

Spin-polarized hydrogen-helium film system: A surface "polaron"

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Experimental investigations of spin-polarized hydrogen $H\downarrow$ are carried out in a sample cell lined with a helium film whose purpose is to shield the $H\downarrow$ atoms from magnetic impurities in the walls. The purpose of the He buffer is to minimize spin flips which cause recombination into the H_2 molecular ground state and degradation of the sample. In this paper, we focus our attention on the $H\downarrow$ - ${}^4\text{He}$ film system and investigate the consequences of allowing the adsorbed $H\downarrow$ atom to interact with a *dynamic* He surface. The coupling between the $H\downarrow$ atom and the modes of the film raises the possibility that the atom will find it energetically favorable to be localized in the plane parallel to the surface of the film, i.e., to form a $H\downarrow$ polaron. Using a simple model for the atom-film interaction, we show that polaron production can be important. We estimate the order of magnitude for the parallel binding energy and find that it can be comparable to the experimentally measured binding energy. We call attention to the important characteristics of the particle-film system that influence the size and nature of the polaron; e.g., the effect of evolution of the film from ${}^4\text{He}$ to ${}^3\text{He}$ and the consequences of the mobility of the surface modes. Where polaron effects are important the parallel binding problem and the perpendicular binding problem are strongly interdependent and cannot be factored. We then use a simple perturbation theory argument to calculate the polaron effective mass. We finally calculate the ripplon-mediated $H\downarrow$ - $H\downarrow$ interaction and show that it is short ranged and attractive.

I. INTRODUCTION

A major experimental effort¹ is underway to attempt to create long-lived samples of spin-polarized hydrogen $H\downarrow$. An important element of this experimental effort is the construction of a suitable sample space, i.e., a sample space that minimizes the spin-flip transition rate (since the presence of "wrong" spin atoms degrades the sample through $H + H \rightarrow H_2$ recombinations).¹⁻³ The current generation of experiments form the sample in a ${}^4\text{He}$ sample chamber, that is, a chamber with walls covered by a liquid- ${}^4\text{He}$ film. Thus, one of the important environments which atoms of the $H\downarrow$ sample continually visit is the ${}^4\text{He}$ film surface. At suitably low temperatures it is quite probable that the first high-density $H\downarrow$ forms in the vicinity of the ${}^4\text{He}$ film surface due to the van der Waals physisorbed state; also, it is near the surface that several important spin-flip mechanisms (forbidden elsewhere) can operate.⁴ Thus, in this paper we focus attention on the behavior of a $H\downarrow$ atom located near the surface of a ${}^4\text{He}$ film.

Guyer and Miller⁵ and Mantz and Edwards⁶ have calculated the binding of a single $H\downarrow$ atom to

the ${}^4\text{He}$ surface. [The $H\downarrow$ is found to be weakly bound in qualitative agreement with experiment⁷ (see Table I).] In these calculations the emphasis was placed on properly accounting for the *static* surface profile and for the effects of strong short-range correlations on the interaction energy. Here we look beyond the $H\downarrow$ *static* surface picture and we consider the interaction of the $H\downarrow$ atom with a *dynamic* surface: a surface described by quantized

TABLE I. Comparison of the calculated and measured binding energies of atomic hydrogen to the surface of liquid ${}^4\text{He}$.

	H	D	T
Expt.	0.93 ± 0.5^a 0.25 ± 0.02^b	2.5 ± 0.4^c	
ME ^d	0.63	1.39	1.83
GM ^e	0.1	1.1	2.4

^aReference 7 ($H\downarrow$ on ${}^4\text{He}$).

^bReference 21 ($H\downarrow$ on ${}^3\text{He}$).

^cReference 22.

^dMantz and Edwards, Ref. 6.

^eGuyer and Miller, Ref. 5.

deformation modes.⁸ If the $H\downarrow$ atom is completely delocalized in its motion parallel to the surface, then it pushes in the same way on all parts of the surface with a force at each point proportional to the probability that it sits at that point, i.e., proportional to A^{-1} where A is the area of the surface. The $H\downarrow$ atom pushes uniformly on the whole surface and drives the $k=0$ mode of the film. If the $H\downarrow$ atom motion becomes localized in a plane parallel to the surface, then it pushes on the surface beneath itself with a force that becomes larger as the atom becomes more localized. A localized $H\downarrow$ atom distorts the surface, drives the finite wave-vector modes of the film, and exerts maximum force on the nearby elements of the surface. As the $H\downarrow$ atom configuration evolves from completely delocalized to completely localized the surface-parallel contribution to the kinetic energy goes from 0 to $+\infty$. Simultaneously, however, the increasingly localized $H\downarrow$ atom will exert stronger and stronger forces which will tend to deform the surface (move it out of the way of the $H\downarrow$ atom) and lead to a *reduction* in the energy of interaction between the atom and the film. See Fig. 1. It is possible that a compromise between the kinetic energy expense and potential energy advantage of parallel localization is achieved for a $H\downarrow$ atom of finite extent in a plane parallel to the surface; i.e., there is a $H\downarrow$ "polaron."⁹ It is the purpose of this paper to explore this possibility and its consequences.

It is clear from these brief remarks that the qualitative and quantitative properties of the film and the modes it can support are an important part of this discussion. In Sec. II we write down and discuss the Hamiltonian we employ to describe (1) the film and its modes, (2) the $H\downarrow$ atom, and (3) the $H\downarrow$ atom-film coupling. In Sec. III we discuss the film and its modes. Special attention is focused on the driving force for the film modes (the force exerted by the substrate and on the force due to surface tension) and on the relationship of the wavelength of the modes to the film thickness. Both the dispersion relation for the film modes and the amplitude of the film's response to an external force driving its modes are influenced by these details. In Sec. IV we discuss the $H\downarrow$ polaron. We make an approximate calculation of its energy and draw particular attention to the possibility that substantial parallel localization of the $H\downarrow$ induces an important coupling between the perpendicular problem (Guyer and Miller,⁵ Mantz and Edwards⁶) and the parallel problem. In Sec. V we

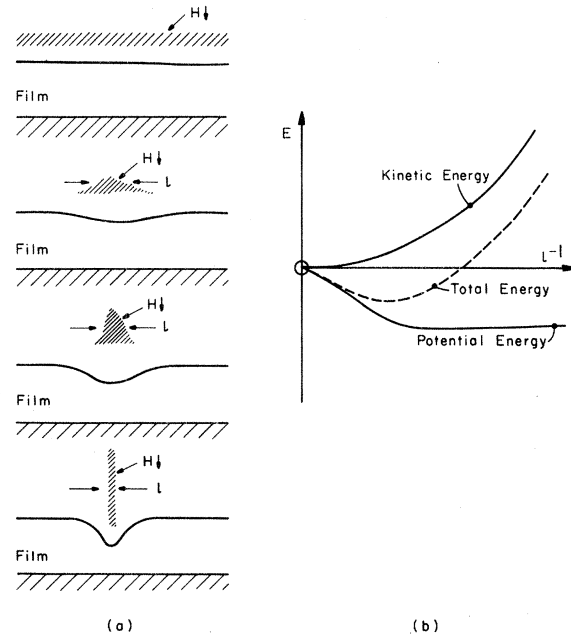


FIG. 1. $H\downarrow$ polaron. In (a) various possibilities for the parallel structure of a particle are considered. When the particle is nonlocal, $l \rightarrow +\infty$, it has zero parallel kinetic energy and zero potential energy, (1). The energy as a function of l is in (b). As it becomes localized the particle deforms the surface and reduces its potential energy, as in (2) and (3). When the particle is highly localized it has large kinetic energy, as in (4). The total energy could have a minimum in a localized structure between (1) and (4).

calculate the effective mass of the $H\downarrow$ and discuss the $H\downarrow$ - $H\downarrow$ interaction communicated by the modes of the film (it is a short-range interaction because of the nature of the $k \rightarrow 0$ modes of the film). Our concluding remarks are in Sec. VI.

