Quantum Reflection: Focusing of Hydrogen Atoms with a Concave Mirror

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We use a concave spherical mirror to focus on a 18-mm-diam beam of H atoms down to 0.5 mm. The mirror consists of a fused-quartz substrate polished to optical precision and coated with a liquid-^{4}He film to obtain high reflectivity. The temperature dependence of the focused beam intensity enables us to study the influence of the dynamic surface roughness on the reflection of the H atoms. Both zero-point fluctuations and thermal excitations turn out to be of importance. A monolayer of ^{3}He does not significantly affect the results.

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Hydrogen atoms (H) colliding with the surface of liquid ^{4}He are particularly well suited to study the quantum regime of atom-surface scattering. This arises from the small mass of the H atom, the weak interaction between H and helium, and the possibility to cool H gas to subkelvin temperatures. Moreover helium is easily purified and liquid-^{4}He surfaces are highly reproducible and can be accurately described in terms of elementary excitations. For vanishingly low incident energy theory predicts H atoms to scatter elastically from the helium surface with a probability approaching unity.[1] This is a purely quantum-mechanical reflection phenomenon, which suggests the feasibility of a near-perfect atomic mirror. Classically, under similar conditions the atoms should stick to the surface as surface excitation is unavoidable.

Quantum reflection of both ^{4}He and ^{3}He beams at the liquid-vacuum interface of liquid helium has been observed by varous authors,[2] in particular, by Edwards and collaborators who studied the energy dependence of the effect. However, high reflectivity was not achieved except at grazing incidence (20% at an angle of 87.5°) due to absorption of the beams by the liquid even at the lowest operating temperatures.[3]

In this Letter we demonstrate for the first time focusing of a highly divergent beam of cold ($T<0.5$ K) hydrogen atoms by the use of a hemispherical concave substrate of optical quality coated with a film of superfluid helium to obtain high reflectivity. At normal incidence we measure a lower limit of 80% for the specular reflectivity. We discuss how static surface roughness due to the substrate and dynamic surface roughness due to the helium film limit this reflectivity. Mirror and diaphragm enable us to study the specularly reflected beam within an acceptance angle of about 10 mrad. By measuring the specular reflectivity as a function of temperature we obtain for the first time experimental evidence for direct inelastic scattering induced by thermal ripplons.

The theory for scattering of low-energy H atoms from the surface of liquid ^{4}He is the subject of several papers in the literature.[4-9] The H-He interaction is extremely weak and gives rise to a surface adsorption potential which only supports a single bound state.[10] Quantum reflection arises since at low energy the scattering amplitude for scattering by zero-point fluctuations or thermal ripplons is small.

Indirect evidence for the occurrence of quantum reflection of H from liquid-helium surfaces has been obtained experimentally by measurements of the sticking coefficient s,[11-13] the probability for surface adsorption, as well as measurements of the accommodation coefficient α,[14-17] a measure for the efficiency of energy transfer in atom-surface collisions. The coefficients s and α may be related using the important theoretical result that sticking is the dominant channel for heat exchange below 0.5 K.[7-9] If s depends linearly on temperature, this leads to the relation $\alpha=\frac{3}{2}s$.[9]

The most complete set of data for the sticking coefficient was obtained by Berkhout *et al.*,[13] who observed a linear temperature dependence, $s/T=0.33(3)$ K^{-1}, both for the surface of pure ^{4}He between 145 and 526 mK and for surfaces of ^{3}He-^{4}He mixtures between 73 and 174 mK. The results for ^{4}He surfaces are in fair agreement with theory[4,5] if the Morse potential is adjusted to yield a 1-K binding energy. Further support for the theoretical model is obtained by comparing the results for the sticking coefficient with the data of Helffrich *et al.*[17] for the accommodation coefficient of H atoms on ^{4}He surfaces between 180 and 400 mK. On the basis of the relation $\alpha=\frac{3}{2}s$, both sets of experimental data are in full agreement (3% level) with one another.

The measurements of s and α enable an estimate for the specular reflectivity at normal incidence $R_\perp$ using the *Ansatz* $R_\perp=1-s_\perp$. The experimental values for s and α represent an average over all angles of incidence. Theory[4] may be used to relate the sticking probability at angle of incidence θ, $s(\theta)$, to the angular-average s, yielding $s(\theta)=1.5s\cos(\theta)$. Using this, the sticking at

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normal incidence as implied by our previous results[13] is given by $s_\perp = \gamma_1 T$ with $\gamma_1 = 0.50(5)\ \mathrm{K}^{-1}$. We thus calculate $R_\perp \approx 0.95$ at $T = 100$ mK.

