VELOCITY DISTRIBUTION OF ALKALI ATOMS IN MICROMETRIC THIN CELLS

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1. Introduction

When one considers a gas at thermal equilibrium on the basis of the kinetic theory of gasses as a part of thermodynamics, it is natural that one should assume that the particles' velocity distribution is Maxwellian, and also that the velocity vector distribution is isotropic. These two points may not be perfectly equivalent, as the anisotropic shape of the container may induce anisotropy in the vector distribution. The kinetic theory only considers ideal containers, i.e. ideal surfaces at the boundary of the gas region. It is by a flux equilibrium between atoms incoming to and departing from the surface that one derives the Knudsen law for a rarefied gas ("molecular regime"), namely, a "cosθ" probability for a departing atom direction, with θ being the angle between the departing atom trajectory and the normal to the surface. This "cosθ" law is well-known, but has no connection with the microscopic description (for a discussion on these topics, see e.g. the review by Comsa and David [1]). It cannot be applied to accommodation effects (surface and gas at different temperatures, as often occurs in aeronautical studies), nor can it be used in a detailed study of the atomic desorption from a well-characterized surface; nevertheless, it remains widely adopted when various averagings have to be introduced in the modeling. The tradition for using such a "cosθ" model is so strong that detailed desorption studies performed under high vacuum, rather than in a thermal gas surrounding, often describe the angular behavior along a "cosθ" expansion, i.e. a $f(\theta)$ law as $f(\theta) = \Sigma_n a_n(\cos\theta)^n$ (see [1], and for examples of high value of the exponent *n*, see e.g. [2]).

Experimentally, it is not easy to perform the corresponding measurements, which demand a gas density low enough to make genuine surface effects observable. The high sensitivity of laser spectroscopy, and its ability to resolve different atomic velocities, make it an attractive tool for experimental tests of this law. At least two such experiments have found agreement with the "cosθ" law, one [3] with a thermal gas cell (with not many details about the nature of the surface), the other one [4] more closely connected with desorption processes on a specific surface.

In all cases, the angular dependence has not been specifically tested for atoms departing from the surface following a grazing incidence. Such a situation of grazing incidence appears to be of a specific interest. Indeed, there is nowadays a sustained activity of using laser spectroscopy to study a gas confined close to a surface [5-11].

For the techniques relying on the confinement of the vapor, the contribution of atoms with velocities nearly parallel to the wall, i.e. atoms leaving the surface following a grazing incidence, is enhanced and can even be made totally dominant, notably in non-linear spectroscopic schemes [6, 8-11], or when a FM technique is coupled to linear spectroscopy [7]. Relatively to the normal velocity component, the atoms with flight nearly parallel to the wall are "slow atoms". For a confinement close to a surface, one can expect that the roughness of the surface on the one hand, and the van der Waals attraction exerted by the surface on the other, would limit the existence of such "slow" atoms, notably when assuming linear atomic trajectories.

2. Principle of the experimental measurement

In a dilute vapor cell of a micrometric thickness irradiated under normal incidence by a single beam, the optical pumping to a non-absorbing level, such as a ground state hyperfine level of an alkali vapor (e.g. the clock transition of Cs at 9.192 GHz), is governed by its transient evolution [6,8]: only slow atoms reach a steady state (the medium is no longer absorbing), and the optical pumping is hence a velocity-selective process. The major assumption is that the atom trajectories are wall-to-wall, as atom-atom collisions are neglected owing to the dilute nature of the vapor. In principle [6,8], the thicker the cell, the narrower the velocity selection can be, provided the vapor remains sufficiently dilute. We have already described in detail [8] the effectiveness of this velocity selection. However, the natural width of the transition makes difficult the optical observation of atoms slower than the "natural" velocity selection v_{nat}, defined by $v_{nat} = u_{th}\gamma_{nat}/\Gamma_{Dopp}$ with γ_{nat} being the natural width of the transition, Γ_{Dopp}, its Doppler width, and u_{th}, the thermal velocity. To observe these slow atoms, one should rather consider a mechanical velocity selection, assuming a known (and constant) thickness L, within which an atom "marked" by a pump beam must propagate to a distance R in order to be probed [9].

Here, we consider a situation in which the pump beam is ring-shaped, and the probe beam, sent under normal incidence, is located precisely at the center of the pump beam ring (figure 1).

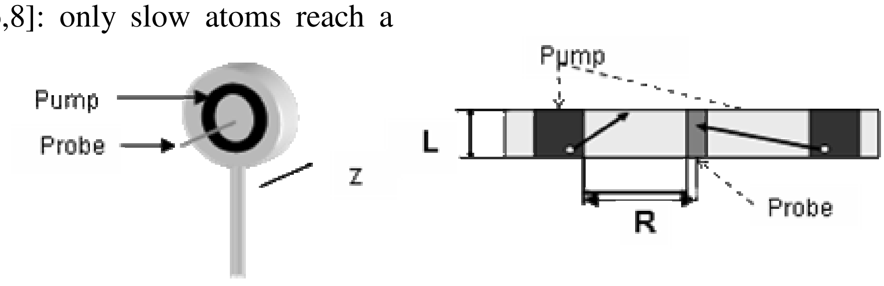

Figure 1. Principle of the experiment; a thin cell of thickness L much smaller than the overall diameter of the cell is irradiated by a ring-shaped pump beam. Only atoms with a slow normal velocity can leave the pump region and reach the axial probe region.

