Landau Theory of a System with Two Bilinearly Coupled Order Parameters in External Field: Exact Mean Field Solution, Critical Properties and Isothermal Susceptibility

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We present simple description of a system with two bilinearly coupled order parameters in external field based on an exact mean-field solution of the Landau Hamiltonian. It reproduces the qualitative form of the "field-temperature" phase diagram given by a molecular-field model and by more sophisticated theories and experiments on metamagnets. The solution gives the same critical exponents as the molecular-field theory, but it is not restricted to the magnetic systems only and it is easier to handle, since it formulates the results in explicit analytical form. The susceptibility in this model does not diverge at the second order transition line (far from a higher order critical point separating the second and first order transition lines), but jumps down from the lower temperature wing to the higher temperature one. The jump amplitude is proportional to the square of the field in small fields and diverges in large fields close to the higher order critical point.

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A Landau Hamiltonian approach with two coupled order parameters has been used to describe phase transitions in antiferroelectrics [1, 2] and complex magnetic systems [3–7], lattice deformations coupled to magnetic [8–10] or superconducting transitions [11], surface phase transitions in interacting membranes [12, 13], and other phenomena [14–18]. The coupling between order parameters might have different functional forms depending on the particular physical mechanism of the interaction and on the symmetry constraints. Bilinear [1, 2, 9, 12, 13, 18], linear-quadratic [8], biquadratic [14, 16] and more complex [17] types of coupling have been considered.

To the best of our knowledge, no exact solution in an external field has been reported even for the simplest system with bilinear coupling. On the other hand, this case corresponds to a model of a metamagnet (antiferromagnet with strong anisotropy). For metamagnets a number of results have been obtained within the molecular-field model and within more sophisticated theories establishing the phase diagram of the system and the pertinent critical exponents (for a review see [19, 20]). These theories appear to be in good agreement with the experimental data [19, 20], but many of the often cumbersome solutions have been obtained by means of multiple approximations making the picture less transparent. Some of the solutions are not even given in explicit form.

The goal of this paper is to demonstrate that the Landau theory for a system with two bilinearly coupled order parameters has an exact analytical solution which gives a very transparent description directly applicable to many systems, not only those of the magnetic origin. The critical properties of this model do not differ from those prescribed by the molecular-field theories for metamagnets, but more results can be obtained in analytical form. For example, a very simple law is obtained for the susceptibility jump at a second-order phase transition.

We consider the Landau Hamiltonian

$$F = a(p^2 + q^2) + \frac{b}{2}(p^4 + q^4) + 2\gamma p q - h(p + q). \tag{1}$$

where p and q are the two order parameters, $\gamma > 0$ is the coupling constant, and h the external field, $b > 0$,

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and a can change the sign with temperature variation. In [13] we obtained an exact solution for the order parameters not found earlier for a formally similar Landau Hamiltonian [2]. Here we use this solution to build a complete phase diagram in the (h, a) plane, to calculate susceptibilities, and to analyse the critical properties. To preserve generality, we will express most of the results in terms of the formal parameters a, b, γ instead of temperature. However, to illustrate simplest temperature effects, for some estimates we will assume that a is the only temperature dependent parameter in (1) and use the Landau expansion $a \approx A(T/T_0 - 1)$, which is valid, strictly speaking, only at temperatures close to T_0.

The system of equations arising from minimization of the free energy has "symmetric" and "nonsymmetric" solutions [13]. The symmetric solution is

$$p = q = \alpha_s + \beta_s, \tag{2}$$

$$\alpha_s = \left[\frac{h}{4b} + \left(\frac{(a+\gamma)^3}{27b^3} + \frac{h^2}{16b^2}\right)^{\frac{1}{2}}\right]^{\frac{1}{3}},$$
$$\beta_s = \left[\frac{h}{4b} - \left(\frac{(a+\gamma)^3}{27b^3} + \frac{h^2}{16b^2}\right)^{\frac{1}{2}}\right]^{\frac{1}{3}}. \tag{3}$$

where in the case of complex α_s and β_s one must choose any pair of values which satisfy the condition $\alpha_s \beta_s = -(a+\gamma)/3b$. The nonsymmetric solutions are

