

# Quantum Sticking, Scattering, and Transmission of $^4\text{He}$ atoms from Superfluid $^4\text{He}$ Surfaces

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We develop a microscopic theory of the scattering, transmission, and sticking of  $^4\text{He}$  atoms impinging on a superfluid  $^4\text{He}$  slab at near normal incidence, and inelastic neutron scattering from the slab. The theory includes coupling between different modes and allows for inelastic processes. We find a number of essential aspects that must be observed in a physically meaningful and reliable theory of atom transmission and scattering; all are connected with multiparticle scattering, particularly the possibility of energy loss. These processes are (a) the coupling to low-lying (surface) excitations (ripples/third sound) which is manifested in a finite imaginary part of the self-energy, and (b) the reduction of the strength of the excitation in the maxon/roton region. [S0031-9007(98)05555-0]

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The dynamics of liquid  $^4\text{He}$  films and the bulk fluid near its free surface continues to be of considerable interest. Experimental information is available pertaining to the scattering of helium atoms from helium surfaces and films [1–5]; from the dynamics of localized excitations within the fluid, including excitation scattering from the surface and quantum evaporation [5–8]; and from inelastic neutron scattering at grazing angles from adsorbed films [9–12]. Moreover, information concerning the condensate fraction in helium may be obtainable directly from elastic transmission of  $^4\text{He}$  atoms through superfluid  $^4\text{He}$  slabs [13]. The fact that helium slabs have been made in the laboratory [14] leads to the prospect that the dynamic probes previously applied to adsorbed films and bulk surfaces of helium can now be applied to slabs. The slabs should produce simpler and thus easier to interpret results than the adsorbed films.

We report here on the results of a manifestly microscopic theoretical analysis of the dynamics of such a slab at zero temperature. We find that atom scattering processes are dominated by multiparticle events, particularly the coupling to ripples. This is in qualitative agreement with the conclusions of Edwards and collaborators [1–4] in their results for the related helium atom scattering from the free surface of bulk helium. Our results not only provide insight into the transmission and sticking of a helium beam that was not previously available from theory or experiment, it also provides detailed predictions for the anticipated experiments on slabs.

The theoretical method used here has been successfully applied to the bulk [15,16], to adsorbed films [17,18], and to droplets [19]; in the latter two it was successful in making detailed and correct predictions about surface states (ripples and third sound) which also play a very important role in the slab geometry. The theory is adapted here to the slab geometry, including scattering states. We outline our theory and present some illuminating results

for waves at normal incidence which also provide insights into quantum evaporation.

We consider the ground state of a slab of superfluid  $^4\text{He}$  of particle number  $1.5 \text{ \AA}^{-2}$ , corresponding to a thickness of approximately  $80 \text{ \AA}$ , and the dynamic structure function that would be measured for neutron momentum loss perpendicular to the slab. The density profile of this slab in the ground state is shown in Fig. 1. The relevant excited states may be written as

$$\Psi_\lambda(\mathbf{r}_1, \dots, \mathbf{r}_N) = F_\lambda(\mathbf{r}_1, \dots, \mathbf{r}_N)\Psi_0(\mathbf{r}_1, \dots, \mathbf{r}_N), \quad (1)$$

where  $\Psi_0$  is the ground state of the slab or an appropriately optimized representation of the ground state,  $F_\lambda$  is a complex *excitation operator*, and  $\lambda$  represents the quantum numbers for these excited states. The excitation energy for these states is given by

$$\epsilon_\lambda = \frac{\langle \Psi_\lambda | H - E_0 | \Psi_\lambda \rangle}{\langle \Psi_\lambda | \Psi_\lambda \rangle} = \frac{\sum_{i=1}^N \langle \Psi_0 | \frac{\hbar^2}{2m} |\nabla_i F_\lambda|^2 | \Psi_0 \rangle}{\langle \Psi_\lambda | \Psi_\lambda \rangle}. \quad (2)$$

