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Exploring the van der Waals Atom-Surface attraction in the nanometric range

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PACS :	42.50.Xa	Optical tests of quantum theory
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The van der Waals atom-surface attraction, scaling as $C_3 z^{-3}$ for z the atom-surface distance, is expected to be valid in the distance range 1-1000 nm, covering 8-10 orders of magnitudes in the interaction energy. Thanks to a Cs vapor nanocell we analyze the spectroscopic modifications induced by the atom-surface attraction on the $6P_{3/2} \rightarrow 6D_{5/2}$ transition. The C_3 value is found to be independent of the thickness in the explored range 40-130 nm, and is in agreement with an elementary theoretical prediction. We discuss the specific interest of exploring short distances and large interaction energy.

The dipole-induced dipole attraction between neutral bodies is a key phenomenon in the ultimate cohesion of matter and is at the origin of covalent crystals and van der Waals molecules. This van der Waals (vW) type of attraction between fluctuating atomic dipoles is a precursor of the general Casimir interaction [1], whose paramount importance in the quantum theory of vacuum has been recognized recently, encompassing effects related to the need of a "cosmological constant" in general relativity theory, and speculations about the detection of a possible fifth force[2]. The interest for the measurement of the Casimir effect has been largely renewed with the recent upsurge of precision experiments [1,3], whose interpretation now requires to take into account various corrections such as the non-ideality of the materials, surface roughness, and non-zero temperature corrections. Simultaneously, the knowledge of the vW attraction between atomic particles, spanning as $-C_6r^{-6}$ (with r the interatomic distance), now appears as a fundamental need to predict the collisional behavior of a collection of atoms [4], ultimately connected to the possibility for cold atoms to condense according to a Bose-Einstein statistics. Between these two related fundamental problems, an intermediate situation is provided by the atom-surface vW interaction, predicted to span as $-C_3z^{-3}$ (z : the atom-surface distance). To theoretically estimate the C_3 value from the knowledge of atomic structures, an electrostatic mirror image approach is usually satisfactory in many cases. The z^{-3} dependence is expected to be valid for distances allowing to ignore the structural details of the surface (typically ≥ 1 nm), up to the micrometric range, when retardation effects impose the more complete Casimir-Polder description [5].

It remains fascinating to note that, although the vW z^{-3} attraction law should cover about 8-10 orders of magnitude of energy, little has been done to test this predicted dependence in an effective situation. The various developments in nanotechnologies and nanosciences should make it more important to measure effectively these remote forces, as for the Casimir force already known to be a possible limiting factor in MEMS technology [6]. In

the shorter distance limit, the vW attraction is only an asymptotic "long-range" tail of the atom-surface potential, to the exact shape of which surface physics experiments are insensitive [7]. For longer distances, and after pioneering principle experiments using deflection of an atomic beam [8], precision experiments tested the spatial dependence on a beam of Rydberg atoms [9] channeled between two plates separated by at least 500 nm. Following the blossom of experimental measurements of vW interaction exerted on a long-lived level [10,11], the spatial dependence of vW interaction between a ground state Cs atom and a metal surface was recently investigated in the 20-100 nm range [12]. Reflection spectroscopy at an interface, in which we have been involved for a long time [13], offers a unique capability to probe the vW interaction for short-lived excited states, possibly exhibiting exotic behaviors such as vW repulsion [14]. However, the depth of the probed region remains fixed, as governed by the wavelength of the optical probe. Here we report on a method of spectroscopy in a vapor cell of a nanometric thickness ("nanocell") [15], with which we explore the thickness dependence of the vW interaction in the range $d = 40\text{-}130$ nm.

Following the development of extremely thin vapor cells, specific features in linear spectroscopy were demonstrated [15], including Doppler-free properties under normal incidence and an intimate mix-up of absorption and reflection signals [16] through intrinsic Fabry-Perot effects. Transmission (S_T) and reflection (S_R) signals are anyhow derived from independent spatial integrations of the (complex) resonantly induced oscillating atomic dipole response $p(z)$. This response, naturally sensitive to the detuning between the excitation frequency and the atomic resonance (*e.g.* through a resonant Lorentzian response $[\gamma/2 + i(\omega - \omega_0(z))]^{-1}$, with γ the optical width, ω the incident frequency, and $\omega_0(z)$ the vW shifted atomic resonance), is usually nonlocal [16], owing to the transient atomic behavior over the wall-to-wall trajectories. Hence, S_T and S_R integrate a complex interplay of natural width, Doppler broadening and velocity distribution, pressure effects (broadening and/or shift), and spatial