$$p = \frac{3h}{4[3b(\alpha_n + \beta_n) + 2\gamma - a]} \pm \frac{1}{2}\sqrt{3(\alpha_n + \beta_n) - \frac{2a}{b}},$$
$$q = \frac{3h}{4[3b(\alpha_n + \beta_n) + 2\gamma - a]} \mp \frac{1}{2}\sqrt{3(\alpha_n + \beta_n) - \frac{2a}{b}}, \tag{4}$$

where

$$\alpha_n = \frac{2\gamma - a}{3b} \cdot \left[1 - \frac{27h^2 b}{8(2\gamma - a)^3}\left\{1 - \left(1 - \frac{16(2\gamma - a)^3}{27h^2 b}\right)^{\frac{1}{2}}\right\}\right]^{\frac{1}{3}},$$
$$\beta_n = \frac{2\gamma - a}{3b} \cdot \left[1 - \frac{27h^2 b}{8(2\gamma - a)^3} \cdot \left\{1 + \left(1 - \frac{16(2\gamma - a)^3}{27h^2 b}\right)^{\frac{1}{2}}\right\}\right]^{\frac{1}{3}}, \tag{5}$$

and $\alpha_n \beta_n = (2\gamma - a)^2/9b^2$. (We exclude from the consideration an additional symmetric solution, $p = q$, with the sign opposite to the external field, which exists at $a < -\gamma$. This solution gains stability at $a < -2\gamma$ and small h, but even then it is only metastable, having higher free energy than the solution given by (2), (3) [13].)

The stability conditions for the symmetric and nonsymmetric solutions define a phase diagram of the system in the (a, h) plane. This diagram, shown in Fig. 1, has four regions. In region I only the symmetric phase is stable. In region II only the nonsymmetric phase is stable. The solid line separating these two regions is given by the equation

$$h_{I-II} = h_c(a) = \frac{4}{3\sqrt{3b}}(a + 2\gamma)\sqrt{\gamma - a},$$
$$a \geq \frac{2}{3}\gamma. \tag{6}$$

This is a line of a second-order transition. In region III the symmetric phase is stable and the nonsymmetric phase is metastable; the situation reverses in region IV. The thin line separating regions III and IV is a line of a first-order transition calculated numerically from the condition of the equality of the free energies. The dashed lines, defining the stability limits of the symmetric and nonsymmetric phases, are given by

$$h_{II-IV} = \frac{4}{3\sqrt{3b}}(a + 2\gamma)\sqrt{\gamma - a},$$
$$-2\gamma \leq a < \frac{2}{3}\gamma, \tag{7}$$

$$h_{I-III} = \frac{4}{3\sqrt{3b}}(2\gamma - a)^{3/2}, \quad a < \frac{2}{3}\gamma. \tag{8}$$

All four curves meet at a higher order critical point

$$a_* = \frac{2}{3}\gamma, \quad h_* = \frac{32\gamma^{3/2}}{27b^{1/2}}, \tag{9}$$

where they have a common tangent. The solid curve in Fig. 1 reproduces the well known phase diagram of a metamagnet [19–21], while the dashed curves confine the metastable regions, most easily identified in the Landau theory.

The phase transition from the symmetric to the nonsymmetric phase occurs as a second-order transition at $h \leq h_*$ and as a first-order transition at $h > h_*$. The trajectories of the order parameters in the (p, q) plane with the variation of h along the paths shown in Fig. 1a are plotted in Fig. 1b, c. These trajectories illustrate the continuous $(1 \rightarrow 2)$ and the jump-wise $(3 \rightarrow 4)$ breakdown of the system symmetry at these transitions.

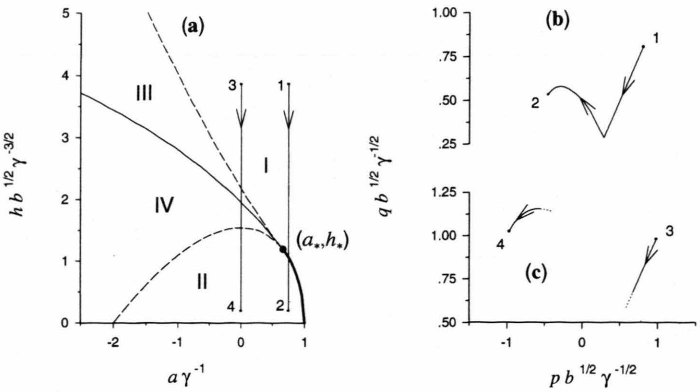

Fig. 1. Phase diagram (a) in the "temperature"-field (a, h) plane. The solid line is a line of a second order transition from a symmetric ($p=q$, region I) to nonsymmetric ($p\neq q$, region II) phase. The thin line is a line of a first order transition. In two neighboring regions III and IV, bounded by dashed lines, both the symmetric and nonsymmetric phases can exist; one phase is stable and the other is metastable. Trajectories (b, c) illustrate variation of the order parameters with h decreasing along the paths 1→2 or 3→4 shown on the phase diagram (*a*). Dotted lines show metastable solutions.

