

## Probing an atomic gas confined in a nanocell

I Maurin, P Todorov, I Hamdi, A Yarovitski, G Dutier, D Sarkisyan\*, S Saltiel\*\*,  
M-P Gorza, M Fichet, D Bloch and M Ducloy

Laboratoire de Physique des Lasers, UMR 7538 du CNRS et de l'Université Paris13,  
99 Av JB Clément, F-93430 Villetaneuse, France

\* Institute for Physical Research, Armenian Academy of Sciences,  
Ashtarak2, Armenia

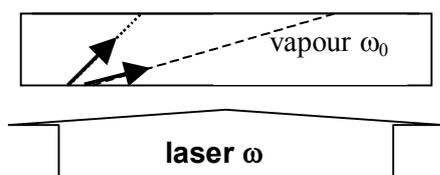
\* Permanent address: Department of Physics, University of Sofia, Sofia , Bulgaria

E-mail: bloch@lpl.univ-paris13.fr

**Abstract.** Since the recent realization of extremely thin vapour cells (local thickness: 20-1000 nm), we investigate the optical properties of these 1-D confined vapours. Aside from their interest for Doppler-free spectroscopy, nanocells offer a new tool to evaluate collisional shift and broadening, yielding an access to the open problem of collisions under confinement. It also allows probing of the atom-surface interaction in a range of unusual short distances. The experimental exploration of the distance dependence, normally evolving according to the  $z^{-3}$  van der Waals (vW) dependence ( $z$  : the atom-surface distance), is worth doing because it could be affected by imperfections of the real surface, such as roughness, adsorbed impurities or charges. A detailed lineshape analysis is now under progress, with tight constraints imposed to the fitting by the twin information brought by simultaneous reflection and transmission spectra. Another issue is a possible resonant enhancement, susceptible to induce a repulsive vW, due to the coupling between atom excitation and a surface mode.

### 1. Introduction: Micro- and nano-cells of dilute vapours

For a very dilute gas, the atomic mean free path can be governed by the container geometry. A specific design, with parallel windows and a very small thickness, enables one to induce a strong anisotropy of the "mean" free path in a vapour cell, with atoms flying from wall to wall. For such thin cells, the response of the "slow" atoms (with respect to the normal velocity, *i.e.* the velocity component along the normal to the windows) to an interaction with resonant light undergoes a relative enhancement, owing to the transient build-up of the resonant interaction, which makes faster atoms less efficient (figure1).



**Figure 1.** Atoms with nearly parallel velocity, insensitive to the Doppler shift, experience a long interaction with the optical field, and exhibit the fully developed steady-state response.

This had provided the principle of a novel method for Doppler-free spectroscopy, applicable to a variety of situations (velocity-dependent optical pumping, linear absorption, two-photon transition, *etc.*) [1] as long as the irradiation is under normal incidence. The initial demonstrations were operated with glass cells of commercial origin, filled-up with alkali-metal vapours, and whose thickness (10-1000 $\mu\text{m}$ ) remained much larger than the optical wavelength  $\lambda$ .

The recent fabrication of extremely thin cells (ETC) of vapour [2], designed with an initial sub-micrometric spacer between two carefully polished parallel windows, has revealed that under the effect of the external atmospheric pressure, the local cell thickness can become extremely small, typically spanning from 20 nm to 1 $\mu\text{m}$ . This truly permits to investigate "nanocells" of vapour, and has opened a realm of novel prospects, that even extends beyond sub-Doppler spectroscopy, with the additional possibility of detecting atom-surface interaction effects in an unexplored range of distances.

Intrinsic to the extremely small thickness of ETCs is an accurate parallelism of the windows (*e.g.* deviation < 1 $\mu\text{m}$  over a 10mm transverse extension) that implies a Fabry-Perot behaviour. On the one hand, this behaviour reveals very convenient to evaluate the local thickness of the nanocell [3], on the other hand, it unavoidably mixes-up the complementary information associated to reflection and transmission spectroscopies [4].

## 2. Spectroscopy in an ETC.

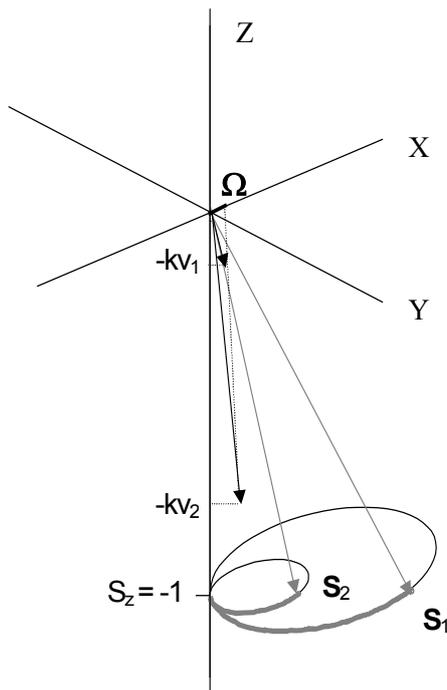
When applying transmission spectroscopy to a micro-cell, the lineshape naturally exhibits sub-Doppler features, whose exact characteristics depend on the cell thickness, type of the considered transition (*e.g.* 2-level vs. 3-level, two-photon or Raman transition), incident intensity (saturation processes), *etc.* Although most often appearing only as a small narrow contribution -peak or dip- over a broad Doppler-broadened background, these sub-Doppler features [1] can usually be singled out through a frequency-derivation of the spectrum, as made possible by FM (frequency-modulated) spectroscopy, yielding a Doppler-free signal.

