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Radiation trapping and spin relaxation of cesium atoms at cell walls

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ABSTRACT We study the spatial profile of atomic fluorescence due to multiple photon scattering in two sealed glass optical cells, one with its internal surface coated with a polysilane film and the other uncoated. The radiation-trapping spatial profile is used as a probe of the excited-state population, and evidences the role of the surface in laser–atomic vapor volume processes. Each cell is distinguished from the other by a depolarization time which characterizes the internal surface state: in the coated cell, the longer depolarization time leads to a trapped fluorescence intensity smaller than in the uncoated cell. A simplified theoretical model taking into consideration the hyperfine polarization of the atoms by the resonant laser, the thermalization by collisions with the cell walls and the trapping of the fluorescence photons by ground-state atoms provides a good description of the observations.

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1 Introduction

Atoms of a low-pressure vapor, in pre-evacuated sealed optical cells, can be considered as isolated in many experiments of atomic spectroscopy. Atomic collisions occur mainly with the cell walls, and in systems where the interaction with a resonant laser gives rise to optical pumping, as in the case of alkali atoms, the role of the surface is essential to ensure the thermalization of the ground hyperfine states and thus to keep a population of atoms interacting with the laser radiation. Nevertheless, the interaction of these atoms with the container is frequently not taken into account in the interpretation of results in experiments exploiting the laser–atom interaction, even when the mechanism of optical pumping is very effective. In this paper we describe an experiment on radiation trapping as an example of the role of the internal surface of the cell in volume processes of laser interaction with resonant alkali vapors.

2 Radiation trapping

In the process of laser interaction with an atomic vapor, the number of atoms in the excited state depends on the

intensity of the laser beam and, in a first approximation, one may expect the spatial fluorescence profile in the vapor to be an image of the beam profile. However, due to the strong absorption cross section of the resonant transitions, particularly for alkali atoms, one can easily observe the fluorescence from atoms not directly illuminated by the laser, but that absorb photons emitted by atoms in the laser-beam volume. As a result, the radiation is trapped in the resonant gas for longer than the atomic transition lifetime, and the optical excitation takes place in a volume largely beyond the limits of the laser beam. In fact, the radiation trapping is a well-known phenomenon and has been studied for decades [1–3]. As an example of its importance, we can mention the effect of photon multiple scattering occurring in optical traps for neutral atoms [4], resulting in a repulsive interaction between the atoms of the cold vapor, which represents a limiting factor to achieving high densities in phase space [5]. Radiation trapping has also been demonstrated to be a destructive mechanism of the polarization of optically pumped alkali atoms [6, 7].

Radiation trapping has been mostly observed and described for high-power lasers, dense vapor or in the presence of a buffer gas [8–10], and much work has focused on the temporal behavior of multiple scattering in different conditions and geometries of the cells [11]. Nevertheless, in stationary conditions the radiation-trapping mechanism also affects the spatial population distribution and the radial profile of resonant scattered light has recently been studied as a function of the atomic density [8]. We report here on the observation of the spatial distribution of the radiation trapping, as an incoherent mechanism in a dilute resonant vapor [12]. We measure the intensity of the radiation scattered by atoms excited only by fluorescence of other atoms and we compare the observations in optical cells with different internal surfaces. We show that the surface plays a non-negligible role in the radiation-trapping intensity, by considering the atoms as three-level systems and taking into account both the optical pumping process and the thermalization on the surface of the ground-state hyperfine sublevels.

3 Experimental set-up and measurements

The experimental set-up is shown in Fig. 1. Two glass cells with the same dimensions are used: their length is 20 mm and their radius 10 mm. Both contain Cs vapor at

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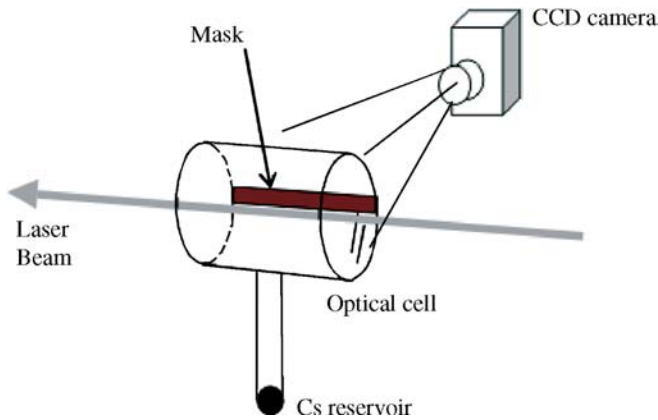


