Quasi-two-dimensional imperfect Bose gas adsorbed on a surface

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We study a submonolayer film adsorbed on a substrate surface as a quasi-two-dimensional (2D) imperfect Bose gas. The ground-state energy and the depletion fraction at zero temperature are calculated. It is found that the depletion is *independent* of the 2D density (in contrast to the 3D case), but is a monotonically increasing function of the substrate potential well depth. Qualitative conclusions are drawn about implications for superfluidity in the case of He on a weak-binding substrate.

I. INTRODUCTION

The subject of superfluidity in ^{4}He films on flat substrates has a long history of surprising discoveries. An intriguing question is the relation between the superfluid properties and the phenomenon of Bose-Einstein condensation (BEC).[1,2] In bulk liquid ^{4}He, we believe that the loss of superfluidity at the λ transition coincides with the disappearance of the condensate.[3] The same is *not* true for a film. While BEC is not possible at finite temperature T in a film of any finite thickness,[4] superfluidity does occur experimentally. This apparently contradictory situation is resolved[5–7] by allowing the film to possess a local condensate wave function which exhibits topological long-range order, but not truly infinite long-range order (equivalent to BEC). An open question is the "practical" matter of knowing theoretical criteria for existence of this superfluid state. Consider, for example, a Bose fluid with such strong interactions that the condensate disappears even at $T=0$. We do not know whether it will be a superfluid at finite T.

This paper addresses the properties of a film of Bose particles adsorbed on a surface. We determine the ground-state properties as a function of the interactions involved: He-He and He-surface. The former is idealized, for simplicity, but we believe the qualitative calculations to be generally accurate.

This paper is a sequel to a previous study,[8] in which the effects of substrate variation was found on the complete monolayer's properties. The principle conclusion was that certain weak-binding substrates have insufficient attraction to nucleate a solid, so that the monolayer phase is liquid. That study assumes the in-plane motion of the He atoms to be strictly two-dimensional (2D). The present paper directs attention to the fact that the monolayer is only quasi-2D, i.e., that the states are diffuse in the direction z perpendicular to the surface. A consequence is that the effective interaction between adatoms weakens as the width w of the single-particle states grows; as a result, the fraction f_0 of particles in the condensate grows:

$$f_0 \equiv N_0/N \ .$$

Here N_0 is the number of particles with 2D momentum equal to zero which lies in the lowest state $i=0$ of motion perpendicular to the surface. The result, shown below, is that the film becomes more ideal for a weak-binding surface than for a strong binding one. The attendant increase of f_0 with increasing w is explored herein. This predicted behavior ought to be observable experimentally. Indeed recent experiments have revealed novel behavior in the properties of He on weak-binding surfaces; these have prompted the present study.[9,10]

There exists a considerable literature of the 2D Bose problem. This includes study of the case of hard-disk and Lennard-Jones interactions and the role of heterogeneity.[11–13] To our knowledge there has been no explicit assessment of the effects on the condensate properties[14] of the out-of-plane motion. In this paper, we use a simplified model to study the quasi-2D imperfect gas, incorporating explicitly the influence of the substrate potential.

II. MODEL CALCULATION

We assume that the substrate is a structureless continuum occupying the region $z<0$. The He physisorption potential can be described approximately in the analytical form of a shifted-Morse (SM) potential. This has proved to be a good approximation to the real potential near the minimum.[15,16] Since we are concerned here with only the low-lying binding states, it is adequate here. The potential has the following form:

$$V_{\rm SM}(z)=D(e^{-2\alpha(z-z_0)}-2e^{-\alpha(z-z_0)}-\Delta) \ . \tag{1}$$

Its eigenvalues and eigenstates are:[16]

$$E_i^{\mathrm{SM}}=-D\left[\Delta+\left[1-\frac{2i+1}{b}\right]^2\right],\quad i=0,1,2\ldots, \tag{2}$$

