Reflection of hydrogen atoms from the surface of superfluid helium

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(Received 6 April 1989; revised manuscript received 21 August 1989)

We propose a new method for studying the reflection of a hydrogen atom from a superfluid-helium film. Starting from the narrow width of the reflected angular distribution recently found experimentally, we tentatively extrapolate to the extreme limit of low ripplon wave numbers in which the adiabatic or degenerate-internal-states approximation becomes valid. We obtain simple closed expressions for single- and multiple-ripplon processes, which do not require the integration of a Schrödinger equation for their evaluation and do not depend on the specific form of the potential.

I. INTRODUCTION

In almost all experiments dealing with spin-polarized atomic hydrogen at low temperatures,[1,2] the interaction of H atoms with surfaces of liquid helium plays a crucial role. The density of adsorbed gas, the hyperfine wall shift, the thermal accommodation and sticking coefficients are examples of experimental quantities that play a decisive role in recently developed ideas about, for instance, the achievement of Bose-Einstein condensation,[3] the operation of the cryogenic H maser,[4] and attempts to detect surface spin waves.[5] Even in experiments where contact with material walls is avoided by means of a static-magnetic-field minimum in free space,[6,7] contact with superfluid helium is unavoidable in the course of the transport of the hydrogen gas to the trapping region.

In view of the importance of atom-surface processes, a completely new category of experiments is now being prepared or already carried out, with the purpose of an angle-resolved measurement of inelastic scattering. The purpose of the present paper is to propose a new approach for analyzing such experiments in terms of ripplon creation and annihilation. The conventional approach[8–10] is based on perturbation theory in the deviation from a flat surface. It allows the calculation of the single-ripplon creation and annihilation in first order and the calculation of multiple-ripplon processes, with rapidly increasing effort, in higher orders. The starting point of our investigation was a prominent feature of the experimental data available at that time that involved normal incidence of a beam of cold ($T=0.1$–0.4 K) hydrogen atoms: The inelastic angular distribution turned out to be concentrated in a very small angle of less than 1° around the surface normal, but nevertheless it was unambiguously distinguishable from specular reflection.[11] On the basis of this the total momentum of the ripplon(s) involved in an inelastic-scattering event is low, i.e., at most $10^{-3}a_0^{-1}$. In this paper we speculate on the possibility that the wavelengths of the individual ripplons are so long and the total ripplon energy is so low that some drastic approximations can be made. The validity of the approximations can only be tested by comparison with experiment or by full coupled-channel calculations. From a theoretical point of view it is satisfactory that the sharply peaked result for the angular distribution obtained in the following is at least not in contradiction with our starting point.

The first approximation is the following. If only the very slow (small frequency ω_q) ripplon modes are involved, it is natural to introduce the adiabatic approximation or its variant, the degenerate-internal-states approximation. With some qualifications to be mentioned in the following, the classical picture is that the atom collides elastically with an instantaneous-"frozen" surface, whereas the surface starts to react dynamically after the collision.

The second approximation is based on the above-mentioned long ripplon wavelengths: We assume that the atom-surface potential for the frozen surface is equal to the flat-surface potential, but displaced according to the local height of the surface.

As we shall see, the combination of these two approximations allows us to give simple analytical expressions for the reflected atomic angular distribution connected with single-ripplon and multiple-ripplon processes. Contrary to the conventional formalism, these expressions do not contain the specific atom-surface potential; this indicates that inelastic scattering depends primarily on the surface dynamics and is rather insensitive to the potential. In an early stage of our investigation we were informed[12] that preliminary results obtained on the basis of the conventional analysis along the lines in Refs. 8–10 confirmed very accurately (to less than 1% difference) the potential independence implied by our method.