FIGURE 1 Experimental set-up for observation of the radiation scattered by the atomic vapor in the region outside the laser-beam volume. A mask is placed between the cell and the CCD camera, along the laser-beam direction

the same density of about 4×10^{10} atoms/cm³, and the background pressure is less than 10^{-5} Torr. In these conditions the atom–atom collisions are negligible, so that the atomic thermal equilibrium is obtained only through collisions with the cell surfaces. One of the cells was previously coated with a film of octadecyltrimethoxysilane [13]. The cells are carefully prepared in order to avoid liquid cesium in the body of the cells. A drop of cesium is distilled into the reservoir and during the experiment the body of the cell contains only dry alkaline vapor.

The laser is a continuous-wave free-running diode laser (SDL 5401-G1) emitting around the D_2 cesium line at 852.1 nm, with an emission line width of about 30 MHz. A lens with focal length of 5 mm collimates the laser output and a 2-mm-diameter diaphragm produces a beam with an almost square intensity profile. This beam is sent along the axis of the cell. The experiments are carried out with the laser coupling the $6S_{1/2}$ ($F = 4$) to the $6P_{3/2}$ ($F' = 3, 4, 5$) levels (see Fig. 2).

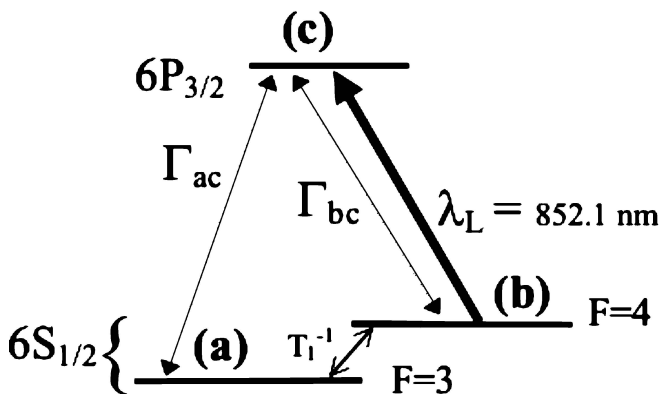


FIGURE 2 Relevant energy levels and elementary processes in the Cs vapor: energy levels of the cesium D_2 transition. The laser (indicated by the bold arrow) is resonant with the $6S_{1/2}$ ($F = 4$) (named (b)) \rightarrow $6P_{3/2}$ ($F' = 3, 4, 5$) (c) transition. The optical pumping transfers population to the $F = 3$ (a) hyperfine ground-state sublevel through spontaneous emission from level (c) to level (a) at a rate Γ_{ac} . The total spontaneous decay rate of the excited level (c) is $\Gamma = \Gamma_{ac} + \Gamma_{bc}$. The relaxation of the electronic polarization occurs through collisions on the cell walls, at a rate $1/T_1$ (process indicated in the figure by a double arrow between the levels (a) and (b))

A CCD camera is used to collect the atomic fluorescence in the lateral regions of the cell. In order to have a good sensitivity for observing the lateral volume of the cell while avoiding the camera saturation, we use a mask ($5 \text{ mm} \times 20 \text{ mm}$) to hide the central region of the cell, i.e. we screen the light emitted by the atoms in the laser-beam volume, and we observe the fluorescence in the lateral regions, i.e. the scattering of the trapped radiation. Images are recorded in identical excitation conditions for the two cells. To compare the intensities of the two fluorescence images we perform preliminary tests to guarantee the linearity of the camera signal with the light intensity. For each series of measurements on the two cells, we also keep the same density of atomic vapor in the cell, as proven by the absorption of a very weak probe beam in the vapor. Then a clear difference in the efficiency of the photon multiple scattering process appears between the coated and the uncoated cells (Fig. 3). To quantify this difference we use