$$\varphi_0=\left[\frac{\alpha}{\Gamma(b-1)}\right]^{1/2} y^{(b-1)/2}e^{-y/2},$$

$$\varphi_1=(b-3)^{1/2}\left[1-\frac{b-2}{y}\right]\varphi_0, \tag{3}$$

$$b=\frac{(8mD)^{1/2}}{\hbar\alpha},\quad y=be^{-\alpha(z-z_0)}, \tag{4}$$

where $D(1+\Delta)$ is the well depth, α is a parameter related to the width of the potential well and z_0 is the position of the potential minimum. In the case of a deep potential well (large D), the low-lying states of the SM potential will be similar to those of a simple harmonic (SH) one. It is much easier to treat the SH potential than the SM one. We present results for it in the Appendix.

The total Hamiltonian of the system can then be written:

$$H=\sum_{i=1}^{N}\frac{\mathbf{P}_i^2}{2m}+\sum_{i<j}^{N}u(|\mathbf{r}_i-\mathbf{r}_j|)+\sum_{i=1}^{N}V_{\mathrm{SM}}(z_i), \tag{5}$$

where N is the total particle number and $u(|\mathbf{r}_i-\mathbf{r}_j|)$ is the adatom-adatom interaction potential. Here we have omitted many-body potential-energy terms, which are generally small for He films.[17] We use the formalism of second quantization to study this system. We introduce single-particle basis states which are solutions of the noninteracting problem:

$$|\beta i\rangle=|\beta\rangle|i\rangle, \tag{6}$$

where $|\beta\rangle$ is the 2D free particle state with a 2D momentum $\mathbf{P}_\beta$:

$$|\beta\rangle=\frac{1}{A^{1/2}}\exp(i\mathbf{P}_\beta\cdot\mathbf{R}/\hbar). \tag{7}$$

Here $\mathbf{R}$ is the 2D (x-y) position vector and A is the area of the surface. The binding state $|i\rangle$ in Eq. (6) is one of the eigenstates of the SM potential. Now we can rewrite the Hamiltonian in Eq. (5) in the second quantized form:[18]

$$H=\sum_{\alpha i}\left[\frac{\mathbf{P}_\alpha^2}{2m}+E_i\right]a_{\alpha i}^\dagger a_{\alpha i}+\tfrac{1}{2}\sum_{\alpha\beta\gamma\lambda}\sum_{ijkl}\langle\alpha i\beta j|u|\gamma k\lambda l\rangle a_{\alpha i}^\dagger a_{\beta j}^\dagger a_{\gamma k}a_{\lambda 1}, \tag{8}$$

where $a_{\alpha i}^\dagger$ and $a_{\alpha i}$ are the creation and annihilation operators of the state $|\alpha i\rangle$, respectively. The major task here is the calculation of the adatom-adatom interaction potential matrix elements. The complicated shape of the realistic two-body interaction potential makes this calculation nontrivial. In order to simplify the problem, we make the crude approximation of a contact interaction, i.e.,

$$u(|\mathbf{r}_i-\mathbf{r}_j|)=u_0\delta^3(\mathbf{r}_i-\mathbf{r}_j). \tag{9}$$

The parameter u_0 provides a measure of the strength and can be determined from the atom-atom scattering length, as described in the following. An identical model is used in the Bogoliubov theory of 3D Bose systems.[19]

Another difficulty is the complicated form of the eigenstates of the SM potential. It is impossible to give a simple analytic result for all of the binding states which can be involved. Fortunately, however, only very few low-lying states arise in the scattering at low temperature and small u_0. Thus we make another approximation: that only the contribution from the ground state φ_0 and the first excited state φ_1 are important; these two wave functions are given in Eq. (3). This approximation should be a good one at very low temperature since the adatoms are unlikely to be excited into higher excited states by thermal excitation or scattering.

