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Universal Incommensurate Structures

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Certain phase transitions in quasiperiodic systems are characterized by *universal* structures. In these cases the functional form of the order parameter corresponding to the modulated phase, $\mathbf{P}(\mathbf{r})$, is determined by the symmetry properties of the system and is independent of the details of the associated Landau–Ginzburg model. Here we consider a simple one-dimensional XY-like model corresponding to this type of phase transition. The universal modulated structure of this model is calculated numerically at various points along the critical line.

KEY WORDS: Universal structures; incommensurate systems; phase transitions; instabilities.

1. INTRODUCTION

Incommensurate or quasiperiodic structures occur in a large variety of physical systems. These include magnetically ordered systems with spiral-like or spin-density-wave structures which are incommensurate with the underlying lattice,[1] intercalation compounds,[3] charge-density-wave systems,[4] and many others. A particularly interesting class of quasiperiodic systems consists of quasicrystals. These newly discovered structures have icosahedral, decagonal, or other noncrystallographic point symmetries.[4] Phase transitions leading to quasiperiodic structures have been studied extensively in recent years. Although a complete classification of these transitions has not yet been carried out, it has been observed that continuous transitions leading to incommensurate structures may be broadly divided into three classes[5-7]: instability, nucleation, and a third, intermediate, class which has common characteristics with both of the first two.

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[3] See ref. 2 for a review of intercalation compounds of graphite.

[4] See refs. 3 for reviews of charge-density waves.

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Instability transitions usually take place when a disordered phase becomes incommensurate. For example, a transition from a paramagnetic phase to one with spin-density-wave (SDW) structure is of this type. The order parameter associated with this transition is the Fourier amplitude $\mathbf{S}(\mathbf{q})$ of the local spin variable $\mathbf{S}(\mathbf{r})$, where $2\pi/|\mathbf{q}|$ is the wavelength of the modulated structure. The wavevector $\mathbf{q}$ is nonzero at the transition temperature and it varies with the temperature T in the ordered phase. Higher harmonics of the primary order parameter, namely $\mathbf{S}(n\mathbf{q})$ with $n>1$, are also nonzero below the transition. The magnetic structure of the ordered phase, and in particular the amplitudes of the higher harmonics, depend on the details of the Hamiltonian associated with the system. However, as the transition is approached from below, the amplitude ratios $|\mathbf{S}(n\mathbf{q})|/|\mathbf{S}(\mathbf{q})|$ vanish for $n>1$. Thus, at the transition, the magnetic structure of the SDW phase is composed of the primary order parameter, with no higher harmonics present. This trivial structure is universal, and is characteristic of the instability-type phase transitions.

On the other hand, nucleation transitions are not associated with a small local order parameter. They are rather described by a condensation of discommensurations or domain-wall-like structures. Such transitions usually take place when an incommensurate structure becomes commensurate. As the transition is approached, the average distance between the discommensurations diverges to infinity, resulting in a continuous transition to a commensurate phase. For example, consider a spiral magnetic structure in an external magnetic field perpendicular to the axis of the spiral. As the magnetic field is increased, the spiral becomes distorted. The magnetic structure is then composed of large ferromagnetic domains oriented along the magnetic field, separated by domain walls in which the local magnetic moment undergoes a rapid 2π rotation. When the field is further increased, the separation between the domain walls increases, and becomes infinite at a critical field. At this point the structure becomes ferromagnetic, namely commensurate with the underlying lattice. Below the transition the structure is characterized by the fundamental Fourier mode $\mathbf{S}(\mathbf{q})$ and its harmonics. However, unlike instability transitions, the wavevector $\mathbf{q}$ vanishes at the critical point. Moreover, the ratios $|\mathbf{S}(n\mathbf{q})|/|\mathbf{S}(\mathbf{q})|$, $n>1$, of the amplitudes of the higher harmonics to that of the fundamental one remain finite at the transition. This is a result of the fact that domain walls are localized objects. The amplitude ratios do not exhibit any universal features, and they depend on the details of the model under consideration.

