

Fluctuation-induced first-order transitions in systems with spatially isotropic competing interactions

Marcia C. Barbosa

*Instituto de Física, Universidade Federal do Rio Grande do Sul, Caixa Postal 15051, 91500 Porto Alegre,
Rio Grande do Sul, Brazil*

(Received 14 October 1991; revised manuscript received 5 December 1991)

We consider a spin system with competing interactions that are isotropic with respect to the axes of a cubic lattice. In the mean-field approximation the model supports paramagnetic, ferromagnetic, and modulated phases separated by a Lifshitz point. The character of this point is investigated in the presence of fluctuations. Using a standard diagrammatic formalism, we find that the paramodulated phase transition should be first order and that the Lifshitz point should be a critical endpoint.

a. Introduction. Models with competing interactions that result in modulated phases have received considerable attention recently. Such phases are present in binary alloys, ferrimagnets, copolymers, and microemulsions.¹

These superstructures can result from competition between ferromagnetic and antiferromagnetic interactions. To study this problem, Upton and Yeomans² introduced an Ising model with isotropic competing interactions that is described as follows: the Ising spins on a cubic lattice have first-neighbor ferromagnetic interactions J , next-neighbor antiferromagnetic interactions along the cubic axes $\kappa_1 J$, and antiferromagnetic interactions along the face diagonals $\kappa_2 J$. On going to a continuous-spin representation by adding a weighting term for each spin, we obtain the following Hamiltonian:

$$H = \frac{1}{2} \int_{\mathbf{q}} u_2 \phi(\mathbf{q}) \phi(-\mathbf{q}) + \frac{1}{4!} u_4 \iiint \phi(\mathbf{q}_1) \phi(\mathbf{q}_2) \phi(\mathbf{q}_3) \phi(-\mathbf{q}_1 - \mathbf{q}_2 - \mathbf{q}_3), \quad (1)$$

where u_4 is a weighting parameter and where

$$u_2 = k_B T - J(\mathbf{q}), \quad (2)$$

and, in this model,

$$\begin{aligned} \frac{1}{2} J(\mathbf{q}) = J [& \cos q_x + \cos q_y + \cos q_z \\ & - \kappa_1 (\cos 2q_x + \cos 2q_y + \cos 2q_z) \\ & - 2\kappa_2 (\cos q_x \cos q_y + \cos q_x \cos q_z \\ & + \cos q_y \cos q_z)]. \end{aligned} \quad (3)$$

Note that when $1 - 4\kappa_1 - 4\kappa_2 < 0$, $J(q)$ has a maximum at $\mathbf{q} = \mathbf{q}_c$. This wave vector for $\kappa_1 < \kappa_2/2$ is given by

$$\cos q_c = (1 - 4\kappa_2)/4\kappa_1, \quad (4)$$

and it is oriented along the x , y or z directions. For $\kappa_1 > \kappa_2/2$, it is oriented along one of the diagonals and is given by

$$\cos q_c = 1/(4\kappa_1 + 4\kappa_2). \quad (5)$$

At $\kappa_1 = \kappa_2/2$, the wave vector can assume any possible direction provided

$$\cos q_x + \cos q_y + \cos q_z = 1/4\kappa_1. \quad (6)$$

In this sense, one might note that as opposed to the axial

next-nearest-neighbor Ising (ANNNI) model,³ \mathbf{q}_c is not restricted to a unique direction.

Mean-field studies of this model indicate that both paramagnetic-ferromagnetic and paramagnetic-modulated phase transitions are continuous and meet the ferromagnetic-modulated first-order transition as $q_c \rightarrow 0$ at a Lifshitz point.²

Renormalization-group analysis carried out to a second-order expansion in $\epsilon = 4 - d$ demonstrates that the paramagnetic-modulated phase transition is first order for both $\kappa_1 > \kappa_2/2$ and $\kappa_1 < \kappa_2/2$, since the initial conditions lie out of the domain of attraction of the stable fixed point.^{4,5} Unfortunately, this result is based on a folding process of the original Brillouin zone in a new one given by $|\mathbf{q}_i| < q_c/2$, which is not valid as one allows $q_c \rightarrow 0$,⁶ and consequently is not valid as one approaches the Lifshitz point.