Under these approximations, the matrix elements can be calculated as

$$\langle\alpha i\beta j|u|\gamma k\lambda l\rangle=\frac{4\pi^2u_0}{A^2}I_{ijkl}\delta^2(\mathbf{K}/\hbar) \tag{10}$$

with $\mathbf{K}=\mathbf{P}_\alpha+\mathbf{P}_\beta-\mathbf{P}_\gamma-\mathbf{P}_\lambda$ and

$$I_{ijkl}\equiv\int_{-\infty}^{\infty}dz\varphi_i\varphi_j\varphi_k\varphi_l, \tag{11}$$

$$I_{0000}=\frac{\alpha}{2^{2b-2}}\frac{\Gamma(2b-2)}{\Gamma^2(b-1)}, \tag{12a}$$

$$I_{0001}=\frac{\alpha}{2^{2b-3}}\frac{\Gamma(2b-3)}{\Gamma^2(b-1)}(b-3)^{1/2}, \tag{12b}$$

$$I_{0011}=\frac{\alpha}{2^{2b-3}}\frac{\Gamma(2b-4)}{\Gamma^2(b-1)}(b-3)(b-2), \tag{12c}$$

$$I_{0111}=-\frac{\alpha}{2^{2b-3}}\frac{\Gamma(2b-5)}{\Gamma^2(b-1)}(b-3)^{3/2}(b-2), \tag{12d}$$

$$I_{1111}=\frac{\alpha}{2^{2b-4}}\frac{\Gamma(2b-5)}{\Gamma^2(b-1)}(b-3)^2(b-2)(3b-5). \tag{12e}$$

The (approximate) proportionality of these matrix elements to α, the potential's inverse width parameter, reflects the fact that broad potentials yield broad wave functions and small matrix elements. The Hamiltonian has now become:

$$H=\sum_{Pi}\left[\frac{\mathbf{P}^2}{2m}+E_i^{\mathrm{SM}}\right]a_{Pi}^\dagger a_{Pi}+\frac{u_0}{2A}\sum_{\substack{\mathbf{P}_1,\mathbf{P}_2\\ \mathbf{P}_1',\mathbf{P}_2'}}'\Big[I_{0000}a_{P_1'0}^\dagger a_{P_2'0}^\dagger a_{P_10}a_{P_20}+I_{0001}(a_{P_1'0}^\dagger a_{P_2'1}^\dagger a_{P_10}a_{P_20}+a_{P_1'0}^\dagger a_{P_2'0}^\dagger a_{P_11}a_{P_20})$$
$$+I_{0011}(4a_{P_1'0}^\dagger a_{P_2'1}^\dagger a_{P_10}a_{P_21}+a_{P_1'1}^\dagger a_{P_2'1}^\dagger a_{P_10}a_{P_20}+a_{P_1'0}^\dagger a_{P_2'0}^\dagger a_{P_11}a_{P_21})$$
$$+I_{0111}(a_{P_1'1}^\dagger a_{P_2'1}^\dagger a_{P_11}a_{P_20}+a_{P_1'0}^\dagger a_{P_2'1}^\dagger a_{P_11}a_{P_21})+I_{1111}a_{P_1'1}^\dagger a_{P_2'1}^\dagger a_{P_11}a_{P_21}\Big]. \tag{13}$$

The primed summation goes over those values of the momenta that conserve the total 2D momentum of the particles: $\mathbf{P}_1+\mathbf{P}_2=\mathbf{P}_1'+\mathbf{P}_2'$.

It is natural to expect that, in the ground state of a slightly imperfect gas, the occupation number $n_{\mathbf{P}}$ for $P\neq 0$ would be small compared to the number n_0, which would be close to the total number N. This enables us to use the method of Bogoliubov.[19] To lowest order, we have:

$$a_{00}^{\dagger}a_{00}=n_0\approx N \tag{14}$$

and

$$a_{00}a_{00}^{\dagger}\approx a_{00}^{\dagger}a_{00} .$$

All the operators in Eq. (13) which involve states other than $|00\rangle$ will give higher order contributions. The lowest-order result for the ground-state energy of the system can be calculated to be:

$$E_0\approx N\left[E_0^{\mathrm{SM}}+\frac{Nu_0}{2A}I_{0000}\right] . \tag{15}$$

