Orientational instability of higher-order commensurate lattices

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(Received 12 January 1984)

Stability of higher-order commensurate structures, formed by a harmonically distorted two-dimensional latice in a periodic uniaxial potential, are investigated. The minimum-energy orientation-angle $\theta_{\min}$ dependence on the lattice constant to the potential period ratio is numerically determined. The number of stable commensurate phases is found to be surprisingly small—in most cases certain incommensurate orientations turn out to be energetically more favorable. A simple criterion is given to single out the stable commensurate phases.

Two-dimensional (2D) structures formed in the presence of an external periodic field—monolayers adsorbed on a crystal substrate,[1] flux-line lattices in periodically modulated superconductors,[2,3] etc.—exhibit a variety of phases. The simplest solid phases are lattices in registry with the imposed periodicity. The low-order commensurate (C) phases and the transition to the incommensurate (I) phase have been extensively studied recently, both experimentally and theoretically. Here we concentrate on the stability conditions for higher-order commensurate phases. We show that most of them are unstable with respect to the formation of an I structure of the same density, but with a different orientation. This effect makes the 2D problem essentially different from the analogous 1D case studied by Theodorou and Rice;[4] in the latter case every C state is stable. On the other hand, our result completes that of McTague and Novaco,[5] who found that the energy of incommensurate structures is strongly orientationally dependent.

Assuming a weak interaction with the external field, we have studied the ground-state structure in the one-harmonic approximation which is meaningful provided that the system is not too close to the simple (first-order) commensurability conditions.[6] The calculation has been performed for the case of a uniaxial pinning potential, relevant for superconductors modulated along a given direction[2,3] and for physisorbed films on anisotropic substrates.[7] Finally, some applications to the case of the substrate potential with hexagonal symmetry, which acts, for example, on the atoms adsorbed on graphite,[1] are discussed.

We consider a 2D system of particles at zero temperature, situated at the positions $\vec{r}_i$ in the x^1-x^2 plane, and interacting *via* the binary interaction $W(|\vec{r}_j-\vec{r}_i|)$. The results are obtained using the Lennard-Jones potential

$$W(r)=4\epsilon\left[\left(\frac{\sigma}{r}\right)^{12}-\left(\frac{\sigma}{r}\right)^{6}\right] \tag{1}$$

for the interaction between two atoms at distance $r=|\vec{r}_j-\vec{r}_i|$. Here ϵ is the depth of the pair potential well and σ is the "hard-core" diameter. However, it should be mentioned that very similar results were obtained in modulated superconductors[8] where the interaction between the flux lines falls off exponentially at large distances. In both cases the corresponding "natural" lattice, which would be formed in absence of the external potential, is hexagonal. The total energy contains the pair interaction between particles

$$W=\tfrac{1}{2}\sum_{i,j}W(|\vec{r}_j-\vec{r}_i|) \tag{2}$$

and the pinning potential energy, which we assume to be of the form

$$V=V_0-V_1\sum_i\cos(\vec{G}\cdot\vec{r}_i)\,, \tag{3}$$

where $\vec{G}=(2\pi/b)\hat{x}^1$ is the modulation vector. We start with the natural lattice of a given density $n=2/(\sqrt{3}a^2)$, and look for its accommodation to the pinning potential. The mth commensurability of the natural lattice is achieved when

$$m\vec{G}=k_1\vec{\tau}_1+k_2\vec{\tau}_2=\vec{\tau}(k_1,k_2)\,. \tag{4}$$

Here m, k_1, and k_2 are integers and $\vec{\tau}_1$ and $\vec{\tau}_2$ are the basic vectors of the reciprocal natural lattice $[\tau_1=\tau_2=4\pi/(\sqrt{3}a)]$. We choose $\vec{\tau}_1$, rotated from the modulation vector $\vec{G}$ through angle θ. The density of a commensurate configuration $(m|k_1,k_2)$ and its orientation are determined by the geometric condition (4) as follows:

$$n_c=n(m|k_1,k_2)=\frac{\sqrt{3}m^2}{2b^2(k_1^2+k_1k_2+k_2^2)}\,, \tag{5}$$

$$\theta_c=\theta_c(m|k_1,k_2)=\tan^{-1}\left(\frac{\sqrt{3}k_2}{2k_1+k_2}\right)\,. \tag{6}$$

The same classification will be used for the distorted lattices, which are commensurate in average.