To obtain direct experimental evidence for high specular reflectivity we designed the experimental cell shown in Fig. 1. H atoms are produced in an rf dissociator operated at 600 mK and situated in the 0.25-T fringe field of a 6-T superconducting magnet. Under the influence of field gradients spin-down-polarized atoms ($\mathrm{H}\downarrow$) are driven downward to the center of the magnet and collected in a buffer reservoir with volume $V_B \approx 10$ cm^3 which is entirely contained in a larger volume, the pumping volume. Atoms escaping through an orifice of 0.5 mm diam in the 0.05-mm-thick bottom of the buffer reservoir give rise to a highly divergent atomic beam in the pumping volume. A small quantity of liquid helium, typically 0.02 cm^3, assures the presence of a saturated film on all surfaces. The gas originating from the beam is continually removed by a "pumping plate" H-flux detector[18] mounted around the neck of the buffer reservoir. All dimensions are chosen to assure that the probability for an atom to reach the pump is about 50× larger than the probability to reenter the buffer volume. The main component of the cell is a concave spherical mirror facing the beam and mounted in the pumping volume. This mirror consists of a helium covered fused-quartz substrate ground to optical precision into a hemispherical shape with radius of curvature 9 mm. A push and pull mechanism driven by an inchworm motor at room temperature enables vertical positioning of the mirror with a resolution of 0.5 μm.

The principle of the experiment is to observe the influence of the position of the mirror on the density decay time of the H gas in the buffer reservoir. If the center of the mirror coincides with the center of the orifice, the atoms in the beam will be specularly reflected back into the reservoir. The net flux through the diaphragm may be expressed as $-dN/dt = \frac{1}{4} n\bar{v}A\chi$, with n the density in the buffer volume, $\bar{v}$ the average atomic speed, A the area of the orifice, and χ a loss factor representing the probability that the atoms are *not* scattered back into the buffer volume. For our mirror 100% reflectivity would correspond to $\chi_g = 0.042$, as limited by geometrical factors such as spherical aberration. For densities up to $n = 10^{14}\ \mathrm{cm}^{-3}$ interatomic collisions in the beam are completely negligible. This analysis implies that the leakage from the reservoir may be largely suppressed with the mirror.

A typical measurement is shown in Fig. 2. We plot the observed loss factor $\chi \equiv \tau_0/\tau \approx -(\tau_0/N)dN/dt$ as a function of the vertical position of the mirror. Here $\tau_0 \equiv 4V_B/\bar{v}A$ is the first-order decay time in the absence of the mirror and τ is the measured first-order decay time. The results clearly demonstrate the occurrence of specular reflection of the atoms. With the mirror far from focusing conditions, the decay time is only slightly larger than that expected in the absence of a mirror. As the mirror is moved to bring the diaphragm into focus, the decay time starts to increase dramatically. From each position scan we may extract the minimal χ value ($\chi_{\min}$) for a given temperature and film thickness. As seen from the figure, an axial displacement of the mirror over 150 μm suffices to reduce the maximum effect by a factor of 2, while the mirror must be positioned with an accuracy of better than 30 μm to determine the maximum effect.

A summary of our results for $\chi_{\min}$ as a function of

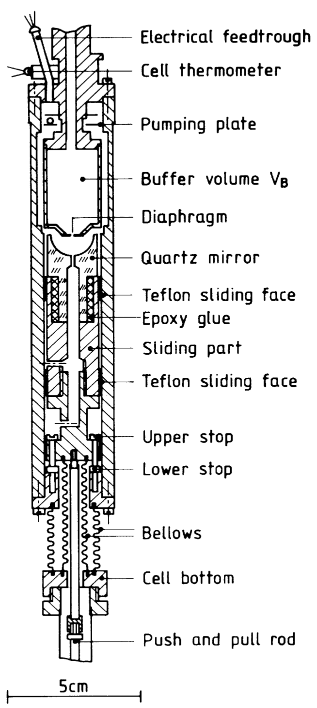

FIG. 1. The experimental cell.

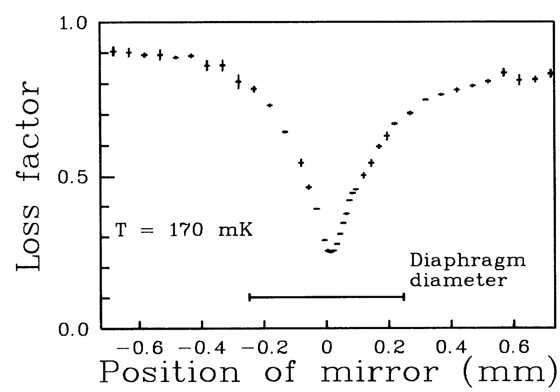

FIG. 2. The loss factor as a function of the vertical mirror position.

temperature is shown in Fig. 3. The triangles represent our data for a saturated ^{4}He film of estimated thickness 11.5 nm.[19] For ideal alignment of the optics ($\chi_g = 0.042$) the temperature dependence of $\chi_{\min}$ predicted on the basis of our earlier results[13] for s is indicated by the dotted line. Here we use for the loss factor due to sticking $\chi_s = s_\perp = \gamma_1 T$ with $\gamma_1 = 0.5$ K $^{-1}$. Most of the discrepancy with the data may be explained by assuming a lateral misalignment of the mirror or by residual roughness due to surface imperfections of the quartz substrate. Both effects cause a temperature-independent loss χ_L from the specular beam,[20] implying that only a lower bound for $R_\perp$ may be extracted from the raw data. A lateral misalignment of 30 μm of an otherwise perfect mirror leads to $\chi_L = 15\%$. The large loss factors obtained for thin undersaturated films (crosses) may well be understood in terms of substrate roughness.

