Measuring the van der Waals forces between a Rydberg atom and a metallic surface

A. Anderson, S. Haroche,* E. A. Hinds, W. Jhe, and D. Meschede
Physics Department, Yale University, P.O. Box 6666, New Haven, Connecticut 06520-8167
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We have observed the deflection of Rydberg atoms towards a metallic surface by the van der Waals force. Cs and Na atoms in states of principal quantum number n were sent between two parallel gold-coated mirrors, spaced by a gap w ($2.1\ \mu m \leq w \leq 8.5\ \mu m$). We measured the value n_m at which the transmission cuts off and from the variation of n_m versus w, we obtained a measure of the atom-surface interaction. For $12 < n < 30$ this interaction is 3–4 orders of magnitude larger than for ground-state atoms, and it obeys the scaling laws of the Lennard-Jones model.

The van der Waals interaction between a metallic surface and an atom is an important process in atomic physics. If the metal behaves as a perfect conductor at characteristic atomic frequencies, the interaction can be viewed, after Lennard-Jones,[1] as resulting from the coupling of the atomic dipole with its electrostatic images in the surface. This coupling gives rise to energy-level shifts proportional to z^{-3} (where z is the atom-surface distance), and the derivative of these shifts with respect to the distance corresponds to the dipole-image van der Waals force $F_{VW} \sim z^{-4}$ that pulls the atom towards the metal. This assumes that z is small compared with any characteristic atomic-transition wavelength so that retardation is negligible.[2] For ground-state atoms or molecules, F_{VW} becomes large when z is smaller than a few Bohr radii. It plays an important role in a variety of physicochemical processes in which atoms come close to surfaces, such as adsorption phenomena.[3] In these processes, however, the simple Lennard-Jones interaction is complicated by short-range atom-surface coupling terms that depend upon the detailed microscopic structure of the surface. At distances larger than a few tens of angstroms, F_{VW} takes its simple asymptotic form, but becomes exceedingly small.

To our knowledge only one experiment has demonstrated the existence of the long-range (> 100 Å) van der Walls attraction between free atoms and a metal. Kusch *et al.*[4] observed the very small deflection experienced by a beam of cesium atoms at grazing incidence, passing within a few hundred angstroms of a gold-coated cylinder. This was a very difficult experiment, with a very low signal due to exceedingly small atomic fluxes in the relevant atom-metal distance range, and only qualitative information about the van der Waals force could be obtained from it.

A simple way of increasing the van der Waals force is to excite the atoms into a Rydberg state with a large effective quantum number n (n is the principal quantum number of the level minus its quantum defect). The electric dipole of the Rydberg atom scales as n^2 and hence the van der Waals force, proportional to the square of this dipole, is expected to increase as n^4 and to scale as $(n/z)^4$, provided z remains small compared with the characteristic Rydberg transition wavelength (in the mm domain). As a result we expect the range of the van der Waals attraction to be extended by a factor equal to the ratio of excited-state to ground-state effective-n values. In this paper we report the first evidence of van der Waals attraction between an excited atom and a metal, using Rydberg atoms with effective quantum numbers varying from 12.5 to 27 observed at typical atom-metal distances ranging from 1 to 3 μm.

The principle of our experiment is to measure the transmission of a beam of excited atoms through a narrow metallic channel. We send a beam of Rydberg atoms (cesium or sodium) through a tunnel of length l made of two plane-parallel gold-coated mirrors separated by a variable distance w ($2.1\ \mu m \leq w \leq 8.5\ \mu m$). This separation is achieved by sandwiching between the mirrors two thin Ni foils used as calibrated spacers.[5] The gold coating is a few hundred angstroms thick, which is enough to ensure high reflectivity at the characteristic Rydberg-atom microwave transition frequencies, making the electrostatic picture of the van der Waals interaction quite adequate. The flatness of the mirrors is $\sim\lambda/10$ over their whole area, where λ is a typical optical frequency. The surfaces irregularities are thus much smaller than the atom-surface distances. Alkali atoms striking the metallic surfaces stick to them regardless of excitation and do not emerge from the other end of the tunnel.[6] The Rydberg atoms that have escaped collision with the walls are detected downstream by electric field ionization using a channeltron electron multiplier (CEM). We have performed two complementary experiments with this setup.