Throughout the text we will talk about the "particle"; we are thinking about $H\downarrow$ but we also keep in mind that the effects we are describing will be qualitatively similar and perhaps more important for $D\downarrow$ and $T\downarrow$ and for other free particles that might reside above the film.

II. ENERGY OF SYSTEM

The energy of the coupled particle-film (PF) system is written as the sum of three terms

$$E = E_F + E_P + E_{PF}, \quad (1)$$

where

$$E_F = \int d^2\rho \left[\frac{\hbar^2}{2m_4} |\nabla_\rho \psi|^2 + naW(|\psi|) - \mu_4 |\psi|^2 \right], \quad (2)$$

$$E_P = \int d^2\rho' \int dz' \frac{\hbar^2}{2m_p} |\nabla\Phi(\vec{x}')|^2, \quad (3)$$

and

$$E_{PF} = \int d^2\rho \int_0^{h(\vec{\rho}, t)} dz \int d\vec{x}' nV(\vec{x} - \vec{x}') \times |\Phi(\vec{x}')|^2. \quad (4)$$

These terms are the film energy E_F , the particle energy E_P , and the energy E_{PF} associated with the coupling of the particle to the film. They are described in detail here. In this connection see Fig. 2.

A. E_F , the film energy

We take the system to be at suitably low temperature so that the film is at $T \ll T_\lambda$ and the ^4He density (assumed to be uniform) is equal to the superfluid density. The height of the film at $\vec{\rho}$ is taken to be¹⁰ (see Fig. 1)

$$nh(\vec{\rho}) = |\psi(\vec{\rho})|^2, \quad (5)$$

where $\psi(\vec{\rho})$ is the complex order parameter of the film. (We use n to denote the average number density of the film $n = a^{-3}$, $a \approx 3.6 \text{ \AA}$). The three terms in Eq. (2) for E_F are (1) the kinetic energy measured by the gradient of ψ , (2) a term proportional to μ_4 , the ^4He chemical potential that determines the equilibrium film profile (see below), (3) the energy of interaction of the film with the sub-

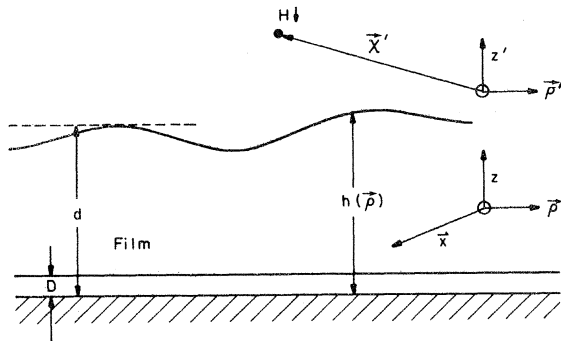


FIG. 2. Notation. The coordinate systems and variables used to describe the particle and film are shown.

strate, i.e.,

$$W_s[\psi(\vec{\rho}, t)] = \alpha \int_D^{h(\vec{\rho}, t)} \frac{dz}{a} \left[\frac{a}{z} \right]^3, \quad (6)$$

where α is the strength of the van der Waals interaction between the ^4He and the substrate (the mobile part of the film is set a distance D away from the substrate), and (4) the energy associated with the production of new film surface

$$W_T[\psi(\vec{\rho}, t)] = \frac{1}{2} \frac{\sigma a^2}{n^2} (\nabla_\rho |\psi|^2)^2, \quad (7)$$

where σ is the surface tension of the ^4He film ($\sigma = 0.378 \text{ ergs/cm}^2$ for bulk ^4He).

B. E_{PF} , the particle-film interaction

The particle-film interaction [see (4)] depends upon the film profile through $h(\vec{\rho})$ and upon the wave function for the particle above the film $\Phi(\vec{x}')$. If we introduce a Hartree-type decomposition for $\Phi(\vec{x}') = \phi(z')\theta(\vec{\rho}')$, then E_{PF} may be divided into two parts

$$E_{PF} = V_\perp + V_\parallel,$$

where

$$V_\perp = \int_D^d dz \int dz' \langle V(z-z') \rangle_\perp |\phi(z')|^2, \quad (8)$$

and

$$V_\parallel = \int d^2\rho \delta h(\vec{\rho}) \int d^2\rho' \langle V(\vec{\rho} - \vec{\rho}') \rangle_\parallel |\theta(\vec{\rho}')|^2. \quad (9)$$

V_\perp is the interaction of the particle with a static film of thickness d and V_\parallel is the interaction of the particle with the modes of the film (the *parallel* excitations) embodied in

$$\delta h(\vec{\rho}) \equiv h(\vec{\rho}) - d.$$

The angle brackets in Eqs. (8) and (9) are defined by

$$\langle \dots \rangle_\perp = \int d^2\rho \int d^2\rho' \dots |\theta(\vec{\rho}')|^2, \quad (10a)$$

$$\langle \dots \rangle_\parallel = \int dz \int dz' \dots |\phi(z')|^2 \delta(z-d), \quad (10b)$$

where the δ function in (10b) fixes the film surface at its equilibrium height $z = d$ [corrections only appear as second order in δh in Eq. (9)]. The perpendicular structure of the particle $\phi(z')$ is determined by the z part of the kinetic energy in Eq. (3) and by the potential energy in Eq. (8),

$$E_\perp[\phi(z')] = \int dz' \frac{\hbar^2}{2m_p} \left[\frac{d\phi}{dz'} \right]^2 + V_\perp. \quad (11)$$

A relatively sophisticated treatment of the perpendicular problem, modeled here by Eqs. (11) and (8), has been given by Guyer and Miller⁴ and Mantz and Edwards.⁵ For the time being we take the perpendicular problem to be solved. The parallel structure of the particle $\theta(\vec{\rho}')$ is determined by the $\vec{\rho}'$ part of the kinetic energy in Eq. (3) and by the potential energy in Eq. (9) [which couples $\theta(\vec{\rho}')$ to the parallel excitations of the film]. Below, we look in some detail at the various terms in Eq. (1).