It has recently been pointed out[7] that a third, intermediate, class of transitions leading to incommensurate structures may exist in certain systems. This class of transitions has common features with both of the first two. In particular, it is characterized by a small local order parameter $\mathbf{P}(\mathbf{q})$

as in instability transitions. However, at the transition the wavevector $\mathbf{q}$ vanishes, and the amplitude ratios $|\mathbf{P}(n\mathbf{q})|/|\mathbf{P}(\mathbf{q})|$, $n>1$, remain finite, as in nucleation transitions. Such transitions occur when the incommensurability is induced by a gradient cubic term of the form $P^2\nabla\cdot\mathbf{P}$ in the corresponding Landau–Ginzburg (LG) model.[7–12] This is in contrast with instability transitions which are induced by quadratic terms, such as Lifshitz terms of the form $(P_x(\partial P_y/\partial z)-P_y(\partial P_x/\partial z))$. It has been argued that this type of phase transition is characterized by *nontrivial universal structure*.[7] In particular, it has been demonstrated that close to the transition point the amplitude ratios $|\mathbf{P}(n\mathbf{q})|/|P(\mathbf{q})|$ become independent of the details of the LG model associated with the system. They are determined by the symmetry properties of the order parameter corresponding to the phase transition. Transitions of this type occur in quartz and in berlinite.[13–16] These crystals exhibit structural transitions from a hexagonal β phase to an incommensurate phase, induced by a $P^2\nabla\cdot\mathbf{P}$-type term. These transitions have been studied in detail in recent years.

In this paper we consider a simple model corresponding to the intermediate-type phase transition. The model is introduced in Section 2. In Section 3 we present our numerical results for the universal incommensurate structure associated with this model.

2. THE MODEL

Consider an isotropic system undergoing a transition to a ferroelectric phase. Let $\mathbf{P}(\mathbf{r})$ be the local polarization vector. The LG model associated with this system is

$$\mathscr{H}=\int d^3r[\alpha P^2+a(\nabla P)^2+\beta P^2\nabla\cdot P+\gamma P^4+\delta P^6] \tag{1}$$

where, as usual $\alpha\sim(T-T_0)$ represents the deviation from the bare critical point T_0, the parameters a, γ, and δ are taken to be positive, and we consider terms up to sixth order in $\mathbf{P}$. The symmetry of this problem is such that the LG model does not have a Lifshitz-type term. This, together with the assumption that $a>0$, implies that the quadratic terms in this model favor ferroelectric, or commensurate, order. The incommensurability is induced by the gradient-cubic term β, which leads to a phase transition of the intermediate type.

The phase diagram associated with this model has been studied in detail.[7–12] It exhibits three phases: a disordered phase in which $\mathbf{P}=0$, a ferroelectric phase, and another phase characterized by a modulated structure. A schematic phase diagram is given in Fig. 1. For small β the phase

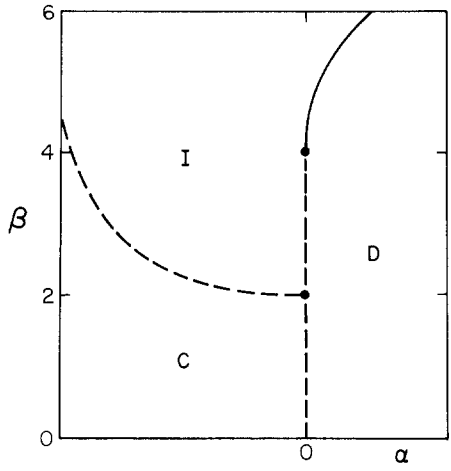

Fig. 1. A schematic phase diagram corresponding to the model (1). It exhibits disordered (D), ferroelectric commensurate (C), and incommensurate (I) phases. Dashed lines denote continuous transitions and solid lines denote first-order transitions.

diagram exhibits a disordered phase (D) at $\alpha > 0$ and a ferroelectric, or commensurate, phase (C) at $\alpha < 0$. The DC transition is an ordinary continuous instability transition. For large β the ferroelectric phase becomes modulated, or incommensurate (I). In the case $\delta = 0$ the transition occurs at $\beta^2 = 4\alpha\gamma$. The DI transition is of the intermediate type discussed above. As β further increases above some critical value β_t, the DI transition becomes first order via a tricritical point. Some features of this phase diagram are modified when thermal fluctuations are taken into account. In particular, renormalization group studies of the Hamiltonian (1) corresponding to the n-component order parameter were carried out[11] in $d = 4 - \varepsilon$ dimensions, $\varepsilon > 0$. It was found that to leading order in ε and for sufficiently small n the model does not possess a stable fixed point. This indicates that in $d = 3$ the DI and the DC transitions may in fact be weakly first order.

