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Complete and incomplete wetting by adsorbed solids

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We carry out two calculations which show that a substrate which attracts a solid adsorbate too strongly prevents that adsorbate from wetting it. In the first, the adsorbed film is modeled microscopically as a set of layers in registry with one another. The appropriate thermodynamic potential is minimized at zero temperature with respect to the common lateral spacing and the individual layer heights for systems of up to 20 layers. For Xe, Kr, and Ar adsorbed on graphite, we find that the film thickness should exceed 20 layers, in agreement with experiment. A similar result is found for Ne, in disagreement with experiment. For atoms attracted very strongly, we find a thickness less than 20 layers, and therefore incomplete wetting behavior. In the second calculation, the film is modeled as an elastic continuum. We show that strain caused by the substrate potential induces a long-range force which, in general, prevents complete wetting of a solid film.

I. INTRODUCTION

There has been much interest recently[1,2] in determining the circumstances under which an adsorbate, at its bulk gas-solid coexistence, will wet a substrate (i.e., form a film of macroscopic thickness upon it). In almost all current descriptions, and most pertinently in lattice-gas models,[1] the question of whether the adsorbate will or will not wet is directly correlated with the strength of substrate-adsorbate interactions relative to adsorbate-adsorbate interactions. Strong substrates lead to wetting (sometimes denoted complete wetting), and weaker ones do not (incomplete wetting).

Recent experiments[3] to determine the wetting behavior of several adsorbates on a graphite substrate gave results which were unanticipated by these descriptions. While a change in behavior from incomplete to complete wetting was observed as the ratio of interaction strengths was increased as expected in such theories, further increase in strength brought a return from complete to incomplete wetting. In particular, while Ar, Kr, and Xe were observed to wet graphite at the low experimental temperatures, Ne, which by any reasonable measure feels a stronger substrate potential, did not. It was suggested[3] that this "reentrant" incomplete wetting must be due to compressional effects which are absent in the simplest lattice-gas calculations, those which only describe two phases. A lattice model[4] which included three phases was able to produce the effect.

In this paper we carry out two very different calculations of the adsorption of structureless atoms, such as the noble gases, on graphite. Both of them should be more accurate than those of simple lattice models. In Sec. II we calculate microscopically the zero-temperature energy $e(h)$ of an adsorbed film of thickness h of up to 20 layers. From this and a calculation of the chemical potential at the bulk gas-solid coexistence, the equilibrium value of the thickness there can be obtained. We do find that as we increase the ratio of the adsorbate-substrate potential to the adsorbate-adsorbate potential, the initial incomplete-wetting behavior changes to complete wetting and then back to incomplete.[5] Much microscopic information can be obtained from these calculations, such as the strain the adsorbate exhibits. Using the best potentials available for the noble gases, we find very good agreement with the Ar, Kr, and Xe data.

In Sec. III we treat the adsorbate film as an elastic continuum and apply linear elasticity theory. This yields the interesting result that a strain produces a long-range force that prevents the system from wetting. This effect also leads to the sequence of incompletely wet, wet, and incompletely wet behaviors as an increasing substrate potential first produces in the film a lateral expansion, then no strain, then a compression. We show that the elastic theory can be made quantitative if the asymptotic form of the potential and one bulk elastic constant are known as well as the actual strain in the first few layers of the film. The latter can be obtained from calculation or experiment. In particular for Ne, we find a finite equilibrium thickness but one which is much larger than obtained experimentally. However, we also find that this thickness is changing rapidly with interaction strength so that small changes in the potential, or the inclusion of quantum effects, could bring the result into agreement.

II. MICROSCOPIC MODEL CALCULATIONS

The determination of the zero-temperature wetting behavior of an adsorbed film proceeds in two stages. The first consists of calculating the chemical potential μ_0 of the bulk rare-gas solid at zero temperature and pressure. In the second, the energy per area $e(h)$ and areal density $n(h)$ of a model of a solid film of h layers adsorbed on the substrate is calculated. The equilibrium thickness of the film at coexistence is determined from the condition that the chemical potential de/dn be equal to μ_0. Equivalently, the minimum of the auxiliary function

$$\hat{\Omega}(h)=e(h)-\mu_0 n(h) \tag{2.1}$$

occurs at the equilibrium value. The substrate is wet if the minimum occurs for infinite values of h and is incompletely wet otherwise. Thus the wetting behavior is obvious from a plot of $\hat{\Omega}(h)$. The minimum value of this function is simply the grand potential $\Omega(\mu_0)$.