(i) Using the beams of two cw dye lasers L_1 and L_2 focused in position A in front of the gap entrance [Fig. 1(a)], we excite the atoms into a Rydberg state before they fly into the tunnel. The excited atoms then travel along the full length $l_0 = 8$ mm of the gap structure. This experiment is performed on cesium atoms. Laser L_1 ($\lambda_1 = 0.455\ \mu m$) excites the atoms from the $6S_{1/2}$ ground state into the $7P_{3/2}$ level. About 13% of the excited atoms decay to the $5D_{5/2}$ level from which laser L_2 ($\lambda_2 = 0.601\ \mu m$) carries the excitation into an nF state. For a given gap width we vary n by tuning λ_2 in the range $15 \lesssim n \lesssim 30$. In order to measure the tunnel transmission factor for each n value we move lasers L_1 and L_2 from position A to position B downstream and

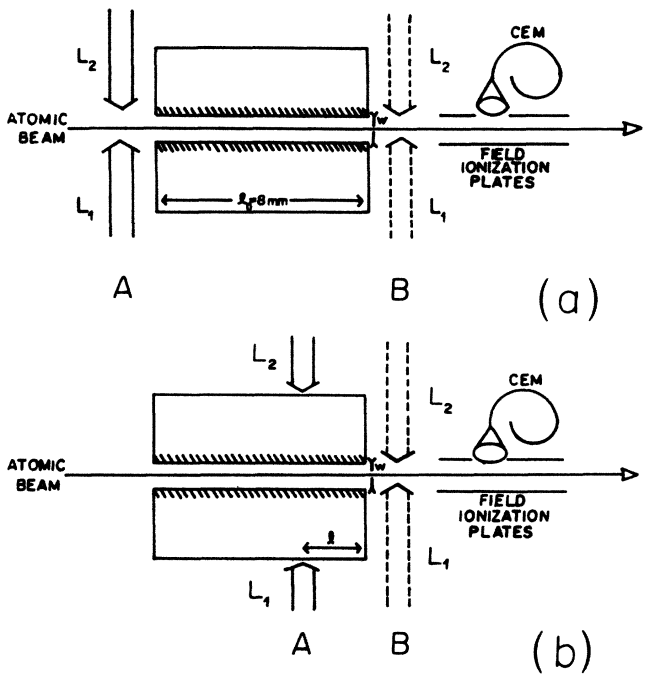

FIG. 1. Scheme of the experimental apparatus for measuring the Rydberg-atom transmission ratio through a metallic tunnel. (a) and (b) show the methods used with Cs and Na, respectively.

determine the ratio S_A/S_B of the corresponding Rydberg-atom detection signals. We make a succession of upstream and downstream measurements, increasing the value of n until the signal S_A drops below our detection sensitivity (we detect usually a few hundred atoms per second in position B and the background signal is smaller than 1 count per second, so that we can measure a minimum transmission factor of the order of 0.1–1 %). We thus determine a cutoff value $n_m(w,l_0)$ corresponding to the "largest" atom for which there exists a trajectory through a tunnel of width w and length l_0.

(ii) In a variant of this experiment, performed on sodium, we prepare the atoms *inside* the tunnel structure, as shown in Fig. 1(b). Two laser beams L_1 and L_2 are in position A and excite the atoms through the semitransparent gold coatings of the mirrors. Laser L_1 ($\lambda_1=0.589$ μm) excites Na to the $3P_{3/2}$ resonant state and laser L_2 ($\lambda_2=0.422$ μm) carries the excitation into the $14S$ level ($n=12.6$). We now vary the length l that the excited atoms must travel in the tunnel by moving the laser-interaction point away from the downstream position B. We measure the Rydberg transmission as a function of l and determine the maximum tunnel length l_m for which there is transmission.

Now we analyze how the measurement of $n_m(w,l_0)$ or, alternatively, of $l_m(n,w)$ can yield direct information on the van der Waals attraction between a Rydberg atom and a gold surface. The nonretarded interaction potential of an atom in quantum state n,j,m with a plane metallic surface at distance z is

$$U(z)=-\frac{e^2}{16}\langle r^2(1+\cos^2\theta)\rangle_{n,j,m}\frac{1}{z^3}\ , \tag{1}$$

where $e^2=q^2/4\pi\varepsilon_0$ and (r,θ) are the polar coordinates of the Rydberg electron (charge q) in the atomic rest frame quantized along the normal to the surface. For $n\gg j,m$ it is legitimate to neglect the slight variation with j and m of the expectation value. Then we have

