Incommensurate phases near multicritical points

I. L. Kraĭzman and V. P. Sakhnenko
Scientific-Research Institute of Physics, Rostov-on-Don State University

(Submitted 27 May 1984)
Pis'ma Zh. Eksp. Teor. Fiz. **40**, No. 5, 173–175 (10 September 1984)

A condition is formulated for distinguishing order parameters which satisfy the Lifshitz criterion but which induce incommensurate phases. Cascades of phase transitions for four-component order parameters are examined as an example. A suggestion is made regarding the magnetic structure of a modulated phase of $CeAl_2$.

One factor of a symmetry nature which would cause an instability of a spatially homogeneous state of a crystal near lines of second-order phase transitions is a violation of the Lifshitz condition.[1] In this case, as Dzyaloshinskiĭ has shown for magnetic systems, phases arise in which the period of the magnetic order is incommensurate with the period of the crystal lattice.[2] An analogous incommensurability of the "main" lattice and an order period has been found for structural transitions, and in several cases it has been explained by arguments based on the Lifshitz condition.[3] In the present letter we show that there are symmetry reasons for the appearance of incommensurate phases even if the Lifshitz condition holds.

We assume that the symmetry degree $[T]^n$ of the representation T under which the multicomponent order parameter η transforms contains some representation T' which does not satisfy the Lifshitz condition. In this case the thermodynamic potential Φ incorporates mixed invariants which are of degree n in η and which are linear in the components ξ that pertain to T'. Upon the condensation of some of the η_i, terms which are bilinear in the components η_i and ξ_i that have not undergone condensation may be conserved in the Landau potential of the corresponding low-symmetry phase. This phase is accordingly unstable with respect to spatially inhomogeneous distortions. In other words, the original potential Φ contains gradient invariants of the Lifshitz type which are nonlinear in the η_i and which acquire the usual Lifshitz form in the low-symmetry phase. The transition from the low-symmetry phase of interest to other homogeneous phases described by the order parameter η may therefore occur through an incommensurate intermediate phase.

As an example we consider spontaneous orientational transitions in cubic crystals of symmetry O_h^7, described by four-component order parameters of the star of the vector $\mathbf{k}_9 = (2\pi/a)(1/2,\ 1/2,\ 1/2)$. Superstructures pertaining to these $\mathbf{k}$ arise upon antiferromagnetic ordering in cubic Laves phases $R\,Al_2$ (R is a rare-earth ion). The $4D$ irreducible representations of T_i ($i = 1, \ldots 4$) of this star satisfy the Lifshitz condition but contain in $[T_i]^2$ a $6D$ irreducible representation $T_1^{(k_{10})}$ (the designations are in the scheme of Ref. 4) which does not satisfy this condition. According to this symmetry, there can be a gradient invariant of the type

$$\left[\eta_1\eta_2 \frac{\partial(\eta_3\eta_4)}{\partial x} - \eta_3\eta_4 \frac{\partial(\eta_1\eta_2)}{\partial x}\right] + \left[\eta_1\eta_3 \frac{\partial(\eta_2\eta_4)}{\partial y} - \eta_2\eta_4 \frac{\partial(\eta_1\eta_3)}{\partial y}\right] + \left[\eta_1\eta_4 \frac{\partial(\eta_2\eta_3)}{\partial z} - \eta_2\eta_3 \frac{\partial(\eta_1\eta_4)}{\partial z}\right]. \tag{1}$$

The Landau potential for these order parameters contains one anisotropic invariant of fourth degree, $\beta_1 \Sigma_{i=1}^4 \eta_i^4$. On a line of second-order transitions, $\alpha_1(p, T) = 0$, the point $\beta_1 = 0$ separates phases 1 ($\eta 000$) and 2 ($\eta\eta\eta\eta$). At it, phases 3 ($\eta\eta 00$) and 4 ($\eta\eta\eta 0$), corresponding to saddle points of the function $\Sigma_{i=1}^4 \eta_i^4$ ($r^2 = \Sigma_{i=1}^4 \eta_i^2 = \text{const}$), may touch the symmetric phase. All of these phases exist in regions of width $\sim\tau = (T - T_M/T_M)$ near the multicritical point p_M, T_M ($\alpha_1 = \beta_1 = 0$). It can be seen from (1) that in phase 3 ($\eta\eta 00$) the Landau potential contains the Lifshits invariant, with a coefficient on the order of τ. Accordingly, an incommensurate phase (phase also appears in area $\sim\tau$) appears between the phases ($\eta\eta 00$) and ($\eta\eta\eta 0$) near p_M, T_M. The phase diagram is shown in Fig. 1.

We believe that this analysis casts some light on the magnetic phases which are observable in $CeAl_2$ at liquid-helium temperatures. Two suggestions have been made here. First, a 3**k** structure arises with a condensation of three components of the 24-component order parameter of the star, $\mathbf{k} = (2\pi/a)(1/2, 1/2, 1/2)$, and the observed superstructural reflection, which corresponds to $\mathbf{k}_9 = (2\pi/a)(1/2, 1/2, 1/2)$, is due to nonlinear interactions. In other words, the four-component order parameter drops out as nonintrinsic.[5] It is shown in Ref. 6, however, that this assumption contradicts experimental data from a study of $CeAl_2$ subjected to uniaxial stress. For their part, Barbara *et al.*[6] suggest that a mixture of phases is observed at standard pressure below $T_c = 3.8$ K: a homogeneous antiferromagnetic phase and a phase which is modulated along [110]. Actually, as follows from the analysis above, the observable state of the crystal may be a single-phase state with two homogeneous components of the four-

