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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
	34.50.Dy	Interactions of atoms and molecules with surfaces; photon and electron emission; neutralization of ions
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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 for  $\sim 1-1000$  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, extracted independently for various thicknesses ranging from 40 nm to 130 nm, is found to be independent of the thickness. It agrees, but only within a factor of 2, with an elementary theoretical prediction, whose validity is discussed.*

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. The interest for the measurement of the Casimir effect has been largely renewed with the recent upsurge of precision experiments [1,2], 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 [3], 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). The  $C_3$  value can usually be predicted through an electrostatic mirror image approach, satisfactory in many cases, and with the knowledge of atomic structures. The  $z^{-3}$  dependence is expected to be valid for distances allowing to ignore the structural details of the surface (typically  $\geq 1$  nm), up to the micrometric range, when retardation effects impose the more complete Casimir-Polder description [4].

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 [5]. 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 [6]. For the longer distances, and after pioneering principle experiments using deflection of an atomic beam [7], precision experiments tested the spatial dependence on a beam of Rydberg atoms [8] channeled between two plates separated by 500 nm at least. Following the recent blossom of experimental measurements of vW interaction exerted on a long-lived level [9], the spatial dependence of vW interaction between a ground state Cs atom, and a metal surface, was very recently investigated in the 20-100 nm range[10]. Spectroscopy at an interface, in which we have been involved for a long time[11], offers a unique capability to probe the vW interaction for short-lived excited states, possibly exhibiting exotic behaviors such as vW repulsion [12]. However, reflection spectroscopy in a macroscopic cell probes a fixed vicinity from the surface. Here we report on a method of spectroscopy in a vapor cell of a nanometric thickness ("nanocell") [13], that allows us to explore the thickness dependence of the vW interaction in the range  $d = 40-130$  nm.

Following the development of extremely thin vapor cells, specific features in linear spectroscopy were demonstrated [13], including Doppler-free properties under normal incidence. and an intimate mix-up of absorption and reflection signals[14] through intrinsic Fabry-Perot effects. However, transmission ( $S_T$ ) and reflection ( $S_R$ ) signals are derived from independent spatial integrations of the (complex) resonantly induced oscillating atomic dipole response  $p(z)$ . This dipole response  $p(z)$  is naturally sensitive to the detuning between the excitation frequency and the atomic resonance (*e.g.* through a resonant Lorentzian response  $[\gamma/2 + i(\omega - \omega_0(z))]$ <sup>-1</sup>, with  $\gamma$  the optical width,  $\omega$  the incident frequency, and  $\omega_0(z)$  the vW shifted atomic resonance). The dipole response  $p(z)$  is usually nonlocal [14], owing to the transient atomic behavior over the wall-to-wall trajectories. This implies a complex interplay between the atomic response, the spatial dependence of the vW potential, the natural width,

the Doppler broadening, and the pressure effects (broadening and/or shift). Hence, the expectation of a  $1/d^3$  dependence of the line-shift (relatively to the bulk resonance) is satisfied only for a *beam* of atoms flying at mid-distance from the two windows (see e.g. [8]). For a *gas* cell, the vW shift, minimal at  $z = d/2$ , is spatially inhomogeneous, and the lineshapes undergo severe distortions and asymmetries along with a vW broadening. In this sense, the observation of an elementary  $1/d^3$  dependence of the position of the peak of  $S_T(\omega)$  [15] is not a sufficient evidence of the vW dependence. Fig. 1 shows anyhow that when the vW interaction dominates over other causes of broadening, the spectral shift varies with an approximate  $1/d^3$  dependence, making the 40-130 nm range sufficient (for our parameters) to explore a factor  $\geq 30$  in the energy of surface interaction. In a complete calculation [16], we have evaluated the theoretical lineshapes  $S_T$  and  $S_R$  for a real gas, including atomic motion and multiple electrostatic images for the vW potential (actually, the difference with an elementary two-wall potential, *i.e.*  $V(z) = -C_3[z^{-3} + (d-z)^{-3}]$ , does not exceed 5 % when the dispersive properties of the dielectric windows [12] are neglected). The comparison between the experimental lineshapes and the theoretical ones is the core of the present work.