$$\langle r^2(1+\cos^2\theta)\rangle_{n,j,m}\simeq\frac{10n^4}{3}a_0^2\ , \tag{2}$$

where a_0 is the Bohr radius. Consider now an atom (mass M, effective quantum number n) initially ($t=0$) at distance z_0 from a single metallic surface with zero normal velocity. As the atom falls towards the surface, the relation between t and z is given by straightforward integration of the equation of motion. From the energy-conservation equation

$$\tfrac{1}{2}M(dz/dt)^2=U(z_0)-U(z)\ , \tag{3}$$

and taking into account Eqs. (1) and (2), we readily find the "falling" time

$$t_0=0.74(12M/5)^{1/2}\frac{z_0^{5/2}}{n^2ea_0}\ . \tag{4}$$

If the atom has an initial longitudinal velocity v, it will travel a distance l along the surface without collision provided l/v is smaller than the time t_0 defined by Eq. (4). This leads to a simple scaling law linking z_0, l, v, and n for atoms escaping collision:

$$\frac{n^4l^2}{z_0^5}\le 1.3\frac{Mv^2}{(ea_0)^2}\ . \tag{5}$$

In a tunnel made of two parallel surfaces separated by a gap w, the attractions to both walls will tend to balance each other but the interaction with the closer mirror will rapidly dominate. As the transmission decreases, the last atoms to cross the gap are those that start with $z_0\sim w/2$, so we expect a scaling law similar to Eq. (5) linking n, l, and w. After replacing Mv^2 by its mean value proportional to k_BT (T is the alkali oven temperature), we have

$$\frac{n^4l^2}{w^5}\le\eta\frac{k_BT}{(ea_0)^2}\ , \tag{6}$$

where η is a dimensionless factor of the order of unity, that is, independent of the atomic species (Na or Cs).

The qualitative arguments above are confirmed by a numerical integration of the equation of motion for atoms in the tunnel, taking into account the attraction to both mirrors and the atomic velocity dispersion. For each set of the fixed parameters n, l, and w we have computed an ensemble of atomic trajectories corresponding to 400 choices of initial conditions. Figure 2(a) shows an ensemble of trajectories in a 5.4 μm wide, 8 mm long tunnel for $n=20$. We obtain a theoretical "van der Waals transmission fraction" $f_{\rm vW}(n,l,w)$ by counting the fraction of trajectories that emerge without wall collision. Figure 2(b) shows the variation of $f_{\rm vW}(n,l,w)$ with n for $w=5.4$ μm and $l=8$ mm, and as expected it decreases to-

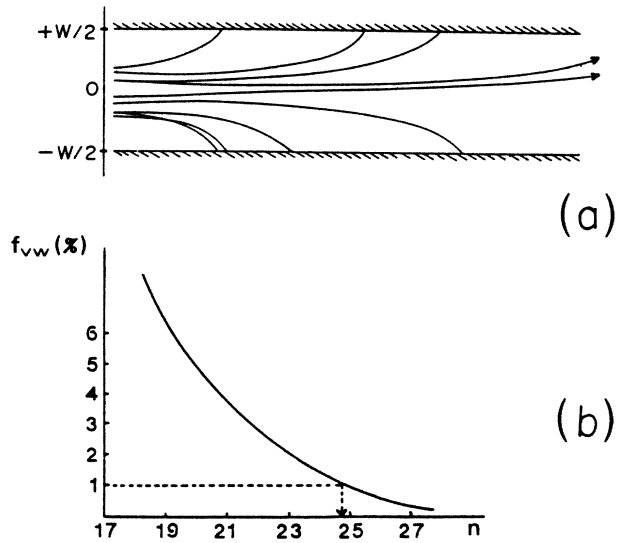

FIG. 2. (a) Examples of atomic Cs trajectories through a gap of width $w=5.4$ μm and length $l=8$ mm. The principal quantum number $n\sim 20$. A thermal velocity distribution corresponding to $T=350$ K is assumed for the atoms entering the tunnel. (b) Calculated transmission ratio f_{VW} as a function of n for $w=5.4$ μm.