II. METHOD

Consider an H atom incident from the $z>0$ half space upon a ^{4}He surface. The eigenvalue equation with which this system is commonly described is

$$\left[-\frac{\hbar^2}{2m_{\mathrm{H}}}\Delta+H_{\mathrm{surf}}(h,\partial/\partial h)+V(\mathbf{r},h)\right]\Psi(\mathbf{r},h) = E\Psi(\mathbf{r},h)\ . \quad (1)$$

The dynamical modes of the surface are described by the surface Hamiltonian H_{surf}. The elementary excitations

are ripplons, associated with fluctuations in film height $h(\rho)\equiv h(x,y)$:

$$h(\rho)=\frac{1}{\sqrt{A}}\sum_{\mathbf{q}} h_{\mathbf{q}} e^{i\mathbf{q}\cdot\rho} , \tag{2}$$

where

$$h_{\mathbf{q}}=\left[\frac{\hbar q \tanh(qD)}{2\rho\omega_q}\right]^{1/2}(r_{\mathbf{q}}^{\dagger}+r_{-\mathbf{q}}) . \tag{3}$$

We use periodic boundary conditions in the x-y plane with period L for both the atom and the surface. The associated area is $A=L^2$. Furthermore, ρ is the bulk ^{4}He density and D is the film thickness, while $r_{\mathbf{q}}^{\dagger}$ and $r_{\mathbf{q}}$ are the creation and annihilation operators for a ripplon. In Eq. (1) h is a short-hand notation for the collection $\{h_{\mathbf{q}}\}$ for all wave vectors.

We are looking for scattering solutions of Eq. (1) having the asymptotic form

$$\Psi(\mathbf{r},h)\underset{z\to\infty}{\sim} e^{i\mathbf{k}_i\cdot\mathbf{r}}\phi_i(h)+\text{scattered waves} \tag{4}$$

for atomic positions $\mathbf{r}$ far from the surface. ϕ_i is the initial surface eigenstate. The Schrödinger equation can be turned into a coupled-channels problem by expanding Ψ in terms of basis functions with z-dependent coefficients:

$$\Psi(\mathbf{r},h)=\sum_j u_j(z)e^{i\boldsymbol{\kappa}_j\cdot\rho}\phi_j(h) . \tag{5}$$

Here and in the following we use the notation $\mathbf{k}_j=(\boldsymbol{\kappa}_j,k_{jz})$ and $\boldsymbol{\kappa}_j=\boldsymbol{\kappa}_i+\mathbf{Q}_i-\mathbf{Q}_j$, $\mathbf{Q}_j$ representing the total momentum of ripplons present in the surface eigenstate ϕ_j.

Instead of solving Eq. (1) as a coupled-channel problem, we assume that the process of inelastic scattering can be described satisfactorily on the basis of the adiabatic approximation. This corresponds to the assumption that the energy differences of all surface eigenstates significantly coupled in the collision are negligible. Their energy eigenvalues can then be replaced by a common value ε. The surface Hamiltonian is thus effectively a constant multipled by the unit operator and we may turn to a new basis relative to which h, and consequently the atom-surface interaction, is diagonal. In the theory of the excitation of surface waves in atomic nuclei by nuclear reactions this is referred to as the adiabatic approximation.[13] We have shown in recent papers[14,15] that it can also be applied with considerable success to spin exchange and dipolar relaxation by H+H collisions in the subkelvin regime. It proved to be excellent even in the zero-temperature limit, provided some adjustments are introduced in the spirit of multichannel-quantum-defect theory, which implies that the replacement of internal eigenvalues is only made in the limited spatial region where the channel coupling occurs. The results of our previous extensive calculations[14,16] for H+H inelastic scattering could thus be found via much simpler closed formulas containing only singlet and triplet phase shifts, obtainable from uncoupled radial equations. On the basis of this experience we recently applied the degenerate-internal-states (DIS's) approximation to the scattering of dressed H atoms[17] in a microwave trap. In this paper we extend it to inelastic scattering of an H atom from a superfluid-helium surface.