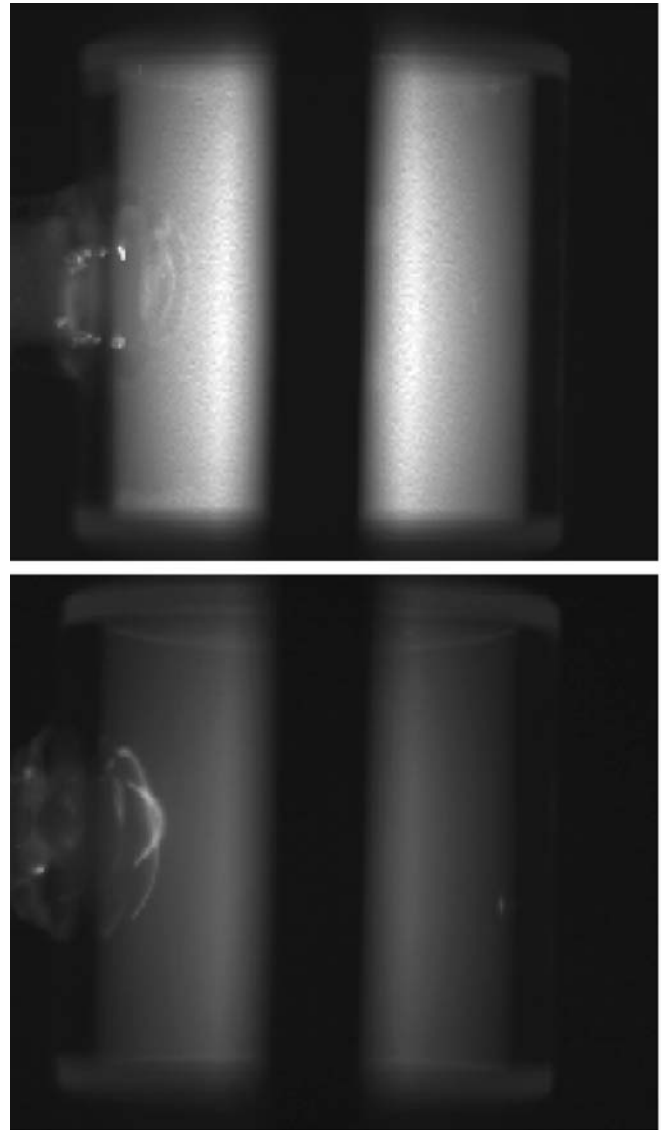


FIGURE 3 Fluorescence from cesium atoms in the lateral regions of cylindrical cells as a narrow resonant laser beam is sent through the low-pressure cesium vapor. *Top*: ordinary glass cell; *bottom*: cell coated with a film of octadecyltrimethoxysilane

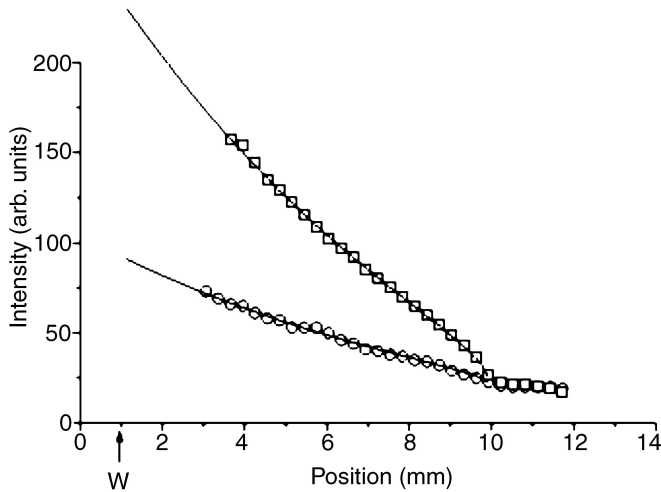


FIGURE 4 Fluorescence profiles due to the multiple scattering in the lateral volume of the cell, as a function of the radial distance from the laser axis, x . The laser intensity is 50 mW/cm^2 . The two fluorescence profiles have been recorded in the same condition of 72% of linear absorption at line center. The fluorescence from the laser region ($-w \leq x \leq w$) is hidden by a larger mask (see Fig. 1). Experimental data: *squares*, uncoated cell; *circles*, coated cell. Calculated profiles (see text): *full lines*

