Ground State, Spectrum, and Phase Diagram of Two-Dimensional Incommensurate Crystals

V. L. Pokrovsky[a]
Baker Laboratory and Materials Science Center, Cornell University, Ithaca, New York 14850

and

A. L. Talapov
Landau Institute of Theoretical Physics, 142432 Chernogolovka, U. S. S. R.

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A model of a monolayer of atoms adsorbed on a crystalline surface is discussed. When the initial incommensurability, δ, between a reciprocal lattice vector of the monolayer and one of the substrate is small, the monolayer has a one-dimensional superstructure with period depending on δ. For $\delta > \delta_c(T)$ the state is incommensurate; $\delta_c(T)$ decreases and vanishes at a T_c given by the elastic moduli of the monolayer. The spectrum of small oscillations has two acoustic branches, one vanishing at the phase transition.

Incommensurate crystals revealed experimentally in many physical systems[1] have become the subject of considerable theoretical interest. The simplest microscopic model of an incommensurate crystal has been proposed by Ying[2] and investigated in some detail in the works of Pokrovsky,[3] Pokrovsky and Uimin,[4] and Novaco and McTague.[5] They succeeded in proving the existence of an acoustic branch of the spectrum in the framework of perturbation theory. A nonperturbative approach for one-dimensional systems has been presented practically simultaneously by Bulayevsky and Khomsky,[6] Theodorou and Rice,[7] and the present authors.[8] In this work an analogous two-dimensional problem is considered. We hope to apply it to describe the experimentally observed two-dimensional incommensurate crystals of the atoms adsorbed on bulk crystal surfaces (for details see the reviews).[9] Here a brief summary of assumptions and results will be given. The details will be published elsewhere.

We consider a lattice of atoms, situated on a plane, interacting with elastic forces. The interaction with the substrate is described by a periodic potential and assumed to be weak compared to a characteristic elastic energy. The ground state, spectrum, and phase diagram may be calculated explicitly provided one of the reciprocal lattice vectors $\vec{p}_0$ of the atomic lattice is close to some reciprocal lattice vector $\vec{q}_0$ of the substrate. Only one pair of vectors is assumed to be close. This is, typically, the case if the atomic lattice and the substrate lattice are of different symmetry. Only "resonance" deformations associated with the periodicity along $\vec{p}_0$ will be substantial. These deformations are slowly varying functions of distance. Thus, continuum elasticity theory may be applied. The potential energy has the form

$$U = \int \left\{ \frac{\lambda}{2}\left(\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y}\right)^2 + \frac{\mu}{2}\left[\left(\frac{\partial u}{\partial x} - \frac{\partial v}{\partial y}\right)^2 + \left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x}\right)^2\right] + V(u + \delta x) \right\}. \tag{1}$$

The quadratic differential form here represents the elastic energy; u and v are components of displacement; λ and μ are the Lamé coefficients, while V is a periodic function with the period, a, of the substrate. Finally, δ is a measure of the incommensurability, $\delta = |q_0 - p_0|a$; V and δ will be considered as small parameters.

The first problem is to minimize the potential energy (1). The solution of this problem depends on the parameter δ. We introduce a critical value

$$\delta_c = \tfrac{1}{2}[(\lambda + \mu)/\lambda\mu]^{1/2}\langle [V(\varphi) - V_{\min}]^{1/2}\rangle, \tag{2}$$

where the brackets denote an averaging over the period a, i.e., $\langle f(\varphi)\rangle = a^{-1}\int_0^a f(\varphi)\,dx$, and $V_{\min}$ is the minimum value of $V(\varphi)$. An incommensurate lattice is preferable for $\delta > \delta_c$. The density and the displacements in the incommensurate phase are modulated along a direction which forms an angle, θ, with the vectors $\vec{q}_0$ and $\vec{p}_0$ (which are parallel), where

$$\cos^2\theta = (1 + \sigma)^{-1} \tag{3}$$

and $\sigma = (\lambda - \mu)/(\lambda + \mu)$ is the Poisson coefficient. The period of the superstructure is equal to

$$L = a\mu^{1/2}\langle [\mathcal{E} + V(\varphi)]^{-1/2}\rangle \tag{4}$$

and the constant $\mathcal{E}$ is related to the initial incommensurability by

$$\delta = \tfrac{1}{2}[(\lambda + \mu)/\lambda\mu]^{1/2}\langle[\mathcal{E} + V(\varphi)]^{1/2}\rangle. \tag{5}$$