In order to study the structure of the modulated phase near the DI line, we consider the region $\alpha < 0$ and $(4\alpha\gamma)^{1/2} < \beta < \beta_t$ in the parameter space. We introduce rescaled variables for the polarization

$$\mathbf{P} = (-\alpha/\gamma)^{1/2}\,\boldsymbol{\sigma} \tag{2a}$$

and for $\mathbf{r}$,

$$\mathbf{r} = (-a/\alpha)^{1/2}\,\mathbf{x} \tag{2b}$$

In terms of these rescaled variables the Euler–Lagrange equation associated with the Hamiltonian (1) is

$$\nabla^2\boldsymbol{\sigma} + \boldsymbol{\sigma} - 2\boldsymbol{\sigma}\sigma^2 + \lambda[\nabla(\sigma^2/2) - \boldsymbol{\sigma}(\nabla\cdot\boldsymbol{\sigma})] - \frac{3\alpha\delta}{\gamma^2}\sigma^4\boldsymbol{\sigma} = 0 \tag{3}$$

where $\lambda = (\beta^2/a\gamma)^{1/2}$. This equation determines the structure of the modulated phase. Close to the DI transition line the parameter α is small and the last term in this equation may be neglected. Therefore, at the transition, the structure of the scaled polarization $\boldsymbol{\sigma}$ is determined by Eq. (3) with $\alpha = 0$. This structure is *universal* and does not depend on the details of the LG model associated with the system. In particular, higher order terms in $\mathbf{P}$ and its derivatives do not affect the modulated structure close to the transition line, since they contribute terms to the Euler–Lagrange equations which scale with some positive power of α. They may thus be neglected at the transition. However, the universal structure does depend on the parameter λ, and it varies along the DI line. This aspect of the intermediate-type transition is quite general, and is not specific to the ferroelectric system considered here. In any LG model corresponding to an intermediate-type transition the universal structure is determined by the terms of order four or less in the order parameter, and higher order terms may be neglected. The universal structure is therefore determined by the symmetry of the problem under consideration via the parameters (such as λ in our case) which occur in the *truncated* LG model. The universal structure associated with the β-incommensurate transition in quartz and in berlinite has been considered in previous studies.[13] Unfortunately, it was found that the corresponding Euler–Lagrange equation has a trivial, harmonic-free, solution. The universal structure in this case is therefore composed of the fundamental harmonic, as in ordinary instability transitions.

3. NUMERICAL RESULTS

In order to study the universal structures which may be obtained in intermediate-type transitions, we consider a somewhat simplified version of the Hamiltonian (1). We take the polarization $\mathbf{P}$ to be an XY vector, $\mathbf{P} = (P_x, P_y)$, and look for one-dimensional solutions of the form $\mathbf{P}(y)$. The Euler–Lagrange equation (3) takes the form

$$\frac{d^2\sigma_x}{dy^2} + \sigma_x - 2\sigma^2\sigma_x - \lambda\sigma_x\frac{d\sigma_y}{dy} = 0 \tag{4a}$$

$$\frac{d^2\sigma_y}{dy^2} + \sigma_y - 2\sigma^2\sigma_y + \lambda\sigma_x\frac{d\sigma_x}{dy} = 0 \tag{4b}$$

where $\sigma^2 = \sigma_x^2 + \sigma_y^2$. We have integrated these equations numerically and determined the modulated structure at various points along the DI line. This is done by first finding a periodic solution of Eq. (4) with period $2\pi/q$, for some wavenumber q. The free energy associated with this solution is calculated, and the wavenumber q of the solution which minimizes the free energy is determined. This procedure is repeated for several points $2 \leqslant \lambda \leqslant \lambda_t$ along the DI line, where $\lambda_t = 4$ corresponds to the tricritical point. The results for the universal structure are summarized in Fig. 2. The corresponding wavenumber $q(\lambda)$ which minimizes the free energy is given in Fig. 3. Note that q is associated with the rescaled polarization $\sigma(y)$. The