The calculation of the bulk chemical potential is straightforward. The interaction between rare-gas atoms is assumed to be pairwise additive, and a particular form of this interaction is chosen. We have used both the usual Lennard-Jones potential and the more accurate Aziz potential.[6] The bulk energy is obtained by performing the appropriate sums over the face-centered-cubic lattice, and the chemical potential follows from differentiation.

The calculation of the energy of the film is less simple, and we proceeded as follows. The interaction between the noble-gas and substrate atoms was taken to be of the Lennard-Jones form. We used the parameters of Cole and Klein[7] but ignored anisotropy terms. The effects of the substrate symmetry were first investigated by simulating the adsorption of particles on a structured substrate. Thus the substrate potential was obtained by performing the necessary sums over the three-dimensional graphite structure. Particles were added and the minimum of the energy per area obtained by an iterative procedure. Periodic boundary conditions were used to contain the atoms parallel to the substrate. The system could contain 16 atoms per layer at close packing. The calculation was repeated with a substrate potential which was averaged over the unit cell. There was little difference in the values of $e(h)$, as expected, and no difference in the behavior with h. From then on we employed the simpler structureless substrate potential.

To speed the process of energy minimization, we modeled the adsorbed film of h layers as one in which all layers were in registry with one another in an fcc stacking. The minimization parameters were then the one common lattice spacing in the lateral direction of all h layers and the h different heights above the substrate of the h layers. This model assumes that dislocations are energetically unlikely, as is a smooth variation of lateral lattice parameter with layer height. Both of these assumptions are justified in the appendices.

Our results for the calculation in which the adsorbate atoms interacted via an Aziz potential are shown in Fig. 1. The function $\hat{\Omega}(h)$ is plotted versus h for Xe, Kr, Ar, and Ne. Both quantities are measured in units of the appropriate adsorbate-atom interaction parameter. It can be seen that the function $\hat{\Omega}(h)$ is monotonically decreasing out to 20 layers. From these results for $h \leq 20$, it might appear that the minimum of this function is at infinite values of h, from which one would conclude that these noble gases wet graphite at zero temperature. Actually, as we will see in Sec. III, all of these functions have a minimum at a finite value of h so that, strictly, the substrate is incompletely wet. However, this minimum occurs for sufficiently large values of h that current experiment would be unable to detect the finite thickness of the film. The above results agree well with experiment[3] for the cases of Xe, Kr, and Ar. This agreement is not a trivial result in the case of Xe, for which the Xe-graphite interaction appears to be so weak compared with the Xe-Xe interaction that an incomplete wetting might be expected. In fact, if the cruder Lennard-Jones potential is used instead of the more accurate Aziz potential, one obtains the results of Fig. 2, which show that in such a model Xe clearly exhibits nonwetting behavior. Furthermore, as seen in the figure, one obtains the same result for Kr and Ar, and obtains wetting behavior only for Ne. Thus use of this potential gives results for each of the gases which disagree with experiment. In retrospect this is due to the overestimation of the strength of the potential tail by the Lennard-Jones potential. While the results of our calculation using the Aziz potential are in good agreement with experiment for Xe, Kr, and Ar, they are

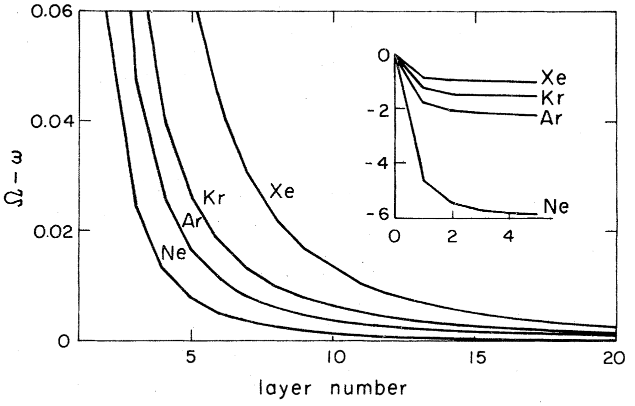

FIG. 1. The auxiliary function $\hat{\Omega}(h)$, in units of ϵ/σ^2, is shown vs layer number of noble gases adsorbed on graphite. In the lower portion of the figure, the vertical scale has been shifted by an amount ω different for each gas to facilitate comparison by eye. The atom-atom interaction is the Aziz potential.