With the advent of sub-micrometric thin cells, the whole spectrum easily turns to a pure Doppler-free spectrum. In fluorescence detection, the width of the excitation process decreases continuously when the ETC thickness is made shorter: this essentially corresponds to a more demanding velocity selection. This decrease can be typically observed down to the natural width, up to the regime when the interaction time is too limited, even for slow atoms, and implies finally a broadening for the smallest thickness. In contrast, the transmission spectrum, aside from the already mentioned Fabry-Perot effect (with a  $\lambda/2$  periodicity), exhibits complex variations with the nanocell length. Indeed, the linear atomic response features a *coherent* spectroscopic narrowing, that extends, to the optical domain, the original observation by Romer and Dicke [5] in the microwave domain, of a *coherent* spectroscopic narrowing in a gas sample with a thickness  $\sim \lambda/2$ . Additionally, a periodical revival (with a  $\lambda$ - pseudo-periodicity) of the Dicke-narrowing [3,6] was demonstrated.

This coherent spectral narrowing [3,6,7] relies on the fact that, when an atom leaves the wall, the sudden atom excitation induced by *on-resonance* light starts to precess in phase with the electromagnetic field at the wall position, but falls out-of-phase with the local driving field under the effect of the atomic motion, with a phase mismatch finally reaching  $kL$  ( $L$ : the cell thickness,  $k$  the wave number). Such a description, relying on a time domain analysis rather than the more common frequency-domain analysis, is typical of the Dicke approach of the Doppler effect (see *e.g.* [8]) and enables one to recognize that the sub-wavelength confinement makes the Doppler effect unobservable.

A simple Bloch vector model [6] applied to ETC spectroscopy, permits to recover major predictions, and even provides hints for more complex situations. One knows that the Bloch vector  $\mathbf{S}(z, v_z) = (S_x, S_y, S_z)$  characterizing the local density matrix of a two-level atom, precesses along the pseudo-magnetic field  $\mathbf{B}$ , according to  $d\mathbf{S}/dt = \mathbf{S} \times \mathbf{B}$ , with  $\mathbf{B} = (\Omega, 0, \delta = \delta_0 - kv_z)$ , where  $\Omega$  is the Rabi frequency, and  $\delta$  the frequency detuning between the irradiation and the resonance of the two-level system. In the Bloch vector model, the  $S_y$  component is associated to the component of the complex optical coherence needed to calculate the absorption, while  $S_z$  is the population inversion, characterizing the local amount of instantaneous fluorescence. In an ETC, the absorption, and

fluorescence, are respectively estimated from  $\int_0^L S_y(z)dz$  and from  $\int_0^L [1+S_z(z)]dz$  for each velocity group. Hence, on line centre ( $\delta_0=0$ ), one has  $\mathbf{B} = (\Omega, 0, -kv_z)$ , and the Bloch vector, precessing around  $\mathbf{B}$  on the relevant cone, travels an angle independent of the velocity: the angular velocity dependence  $kv_z$  is indeed compensated for by an interaction time  $L/v_z$  (figure 2). Hence, the local absorption, as governed by  $S_y$ , accumulates up to a length  $\lambda/2$ , while the cell region between  $\lambda/2$  and  $\lambda$  interferes destructively with the first  $\lambda/2$  section of the cell, and so on, explaining the revival. For a frequency-detuned irradiation ( $\delta_0 \neq 0$ ), the precession angle of the atomic excitation becomes velocity-dependent, leading to a smoother length dependence of the signal. This justifies for  $L=\lambda/2$  the contrast between the line centre and the wings, and the apparent "Dicke narrowing" of the lineshape. This leads to the  $L = (2n+1)\lambda/2$  revival of the narrow peaks. The dipole relaxation actually reduces the contrast of this lineshape periodicity. As far as the fluorescence emission is concerned, the various cell regions add incoherently, implying a continuous growth of the fluorescence with the cell length. In addition, the fluorescence is a second-order process, limited through a genuine velocity selection to those atoms slow enough to undergo a successive absorption and emission process. This makes its lineshape narrower than the transmission profile, even for the optimal  $L= \lambda/2$  situation, and comparable to the one obtained in FM transmission, when the central logarithmic singularity is turned into a Doppler-free contribution.