The total loss factor $\chi_{\min}$ for N-independent loss mechanisms (spherical aberration, sticking, etc.) with loss factors χ_i may be written as

$$\chi_{\min} = 1 - \prod_{i=1}^{N} (1 - \chi_i) . \qquad (1)$$

Assuming $s_\perp = \gamma_1 T$ and fixing $\gamma_1 = 0.5$ K $^{-1}$, for $\chi_L = 23\%$ a least-squares fit to the data is obtained (dash-dotted line). Agreement remains unsatisfactory. Alternatively, assuming a linear temperature dependence, $s_\perp = \gamma_1 T$ and treating γ_1 and χ_L as free parameters, fitting Eq. (1) to the data yields the dashed line characterized by $\chi_L = 17\%$ and $\gamma_1 = 0.74(5)$ K $^{-1}$, which deviates significantly from our previous determination of s by a factor of 1.5.[21] Hence we find that apart from the sticking-desorption process another inelastic scattering channel has to be of importance. By exclusion this has to be direct (nonsticking) inelastic scattering by ripplons. The loss factor associated with this process we shall denote by χ_{in}.

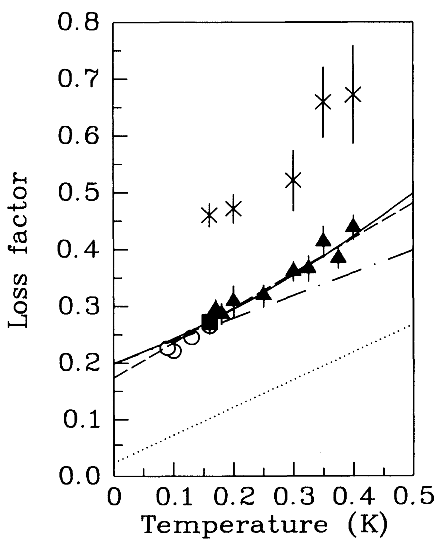

FIG. 3. Measured loss factors as a function of temperature. Crosses, results on thin pure ^{4}He film; triangles, results on saturated (115 Å) pure ^{4}He films; square (160 mK), result on saturated ^{4}He films with partial ^{3}He monolayer coverage; circles, results on saturated ^{4}He films with full ^{3}He monolayer coverage. The various curves are discussed in the text.

To gather support for this hypothesis we applied the perturbative scattering theory to direct inelastic scattering. In view of the high angular resolution of our experimental setup even ripplons with wavelength as long as $\lambda = 300$ nm carry sufficient momentum to deflect H atoms out of the specular beam as defined by the diaphragm. At wavelengths $\lambda \gtrsim 50$ nm the dynamic surface roughness is dominated by thermal ripplons rather than by zero-point fluctuations which are responsible for the sticking process. The direct inelastic scattering processes neither show up in measurements of α, since they are quasielastic, nor in measurements of s by the capillary-flow method, since to first order the momentum along the capillary axis is conserved. We calculate $\chi_{\text{in}}(T) = \gamma_2 T^2$ with $\gamma_2 \approx 0.7$ K $^{-2}$ for the probability at normal incidence to scatter out of the specular beam.[22] This result is independent of the exact shape of the H-He potential.[22,23] A least-squares fit to the data, assuming $\chi_s = \gamma_1 T$ with fixed $\gamma_1 = 0.5$ K $^{-1}$ and $\chi_{\text{in}} = \gamma_2 T^2$, yields $\gamma_2 = 0.5(1)$ K $^{-2}$ and $\chi_L = 0.20$ (solid curve). This curve satisfactorily describes our data. A least-squares fit to the data treating all three parameters χ_L, γ_1, and γ_2 as free does not produce meaningful results due to the scatter in the data. Evidence for the absence of static surface roughness was obtained by increasing the film thickness by a factor of 2, which had no effect on the observed loss factor. This implies a lateral misalignment of the mirror of 40 μm. We conclude that the specular reflectivity of our mirror is given by $R_\perp = 1 - \gamma_1 T - \gamma_2 T^2$ with $\gamma_1 = 0.5$ K $^{-1}$ and $\gamma_2 = 0.5$ K $^{-2}$. For $T = 160$ mK we calculate $R_\perp = 0.91$ within the solid angle suspended by 10 mrad. Remarkably, no effect on the reflectivity was observed by adding up to 0.1% ^{3}He to the helium in the cell. Under these conditions the presence of a full monolayer of ^{3}He is to be expected on the surface of the mirror. A similar lack of influence of ^{3}He was observed in our capillary-flow experiment.

We believe that the present work clearly demonstrates that the hydrogen atom could play a prominent role in "particle optics" experiments.[24] High reflectivity, even at normal incidence, offers excellent prospects for the use of components such as mirror collimators or grating monochromators.

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