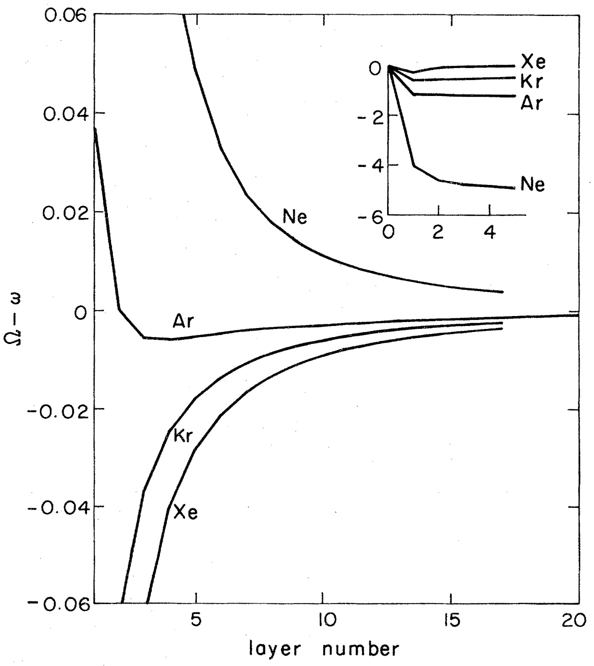

FIG. 2. Same as Fig. 1 except that the interaction is given by the Lennard-Jones potential.

in disagreement in the case of Ne. As we shall see below, the equilibrium thickness of the Ne film changes rapidly with changes in the interaction strength and it is possible that quantum effects, which would alter the strength of an effective classical potential, could be responsible for the observed incomplete-wetting behavior.

It is straightforward to extend our calculations to arbitrary structureless gases adsorbed on graphite. We show in Fig. 3(a) the results for the equilibrium thickness h versus the ratio of the integrated strengths of the atom-substrate and atom-atom potentials. As a measure of this ratio, we have used

$$R=(\epsilon_{S\text{-}A}/\epsilon_{A\text{-}A})(n_S\sigma^3_{S\text{-}A}/n_A\sigma^3_{A\text{-}A})(\sigma_{S\text{-}A}/\sigma_{A\text{-}A}) ,$$

where the densities of substrate and adsorbate have been denoted n_S and n_A, and the strength and range parameters of the potentials, taken from Ref. 7, are ϵ and σ, respectively. The subscripts identify the atom-atom or substrate-atom potential. The results show clearly the sequence of nonwetting, wetting, and nonwetting behaviors with increasing substrate strength. The ratio appropriate for Ne is 2.6 which, with Fig. 3, indicates that the Ne thickness is very sensitive to the potential strength. The abrupt jump in $\hat{h}$ seen for very weak potentials occurs when the value of $\hat\Omega(h)$ at the minimum exceeds $\hat\Omega(0)=0$. Then the equilibrium state corresponds to no adsorption whatsoever.

Another result of interest which can be extracted from these calculations is the lateral strain of the film relative to the bulk. This is shown in Fig. 3(b). Note that Xe, Kr, and Ar are dilated with respect to bulk, while Ne is compressed.

It is interesting to note that the lateral dilation or compression [Fig. 3(b)] is rather small. This being the case, one can ask how well a simple lattice-gas calculation, which ignores this difference between film and bulk, does in predicting the wetting behavior of the noble gases. The answer is that it does surprisingly well. In the lattice-gas model, the function $\hat\Omega(h)$ has the zero-temperature form

$$\hat\Omega(h)=\sum_{l=1}^{h}\phi(l) , \tag{2.2}$$

where $\phi(l)$ is the difference of the interactions between an adatom which is a distance l from a half-space of substrate and one which is the same distance from a half-space of bulk adsorbate. If this expression is evaluated by replacing sums by integrals (using the bulk for the film density) and employing the Aziz potential for the atom-atom interaction, then one finds that $\hat\Omega(h)$ is a decreasing function of h with its minimum at infinite h for the four noble gases considered. That is, the simple lattice-gas calculation gives the same result as the more elaborate one *for the cases of the noble-gases.* For cases in which the ratio of potential strengths is much larger than that encountered in adsorption of noble gases on graphite, the simple lattice-gas model is inadequate, since it cannot produce the reentrant incomplete wetting behavior which is obtained by the more complete calculation.