**Figure 2.** This representation of the motion of the Bloch vector ( $\mathbf{S}$ ) shows that it rotates the same angle for small ( $v_1$ ) and large ( $v_2$ ) velocities when precessing around the pseudo magnetic field. The integrated  $S_y$  component increases up to an angle  $\pi$ , and the path described by  $\mathbf{S}$  reveals proportional to  $1/v_z$  (for negligible Rabi frequency  $\Omega$ ): this explains the logarithmic singularity.

Saturation effects in nanocells, experimentally investigated in some works [6,9], tend to washout the main features of the Dicke coherent narrowing, although the resonances remain narrow. This washout can also be understood [10] with the Bloch vector model. Incoherent transfer through optical pumping, as occurs in an *open* two-level system, modifies the length of the Bloch vector, and tends to randomize the interferences between the various spatial and velocity contributions. For a strongly-driven *close* two-level system, the Bloch vector model shows that the optimal cell length for a given velocity group is no longer  $\lambda/2$ , and becomes velocity-dependent. Moreover, for a degenerate two-level system, one has to consider various time scales owing to the different Rabi frequencies involved with the Zeeman sub-states. Although saturation effects remain in the frame of a coherent excitation,

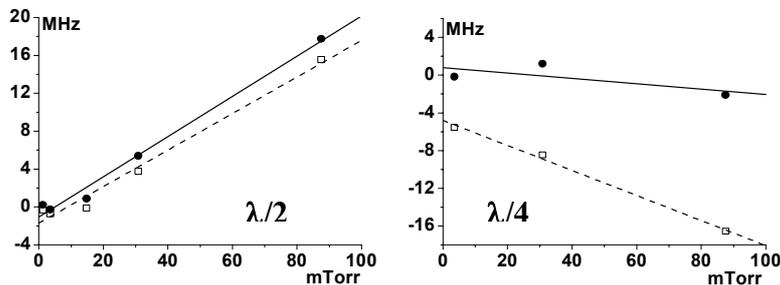
the thin cell spectroscopy intrinsically explores the dynamics of the atomic response, on a time scale that depends on the velocity distribution. This complexity of the time sequence of the nonlinear process tends to hinder any recognizable Dicke-type pseudo-periodicity.

### 3. Atomic collisions under confinement

Spectroscopy close to an interface, notably Selective Reflection (SR) spectroscopy [11], permits to probe strongly absorbing gases, because the probed region is made small (e.g. on the order of the wavelength in SR spectroscopy). In particular, it has been a choice technique for a long time to evaluate collisional broadening and shift in an optically dense gas. A notable advantage [12] is that the information is obtained close to the resonance line-centre and not in the far spectral wings, as it would be in a volume experiment.

There are actually few such collisions studies, even in the elementary case of the Cs resonance lines. In particular, the pressure shift had not been widely explored because it usually represents only a small fraction of the collision broadening, while its value and sign possibly depend on the considered hyperfine components [13]. This has provided us a motivation to study collision effects in an ETC, in particular on the Cs D<sub>1</sub> line (894 nm), that had not been studied systematically in previous works. The possibility of a comparison with the well-known method of SR spectroscopy is also a way to check the validity of current interpretations of ETC spectroscopy. A precise comparison, with truly identical experimental conditions, is deferred to the near future, with a specially designed Cs cell including a nanometric thickness region, and a more macroscopic region suitable for SR spectroscopy.

Until now, we have conducted preliminary experiments [10] on the well-resolved Cs D<sub>1</sub> line, enabling a comparison of a  $\lambda/2$  and  $\lambda/4$  thickness. For a given spectrum obtained in an ETC, the modelling of the spectral lineshape allows one to extract both the homogeneous width (and hence the overall pressure broadening) and the shift relatively to the atomic resonance in a low-density volume experiment. This implies a simultaneous analysis of the Fabry-Perot effects, Dicke narrowing and surface interaction. The pressure dependence of the resonance broadening and shift is expected to remain linear in a large regime of density. The introduction, in the predicted lineshapes, of a van der Waals (vW) atom-surface potential known to evolve like  $V(z) = -C_3 z^{-3}$  ( $z$ : the atom-surface distance) for a single wall, with a typical value  $C_3 \sim 1\text{-}2 \text{ kHz}\cdot\mu\text{m}^3$  for the D<sub>1</sub> line, imposes a small but visible perturbation (distortion and shift) for a  $\lambda/4$  thickness, that is comparable to the vW effect in SR spectroscopy. For a  $\lambda/2$  thickness, the vW potential has a negligible effect. Preliminary fittings of the experimental lineshapes seem indeed slightly improved when the vW interaction is included for the  $\lambda/4$  thickness [10], while no difference is observed for a  $\lambda/2$  cell. Remarkably, taking into account the



**Figure 3.** A comparison between the estimated resonance shifts, as appearing in a fitting of the ETC transmission lineshapes for  $\lambda/2$  and  $\lambda/4$  cell. The points represented by  $\square$  are obtained when neglecting the vW interaction, the points represented by  $\bullet$  take this interaction into consideration. vW shift induces not only a slight change in the estimated pressure broadening, but strong modifications of the extrapolated pressure shift, with a notable difference between the shifts estimated for a  $\lambda/2$  and  $\lambda/4$  thickness (see figure 3). Such a behaviour is sensitively independent of the fine

adjustment of the  $vW$  interaction coupling coefficient, enabling by the way one to cancel the residual shift at null pressure, remaining otherwise unexplained.