the images captured by the CCD camera and select a line orthogonal to the laser axis. We plot the fluorescence intensity as a function of the radial distance from the laser axis. In Fig. 4 the two fluorescence profiles have been recorded in the same condition of 72% of linear absorption, which corresponds to the cesium-vapor density at room temperature ($T = 298 \text{ K}$) in the bare Pyrex cell. It has been observed by groups working with silanized or paraffined cells that the alkali-vapor pressure in this kind of treated container is always smaller than the pressure of alkali vapor in equilibrium with the molten metal. This is attributed to the absorption of the atoms by the polymer coating [14]. So, in order to have the same cesium density in our two cells, the reservoir temperature of the coated cell has to be slightly increased ($\Delta T \approx 40 \text{ K}$). The atomic density is inferred from the measurement of the transmission of a very weak thin probe beam through the cell. The intensity of this probe beam is sufficiently low ($< 0.5 \text{ mW/cm}^2$) that the optical pumping is negligible and that the absorption does not depend on the relaxation time on the surface and has therefore the same dependence on the atomic density in the two cells. In these conditions, we observe a ratio of about three between the fluorescence intensities in the two cells. As the laser line width is narrower than the Doppler width of the excited $6P_{3/2}$ level, we repeated the fluorescence measurements, as those of Fig. 4, for different frequencies of the laser in the Doppler profile. We found, within the experimental error, the same ratio between the fluorescence signals obtained from the two different cells.

4 Hyperfine relaxation on cell surfaces

The only difference between the two cells lies in their internal surface state, so that the difference observed between the intensities of the fluorescence in these two cells is an effect of the interaction of the optically pumped cesium atoms with the cell walls. The energy splitting of the two hyper-

fine sublevels ($F = 3$ and $F = 4$) of the cesium ground state is about 0.038 meV (9.2 GHz) and thus much less than the thermal energy at room temperature ($k_B T = 25 \text{ meV}$). Their thermal populations are therefore in the ratio of their degeneracies, i.e. of the same order of magnitude. On the other hand, the ground-state level separation is much larger than the Doppler broadening at room temperature, so that a laser field resonant with a transition from one of the hyperfine levels will transfer part of its population to the other, non-resonant hyperfine level, through spontaneous decay of the excited level, thus creating a disequilibrium in the ground hyperfine populations (optical pumping, see [15]). The fast re-equilibrium of the populations of the two ground-state sublevels usually hides this three-level optical process, frequently neglected in the descriptions of laser-atom experiments. Conversely, in some experiments where this thermalization can not be achieved, as in magneto-optical traps for neutral atoms [4], a repumping laser is essential to attain a stationary regime of the laser-atom interaction.

At low pressure, the thermalization of the fundamental hyperfine populations can be obtained through collisions with atoms of a buffer gas or, as is the case in our experiments, through collisions with the cell walls: atoms incident on a surface stay adsorbed on it during a mean adsorption time which depends on the surface state [16]. While adsorbed at the surface, atoms undergo fluctuating magnetic forces which tend to thermalize the electronic spin of the ground-state atoms. Silane films deposited on metallic or dielectric surfaces are known to reduce the adsorption time on these surfaces and thus to increase the lifetime of the atomic polarization in containers with so-coated internal walls [17]. In order to characterize each cell of the radiation-trapping experiment described above, we carried out measurements of the depolarization time of electronic spin of the cesium atoms on the cell walls. A pump/probe temporal technique is used: a large pump beam (about 10-mm diameter) polarizes the electronic spin of a fraction of the cesium atoms, through the mechanism of optical pumping, and is then turned off. A narrow, low-intensity probe beam monitors the relaxation of the population difference between the two ground-state hyperfine sublevels. This experiment permits us to directly measure the temporal evolution to thermal equilibrium of the populations of each hyperfine level of the fundamental state, after optical pumping by the pump laser. We measured relaxation times of $\sim 2 \text{ ms}$ and $\sim 40 \mu\text{s}$ for atoms in the coated and the uncoated cells, respectively. The time to restore the population of the resonant sublevel in the coated cell is longer by about two orders of magnitude than in the uncoated cell, and is responsible for the availability of fewer atoms (in the ground sublevel b , as indicated in Fig. 2) to absorb the laser radiation in this cell compared to the cell with bare walls.

5 Rate equations for the populations

To describe the fluorescence observed in alkali-atom vapor in the presence of resonant lasers we consider the atoms as three-level systems (two ground-state sublevels and one excited state, see Fig. 2) where the mechanisms of optical pumping, radiation trapping and hyperfine thermalization on the surfaces occur.