The resulting incommensurability may be represented by the value $\langle \partial\varphi/\partial x\rangle = a/L$, where $\varphi = u + \delta x$. Besides this, there is a constant longitudinal deformation along the y axis, namely,

$$\partial v/\partial y = \sigma\delta. \tag{6}$$

The energy of the incommensurate crystal is

$$U_1 = -\mathcal{E} + [2\lambda\mu/(\lambda + \mu)]\delta^2. \tag{7}$$

When δ tends to δ_c, the period L diverges as $-\ln(\delta - \delta_c)$ and the parameter $\mathcal{E}$ tends to $-V_{\min}$. For $\delta < \delta_c$ a *commensurate* crystal becomes preferable. Its energy is

$$U_Z = V_{\min} + [2\lambda\mu/(\lambda + \mu)]\delta^2. \tag{8}$$

In the commensurate phase ($\varphi = \text{const}$ and $v = 0$) the transition between commensurate and incommensurate phases is of second order. Physically it is governed by the concentration of atoms.

The total Hamiltonian $\mathcal{H}$ differs U [see Eq. (1)] by kinetic energy terms. Taking these into account, we find that the spectrum of small oscillation for the incommensurate phase consists of two acoustic and, generally, many optical branches. In the long-wave limit the spectrum of the acoustic branches is, explicitly,

$$\begin{aligned} \omega_1^{\,2} &= \frac{(\lambda + \mu)^2}{2\lambda\rho}[c^2(p - q\sqrt{\sigma})^2 + 4\sigma q^2], \\ \omega_2^{\,2} &= \frac{\lambda + \mu}{\rho}\left[\frac{\lambda + \mu}{4\mu}(p - q\sqrt{\sigma})^2 + q^2\right], \end{aligned} \tag{9}$$

where p and q are the wave-number components and

$$c^{-2} = 2\mu\left(\frac{\lambda}{\lambda + \mu}\right)^{1/2} a\delta\,\frac{d(L^{-1})}{d\mathcal{E}}. \tag{10}$$

The first frequency, ω_1, corresponds to oscillations along the direction of density modulation. As expected, it vanishes at the phase transition. Below the phase transition only an optical branch of these oscillations remains with a gap $\Delta = (V_0''/\rho)^{1/2}$, where V_0'' is the second derivative of $V(\varphi)$ at the minimum.

Thermal fluctuations destroy the long-range order in the incommensurate state. Nevertheless, the incommensurate phase does exist in some region of temperature and concentration. The first problem to solve is the dependence of the critical value, δ_c, on the temperature. It can be found using the renormalization-group approach, which was developed for the similar problem of the sine-Gordon field by Wiegmann.[10] For further consideration we take $V = V_0\cos(2\pi\varphi/a)$. The resulting phase diagram is shown schematically in Fig. 1. The commensurate and incommensurate phases are denoted by C and I, respectively. Everywhere along the phase boundary a phase transition of second order is predicted by the theory. The critical line ends at the points $T = T_0 = 4\mu(\lambda + \mu)a^2/\pi(\lambda + 2\mu)$, $\delta = 0$, $T = 0$, $\delta = \delta_c(0)$, where $\delta_c(0)$ is defined by (2). If T is not too close to the critical temperature $[|T - T_0| \gg T_0 V/(\gamma + \mu)]$, the analytic expression for $\delta_c(T)$ is