Because extrapolation from experimental data can be sensitive to various hindered phenomena, more systematic investigations -implying systematic variations of the cell thickness, and of the atomic density- are needed before concluding that one truly observes a thickness-dependent collisional behaviour. However, such an hypothetic possibility is worth discussing from a general point of view. An atom-atom collision, in its principle, is not very different from an atom-surface collision, especially at long distance. In particular, the atom-atom  $vW$  interaction is nothing else than the dipole-dipole coupling between the fluctuating atom-dipole and the correlated induced (and fluctuating) dipole in the atomic perturber. As long as an electromagnetic boundary lies in the vicinity of the two interacting atoms, the correlation between the fluctuations seen by the atom and its perturber can follow a variety of paths, from the direct propagation, to paths involving reflection(s) on the boundary (see e.g. [14]). Physically, these additional contributions can be understood as equivalent to a collision process where at least one of the atom is replaced by its electric image. In the principle, these contributions have to be considered when the distance to the reflecting plane remains smaller than the wavelength of the relevant (virtual) atomic transitions. Anyhow, the efficient distance in collision processes may rather be related with the square root of the collision cross-section. To our knowledge, no evaluation of this confinement effect has ever been performed in the situation of a confined vapour, with the required averaging over atomic positions and velocities. Note also that the implicit assumption that the atom dynamics -*i.e.* the velocity distribution and the population distribution among various sublevels- is independent of the cell thickness, may turn to be questionable for a very confined vapour.

#### 4. Exploring the atom-surface van der Waals interaction at small distances

It has already been briefly recalled in section 3 that in ETC spectroscopy, the atom-surface interaction is dominated by the universal long-range  $vW$  attraction [11] between a fluctuating atom dipole and its image. The major interest of these nanocells for the probing of the  $vW$  interaction is that it extends the possibility to probe short-lived excited states, typical of optical methods such as SR spectroscopy, to an unusual range of short distances. Indeed, the  $z^{-3}$  scaling normally applies in a typical 1-1000 nm range, *i.e.* as long as retardation (Casimir) effects are negligible and before occurrence of the short-range effects depending upon the structural (atomic) details of the surface. It hence implies a validity covering a huge energy range, that is particularly worth to be tested. With ETC spectroscopy, the thickness can normally be arbitrarily chosen through the choice of the container thickness. This is in contrast with SR spectroscopy, with its probed depth  $\sim \lambda/2\pi$  governed by the wavelength of observation. This interest is enhanced by the fact that the choice of methods to investigate the long-range atom-surface interaction is actually scarce [11], in spite of the ubiquity of the  $vW$  interaction. Most common methods rely on mechanical deflection, making them applicable only to ground state atoms or long-lived states. Practically, the only accurate measurements of the distance law of the interaction have been limited to the 3000-500 nm range, with Rydberg atoms that are strongly interacting and long-lived, allowing for a detection that is partly mechanical [15]. With nanocells, one envisions the possibility of probing distances at least an order of magnitude smaller, reaching a distance range where various short-range effects could limit the effectiveness of the  $z^{-3}$  scaling.

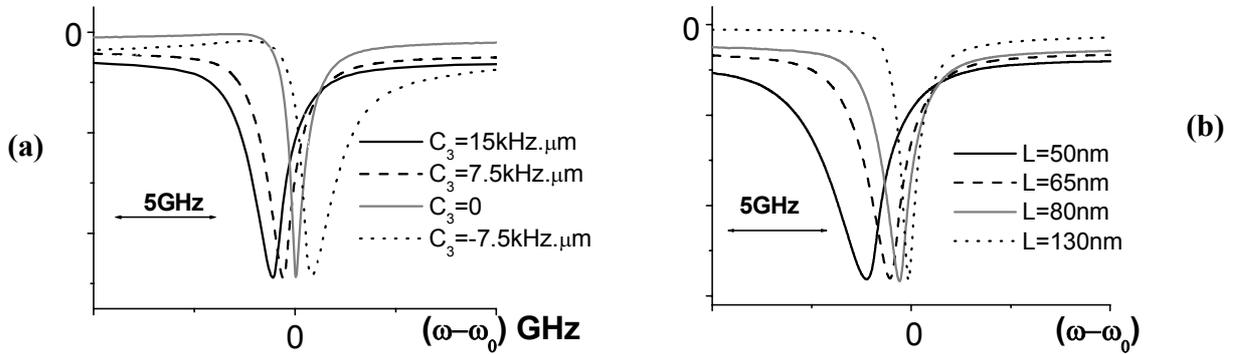
We had already reported on various evidences of the atom-surface interaction in nanocell spectroscopy. Our most recent analyses [10] show that turning from the observation [16] of the surface interaction, to a significant measurement, is now feasible. A first step was to analyze the transmission behaviour in nanocells on the Cs  $D_1$  resonance line. When the local cell thickness turns to be very small ( $\sim 50$ -100nm), a lineshape distortion and a red-shift (up to 200MHz) are observed, that largely exceed those observed through SR spectroscopy. Moreover, the spectral lineshapes appear in good agreement with a theoretical model that assumes a known strength of the van der Waals ( $vW$ ) interaction. This theory simply assumes a thermal distribution of atoms flying wall-to-wall, and integrates the transient atomic response while taking into account a  $vW$  potential. The  $vW$  potential itself is modelled according to an electrostatic description including the interaction with the multiple images induced in the two reflecting walls. These transmission experiments have been extended to