Then, in order to include fluctuations properly in this region, following Hornreich, Luban, and Shtrikman work (they have studied the $\kappa_1 = \kappa_2/2$ case),⁷ we applied a renormalization-group expansion in $\bar{\epsilon} = 8 - d$ (as $q_c \rightarrow 0$, the bare upper critical dimension is $d_+ = 8$), and we found that the same fixed point obtained for the $\kappa_1 = \kappa_2/2$ case can be used for the $\kappa_1 \neq \kappa_2/2$ case. This characterizes an isotropic Lifshitz point of Ising type. However, since in this case $d \approx 8$, the analysis based on $\bar{\epsilon} \ll 1$ cannot reveal the weak first-order disordered-modulated phase transition arising from fluctuations, which is expected below $d_c = 4$, that is, below the lower critical dimension of the $\bar{\epsilon}$ theory.^{4,5}

Given the inconclusiveness associated with this result, Levin and Dawson⁵ proposed that the three phases meet at a point in the universality class of a $2n$ model with $n = 7$ for the $\kappa_1 > \kappa_2/2$ case, assuming that $q_c \neq 0$. The problem with this analysis is that it has no control in the initial conditions and one cannot ensure that the initial parameters are not out of the domain of attraction of a stable fixed point.⁵

One might also point out that extensive Monte Carlo simulations have been carried out, indicating a clear Ising-like disordered ferromagnetic and a weak first-order disordered-modulated phase transition, but in the region between these limits the results are inconclusive.⁸ In this sense, one can see that the character of the Lifshitz point

region is still an open question.

One can also ask about the line $\kappa_1 = \kappa_2/2$ that was left aside in the previous analysis. In this limit, the model can be mapped into a lattice model for microemulsion, which has been receiving much attention recently.⁹ In this case, there are an infinite number of critical modes given by Eq. (6) and consequently, one cannot use the folding without generating overlaps between them. In this sense, the $\kappa_1 = \kappa_2/2$ line must be investigated by other means. One might also point out that, in this special case, as one approaches the Lifshitz point region, even results in the $\bar{\epsilon} = 8 - d$ expansion are not conclusive.^{4,10} Besides, Monte Carlo simulations in $d = 3$ dimensions are contradictory.^{11,12}

In this note we fix $\kappa_1 = \kappa_2/2$, the so-called microemulsion limit, and we analyze the paramagnetic-modulated phase transition assuming $q_c \ll 1$ in $d = 3$ dimensions.

$$H = H_1(\bar{\phi}) + H_1(\psi) + H_2, \quad (7)$$

$$H_1(\psi) = \int_{\mathbf{q}} \frac{1}{2} [r_0 + (q^2 - q_c^2)^2] \psi(\mathbf{q}) \psi(-\mathbf{q}) + \frac{1}{4!} u_0 \int \int \int \psi(\mathbf{q}_1) \psi(\mathbf{q}_2) \psi(\mathbf{q}_3) \psi(-\mathbf{q}_1 - \mathbf{q}_2 - \mathbf{q}_3), \quad (8)$$

$$H_2 = \int_{\mathbf{q}} [r_0 + (q^2 - q_c^2)^2] \psi(\mathbf{q}) \bar{\phi}(-\mathbf{q}) + \frac{u_0}{6} \int \int \int [\psi(\mathbf{q}_1) \psi(\mathbf{q}_2) \psi(\mathbf{q}_3) \bar{\phi}(-\mathbf{q}_1 - \mathbf{q}_2 - \mathbf{q}_3) + \frac{3}{2} \psi(\mathbf{q}_1) \psi(\mathbf{q}_2) \bar{\phi}(\mathbf{q}_3) \bar{\phi}(-\mathbf{q}_1 - \mathbf{q}_2 - \mathbf{q}_3) + \psi(\mathbf{q}_1) \bar{\phi}(\mathbf{q}_2) \bar{\phi}(\mathbf{q}_3) \bar{\phi}(-\mathbf{q}_1 - \mathbf{q}_2 - \mathbf{q}_3)], \quad (9)$$