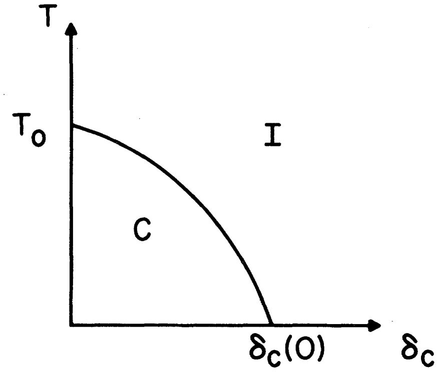

FIG. 1. Phase diagram in the (T, δ) plane showing the critical locus and the commersurate (C) and incommensurate (I) phases.

$$\delta_c(T) = \frac{[T(\lambda + \mu)]^{1/2}}{\pi a(\lambda\mu)^{1/2}}\left(\frac{V_0 a^2}{T}\right)^{1/2[1-(T/T_0)]}. \tag{11}$$

The gap Δ in the spectrum of oscillations of the commensurate crystal is proportional to $\delta_c(T)$.

The elastic moduli do not depend significantly on temperature. The resulting incommensurability may be found by using a transformation from Bose to Fermi operators, similar to that used by Luther and Emery[11] for the sine-Gordon field and by Luther and Pokrovsky[12] for the analogous one-dimensional problem. The resulting incommensurability $\langle \partial\varphi/\partial x\rangle$ near the phase transition line behaves as $[\delta - \delta_c(T)]^{1/2}$ or $[T - T_c(\delta)]^{1/2}$. It should be recalled that, on neglecting the fluctuations, $\langle \partial\varphi/\partial x\rangle$ behaves as $[\ln(\delta - \delta_c)]^{-1}$. The discrepancy between these two results can be explained by the increasing amplitude of fluctuations in the close vicinity of the critical curve. Similar considerations for quantum fluctuations were presented in our previous work.[8]

Our analysis may be directly applied to the lattice of vortices in a superconducting film with a periodically modulated thickness.[13] Second-order commensurate/incommensurate transitions have been observed experimentally for molecules H_2 and D_2, atoms He^3 and Kr, and some others adsorbed on a Grafoil substrate.[9] Our theory cannot be applied directly to these cases because two pairs of basic reciprocal lattice vectors become close simultaneously. The appropriate calculations are now in progress.

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(a) Permanent address: Landau Institute of Theoretical Physics, 142432 Chernogolovka, U. S. S. R.

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Absence of Substrate Nucleation for bcc Solid ^3He on Grafoil

J. Landau and Y. Eckstein
Department of Physics, Technion–Israel Institute of Technology, Haifa, Israel

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In contrast to the case of ^4He, we find no evidence for spontaneous nucleation of a uniform surface solid layer from the liquid phase of ^3He near Grafoil substrates. However, surface solid could be detected after forcing the rapid growth of bulk solid from external points of nucleation. The experiment covers the temperature range between 30 and 320 mK at pressures just below the bulk melting curve.

The role of a Grafoil substrate in nucleating solidification from superfluid ^4He was studied recently by Landau and Saam[1] (L-S). The principal conclusion was that uniform nucleation of a surface solid phase always occurs as the pressure approaches the bulk melting curve. If the general analysis of Dash[2] and Peierls[3] are applied to this problem, one concludes that the growth of the ^4He surface solid is analogous to their Class-I adsorption process (i.e., unimpeded growth of the adsorbed layer). The principal purpose of this experiment was to test whether the Class-I behavior also applied to nonsuperfluid ^3He on Grafoil. We found, to our surprise, that ^3He does not show any sign of uniform nucleation of a solid phase near the bulk melting curve. These results, combined with the observation from neutron diffraction studies[4-7] that the completed monolayer of noble gases, including ^3He and ^4He, adsorbed on Grafoil displays a triangular symmetry, suggest that continuous growth of a liquid-solid interface is possible only when the symmetry of the monolayer matches the symmetry of the bulk solid lattice. The hexagonal structure of solid ^4He matches with a triangular monolayer while the cubic structure of ^3He does not. Thus the adsorptive solidification of ^3He on Grafoil should be classified as Class II in the Dash-Peierls scheme.

In the L-S experiment the nucleation of solid ^4He layers on Grafoil in equilibrium with bulk superfluid ^4He was studied using equation-of-state–type measurements. They observed isopycnals[8] just below the bulk melting curve, find-