Experimental Observation of Quantum Reflection far from Threshold

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We present first experimental evidence for quantum reflection, originating exclusively from an attractive potential between an atom and a solid surface, at energies far from the threshold $E_i \to 0$. The system of light and stable ^{3}He atoms scattering from an α-quartz crystal allows confirmation of recent theory on quantum reflection up to its asymptotic behavior, determined by the nonretarded van der Waals potential $-C_3/r^3$. From the data, the gas-solid interaction potential is deduced quantitatively, covering the energy region, in which retardation plays a role.

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Surprising quantum phenomena may occur when the wave nature of an atom becomes dominant with respect to its classical, particlelike behavior. An example is above-barrier reflection of slow atoms, with incident kinetic energy exceeding the barrier height [1]. This effect was recently discussed by Côté *et al.* for an evanescent wave atomic mirror [2,3]. Interestingly, the barrier does not need to be repulsive [4]. In the quantum regime, reflection also takes place from a purely attractive potential, which falls off with distance r faster than r^{-2}.

Therefore, a wave function impinging on a surface may be partially reflected already from the attractive well of the interaction, before reaching the repulsive wall. This quantum reflection (QR) is predicted to increase with smaller incident energy E_i. As a consequence, the probability s of the particle sticking to the surface drops to zero. In the MIT experiment on H atoms scattering from the liquid surface of He [5], it was confirmed that, at very low energies (as compared to the unique bound state of the system) $s \propto E_i^{1/2}$. At higher energies, $s(E_i)$ changes form, depending on the parameters of the attractive potential tail [6]. It should be noted, however, that using the reflection probability defined in these experiments as $R = 1 - s$, it is generally difficult to discriminate between reflections originating from the attractive and those from the repulsive branch of the potential.

With new quantum optics experiments on ultracold collisions it has become very important to investigate QR from an attractive potential only, without the influence of the repulsive wall (e.g., for understanding loss processes in creating 2D quantum gases [7]). The first experiment hereto, recently performed by Shimizu [8], involves metastable neon atoms scattering from glass and silicon surfaces. Since the metastable atoms quench when approaching the surface, long before they reach the repulsive wall, the detected QR originates from the attractive tail only. In the following experiment, we scatter and detect ground state ^{3}He atoms, which can come very close to the surface without such losses. Still, only the atoms reflected from the attractive branch of the interaction potential are measured, by virtue of the fact that the quartz surface we used is rough. Helium being highly sensitive to surface disorder guarantees that any atom approaching the repulsive wall of the rough surface is diffusively scattered into the background very efficiently.

In earlier theoretical works, Böheim and Brenig and Carraro and Cole [9,10] establish an extremely low critical normal kinetic energy required for observing quantum reflection from surfaces, on the order of 10^{-6} of the interaction potential depth V_0. Their value is based on a coarse criterion for the breakdown of the WKB approximation ($|d\lambda_{\mathrm{dB}}(r)/dr| \approx 1$) and therefore represents merely a rough and rather conservative estimate of the critical energy. Shimizu's data were obtained in a lower energy regime and are thus no test for this criterion. A more accurate theory on QR, formulated recently by Côté, Friedrich, and co-workers [11,12], predicts a much more gradual transition from the classical into the quantum regime. In this Letter, we present the first experimental evidence for QR at energies E_i up to $10^{-3} \times V_0$, mapping out this transition precisely. The implications for S-wave scattering in ultracold atom-atom collisions is discussed in detail in [13]. We here explicitly prove the validity of this new theory far from threshold, which may have important practical consequences. In addition, the exact gas-surface potential parameters are deduced, providing information on the surface properties.

The region within the interaction potential, in which QR takes place, strongly depends on the kinetic energy of the incident particle. The center of this reflection region is situated at some distance close to r_0, where the kinetic energy equals the potential one [9,12]. In order to observe reflection from the attractive branch of a hard wall potential it follows that the kinetic energy of the atom must be smaller than V_0. For realistic systems there is an additional effect: soft repulsive interaction alters the form of the well near the minimum to be flatter than r^{-3}, which leads to a decrease of the critical energy below V_0. When reducing the incident kinetic energy, the reflection region moves towards larger distances, i.e., away from the surface, and the reflection amplitude increases. By varying the incident energy the attractive

part of the interaction potential range can thus be sensitively probed.