III. THE FILM

A. Modes of the film

The structure of the film and the modes of the interacting film (with no particle-film coupling) are found from Eq. (2).¹⁰ Upon writing

$$h(\vec{\rho}, t) = d + \delta h(\vec{\rho}, t),$$

$$\psi(\vec{\rho}, t) = \psi_0 + \delta\psi(\vec{\rho}, t),$$

$$|\psi|^2 = nh(\vec{\rho}, t)$$

and then expanding Eq. (2) to second order in $\delta\psi$, we find

$$E_F = E_F^{(0)} + E_F^{(1)} + E_F^{(2)}, \quad (12)$$

where $E_F^{(0)}$ depends upon d , $E_F^{(1)}$ is linear in $\delta\psi(\vec{\rho})$, and $E_F^{(2)}$ is quadratic in $\delta\psi(\vec{\rho})$. For $E_F^{(1)}$ we have

$$E_F^{(1)} = - \int d^2\rho \left[\mu_4 + \alpha \left(\frac{a}{d} \right)^3 \right] \psi_0 (\delta\psi + \delta\psi^*), \quad (13)$$

where we have dropped the surface tension contribution to W . If we take μ_4 as given and set the coefficient of $\delta\psi(\rho)$ equal to zero, we find d as a function of μ_4 ; the film thickness is determined by μ_4 . Use of the value of d from Eq. (13) in $E_F^{(0)}$ yields the energy that resides in the static film. The modes of the film are found from the quadratic term in Eq. (12). For $E_F^{(2)}$ we find

$$E_F^{(2)} = \int d^2\rho \left[\frac{\hbar^2}{2m_4} |\nabla\delta\psi|^2 + \frac{3}{2}\alpha \left(\frac{a}{d} \right)^3 (\delta\psi + \delta\psi^*)^2 \right]. \quad (14)$$

We now introduce explicitly the real and imaginary parts of the order-parameter fluctuation:

$$\delta\psi(\vec{\rho}, t) = \sqrt{nd} [\eta(\vec{\rho}, t) + i\xi(\vec{\rho}, t)]. \quad (15)$$

Thus,

$$E_F^{(2)} = nd \int d^2\rho \left[\frac{\hbar^2}{2m_4} [(\nabla\eta)^2 + (\nabla\xi)^2] + 6\alpha \left(\frac{a}{d} \right)^3 \eta^2 \right]. \quad (16)$$

The equations of motion for small amplitude disturbances are found from

$$i\hbar \frac{\partial\psi}{\partial t} = \frac{\delta E_F^{(2)}}{\delta\psi^*}$$

or equivalently,

$$\hbar \frac{\partial\eta}{\partial t} = \frac{1}{2nd} \frac{\delta E_F^{(2)}}{\delta\xi},$$

$$\hbar \frac{\partial\xi}{\partial t} = - \frac{1}{2nd} \frac{\delta E_F^{(2)}}{\delta\eta}.$$

Thus, we obtain the continuity equation

$$\frac{\partial\eta}{\partial t} + \vec{\nabla}_\rho \cdot \vec{v} = 0, \quad (17)$$

and the equation of motion

$$\frac{\partial}{\partial t} \frac{\hbar}{2m_4} \xi = - \frac{3\alpha}{m_4} \left(\frac{a}{d} \right)^3 \eta + \left(\frac{\hbar}{2m_4} \right)^2 \nabla_\rho^2 \eta, \quad (18)$$

where $\vec{v} = \vec{\nabla}_\rho (\hbar/2m_4)\xi$ is the velocity of the fluid in the film parallel to the surface. Equations (17) and (18) are combined to yield the dispersion relation for third sound

$$C_3^2 \equiv \left(\frac{\omega}{k} \right)^2 = \frac{3\alpha}{m_4} \left(\frac{a}{d} \right)^3. \quad (19)$$

[We have dropped the second driving term in Eq. (18).] C_3^2 is proportional to the van der Waals force on the upper edge of the film $C_3^2 \propto d^{-3}$.

In Eq. (19) we have an expression for $\omega(k)$ when the film is driven at the upper edge by the van der Waals force and when the wavelength of the disturbance is long compared to the film thickness d . We will continue to pursue a calculation subject to these and further specializations. Below we will indicate how the relevant results are modified to deal with a driving force due to surface tension and the effects of the wavelengths which are short compared to the film thickness.

The modes of the film are quantized by introducing the definitions

$$\eta(\vec{\rho}) = \sum_k (\eta_k e^{i\vec{k}\cdot\vec{\rho}} + \eta_k^* e^{-i\vec{k}\cdot\vec{\rho}}) \quad (20)$$

and

$$\vec{v}(\vec{\rho}) = \sum_k (\vec{v}_k e^{i\vec{k}\cdot\vec{\rho}} + \vec{v}_k^* e^{-i\vec{k}\cdot\vec{\rho}}) \quad (21)$$

and writing the continuity equation in the form $\eta_k = \vec{k}\cdot\vec{v}_k/\omega_k$. We find from Eq. (16)

$$E_F^{(2)} = N \sum_k \frac{2m_4\omega_k^2}{k^2} (\eta_k \eta_k^* + \eta_k^* \eta_k). \quad (22)$$

Upon defining $C(k)$ through $\eta_k = C(k)a_k$, $\eta_k^* = C(k)a_k^\dagger$ we find

$$E_F^{(2)} = \sum_k \hbar\omega_k (a_k^\dagger a_k + \frac{1}{2}), \quad (23)$$

with

$$C(k)^2 = \frac{\hbar k^2}{4m_4 N \omega_k} \quad (24)$$

and ω_k given by Eq. (19). It is useful to note that Eqs. (20) and (24) lead to

$$\eta(\rho) = \sum_k \left[\frac{\hbar k^2}{4m_4 N \omega_k} \right]^{1/2} (a_k + a_k^\dagger) e^{i\vec{k}\cdot\vec{\rho}}. \quad (25)$$

We call particular attention to the amplitude $C(k)$ that relates the creation or destruction of a surface excitation to $\eta(\rho)$ [or $\delta h(\rho)$]. The amplitude goes as \sqrt{k} , $C(k) \propto (k^2/\omega_k)^2$, so that $\delta h(\rho) \rightarrow 0$ as the wave vector of the excitation approaches zero. This result is a consequence of the nature of the driving force in Eq. (16) (i.e., this driving force is proportional to η^2) and it has important consequences for the form of the film-mediated interaction between adsorbed particles.

As remarked on above, the equations that we have developed are subject to some specialization. The essential features of the development are un-

changed when two additional effects are taken into account. These additional effects are: (1) modification of the forces which drive the film to include surface tension [the second term in W , W_T from Eq. (7)] and (2) modification of the flow pattern of the fluid that occurs as the wavelength of the disturbance becomes comparable to or smaller than the film thickness. Before we proceed to discuss the consequences of accounting for (1) and (2) above, let us pause to define the language we will use in the developments below. It is conventional to refer to the modes of the film that are driven by the substrate force on the upper surface as *third sound*.¹¹ The modes that are driven by the surface tension and that are at $kd \gg 1$ (wavelength short compared to d) are capillary waves. The quanta of these surface excitations are ripples. Those modes of the film with wavelengths that are long compared to the film thickness are *shallow modes*¹² (actually the modes are of a shallow film) and those with wavelengths that are short compared to the film thickness are *deep modes*. The films we will consider are driven at the upper surface by forces from the substrate and by the force associated with surface tension. They can support modes that have wavelengths that are long or short compared to the film thickness. We refer to these four possibilities using a language that describes the wavelength-thickness relationship and the source of the driving force; i.e., long substrate (LS), short substrate (SS), long (surface) tension, (LT) and short (surface) tension, (ST) modes. See Table II. Below we refer to a generic excitation of the film as a ripplon.

