Long-range atom-surface bound states

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We study the photoassociative process between *cold atoms and a surface*. A mechanism to confine free cold atoms in a planar waveguide, characterized by the long-range atom-surface van der Waals potential and based on radiative transitions, is presented. We propose that the vibrational states of this potential can be probed by reflection spectroscopy, giving information about the C_3 van der Waals coefficients.

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The successful use of laser light to control the external degrees of freedom of neutral atoms [1], slowing them down to the recoil limit, permits one to obtain atoms with de Broglie wavelength comparable to the optical wavelength. This opens the way to new experiments in fields such as nonlinear optics, atomic collisions, and metrology, as well as to new domains such as Bose-Einstein condensation in vapors [2] and atom optics [3]. In atom optics, laser light has been used, or proposed to be used, to reflect or diffract matter waves or even to confine atoms in two-dimensional (2D) waveguides, as for example in the recent articles on planar [4], cylindrical [5], and spherical [6] waveguides using evanescent waves. Lately a planar waveguide has been proposed utilizing a static magnetic field [7], which unlike the previous ones, does not induce heating of the atomic sample by spontaneous emission. For processes that take place close to the surface at distances shorter than the optical wavelength, it is very important to consider the atom-surface van der Waals interaction. For instance, atom mirrors based on the dipolar force of an evanescent wave present a difference of more than 60% in their barrier height which is attributed to the van der Waals interaction [8]. Despite its importance, only a few measurements have been performed to obtain the C_3 van der Waals coefficient in the $-C_3/z^3$ surface potential for the *lowest resonant* levels [9,10], which may be a consequence of the experimental difficulties to control atom-surface distance [11,12]. In measurements with atoms in the ground state the mechanical effects are "contaminated" by the unavoidable z^{-4} dependence of the Casimir term [13]. For Rydberg states ("large" atoms) precise measurements of the van der Waals surface interaction has been made by the Yale group [14].

Currently, atoms at temperatures below 1 mK are easily obtained with present cooling techniques, which allows one to consider new dynamic regimes for interatomic collisions [15], already observed in cold atoms photoassociation [16] and in the formation of long-range cold molecules [17]. In this paper we study the interaction between atoms with a large de Broglie wavelength and a dielectric surface via the van der Waals potential. The transfer induced by Raman transition of free cold atoms to the bound states of the surface well is considered. We calculate the efficiency of the loading mechanism and discuss a possible optical technique to probe the energy of these vibrational states.

The atom-surface potential is considered to be the sum of the long-range van der Waals attraction and a short-range exponential repulsion $V(z) = Ae^{-\alpha z} - C_3/z^3$, where z is the coordinate normal to the surface, while A and α are constants of the short-range potential [18]. The van der Waals attraction is the predominant interaction for distances smaller than $\lambda/2\pi$ from the surface [19], as considered in this work. In Fig. 1 we show the short-range repulsive, van der Waals and the resultant $V(z)$ potential forms. In order to determine the atomic center-of-mass wave functions and their energies, we numerically solve the one-dimensional (1D) Schrödinger equation:

$$-\frac{\hbar^2}{2m}\frac{d^2\Psi(z)}{dz^2} + V(z)\Psi(z) = E\Psi(z) \qquad (1)$$

for cesium atom parameters. We use 2 kHz $(\mu\mathrm{m})^3$ and 4 kHz $(\mu\mathrm{m})^3$ as the van der Waals C_3 coefficients for the $6S_{1/2}$ ground state and the $6P_{3/2}$ excited state, respectively, as extrapolated from measurements on a sapphire window [10]. Long-range bound-state eigenvalues with their respective atomic center-of-mass wave function, obtained from the numerical solution of the Schrödinger equation, are dis-
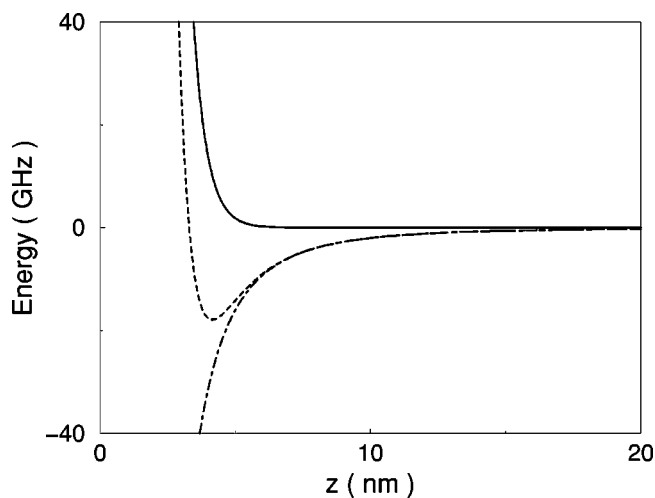