The functions  $F_\lambda$  are solutions of an effective Schrödinger equation obtained by functionally minimizing this excitation energy with respect to  $F$  [20], by using correlated

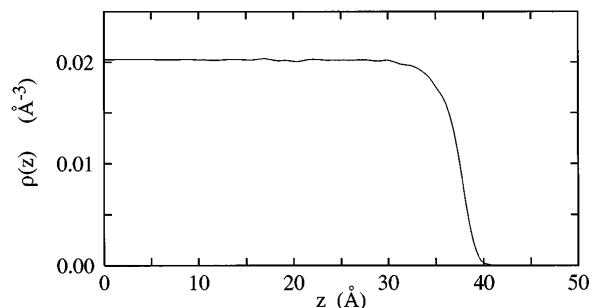


FIG. 1. The density profile of the  $^4\text{He}$  slab used here. The profile is symmetric around  $z = 0$ .

basis function (CBF) perturbation theory [15], or by extremizing the action [16,17].

It is well known from the theory of the phonon-roton spectrum of bulk liquid  $^4\text{He}$  that these low excited states are quantitatively accounted for by a wave function of this type containing one- and two-body terms in  $F_\lambda$  [15–18]:

$$F = \sum_{i=1}^N f_1(\mathbf{r}_i) + \sum_{i<j=1}^N f_2(\mathbf{r}_i, \mathbf{r}_j). \quad (3)$$

Retaining only  $f_1$  in the bulk case produces the familiar Bijl-Feynman spectrum  $\epsilon_{\mathbf{k}} = \hbar^2 k^2 / [2mS(k)]$ , where  $k$  is the wave number for the bulk excitation and  $S(k)$  is the zero temperature x-ray structure factor. This excitation energy is quantitatively correct at long wavelengths and qualitatively correct in the maxon-roton regime (cf. Fig. 2). Including  $f_2$  is sufficient to correct most of the residual disagreement with experiment [15,16,18].

The work reported herein is a further adaptation of the above procedure for studying transmission, reflection and sticking of incident particles. This is achieved by solving our equations with the boundary condition that there is an incoming particle beam of specified energy and unit incoming flux, and an outgoing particle beam. Far from the slab the particle is a plane wave with the wave number determined by the energy and the direction of propagation.

For simplicity, in this Letter we focus on the elastic transmission and reflection of states with normal incidence. Thus we may describe some of our results in terms of reflection and transmission amplitudes  $R$  and  $T$ , respectively. At first glance we appear to be describing a one-dimensional quantum mechanical scattering problem with the helium slab serving as a well or barrier, but the actual situation is far richer: Since this scattering “well”

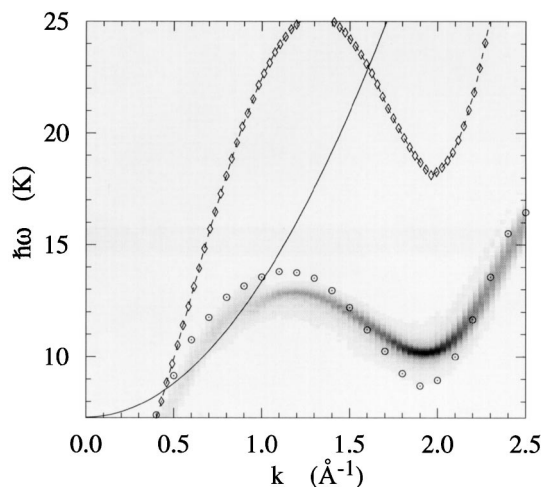


FIG. 2. A grey-scale map of the dynamic structure function  $S(k_\perp, \omega)$  for momenta  $k_\perp$  perpendicular to the slab, and energies above the evaporation barrier  $\hbar\omega > -\mu$ . Also shown are the experimental phonon-roton spectrum [21], the Feynman spectrum  $\hbar^2 k^2 / [2mS(k)]$  (dashed line with diamonds), and the kinetic energy of the incoming particle  $-\mu + \hbar^2 k^2 / 2m$  (solid line).