To describe the modes of the film we employ the hybrid dispersion relation

$$\omega^2 = \left[\frac{C_3^2}{d} + \frac{\sigma a^3 k^2}{m_4} \right] k \tanh kd, \quad (26)$$

TABLE II. Notation for modes of a film. λ is the wavelength of the mode and d is the film thickness.

Old name	Driving force	Size	New name
shallow third sound	substrate	$\lambda \gg d$	long, substrate (LS)
deep third sound	substrate	$\lambda \ll d$	short, substrate (SS)
shallow capillary wave	surface tension	$\lambda \gg d$	long, surface tension (LT)
deep capillary wave	surface tension	$\lambda \ll d$	short, surface tension (ST)

which gives the four limits correctly ($n=a^{-3}$):

$$\begin{aligned}
 \text{LS: } & \sigma=0, \quad kd \ll 1, \quad \omega^2 = C_3^2 k^2, \\
 \text{SS: } & \sigma=0, \quad kd \gg 1, \quad \omega^2 = C_3^2 k/d, \\
 \text{LT: } & C_3^2=0, \quad kd \ll 1, \quad \omega^2 = \frac{\sigma(ka)^3}{m_4} kd, \\
 \text{ST: } & C_3^2=0, \quad kd \gg 1, \quad \omega^2 = \frac{\sigma(ka)^3}{m_4}.
 \end{aligned} \tag{27}$$

While the dispersion relation changes its form as the wavelength and driving force change, the amplitude $C(k)$, which relates the creation (destruction) of an excitation of the film to the amplitude of the surface [see Eq. (25)] depends only on kd . We write the hybrid amplitude

$$C(k)^2 = \frac{\hbar k (\tanh kd)}{4m_4 N \omega d}, \tag{28}$$

which gives the two limits correctly:

$$\begin{aligned}
 \text{LS, LT: } & kd \ll 1, \quad C(k)^2 = \frac{\hbar k^2}{4m_4 N \omega}, \\
 \text{SS, ST: } & kd \gg 1, \quad C(k)^2 = \frac{\hbar k}{4m_4 N \omega d}.
 \end{aligned} \tag{29}$$

The various limiting forms exhibited here Eqs. (27) and (29) can be derived by following the steps outlined in Eqs. (16)–(24), and incorporating three modifications: (1) the change from the substrate driving force to the surface tension driving force introduces a factor of k^2 in the last term in Eq. (16), (2) the transition from long-wavelength to short-wavelength modes changes the kinetic energy in the flow pattern¹³ (an amount of fluid proportional to k^{-1} instead of d flows in response to the forces), and (3) because of the change in flow pattern the continuity equation changes from $\delta \dot{h}_k + d(ikv_k) = 0$ to $\delta \dot{h}_k + k^{-1}(ikv_k) = 0$; i.e., the $\eta_k - v_k$ relationship changes [see below Eq. (21).]

The ω - k relation in Eq. (26) has two crossovers as k increases. At

$$k_1 a = \left[\frac{m_4 C_3^2}{\sigma a^2} \right]^{1/2} \frac{a}{d} \approx 5 \left[\frac{a}{d} \right]^2, \tag{30}$$

there is a crossover from long-wavelength substrate driven modes to long-wavelength surface-tension driven modes; we use $m_4 C_3^2 = 3\alpha(a/d)^3$, $3\alpha \approx 10^{-14}$ erg, and $\sigma = 0.38$ ergs/cm² to obtain the numerical estimate on the right-hand side of Eq. (30). At

$$k_2 a = \frac{a}{d} \tag{31}$$

there is a crossover from long-wavelength modes ($kd \ll 1$) to short wavelength modes. Except for the thinnest films, we always have $k_1 < k_2$. Thus as k increases, the sequence is LS→SS→ST. See Fig. 3.

We close this description of the modes of the film with several observations. (1) Both the substrate and surface-tension driven modes are relatively soft. At $ka \rightarrow 1$, $\hbar C_3 k \approx 3(a/d)^{3/2} \ll 1$ K for $d \gg a$. At $ka \rightarrow 1$ the energy in the surface tension driven modes is of order $\sigma a^2 \approx 5$ K. Disturbances of the surface on a length scale of order a or smaller may bring into play energies (and forces) that are quite a bit larger than those that are found from $\hbar\omega$ by extrapolating the ω in Eq. (26) to $ka \approx 1$.¹⁴ We will sometimes use a k cutoff at $ka \approx 1$ to examine the consequences of the treatment of the large wave-vector modes; (see, e.g., Sec. VB). (2) As $k \rightarrow 0$ the dispersion relation goes unambiguously to $\omega = C_3 k$, the LS mode, and $C(k) \sim \sqrt{k}$. See the discussion at the conclusion of Sec. VI.

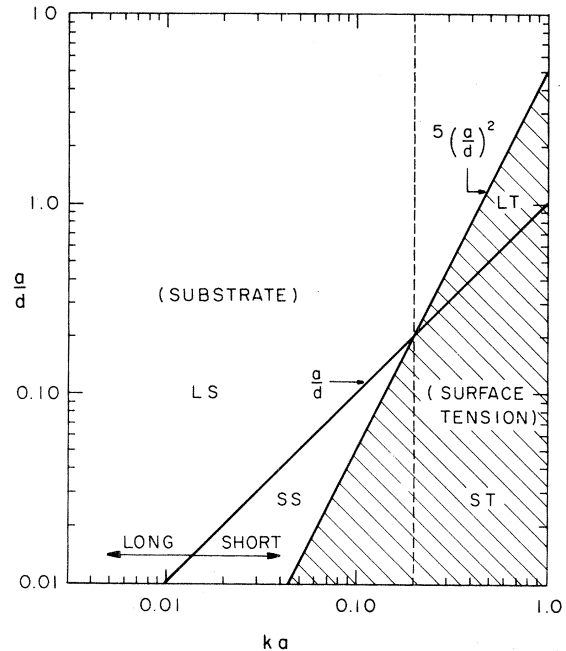


FIG. 3. Crossovers and the dispersion relation. As described in the text the modes of the film evolve in a way that depends upon the film thickness d and on the mode wavelength ka . The crossover lines are shown as a function of a/d and ka [see Eqs. (30) and (31)]; however, its precise location depends upon details that cannot be assessed in general.