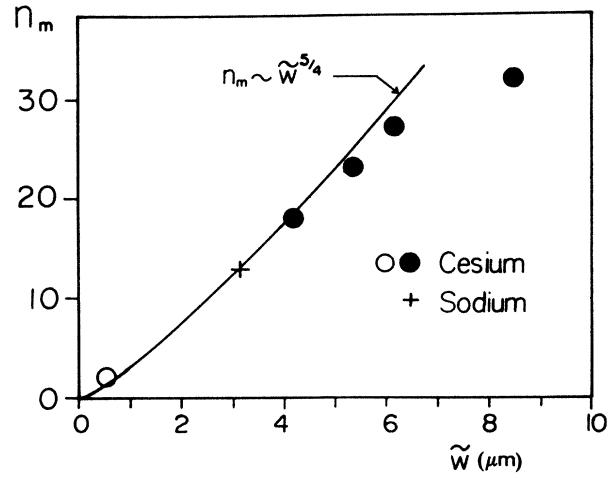

FIG. 3. Maximum Rydberg principal quantum number n_m vs scaled tunnel width $\tilde{w}$. The solid line is theoretical [Eq. (7)], corresponding to a cutoff in transmission $f_{VW}=1\%$. Circles are experimental values for Cs, obtained after correction for the radiative decay of the Rydberg atoms in the gap, the plus sign corresponds to Na data. The unshaded circle indicates the ground-state cutoff point, estimated from Ref. 4. Note the agreement between van der Waals theory and experiments on narrow gaps. Deviation from theory for larger gaps is attributed to stray electric fields.

wards zero with increasing n. The tail of f_{VW} for large n corresponds to the fastest atoms in the velocity distribution of the atomic beam. In order to be definite and because it corresponds to the experimental sensitivity, we choose $f_{VW}=1\%$ as the cutoff limit that defines the value of $n_m(w,l_0)$. The results of these calculations, performed for various values of l and w, agree with the scaling law [Eq. (6)] and yield the numerical value $\eta=0.74$. In the case of our Cs measurements the scaling law takes the form $n_m^4 \propto w^5 T/l_0^2$, with $T=350$ K and $l_0=8$ mm. We define a scaled width $\tilde{w}=w[(8\text{ mm})/l_m]^{2/5} \times[T/(350\text{ K})]^{1/5}$ that allows us to write a simple, more general scaling law

$$n_m=3.0\tilde{w}^{5/4}. \tag{7}$$

This is plotted in Fig. 3 and constitutes a universal curve to which all our experimental data can be compared.

Before making this comparison, we must briefly discuss the influence of the Rydberg-atom's finite lifetime on our measurements. The Rydberg-atom transmission we actually measure is not the purely ballistic fraction $f_{VW}(n,l,w)$. There is also a radiation loss factor $f_{rad}(n,l)$ because the Rydberg atoms are radiatively unstable and have a probability of decaying to states with $n' \ll n$ during the crossing time $\tau=l/v$. These states are not detected by the field-ionization detector and so the decay process reduces the observed transmission factor below the ballistic value f_{VW}. The radiative decay of Rydberg states is a combination of spontaneous emission and blackbody radiation-induced transitions.[7] The former transfers atoms directly into undetected bound states whereas the latter redistribute them into a larger set of detected Rydberg states with $n'\sim n$ and various angular momenta ($j'><j$). As a result of these processes, the atoms that remain in Rydberg levels evolve into a mixture of states with different angular momenta, having on the average a much longer spontaneous-emission lifetime than the state initially excited. The van der Waals force is not appreciably modified by the induced processes since n does not change by a significant amount and j remains much smaller than n. It is thus reasonable to assume that the trajectories of these Rydberg-atom superpositions are still well characterized by their initial quantum number n. In an independent experiment, we measured the "effective lifetime" of these blackbody radiation-induced superpositions in cesium and found that it is in the range 10–40 μs for n varying between 15 and 35. As a result the radiative decay factor during the $\sim$25-μs flight time through the $l_0=$ 8-mm tunnel is between 0.1 ($n\sim 15$–20) and 0.5 ($n\sim 30$). It follows that the 0.1% minimum transmitted fraction measured in our experiment corresponds to ballistic factors in the range 0.2–1 %. Specifically, a ballistic fraction $f_{VW}=1\%$ corresponds to experimental transmission fractions $f=f_{VW}f_{rad}\sim 0.1\%$ for $n\sim 15$–20, $f\sim 0.25\%$ for $n\sim 25$, and $f\sim 0.5\%$ for $n\sim 30$–35. After making this correction our results in the cesium experiment ($l_0=8$ mm) are $n_m=18\pm1$, 22 ± 1, 27 ± 1, and 32 ± 1 for $W=4.2$ μm, 5.4 μm, 6.2 μm, and 8.5 μm, respectively. These data are shown as solid circles in Fig. 3.