Now we calculate the next order of approximation. To simplify the calculation, we omit terms involving interlayer scattering operators: $a_{P1}^{\dagger}a_{P'0}$ and $a_{P0}^{\dagger}a_{P'1}$. Since the probability of interlayer scattering is very low, the effect of this omission should be negligible. Furthermore, a correction to Eq. (14) from the lowest-order term $a_{00}^{\dagger}a_{00}^{\dagger}a_{00}a_{00}$ should be included:[18]

$$a_{00}^{\dagger}a_{00}^{\dagger}a_{00}a_{00}\approx N^2-2Na_{01}^{\dagger}a_{01}-2N\sum_{P\neq 0}(a_{P0}^{\dagger}a_{P0}+a_{P1}^{\dagger}a_{P1}) \tag{16}$$

Other contributions come from $\mathbf{P}\neq 0$ terms. Finally we have [see Eq. (11.2.9) in Ref. 18]:

$$H=E_0+\varepsilon_{01}a_{01}^{\dagger}a_{01}+\sum_{P\neq 0}[K_0(P)a_{P0}^{\dagger}a_{P0}+S_0(a_{P0}^{\dagger}a_{-P0}^{\dagger}+a_{P0}a_{-P0}+2a_{P0}^{\dagger}a_{P0})]$$
$$+\sum_{P\neq 0}[K_1(P)a_{P1}^{\dagger}a_{P1}+S_1(a_{P1}^{\dagger}a_{-P1}^{\dagger}+a_{P1}a_{-P1}+2S_{01}a_{P1}^{\dagger}a_{P1})] , \tag{17}$$

where E_0 is defined in Eq. (15) and

$$\Delta E^{\mathrm{SM}}\equiv E_1^{\mathrm{SM}}-E_0^{\mathrm{SM}}=4D(b-2)/b^2 , \tag{18}$$

$$\varepsilon_{01}=\Delta E^{\mathrm{SM}}+2S_1S_{01} , \tag{19}$$

$$K_0(P)=\frac{P^2}{2m},\quad K_1(P)=\frac{P^2}{2m}+\Delta E^{\mathrm{SM}} , \tag{20}$$

$$S_0=\frac{u_0N}{2A}I_{0000},\quad S_1=\frac{u_0N}{2A}I_{0011},\quad S_{01}=2-\frac{I_{0000}}{I_{0011}} . \tag{21}$$

In order to diagonalize the Hamiltonian in Eq. (17), we apply the Bogoliubov linear transformation to both the $|P0\rangle$ and $|P1\rangle$ states:

$$b_{Pi}=\frac{a_{Pi}+\gamma_{Pi}a_{-Pi}^{\dagger}}{(1-\gamma_{Pi}^2)^{1/2}},\quad b_{Pi}^{\dagger}=\frac{a_{Pi}^{\dagger}+\gamma_{Pi}a_{-Pi}}{(1-\gamma_{Pi}^2)^{1/2}} , \tag{22}$$

where $i=0$ and 1 and:

$$\gamma_{P0}=[K_0(P)+2S_0-\varepsilon_0(P)]/2S_0 , \tag{23}$$

$$\varepsilon_0(P)=[K_0^2(P)+4S_0K_0(P)]^{1/2} , \tag{24}$$

$$\gamma_{P1}=[K_1(P)+2S_1S_{01}-\varepsilon_1(P)]/2S_1 , \tag{25}$$

$$\varepsilon_1(P)=\{[K_1(P)+2S_1S_{01}]^2-4S_1^2\}^{1/2} . \tag{26}$$

The diagonalized Hamiltonian now becomes

$$H=E_0+\varepsilon_{01}a_{01}^{\dagger}a_{01}+\sum_{P\neq 0}[\varepsilon_0(P)b_{P0}^{\dagger}b_{P0}+\varepsilon_1(P)b_{P1}^{\dagger}b_{P1}] . \tag{27}$$