The probe beam intensity should be non-saturating. To count the pumped atoms travelling across the probe beam, one should assume that the probe absorption takes place in a steady-state mode: practically, as long as the cell thickness largely exceeds the wavelength [7], the transient build-up of absorption can be neglected, especially for the slow atoms. When scanning the frequency of the (normal incidence and minimum diameter avoiding saturation) probe beam, the atoms pumped at a distance appear only as a Doppler-free resonance. The pump beam should provide an efficient transfer and must be strongly saturating. For an alkali-metal resonance and a non-cycling transition, it is easy to reach these strongly saturating intensities, owing to the very long relaxation time constants of the optical pumping process. This standard hypothesis of a negligible relaxation of the optical pumping requires that the pump beam internal diameter must be well defined in order to avoid optical pumping of atoms located in the dark region separating the pump and probe beams.

In an independent work [11], a calculation of the line-shape has been performed as a function of the *L* and *R* parameters, under the usual assumption that the velocity distribution in the thin cell obeys a 3D-Maxwellian distribution. Here, we concentrate on the idea that some slow atoms may be missing, and we analyze the influence of a missing "slice" in the velocity distribution of slow atoms.

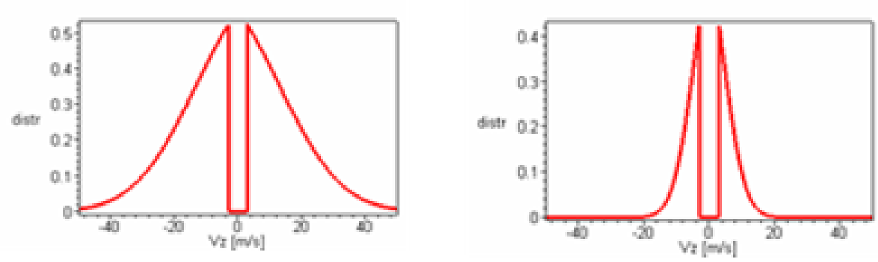

Figure 2. Velocity distribution of atoms reaching the probe region, after having travelled from the pump region. A Maxwellian distribution is assumed, with a thermal velocity u_{th}=250 m/s. The *R/L* ratio is 10 (left) or 30 (right), assuming an initial velocity distribution in which atoms with $|v_z| < 3$m/s do not exist. The pump intensity is assumed to be so strong that the calculation does not depend on the pump frequency detuning.

Figure 2 shows the relative velocity distribution of pumped atoms in the situations *R/L* = 10, and *R/L* = 30. The probe spectrum is expected to be Doppler-free, or at least to exhibit only a weak residual Doppler broadening if the *R/L* ratio remains small. Assuming a modified velocity distribution $f(v_z) = 0$ for $|v_z| \leq v_{missing}$, and $f(v_z) = (u_{th}\pi^{1/2})^{-1}\exp(-v_z^2/u_{th}^2)$, for $|v_z| > v_{missing}$ [11], in which too "slow" (normal velocity) atoms are missing, one derives the distribution of the (normal) velocity of pumped atoms travelling to the probe region. Figure 3 shows that when varying *R/L* (e.g, by changing the pump beam size), the amplitude of the probe absorption, as measured on resonance, yields a sensitive indication of the missing velocities.

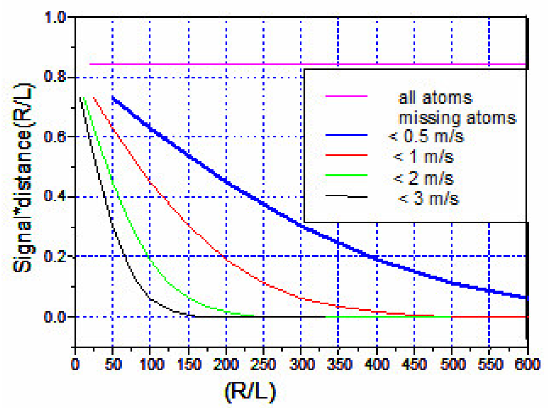

Figure 3. Amplitude of the probe absorption as a function of *R/L*, for various distributions of (normal) atomic velocities. The height is normalized, so that for a distribution with no loss of slow atoms, the expected reduction of

signal when increasing the pump-probe separation is already taken into account. One has taken u_{th} = 250 m/s.