where, assuming that the wave vector q_c is small, we rewrite from Eq. (2) as

$$r_0 = [k_B T / J - 6(1 - 5\kappa_1)] / \kappa_1 - [(1 - 12\kappa_1) / 4\kappa_1]^2 \quad (10)$$

and the coupling u_4 as

$$u_0 = u_4 / (2\kappa_1 J)^{1/2}. \quad (11)$$

Now, the equation of state can be determined from

$$h_q = \partial F(\bar{\phi}) / \partial \bar{\phi} = \langle \partial H(\bar{\phi}, \psi) / \partial \bar{\phi} \rangle, \quad (12)$$

and it is given by

$$h(q) = [r_0 + (q^2 - q_c^2)^2] \bar{\phi}(\mathbf{q}) + \frac{1}{6} u_0 \int_{\mathbf{q}_1} \int_{\mathbf{q}_2} \bar{\phi}(\mathbf{q}_1) \bar{\phi}(\mathbf{q}_2) \bar{\phi}(-\mathbf{q} - \mathbf{q}_1 - \mathbf{q}_2) + \frac{1}{2} u_0 \int_{\mathbf{q}_1} \int_{\mathbf{q}_2} \langle \psi(\mathbf{q}_1) \psi(\mathbf{q}_2) \rangle \bar{\phi}(-\mathbf{q} - \mathbf{q}_1 - \mathbf{q}_2) + \frac{1}{2} u_0 \int_{\mathbf{q}_1} \int_{\mathbf{q}_2} \langle \psi(\mathbf{q}_1) \psi(\mathbf{q}_2) \psi(-\mathbf{q} - \mathbf{q}_1 - \mathbf{q}_2) \rangle. \quad (13)$$

Now, in order to study the equation of state in Eq. (13), we have to compute $\langle \psi(\mathbf{q}_1) \psi(\mathbf{q}_2) \rangle$, as well as $\langle \psi(\mathbf{q}_1) \psi(\mathbf{q}_2) \psi(-\mathbf{q} - \mathbf{q}_1 - \mathbf{q}_2) \rangle$ to all orders in a loop expansion. This would be an impossible task, if we had to include all diagrams. Fortunately, as we will show next, we can eliminate most of them by simply assuming that the wave vector q_c is small but not zero. Let us begin by computing the two points correlation function $\langle \psi(q) \psi(-q) \rangle$. First, for simplicity, let us look at the

We employ a diagrammatic method, and we control divergences by a Hartree approximation.¹³ With this strategy, we compute the equation of state, as well as the free energy of each phase and compare them.

b. Equation of state and free energy. In this section we fix $\kappa_1 = \kappa_2/2$ and analyze the phases and transitions between them by including fluctuations. In order to do so, we compute the equation of state and the thermodynamic potential.

Following previous mean-field analyses,² we already know that three phases, i.e., disordered, ferromagnetic, and modulated phases, are present. In this case, in order to study the effect of fluctuations on the paramagnetic-modulated phase transition, one may rescale ϕ by a factor $(2\kappa_1 J)^{1/2}$ and write $\phi = \bar{\phi} + \psi$ with $\langle \phi \rangle = \bar{\phi}$, where $\langle \dots \rangle$ means, as usual, the thermal average. Then, the Hamiltonian, Eq. (1), will be given by

diagonal elements. To one loop order, one has

$$\langle \psi(q) \psi(-q) \rangle^{-1} = [r_0 + (q^2 - q_c^2)^2] + \frac{1}{2} u_0 \int_{\mathbf{q}_1} \frac{d^3 q_1}{[r_0 + (q_1^2 - q_c^2)^2]}. \quad (14)$$