Our experiments were conducted on the  $6P_{3/2} \rightarrow 6D_{5/2}$  transition of Cs (917 nm) (requiring a simultaneous pumping on the at 852 nm  $D_2$  resonance  $6S_{1/2} \rightarrow 6P_{3/2}$  at 852 nm). The vW shift of the transition mainly originates in the vW interaction exerted onto the high-lying 6D level [12], and the explored distance range makes it dominant over other broadenings. The  $6D_{5/2}$  sublevel was chosen because it does not exhibit strong resonances with the YAG windows of the nanocell. Because the thickness of a nanocell varies locally [13], investigation of various spots in it, under unchanged experimental conditions, is a way to monitor the influence of the thickness. The local thickness  $d$  is determined in a two-wavelength reflection measurement (auxiliary He-Ne, and 852 nm beam) [13]. 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 the bulk. For each spot, in an approach analogous to the one developed in [12], several temperature conditions 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  $\nu W$  shift and lineshape restrict to a narrow-range the  $C_3$  values providing a satisfactory (least-square) fitting. The dominant  $\nu W$  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 [17] and amplitude as well [18]; this consistency is quite remarkable because it appears in spite of independent determinations for the fitting, and of unrelated lineshapes; (ii) as a key evidence of the  $\nu W z^{-3}$  behavior, the optimal fitting  $C_3$  values are found to be independent of the thickness (fig.2). One observes an accurate convergence to a  $\sim 7.5$  kHz. $\mu\text{m}^3$  value for small thickness. The larger uncertainty ( $\pm 2$  kHz. $\mu\text{m}^3$ ) appearing for  $d \geq 80$ nm, when the  $\nu W$  shift is not very large, mostly originates in the choice of an appropriate SA reference. Indeed, the  $D_2$  pumping in the nanocell may induce an unbalanced, but hard to control, population of the various  $6P_{3/2}$  hyperfine sublevels (h.f.s. up to 450 MHz). An additional illustration of the overall consistency obtained in the fittings is provided in fig.3, where spectra recorded at various thicknesses in otherwise identical conditions are all fitted with a single set of parameters. The residual slight discrepancies that can be observed appear only because of the choice of a

unique set of fitting parameters, and disappear, with no increase in the  $C_3$  uncertainty, with more individual adjustments.

Notwithstanding the excellent agreement between the experimental data and the theoretically modeled curves, some reproducibility issues are worth being discussed. Indeed, the measured thickness averages over the laser spot (diameter  $\sim 100 \mu\text{m}$ ) a local nanometric wall-to-wall distance  $d$ , sensitive to local surface roughness (estimated to 1-2 nm) and defects in planarity or parallelism (the minimal observed slope of the nanocell is  $\sim 10 \text{ nm}$  for 1mm [13]). This may result in variations of the observed vW interaction 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 [8] 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 for erratic signals [19] in an experiment derived from [8] but with an atomic beam channeled between dielectric walls. Here, the nanocell technology allows the comparison between experiments supposed-to-be identical with respect to the nominal (average) thickness, instead of averaging over the whole length traveled by the atomic beam [8], or on a large spot size as with bouncing of cold atoms [10]. We have observed a perfect spectral reproducibility for the various investigated spots with  $d \geq 65 \text{ nm}$ . At  $d = 50 \text{ nm}$ , several spots -but not all of them- are found to 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, imperfections in the reproducibility have not prevented us from the possibility of a fitting for individual spot, as exemplified in fig.3. Remarkably, for these small thicknesses that induce a very strong vW

interaction, minor changes in the fitting parameters suffice to interpret visible variations in the lineshapes. Hence, 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.

It is also worth comparing our estimated  $C_3$  value ( $\sim 7.5 \text{ kHz}\cdot\mu\text{m}^3$ ) to theoretical predictions, yielding for a Cs atom in front of a single ideal reflector:  $C_3(6D_{5/2}) = 24 \text{ kHz}\cdot\mu\text{m}^3$ ,  $C_3(6P_{3/2}) = 4 \text{ kHz}\cdot\mu\text{m}^3$ , and in front of a YAG window [20] :  $C_3^{\text{YAG}}(6D_{5/2}) = 17 \text{ kHz}\cdot\mu\text{m}^3$ ,  $C_3^{\text{YAG}}(6P_{3/2}) = 2 \text{ kHz}\cdot\mu\text{m}^3$ . This leads for our *spectroscopic* experiment [*i.e.*  $C_3 = C_3(6D_{5/2}) - C_3(6P_{3/2})$ ] to a predicted  $C_3$  value  $\sim 15 \text{ kHz}\cdot\mu\text{m}^3$ , a value about twice larger than found in our experiments. Several reasons are susceptible to explain this slight discrepancy:

(i) on the experimental side, the modeling assumes the initial level to be populated in a spatially uniform manner and with a thermal velocity distribution. This may be not totally valid for an initial level such as the resonant  $6P_{3/2}$  level, that has required a prior pumping that could be more efficient for the slowest atoms. However the quality of the fittings, and the relative insensitivity of the vW shift to the velocity distribution (see fig. 1), show that the discrepancies with the assumptions of the model should remain of a modest influence.

(ii) On the theoretical side, the evaluation of the dielectric image factor applicable to  $Cs(6D_{5/2})$  is delicate [20]: even for a single wall, it requires a precise knowledge of the YAG spectrum and of its surface modes, notably in the  $\sim 15\mu\text{m}$  region, where a virtual *emission* couples  $Cs(6D_{5/2})$  to 7D. Moreover, in a nanocell [21], the coupling is actually between atomic transitions and a guided mode structure: the elementary two-wall model, or the refined multiple electrostatic images model, may become partly improper to deal with the resonant couplings occurring through the dispersive properties of YAG.