experiments in the FM mode, whose improved spectral resolution is of particular interest for large thickness (*i.e.* when the vW shift remains small), and to reflection spectra: for the smallest thicknesses, reflection spectra are indeed recorded over a low level of non resonant reflection as due to Fabry-Perot interferences, and offer a competitive possibility to observe the vW shift [16].

We are now investigating the stronger vW shift induced on high-lying excited states, such as Cs(6D), that is probed at 917 nm ( $6D_{5/2}$ ) or 921 nm ( $6D_{3/2}$ ) after a prior excitation on the Cs  $D_2$  line  $6S_{1/2} \rightarrow 6P_{3/2}$  at 852 nm. For these transitions, the vW interaction should be about an order of magnitude larger than for the  $D_1$  line. Moreover, for small cell thickness, it is susceptible to provide the dominant broadening, owing to the spatial inhomogeneity of the vW shift. Thanks to a laser diode specially designed for a broad range tuneability (through step-by-step frequency changes), we could observe the broadened transmission (and reflection) spectra for various ETC thicknesses, down to 20 nm. Depending on the thickness, the observed (red) shift can be in excess of 10's of GHz -*i.e.* an energy shift 2 orders of magnitude larger than previously observed [15]- . Also, the apparent width of the largely distorted lineshape simultaneously increases for short thicknesses, remaining an approximately constant fraction of the frequency shift (*i.e.* vW induced inhomogeneous broadening). The observed shift -as measured by evaluating the frequency of the transmission peak- shows an approximate  $L^{-3}$  dependence, in agreement with a simple theoretical modelling: at a given relative position  $z/L$ , the shift is indeed predicted to evolve like  $L^{-3}$ , but the behaviour of the overall lineshape, with its weighted integration of different spatial contributions, is more difficult to evaluate.

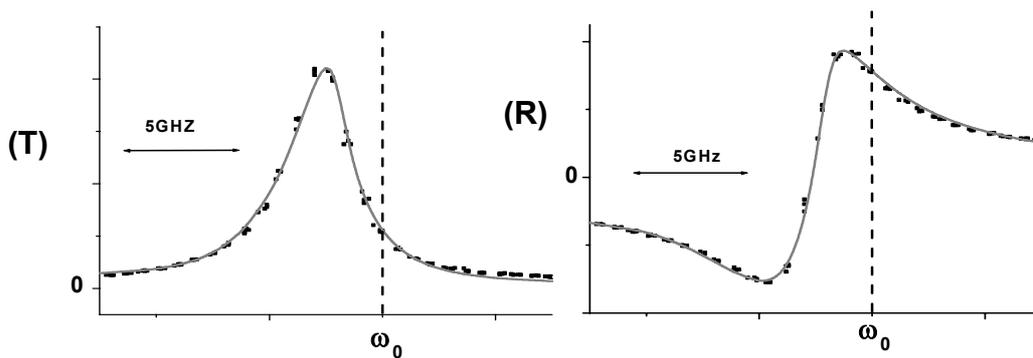
An accurate evaluation of the surface interaction effect obviously requires more systematic experiments, because the observed lineshapes can be affected by various process, such as the pumping into the intermediate states - spatial inhomogeneities of the pumping, non thermal velocity distribution in the intermediate state -, or by the high vapour density required for these experiments, implying broadening and shift. This is why we have turned to a series of experiments for a limited number of accurately-controlled cell thickness (presently in the range 40-130 nm) with investigation of the pressure effects, and control of the effects of the pumping light: the pumping in the  $6P_{3/2}$  state is expected to be nearly thermal when the pump absorption is weak, and when lineshapes are independent of the pumping power. Moreover, the careful control of a stabilized pump frequency helps to perform a reproducible selection among the various hyperfine sublevels of the resonant state. These conditions permit to extend the modelling, previously used for a resonance transition, to transitions between excited states which are sensitive to a much stronger vW interaction. On this basis, it is possible to extract an acceptable range of vW strengths from each individual spectrum, and to check if it remains constant under phenomenological spectral changes as induced by a change in the Cs density. Similarly, one can also check the consistency of the estimated widths with a pressure broadening, or even the linearity of the pressure-induced shift (actually, the vW shift and its induced inhomogeneous broadening are clearly dominant, at least for small thickness experiments, and the pressure shift can be most often neglected).