Eq. (27) is just the ground-state energy

$$E_0=N(E_0^{\mathrm{SM}}+S_0)+\sum_{P\neq 0}\left[\frac{\gamma_{P0}^2}{1-\gamma_{P0}^2}K_0(P)-\frac{2\gamma_{P0}}{1+\gamma_{P0}}S_0\right]$$
$$+\sum_{P\neq 0}\left[\frac{\gamma_{P1}^2}{1-\gamma_{P1}^2}K_1(P)-\frac{2\gamma_{P1}(1-\gamma_{P1}S_{01})}{1-\gamma_{P1}^2}S_1\right] \tag{28}$$

plus the energy of a gas of independent quasiparticles. We calculate the depletion number N_d, i.e., the number of particles which are not in the single-particle ground state. Since the occupation number of any single excited state is very small compared to that of the ground state ($\sim N$), we can omit the contribution from $|01\rangle$ state. The summation over all of the excited states with nonzero lateral momentum $\mathbf{P}$, however, will give a finite result. We define here N_{d0} and N_{d1} as the total numbers of particles in the surface-normal states $|0\rangle$ and $|1\rangle$ having $P\neq 0$, respectively. These numbers can be calculated as follows:[18]

$$N_{d0}=\sum_{P\neq 0}\frac{\gamma_{P0}^2}{1-\gamma_{P0}^2}\approx\frac{A}{h^2}\int d^2\mathbf{P}\frac{\gamma_{P0}^2}{1-\gamma_{P0}^2}$$
$$=\frac{mA}{2\pi\hbar^2}S_0=N\frac{\alpha a}{2\pi^{1/2}}\frac{\Gamma(b-1/2)}{\Gamma(b-1)} , \tag{29}$$

$$N_{d1}\approx\frac{A}{h^2}\int d^2\mathbf{P}\frac{\gamma_{P1}^2}{1-\gamma_{P1}^2}=\frac{2}{(x_0^2-4)^{1/2}+1}\frac{mA}{2\pi\hbar^2}S_1 , \tag{30}$$

$$x_0=\Delta E^{\mathrm{SM}}/S_1+2S_{01} . \tag{31}$$

Here we have introduced a (3D) scattering length a:[18]

$$a=\frac{m}{4\pi\hbar^2}\int d\mathbf{r}\,u(r)\exp(i\mathbf{p}\cdot\mathbf{r}/\hbar)=\frac{mu_0}{4\pi\hbar^2}\ . \tag{32}$$

The major contribution to the depletion is N_{d0}. Numerical calculations shown below indicate that N_{d1} is less than 10% of N_{d0}. Hence the depletion fraction of this 2D imperfect gas can be approximated as

$$\frac{N_d}{N}=\frac{N_{d0}+N_{d1}}{N} \tag{33}$$

$$\approx\frac{a\alpha}{2\pi^{1/2}}\frac{\Gamma(b-1/2)}{\Gamma(b-1)}\ . \tag{34}$$

It is very interesting that this depletion fraction is independent of the (2D) density N/A of the system. We may contrast this with the 3D for which case the result is a function of the (3D) density n_{3D}:[18]

$$\left[\frac{N_d}{N}\right]_{3D}\approx\frac{8}{3\pi^{1/2}}(n_{3D}a^3)^{1/2}\ . \tag{35}$$

The origin of this different dependence can be seen from the following heuristic argument. The characteristic interaction energy per particle is roughly

$$\bar{\varepsilon}\approx 2\pi a n\hbar^2/m \tag{36}$$

[see Eq. (11.2.22) in Ref. 18]. An *estimate* of the depletion fraction can be obtained by dividing by N the total number of single-particle states with energy less than $\bar{\varepsilon}$, i.e.,

$$\frac{N_d}{N}\cong\frac{1}{N}\int_0^{\bar{\varepsilon}}d\varepsilon g(\varepsilon)\ . \tag{37}$$

This would be exact if states with energy less (greater) than $\bar{\varepsilon}$ were singly occupied (unoccupied). Let the noninteracting single-particle density of states $g(\varepsilon)$ be

$$g(\varepsilon)\propto\varepsilon^{\gamma}\ . \tag{38}$$

Hence

$$\frac{N_d}{N}\propto\frac{\bar{\varepsilon}^{\gamma+1}}{n}\propto n^{\gamma}\ . \tag{39}$$

Thus the power of n is just that of ε in the density of states. $\gamma=0$ in 2D so there is *no* dependence on n. In 3D, $\gamma=\frac{1}{2}$, consistent with Eq. (35). In 1D, $\gamma=\frac{1}{2}$; therefore BEC is destroyed with even infinitesimal interactions and small n.