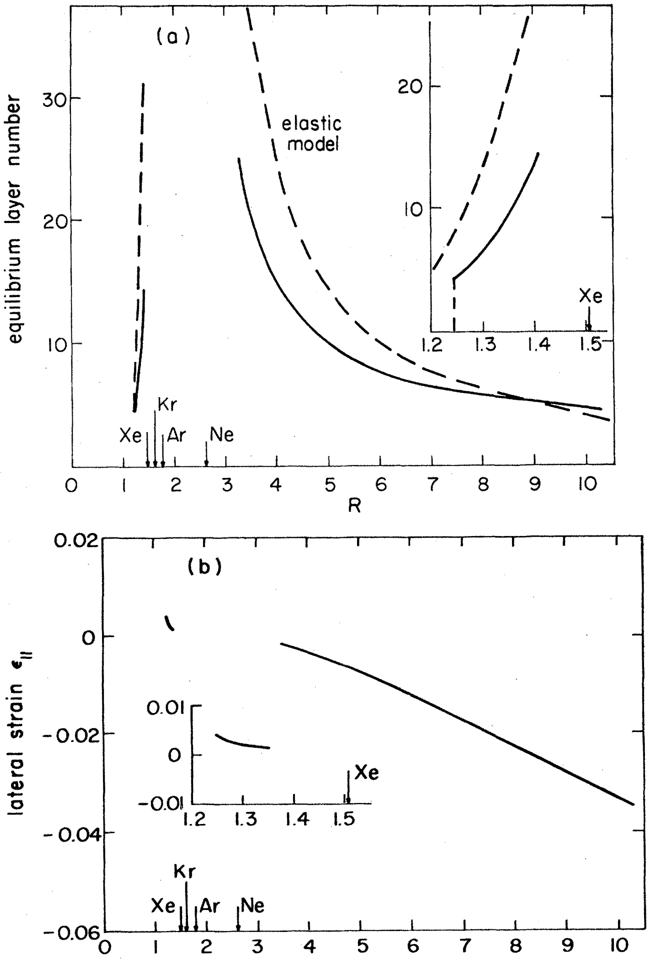

FIG. 3. (a) Equilibrium layer number of film adsorbed on graphite vs integrated potential strength. Solid lines are the interpolated results of microscopic calculation; dashed line is the result of elastic calculation. Layer number for the lattice is h in units of 0.9σ appropriate for fcc stacking. (b) Lateral strain of film adsorbed on graphite at equilibrium vs integrated potential strength.

III. LINEAR ELASTIC THEORY

In this section, the adsorbate is modeled by a continuous system of height h in the presence of a substrate. As in the preceding section, we take the vertical strain $\epsilon_\perp$ to be z dependent while the lateral strain $\epsilon_\parallel$ is constant. Since the film is considered to be of fcc structure with principal axes parallel and perpendicular to the substrate, the stress and strain tensors are diagonal. The auxiliary function $\hat\Omega(\epsilon_\perp,\epsilon_\parallel,h)$ is

$$\hat\Omega(\epsilon_\perp,\epsilon_\parallel,h)=\frac{1}{1+2\epsilon_\parallel}\int_0^h dz\left[\mu(\epsilon_\perp^2+2\epsilon_\parallel^2)+\frac{\lambda}{2}(\epsilon_\perp+2\epsilon_\parallel)^2 + n_A\frac{d\phi}{dz}\int_0^z\epsilon_\perp(z')dz' + n_A\phi(z)\right] , \tag{3.1}$$

where μ and λ are Lamé constants and n_A is the density of the unstrained film. The potential $\phi(z)$ is the continuum generalization of the lattice-gas potential of Eq. (2.2). The conditions of equilibrium, that the body forces balance the external forces, yields the equation

$$n_A \frac{d\phi}{dz} - (2\mu+\lambda)\frac{d\epsilon_\perp}{dz} = 0 \,,$$

which determines the z dependence of the vertical strain according to

$$(2\mu+\lambda)\epsilon_\perp(z) - n_A \phi(z) = \text{const} \,.$$

The boundary condition of zero pressure at $z=h$ determines the constant so that

$$(2\mu+\lambda)\epsilon_\perp(z) = n_A[\phi(z)-\phi(h)] - 2\lambda\epsilon_{||} \,.$$