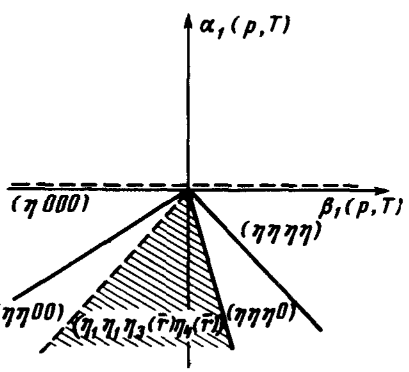

FIG. 1. Cascade of phase transitions near a multicritical point containing an incommensurate phase [an irreducible representation T_i ($i = 1, \ldots 4$) of the star $\mathbf{k}_9 = (2\pi/a)(1/2, 1/2, 1/2)$ of space group O_h^7]. Solid lines—First-order phase transitions; dashed lines—second-order phase transitions; hatching—incommensurate phase.

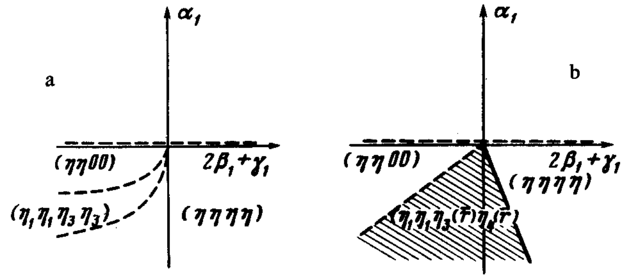

FIG. 2. Displacement of the corner phase by the incommensurate (hatched) phase near a four-phase point. The irreducible representation T_i ($i = 1, \ldots 4$) of the star $\mathbf{k}_{11} = (1/2)\mathbf{b}_2$ of space group D_{4h}^{17}. Solid lines—First-order phase transitions; dashed—second-order phase transitions.

component order parameter η ($\eta_1 = \eta_2 = \eta$) and with two modulated components $\eta_3(y, z)$ and $\eta_4(y, z)$. It follows from the phase diagram, however, that such a phase could not arise directly from the symmetric phase upon a second-order phase transition: There must be phases with a homogeneous order between them. The existence of such phases in $CeAl_2$ can be suggested on the basis of an extrapolation to $x \to 0$ of experimental data on phase transitions in $Ce_{1-x}Pr_xAl_2$. This extrapolation furnishes evidence of a commensurate antiferromagnetic structure above $T_c = 3.8$ K (Ref. 7). Finally, we note that phase-transition cascades analogous to that in Fig. 1, with an alternation of commensurate and incommensurate (long-period) phases, should be typical of order parameters of dimensionality $n \geqslant 4$. In the O_h^7 group, for example, all the order parameters with $n \geqslant 4$ either do not satisfy the Lifshitz condition or allow the existence of invariants of the type in (1).

Incorporating nonlinear gradient invariants of the Lifshitz type may also lead to more radical changes in the phase diagrams. In the simplest case of a tetragonal anisotropy, $\beta_1 \Sigma_{i=1}^4 \eta_i^4 + \gamma_1(\eta_1^2\eta_2^2 + \eta_3^2\eta_4^2)$, for example, $(\eta\eta 00)$ and $(\eta\eta\eta\eta)$ phases border each other on the second-order phase-transition line at the point $\alpha_1 = 0$, $2\beta_1 + \gamma_1 = 0$. When only homogeneous states are taken into account, the phase diagram in the α_1, $2\beta_1 + \gamma_1$ plane is as shown in Fig. 2a. If Φ contains invariants of the type in (1), however, the phase diagram changes qualitatively: Instead of the corner phase $\eta_1\eta_1\eta_3\eta_3$ of "width" $\sim\tau$ an incommensurate phase of width $\sim\tau$ appears between $(\eta\eta 00)$ and $(\eta\eta\eta\eta)$ phases (Fig. 2b). A situation of this sort occurs, for example, for the four-component order parameter of the star $\mathbf{k}_{11} = (1/2)\mathbf{b}_2$ of the group D_{4h}^{19}.

[1]E. M. Lifshitz, Zh. Eksp. Teor. Fiz. **11**, 253, 269 (1941).
[2]I. E. Dzyaloshinskiĭ, Zh. Eksp. Teor. Fiz. **46**, 142 (1964) [Sov. Phys. JETP **19**, 960 (1964)].
[3]A. P. Levanyuk and D. G. Sannikov, Fiz. Tverd. Tela (Leningrad) **18**, 423 (1976) [Sov. Phys. Solid State **18**, 245 (1976)].
[4]O. V. Kovalev, Neprivodimye predstavleniya prostranstvennykh grupp. (Irreducible Representations of Space Groups) Kiev, 1961.
[5]S. M. Shapiro, E. Gurewitz, R. D. Parks, and L. C. Kupferberg, Phys. Rev. Lett. **43**, 1748 (1979).
[6]B. Barbara, M. F. Rossignol, Y. X. Boucherle, and C. Vettier, Phys. Rev. Lett. **45**, 938 (1980).
[7]B. Barbara, Y. X. Boucherle, J. L. Buevoz, M. F. Rossignol, and J. Schweizer, J. Magn. Magn. Mat. **14**, 221 (1979).

Translated by Dave Parsons
Edited by S. J. Amoretty