In the sodium experiment (oven temperature $T=470$ K) we have found that the 14S Na Rydberg level ($n=12.6$) propagating in a $w=2.1$ μm wide channel is cut off at $l_m=3.2$ mm. Using the scaling laws discussed above, this result corresponds to the cross in Fig. 3. We also show for comparison on the same figure the corre-

sponding cutoff value for the cesium ground state ($n=1.96$). Using the data from the experiment by Kusch *et al.*[4] we have estimated the cesium ground state to metal van der Waals force and have found that a f_{VW} ratio of 1% must correspond for a $l_0=8$ mm long tunnel to $w=0.6$ μm.

The agreement between experiment and theory exhibited in Fig. 3 in the effective quantum-number range $n=2-27$ shows that Rydberg atoms up to $n=27$ experience the dipole atom-metal force at micrometer distances from metallic mirrors. The factor of 5–10 increase in gap size between the (estimated) ground-state point and the measured Rydberg data reflects an increase of 3–4 orders of magnitude in the van der Waals force obtained by exciting the atoms from their ground state to excited levels in the range $n=12-25$. Within the precision of our experiment, there is no significant departure in this range of n values from the Lennard-Jones model assuming that the gold surface is a perfect conductor at the Rydberg electron characteristic frequency. Our results are compartible with a $w^{5/4}$ scaling law for n_m, although our precision does not allow us to distinguish this clearly from, say, a linear variation. Nor are we able to see any differences between Cs and Na because the van der Waals interaction depends only upon the effective quantum number of the atom at our level of precision.

The atomic sizes $a_0 n_m^2$ corresponding to the cutoff data for gaps $w=4.2$, 5.4, and 6.2 μm are, respectively, 200, 250, and 300 Å, i.e., about 200 times smaller than the gap widths. This means that the transmission cutoff indeed indicates the deflection of atoms onto the surface and not a geometrical size effect such as that observed in an earlier transmission experiment[8] performed with very short holes ($l \simeq w$).

With the largest gap we have studied ($w=8.5$ μm), the cutoff occurs for smaller atoms than predicted by the van der Waals force theory (a tendency also noticeable in the 6.2-μm-gap point). This suggests that the Rydberg atoms are then sensitive to perturbations of another nature, which we believe are related to stray electric fields. For example, large gradient fringe fields at the mirror gap entrance could induce forces deflecting the atoms onto the mirrors. Alternatively, a uniform field across the gap could destroy the atoms by field ionization.[9] The strength of the stray fields is unknown but may be quite large and influenced by alkali atom deposition (10 mV across a 5-μm gap corresponds to 20 V/cm). The important point to notice, however, is that these fields should scale as $1/w$ and their gradients as $1/w$ or $1/w^2$. The effect produced by them on the atoms should thus decrease with w much less rapidly than the van der Waals force, which scales as $1/w^4$. Thus the stray field effects are bound to overcome the van der Waals force for wide-enough gap structures. Our data indicate that this occurs around $w=6$ μm. The agreement between our experimental points and the van der Waals theory below that point indicates that in the 0–3 μm atom-surface range (0–6 μm gap) the van der Waals interaction dominates any stray field effect.

The use of a narrow tunnel in this "atom-surface collision" experiment instead of a single plane mirror has the effect of collimating the atomic beam very well. The angle θ between the atomic velocity and the tunnel axis is smaller than $w/2l \sim 5\times 10^{-4}$ rad. By virtue of this very strong collimation we select atoms whose average velocity normal to the surfaces is smaller than $v\theta \sim 15$ cm/s. The corresponding transverse kinetic energy $k_B T\theta^2$ is of the order of only 10 μK, making it possible to probe exceedingly small atom-metal interactions. The next step in this study will be to use the same kind of apparatus to measure the van der Waals energy shifts instead of the van der Waals forces. These shifts, which depend strongly upon the atom-metal distance, are of the order of a few tens of MHz at micrometer distances from the mirrors for $n \sim 20$. This means that a laser beam slightly detuned from the free-atom Rydberg transition frequency will be absorbed by atoms located in the tunnel at a well-defined distance from its walls. Irradiation of the atoms with monochromatic laser light inside the tunnel should thus provide a simple way of selecting atom-metal distances and of studying the van der Waals interaction more precisely.

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*Also at Ecolé Normale Supérieure, Paris, France.

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