This form of the vertical strain is substituted into Eq. (3.1) to yield $\hat{\Omega}(\epsilon_{||},h)$. The dependence of this function on h and $\epsilon_{||}$ for small $\epsilon_{||}$ and $1/h$ is

$$\hat{\Omega}(\epsilon_{||},h) = \hat{\Omega}_0 + B(2\epsilon_{||}S + \epsilon_{||}^2 h) + B\left[\frac{C\bar{n}_A}{2Bh^2} - 4\epsilon_{||}^2 S - 2\epsilon_{||}^3 h\right] ,$$

where

$$\hat{\Omega}_0 = n_A \int_0^\infty \phi\, dz - \frac{n_A^2}{2(2\mu+\lambda)} \int_0^\infty \phi^2 dz \,,$$

$$S = \frac{-1}{2\mu+3\lambda}\left[n_A \int_0^\infty \phi\, dz - \frac{n_A^2}{4\mu}\int_0^\infty \phi^2 dz\right] , \qquad (3.2)$$

$$B = 2\mu(2\mu+3\lambda)/(2\mu+\lambda) \,,$$

and $\phi(z) = -C/z^3$ for large z.

Minimization of $\hat{\Omega}(\epsilon_{||},h)$ with respect to $\epsilon_{||}$ for a given h determines the lateral strain $\epsilon_{||}(h)$. For h large, we obtain

$$\epsilon_{||}(h) = -S/h - (S/h)^2 \,. \qquad (3.3)$$

Note that the lateral strain is negative corresponding to compression for effective potentials ϕ whose integrated strength is negative (strong substrates), and is positive corresponding to expansion for effective potentials whose integrated strength is positive (intermediate substrates). Substitution of $\epsilon_{||}(h)$ into $\hat{\Omega}(\epsilon_{||},h)$ yields the auxiliary function $\hat{\Omega}(h)$. At large h this function has the form

$$\hat{\Omega}(h) = \hat{\Omega}_0 - \frac{BS^2}{h} + \frac{B}{h^2}\left[\frac{C}{2B} - 2S^3\right] . \qquad (3.4)$$

The asymptotic value of $\hat{\Omega}(h)$ is approached from *below* so that the minimum of this function is not in general at infinite values of film thickness; that is, the solid film does not wet the substrate.[8] Another way to state this result is that the strain induced by the potential produces a long-range attraction between the film-vapor and film-substrate interfaces which decays as $1/h$. Such an attraction is always asymptotically stronger than the van der Waals and thermal forces, which favor wetting.[2]

The equilibrium value of the thickness for large thickness is

$$h = \frac{C}{BS^2} - 4S \,. \qquad (3.5)$$

This result shows that reentrant incomplete-wetting behavior is obtained for effective potentials ϕ, which are asymptotically attractive ($C>0$), but whose integrated strength can be varied from positive to negative. In such a case, the film is initially expanded, and the strain causes incomplete wetting. As the substrate potential is made more attractive, the strain becomes smaller and will vanish for some potential. The adsorbate now wets the substrate. As the potential is made even stronger, the film becomes compressed, and once again the strain causes the wetting to be incomplete.

We now ask whether this linear elastic theory can be made quantitative. One cannot simply substitute real potentials into the equations because the integrals, which arise from treating the film as a continuum, are too sensitive to the lower limit of integration. However, a reasonable estimate can be obtained from Eq. (3.5) which requires only the three numbers C, B, and S. The first is easily obtained because the effective potential $\phi(z)$ is the difference of the interactions between an adatom a distance z from a half-space of substrate and of bulk adsorbate. As both of these potentials fall off as z^{-3} with known amplitudes, the constant C is known. The parameter B, as given in Eq. (3.2), is simply a combination of Lamé coefficients of the bulk adsorbate. This expression is simplified considerably using the fact that for simple structureless adsorbates which form a fcc lattice, such as the noble gases, the Lamé coefficients are related by[9] $\mu \simeq 3/2\lambda$, so that $B \simeq 3\mu$. Lastly the value of S, which can be shown to be proportional to the stress in the system, can be obtained from Eq. (3.3) if the strain is known for a few different thicknesses. We have evaluated S from our microscopic calculations of Sec. II. We found that it obeys Eq. (3.3) down to a very few layers and scales linearly with the ratio of the integrated substrate-atom to atom-atom potential:

$$S = 0.0229[R - 1.88] \,.$$

Note that this dependence on R implies that the equilibrium height h depends on two parameters; the asymptotic potential strength C and the integrated strength R. This contrasts with the lattice-gas model in which the height depends only on one parameter. We have taken $|C|/R$ to be that of neon for hypothetical noble gases with $R_{\text{Ne}} < R$, and to be that of xenon for hypothetical gases with $R_{\text{Xe}} > R$. Lastly, B was obtained by calculating the Lamé constants in the fcc crystal in which atoms interacted with the Aziz potential. Results of this calculation are shown as dotted lines in Fig. 3(a). The film thickness h is plotted versus the ratio of integrated potential strengths, R. As can be seen, the agreement is fair. It should be noted from the qualitative agreement alone that the equilibrium heights obtained in the microscopic calculation are limited by the stress for both the cases of expanded and compressed films.

To summarize, the actual stress and the known long-distance behavior of the potentials are sufficient to obtain a good estimate of h from Eq. (3.5). Further, the needed stress S can be obtained from calculation or experiment on a system of *only a few layers.*

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APPENDIX

In the models above, we ignored the effects of lateral relaxation of finite crystallites, a relaxation which results in the variation of the horizontal lattice constant with vertical coordinate z. We also ignored the appearance of dislocations which also permit such a variation. We now consider these two possibilities and show that is reasonable to ignore them.

1. Relaxation of finite crystallites

For a domain in which the ratio of its radius r_0 to height h is very large, one expects that all layers undergo a common horizontal expansion or compression. If this ratio is of order unity or less, however, the shape of the domain might well distort appreciably in the presence of the substrate potential. In order to quantify the distinction between these two types of elastic response, we employ the model of a cylindrical crystallite described by isotropic elastic constants μ and λ. The equation of equilibrium is

$$\mu\nabla^2\vec{u}+(\mu+\lambda)\nabla(\nabla\cdot\vec{u})=\nabla\phi_{\mathrm{ex}}\ , \tag{A1}$$

where u is the displacement and ϕ_{ex} is the external potential, which is assumed to fall off rapidly with height. We take the effect of this potential to be negligible inside the body, but will retain an arbitrary in-plane deformation of the bottom face of the cylinder of the crystallite as a boundary condition in order to represent the lateral strain it induces.

To find a solution to this equation, we proceed as in Ref. 10. The neglect of the potential inside the body implies that $u_z=0$. The radial component of the displacement is obtained from the radial part of Eq. (A1) which, in cylindrical coordinates, becomes Bessel's equation of order of 1 with solutions

$$u_l(r,z)=J_1(x_l r/r_0)\cosh[\gamma x_l(h-z)/r_0]\ ,$$

where $\gamma^2=(2\mu+\lambda)/\mu=\frac{8}{3}$ for $\mu=3/2\lambda$, and where the condition of zero radial stress at the walls of the cylinder determines the x_l. An arbitrary displacement $u(r,0)$ imposed as a boundary condition at the bottom face, $z=0$, can be expanded in this set of functions;

$$u(r,0)=\sum_l a_l J_1(x_l r/r_0)\cosh(\gamma x_l h/r_0)\ .$$

The lth term contributes a deformation of the top face relative to the bottom face of

$$u_l(r,h)/u_l(r,0)=1/\cosh(\gamma x_l h/r_0)\ .$$

By definition, the limit of thin cylinders occurs when this ratio is near unity. Provided that the major contribution to $u(r,0)$ comes from small l, this limit is obtained for $R/h \gtrsim x_l\gamma$, which should certainly be well below 100. For example, in the case of a uniform dilation of the bottom face, the relative contribution of the terms in $u(r,0)$ are $a_l=1,-0.05,0.03,0.08,0.01,\ldots,$ with higher terms decreasing faster than exponentially. In this case an aspect ratio $h/r_0=0.1$ is already in the limit of thin cylinders. The films examined in experiment with domain sizes on the order of hundreds of lattice spacings or more should be well described as thin in the above sense for thicknesses up to 20 layers at least.