is composed of helium atoms, this is a generically non-local problem when viewed at the one-body level. The Bose exchange symmetry of the incoming atoms with those in the slab requires a full symmetrization, as one sees in the summations in Eq. (3). Moreover, the well is dynamic: The incoming particle may produce excited states, corresponding to inelastic processes, which may result in the capture of the particle and/or the emission of particles in states other than the elastic channel; this includes multiexcitation channels in which the individual states carry momentum parallel to the surface. Of particular importance are the effects of surface states, where the incoming particle may stick, and the structure of the surface, which can disperse the incoming particle into a number of states of quasimomentum differing significantly from the momentum of the incoming particle.

Since the states that we are exploring will also be excited by inelastic neutron scattering, it is useful to first consider the dynamic structure factor  $S(\mathbf{k}, \omega)$  that would be measured by neutrons scattered with momentum change  $\hbar\mathbf{k}$ . This is obtained theoretically by using linear response theory to obtain the density-density response function  $\chi(\mathbf{k}, \omega)$  together with the relation  $S(\mathbf{k}, \omega) = -\text{Im}[\chi(\mathbf{k}, \omega)/\pi]$ . In the case of the bulk superfluid at low temperatures,  $S(k, \omega)$  has a very sharp spectrum that maps out the bulk phonon-roton excitation energy as well as broad multiexcitation strength at higher energies. Neutron scattering from adsorbed helium films produces a similar phonon-roton structure when studied as a function of the parallel momentum transfer  $\hbar\mathbf{k}_\parallel$  in grazing angle scattering [18]. However, the layered structure of these films broadens this phonon-roton structure and produces surface and layer modes which are also detected in the neutron scattering, but which complicate the analysis [18] and interpretation of the data. There is no layering in the slab (cf. Fig. 1); thus the broadening of the phonon-roton structure is significantly reduced for grazing angle scattering. Nevertheless, surface modes are still present and would be observed similarly to the adsorbed film system. However, if there is significant momentum transfer perpendicular to the surface, one would expect significant surface effects on  $S(\mathbf{k}; \omega)$  particularly for relatively thin slabs. Nonetheless it can be seen from Fig. 2 that the calculated structure in  $S$  for our 80 Å slab has substantial strength in the vicinity of the bulk phonon-roton spectrum for perpendicular momentum transfer  $\mathbf{k}_\perp$ , though it is noticeably broadened and weakened. The fact that  $S(\mathbf{k}_\perp, \omega)$  is effectively parametrizable in terms of  $\mathbf{k}_\perp$  should not be interpreted to indicate that this is a good quantum number. Nevertheless it is clear from the figure that it is useful to use  $\mathbf{k}_\perp$  to approximately classify these modes.

To examine the propagation of a helium atom normally incident at energy  $\hbar\omega$ , we first exhibit the wave equation satisfied by the one-body part of the excitation function  $F$ . Defining the auxiliary function  $\psi(\mathbf{r}) = f_1(\mathbf{r})/\sqrt{\rho_1(\mathbf{r})}$ , the equation for  $\psi$  is:

$$\frac{\hbar^2}{2m} \left\{ -\nabla^2 + \frac{\nabla^2 \sqrt{\rho_1(\mathbf{r})}}{\sqrt{\rho_1(\mathbf{r})}} \right\} \psi(\mathbf{r}) + \int \Sigma(\mathbf{r}, \mathbf{r}'; \omega) \psi(\mathbf{r}') d^3 r' = \hbar \omega \left\{ \psi(\mathbf{r}) + \int d^3 r' [g_2(\mathbf{r}, \mathbf{r}') - 1] \sqrt{\rho_1(\mathbf{r}) \rho_1(\mathbf{r}')} \psi(\mathbf{r}') \right\}, \quad (4)$$