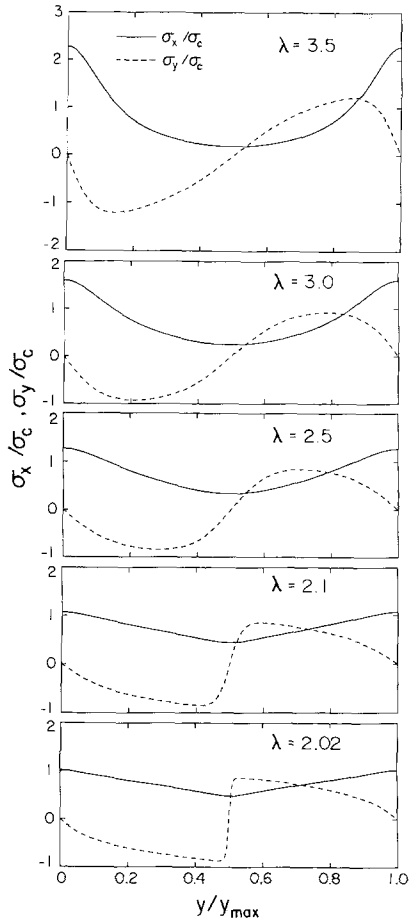

Fig. 2. The universal modulated structure at various points λ along the DI line. Here $Y_{max} = 2\pi/q$, where q is the wavenumber of the modulated structure.

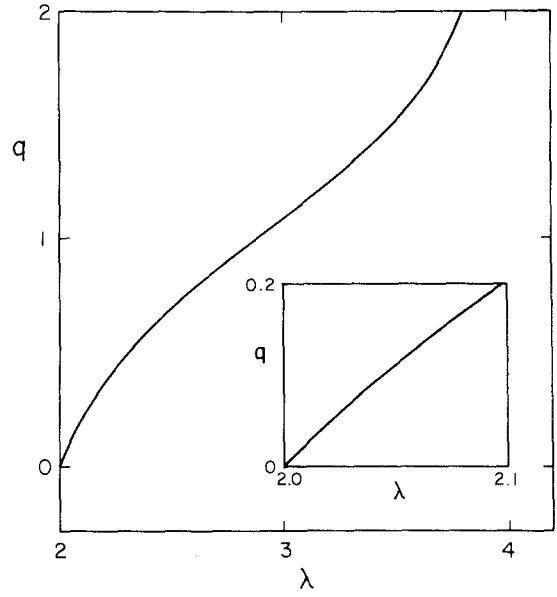

Fig. 3. The wave number q of the modulated structure along the DI line.

wavenumber corresponding to the unscaled polarization $\mathbf{P}(\mathbf{r})$ is given by $q(-\alpha/a)^{1/2}$ and it vanishes in the limit $\alpha \to 0$.

As is evident from Fig. 2, the universal structure is nontrivial, namely, it has a nonnegligible contribution from higher harmonics. This contribution is more pronounced close to the CI transition, $\lambda = 2$, and to the tricritical point, $\lambda = \lambda_t$.

The modulated structure may be calculated perturbatively in the vicinity of the CI line, namely near $\lambda = 2$. In this limit the wavenumber q approaches zero, indicating a transition to the commensurate ferroelectric phase. Taking $\lambda = 2 + \varepsilon$, where $\varepsilon > 0$ is a small parameter, one finds that to leading order in ε, Eq. (4) has a solution of the form

$$\begin{aligned} \sigma_x &= \frac{1}{\sqrt{2}} + \varepsilon \sigma_1(y) \\ \sigma_y &= \sqrt{\varepsilon}\, \sigma_2(y) \\ q &= \sqrt{\varepsilon}\, q_1 \end{aligned} \tag{5}$$

where $\sigma_1(y)$ and $\sigma_2(y)$ satisfy some nonlinear coupled differential equations. Therefore, near the CI line, the modulated structure is roughly composed of a ferroelectric σ_x component accompanied by a small modulated σ_y. The CI transition is of peculiar, possibly intermediate-type, nature. It would be interesting to study this transition in more detail.

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