The depletion fraction in Eq. (34), however, is not a universal constant. It is actually a function of the shape and depth of the substrate potential. While the width α^{-1} is not a very sensitive parameter for different substrates, the well-depth ranges from several K to of order 100 K.[8] The well-depth dependence of the depletion fraction is complicated by the gamma functions in Eq. (34). A numerical calculation is needed to reveal the whole behavior.

We choose the following parameters in our calculation: the potential well width $\alpha=2$ Å^{-1} and scattering length $a=2.2$ Å.[18] Equation (35) then yields for 3D He a depletion of 35 percent, compared to the 90 percent found from more accurate calculations.[1,3] While N_{d0} is independent of the 2D density, N_{d1} is a function of N/A [see Eqs. (30) and (31)]. We calculate N_{d1} at different densities ($N/A=0.03$ Å^{-2} and 0.077 Å^{-2}) and find that the results are always less than 10% of N_{d0}. This result is confirmed by a calculation of SH potential in the reference. It is very reasonable since most particles stay in the surface-normal ground state. This is consistent with the neglect of contributions from states higher than the first excited state.

In Fig. 1, we show the depletion fraction calculated from Eq. (34) as a function of the well depth D. A monotonically increasing function is observed. At large D, the value is larger than one. The small depletion assumption is obviously responsible for this erroneous behavior. It is also interesting to note the asymptotic behavior of the depletion in the limit of very large D. In such a case we have:

$$\frac{N_d}{N}\to\frac{a\alpha e^{-1/2}}{2\pi^{1/2}}\frac{(b-1/2)^{b-1}}{(b-1)^{b-3/2}}\Longrightarrow a\alpha b^{1/2}\propto a\alpha^{1/2}D^{1/4}\ , \tag{40}$$

where Eq. (4) has been used. This asymptotic dependence is the same as that of a simple harmonic potential, as expected; see the Appendix.

The monotonic increase of the depletion fraction with D is expected. Large D corresponds to highly localized single-particle states; hence the two-body scattering is large enough to excite relatively many particles to excited states. This means a large depletion. On the other hand, adatoms are less confined in the case of a shallow potential well. By spreading over a large z domain, this scattering and the depletion are much reduced.

The spreading of the adatoms reflects the fact that our system is not exactly a 2D gas. The spatial extent of the wave functions can be estimated from a width parameter w:

$$w=\left[\int dz\,z^2\varphi_0^2-\left[\int dz\,z\varphi_0^2\right]^2\right]^{1/2}$$

$$=\frac{1}{\alpha}[\zeta(2,b-1)]^{1/2}\ , \tag{41}$$

where $\zeta(a,b)$ is the Riemann's zeta function.[20] A 3D density can then be defined as

$$n_{3D}\equiv\frac{N}{Aw}\ . \tag{42}$$

This is determined by both the 2D density and the potential well. The depletion is seen in Fig. 2 to be correlated with n_{3D}, but not a function of it alone.

We may summarize our results as follows. We have treated a submonolayer film by applying the quasi-2D analog of the Bogoliubov approximation. We find that the depletion fraction is independent of the 2D density, but strongly dependent on the substrate potential; small D corresponds to small depletion. The qualitative implications for real He films are similar; z-wise delocalization leads to an attenuated He-He interaction. The effect on the fragile 2D liquid-vapor condensation (binding energy one tenth of the 3D result) has been previously noted.[14] If