IV. THE PARTICLE-FILM INTERACTION: THE POLARON

The particle couples to the modes of the film through $V_{\parallel}[\theta(\vec{\rho})]$ [see Eq. (9)]. In the following, we shall only consider the linear (one ripplon) coupling terms. Thus, we set $\delta h(\vec{\rho}) = 2d\eta(\vec{\rho})$ and find

$$V_{\parallel} = 2nd \int d^2\rho \eta(\vec{\rho}) F(\vec{\rho}) \\ = \frac{d}{a} \sum_k (\eta_k F_{-k} + \eta_k^* F_k), \quad (32)$$

where

$$F(\vec{\rho}) \equiv \int d^2\rho' \langle V(\vec{\rho} - \vec{\rho}') \rangle_{\parallel} |\theta(\vec{\rho}')|^2 \\ = \frac{a^2}{A} \sum_k F_k e^{-i\vec{k} \cdot \vec{\rho}}. \quad (33)$$

If we now substitute a_k and a_k^\dagger as defined below Eq. (22), we obtain

$$V_{\parallel} = \sum_k [E(k)a_k + E(-k)a_k^\dagger], \quad (34)$$

where we have defined

$$E(k) \equiv \frac{d}{a} C(k) F_{-k}, \quad (35)$$

and $C(k)$ is from Eq. (28). We now add this contribution to the film energy to find

$$E' = \sum_k [\hbar\omega_k (a_k^\dagger a_k + \frac{1}{2}) \\ + E(k)a_k + E(-k)a_k^\dagger]. \quad (36)$$

E' is easily brought into diagonal form by introducing the operators $b_k = a_k + E(-k)/\hbar\omega_k$. Thus, we find

$$E' = \sum_k \hbar\omega_k (b_k^\dagger b_k + \frac{1}{2}) + U_0[\theta], \quad (37)$$

where

$$U_0[\theta] = - \sum_k E(k) \frac{1}{\hbar\omega_k} E(-k) \quad (38)$$

acts as a single particle potential that can bring about the parallel localization of the particle. In competition with this energy reduction is the parallel kinetic energy in Eq. (3). Thus, the parallel wave function is found from the energy

$$E_{\parallel} = \int d^2\rho \frac{\hbar^2}{2m_p} |\vec{\nabla}_{\rho}\theta(\vec{\rho})|^2 + U_0[\theta(\vec{\rho})]. \quad (39)$$

We refer to the problem set by Eq. (39) as the *parallel problem*. The Euler-Lagrange equation

$\delta E_{\parallel}/\delta\theta^*(\vec{\rho}) = 0$ for the optimal θ can now be easily obtained. We shall, however, take a less direct route which may better illustrate the essential physics.

Let us consider a simple contact interaction

$$V(\vec{x} - \vec{x}') = V_0 a^3 \delta(\vec{x} - \vec{x}')$$

to describe the particle-film potential. With this form, we obtain from Eq. (33)

$$F(k) = (V_0 a |\phi(d)|^2) P(k), \quad (40)$$

where

$$P(k) = \int d^2\rho e^{i\vec{k} \cdot \vec{\rho}} |\theta(\vec{\rho})|^2, \quad (41)$$

is the form factor for the parallel wave function. The potential $U_0[\theta]$ can be written:

$$U_0[\theta] = - [V_0 a |\phi(d)|^2]^2 \\ \times \left[\frac{d}{a} \right]^2 \sum_k \frac{C^2(k) P^2(k)}{\hbar\omega_k}. \quad (42)$$

In order to assess the effects of localization we shall use a simple Gaussian form for the parallel wave function:

$$\theta(\rho) = \left[\frac{\gamma}{\pi} \right]^{1/2} e^{-(1/2)\gamma\rho^2}. \quad (43)$$

Using the Gaussian in Eq. (41), we find $P(k) = \exp(-k^2/4)$ and thus

$$E = \frac{\hbar^2\gamma}{2m_p} - [V_0 a |\phi(d)|^2]^2 \left[\frac{da}{8\pi m_4 C_3^2} \right] \gamma. \quad (44)$$

In the completely delocalized limit $\gamma \sim 1/A$ and thus E vanishes (it goes to zero inversely as the area of the system). In order to obtain localization we need to find a state with $E < 0$. In this simple model we cannot obtain an estimate for the width γ since both the kinetic and potential energies are linearly proportional to γ . We can, however, obtain an estimate of the value of $d \equiv d_c$ where for $d > d_c$ polaron production becomes energetically favorable. Thus, from Eq. (44) we find

$$\left[\frac{d_c}{a} \right] = 8\pi \frac{\left[\frac{\hbar^2}{2m_p a^2} \right] (m_4 C_3^2)}{[V_0 a |\phi(d)|^2]^2}, \quad (45)$$

(before we have extracted the d dependence in C_3^2). We can see qualitatively from Eq. (45) that the larger the energy cost in localization ($\sim \hbar^2/m_p$) re-

relative to the particle-film interaction and the larger the energy cost in distorting the surface ($\sim m_4 C_3^2$) relative to the particle-film interaction, the more difficult it is to produce polarons (i.e., one must go to thicker and thicker films). Let us now use $m_4 C_3^2 = 3\alpha(a/d)^3$ in (45)

$$\left(\frac{d_c}{a}\right) = \left[\frac{8\pi \frac{\hbar^2}{2m_p a^2} 3\alpha}{[V_0 a |\phi(d)|^2]^2} \right]^{1/4}, \quad (46)$$

and using $\alpha = 25$ K (glass substrate), $a = 3.6$ Å and $V_0 a |\phi(d)|^2 \approx 2$ K we find $d_c/a \approx 5-6$ layers. In Fig. 4 we plot $E_{||}$ as a function of γa^2 and d/a for the above values of the various parameters. At a given localization the binding energy decreases with increasing film thickness. This behavior reflects the softening of the ripplon spectrum with

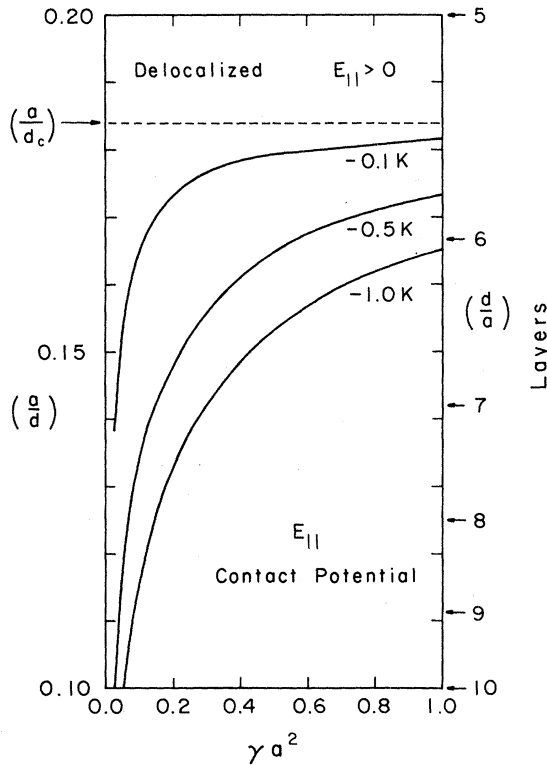


FIG. 4. The parallel binding energy $E_{||}$ [Eq. (44)] using a Gaussian for the $H\downarrow$ distribution function, as a function of film thickness d/a and localization a^2 . Using the values $V_0 a |\phi(d)|^2 = 2$ K and $\alpha = 25$ -K (layer)³, we find polaron production energetically unfavorable for $d/a \leq 6$ layers. For thick films, localization can lead to values for $E_{||}$ of the same order of magnitude as the measured (Ref. 6) binding energy 0.9 K.

increasing d ($m_4 C_3^2 \sim d^{-3}$); i.e., the thicker the film, the easier it is to distort the surface.