At energies E_i close to V_0, i.e., far from threshold, the reflection coefficient shows asymptotic behavior. This was analytically calculated by Pokrovskii *et al.* [1] for above-barrier reflection in the general case, and by Friedrich *et al.* for the attractive atom-surface interaction [12]. The reflection coefficient from a homogeneous attractive potential $-C_n/r^n = -\frac{\hbar^2}{2m} \times (\beta_n)^{n-2}/r^n$, which is a function of the distance r from the surface, takes the asymptotic form

$$|R_n^{\text{asymp}}|^2 = \exp[-2B_n(k_i\beta_n)^{1-(2/n)}], \tag{1}$$

where the constant B_n depends on the power n of the potential. The primary normal kinetic energy E_i of the incident atom with mass m and the strength of the homogeneous potential are expressed in terms of the wave number $k_i = [(2m/\hbar^2) \times E_i]^{1/2}$ and the length parameter $\beta_n = [(2m/\hbar^2) \times C_n]^{1/(n-2)}$, respectively, so that the product $k_i\beta_n$ is dimensionless. The parameters of the reflecting potential can thus be determined by measuring the asymptote (1) as a function of the normal incident kinetic energy.

The long range attractive part of the interaction potential between a neutral atom and the surface of a solid is in general not homogeneous but predicted to be well described by the Casimir–van der Waals potential [8,12]:

$$V(r) = -\frac{C_4}{r^3(r+l)}, \tag{2}$$

where l is the transition length between the two homogeneous parts of the potential: the van der Waals potential $-C_3/r^3$ at the distance $r \ll l$, and the retarded potential $-C_4/r^4 = -C_3 l/r^4$ at $r \gg l$ due to the Casimir effect.

This inhomogeneous potential yields two separate reflection coefficient asymptotes of the form (1), determined by the r^{-4} and r^{-3} parts, each one having its own validity range:

$$|R_4^{\text{asymp}}(k_i)|^2 \quad \text{for } G_4 \ll k_i\beta_4 \ll \rho^2, \tag{3}$$

$$|R_3^{\text{asymp}}(k_i)|^2 \quad \text{for } \rho^3 \ll k_i\beta_3 < \left(\frac{\beta_3}{a}\right)^{3/2}. \tag{4}$$

The distance a denotes the position of the potential minimum and the constants B_n, which enter in (1), are calculated to amount to $B_4 = 1.694\,43$ and $B_3 = 2.240\,50$. The lower limit $G_4 = 0.35$ [14] for $k_i\beta_4$ defines the region of relative high energies, $E_i \gg (2m/\hbar^2)^2 \times G_4/C_4$, where the reflection coefficient takes the analytic form (1).

The dimensionless parameter

$$\rho = \frac{\beta_3}{\beta_4} = \sqrt{\frac{2m}{\hbar^2}}\frac{C_3}{\sqrt{C_4}} \tag{5}$$

is characteristic for the atom-surface system in general and determines the asymptotic behavior for reflection: at $\rho^2 \approx \beta_3/a \gg G_4$ only the asymptote (3) can be observed, whereas when $\rho^2 \not\gg G_4$ the asymptotic behavior (4) dominates.

In order to measure the asymptote (4), determined by the nonretarded van der Waals potential only, the incident atom should have a normal incident energy $E_i \gg C_3^4/C_4^3 = C_3/l^3$. In the entire validity range of (3) and (4) for high energy QR, the incident atom is reflected relatively close to the surface. In the recent experiment by Shimizu [8] exactly this asymptotic region was not accessible, because the metastable Ne atoms they used decay at the distance of some nm from the surface.