dependence of the vW potential as well. For a *gas* cell, the vW shift, minimal at $z = d/2$, is spatially inhomogeneous, and the lineshapes undergo severe vW broadenings with distortions and asymmetries. In this sense, the observation of an elementary $1/d^3$ dependence of the position of the peak of $S_T(\omega)$ [17] is not a sufficient evidence of the vW dependence. Only for a *beam* of atoms flying at mid-distance from the two windows (see e.g. [9]) a $1/d^3$ dependence of the line-shift (relatively to the free-space resonance) is a rigorous expectation. However, fig. 1 shows that when the vW interaction dominates over other causes of broadening, the spectral shift follows a $1/d^3$ dependence rather well, making the 40-130 nm range sufficient (for our parameters) to explore a factor ≥ 30 in the energy of surface interaction. We have evaluated the theoretical lineshapes S_T and S_R for a real gas and the comparison between the experimental lineshapes and the theoretical ones is the core of the present work. Our complete calculation includes the atomic motion, and multiple electrostatic images for the vW potential (although the two-wall potential, *i.e.* $V(z) = -C_3\{z^{-3} + (d-z)^{-3}\}$, differs by less than 5 % when the dispersion of the dielectric window [14] is negligible). Our experiments were conducted on the $6P_{3/2} \rightarrow 6D_{5/2}$ transition of Cs (917 nm) (requiring a pumping on the 852 nm D_2 resonance $6S_{1/2} \rightarrow 6P_{3/2}$). The vW shift of the transition mainly originates in the vW interaction exerted onto the high-lying 6D level [14], and dominates over other broadenings in the explored distance range. The $6D_{5/2}$ sublevel was chosen because it does not exhibit atomic couplings in strong resonances with the YAG windows of the nanocell. Because the thickness of a nanocell varies locally [15], translating the nanocell allows monitoring the influence of the thickness under unchanged experimental conditions. The local thickness d is determined in a two-wavelength reflection measurement (auxiliary He-Ne, and 852 nm beam) [15]. The accuracy (1-5nm) notably depends on the local window parallelism at the considered spot. S_T and S_R spectra are simultaneously recorded, and thicknesses $d = 40$ nm, 50 nm, 65 nm, 80 nm, 100 nm and 130 nm were analyzed. An auxiliary 917 nm saturated absorption (SA) signal in a

macroscopic cell provides a frequency reference for the transition in free-space. For each spot, in an approach analogous to the one developed in [14], several Cs densities *-i.e.* temperature- were investigated. This allows a consistent extraction of the C_3 measurement in which the influence of Cs pressure is removed. Similarly, a variety of pumping conditions was explored to assess the spatial homogeneity of the $6P_{3/2}$ population, as our modeling assumes a homogeneous and thermal population of atoms.

The analysis relies on fittings of the experimental spectra with the family of C_3 curves calculated for a given thickness d . The strong constraints on the νW shift and lineshape restrict to a narrow-range the C_3 values providing a satisfactory (least-square) fitting. The dominant νW spatial broadening makes fittings nearly insensitive to the adjustment of the width of the optical transition. The main results of our systematic analysis are two-fold : (i) for a given experiment, S_T and S_R signals are consistent in shape [18] and amplitude as well [19]; this consistency is quite remarkable because it appears in spite of independent determinations for the fitting, and of unrelated lineshapes; (ii) the optimal fitting C_3 values are found to be independent of the thickness. In fig.2, one observes that the various experiments accurately yield a $\sim 14 \pm 3 \text{ kHz} \cdot \mu\text{m}^3$ value. For the smallest thickness (*i.e.* dominant νW contribution) these values are, as expected, independent on the Cs pressure, although a pressure broadening can be observed. For $d \geq 80\text{nm}$, the νW effect becomes partly hindered by pressure effects, with a trend to an apparent lower C_3 value when Cs density increases, and its accuracy is affected by uncertainties concerning the SA reference frequency : owing to nonlinearities in the D_2 pumping, selective pumping to the various $6P_{3/2}$ hyperfine sublevels (h.f.s. up to 450 MHz) may occur. Figure3 illustrates the consistency of our results, where a single set of parameters fits all spectra recorded at various thicknesses in otherwise identical conditions. Note that the residual discrepancies would disappear with more individual adjustments, while not increasing in the C_3 uncertainty.

