynamically defined by

$$M(\mathbf{r})=\frac{\delta}{\delta\lambda(\mathbf{r})}\ln Z=\langle\mu(\mathbf{r})\rangle.\quad(4.11)$$

Applying the transformation (3.6) we obtain

$$M(\mathbf{r})=\bar\mu(z)+\langle\nu(\mathbf{r})\rangle.\quad(4.12)$$

Within the harmonic approximation the second term vanishes and the magnetization reduces to the $\bar\mu$ function (3.4).

We define the two-point reduced correlation function by

$$G(\mathbf{r},\mathbf{r}')=\langle(\mu(\mathbf{r})-\bar\mu(z))(\mu(\mathbf{r}')-\bar\mu(z))\rangle=\langle\nu(\mathbf{r})\nu(\mathbf{r}')\rangle.\quad(4.13)$$

The average in this equation and in (4.12) is given through

$$\langle F\rangle=\frac{\int\delta\nu e^{-(S_0+S')}F}{\int\delta\nu e^{-(S_0+S')}},\quad(4.14)$$

where F denotes an arbitrary functional of ν. Using (4.3) we may calculate the right-hand side of (4.13) within the harmonic approximation

$$G_0(\mathbf{r},\mathbf{r}')=\sum_{n\mathbf{p},m\mathbf{q}}\langle\xi_{n\mathbf{p}}\xi_{m\mathbf{q}}\rangle_0\nu_{n\mathbf{p}}(\mathbf{r})\nu_{m\mathbf{q}}(\mathbf{r}')=\sum_{n\mathbf{p}}\frac{\nu_{n\mathbf{p}}(\mathbf{r})\nu_{n\mathbf{p}}^*(\mathbf{r}')}{E_{n\mathbf{p}}},\quad(4.15)$$

which is identified as the Green's function of the "Hamiltonian" (3.10) at zero energy, $G(\mathbf{r},\mathbf{r}'|\omega=0)$. Separating the transverse part of the wave functions we write

$$G_0(\mathbf{r},\mathbf{r}')=\frac{1}{(2\pi)^2}\int d^2\mathbf{p}\times e^{i[p_x(x-x')+p_y(y-y')]}G_{0\mathbf{p}}(z,z').\quad(4.16)$$

$G_{0\mathbf{p}}(z,z')$ is calculated using residue techniques:

$$G_{0\mathbf{p}}(z,z')=\frac{e^{-\omega_\mathbf{p}|t-t'|}}{2a(\epsilon+\gamma)^{1/2}\omega_\mathbf{p}}(\omega_\mathbf{p}^2-1)^{-1}(\omega_\mathbf{p}^2-4)^{-1}\times[1-\omega_\mathbf{p}^2-3\tanh^2 t-3\omega_\mathbf{p}\,\text{sgn}(t-t')\tanh t]\times[1-\omega_\mathbf{p}^2-3\tanh^2 t'+3\omega_\mathbf{p}\,\text{sgn}(t-t')\tanh t'].\quad(4.17)$$

The contribution of bound states to the correlations is seen more clearly by looking at the autocorrelation $G_0(\mathbf{r},\mathbf{r})$. From (4.17) we deduce

$$G_{0\mathbf{p}}(z,z)=\frac{1}{2a(\epsilon+\gamma)^{1/2}\omega_\mathbf{p}}\Big\{1+\frac{3}{\omega_\mathbf{p}^2-1}\tanh^2 t(1-\tanh^2 t)+\frac{3}{\omega_\mathbf{p}^2-4}(1-\tanh^2 t)^2\Big\}.\quad(4.18)$$

The second and third terms representing the bound-state contributions are large in those regions where the corresponding wave functions are concentrated. The behavior of the full autocorrelation function is of particular interest in the limit of vanishing field ($\gamma \to 0$). An elementary calculation leads to the result

$$G_0(\mathbf{r},\mathbf{r}) = C_1 + C_2 \tanh^2 t(1-\tanh^2 t) + \left[C_3 - \frac{3\sqrt{\epsilon}}{16\pi a^2}\ln\gamma\right](1-\tanh^2 t)^2, \quad (4.19)$$

where $C_{1,2,3}$ are constants. Obviously the logarithmic divergence in the last term is due to the large fluctuations caused by the instability of the interface in a vanishing external field. As was expected, the divergence is exactly the same as found by Buff, Lovett, and Stillinger.[2]