In the vicinity of each first-order ($m=1$) commensurability of the natural lattice, i.e., when $\vec{\tau}\simeq\vec{G}$ so that the misfit between the two periodicities $p=|\vec{G}-\vec{\tau}|$ is smaller than the critical value p_c, a new first-order commensurate configuration is formed, with all the particles in the minima of the pinning potential. The adjacent I phases consist of large nearly commensurate domains, separated by solitonic walls.[9] With increasing p, but still in the long-wavelength regime ($p/G\ll 1$) the solitonic lattice becomes more dense and equivalent to the harmonically distorted natural lattice.[6] In the weak pinning limit such harmonic structure may persist as a ground state even in the short-wavelength regime ($p\sim G$). Thus, considering the I phases which are not

close to the principal commensurabilities, as well as the higher-order ($m \geq 2$) C phases, we can restrict ourselves to the stationary particle positions given by

$$\vec{r}_i = \vec{r}_i^{\,0} + \vec{v}_i + \vec{u}\sin[\vec{G}\cdot(\vec{r}_i^{\,0} + \vec{v}_i) + \Phi'] + \vec{r}_0 \ , \qquad (7)$$

where $\vec{r}_i^{\,0}$ are the particle positions in the natural lattice, rotated from G through angle θ (cf. Fig. 1). The position of the resulting configuration relative to the periodic potential (3) can be described by the phase variable $\Phi = \vec{G}\cdot\vec{r}_0$. In the I configurations ($n \neq n_c$, or $\theta \neq \theta_c$ for $n = n_c$) this phase is undetermined in the ground state, as in the analogous 1D problem (Frank–van der Merwe model), while in the C configurations this phase is pinned. When the natural lattice is incommensurate, the displacements $\vec{v}_i$ describe a small and unimportant homogeneous deformation, which is neglected in what follows. When $n \approx n_c$, one can consider the configuration to be incommensurate or allow instead for the displacements $\vec{v}_i$ which make the new structure commensurate:[8]

$$(x_i^1)^0 \rightarrow (x_i^1)^c, \quad (x_i^2)^0 \rightarrow (x_i^2)^c(1+\alpha) \ , \qquad (8)$$

where $(x_i^1)^c$ and $(x_i^2)^c$ are the projections of the commensurate natural lattice vectors on the directions parallel and perpendicular to the modulation axes, respectively. In all cases the suitably translated and oriented natural lattice is harmonically distorted. The parameters θ, α, $\vec{u}$, Φ and Φ' are determined by minimizing the resulting configuration energy. Since for a general C state the potential which pins the phase decreases as a power of m,[9] the high-order C phases become undistinguishable from the incommensurate ones. Thus we discuss the second- and third-order phases only. For the commensurate (in average) configurations with $\theta = \theta_c$ and n equal or close to n_c, the energy gain per unit area is given by

$$\Delta E_c = \Delta E + \Delta E' \ . \qquad (9)$$

The harmonic distortion-induced energy gain ΔE consists of two contributions: a negative one, due to the variation of the pinning energy, and a positive one, due to the increase of the interaction energy. As long as the linear one-harmonic approximation is valid, the first contribution is, in absolute value, twice as large as the second one. The additional increase $\Delta E'$ of elastic energy, related to the displacements $\vec{v}_i$ [Eq. (8)] is given by

$$\Delta E' = \frac{2\mu(\lambda+\mu)}{\lambda+2\mu}\left[1-\left(\frac{n_c}{n}\right)^{1/2}\right]^2 \ , \qquad (10)$$

where λ and μ are the Lamé coefficients of the natural lattice.