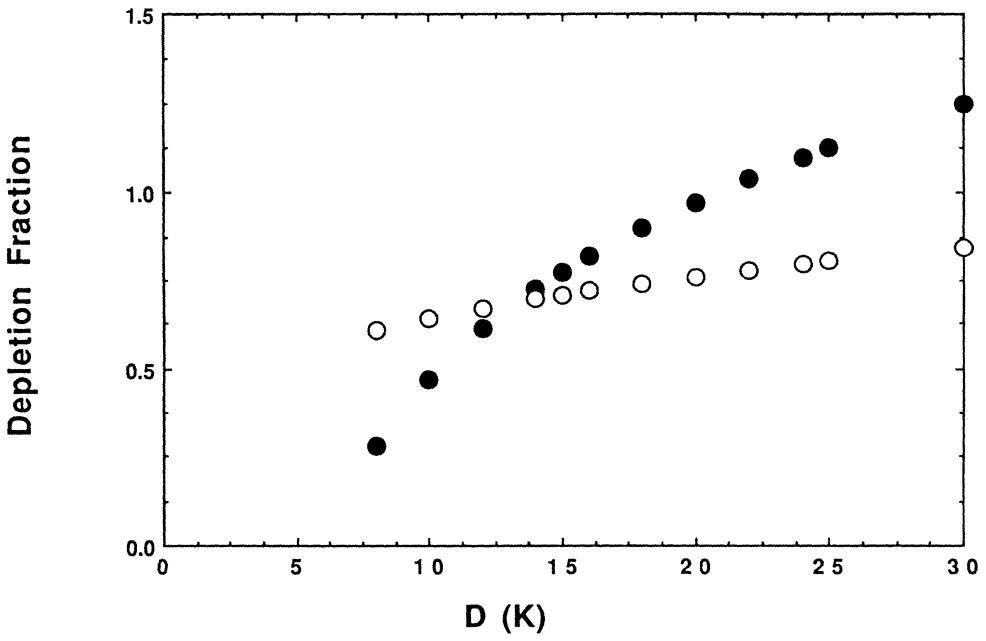

FIG. 1. The depletion fraction N_d/N, calculated from Eq. (34) (solid circle) and Eq. (A5) (open circle), as a function of the substrate potential well-depth D. The fraction is sometimes larger than 1 is due to the crudeness of our model; see the text.