FIG. 1. Atom surface interaction potential for a dielectric and a neutral atom as a function of z. The solid, dot-dashed, and dashed lines correspond to the short-range repulsive, van der Waals, and resultant interaction potentials, respectively, for $A = 4 \times 10^{13}$ Hz, $\alpha = 2 \times 10^9$ m^{-1}, and the C_3 values given in the text.

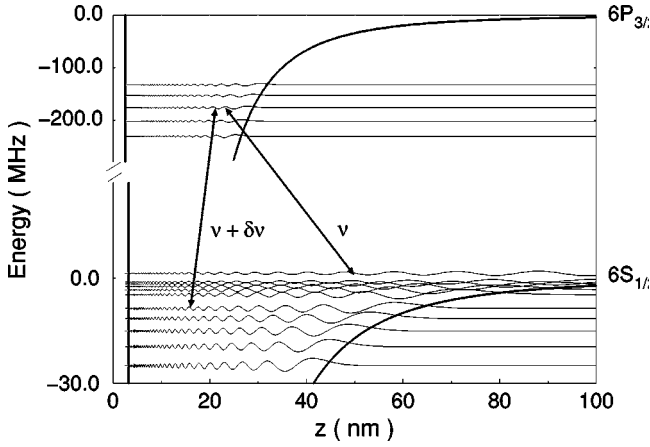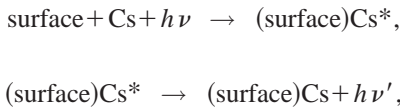*Electronic address: oria@otica.ufpb.br

FIG. 2. The Cs-dielectric surface potentials and their wave functions, corresponding to $6S_{1/2}$ and $6P_{3/2}$ cesium states for the free atom. The vibrational levels are obtained for $A = 4 \times 10^{13}$ Hz and $\alpha = 2 \times 10^9$ m^{-1}. The arrows represent the laser of frequency ν coupling the states $|\psi^g_{free}\rangle$ and $|\psi^e_{v'}\rangle$ and the laser of frequency $\nu + \delta\nu$ coupling the states $|\psi^e_{v'}\rangle$ and $|\psi^g_v\rangle$, resulting in a Λ configuration Raman transition.
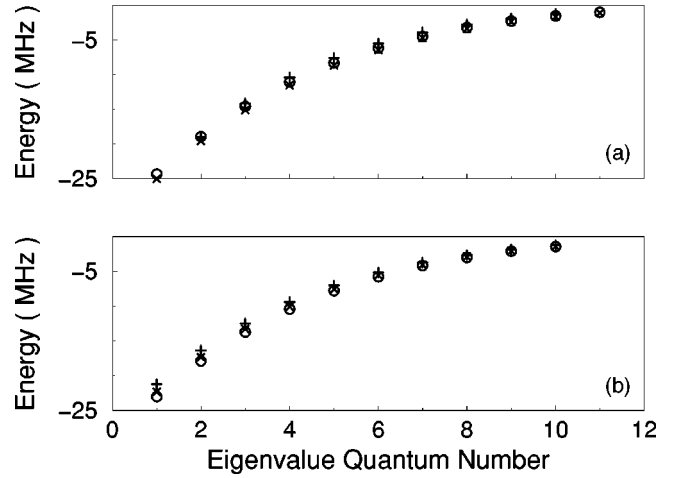

FIG. 3. Dependence of the eigenvalues in the range -25 to -1 MHz as a function of the repulsive short-range potential $A e^{-\alpha z}$ parameters: (a) for $A = 4 \times 10^{13}$ Hz and $\alpha = 2 \times 10^9$ m^{-1}($\times$), 1×10^9 m^{-1}($\bigcirc$), and 4×10^8 m^{-1}(+); (b) for $\alpha = 1 \times 10^9$ m^{-1} and $A = 4 \times 10^{15}$ Hz($\times$), 4×10^{17} Hz($\bigcirc$) and 4×10^{19} Hz(+).

played in Fig. 2. Obviously the deeper levels strongly depend on the strength A and range α^{-1} of the short-range interaction. However, due to the asymmetry of the potential, the upper states are much less sensitive to the short-range repulsive potential parameters as we can see in Fig. 3, where we display the energy of the states in the interval between -25 MHz and -1 MHz for a wide range of A and α values. Since the energy levels remain almost unchanged for the entire range considered, by fitting energy spacing measurements of the long-range vibrational levels, one is able to obtain the C_3 van der Waals coefficient [20].