On the basis of the adiabatic approximation the solution of Eqs. (1) and (4) is

$$\Psi(\mathbf{r},h)=\psi_{\mathbf{k}_i^0}(\mathbf{r};h)\phi_i(h) , \tag{6}$$

where the ψ function is the frozen-surface atomic-scattering wave function with the plane-wave part $\exp(i\mathbf{k}_i^0\cdot\mathbf{r})$, which depends parametrically on h and satisfies

$$\left[-\frac{\hbar^2}{2m_H}\Delta+V(\mathbf{r},h)\right]\psi_{\mathbf{k}_i^0}(\mathbf{r};h)=(E-\varepsilon)\psi_{\mathbf{k}_i^0}(\mathbf{r};h) . \tag{7}$$

Equivalently, expanding the product wave function (6) as in Eq. (5) with coefficients given by

$$u_j(z)e^{i\boldsymbol{\kappa}_j\cdot\rho}\equiv\langle\phi_j|\psi_{\mathbf{k}_i^0}(\mathbf{r};h)|\phi_i\rangle , \tag{8}$$

the set of functions $u_j(z)$ satisfies the above-mentioned coupled-channels equations in the case of degenerate surface states. For the sake of generality we allow $\mathbf{k}_i^0$ to be different from $\mathbf{k}_i$, corresponding to a more general choice for ε. The functions $u_j(z)$ have the asymptotic form

$$u_j(z)\underset{z\to\infty}{\sim}\delta_{ji}e^{-ik_{jz}^0 z}-S_{ji}e^{ik_{jz}^0 z} . \tag{9}$$

The S-matrix element S_{ji} is the adiabatic approximation to the exact S_{ji} determining the asymptotic form of the coupled solutions $u_j(z)$ for nondegenerate surface states. In the DIS's approximation one would first extract $\sqrt{k_{iz}}$ and $\sqrt{k_{jz}}$ before approximating the remaining coefficient by the adiabatic value. Such factors also play a role in the $\sqrt{T}$ decrease of the sticking probability at low temperature and are determined by the behavior of the "on-the-energy-shell" S_{ji} value close to the i and j thresholds, i.e., for small k_{iz} and k_{jz}. The DIS's assumption means essentially that the relevant ranges of wave numbers are so small that after extracting factors which would make $S_{ji}-\delta_{ji}$ equal to zero, the remaining part can be considered as a constant.

Substituting Eq. (9) in Eq. (5) we find for Ψ the asymptotic behavior

$$\Psi(\mathbf{r},h)\underset{z\to\infty}{\sim}e^{i\mathbf{k}_i^0\cdot\mathbf{r}}\phi_i(h)-\sum_j S_{ji}e^{i\mathbf{k}_j^0\cdot\mathbf{r}}\phi_j(h) . \tag{10}$$

Note that this expression takes into account the creation or annihilation of an arbitrary number of ripplons: ϕ_j represents an arbitrary surface eigenstate. From Eq. (8) it can be seen that we do not restrict ourselves to single-ripplon processes: The ψ function, when expanded in the field coordinates $h_{\mathbf{q}}$, also contains higher than first-order terms.

It is illustrated to give a more explicit expression for the adiabatic approximation to S_{ji}. Writing the asymptotic form of the frozen-surface scattering wave function as

$$\psi_{\mathbf{k}_i^0}(\mathbf{r};h)\underset{z\to\infty}{\sim}e^{i\mathbf{k}_i^0\cdot\mathbf{r}}+\sum_j f_{\mathbf{k}_i^0\to\mathbf{k}_j^0}(h)e^{i\mathbf{k}_j^0\cdot\mathbf{r}} , \tag{11}$$

we have

$$S_{ji} = -\langle \phi_j | f_{\mathbf{k}_i^0 \to \mathbf{k}_j^0}(h) | \phi_i \rangle \ . \tag{12}$$

Note that the surface reacts dynamically to the scattering process, although the atom is scattered by a frozen-surface shape. The classical picture is that the atom follows an orbit determined by the force from a static surface, while the surface starts to react to the sudden opposite force from the atom only after the collision.