The spontaneous symmetry breakdown at the second-order transition can be characterized by an asymmetry parameter, $\eta=p-q$ (usually referred to as staggered magnetization in the context of the theory of metamagnets). It is zero in the symmetric phase. In a small vicinity below the critical line, η follows a critical law

$$\eta\approx\left(\frac{12(\gamma-a)}{b(3a-2\gamma)^2}\right)^{1/4}(h_c(a)-h)^{1/2}. \tag{10}$$

At the higher order critical point $a=2\gamma/3$ and (10) takes the form

$$\eta\approx\sqrt{2}\left(\frac{\gamma}{b^3}\right)^{1/8}(h_*-h)^{1/4}. \tag{11}$$

These critical indices coincide with those given by the molecular-field theory of metamagnets [20].

Note that this model due to its symmetry can be reduced to a one-dimensional problem with the single critical order parameter, η (see e.g. [21]). We, however, keep the notion of two coupled order parameters that is widely accepted in the literature [1–18] and that allows to preserve the generality important for lower symmetry problems.

In weak external fields and with weak coupling between the order parameters, the Landau approximation for $a(T)$, $a\approx A(T/T_0-1)$, allows one to evaluate the temperature dependencies as well. Then (6) gives the temperature of the second order transition,

$$T_c\approx T_0\left(1+\frac{\gamma}{A}-\frac{3b}{16A\gamma^2}h^2\right), \tag{12}$$

demonstrating the reduction of the critical temperature in an external field. Quadratic dependence of this type on the external field is known in the molecular theory of antiferromagnetism [22]. It was also obtained in an Ising model [23]. The critical behavior of the asymmetry parameter below T_c is given by

$$\eta\approx 2\sqrt{\frac{aA}{b(3a-2\gamma)}}\left(\frac{T_c-T}{T_0}\right)^{1/2}, \tag{13}$$

which at the higher order critical point takes the form

$$\eta\approx 2\left(\frac{A\gamma}{3b^2}\right)^{1/4}\left(\frac{T_c-T}{T_0}\right)^{1/4}. \tag{14}$$

Both critical exponents are, naturally, the same as in the molecular-field approach. The isothermal susceptibility,

$$\chi=\partial(p+q)/\partial h, \tag{15}$$

can be expressed through the solutions for the order parameters, (2)–(3) or (4)–(5), as

$$\chi_s=\frac{1}{a+\gamma+3bp^2}, \tag{16}$$

$$\chi_n=\frac{1}{4a-2\gamma-6bpq} \tag{17}$$

for the symmetric and nonsymmetric phases, respectively. The plots for the susceptibilities as functions of the external field are shown in Figs. 2a, b (along the order parameter trajectories illustrated in Figure 1).

The "temperature" dependence of the susceptibility at three characteristic values of the external field ($h=0$, $0<h<h_*$, $h>h_*$) is plotted in Figures 3a, b, c. Note that the spike at the critical temperature [24] is seen only at $h=0$. In any nonzero field the susceptibility exhibits a break at the second-order transition,

$$\Delta\chi=\chi_n-\chi_s=\frac{3(\gamma-a)}{2\gamma(3a-2\gamma)}>0. \tag{18}$$

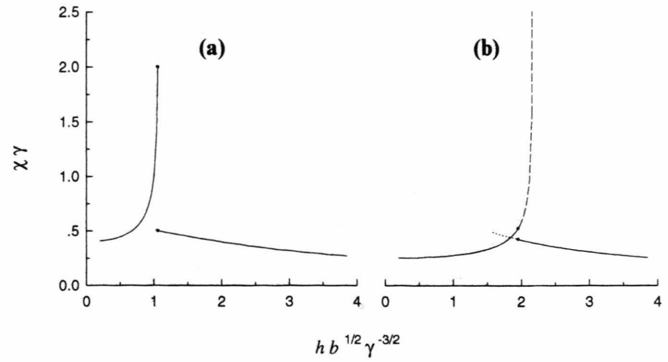

Fig. 2. Susceptibilities as a function of the external field plotted along the trajectories $1 \rightarrow 2$ (a) and $3 \rightarrow 4$ (b) from Fig. 1. Solid circles mark the susceptibilities at phase transition points. Dashed and dotted lines in Fig. 2b show susceptibilities for metastable solutions. The susceptibility for metastable nonsymmetric phase diverges with increasing field as the field approaches the limit of stability for this phase.