Here, we report on the first observation of QR of neutral helium atoms from an α-quartz crystalline surface in the high energy region, far from threshold. In this system the asymptotic behavior of the reflection coefficient is determined by the nonretarded van der Waals potential only.

The experimental results presented here are obtained on an apparatus designed for surface studies, using the novel atomic beam spin echo technique [15]. In this machine, the nuclear magnetic moments of ^{3}He atoms are manipulated, so as to obtain detailed information on changes in the particle's energy before and after scattering [16]. For the data here, however, the actual spin echo part of the ^{3}He spectrometer is of importance only in as much as it allows us to determine the velocity distribution in the beam precisely. The atomic ^{3}He beam is produced in a 500 μm diameter nozzle source, cooled by a 4.2 K ^{4}He bath cryostat and detected in a commercial mass spectrometer with a saturation rate of 2 MHz. The target crystal is mounted in the scattering chamber, half way between source and detector, and can be manipulated around the three Cartesian axes for incident angle θ_i, in-plane and azimuthal orientation. The detector can be rotated in the horizontal plane to include a total scattering angle $90° \leq (\theta_i + \theta_f) \leq 180°$ with the incident beam. Since the rotation axes of incident and scattering angle are aligned to coincide, the specularly reflected He atoms can be followed directly in a so-called $(\theta - 2\theta)$ scan, with an angular resolution of $\Delta\theta_f \approx 0.17°$. Further details on the ^{3}He spectrometer will be presented elsewhere. A sketch of the experimental setup is given in Fig. 1.

The QR experiment is performed on an α-quartz single crystal having a diameter of 25 mm, a thickness of 1 mm, and a polish on both sides. The ^{3}He-beam average kinetic

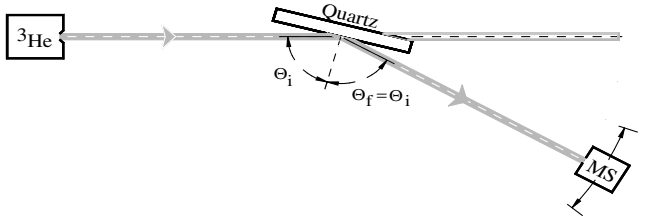

FIG. 1. Schematic representation of the experimental setup.

energy $E_0 = 0.63$ meV amounts to approximately 10% of the He-quartz interaction potential well depth $V_0 = 9.6$ meV, reported in literature [17]. The atomic beam has a wavelength distribution with a relative width of circa 20% at an average de Broglie wavelength λ_{dB} of 6 Å. Atomic force microscopy measurements, performed prior to chemical etching of the quartz sample, indicate a randomly stepped surface structure. The terrace width is of the order 100 nm and their height is Gaussian distributed with width ≈ 12 Å. Because of an atomic roughness within the terraces, there is no specular reflection from the repulsive potential wall. Indeed, when scattering electrons (LEED), ^{4}He or ^{3}He atoms close to normal incidence from the surface, no reflectivity could be detected. However, upon incrementing θ_i beyond 84° a rapidly growing ^{3}He specular intensity is measured. By increasing the impinging angle, the incident kinetic energy of the atom perpendicular to the surface $E_i = E_0\cos^2\theta_i$ is decreased. For θ_i ranging from 84° to 89.73°, this means a reduction of the average normal energy from 6.9 μeV down to 14 neV, corresponding to 10^{-3}, respectively, 10^{-6} of V_0. The angular width of the reflected peak at this grazing incidence is machine limited. No broadening of the specular peak, as measured for classical reflection from stepped surfaces [18], is observed. The coherence length (or transfer width) for specular reflection, $\omega = \lambda_{dB}/(\Delta\theta_i \times \cos\theta_i)$, is 0.2 μm at normal incidence and ranges from 2 up to 42 μm for the angular range in which QR is measured. The surface area illuminated by the atomic beam and the fraction of atoms actually involved in the scattering experiment depend on the incident angle. This was determined in an independent measurement and taken into consideration when analyzing the data.