A second closely related approximation we now introduce is based on the fact that the ripplon wavelengths involved are at least of the order $1000a_0$ and therefore large both relative to the amplitude of the surface fluctuations and relative to the range of the interatomic H-^{4}He interaction along the surface. On this basis we write the atom-surface interaction as

$$V(\mathbf{r},h) \simeq V_0(z - h(\boldsymbol{\rho})) \ , \tag{13}$$

where $V_0(z)$ is the potential due to a flat surface; i.e., the flat-surface potential is simply displaced by the surface height at the horizontal position of the atom. In addition, the frozen-surface scattering wave function now behaves as for a flat but displaced potential, i.e., all k_{jz}^0 are equal ($\equiv k$) and

$$\psi_{\mathbf{k}_i^0}(\mathbf{r};h) \simeq e^{-ikh} u_k(z-h) e^{i\boldsymbol{\kappa}_i \cdot \boldsymbol{\rho}} \ , \tag{14}$$

where $h = h(\boldsymbol{\rho})$ and $u_k(z)$ is the one-dimensional scattering wave function for $V_0(z)$. The factor e^{-ikh} ensures that the plane-wave part of ψ has its correct form $\exp(i\mathbf{k}_i^0 \cdot \mathbf{r})$.

Equation (14) may be used to expand ψ in "horizontal" momentum states. Outside the range of the potential, we then find the expansion (11) with

$$f_{\mathbf{k}_i^0 \to \mathbf{k}_j^0}(h) = -e^{2i\delta(k)} \frac{1}{A} \int d\boldsymbol{\rho} e^{i(\boldsymbol{\kappa}_i - \boldsymbol{\kappa}_j)\cdot\boldsymbol{\rho}} e^{-2ikh(\rho)} \ . \tag{15}$$

In this equation $e^{2i\delta(k)}$ is the reflection amplitude associated with the potential $V_0(z)$:

$$u_k(z) \underset{z \to \infty}{\sim} e^{-ikz} - e^{2i\delta(k)} e^{ikz} \ . \tag{16}$$

Note that in the present approach the scattering amplitude (15) and thus the S-matrix element (12) separate into a potential-dependent phase factor and a factor depending on the surface dynamics, i.e., the ripplon dispersion relation. The former drops out in calculating probabilities. This insensitivity to potential shape is one of our main conclusions.

It is of interest to compare the potential sensitivity of the reflection angular distribution discussed in this paper with that for the nonsticking part of the thermal accommodation coefficient calculated in Ref. 10. This nonsticking part a^{NS} is an integral over the same transition probability and was found to differ by about a factor of 2 for various choices of the adsorption potential. This might seem to contradict the weak potential sensitivity predicted here. Note, however, that the accommodation coefficient uses the energy transfer $\hbar\omega_q$ as a weight factor. In addition, contrary to the reflection angular distribution, which is a sum of the ripplon-creation and ripplon-annihilation terms, it is a difference of such contributions that is represented by the factor $e^{\Delta\beta\varepsilon_q} - 1$ in Goldman's expression for a^{NS}. This gives rise to a second $\hbar\omega_q$ factor. The quantity a^{NS} thus places greater weight on higher-energy transfers than the inelastic-scattering angular distribution that we study in this paper, so that deviations from the extreme low-q behavior are expected to be larger. In fact, in calculating a^{NS} by taking the $q \to 0$ limit of the transition matrix-element squared [cf. Eq. (17)], the Boltzmann integrations for perpendicular and parallel atomic momenta can be carried out analytically and we find

$$a^{\mathrm{NS}} = 2\zeta(3) m_{\mathrm{H}} k_B^2 T^2 / (3\pi\gamma_0 \hbar^2) = 1.94 \times 10^{-2} \ T^2/K^2 \ ,$$