3. Experimental set up and results

We are presently implementing the corresponding experiments on Cs vapor confined in a thin cell with sapphire windows, with the spectroscopic measurements to be performed on the Cs D_2 line. The cell is a cylindrical one, with a diameter of ~ 3cm, with Cs deposited in a bottom reservoir (as shown schematically in figure 1). The windows are made of standard high-quality polished sapphire. The roughness was estimated at ~5 nm. We measured interferometrically the thickness to be ~19 µm. When performing this thickness measurement at various points, we found that the deviation from parallelism did not to exceed 100 nm over 1 mm. The cell (and the Cs reservoir) was generally heated up to ~60 – 70 °C to increase the Cs atomic density. From our experience with comparable cells, neither the thickness nor the parallelism should be thus affected. For the spectroscopic measurements, we used a single laser tunable around the D_2 line (852 nm), namely, a DFB type diode laser. It is a narrow linewidth (< 3 MHz) semiconductor laser delivering a relatively high power (150 mW) sufficient to provide an intense (highly saturating) pump beam. The laser beam is split into a standard weak probe beam and a pump beam modulated by a mechanical chopper (typically at 2 kHz), and shaped into a diverging ring by optical elements (see figure 4).

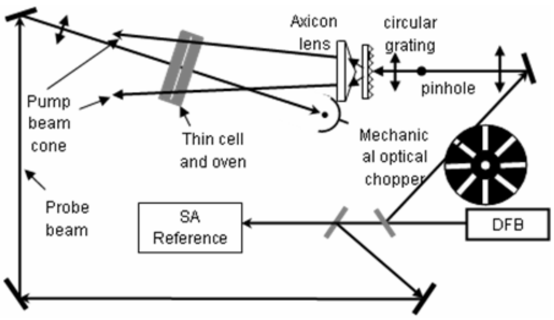

Figure 4. Experimental set-up.

The probe transmission is detected using a lock-in scheme synchronous with the chopping of the pump. The most essential point in the set-up is the shaping of the pump beam, with the aim to convert as much power as possible into a ring-shape geometry. For this purpose, we implemented a special transmission grating with circular grooves, which converts the beam into (conically) diverging diffraction rings. An axicon lens allows a reduction of the diverging rings (the half-angle is ~11° for the first order).

Figure 5 shows a spectrum of the probe beam transmission across the Cs D_2 line, starting from the ground state $F_g = 4$, for a pump-probe spatial separation ~1.5 mm. One distinctly observes the sub-Doppler contribution of the separate hyperfine lines. We note that the $F_g = 4 - F_e = 4$ transition appears to be the larger one, while for a linear probe spectrum one expects the $F_g = 4 - F_e = 5$ transition to be larger than the $F_g = 4 - F_e = 4$. Actually, when using only a single laser, the efficiency of the pump beam on the non-cycling $F_g = 4 - F_e = 5$ transition is lower than for the $F_g = 4 - F_e = 4$ transition (and the $F_g = 4 - F_e = 3$ transition). We also note that there is a residual Doppler background, probably to be attributed to atoms which have collided with the wall without losing their excitation received from the pump beam.

Another notable point to be considered when discussing figure 5 is the presence of a weak quadrature signal that is indicative

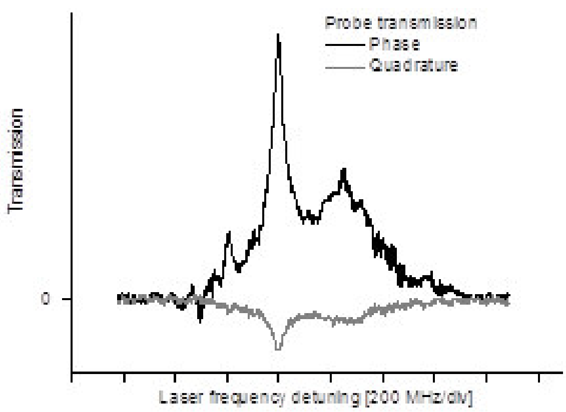

Figure 5. Probe beam transmission spectrum across the Cs D_2 line from $F_g = 4$ to (left-to-right) $F_e = 3, 4, 5$. The pump-probe separation is ~1.5 mm. The Cs source temperature is 60 ºC.

of the stronger and narrow contribution of the $F_g = 4 - F_e = 4$ transition. For our pump-probe separation (~1.5 mm), an atom's time-of-flight between the pump and the probe can be estimated to be ~7.5 µs, a value in agreement with the phase shift of the signal (as can be measured in fig. 5) with respect to the 2 kHz pump modulation.

4. Conclusions

We were able to observe the response of atoms flying nearly parallel to the wall, with the level of angular selectivity obtained mechanically in fig. 5 (~1.5 mm over a 19 µm thickness) corresponding to the observation of atoms with a normal velocity ~3 m/s, smaller than the one just expected from an optical selection. It is already closely comparable to the one reported in [10] in an experiment where the cell thickness had not been precisely controlled.

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