For small reduced amplitudes, $t = (\vec{G}\cdot\vec{u}) \ll 1$, ΔE can be approximated by

$$\Delta E = -\frac{nV_1^2}{4}(1+\delta_{m,2})(\vec{G}\hat{D}^{-1}\vec{G}) \ , \qquad (11)$$

where $\hat{D}^{-1}$ is the inverse of the dynamical matrix $\hat{D}$. The components of $\hat{D}$ are given by

$$D_{\alpha\beta} = \sum_i \frac{\partial^2 W}{\partial X_i^\alpha \partial X_i^\beta}[1-\cos(\vec{G}\cdot\vec{r}_i^{\,0})] \quad (\alpha,\beta = 1,2) \qquad (12)$$

in the coordinate system $X^1 - X^2$ of the lattice (cf. Fig. 1). Within the linear harmonic approximation the energy gain is quadratic in V_1, whereas the reduced amplitude t is linear:

$$t = V_1(\vec{G}\hat{D}^{-1}\vec{G}) \ . \qquad (13)$$

For the incommensurate configurations the energy gain is simply ΔE:

$$\Delta E_I = \Delta E \ . \qquad (14)$$

The orientation of the lattice, fixed geometrically for the commensurate configurations only, enters the calculation *via* Eq. (12), since $\vec{G} = (G\cos\theta, -G\sin\theta)$ in the coordinate system of the lattice. Notice also that the equilibrium translation, undetermined in the incommensurate phase, is for $m = 2$ different from that for $m = 3$, since $\Phi = \pi/2$ and $\Phi' = -\pi/2$ for $m = 2$, whereas $\Phi = -\pi/3$, $\Phi' = 2\pi/3$ for $m = 3$.

In the continuum approximation, the minimization of ΔE [Eq. (11)] gives

$$\cos\theta_{\min} = \frac{1+\tilde{\tau}^2(1+2\eta)}{\tilde{\tau}[2+\eta(1+\tilde{\tau}^2)]} \ , \qquad (15)$$

where $\tilde{\tau} = \tau/G$ and $\eta = (\lambda+\mu)/\mu$. The same expression for $\theta_{\min}$ was obtained in the elastic limit by McTague and Novaco[5] for a 2D substrate potential with a set of rotationally equivalent $\vec{G}$ vectors.

To investigate the short-wavelength regime of deformation one has to use instead a discrete approach. So we have evaluated numerically the optimum lattice oreintation $\theta_{\min}$ as a function of lattice density n. Whenever $n = n_c$, we have compared $\Delta E(\theta_{\min})$ with ΔE_c. It turns out that in many cases the commensurate configurations are unstable, i.e., $|\Delta E_c| < |\Delta E_I(\theta_{\min})|$. This happens whenever a better accommodation to the pinning potential can be achieved by a suitable orientation $\theta \neq \theta_c$. The results presented in Fig. 2 are valid, strictly speaking, in the weak pinning limit ($V_1 \rightarrow 0$) only; otherwise, new commensurate phases with the same orientation θ_c are formed in the vicinity of each stable C configuration. The width of the corresponding plateau of the $\theta_{\min}(n)$ curve can be calculated from Eqs.

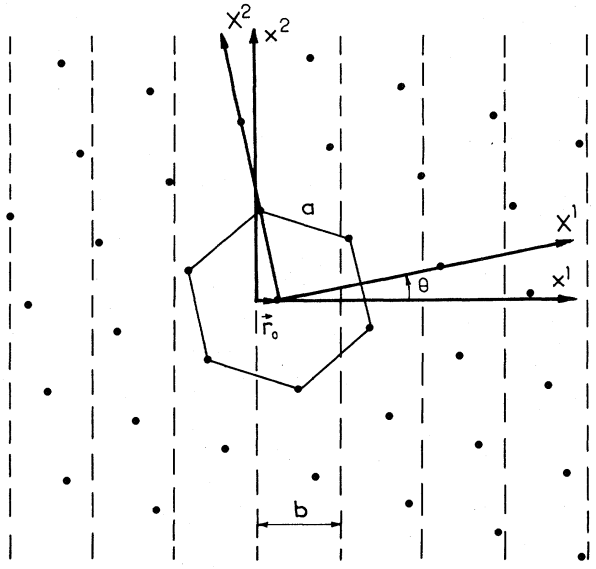

FIG. 1. Natural lattice of constant a in a potential of period b. Dashed lines represent the minima of the potential. The modulation vector $\vec{G}$ is oriented along the x^1 axis.