Since the minimum of the propagator is attained at a surface $|\mathbf{q}| = q_c$, the most significant contribution to the integral comes from this region. Then, using a standard harmonic approximation, one has

$$u_0 \int_{\mathbf{q}_1} \frac{d^3 q_1}{[r_0 + (q_1^2 - q_c^2)^2]} = u_0 \int_{q_1} \frac{d^3 q_1}{[r_0 + 4q_c^2(q_1 - q_c)^2]} + O\left(\frac{u_0}{q_c^2}\right) = u_0 q_c / 4\pi r_0^{1/2} + O(u_0 / q_c^2), \quad (15)$$

where we assume that

$$r_0 \ll q_c^6. \quad (16)$$

This condition makes it possible to confine the problem to the region close to $\mathbf{q} = \mathbf{q}_c$ and still perform the calculations analytically. Obviously more complicated skeleton diagrams or diagrams with ladder loops may also contribute as we consider higher orders in the loop expansion in Eq. (14). These terms can appear in two forms: with and without external momentum dependence. In the first case, one has to add to Eq. (14) terms of the form

$$u_0^2 \int \frac{d^3 q_1 d^3 q_2}{\{r_0 + [q_1^2 - q_c^2]^2\} \{r_0 + [q_2^2 - q_c^2]^2\} \{r_0 + [(q_1 - q_2 - q)^2 - q_c^2]^2\}} \sim \frac{u_0^2}{r_0^{3/2}}. \quad (17)$$

Assuming now that (we will return to this point later)

$$u_0 q_c / 2r_0^{3/2} \sim 1, \quad (18)$$

the expression in Eq. (17) will exhibit relative order $r_0^{1/2}/q_c^2 \ll 1$ when compared with Eq. (15) if we assume the approximation in Eq. (16). From this one can readily note that more complicated diagrams of $n - 1$ loops have relative order $(u/q_c r_0)^n$ when compared with Eq. (15). Then, using Eq. (18) and Eq. (16), all these terms that depend on external momentum will be small when compared with last term in Eq. (14). Besides this small contribution, one also has a channel of terms without external momentum dependence given, for example, by

$$u_0^2 \int_{q_2} \frac{d^3 q_2}{[r_0 + (q_2^2 - q_c^2)^2]} \int \frac{d^3 q_1}{2\{r_0 + [q_1^2 - q_c^2]^2\}^2} \\ = (u_0^2 q_c / 4\pi r_0^{1/2}) \Pi \sim u_0^2 q_c^2 / r_0^2, \quad (19)$$

where Π represents “a bubble” diagram with no external momentum. Equation (19), in spite of the fact of being a two-loop order term, if one uses Eq. (18), exhibits relative order $O(1)$ when compared with Eq. (15). One can easily verify that this sort of important contribution will appear

$$\langle \psi(q_1) \psi(q_2) \rangle = \frac{1}{2} u_0 \{ \bar{\phi}(q_1) \bar{\phi}(q_2) / [r_0 + (q_1^2 - q_c^2)^2] [r_0 + (q_2^2 - q_c^2)^2] \} [1 + O(u_0/r)], \quad (22)$$

higher orders in the loop expansion will exhibit diagrams, where the propagators being integrated simultaneously depend on the external momenta and consequently do not coincide. In that case, we obtain, by using Eq. (18), that these terms will be $O(r_0/q_c)$ when compared with the zero-loop contribution, and consequently they will be negligible. Next, in order to complete the equation of state, the computation of $\langle \psi(q_1) \psi(q_2) \psi(q_3) \rangle$ is also needed. Using a similar analysis, we can see that the last term in Eq. (13) also depends on external momentum and is also small.

Finally, we have found, although using a different initial propagator, that the model we are considering here has an equation of state with the same form as Brazovskii's equation,¹³ namely,

$$h(q) = r(q_c) \bar{\phi}(q) + \frac{1}{6} u \int \int \bar{\phi}(q_1) \bar{\phi}(q_2) \bar{\phi}(q - q_1 - q_2) \\ - \frac{u}{2} \int \bar{\phi}(q_1) \bar{\phi}(-q_1) \bar{\phi}(q), \quad (23)$$

where

$$r = r_1 + \frac{u}{2} \int \bar{\phi}(q_1) \bar{\phi}(-q_1). \quad (24)$$

Now, having completed Eq. (13), we can study the paramagnetic-modulated phase transition properly.

in all orders in loop expansion in powers of $u_0 \Pi$ and must then be included. Since they do not have any external momentum dependence, we can take care of these terms by redefining the coupling u_0 as

$$u = u_0 [(1 - u_0 \Pi) / (1 + u_0 \Pi)], \quad (20)$$

where $\bar{\Pi} = 1/(1 + u_0 \Pi)$ represents a sum of this series of Π diagrams specified by Eq. (19).