Figure 2 shows the resulting reflection coefficient as a function of the dimensionless average normal wave number $k_i a = 2\pi\cos\theta_i(a/\lambda_{dB})$, with $a = 2.65$ Å being the position of the potential minimum [17]. For a constant energy beam, the normal wave number is varied by changing the incident angle θ_i. Open circles in the figure represent the experimental data from the randomly stepped surface. Since the step height distribution is Gaussian with width $\sigma \lesssim 12$ Å, the terraces are wide and the illuminated surface area consists of a large number of them; the reflection coefficient of the rough surface (open circles) can be related to that of a smooth one (full circles), through

$$|R_{\text{rough}}(k_i)|^2 = e^{-4\sigma^2 k_i^2} \cdot \frac{\int_{0°}^{\theta} f(\theta)d\theta}{\int_{0°}^{90°} f(\theta)d\theta} \cdot |R_{\text{smooth}}(k_i)|^2. \quad (6)$$

The first term quantifies the reduction of the reflection coefficient due to dephasing of the wave function upon scattering from terraces at different heights. This effect is more pronounced at higher energy, because then the de Broglie wavelength normal to the surface, $\lambda_{dB}/\cos\theta_i$, becomes comparable to σ. The second factor in (6) takes into account the loss of atoms hitting the steps from the side and becomes noticeable only at grazing incidence. Here, $\theta = (90° - \theta_i)$ and $f(\theta) = (L/\sigma)/\sqrt{2\pi} \times \exp[-(L\tan\theta/\sigma)^2/2]$ describes the probability that a step has height $L\tan\theta$. In Fig. 2, the average terrace width L is taken to be $L = 75$ nm and $\sigma = (10 \pm 2)$ Å. The error bars on the corrected data (full circles) contain both the statistical error and the uncertainty in σ.

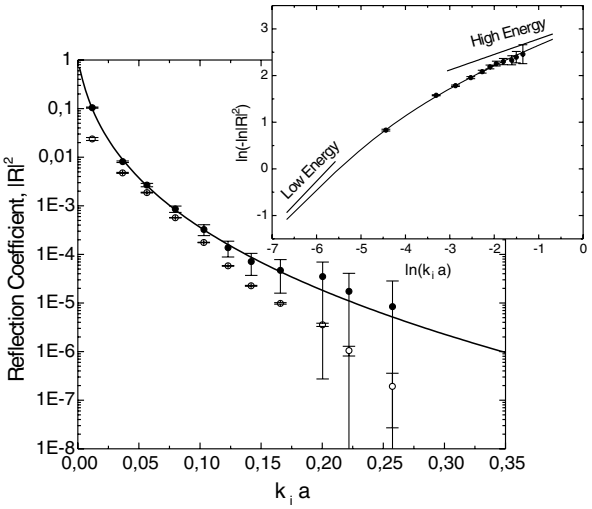

FIG. 2. Reflection coefficient as a function of the dimensionless incident wave number $k_i a$ ($\propto \cos\theta_i$). Open circles: experimental data from the stepped surface. Full circles: corrected data, representing QR from the smooth surface. Solid line: computer simulation using the potential (2) parametrized with $C_4 = 23.6$ eV Å^4 and $l = 10$ nm. Inset: replot of the same data. The straight of slope $= 1/3$ shows the asymptote (4) with $\beta_3 = 347$ Å; the straight at small $\ln(k_i a)$, with slope $= 1$ and ordinate axis intercept $\approx \ln(2.4\beta_3/a)$ for $\rho \approx 1.9$ [12], is the near-threshold asymptote.

We can now directly compare the corrected experimental data with a computer simulation for QR from a smooth surface (solid line). Our calculation is based on the method suggested in [11] for an attractive potential of the form (2) and shows very good agreement with the data. This method matches the WKB wave function to the exact solution of the Schrödinger equation in every point of the interaction. We have seen no significant difference when including the entire wavelength distribution (as determined using the spin echo technique) in the simulation instead of just the average value of λ_{dB}.