As can be seen from the various theoretical spectra shown in figure 4, in the regime of small thickness, and strong vW interaction, there is an imposed relation between the shift (e.g. as measured with the position of the peak of the transmission lineshape) and the width of the lineshape, essentially governed by the spatial inhomogeneity of the vW interaction. In other words, before any optimization of the parameters in a fitting process, one can recognize if an experimental lineshape will be acceptably fitted with the theoretical model, or if the modelling is inadequate. This obviously imposes tight constraints to the acceptable fits. Moreover, for our series of experiments, as due to a vW strength largely exceeding the Doppler broadening, the pressure broadening, although observable, modifies only marginally the predicted lineshapes.



**Figure 4.** Theoretical transmission lineshapes through an ETC : **(a)** for a 80nm thickness and the various indicated vW strength ; **(b)** for the various indicated thicknesses, and a constant vW interaction  $C_3=7.5\text{kHz}\cdot\mu\text{m}^3$ . The  $C_3$  value corresponds to the 1-wall vW strength. The lineshapes are quasi-independent of the (non resonant) reflection coefficient of the windows (here  $r=0.5$ ). The Fabry-Perot effects included in the modelling explain the asymmetry for  $C_3=0$  in (a). For normalization purposes, the amplitudes are multiplied by a respective factor: (a) 1 for  $C_3=0$ ; 2.55 for  $C_3=7.5\text{kHz}\cdot\mu\text{m}^3$ , 3.28 for  $C_3=15\text{kHz}\cdot\mu\text{m}^3$ , 2.84 for  $C_3=-7.5\text{kHz}\cdot\mu\text{m}^3$ ; and: (b) 1 for 130 nm, 2.96 for 80 nm, 4.97 for 65 nm, 10.1 for 50 nm.

A specificity of ETC spectroscopy is that it allows for the simultaneous recordings of the transmission and reflection lineshapes. These lineshapes result [4] from a very different combination of absorption-related and dispersion-related lineshapes. As illustrated in figure 5, for the transition to the  $6D_{5/2}$  level (917 nm line), consistent fittings for one type of signal (e.g. reflection) are actually obtained with those parameters extracted from the other signal (transmission). This provides a major evidence for the satisfactory description obtained with our modelling. Detailed tests of the numerical fittings are presently under progress to evaluate to which extent the  $z^{-3}$  dependence is verified in our range of thickness. For a 65 nm-130 nm thickness range (i.e. the vW potential varies by an order of magnitude), the estimated  $C_3$  value seems to remain constant within a factor 2, and to be in agreement



**Figure 5.** Experimental transmission (T) and reflection (R) spectra (black squares) as recorded on the Cs  $6P_{3/2}$ - $6D_{5/2}$  line (917 nm) for a 50 nm cell thickness, and corresponding fits (grey dashed line). The nanocell temperature (220°C) is low enough to make pressure shift, and even pressure broadening, negligible. The model assumes a "thermal" pumping into Cs ( $6P_{3/2}$ ). The fitting parameters are the same for transmission and reflection

with the theoretical expectation at an interface with YAG windows. More precise evaluations are now under progress. Note that the accuracy for the relatively large thickness is partially altered because of the hyperfine structure of the pumped  $6P_{3/2}$  level, which turns to be comparable with the average shift

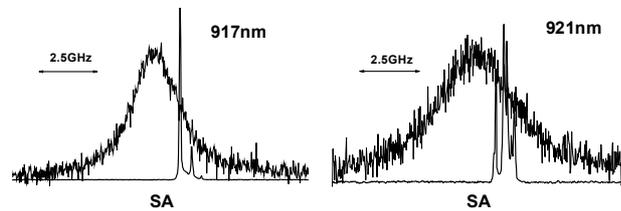
It should be emphasized that for extremely small thicknesses, which make our ETCs genuine 1-D "nanocells", the reproducibility of the recorded spectra becomes really an issue. For a series of experiments performed on a nanocell with YAG windows, lineshape reproducibility has been clearly verified when comparing different cell spots of equal thickness, as long as this thickness is 65 nm or more. The comparison of "50 nm" spots shows some variations in the lineshapes, although several spots appear able to provide nearly identical responses, such as the one selected as the "valid" response for 50 nm (see figure 5). For differing 40 nm spots, no effective reproducibility has been found. Rather, the apparent shift, relatively to the free-atom resonance, varies from spot to spot, by a factor that anyhow remains smaller than 2 in all cases. In spite of the accuracy of the thickness measurement, that apparently reaches in some cases 1-2 nm, the measurement intrinsically provides a thickness *averaged* on the optical spot size (typical beam diameter is 100-200  $\mu\text{m}$ ). Hence, the non-measured local irregularities affecting the window profiles (*i.e.* roughness, evaluated over various characteristic lengths), may induce local fluctuations in the atom-surface distance, expected to produce dramatic effects on the averaged atomic spectra, with respect to a vW potential essentially sensitive to the *inverse cube* of the distance (distance fluctuations between 35 and 45 nm correspond to a doubling of the vW shift).