From the discussion here it is clear that if the long wavelength modes of the film alone are involved, then a polaron will form for suitably thick films. The size of the polaron will be comparable to the size of the modes that support it (i.e., to the wavelength of the modes to which the parallel structure of the particle is coupled). If the long wavelength modes alone are involved, then the polaron will become larger and larger as the film is thickened and its binding energy to the film will become smaller and smaller. Such a polaron is not much different from a free particle. However, we are looking for the possibility of parallel localization that involves energies that are important to the surface problem, i.e., energies that are of the order of the surface binding energy, e.g., 1 K. From $\hbar^2 k^2 / 2m_p \approx 1$ K, $k \approx 10^8$ cm⁻¹, we see that important polaron effects occur if the particle can couple advantageously to modes of the surface at $ka \approx 1$, i.e., to the short-wavelength modes of the surface. The elasticity of these modes, the measure of the forces the film brings to bear to prevent distortion, is much larger than that of the long-wavelength modes, and is independent of the film thickness. Important polaron effects will occur if the potential energy

$$U_0 \approx - \frac{[V_0 a \phi(d)^2]^2}{\sigma a^2}$$

is greater than the kinetic energy $\hbar^2 / 2m_p a^2$. This potential energy drives the particle toward the polaron state and depends importantly on the characteristics of the system: (1) $H\downarrow$ on ⁴He and $H\downarrow$ on ³He have different values of $\phi(d)^2 a$. $T\downarrow$, for example, on the average resides closer to the ⁴He surface than $H\downarrow$, etc., and (2) by considering a ³He-⁴He mixture one can adjust the surface tension¹⁵ in a continuous manner between the two pure limiting cases. In addition there are considerations of another genre. The modes of a ⁴He film are fast and undamped so that the particle and its "dressing" can move together. The short-wavelength modes of a ³He film are overdamped, however, and the particle may have to leave the modes of the film behind in order to move.

Finally we note that the potential energy that gives advantage to parallel localization [Eq. (44)] is driven by $\phi(d)^2 a$, the probability that the particle is on the surface. This probability is governed by the perpendicular problem, Eq. (11). Thus, the perpendicular problem, Eq. (11), and the parallel problem,

Eq. (39), are not independent. In this paper we will not attempt to deal with the coupled perpendicular problem using the detailed numerical methods which were employed in the relatively sophisticated treatments of the perpendicular problem alone. We will be content here to draw attention to the physical ideas that underlie a discussion of the binding of a particle to the ^4He surface with a simple model of the important relationships.

V. THE PARTICLE

A. The effective mass

In the development above, attention has been focused on the coupling of a particle above the surface to the modes of the surface and on the potential energy that results from this coupling. Our picture has been that of a single *static* particle above the surface. We find that this particle may couple strongly enough to the surface to cause parallel localization and an attendant localized surface distortion, as seen in Fig. 1. When the particle moves it carries this localized surface distortion along, and as a consequence, it has an effective mass. To estimate this effective mass we shall use a simple perturbation theory approach.¹⁶

From Eqs. (23) and (34) we can cast the particle-film problem in the operator form

$$H = H_0 + H', \quad (47)$$

where

$$H_0 = \sum_k \hbar\omega_k (a_k^\dagger a_k) + \frac{\hat{p}^2}{2m_p}, \quad (48)$$

$$H' = \sum_k [E(k)a_k + E(-k)a_k^\dagger], \quad (49)$$

the a_k^\dagger (a_k) create (destroy) ripplons, \hat{p} is the hydrogen atom momentum operator, and we have neglected the zero-point term in Eq. (48). The eigenfunction of H_0 can be written

$$\psi_{\vec{p}}^{(0)} = \theta_{\vec{p}} |0\rangle \equiv |\vec{p}; 0\rangle \quad (50)$$

which denotes a state with no ripplons ($|0\rangle$ is the film vacuum) and a hydrogen atom with momentum \vec{p} ($\theta_{\vec{p}}$ is now a two-dimensional plane wave state). We can proceed by building a basis with total momentum \vec{p} and increasing numbers of ripplons:

$$\begin{aligned} \theta_{\vec{p}-\vec{k}} a_k^\dagger |0\rangle &= |\vec{p}-\vec{k}; \vec{k}\rangle, \\ \theta_{\vec{p}-\vec{k}-\vec{l}} a_k^\dagger a_l^\dagger |0\rangle &= |\vec{p}-\vec{k}-\vec{l}; \vec{k}, \vec{l}\rangle, \end{aligned} \quad (51)$$

etc. If we keep only the single ripplon states, then the wave function, in first-order perturbation theory, can be written

$$\psi_{\vec{p}}^{(1)} = \psi_{\vec{p}}^{(0)} + \sum_q' \frac{H'_{q0}}{\left[\frac{p^2}{2m_p} - \epsilon_q \right]} |\vec{p}-\vec{q}; \vec{q}\rangle, \quad (52)$$

where

$$H'_{q0} = e^{i\vec{q}\cdot\vec{p}} E(-q), \quad (53)$$

and

$$\epsilon_q = \frac{(\vec{p}-\hbar\vec{q})^2}{2m_p} + \omega_q. \quad (54)$$

The second-order energy shift is thus given by

$$\begin{aligned} \Delta E &= \sum_q' \frac{|H'_{q0}|^2}{\left[\frac{p^2}{2m_p} - \epsilon_q \right]} \\ &= - \sum_q \frac{|E(q)|^2}{\left[\hbar\omega_q + \frac{\hbar^2 q^2}{2m_p} - \frac{\vec{p}\cdot\hbar\vec{q}}{m_p} \right]}. \end{aligned} \quad (55)$$

We shall only consider the case of slow atoms (i.e., no Cerenkov behavior) and thus we can develop ΔE in a power series in p^2 :

$$\Delta E = \Delta E^{(0)} + \Delta E^{(2)} + \dots, \quad (56)$$

where $\Delta E^{(2n)}$ is proportional to p^{2n} and we shall omit terms of higher order than p^2 . In order to compute ΔE , we need a specific model for the $E(q)$ in Eq. (55). For simplicity we shall use the contact potential which was introduced in Sec. IV. Thus, from Eqs. (35), (40), and (41):

$$|E(q)|^2 = \left(\frac{d}{a} \right)^2 \frac{[V_0 a |\phi(d)|^2]^2}{4m_4 C_3 N} \hbar q e^{-q^2/2\gamma} \quad (57)$$

with the Gaussian form in Eq. (43) for the parallel wave function. This form of the wave function is appropriate to the unperturbed state $\psi_{\vec{p}}^{(0)}$ in the limit $\gamma a^2 \ll 1$; i.e., for a state of modest parallel localization, for which only the long-wavelength modes of the film are important. To be consistent with this we have taken $\omega_q = C_3 q$. Substituting Eq. (57) in Eq. (55) we find

$$\Delta E^{(0)} = U_0 + O((\gamma a^2)^{3/2}) \quad (58)$$

and

$$\Delta E^{(2)} = \frac{U_0}{m_p C_3^2} \left[\frac{p^2}{2m_p} \right] + O((\gamma a^2)^2), \quad (59)$$

where U_0 is the parallel potential defined in Eqs. (39) and (44). Thus, the energy for the parallel problem can be written

$$E_{||} = \frac{p^2}{2m_p} + U_0 + \frac{U_0}{m_p C_3^2} \frac{p^2}{2m_p}$$

from which we can extract the effective mass m_p^* ,

$$\frac{m_p^*}{m_p} = \frac{1}{1-\epsilon},$$

where

$$\begin{aligned} \epsilon &= \frac{-U_0}{m_p C_3^2} = \frac{d}{a} \left[\frac{V_0 a |\phi(d)|^2}{m_4 C_3^2} \right]^2 \\ &= \left[\frac{V_0 a |\phi(d)|^2}{3\alpha} \right]^2 \left[\frac{d}{a} \right]^7. \end{aligned} \quad (60)$$