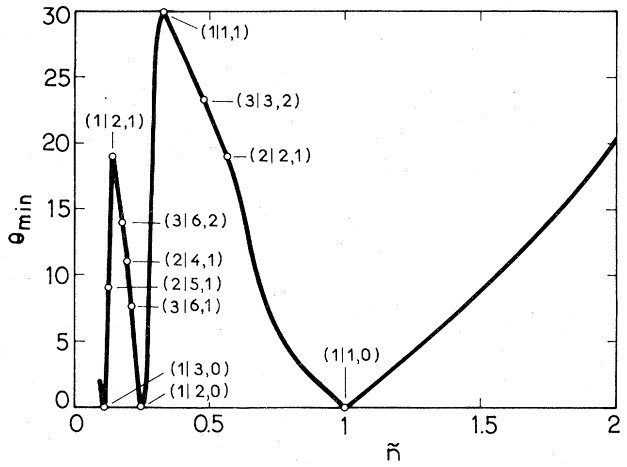

FIG. 2. Minimum energy orientation angle $\theta_{\min}$ as a function of the reduced density $\tilde{n}=n/n(1|1,0)=4b^2/3a^2$. The stable commensurate structures of the first, second, and third order are indicated.

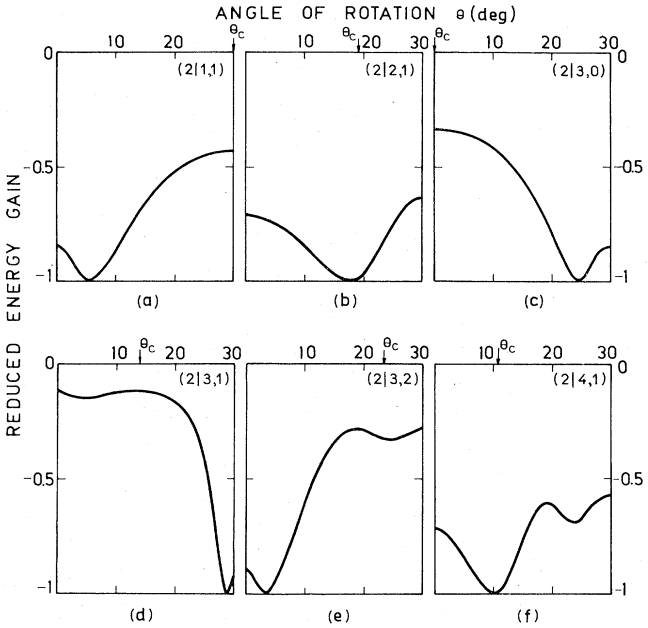

FIG. 3. Plots of the reduced energy gain $\Delta E/|\min\Delta E|$ vs orientation angle θ. Six representative values of the reduced density

$$\tilde{n}=\tilde{n}_c(2|k_1,k_2)=4/(k_1^2+k_1k_2+k_2^2)\ ,$$

such that second-order commensurability can be achieved at suitable $\theta=\theta_c$, are chosen. The lattice is at zero pressure. (b) and (f): stable commensurate phases; (a), (c), (d), and (e): unstable commensurate phases.