Notwithstanding the excellent agreement between the experimental data and the theoretically modeled curves, some reproducibility issues are worth being discussed. Indeed, the measured thickness is a laser-spot- average (diameter $\sim 100 \mu\text{m}$) of a local nanometric wall-to-wall distance d . Because of the local surface roughness (estimated to 1-2 nm), and the defects in planarity or parallelism (the minimal observed slope of the nanocell is $\sim 10 \text{ nm}$ for 1mm [15]), the vW interaction could vary for comparable spots as $\langle d \rangle^{-3} \neq \langle d^3 \rangle$. Testing the reproducibility is also a way to monitor the influence of local residual stray electric fields at the surface: it was indeed recognized in [9] that imperfections of the metal coatings are sources of a Stark shift competing with the vW interaction for excited Cs levels such as Rydberg levels. Stronger residual fields are expected for a dielectric window. Static charges and random distribution of crystalline domains have been apparently responsible [20] for erratic signals in an experiment derived from [9], but with an atomic beam channeled between dielectric walls. Here, the nanocell technology allows the comparison between supposed-to-be identical experiments with respect to the nominal (average) thickness, instead of averaging over the whole length traveled by the atomic beam [9], or on a large spot size as with bouncing of cold atoms [10,12]. Reproducibility is not questionable for the various investigated spots with $d \geq 65 \text{ nm}$. At $d = 50 \text{ nm}$, several spots -but not all of them- produce rigorously identical spectra: they are those here analyzed, as apparently immune to random defects -*e.g.* roughness, local Stark shifts- of the surface. For $d = 40 \text{ nm}$, sensitive variations are observed from spot to spot. However, except for a few erratic behaviors clearly out of the considered family of lineshapes, minor irreproducibilities do not prohibit to fit a single lineshape, as exemplified in fig.3. Remarkably, for these small thicknesses (*i.e.* very strong vW interaction), minor changes in the fitting parameters suffice to interpret visible variations in the lineshapes. This is why we estimate that the major causes of irreproducibility are

connected to uncertainties, at the level of a few nm, in the local geometry of the two planar windows, rather than to the effect of stray fields.

Our estimated value ($C_3 = 14 \text{ kHz} \cdot \mu\text{m}^3$) is also in very good agreement with the usual theoretical prediction. For a Cs atom in front of a single ideal reflector, one predicts $C_3(6D_{5/2}) = 24 \text{ kHz} \cdot \mu\text{m}^3$, $C_3(6P_{3/2}) = 4 \text{ kHz} \cdot \mu\text{m}^3$, and in front of a YAG window [21] : $C_3^{\text{YAG}}(6D_{5/2}) = 17 \text{ kHz} \cdot \mu\text{m}^3$, $C_3^{\text{YAG}}(6P_{3/2}) = 2 \text{ kHz} \cdot \mu\text{m}^3$, leading to a C_3 value of $15 \text{ kHz} \cdot \mu\text{m}^3$ for our *spectroscopic* experiment [*i.e.* $C_3 = C_3(6D_{5/2}) - C_3(6P_{3/2})$]. Although remarkable, this agreement could be partly coincidental in view of various pitfalls, specific to our experiment, or of a more general nature and affecting the theoretical estimates. Evaluating the dielectric image factor applicable to Cs($6D_{5/2}$) is delicate [21] because the situation is not purely nonresonant: the virtual *emission* coupling Cs($6D_{5/2}$) to 7D (in the $\sim 15 \mu\text{m}$ region) requires the precise knowledge of the YAG spectrum and of its surface modes. Moreover, in a nanocell [22], the atom couples to a guided mode structure, and the two-wall model, or its straightforward extension to multiple electrostatic images model, may reveal too elementary. Also, the orientation of the atom is commonly assumed to be isotropic, implying equal contributions for the dipole fluctuating along the normal, and parallel to the interface. However, the atomic state could undergo a Zeeman polarization under the influence of the D_2 line pumping (the irradiation, being under near normal incidence, is polarized *parallel* to the surfaces), or through the polarized excitation of the 917 nm laser. This would restrict the summing over the virtual dipole transitions connecting the $6D_{5/2}$ level to some directions only, hence reducing the observable C_3 value [22]. More generally, a recent work [23] shows that at the smaller distances, C_3 should exhibit a spatial dependence $C_3(z)$, owing to the non retarded, and hence stronger, contribution of the highly energetic transitions involving the electronic core [4]. This dependence, not presently reported [10,12] in experiments, is expected to be more sensitive for a ground state than in our case of a high-lying state [13].