The effective mass increases with increasing film thickness, which reflects the fact that the surface of a thicker film can be distorted more easily than that of a thinner film. Note, however, that Eq. (60) is valid only for *very* thin films. For films which are more than a few layers thick, the energy shift calculated is greater than the unperturbed energy. Using the full dispersion relation [Eq. (26)] and the hybrid amplitude [Eq. (28)] in Eq. (55) curbs this effect somewhat. Nevertheless, the energy shift is comparable to the unperturbed energy for fairly thin films [$d < 10-15$ layers, depending on the estimate of $V_0 a |\phi(d)|^2$]. This suggests that for thicker films the parallel potential energy in Eq. (9) cannot be treated as a perturbation, and that its contribution to the binding energy may be significant. See the remarks at the end of Sec. IV and in the conclusion. A large surface effective mass could have extremely important effects for surface rate processes.¹⁸

Following Pines,¹⁶ we can use our perturbation theory results to try to identify the coupling constant for the H-rippion system. To this end we calculate \bar{N} , the average number of virtual ripples which follow the physisorbed particle:

$$\begin{aligned} \bar{N} &= \left\langle \sum_k a_k^\dagger a_k \right\rangle \\ &= \sum_k \frac{E(k)^2}{\left[\hbar\omega_k + \frac{\hbar^2 k^2}{2m_p} - \vec{p} \cdot \hbar\vec{k} \right]^2}. \end{aligned} \quad (61)$$

Thus, in the delocalized limit for a slow particle, we find

$$\bar{N} = \left[\frac{\pi}{2} \right]^{1/2} \left[\frac{|U_0|}{\hbar C_3 \gamma^{1/2}} \right] + O((\gamma a^2)^{3/2}), \quad (62)$$

a measure of the degree of localization.

We have consistently described the physisorbed atom as a "slow" particle. Let us consider this for the case of a well-localized particle; then, we may simply ask whether the large k modes of the film can keep up with the particle. Using the ST dispersion relation $\omega^2 = \sigma(ka)^3/m_4$, we have

$$v(k) = \frac{3}{2} \left[\frac{\sigma a^{2'}}{m_4} \right]^{1/2} (ka)^{1/2}, \quad (63)$$

which is to be compared to the particle velocity

$$v_p(k) = \frac{\hbar k}{m_p^*} = \frac{\hbar}{m_p^* a} (ka). \quad (64)$$

The velocities in Eqs. (63) and (64) are plotted as functions of ka in Fig. 5. (For the purposes of this figure, we have taken $m_p^* = m_p$.) The figure indicates that the particle velocity will be less than the surface mode velocity for all wave vectors of interest.

B. The particle-particle interaction

We now consider the case of N_p particles adsorbed to the helium surface. In the dilute particle limit, $N_p a^2/A \ll 1$, we may write the distribution function $P(\vec{\rho})$ as

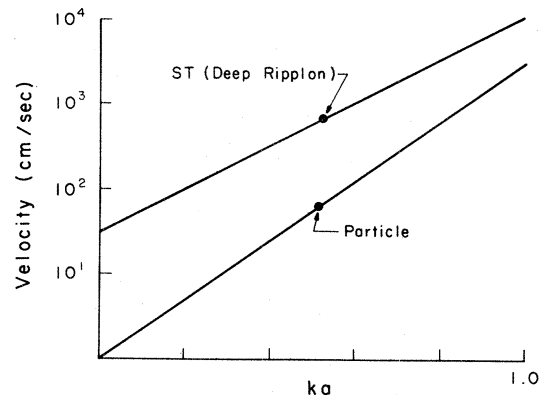


FIG. 5. Velocities. The particle velocity and the velocity of the short-wavelength modes are shown as a function of ka . If the particle is well localized it drives the short-wavelength modes to form the distortion of the surface. These modes are faster than the particle and will not be left behind as it moves.

$$P(\vec{\rho}) = \sum_{i=1}^{N_p} P_i(\vec{\rho})$$

$$\equiv \sum_{i=1}^{N_p} |\theta(\vec{\rho} - \vec{R}_i)|^2, \quad (65)$$

where the i th particle is located at lateral position \vec{R}_i . If we now proceed as we did above in the case of a single ad atom, then the function $E(k)$ introduced in Eq. (35) becomes

$$E(k) \equiv \sum_{i=1}^{N_p} E_i(k)$$

$$= \left[\frac{d}{a} \right] C(d) \sum_{i=1}^{N_p} e^{i\vec{k} \cdot \vec{R}_i} F_i(-k), \quad (66)$$

where

$$F_i(k) \equiv V_{||}(k) P_i(k), \quad (67)$$

and $V_{||}(k)$ and $P_i(k)$ are defined by

$$\langle V(\vec{\rho} - \vec{\rho}') \rangle_{||} = \frac{a^2}{A} \sum_k V_{||}(k) e^{-i\vec{k} \cdot (\vec{\rho} - \vec{\rho}')}, \quad (68)$$

$$P_i(\vec{\rho}) = \frac{1}{A} \sum_k P_i(k) e^{-i\vec{k} \cdot (\vec{\rho} - \vec{R}_i)}. \quad (69)$$

The potential $U[\theta]$ can be written in terms of $E(k)$ precisely as was done in Eq. (38):

$$U[\theta] = - \sum_k E(k) \frac{1}{\hbar\omega_k} E(-K). \quad (70)$$

We can now substitute Eq. (66) into (70) and separate the diagonal from the nondiagonal terms. Thus,

$$U[\theta] = N_p U_0 + U_{\text{eff}}(\vec{R}_1 \cdots \vec{R}_{N_p}), \quad (71)$$

where U_0 the diagonal piece is the same function as Eq. (38) and U_{eff} the nondiagonal piece can be written

$$U_{\text{eff}}(\vec{R}_1 \cdots \vec{R}_N) = - \sum_{i,j} \sum_k e^{-i\vec{k} \cdot (\vec{R}_i - \vec{R}_j)} E_i(k) \frac{1}{\hbar\omega_k} E_j(-k). \quad (72)$$

U_{eff} is a particle-particle interaction mediated by the helium surface deformations (i.e., the "mattress" effect).