V. DISCUSSION

We have treated an Ising-like magnetic model in the presence of an external space-dependent magnetic field. As we were particularly interested in the interfacial structure, we had to choose an external field which favors the separation of two phases in space. It turned out that we could treat the inhomogeneous case with almost full rigor, at least in the harmonic approximation. The validity of this approximation depends essentially on the temperature range in which we are interested. It is clear that in the critical region all molecular-field treatments break down and one has to use more refined methods. Nevertheless we believe that our treatment is correct at low temperatures.[9] As was shown in the last section, we recover in our microscopic approach the predictions of Buff, Lovett, and Stillinger[2] in their treatment of interfacial structure in fluid systems. We believe that the coincidence of results is not limited to the harmonic approximation in both treatments. The results of Buff, Lovett, and Stillinger[2] at least remain valid in a rigorous non-harmonic treatment of a two-dimensional system; this is shown in a forthcoming publication of the author in collaboration with J. S. Langer.[10]

Whether our results remain valid in a calculation going beyond the harmonic approximation may be doubted. The perturbation expansion of the full free energy (4.1) is straightforward, if one uses methods developed by Langer for a similiar problem.[11] This would lead to a diagrammatic expansion in terms of the Green's function $G_0(\mathbf{r},\mathbf{r}')$ (4.15). As the energy denominator of the first bound-state contribution to G_0 leads to a logarithmic term (4.19) in small fields ($\gamma \to 0$), it is obvious that finite-order perturbation theory breaks down completely in this limit. It remains to be seen whether a partial summation of diagrams may overcome the difficulty. We hope to return to this problem in the future.

[9] As we stay away from the Curie temperature, the parameter ϵ (2.7) is finite. The effective coupling parameter α/ϵ can therefore be assumed to be small enough to allow for an expansion. Our molecular-field treatment gives the first two dominant contributions to the free energy. As usual, we expect this expansion to be correct only in the asymptotic sense.

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APPENDIX A

The one-dimensional equivalent of the differential equation (3.1) with vanishing external field is given by

$$-a^2\frac{d^2\bar{\mu}}{dz^2} - 2\epsilon\bar{\mu} + 4\alpha\bar{\mu}^3 = 0. \quad (A1)$$

We have to confine ourselves to solutions of this equation which contribute a bulk term to the expression (2.8) at most. This means that the physical solution must be bounded everywhere and constant at infinity. The latter condition implies vanishing derivatives in every order at infinity.

Equation (A1) immediately leads to the first integral:

$$-\tfrac{1}{2}a^2\left(\frac{d\bar{\mu}}{dz}\right)^2 - \epsilon\bar{\mu}^2 + \alpha\bar{\mu}^4 = C. \quad (A2)$$

The integration constant C is determined by the boundary condition

$$C = -(\epsilon^2/4\alpha) \quad \text{or} \quad 0. \quad (A3)$$

Using the first value we get the meaningful solution

$$\bar{\mu}(z) = \pm\left(\frac{\epsilon}{2\alpha}\right)^{1/2}\tanh\frac{\sqrt{\epsilon}}{a}(z-z_0), \quad (A4)$$

where z_0 is still arbitrary. The second value in (A3) leads to the unphysical solution

$$\bar{\mu}(z) = \pm\left(\frac{\epsilon}{\alpha}\right)^{1/2}\sec\frac{(2\epsilon)^{1/2}}{a}(z-z_0) \quad (A5)$$

which is not bounded and therefore must be discarded. It should be mentioned that Eq. (A1) also has the trivial solution

$$\bar{\mu} \equiv 0 \quad (A6)$$

which has to be discarded because the corresponding second variation of $S\{\mu\}$ (2.8) is not positive.