in which $\gamma_0 = 3.78 \times 10^{-4} \ J/m^2$ is the ^{4}He surface tension. This is the expression obtained by Castaing and Papoular[18] in the limit where the H atom is treated as a classical particle, divided by $(\pi^4/30)/\zeta(3) = 2.70$.[19] It agrees reasonably with Goldman's results. In any case it may be used to estimate the fraction of a^{NS} which is due to the small range of q values $0 < q < q_{\max}$ corresponding to the 1° angle around specular reflection which interests us in connection with the reflection experiment. This fraction turns out to be

$$\gamma_0 \hbar^2 q_{\max}^3 / [4\zeta(3)\rho(k_B T)^2]$$

with ρ the ^{4}He density. Its order of magnitude varies from 10^{-6} to 10^{-5} in the experimental temperature range. Clearly, the potential-independent part of a^{NS} is predominantly determined by much higher q values. The potential-dependent part of the transition matrix element is of even higher order in q, so that the potential sensitivity of the low-q contribution to a^{NS} may be expected to be smaller than that exhibited by Goldman's results by a factor of at least 10^5. We conclude that a restriction of the q integral in the nonsticking part of the accommodation coefficient to our q range would reduce the potential dependence to the previously mentioned level of less than 1%.

The fact that our inelastic angular distribution does not depend on the potential enables us to give explicit expressions for the final results. As an example we work out Eq. (15) to first order in h for the case of normal incidence ($\boldsymbol{\kappa}_i = 0$). The DIS's probability for inelastic scattering per steradian is then given by

$$P(\theta) = k_{f+}^3 k_i \cos^2\theta \frac{\hbar q_+ \tanh(q_+ D)}{2\pi^2 \rho \omega_{q_+}} (n_{\mathbf{q}_+} + 1) + k_{f-}^3 k_i \cos^2\theta \frac{\hbar q_- \tanh(q_- D)}{2\pi^2 \rho \omega_{q_-}} n_{\mathbf{q}_-} \ . \tag{17}$$

The plus and minus subscripts refer to the one-ripplon creation and annihilation, respectively. The final wave numbers are determined by energy conservation

$$k_{f\pm}^2 - k_i^2 \pm \frac{2m_H}{\hbar} \omega_{k_{f\pm}\sin\theta} = 0 \ .$$

while $q_\pm = k_{f\pm}\sin\theta$ are ripplon wave numbers.

Thermal averaging is easily carried out numerically on the basis of Eq. (17). Explicit expressions can be given in two angular regimes, depending on the predominance of either the van der Waals or surface-tension term in the ripplon dispersion relation. For small θ ($\theta<0.5°$ at $T=0.1$ K)

$$\langle P(\theta)\rangle_{\rm th}=\frac{24m_H^2k_B^3}{\pi^2\rho g\hbar^4}T^3\cos^2\theta\ , \tag{18}$$

while for larger angles ($\theta>0.5°$ at $T=0.1$ K) we have a rapid falloff:

$$\langle P(\theta)\rangle_{\rm th}=\frac{4m_Hk_B^2}{\pi^2\gamma_0\hbar^2}T^2\frac{1}{\tan^2\theta}\ , \tag{19}$$

the angular width at half maximum being less than 1° for the experimental temperature range 0.1–0.4 K and film thickness 115 Å.[11] Equations (17), (18), and (19) clearly exhibit the underlying reason for the dominant role of the long-wavelength ripplons: The strong increase of their number n_q for decreasing wave number q at the low temperatures considered. Note that both in Eq. (18) and in Eq. (19) one $\cos\theta$ factor comes in because the DIS's approximation is different from the adiabatic one.