In order to examine the qualitative features of U_{eff} , we shall specialize to the case of two ad atoms and the particle-film contact interaction as introduced above. Then

$$V_{||}(k) = V_0 a |\phi(d)|^2, \quad (73)$$

and

$$U_{\text{eff}}(R) = - \left[\frac{d}{a} \right]^2 (V_0 a |\phi(d)|^2)^2 \frac{1}{(2Nm_4 C_3^2)} \sum_k \cos(\vec{k} \cdot \vec{R}) P(k)^2, \quad (74)$$

where $\vec{R} = \vec{R}_2 - \vec{R}_1$ is the distance between particles and we use the same $P(k)$ for each particle [i.e., the same γ as in Eq. (43)]. Converting the summation over k into an integral, Eq. (74) can be written

$$U_{\text{eff}}(R) = (U_0/\pi\gamma) \int d\vec{k} \cos(\vec{k} \cdot \vec{R}) P(k)^2. \quad (75)$$

(Note, the quantity in parenthesis is independent of γ .) We now consider U_{eff} in the localized and delocalized limits.

In the localized limit $P(k) \sim 1$ for $ka < 1$. This condition can be introduced into Eq. (75) by setting $P(k) = 1$ along with a momentum cutoff at $k = \bar{k}$, $\bar{k}a \approx 1$. Then

$$U_{\text{eff}}(R) = (2U_0/\gamma) \frac{\bar{k}}{R} J_1(\bar{k}R). \quad (76)$$

For large $\bar{k}R$, this function decays algebraically

like $R^{-3/2}$, since

$$J_1(\bar{k}R) \sim (2/\pi\bar{k}R)^{1/2} \cos(\bar{k}R - 3\pi/4).$$

(The oscillation in sign is a consequence of the sharp momentum cut off.) In a thick film we should take account of the ST contribution to the dispersion relation [see Eq. (27)]. Thus, we may substitute

$$\omega^2 = C_3^2 \frac{k}{d} (1 + k^2 l d),$$

where

$$l \equiv \left[\frac{\sigma a^2}{m_4 C_3^2} \right] a, \quad (77)$$

into Eq. (72) and omit the momentum cutoff. This yields

$$U_{\text{eff}}(R) = (2U_0/\gamma) \frac{1}{dl} K_0(R/\sqrt{dl}), \quad (78)$$

where K_0 is a modified Bessel function. For large separation distances,

$$K_0(z) \sim \sqrt{\pi/(2z)} \exp(-z),$$

i.e., the effective interaction is short ranged. For particles that are close to one another, $R/\sqrt{ld} \ll 1$, $U_{\text{eff}}(R) \propto \ln R$.

In the delocalized limit, we can return to the Gaussian model which has $P(k) = \exp(-k^2/4\gamma)$. Thus,

$$U_{\text{eff}}(R) = 2U_0 \exp(-\frac{1}{2}\gamma R^2), \quad (79)$$

which for $\gamma R^2 \ll 1$ is an oscillator potential. [This model could also be applied to the localized case where the Gaussian would play the role of a smooth momentum cutoff. Then in contrast to Eq. (76) U_{eff} is very short ranged.]

We note, in passing, that in the limit of a semi-infinite film $l \rightarrow \infty$ since $C_3 \rightarrow 0$ [see Eq. (77)] and thus the surface would seem to become unstable. This unphysical result has been remarked on by Widom¹⁹ and discussed in detail by Cole.²⁰ This behavior is of interest for the thermodynamics of surfaces but is not of importance for the system we are studying. It should also be stressed that a crucial element in determining the form of the effective interaction is the k dependence of the amplitude $C(k)$ [see Eq. (28)] which couples the surface excitations to surface displacements. The $C(k)$ do not go like $k^{-1/2}$ as do phonons and so we do not obtain a $\ln R$ interaction.

VI. CONCLUSION

In this paper, we have described qualitatively the consequences of coupling a $H\downarrow$ atom to the modes of a ^4He film. The modes of the film (ripplons) were discussed in some detail with particular attention paid to how the restoring forces and the relative depth of the film influence the dispersion relation $\omega(k)$ [see Eq. (26)] and the ripplon amplitude $C(k)$ [see Eq. (28)]. It is clear from the results of Sec. V that the characteristics and behavior of the adsorbed system depend importantly on the form of $\omega(k)$ and $C(k)$.

If a $H\downarrow$ atom couples strongly enough to the ^4He film, parallel localization can occur: i.e., a $H\downarrow$ polaron is formed. The strength of the interaction that makes a localized state stable U_0 [see Eq. (39)], depends upon the (correlated) interaction of the $H\downarrow$ atom with the surface together with the elasticity of the surface. Thus, the $H\downarrow$ atom is attracted toward the surface by the van der Waals

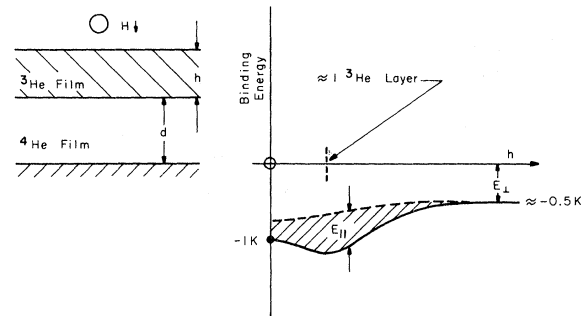


FIG. 6. Binding energy as a function of ^3He thickness. A ^3He layer, riding on top of a ^4He film, can respond to the long-wavelength modes of the ^4He film; it cannot respond to the short-wavelength modes. Thus, to the degree that the parallel binding of the $H\downarrow$ is due to the short-wavelength modes in the film, this component of the binding will go to zero as the film is thickened.

force between it and the film $V_{\parallel}(k) \rightarrow V_0 a |\phi(d)|^2$ (for the contact potential model). This “potential” depends explicitly on $|\phi(z)|^2$ and couples the parallel and perpendicular problem (in this same manner, of course, V_{\perp} depends explicitly on $|\phi(\rho)|^2$ [c.f., Eq. (10)]). Thus, the parallel localization problem and the perpendicular binding problem are coupled and interdependent. These two problems may be taken to be approximately independent if the energies involved are very different (e.g., $E_{\perp} \gg E_{\parallel}$). Our estimates indicate that it is possible that the energies involved are not very dissimilar and so this separation is not valid. A measurement of the $H\downarrow$ binding energy as a function of film thickness and composition might be very illuminating in this respect. (Indeed, if polaron production is occurring, the surface recombination rate might first decrease with decreasing film thickness.) An alternative way to alter the surface modes and thus probe for the existence of polarons is to add ^3He to the film.^{15,21} The addition of a ^3He component tends to soften the surface modes so that one might expect an increased tendency towards parallel binding. However, although the (normal) ^3He film will support $k > 0$ long-wavelength modes, it probably will not support the mobile large wave-vector modes that represent a distortion of the upper ^3He surface. Thus, a thick ^3He film above ^4He may bind a $H\downarrow$ atom only in the perpendicular direction (see Fig. 6).

ACKNOWLEDGMENT

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