(iii) In the standard theoretical evaluation, an equal contribution of the dipole fluctuations along the normal, and parallel to the interface is assumed. However, because the  $D_2$  line



pumping laser is polarized *parallel* to the surfaces –near normal incidence irradiation- , and with the polarized excitation of the 917 nm laser, the preparation of Cs ( $6D_{5/2}$ ) atoms in the nanocell is most probably non-isotropic with respect to the various Zeeman sub-states. Hence, the summing over the virtual dipole transitions connecting the  $6D_{5/2}$  level may be restricted to some directions only. This would reduce, by up to a factor of  $\sim 2$  [21], the observable value of  $C_3$ .

In conclusion, linear spectroscopy in a nanocell has enabled us to demonstrate the  $z^{-3}$  shape of the vW potential, in a distance range (40-130 nm) for which the energy shift spans from  $\sim 150$  MHz to  $\sim 5$  GHz (for 2 walls) and covers a factor  $\sim 30$ . In spite of negligible stray fields, the experimental  $C_3$  value is found in discrepancy with an elementary prediction. This should impulse more elaborate theoretical developments. The rich physics of the interaction with dielectric media, including a possible resonant coupling with surface modes, can also be addressed with our technique. More generally, the distances investigated here, falling an order of magnitude below those explored years ago for excited atoms in Rydberg levels [8], compares favorably with the range currently investigated in precision Casimir measurements [2] and in the recent vW interaction experiments with ground state atoms [10]: the vW shift observed in nanocells is indeed equivalent to the one for a single wall at  $\sim 0.3 d$  (see fig.1). Moreover, for short distances, vW measurements based upon the reflection of slow atoms [9-10] are intrinsically limited by the considerable forces needed to equilibrate the vW attraction, and the current atomic force microscopy techniques developed for the measurement of Casimir interaction would hardly be compatible with a high pressure (the standard calculation predicts a Casimir interaction exceeding the atmospheric pressure for  $d \leq 10$  nm). In contrast, nanocell spectroscopy appears feasible for even smaller nanocell thickness ( $d \sim 20$ -30 nm is reported in [15]). Finally, it is worth noting that vW strength is huge at small distances (from our experiments, the atomic acceleration 20 nm away from one

of the wall can be estimated to  $\sim 4 \cdot 10^7$  g) and may induce a gradient of density for the atomic gas in the extreme vicinity with the surface, and that more generally, the very short distance regime of Casimir-related interaction remains under debate [22].

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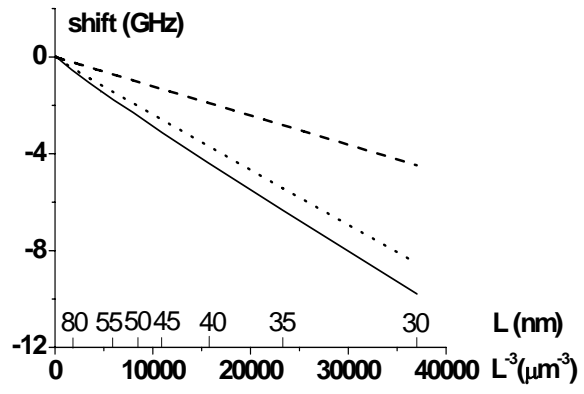
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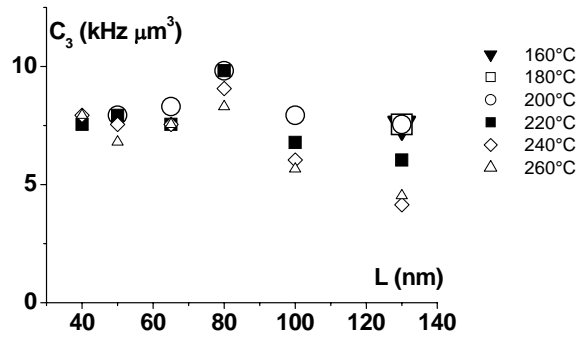
### Figure captions

Figure 1:  $\nu W$ -induced spectral shift between the  $S_T$  peak and the free-atom resonance as a function of the thickness. The calculation is performed for:  $C_3 = 7.5 \text{ kHz} \cdot \mu\text{m}^3$ ,  $\gamma = 300 \text{ MHz}$ . The dashed line is for a beam of atoms flying at mid-distance  $d/2$ , the dotted line for a gas of motionless atoms, the full line for a gas of atoms flying wall-to-wall with a 250 MHz Doppler width.

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). 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.

M.Fichet *et al*, Figure 1

M.Fichet *et al*, Figure 2

M.Fichet *et al*, Figure 3