where  $\rho_1$  and  $g_2$  are the density and pair distribution of the ground state, and  $\Sigma(\mathbf{r}, \mathbf{r}'; \omega)$  is the self-energy. This equation has the appearance of a one-body Schrödinger equation with a nonlocal, non-Hermitian “optical potential,” which has its origin in the fact that this is a many-body system. The derivation and the approximations we use to calculate the self-energy may be found in Ref. [18]. In our approximation, it has the form

$$\Sigma(\mathbf{r}, \mathbf{r}'; \omega) = -\frac{1}{2} \sum_{mn} \frac{V_{mn}(\mathbf{r}) V_{mn}(\mathbf{r}')}{\hbar(\omega_m + \omega_n - \omega) - i\eta}, \quad (5)$$

where the  $V_{mn}(\mathbf{r})$  are three-phonon vertex functions derived in Ref. [18], and  $\omega_m$  are the excitation energies of the background; note that the state sums go over both phonon- and ripplon-type excitations and include, in particular, all parallel momenta. This feature is manifested in the energy denominator which can cause the self-energy to be complex.

The excited states consist of bound and continuum states. The scattering states of a  $^4\text{He}$  atom from the slab are continuum states. Thus Eq. (4) is solved subject to the boundary conditions

$$\psi(\mathbf{r}) = \begin{cases} e^{ikz} + R e^{-ikz} & \text{for } z \rightarrow -\infty \\ T e^{ikz} & \text{for } z \rightarrow \infty \end{cases}, \quad (6)$$

where  $k$  is the positive root of  $\omega = \hbar k^2/2m$ .

We have carried out calculations including both the full, complex self-energy as well as the simpler Feynman approximation. The latter is equivalent to setting  $f_2(\mathbf{r}, \mathbf{r}') = 0$  in the definition of the excitation factor  $F$ , which gives  $\Sigma(\mathbf{r}, \mathbf{r}'; \omega) = 0$  in Eq. (4), reducing the equation to the one used by Edwards and Fatouros [2] to describe scattering from the surface of the bulk liquid. At that level, the stationary scattering states have the unphysical property that the elastic single-particle flux is conserved, i.e.,  $|R|^2 + |T|^2 = 1$ .

Our results for  $|R|$  and  $|T|$  are summarized in Fig. 3. It is seen in the middle panel of Fig. 3 that the reflection coefficient  $|R|$  undergoes oscillations similar to those seen in scattering from wells and barriers in one-dimensional, one-body quantum mechanics. The details are of course different due to the fact that the effective dispersion relation inside the slab is quite different from the free-particle spectrum, as can be seen in Fig. 2.

The self-energy has dramatic effects on the results:

The *real part* of the self-energy comes from virtual processes that dress the Feynman states by allowing for fluctuations of the short-ranged structure of the system. The consequence is a significant improvement of the single excitation energies, as seen in Fig. 2.

The *imaginary part* of the self-energy comes from the existence of multiexcitation states with total energy

equal to the single-excitation state. Thus a single particle impinging on the surface has channels for decaying into these multiexcitation states. [In our nomenclature, a multiexcitation state is one in which the excitation function  $F$  is primarily a product of single excitation  $F$  factors. Thus, e.g., a two-excitation state would be characterized by a dominant term in Eq. (3) of the form  $f_{2,\omega}(\mathbf{r}, \mathbf{r}') = f_{1,\omega_a}(\mathbf{r}) f_{1,\omega_b}(\mathbf{r}') + f_{1,\omega_b}(\mathbf{r}) f_{1,\omega_a}(\mathbf{r}')$ , where  $\omega = \omega_a + \omega_b + O(N^{-1})$ .] One consequence of this is that the incoming atom can “stick” in the slab by decaying to two or more bound states. Similarly, it can decay into bound states and an emitted particle of lower energy. This leads to a significant reduction of  $|R|^2 + |T|^2 < 1$  as is seen in the top panel of Fig. 3. The difference  $1 - [|R|^2 + |T|^2]$  is a measure of the atomic sticking plus real inelastic scattering and transmission.