2. Dislocations

We now estimate the energy associated with the creation of a dislocation, since such excitations might be created to relieve the lateral strain. We consider only nearest-neighbor interactions and describe the registry of a first layer against a second by locating the atoms of the first layer at a sublattice of sites on a honeycomb lattice. A reasonable model for an isotropic compression of the first layer relative to the second is a triangular network of walls across which the sublattice occupations are changed. Note that this triangular network of walls arising from atoms on honeycomb sites is completely analogous to the honeycomb array of walls arising from atoms on triangular sites.[11] For a network of dislocation segments of length S, the change in area, $\delta A/A$, is approximately $2\epsilon_{\parallel}=r_{\mathrm{NN}}/S$, for $S\gg r_{\mathrm{NN}}$, where r_{NN} is the nearest-neighbor distance. The atoms within the domain wall are displaced by a distance of order $w\simeq r_{\mathrm{NN}}/\sqrt{3}$. We approximate the total energy of this wall as

$$\int dx[A\sin^2(\pi x/w)+B(dy/dx)^2]\ ,$$

where x is the position along the row of atoms and y is the displacement perpendicular to the row. The first term represents the contribution due to the corrugation of the adjacent layer. The amplitude $A\simeq0.5\epsilon_{AA}$ if only nearest-neighbor interactions are considered. The second term is essentially the shear energy of the first layer. Calculating the shear energy of a uniformly sheared monolayer, we find $B\simeq20\epsilon_{AA}$. The solution minimizing the energy subject to the boundary condition $y(-\infty)=0, y(\infty)=w$, is

$$\cos(\pi y/w)=-\tanh[(A/B)^{1/2}\pi x/w]\ ,$$

which has a width $\Delta x\simeq4(B/A)^{1/2}\omega/\pi\simeq5r_{\mathrm{NN}}$, and a dislocation energy of approximately $0.6\epsilon_{AA}/r_{\mathrm{NN}}$ per unit length of dislocation.

For the triangular network of dislocations, we arrive at a rough estimate of the core energy per unit area necessary for a compression $\delta A/A$ of $(2\epsilon_{AA}/r_{\mathrm{NN}}^2)\delta A/A$. In the numerical calculations of Sec. II the changes in the lateral strain and in the energy from layer to layer were so small as to exclude the possibility of dislocations such as these.

[1] A guide to the literature up to 1982 can be found in R. Pandit, M. Schick, and M. Wortis, Phys. Rev. B **26**, 5112 (1982). More recent literature is cited in Ref. 2.

[2] M. P. Nightingale, W. F. Saam, and M. Schick, Phys. Rev. Lett. **51**, 1275 (1983).

[3] J. L. Seguin, J. Suzanne, M. Bienfait, J. G. Dash, and J. A. Venables, Phys. Rev. Lett. **51**, 122 (1983); M. Bienfait, J. L. Seguin, J. Suzanne, E. Lerner, J. Krim, and J. G. Dash, Phys. Rev. B **29**, 983 (1984).

[4] C. Ebner, C. Rottman, and M. Wortis, Phys. Rev. B **28**, 4186 (1983).

[5] Calculations similar in spirit but differing in detail have been carried out by R. J. Muirhead, J. G. Dash, and J. Krim, Phys. Rev. B **29**, 5074 (1984).

[6] R. A. Aziz, Mol. Phys. **38**, 177 (1979). The potential in this paper is for Kr. We have obtained it for the other noble gases by scaling up the strength and range parameters which occur by the ratios of the analagous Lennard-Jones parameters which are given in Ref. 7.

[7] M. W. Cole and J. R. Klein, Surf. Sci. **124**, 547 (1983).

[8] This result has also been obtained by D. A. Huse (unpublished).

[9] M. L. Klein and T. R. Koehler, in *Rare Gas Solids,* edited by M. L. Klein and J. A. Venables (Academic, London, 1976), Vol. 1.

[10] J. G. Dash, J. Suzanne, H. Shecter, and R. E. Peierls, Surf. Sci. **60**, 411 (1976).

[11] J. Villain, in *Ordering in Strongly Fluctuating Condensed Matter Systems,* edited by T. Riste (Plenum, New York, 1980).