### 5. Resonant atom-dielectric coupling in nanocells

In the modelling of ETC spectroscopy used above, the local shift of the atomic resonance induced by the surface interaction relies on an electrostatic description of the vW interaction, and includes a multiple image approach to take into account the two neighbouring interacting surfaces. Such a vW description leads to an overall spatial dependence of the vW interaction that follows a special function (the transcendental Lerch function [17]). This description is obviously more precise than the simple addition of the individual potential exerted by each of the walls, even if the overall spectral predictions are most often not very different.

Actually, this electrostatic approach applies only as long as the resonant couplings between the dielectric surface and the relevant virtual atomic transitions are not considered. Indeed, the vW atom-surface interaction is a dipole-dipole interaction, originating from the coupling between the atom dipole fluctuations (to be expanded along the virtual electric dipole transitions) and the correlated fluctuations induced in the surface. Resonances in the electromagnetic coupling between a surface polariton mode and an atom fluctuation (in virtual emission), have been shown to lead to giant and possibly repulsive interaction [18,19], along with an increased decay rate from the considered excited state [20], as governed by a surface response  $(\epsilon-1)/(\epsilon+1)$ , with  $\epsilon$  the *complex* dielectric permittivity. In particular, the 12.15 $\mu\text{m}$  virtual emission of Cs( $6D_{3/2}$ ) towards ( $7P_{1/2}$ ) makes a sapphire surface strongly repulsive for a Cs( $6D_{3/2}$ ) level, while a YAG surface is expected to exert only a weak repulsion [18]. Simultaneously with major changes for the vW interaction induced by the *virtual* resonant coupling, a *real* energy transfer between the atomic excitation and the surface mode may occur, implying a  $z^{-3}$  lifetime shortening as due to a surface-induced quenching. Conversely, the Cs( $6D_{5/2}$ ) level, with its main virtual emission at 14.6  $\mu\text{m}$ , is expected to remain attracted by a surface such as sapphire or YAG. This predicted difference between the two fine-structure components of the Cs(6D) level had not been investigated before, as due to a lack of adequate sources.

A series of experiments on the 921 nm line  $6P_{3/2} - 6D_{3/2}$  in a YAG nanocell has been performed, exploring a similar variety of situations as for the experiments on the 917 nm line (figure 6). A detailed analysis of the recorded lineshapes is still in progress. The theoretical modelling of the interaction potential reveals more delicate than in the non resonant electrostatic situation. At first sight, one anyhow notices that in spite of a predicted repulsion, no blue shift of the line is specifically observed. Actually, this does not preclude a repulsive behaviour (upward convexity of the potential),



**Figure 6.** Comparison between the 917 nm and 921 nm spectra, recorded in a 65nm-thick cell in the same experimental conditions ( $T=220^{\circ}\text{C}$ ). The SA spectra are reference saturated absorption spectra (note the effect on the reference of the slight differences in the hyperfine structure of Cs ( $6D_{3/2}$ ) and Cs ( $6D_{5/2}$ )). In our conditions, the shift and lineshape distortion purely originate in the surface interaction. The 921 nm linewidth is broader than the 917 nm one in spite of a smaller shift.

because with two repulsing walls, it has been shown [10] that the total potential can exhibit a repulsive behaviour at any distance while inducing a red-shift close to the nanocell center. Also, the comparison with the 917 nm transition shows that, in similar conditions, the signal has much smaller amplitude - not entirely explained by smaller transition probabilities in the free-space-, and that the signal width is nearly twice larger, in spite of an observed shift that remains comparable to, or smaller than the one observed at 917 nm. This is a possible first indication, that remains to be confirmed by a further analysis, of a surface-induced energy transfer occurring within an nanocell, when the two walls simultaneously interact with the excited atom. At least, the observed ratio between width and shift shows that the non resonant model valid for the 917 nm line is not applicable here.

## 6. Conclusion

In spite of the fact that nanocells permit to study atomic vapour at very unusual distances from a surface, essential techniques of high-resolution spectroscopy are proved to remain applicable with rather minor changes. The detailed analysis of the experimental spectra shows that this novel spectroscopy can effectively provide a unique tool to investigate problems not attacked previously, as the one of collision under confinement, or effective distance-dependence of the atom-surface interaction in the sub-100 nm range. Extensions of our investigations to the physics of resonant gases under strong multi-dimensional confinement, that can be approached with systems such as holey fibres or porous media [21], should offer renewed surprises, of interest for both fundamental and applied physics.

## Acknowledgments

This work has been partially supported by European contract HPRN-CT-2002-00304, as part of the FASTNet programme. We also acknowledge discussion with A. Lezama in the frame of the French-Uruguayan ECOS programme (U00-E03), and with S. Cartaleva in the frame of French-Bulgarian project RILA 2005.