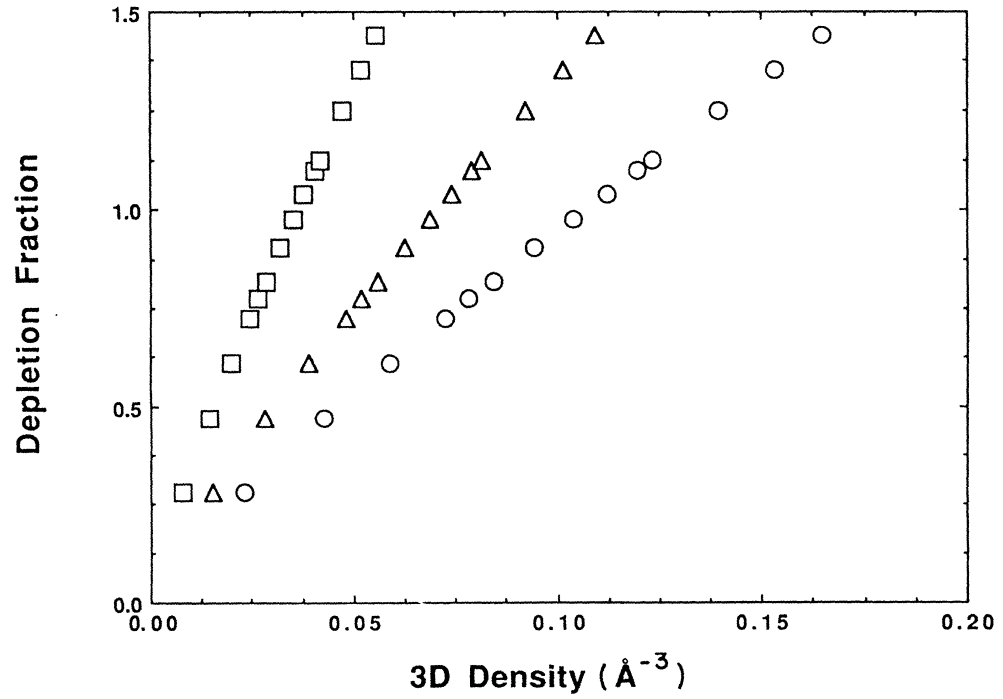

FIG. 2. The depletion fraction N_d/N, calculated from Eq. (34), as a function of the effective 3D density, defined as in Eq. (42). Different data sets are calculated at constant 2D densities ρ: squares are at $\rho=0.026$ Å^{-2}, triangles are at $\rho=0.051$ Å^{-2}, and circles are at $\rho=0.077$ Å^{-2}.

the D value is quite small, the liquid-vapor coexistence region may disappear, leading to a weakly interacting Bose gas of considerable fundamental interest. Particularly ideal substrates are alkali or H_2 surfaces.[8] Indeed Shirron *et al.*[9] found that the nonsuperfluid (inert) layer of He on H_2 was unprecedentedly small, consistent with the present idea. The alkali surfaces should be even more interesting because the D values are even smaller (<15 K versus 30 K for H_2).[21,22]

Note added in proof. Quantitative estimates are presented in a forthcoming publication we have written.

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APPENDIX: RESULTS FROM A SIMPLE HARMONIC POTENTIAL

Instead of the complicated SM potential Eq. (1), we can also use a simple harmonic (SH) model to approximate the substrate potential. It should give the same qualitative results when only the low-lying states are concerned. The potential, eigenvalues and eigenfunctions are

$$V_{\rm SH}(z)=-D(1+\Delta)+\tfrac{1}{2}m\omega^2(z-z_0)^2 , \tag{A1}$$

$$E_i^{\rm SH}=(i+\tfrac{1}{2})\hbar\omega-D(1+\Delta) , \tag{A2}$$

$$\Psi_i(z)=(2^i b i!)^{-1/2}H_i(z/b)\exp(-z^2/2b^2) , \tag{A3}$$

$$b=(\pi\hbar/m\omega)^{1/2} ,$$

where $H_n(x)$ is the Hermite polynomials. In order to compare with the SM potential used in the text, we chose the parameter of the SH potential so that they give the same ground state energy. This yields

$$\omega=D\left[\frac{4\alpha}{(8mD)^{1/2}}-\frac{2\hbar\alpha^2}{8mD}\right]\approx 4\alpha(D/8m)^{1/2} . \tag{A4}$$

The matrix elements I_{ijkl} can be calculated in terms of length b, which is $(2\pi)^{1/2}$ times the root-mean-square displacement of an isolated atom. The result of $2^{1/2}bI$ are $1(ijkl=0000)$, $\frac{1}{2}(0011)$, $3(1111)$, and zero (if there is an odd number of excited states). The energy and depletion calculations are the same as in the text.

The formula for depletion in this case are the same as in Eqs. (29)–(31), with different expressions for S_0 and S_1, and particularly, $S_{01}=0$. We have

$$\frac{N_{d0}}{N}=\frac{a}{2^{1/2}b} \tag{A5}$$

$$\frac{N_{d1}}{N}=\frac{a}{2^{3/2}b}\frac{2}{(x_0^2-4)^{1/2}+1} , \tag{A6}$$

$$x_0=\frac{\Delta E^{\rm SH}}{S_1}=\left[\frac{N}{A}\right]^{-1}\frac{2b}{a} . \tag{A7}$$

With Eq. (A4) and typical values for He films cited in text, we have $x_0\sim 15$; hence N_{d1} is less than ten percent of N_{d0}. With Eq. (A4), we also have

$$\frac{N_{d0}}{N}\Longrightarrow a\alpha^{1/2}D^{1/4} . \tag{A8}$$

This is the same dependence as in the asymptotic behavior of the SM potential, Eq. (40). The results of the SH potential are also shown in Fig. 1. The same qualitative behavior is observed. Compared to the SM potential, of course, the SH potential is simpler to use.

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