With the three relevant levels denoted a , b and c (see Fig. 2), we can write the stationary 1D rate equations for their population densities n_a , n_b and n_c :

$$\frac{dn_a}{dt} = 0 = \Gamma_{ac} n_c + \frac{n_b - n_a}{T_1} - \Gamma_{ac} \int dx' n_c(x') G_{ac}(n_a, n_c, x, x'), \quad (1)$$

$$\begin{aligned} \frac{dn_c}{dt} = 0 = & -\Gamma n_c + \alpha \Gamma (n_b - n_c) \\ & + \Gamma_{bc} \int dx' n_c(x') G_{bc}(n_b, n_c, x, x') \\ & + \Gamma_{ac} \int dx' n_c(x') G_{ac}(n_b, n_c, x, x'), \end{aligned} \quad (2)$$

$$n = n_a + n_b + n_c. \quad (3)$$

This last equation expresses the conservation of the total number of atoms in the cell. In (1) and (2), $\Gamma = \Gamma_{ac} + \Gamma_{bc}$, where Γ_{ac} (Γ_{bc}) is the spontaneous decay rate of the excited population to the level a (b), and Γ is the total decay rate of n_c . The variable x is the radial distance to the axis of the laser. The 1D treatment is a good approximation for small average number of scatterings [18], as is the case in our very low optical density vapor. In (1), the first term represents the spontaneous emission from state c to state a . The second term accounts for the thermalization of the hyperfine sublevels a and b , with a time constant T_1 [19]. In our case this thermalization is entirely due to the collisions of the atoms with the cell walls. The last term represents the radiation-trapping process and $G_{ac}(n_a, n_c, x, x') dx'$ is the probability that a photon at the frequency of the transition $a \leftrightarrow c$, emitted at position x' , is absorbed at position x where the populations are evaluated. The second term in (2) represents the absorption and stimulated emission due to the pumping laser. The other terms may be attributed the same interpretation as in (1), except for the thermalization term which does not directly act on the excited population dynamics. We assume a complete frequency redistribution, i.e. the frequency of the spontaneously emitted photon is independent of the frequency of the absorbed photon. The low optical density in our experiments ($\bar{k}R \sim 1.4$, where \bar{k} is the average atomic absorption coefficient and R the radius of the cell) permits one to make the mean-free path approximation for the photon [18, 20], where the frequency-dependent absorption coefficient is replaced by its frequency-average value. Under these assumptions, we can write:

$$G_{ac}(n_a, n_c, x, x') = \frac{1}{2} \bar{k}_{ac} \exp\{-\bar{k}_{ac} |x - x'|\}, \quad (4)$$

where the exponential term describes the absorption of the photon as it passes from position x' to position x . \bar{k}_{ac} is the frequency average of the absorption coefficient for the $a \leftrightarrow c$ transition, $k_{ac}(\nu)$, where:

$$k_{ac}(\nu) = k_0 \left(\frac{g_c}{g_a} n_a - n_c \right) F(\nu), \quad (5)$$

with $k_0 = (\lambda^2/4)\Gamma$, g_i the statistical weight of level i ($i = a, b, c$) and $F(\nu)$ the normalized Doppler-broadened line shape:

$$F(\nu) = \frac{\lambda}{2\pi^{3/2} v_{th}} \exp\left(-\frac{(\nu - \nu_0)^2}{\nu_0^2} \frac{c^2}{v_{th}^2}\right). \quad (6)$$

In (6) λ is the wavelength of the pumping light, ν its frequency and $v_{th} = (2k_B T/m)^{1/2}$ the mean velocity of the atoms of mass m in the vapor at temperature T . k_B is the Boltzmann constant.

We are interested in the spatial distribution of excited atoms in the cell, experimentally expressed by the spatial distribution of the trapped fluorescence. We must therefore solve the coupled equations (1)–(3), using for the relaxation time T_1 the values we experimentally obtained (see Sect. 4).

In a simple approach to solve these equations, we make the following assumptions.