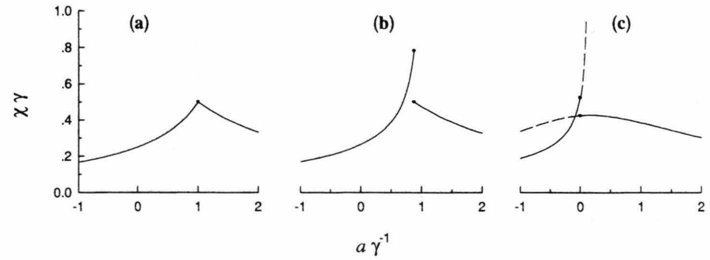

Fig. 3. "Temperature"-dependence of susceptibilities at $h=0$ (a), $h=0.77\,\gamma^{3/2}\,b^{-1/2}$ (b), and $h=1.95\,\gamma^{3/2}\,b^{-1/2}$ (c). Solid circles mark the susceptibilities at second- (a), (b) and first- (c) order transition points. Dashed lines show susceptibilities for metastable solutions.

In particular, in a small field the jump near the critical temperature, T_c, increases quadratically,

$$\Delta\chi = \chi(T_c^{-0}) - \chi(T_c^{+0}) \approx \frac{9}{32}\frac{b}{\gamma^4}h^2. \tag{19}$$

Within the Landau approximation for $a(T)$ we can calculate the heat capacity C. At the second-order transition it does not diverge, but as it is well known exhibits a jump,

$$\Delta C = C(T_c^{-0}) - C(T_c^{+0}) \approx \frac{2\,T_c\,A^4\,(T_c - T_0)^2}{b\,\gamma\,T_0^3\,(3\,A\,(T_c - T_0) - 2\,\gamma\,T_0)} \tag{20}$$

which does not vanish at $h=0$, $\Delta C(h=0) = (2\,A^2\,T_c)/(b\,T_0^2)$.

The hysteresis of the susceptibility, which accompanies the first-order transition at $h > h_*$ (see (9)), is illustrated in Figure 2b. The metastable branches are shown by dotted lines. Note that the susceptibility in the nonsymmetric phase, χ_n, diverges with increasing field as this metastable branch approaches its existence limit.

From (16)–18) one can see that the susceptibility in the nonsymmetric phase diverges at the higher order critical point defined by (9)

$$\chi_n(h) \approx \frac{1}{4}(\gamma\,b)^{-1/4}(h_* - h)^{-1/2}. \tag{21}$$

The susceptibility in the symmetric phase remains finite, so that the suceptibility jump diverges as well. The same is true for the heat capacity. In the case of weak coupling, $\gamma \ll A$, the Landau approximation for $a(T)$ in the vicinity of this point gives

$$\chi_n(T) \approx \frac{1}{8}\sqrt{\frac{3}{\gamma A}}\left(\frac{T_* - T}{T_0}\right)^{-1/2}, \tag{22}$$

$$C_n(h) \approx \frac{4}{9}\frac{A^2\,\gamma^{3/4}\,T_*}{b^{5/4}\,T_0^2}(h_* - h)^{-1/2},$$

$$C_n(T) \approx \frac{2\,T_*\sqrt{3\,\gamma\,A^3}}{9\,b\,T_0^2}\left(\frac{T_* - T}{T_0}\right)^{-1/2}, \tag{23}$$

where

$$T_* = T_0\left(1 + \frac{179}{243}\frac{\gamma}{A}\right). \tag{24}$$

These critical indices are again the same as in the molecular-field theory of metamagnets.

The Landau Hamiltonian given by (1) represents a simple phenomenological model of a metamagnet in an external field. In this context the most interesting qualitative prediction of our calculation is that for any nonzero field the low and high temperature wings of the susceptibility of not merge at the "Neel" point where the susceptibility at the low temperature branch is larger. When the field tends to zero, the susceptibility jump vanishes as the square of the field. When the field increases and approaches the region of a first-order transition, the susceptibility jump diverges.