To summarize, we have demonstrated the z^{-3} shape of the vW potential in a distance range 40-130 nm, for which the energy shift covers a factor ~ 30 , through linear spectroscopy in a nanocell. This technique could allow testing in the same circumstances the rich physics of the interaction with dielectric media, including a possible resonant coupling with surface modes. More generally, the distances investigated here are an order of magnitude below those explored years ago for excited atoms in Rydberg levels [9], and compares favorably with those currently investigated in precision Casimir measurements [3] or with the one addressed in the recent vW interaction experiments with ground state atoms [12]: in our situation, the vW shift, whose minimal value is twice the one undergone by an atom at a distance $d/2$, is equivalent to the one of an atom located at $\sim 0.4d$ from a single wall. The combined effect of the transient regime and the steepness of the vW potential make the central region of the nanocell spectrally dominant, as can be inferred from fig.1. Hence, for each investigated thickness d , the obtained C_3 evaluation is nearly free of spatial averaging. Lowering the atom-surface distance below 10 nm appears a realistic objective ($d \sim 20-30$ nm is reported in [17]). This is in contrast with the vW measurements based upon the reflection of slow atoms [10,12], that are limited by the considerable forces needed to equilibrate the vW attraction and which explore a closest approach distance. Similarly, the techniques used for Casimir interaction, of atomic force microscopy, are hardly compatible with too high a pressure (the standard calculation predicts a Casimir interaction exceeding the atmospheric pressure for $d \leq 10$ nm). Finally, in the small distance regime that we explore here, the interaction energy spans up to ~ 5 GHz (for 2 walls), a value much larger than the one for all previous investigations, for which cooled atoms would be considerably accelerated (20 nm away from one of the wall, $C_3 = 14 \text{ kHz} \cdot \mu\text{m}^3$ yields an acceleration $\sim 8 \cdot 10^7 \text{ g}$). A realm of exotic effects, such as a gradient of density for the atomic gas in the extreme vicinity with the surface, is also opened for these small distances. It is even worth mentioning that, in the absence of experimental evidence,

the regime of Casimir-related interaction remains under debate [24]. This range of very short distances appears also of a significant interest in view of some theories predicting non Newtonian gravity [25].

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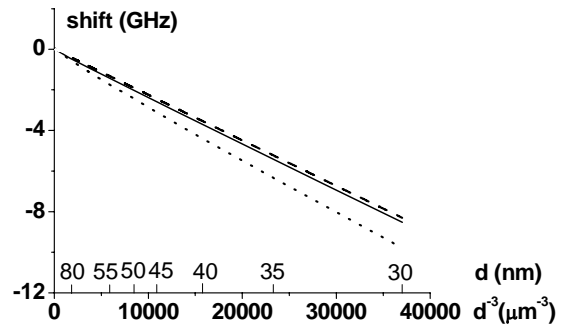
Figure captions

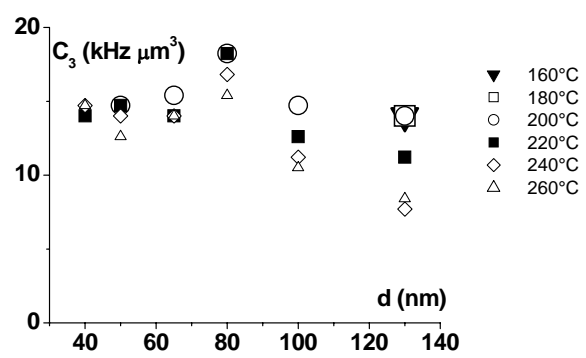
Figure 1: νW -induced spectral shift between the S_T peak and the free-atom resonance as a function of the thickness. The calculation is performed for: $C_3 = 14 \text{ kHz} \cdot \mu\text{m}^3$, $\gamma = 300 \text{ MHz}$.

The dotted line is for a gas of atoms flying wall-to-wall with a 250 MHz Doppler width. The dashed line, and the continuous line, are respectively for a model for beam of atoms flying at mid-distance $d/2$, and for a gas of motionless atoms.

Figure 2 : Optimal fitting C_3 values found for various thickness and Cs temperatures.

Figure 3: Experimental lineshapes (S_T and S_R) recorded on the 917 nm transition for various thicknesses. The frequency scans are continuous, or discrete (for 40 nm and 50 nm). The vertical dashed line is a marker for the SA resonance. The fittings (dotted lines) are based upon the optimal parameters for S_T (50 nm), found to be $C_3 = 14 \text{ kHz} \cdot \mu\text{m}^3$, $\gamma = 300 \text{ MHz}$. Amplitude adjustment factors are introduced to compensate for the thickness dependence of the efficiency of the $6P_{3/2}$ pumping. For 40 nm: respectively 3.3 and 4.8 for S_T and S_R ; for 65 nm : 0.25 and 0.24; for 80 nm: 0.19 and 0.15; for 100 nm: 0.11 and 0.07; for 130 nm: 0.07 and 0.04. The Cs nanocell temperature is 200°C. The pumping frequency is locked onto the $4 \rightarrow 4-5$ crossover of the D_2 line, pump power is $\sim 1 \text{ mW}$ focused on a diameter $\sim 100 \mu\text{m}$, pump absorption reaches 25 % for 130 nm. The 917 nm transmission change is $\sim 5 \cdot 10^{-4}$ for 50 nm.

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