Let us now consider a loading process scheme:

$$\text{surface} + \text{Cs} + h\nu \; \rightarrow \; (\text{surface})\text{Cs*},$$

$$(\text{surface})\text{Cs*} \; \rightarrow \; (\text{surface})\text{Cs} + h\nu',$$

which describes the photoassociative formation of an excited bound state and its deexcitation to an atom-surface electronic ground state. In fact to populate such atom-surface long-range states we consider cold Cs atoms near a dielectric surface interacting simultaneously with two resonant lasers of frequencies ν and $\nu' = \nu + \delta\nu$, as shown in Fig. 2. The incident lasers direction is normal to the surface and consequently the induced dipoles are parallel to the surface. The complete atomic wave function can be written as a linear combination of the product states,

$$|\Psi(t)\rangle = \sum_{i=1}^{N_g} c^g_i(t)|\psi^g_i\rangle \otimes |g\rangle + \sum_{j=1}^{N_e} c^e_j(t)|\psi^e_j\rangle \otimes |e\rangle, \quad (2)$$

where $\{|\psi^g\rangle, |\psi^e\rangle\}$ are solutions of Eq. (1) with i and j running over the N_g ground and N_e excited states, respectively.

In the interaction picture, and taking into account the rotating-wave and dipole approximations, the semiclassical Hamiltonian is

$$H = \hbar \sum_l \sum_{i,j} \{\Omega_l a^\dagger_i a_j e^{i(\omega_l - \omega_{ij})t} + \Omega^*_l a^\dagger_j a_i e^{-i(\omega_l - \omega_{ij})t}\},$$
$$(3)$$

where a ($a^\dagger$) is the vibrational state annihilation (creation) operator, and Ω_l is the Rabi frequency of the laser l. Considering that the potential well is very deep, a quantum Monte Carlo simulation to observe the temporal evolution of a wave packet is computationally impractical. Instead, we numerically solve the time-dependent Schrödinger equation, with atoms initially in a free ground state which is also solution of Eq. (1), using the asymptotical ($z \to \infty$) momentum p_i. Likewise for typical cold atomic samples this state has a momentum dispersion of just a few $\hbar k$, the conclusions obtained here being realistic. The number of ground and excited states considered in each calculation depends on which levels are chosen to be resonant with the lasers, i.e., it depends on the relation between the laser Rabi frequencies, the vibrational levels energy spacing and the excited-states linewidth Γ. In this calculation all states displayed in Fig. 2 are taken into account and we used values of $\Gamma/2\pi = 5.3$ MHz, and $k = 2\pi/\lambda$, with $\lambda = 852$ nm, which correspond to the D_2 Cs transition. The laser of frequency ν is set to be resonant with a free state $|\psi^g_{free}\rangle$ to an excited bound-state $|\psi^e_{v'}\rangle$ transition, where the excited-state separation to the neighbor levels is larger than the linewidth Γ of these states. The laser of frequency $\nu' = \nu + \delta\nu$ is set to be resonant with the excited state $|\psi^e_{v'}\rangle$ to a ground bound-state $|\psi^g_v\rangle$ transition. We choose the lasers to couple the states $|\psi^g_{free}\rangle$ and $|\psi^g_v\rangle$ via a Raman transition, so that these states are well separated and the lasers do not interact with the same levels. Since the only free state we take into account is the initial state of the in-
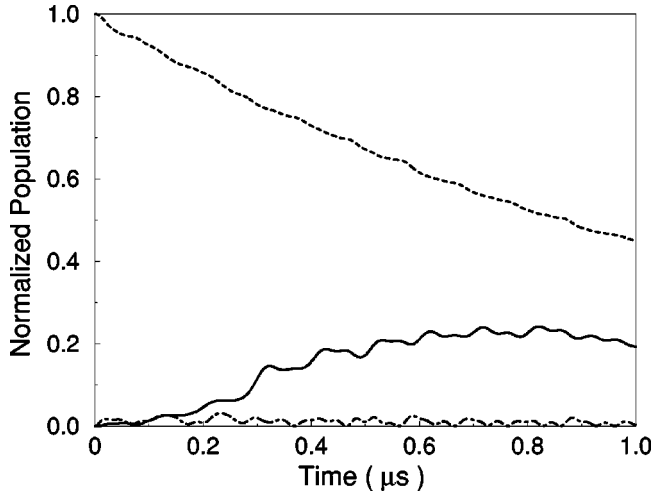
FIG. 4. The temporal evolution of the population for each of three states involved in the Raman transition: the free ground state $|\psi_{free}^g\rangle$ (dashed line), the bound ground state $|\psi_v^g\rangle$ (continuous line), and the bound excited state $|\psi_{v'}^e\rangle$ (dot-dashed line) populations.
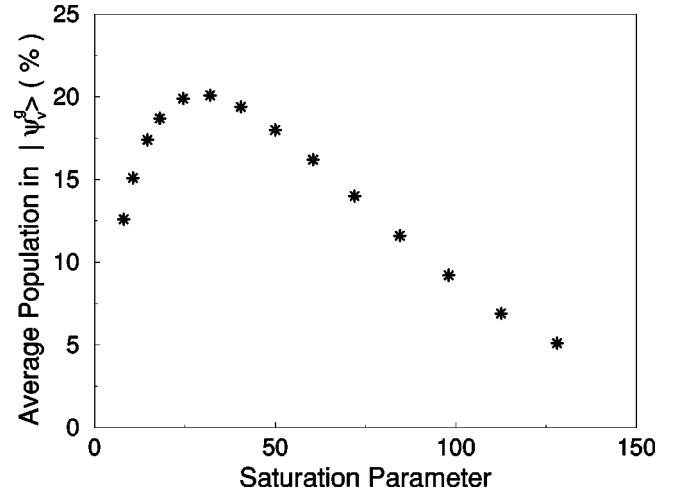