While the presence of the jump can be deduced from the solutions obtained in the molecular-field theory, to our knowledge the dependence of the jump on the field has never been carefully described. The experimental data [20, 25, 26] show a spike rather than a break in the susceptibility, but a certain assymmetry of the low and high temperature wings indicates that there might be a break which is not well resolved (see in particular Fig. 12 of [25] and Fig. 1b of [26]). This spike is interpreted as an indication of weak divergence in the susceptibility [24, 25]. In order to check the prediction of the mean-field theory we have conventionally treated the singularity as a break having estimated the magnetic field dependence for the first three curves from Fig. 12 of [25] (low field limit). The result of this rather crude evaluation, shown in Fig. 4, is consistent with the predicted quadratic dependence of the susceptibility break on the external field. Such analysis, however, should be regarded with extreme caution because the resolution procedure is not accurately defined. Note that impurities in real systems might be a source of a staggered external magnetic field which is known to smear the transition [19] and to transform the break into an asymmetric peak.

More distinct data exist for field-dependent susceptibility curves at different temperatures, see e.g. Fig. 10 [25]. To show the qualitative proximity of the predictions of the Landau theory, in Fig. 5 we plot a family of $\chi(h)$ curves for a set of a-values. Note, however, that a is linear in T only near the critical point. Still, this family of curves is indeed qualitatively similar to the one observed in [25].

Of course, predictions based on a mean-field approach will be most likely inaccurate close to the critical line. In addition, the Landau model does not account for any specific properties of any particular system. Still, a relatively simple, exact solution obtained for a general class of systems can be an instructive reference base for building more accurate and therefore much more complex theories.

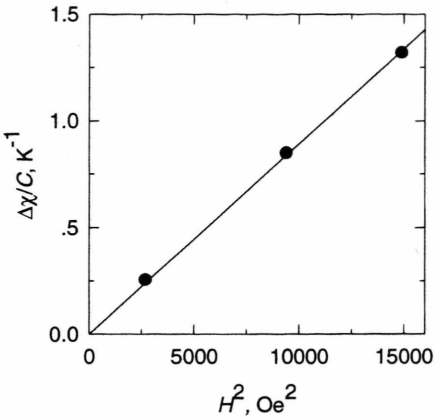

Fig. 4. The parallel susceptibility "jump" in $(C_2H_5NH_3)_2CuCl_2$, $\Delta\chi$, normalized by the Curie constant C, plotted vs. the magnetic field H (as evaluated from Fig. 12 of [25]).

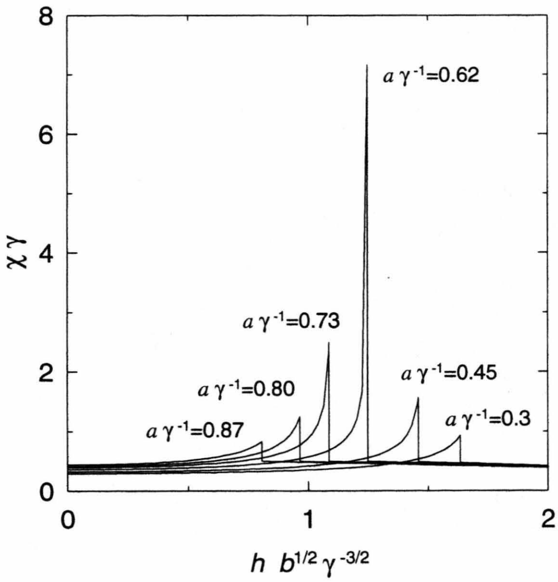

Fig. 5. Field-dependent susceptibility curves shown for different values of a. Susceptibilities are shown for the stable solutions only. Spikes on the curves correspond to phase transition points, a first-order transition at $a\gamma^{-1} < 2/3$ and a second-order transition at $a\gamma^{-1} > 2/3$. At the triple critical point, $a\gamma^{-1} = 2/3$, the susceptibility break diverges. This is illustrated by a sharp increase in the susceptibility break shown at $a\gamma^{-1} = 0.62$. To stress the qualitative similarity with experimental observations, the susceptibilities are shown by continuous lines even though they exhibit a break, corresponding to the vertically dropping line right after the maximum.

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