The C_4 coefficient for the inhomogeneous interaction potential (2) entering into the computer simulation can be written as [8,19]

$$C_4 = \frac{1}{4\pi\varepsilon_0} \cdot \frac{3\hbar c\alpha}{8\pi} \cdot \phi(\varepsilon) \cdot \frac{\varepsilon - 1}{\varepsilon + 1} = 23.6 \text{ eV Å}^4. \quad (7)$$

Herein, $\alpha = 2.3 \times 10^{-41}$ Fm^2 denotes the polarizability of the incident He atom and $\varepsilon = 4.5$ is the dielectric constant of the α-quartz crystal [20]. The terms containing ε in expression (7) correct the interaction with a

dielectric surface for that with a conductive one, whereby $\phi(\varepsilon)$ is found in [19]. The transition length l, the only adjustable parameter in the simulation, is determined to be $l = (10 \pm 1)$ nm in order to give the best agreement with experiment. This value is in perfect agreement with the wavelength $\lambda/(2\pi) = 9.3$ nm corresponding to the atomic transition between the electronic ground and the first excited state in helium. In addition, the important dimensionless parameter ρ characterizing our system and defined in (5) then becomes $\rho = 1.9 \pm 0.2$. ρ being so small, the asymptotic behavior of the reflection coefficient is expected to be determined entirely by the nonretarded interaction potential $-C_3/r^3$. That is, from the two high energy asymptotes only the higher one (4) should be observed. For our system this lies at incident energies $E_i \gg 7 \times 10^{-6} \times V_0 = 69$ neV, corresponding to incident angles $\cos\theta_i \gg 0.011$. Our experimental data acquired at nongrazing incidence approach this asymptote very closely.

Replotting the reflection coefficient on a $\ln(-\ln)$ scale as a function of $\ln(k_i a)$ turns the asymptotic behavior (1) into a straight line, as shown in the inset of Fig. 2. From this, full information on the homogeneous part of the reflecting potential can be obtained: the slope gives the potential power n and the ordinate axis intercept yields the length parameter β_n (and therewith C_n). The high energy asymptote for the nonretarded branch ($n = 3$) in the figure results in a van der Waals coefficient $C_3 = C_4/l = 236$ meV Å^3.

Kunc *et al.* [17] calculate the potential power n to vary from 3.8 to 6 within the distance $r < 50$ Å. Their potential follows (2) with the given parameters only at the distances $\lesssim 10$ Å from the surface. This is perfectly consistent with the authors not including retardation in their model. As an independent check of the resulting attractive potential the parameters for the potential minimum with and without retardation are calculated. The latter show agreement within 3% with the values given in [17].

In contrast, potential parameters can principally not be derived from the near-threshold $E_i \to 0$ asymptote in a single experiment only. On a $\ln(-\ln)$ scale (inset in Fig. 2) this asymptote always has slope one, independent of n. It causes the universal quantum sticking behavior, $s \propto \sqrt{E_i}$, first measured by Yu *et al.* [5].

In conclusion, ground state ^{3}He atoms allow experimental access to QR from the attractive potential far from threshold they sense from an α-quartz surface. We confirm the high energy asymptotic expression given by (1) and show that it is determined by the nonretarded van der Waals potential only. Deviation of the experimental data from this asymptote shows that the interaction potential near the surface falls off steeper than a pure van der Waals potential. This is due to the retardation effect even at the distance of 30 Å above the surface. Our analysis, based on the complete theory on above-barrier QR, shows excellent agreement with the experimental data for the potential coefficients $C_4 = 23.6$ eV Å^4 and $l = (10 \pm 1)$ nm. The interaction potential compares well to the one calculated by Kunc *et al.* [17] in the vicinity of the potential minimum. Moreover, l perfectly matches the transition wavelength from the electronic ground to the first excited state of He.

We have recently installed a highly efficient mass spectrometer [21], allowing us to measure in the even higher energy range, where the influence of the repulsive wall becomes visible. We have now obtained QR data also on metal and semiconductor targets, which we are currently analyzing.

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