FIG. 5. Efficiency of the loading process as a function of the saturation parameter. The maximal transfer rate of 20% is obtained for $\Omega = 4\Gamma$.

coming atom, we suppose that the spontaneous emission will always drive the atom out of the set of states $\{|\psi^g\rangle, |\psi^e\rangle\}$ we are considering, i.e., we neglect the contribution of spontaneous emission to the ground bound-state population. Therefore the total population $P(t) = \sum_{i=1}^{N_g} |c_i^g(t)|^2 + \sum_{j=1}^{N_e} |c_j^e(t)|^2$ will not be preserved.

The temporal evolution of the normalized population of the free, ground, and excited states involved in the Λ system is shown in Fig. 4 for the Rabi frequencies $\Omega_1 = \Omega_2 = 4\Gamma$ and initial atomic momentum $25\hbar k$. The dynamic is then only considered up to 1 μs, the time by which the atom classically would leave the region of interaction. If instead an initial wave-packet distribution were considered, a smaller interaction time would be compensated by a larger overlap between the free and the excited wave functions. Qualitatively this loading dynamic is equivalent to the interatomic long-range photoassociation [15–17]. The main difference for the ground atomic state is that the van der Waals potential varies as $1/r^3$ for the interaction with the surface and as $1/r^6$ with another ground-state atom.

The population transfered to the bound ground state increases until it reaches an almost stable value averaging 20%. This efficiency as a function of the saturation parameter $s = 2\Omega_1^2/\Gamma^2$ for $\Omega_1^2 = \Omega_2^2$ is shown in Fig. 5. The maximum transfer rate obtained being for the saturation parameter $s = 32$. Turning the lasers off after trapping, we obtain tightly confined atoms, if compared with traps using typical optical potentials, with no heating induced by spontaneous emission. Atoms on these vibrational states are confined in a 2D waveguide, where the binding energies and the energy spacing depend on how deep the atoms are in the surface well. The results obtained for the long-range states are of the same order of magnitude as in the recent proposition of a static planar magnetic waveguide [7]: the energy separation ranges from few to tens of MHz, and the ''bond length'' is of the order of 50 nm.