- (i) The atoms in the vapor absorb photons either from the laser beam or emitted by atoms directly excited by the laser, but not re-emitted by atoms in the peripheral volume. This assumption is reasonable in our case of very low optical depth of the vapor. It permits one to solve (1)–(3) separately for the volume of the laser beam (henceforth called the inner volume) and the volume of vapor outside the laser beam (outer volume).
- (ii) In the outer as well as in the inner volume, $n_c \ll n_a, n_b$ and the absorption coefficient is homogeneous in each region.
- (iii) In the inner volume, the densities n_a , n_b and n_c are spatially constant, with the values n_a^0 , n_b^0 and n_c^0 . This assumption is made considering the square spatial profile of the laser beam, its small radius ($w = 1$ mm, to be compared to the radius of the cell, $R = 10$ mm) and assumption (i).

Under these assumptions, we solve (1)–(3) in the outer volume using $\alpha = 0$ and $\bar{k}_{ac} = \bar{k}_{bc} = \bar{k}$.

In this case, (2) becomes:

$$n_c(x) = \frac{\bar{k}}{2} n_c^0 \int_{-w}^w dx' \exp(-\bar{k} |x - x'|), \quad (7)$$

which gives, for $x > w > 0$,

$$n_c(x) = n_c^0 \sinh(\bar{k}w) \exp(-\bar{k}x) \quad (8)$$

and $n_c(-|x|) = n_c(|x|)$. In (8), the density of excited atoms in the outer region depends on the pumping efficiency and on the relaxation time on the walls through the density of excited atoms in the inner volume, n_c^0 . The ratio of the fluorescence intensities in the outer volume in the two cells reproduces the ratio between the populations of the excited level in the inner volume.

In Fig. 4 the experimental fluorescence profiles are fitted by exponentially decaying functions of the distance to the cell axis, taking into account the geometrical (circular section) characteristics of the cell: $A(T_1) \exp(-\bar{k}x) \sqrt{R^2 - x^2}$, with $\bar{k} = 140 \text{ m}^{-1}$ and $R = 10$ mm. The ratio of their amplitudes, $A(T_1 = 40 \mu\text{s})/A(T_1 = 2 \text{ ms})$, is ~ 3 .

In the inner volume, the main term is the one of pumping. The coefficient α can be written [6]:

$$\alpha = \frac{\lambda^2}{4\pi} \frac{I_v}{h\nu} Q_w Q_f Q_a, \quad (9)$$

with I_v the light intensity per unit frequency and Q_w , Q_f and Q_a reduction factors of the pumping rate. Q_f is the reduction factor due to the fact that the frequency width of the laser (~ 30 MHz) is smaller than the Doppler-broadened cesium line width (~ 500 MHz). It can thus be estimated as $Q_f = 30/500 \approx 0.06$. Q_w accounts for the fact that the radius w of the laser beam (1 mm) is much smaller than that of the cell ($R = 10$ mm), reducing the pumping efficiency. For the geometry of our optical set-up ($w = 1$ mm, $R = 10$ mm, $L = 20$ mm), we estimate Q_w as 0.005 (see [6]). Q_a is the reduction factor of the pumping rate due to the absorption of the laser light as it propagates in the resonant vapor:

$$Q_a = \frac{1}{\bar{k}L} \left(1 - e^{-\bar{k}L}\right). \quad (10)$$

Using $\bar{k} = 140 \text{ m}^{-1}$, as deduced above from the fitting of the experimental fluorescence profiles, we find $Q_a = 0.335$.

Following assumption (iii), we evaluate the densities n_a^0 , n_b^0 and n_c^0 at position $x = 0$. Equations (1) and (2) become:

$$\Gamma_{ac} n_c^0 = -\frac{n_b^0 - n_a^0}{T_1} + \Gamma_{ac} \frac{\bar{k}_{ac}}{2} n_c^0 \int_{-w}^w dx' \exp(-\bar{k}_{ac} |x'|), \quad (11)$$

$$\Gamma n_c^0 = \alpha \Gamma (n_b^0 - n_c^0) + \Gamma_{bc} \frac{\bar{k}_{bc}}{2} n_c^0 \int_{-w}^w dx' \exp(-\bar{k}_{bc} |x'|) + \Gamma_{ac} \frac{\bar{k}_{ac}}{2} n_c^0 \int_{-w}^w dx' \exp(-\bar{k}_{ac} |x'|). \quad (12)$$