Now we can understand the assumption Eq. (18). First note that we shall be interested in the region in which u becomes negative, i.e., the region where $u_0 \Pi \sim 1$, since a negative coupling introduces inflections in the free energy. Then a nonzero $\bar{\phi}$ can exist where otherwise only a disordered phase should be present.

One can observe that up to this moment in order to construct an equation of state with fluctuations taken into account we have only needed a renormalization in the coupling, and from Eqs. (14) and (15) a definition of a new parameter, namely,

$$r_1 = r_0 + u(q_c / 8\pi r_1^{1/2}). \quad (21)$$

Now let us return to the complete form of the correlation function $\langle \psi(q_1) \psi(q_2) \rangle$ for $q_1 \neq q_2$. Even if, in principle, one must also include off-diagonal elements to all orders, one can easily see that given that

First, let us introduce an explicit form for $\bar{\phi}$ given by

$$\bar{\phi}(\mathbf{r}) = 2a \cos(\mathbf{q}_c \cdot \mathbf{r}). \quad (25)$$

It is not difficult to show that this unidimensional structure should be a good choice since a nonunidimensional structure is unstable.¹³ One can note that the main feature of Eq. (25) is that, as opposed to the ANNNI case (in the uniaxial models one can use a two-order parameter theory, since we need an amplitude and a unique phase),³ we have here a unidirectional structure where the wave vector \mathbf{q}_c can assume any fixed direction. Here, we need an amplitude a and multiple choices of phases.

Then, with the specification of $\bar{\phi}$ given by Eq. (25), we can analyze the equation of state. Note that the expression for $h(q_c)$ given by Eq. (23) with Eq. (25), namely,

$$h(q_c) = r a - \frac{1}{2} u a^3, \quad (26)$$

besides the usual disordered phase $a = 0$ solution, also exhibits an $a \neq 0$ solution given by

$$a = (2r/u)^{1/2}. \quad (27)$$

Assuming $h = 0$, from Eqs. (26) and (24), one finds that

$$-r_0 = r + u(q_c / 8\pi r^{1/2}), \quad (28)$$

which has two real solutions for r if $-r_0 \geq$

$3(uq_c/16\pi)^{2/3}$.¹³ These solutions (one of them is a maximum and the other one is a minimum) become identical for $-r_0 = 3(uq_c/16\pi)^{2/3}$, where $r = -r_0/3$. For lower values of $-r_0$ only the $a = 0$ solution persists. Now, one has to compare the energies of the $a = 0$ and $a \neq 0$ phases. The free energy of the modulated phase differs from the free energy for the disordered phase by

$$\begin{aligned} \Delta F &= \int_0^a 2hd\bar{a} = \int_{r_1}^r 2h \frac{da}{dr} dr \\ &= -r_1^2/2u - r_1^{1/2}/8\pi - r^2/2u + r^{1/2}/8\pi. \end{aligned} \quad (29)$$

If one analyzes Eq. (29) together with Eq.(24) and Eq. (21), it is not difficult to verify that ΔF will be negative, when $-r_0 > 2(uq_c/8\pi)^{2/3}$ and, consequently, at $-r_0 \approx 2(uq_c/8\pi)^{2/3}$, one has a first-order transition from a disordered to a modulated phase.

c. Summary. In this paper, we have studied the paramagnetic-modulated phase transition present in the Ising model with isotropic competing interactions. This model exhibits incommensurate phases with wave vector $q = q_c$ pointing in any fixed but not specific direction provided $\kappa_1 = \kappa_2/2$. The relevance of fluctuations in such kinds of systems could have been conjectured, since the previously studied cases $\kappa_1 > \kappa_2/2$ and $\kappa_1 < \kappa_2/2$ exhibit first-order transition induced by fluctuations effects.