The change in the transmission coefficient is the most dramatic effect of allowing for decay processes: Hermitian approximations for the self-energy all lead to the feature  $|R|^2 + |T|^2 = 1$ . In this aspect, our predictions differ dramatically from those of the Feynman approximation as well as a more recent attempt [22] to study quantum evaporation within time-dependent density functional theory. Our conclusions agree, however, with those of Edwards and Fatouros [2] when damping of reasonable magnitude is included.

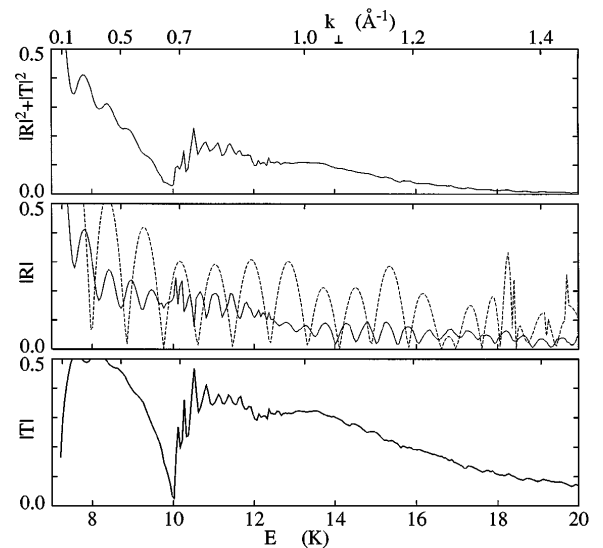


FIG. 3. The absolute value of the transmission and reflection coefficients,  $T$  (lowest figure),  $R$  (middle figure), and the intensity loss  $|R|^2 + |T|^2$  (top figure) is shown as obtained from the CBF calculation (solid lines). We also show the Feynman approximation for  $|R|$  (middle figure, dashed line); note that  $|R|^2 + |T|^2 = 1$  in that approximation.

Another interesting feature seen in Fig. 3 is that the transmission coefficient has notable structure at and above the roton energy which is largely missing from the reflection coefficient. This structure is in part due to the high density of states of the roton, and to the fact that the phonon-roton spectrum is nonmonotonic in this region, giving rise to degeneracies for energies between the roton and the maxon. Moreover the absence of translational invariance in the  $z$  direction results in the incoming plane wave hybridizing with the degenerate states in the slab. This is a surface effect, and would continue to exist for thick films and for the surface of the bulk.

An examination of the wave functions and the structure of the self-energy show that most of the sticking occurs in the surface of incidence, with a further reduction of amplitude at the back surface. By turning off the surface state contributions to the imaginary part of the self-energy (ripples/third sound and other states localized in the surface), we have seen that the main contribution to this surface sticking comes from these surface states.

In conclusion, our adaptation of the microscopic theory of excited states in inhomogeneous liquid  $^4\text{He}$  to describe quantum sticking, scattering and transmission of  $^4\text{He}$  atoms gives a clear picture of the many-body physics of the interaction of a beam of  $^4\text{He}$  atoms with a liquid helium surface. However, the existence of a second surface, at approximately  $80 \text{ \AA}$  behind the first, shown in the present paper, makes a direct comparison to scattering from the free surface of bulk liquid  $^4\text{He}$  ambiguous. A significant fraction of the scattering from the slab occurs at the second surface, contributing some signal to the reflection by backpropagation. Some of the sticking also happens at the second surface. The remaining elastic transmission into the vacuum can also be viewed as quantum evaporation, as can be seen by examining the propagation of localized wave packets, wherein excitations in the interior of the slab, produced by the incoming wave packet at the first surface, propagate to the second surface where helium atoms are evaporated.

The main approximation in this work is that only decay into two-excitation states is included. Opening other inelastic channels would further reduce the amplitude of the elastically scattered and transmitted atoms.

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