We solve (3), (11) and (12) in n_c^0 for the following values of constants and parameters: $\Gamma_{ac} = 13.7 \times 10^6 \text{ s}^{-1}$, $\Gamma_{bc} = 19.2 \times 10^6 \text{ s}^{-1}$, $n = 4.6 \times 10^{16} \text{ m}^{-3}$, $I = 32 \text{ mW/cm}^2$, $\alpha = 2.4 \times 10^{-4}$. We use for the depolarization time T_1 in the uncoated glass cell and in the coated one the values $40 \mu\text{s}$ and 2 ms , respectively, deduced from experimental measurements as described in Sect. 4. In this case of a low atomic density and of a small diameter of the pump beam ($\bar{k}w \ll 1$), the neglecting of the integral terms in (11) and (12) underestimates by 12%–14% the values of the excited populations in each cell, but leads to a ratio similar to the one obtained by considering the integrals.

We find, for the uncoated glass cell, $n_c^0 \approx 6.0 \times 10^{12} \text{ m}^{-3}$ and, for the coated glass cell, $n_c^0 \approx 1.5 \times 10^{12} \text{ m}^{-3}$, which are in the ratio n_c^0 (uncoated)/ n_c^0 (coated) ≈ 4 , reproducing quite well the experimental value of 3. The slight discrepancy with the experimental value may be attributed essentially to the uncertainties in the estimating of the total pumping rate α (spatial profile of the beam and its intensity).

6 Discussion

We showed in Sect. 5 that the density of excited atoms (and thus the fluorescence intensity) in the volume outside the illuminated region is proportional to the density of excited atoms in the volume inside this region, which in turn is directly related to the number of resonant atoms. In the experiment discussed here, the efficient optical pumping mechanism has as a consequence the rapid decreasing of the fluorescence in the cell, unless the population of the resonant transition has been restored. In our experiments, the depolarization occurs essentially through the interaction of the polarized atoms with the internal surfaces of the cell during the adsorption (see Sect. 4). This process is very efficient in ordinary glass cells [21], while silane coatings are known [17] to reduce the energy of adsorption and, as a result, the dwell time, of alkali atoms on cell walls. The ground-state hyperfine populations are then less efficiently re-equilibrated in those coated cells and the number of atoms resonant with the laser is therefore lower than in uncoated cells.

By using the radiation-trapping process as a probe of the excited-state population in the vapor, we have thus evidenced and measured a surface effect. The difference of a factor 50 between the relaxation times of the two cells studied is expressed by a difference of only a factor 3 between the fluorescence intensities. This is essentially due to the very weak pumping rate: the stationary populations result from a competition between pumping (hyperfine polarization) and thermalization by the walls. The dynamics of the population evolution is governed by the slower mechanism, so that the depolarization rate is more effective in this evolution at higher pumping rates. The excited population would for instance be more than 40 times higher in the bare than in the coated cell for a pumping rate $\alpha = 2.4 \times 10^{-2}$, i.e. two orders of magnitude higher than in the experiment discussed here. Another specification of our experiments is that they are carried out at very low atomic density. In this regime, the radiation-trapping term in the rate equations for the atoms in the illuminated volume can be neglected, as it alters the excited population ratio between the two cells by only a few percent. The relative weight of the radiation-trapping term grows with optical density but the approximations made to interpret our experiments are no longer valid when the mean-free path assumed for the photons turns out to be much smaller than the cell radius ($\bar{k}R > 2$ [18]).

7 Conclusion

We observed the spatial distribution of radiation trapping in a dilute cesium vapor in glass cells with different internal surfaces, namely one coated with a silane film and the other uncoated (bare glass). Monitoring the fluorescence of atoms not directly illuminated by the resonant laser beam, we found that the surface state strongly affected the interaction between the laser and the vapor atoms. The mechanisms responsible for this behavior are, on one hand, the hyperfine pumping of the ground-state population by the resonant laser and, on the other hand, the thermalization of this pumping whose efficiency is determined by the interactions of the atoms with the surface. In silane-coated cells this thermalization is less efficient, preserving the hyperfine polarization for longer and thus diminishing the population avail-

able for interaction with the laser. Solving the system rate equations permits us, through comparison with experimental spatial profiles, to determine parameters of the surface interaction. In conclusion, in this work the very important role played by the atom–surface interaction in optical experiments in low-pressure cells is demonstrated. Conversely, this technique shows a way of obtaining information about surface effects by spectroscopically monitoring volume processes in resonant vapors.

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