## References

- [1] Briaudeau S, Bloch D and Ducloy M 1999 *Phys. Rev. A* **59** 3723; Briaudeau S, Saltiel S, Nienhuis G, Bloch D and Ducloy M 1998 *Phys. Rev. A* **57** R3169; Briaudeau S, Bloch D and Ducloy M 1996 *Europhys. Lett.* **35**, 337; see also Briaudeau S et al. 2000 *J. Phys. IV* **10** 145 and Briaudeau S et al. 1998 *Laser Spectroscopy XIII* (Wang YZ et al, eds. , World Scientific,) pp. 33-36
- [2] Sarkisyan D, Bloch D, Papoyan A and Ducloy M 2001 *Opt. Commun.* **200** 201

- [3] Dutier G, Yarovitski A, Saltiel S, Papoyan A, Sarkisyan D, Bloch D and Ducloy M 2003 *Europhys. Lett.* **63** 35
- [4] Dutier G, Saltiel S, Bloch D and Ducloy M 2003 *J. Opt. Soc. Am. B* **20** 793
- [5] Romer RH and Dicke RH 1955 *Phys. Rev.* **99** 532
- [6] Sarkisyan D Varzhapetyan T, Sarkisyan A, Malakyan Yu, Papoyan A, Lezama A, Bloch D and Ducloy M 2004 *Phys. Rev. A*, **69** 065802
- [7] Dutier G, Todorov P, Hamdi P, Maurin I, Saltiel S, Bloch D and Ducloy 2005 Dicke coherent narrowing in two-photon and Raman spectroscopy of thin vapour cells *Preprint* <http://fr.arxiv.org/abs/physics/0506025>.
- [8] Dicke RH 1953 *Phys. Rev.* **89** 472 and Dicke RH 1954 *Phys. Rev.* **96** 530
- [9] Varzhapetyan T, Sarkisyan D, Petrov L, Andreeva C, Slavov D, Saltiel S, Cartaleva S 2005, in SPIE Proceedings of the XIII International School on Quantum Electronics "Laser Physics and Applications", held in Burgas, Bulgaria, Sept 20-24,2004, to appear.
- [10] Hamdi I, Todorov P, Yarovitski A, Dutier G, Maurin I, Saltiel S, Li Y, Lezama A, Varzhapetyan T, Sarkisyan D, Gorza M-P, Fichet M, Bloch D and Ducloy M 2005 *Laser Phys* **15** (7)
- [11] Bloch D and Ducloy M 2005, Atom-Wall Interaction in *Adv. At. Molec. Opt. Phys.*, vol. **50**, (Bederson B and Walther H eds., Elsevier Academic Press, 2005) pp. 91-154
- [12] Akul'shin AM, Velichanskii VL, Zibrov AS, Nikitin VV, Sautenkov VV, Yurkin EK and Senkov NV 1982 *J.E.T.P. Lett.* **36** 303
- [13] Papageorgiou N, Fichet M, Sautenkov VA, Bloch D and Ducloy M 1994 *Laser Phys.* **4** 392 ; see also preliminary results for Cs D<sub>1</sub> line in Dutier G 2003, PhD dissertation, Université Paris 13, unpublished
- [14] Cho M and Silbey RJ 1996 *J. Chem. Phys.* **104** 8730
- [15] Sandoghdar V, Sukenik CI, Hinds EA and Haroche S 1992 *Phys. Rev. Lett.* **68** 3432; Sandoghdar V, Sukenik CI, Haroche S and Hinds EA 1996 *Phys. Rev. A* **53** 1919
- [16] Dutier G, Hamdi I, Segundo PCS, Yarovitski A, Saltiel S, Gorza M-P, Fichet M, Bloch D, Ducloy M, Sarkisyan D, Papoyan A and Varzhapetyan T 2004, in "Laser Spectroscopy. Proceedings of the XVI International Conference" (Hannaford P, Sidorov A, Bachor H and Baldwin K eds., World Scientific, Singapore) pp.277-284
- [17] Nha H and Jhe W 1996, *Phys. Rev. A* **54** 3505 (1996); and *erratum* 1999 **60** 1729
- [18] Failache H, Saltiel S, Fichet M, Bloch D and Ducloy M 1999 *Phys. Rev. Lett.* **83** 5467 and Failache H, Saltiel S, Fichet M, Bloch D and Ducloy M 2003 *Eur. Phys. J. D* **23** 237
- [19] Fichet M, Schuller F, Bloch D and Ducloy M 1995, *Phys. Rev. A* **51** 1553; Wylie M and Sipe JE 1984 *Phys. Rev. A* **30** 1185 and Wylie M and Sipe JE 1985 *Phys. Rev. A* **32**, 2030 (1985) ; Gorza M-P, Saltiel S, Failache H and Ducloy M 2001 *Euro. Phys. J. D* **15** 113-126
- [20] Failache H, Saltiel S, Fischer A, Bloch D and Ducloy M 2002, *Phys. Rev. Lett.* **88** 243603
- [21] Burchianti A, Marinelli C, Bogi A, Brewer J, Rubahn K, Rubahn H-G, Della Valle F, Mariotti E, Biancalana V, Veronesi S and Moi L 2004, *Europhys. Lett.* **67** 983