To discuss the optical observation of such atom-surface bound states one should consider two main aspects of the surface condition: its roughness and temperature. To analyze the actual nature of the surfaces we will refer to the theoretical and experimental results on atomic beam diffraction from solid surfaces [21]. For the mechanism discussed in this paper to load neutral atoms in the surface well we have considered the surface of an ideally planar crystalline transparent dielectric. As usual, we can separate the atomic center of mass coordinates in parallel and perpendicular to the surface, $\mathbf{r} = (\mathbf{R}, z)$, with $\mathbf{R} = (x, y)$. So, to describe the atomic propagation in the van der Waals surface waveguide one should take into account the surface roughness, which results in band structure for the motion in the (x, y) plane, parallel to the surface [22]. Here, nevertheless, the origin (z_0) for the $V(z)$ potential is an average value of the distance to the atomic center of mass [18], i.e., we neglect the surface corrugation in the expression of the wall potential. This is indirectly justified by the very low sensitivity of the near-dissociation energy levels with the short-range potential parameters, as discussed above (cf. Fig. 3). Even for atoms coupled into deeper states (closer to the surface), their wave packet at temperatures below 1 mK [23] is larger than the solid interatomic distances, and therefore, larger than the surface corrugation [24]. With respect to the surface temperature, in our calculations, it was ideally taken as $T = 0$. Meanwhile, we believe that the bound states between cold atoms and a $T \neq 0$ surface may be observed. At first glance, to observe bound states on a surface one may consider the solid temperature T, such that $k_B T$ is less than the energy-levels separation. For the near-dissociation levels treated here this means surface temperatures of the order of 1 mK, which represents very hard experimental conditions. Nevertheless, to observe these bound states we do not need the atom to stay indefinitely physisorbed on the surface and, in fact, the signature of bound states was observed in atomic diffraction experiments for finite temperatures of the surface [25]. Despite sticking studies having been done essentially for light

atoms [26], the atomic interaction with the surface modes can be described by [27] $\Sigma_k f_k(r)(b_k^\dagger + b_k)$, where k indicates the sum on all phonon modes involved, f_k are the coupling constants, and $b^\dagger$ (b) are the phonon creation (annihilation) operators respectively. It was obtained that desorption is essentially due to one-phonon processes [28]. To be observed by the optical techniques proposed here, the bound-states lifetime must be of the order of optical transitions lifetime and this seems to be possible at wall temperatures easily attained in most of the laboratories. Obviously, for high temperatures, only the deeper states will have a lifetime that would allow their observation. In this case the technique discussed here to measure the C_3 van der Waals coefficient may be less precise [29]. In order to observe atoms in the surface bound states, besides the surface thermal excitations coupling, it would also be important to consider the surface contamination condition [30].

To optically probe quantum levels of the physisorbed atoms it is essential to be able to distinguish between the atoms at distances from the surface of less than one optical wavelength, the range of the van der Waals potential, from the other atoms of the cold vapor, usually a cloud of, at least, tens of micrometers [11,12]. Starting from a cold atomic sample very close to a dielectric window and loading the surface well by the Raman process described above, one could use selective reflection (SR) spectroscopy [31] to probe the vibrational levels. The SR signal Φ^{SR} arises from the resonant contribution of the atomic dipoles $p(z)$ to the Fresnel reflection at a dielectric-atomic vapor interface,

$$\Phi^{SR}(\omega - \omega_{ge}) \propto \int_0^\infty dz e^{2ikz} p(z). \qquad (4)$$

Notice that although the integral is calculated over the whole half-space of the vapor ($z \geq 0$) only the atomic dipoles in a layer of thickness $\lambda/2\pi$ significantly contribute to this reflection signal [32]. This spatial selectivity turns the SR particularly adapted to explore processes near an interface. In general one can distinguish between atom-surface interaction, whose SR signal is linearly dependent on the density $n = N_{at}/(\lambda/2\pi)^3$, and the interatomic collisional effects which depend on n^2. Considering a magneto-optical trap (MOT) [33] as the source of atoms, the condition $n \ll 1$ is easily satisfied for all atoms in the probed region, under the van der Waals potential. Previous measurements of the van der Waals shift on atomic lines have been realized, probing by SR atoms with positive energies in the van der Waals potential, i.e., in the continuum of states of the well potential (see Fig. 2) [34]. The SR spectra calculations for the vibrational levels of physisorbed atoms are being performed and will be published elsewhere.

In conclusion, long-range bound atoms in a van der Waals surface potential is of multiple interest: as a dimentionally simple (1D) photoassociative system; as a system in which we can measure the strength of the van der Waals surface forces on the atoms; and as a ''natural'' (intrinsic) static waveguide for atoms in a plane very close to the surface.

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