In order to verify this, we introduced fluctuations in $d = 3$ dimensions. We applied a diagrammatic expansion where most of the diagrams were considered small assuming q_c is not too small. In that case, we showed that the transition is first order. This result is in agreement with the more general case proposed in Ref. 14.

From this, one can see that, at least in $d = 3$, fluctuations change the mean-field result which indicates a continuous transition. Now one can understand the previous analysis where Mukamel and Luban, using usual $d = 8 - \epsilon$ expansion to study the vicinity of the isotropic

Lifshitz point, could not define a susceptibility related to a continuous disordered modulated phase transition.¹⁰ From our point of view, this result indicates that even in $d \approx 8$ dimensions the disordered-modulated phase continuous transition is not stable, and this transition should be first order.

One can, otherwise, argue that our approximation is not valid at the Lifshitz point where $q_c = 0$ and, consequently, we could not say that the disordered-modulated phase transition should stay first order even when $q_c = 0$. However, we would like to point out that the condition Eq. (16) allows us to reach regions in the close vicinity of the Lifshitz point, and even there we find no indication of a continuous transition. In this sense, we might well say that the so-called Lifshitz point could be a triple point (the point where three first-order lines meet) if the paramagnetic-ferromagnetic transition is also first-order or a critical endpoint (the region where a critical line ends in a first-order line), otherwise. Results from Hornreich, Luban, and Shtrikman indicate that even near the so-called Lifshitz-point regime the paramagnetic-ferromagnetic transition is still continuous and, in that sense, one cannot have a triple point.⁷ Then, we can suggest that it should be a reasonable guess that the microemulsions model exhibits a first-order paramagnetic-modulated phase transition that meets the continuous paramagnetic-ferromagnetic transition at an endpoint.

One still has to understand how this changes if one allows $\kappa_1 \neq \kappa_2/2$ at the Lifshitz point. Naturally it is possible to guess that no drastic change will occur and that the endpoint is still there.

I am particularly grateful to Professor Alba Theumann and Dr. John D. Gaffey, Jr. for their critical reading of the manuscript. I want also especially to thank Professor K. A. Dawson for sending me a copy of his work prior to publication. This work was supported by Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq).

¹J. Yeomans, *Solid State Phys.* **41**, 151 (1988).

²Paul Upton and Julia Yeomans, *Phys. Rev. B* **40**, 479 (1989); K. A. Dawson, *J. Phys. A* **36**, 3383 (1987).

³W. Selke, *Phys. Rep.* **170**, 213 (1988).

⁴Marcia C. Barbosa, *Phys. Rev. B* **42**, 6363 (1990).

⁵Y. Levin and K. A. Dawson, *Phys. Rev. B* **42**, 1976 (1990).

⁶M. Droz and M. D. Coutinho Filho, in *Magnetism and Magnetic Materials*, Proceedings of the 21st Annual Conference on Magnetism and Magnetic Materials, AIP Conf. Proc. No. 29, edited by J. J. Becker, G. H. Lander, and J. J. Rhyne (AIP, New York, 1976), p. 465.

⁷R. M. Hornreich *et al.*, *Phys. Rev. Lett.* **35**, 1678 (1975).

⁸K. A. Dawson *et al.*, *Physica A* **165**, 320 (1990).

⁹B. Widom, *J. Chem. Phys.* **84**, 6443 (1986); B. Widom *et al.*, *Physica A* **140**, 26 (1986).

¹⁰David Mukamel and Marshall Luban, *Phys. Rev. B* **18**, 3631 (1978).

¹¹K. A. Dawson *et al.*, *J. Chem. Phys.* **87**, 6211 (1987); **88**, 5149 (1988).

¹²D. Stauffer and N. Jan, *J. Chem. Phys.* **87**, 6210 (1987).

¹³S. A. Brazovskii, *Zh. Eksp. Teor. Fiz.* **68**, 175 (1975) [*Sov. Phys. JETP* **41**, 85 (1975)].

¹⁴Y. Levin, C. J. Mundy, and K. A. Dawson (unpublished).