In view of the results already obtained it is of importance to derive the insensitivity to the potential also in the context of the usual analysis of the scattering of hydrogen atoms by ^{4}He surfaces, which is based on the distorted-wave Born approximation (DWBA). The inelastic transition amplitude is a matrix element of the perturbation

$$\delta V=V(\mathbf{r},h)-V(\mathbf{r},0)\simeq -h dV_0(z)/dz$$

between unperturbed atomic and surface states. In the usual notation of scattering theory we find for the S matrix up to first order in the surface displacement h:

$$S_{ji}=\delta_{ji}\delta_{\kappa_j,\kappa_i}e^{2i\delta(k)}-2\pi i\left[\frac{m_H}{\hbar^2k}\right]\langle\mathbf{k}_j^{(-)},\phi_j|\delta V|\mathbf{k}_i^{(+)},\phi_i\rangle\ , \tag{20}$$

with the δV matrix element equal to

$$\langle\phi_j|\frac{1}{A}\int d\rho e^{i(\kappa_i-\kappa_j)\cdot\rho}\left[\frac{1}{2\pi}\int dz\, u_{-k}(z)\delta V u_k(z)\right]|\phi_i\rangle\ . \tag{21}$$

Here, conforming to our earlier approximations, we have given the final perpendicular wave number the same magnitude as the initial one. According to the analogue of Eq. (20) for one-dimensional potential scattering, the integral between square brackets in Eq. (21) is essentially the first-order change of the one-dimensional reflection amplitude due to a small displacement of the potential. This, however, is also given by

$$e^{2i\delta(k)}e^{-2ikh}-e^{2i\delta(k)}\simeq -2ikhe^{2i\delta(k)}\ . \tag{22}$$

We thus find that the influence of the potential is again only contained in an overall phase factor $e^{2i\delta(k)}$, which drops out when calculating probabilities. The result is identical to Eqs. (17)–(19) with one $\cos\theta$ factor omitted. It is clear from the foregoing that a result such as we have found can certainly not be obtained without a consistent choice of potential for calculating the distorted waves $u_k(z)$ and $u_{-k}(z)$ on one hand, and the potential occurring in the perturbation δV on the other.

III. CONCLUSIONS

In summary, we have presented a new formalism for treating inelastic H-atom scattering from the surface of superfluid ^{4}He in terms of the ripplon creation and annihilation. The method makes use of two approximations, both of which are based on the extreme low-q limit, i.e., the tentative assumption that only very long-wavelength ripplons are involved in the inelastic-scattering process. Expanding the exponential e^{-2ikh} in Eq. (15), using Eqs. (2) and (3) and substituting in Eq. (12), we find simple closed expressions, which do not contain distorted-wave integrals and Green's functions. Compared with the conventional approach, our formalism also makes it possible to calculate multiple-ripplon processes with relatively little effort. Except for the last step, i.e., the separation of the potential part in Eq. (15), our approach applies also to phonon processes.[20] In this case the atom-surface potential does not drop out since the (ρ-dependent) one-dimensional reflection amplitude changes in a more complicated way due to a variation of the bulk ^{4}He density than due to a simple displacement.

After completing this work we obtained a detailed description[2] of the reflection experiment studied in this paper, as well as an analysis[22] on the basis of the conventional distorted-wave approach, which fully confirm the validity of our low-q limit for the one-ripplon part. Reference 22, in particular, confirms via an alternative derivation the potential independence of the low-q distorted-wave results obtained in the present paper.

ACKNOWLEDGMENTS

We acknowledge useful discussions with Jook Walraven and Tom Hijmans about their ongoing inelastic scattering experiment, which was the direct motivation for the present work. This work is part of the research program of the Stichting voor Fundamental Onderzoek der Materie (FOM), which is financially supported by the Nederlandse Organisatie voor Wetenschappelÿk Onderzoek (NWO).

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[19]Although the final expressions for a^{NS} found here and in Ref. 18 differ only by a numerical factor, the q integrands differ also by a factor $\hbar\omega_q/k_BT$. This points to an essential difference in the physical pictures.

[20]We are grateful to Jook Walraven for pointing out this aspect.

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