(9)–(11). We have not evaluated the widths of these regions which are proportional to $(V_1 t^{m-1})^{1/2}$ for $m \geq 3$. This can be shown when the nonlinear harmonic approximation is utilized.[8] For the 1D case the analogous result is given in Ref. 4. As long as the linear harmonic approximation is valid, the stability criterion is independent of the pinning potential amplitude, as can be seen from Eq. (11). There is a strong influence of the orientation of first-order phases on the form of the $\theta_{\min}(\tilde{n})$ curve as seen from Fig. 2. This influence is also manifested in the energy gain ΔE dependence on the orientation θ (Fig. 3). For some of the considered unstable C configurations the corresponding stable I structure is oriented in such a way that $\theta_{\min}$ is close to θ_c of the nearest first-order phase. Let us, for example, consider the configuration $(2|1,1)$ [see Fig. 3(a)], where $\theta_{\min}=5.6°$ is much closer to the neighboring first-order phase orientation $\theta_c(1|1,0)=0$ than the $\theta_c(2|1,1)=30°$. Sometimes, the influence of the two neighboring first-order phases is evident; an example of which is the structure $(2|3,1)$ [see Fig. 3(d)], where the small minimum reflects the vicinity of the $(1|2,0)$ phase, while the deep minimum shows the preponderant influence of the $(1|1,1)$ configurations. A significant example of a stable C configuration is $(2|2,1)$ [see Fig. 3(b)], where $\theta_{\min}$ for the I phase is equal to $\theta_c(2|2,1)=19.11°$.

Within the reduced density $[\tilde{n}=n/n(1|1,0)]$ range $0.1<\tilde{n}<2$, we find that out of 11 second-order geometrically possible C phases only 3 are stable, and out of 26 third-order configurations only 3 are stable also. An important result is that for $\tilde{n}\lesssim 2$ only those C structures for which θ_c falls on the $\theta_{\min}(\tilde{n})$ curve are stable, as seen in Fig. 2. With increasing density the orientational effects become less pronounced. The shape of the $\theta_{\min}(\tilde{n})$ curve is the same as that in the case of modulated superconductors,[8] i.e., it depends neither on the type of interactions nor on the lattice pressure.

The conclusion that there are only a few stable higher-order configurations confirms the conjecture made by McTague and Novaco[4] that "several reported high-order commensurate structures are likely to be incommensurate orientationally epitaxial systems." Neon adsorbed on graphite[10] could be an example of such a system. The reported density of monolayer coverage corresponds to the $\sqrt{7}\times\sqrt{7}$ structure with four atoms per elementary cell, i.e., to the $(2|2,1)$ configuration in our notation for which $\theta_c=19.11°$. This structure is predicted to be a stable one; however, the observed rotation with respect to the graphite axis is about 13°. Notice that the $\theta_{\min}(\tilde{n})$ curve is rather steep and that the pinning is weak here. Hence a possible explanation is that the actual density differs slightly from $\tilde{n}_c$, so that, in fact, an I structure with $\tilde{n}>\tilde{n}_c$ and of the corresponding optimal orientation is observed.

The above results are applicable also to superconductors whose structure is uniaxially modulated.[2,3] In this case, the density of the natural 2D lattice, formed by the centers of the flux lines, can be tuned continuously over a wide range by varying the external magnetic field. The observed[2,3] high and broad principal peaks of the critical current density manifest the presence of the first-order C phase, while only one second-order C phase is clearly seen. This is the $(2|1,0)$ structure occurring at $\tilde{n}=4$, which is, by our calculations, predicted to be a stable one. In the experiments with thin Al films of modulated thickness,[3] the $(2|2,1)$ structure, also predicted to be a stable one, is seen as a small irregularity on the critical current curve.

In conclusion, a generally rare occurrence of the higher-order C phases, in the case of weak pinning where the formation of some lower-order C phase is not a more favorable choice, can be explained by their orientational instability. We expect the influence of the relative lattice orientation to persist in the case of stronger pinning force as well. So, it is probable that the devil's staircase[11] always remains incomplete in the 2D (and even more so in the 3D) periodically modulated systems.

ACKNOWLEDGMENT

We would like to thank Dr. I. Božović for valuable discussions.

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