Compensation of Doppler Broadening by Velocity-Dependent Light-Shifts

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1. Introduction

When a 2-level atom is irradiated by a quasi-resonant light beam, having a frequency ω_L close to the atomic frequency ω_0, its energy levels a and b are shifted. Perturbation theory shows that the so called light-shift ε_a of the lower level a (which is equal to $-\varepsilon_b$) is proportional to the light-intensity I and inversely proportional to the detuning $\omega_L-\omega_0$, provided that this detuning is not too small.

$$\varepsilon_a = - \varepsilon_b \sim I/(\omega_L - \omega_0) \tag{1}$$

Light-shifts have been studied several years ago, before the advent of lasers, in optical pumping experiments using ordinary light sources [1][2]. By choosing a convenient polarization for the light beam, it was possible to shift two Zeeman sublevels of the ground state by different amounts and to detect this effect by a shift of the magnetic resonance curve between these two sublevels. As a consequence of the long relaxation times in the ground state, this resonance curve is very narrow and light-shifts as small as one hertz have been easily measured.

Now, with laser sources, light-shifts have considerably increased, from a few Hz to several GHz [3], and they can be observed directly on optical transitions. Actually, they introduce a limitation in the accuracy of high resolution spectroscopic measurements and they have to be avoided or, at least, carefully controlled.

On the other hand, during the last few years, new physical effects using light-shifts have been proposed and studied. For example, atomic beam deflection experiments using transverse dipole forces, and discussed in this meeting [4], may be interpreted in terms of position-dependent light-shifts. An intensity gradient $I(\vec{r})$ of the laser beam introduces a $\vec{r}$-dependent light-shift $\varepsilon_a(\vec{r})$ of the ground state. When the detuning is large enough, $\varepsilon_a(\vec{r})$ appears as a potential energy for the atom, giving rise to dipole forces, $-\vec{\nabla}\varepsilon_a(\vec{r})$, which can deflect an atomic beam.

In this paper, we discuss another effect using velocity-dependent light-shifts for compensating the Doppler broadening of an optical transition. We will briefly outline the principle of such a scheme which is analyzed in more details in references [5] and [6] (see also [7]). Experimental evidence for the narrowing mechanism has been obtained recently [8]. We present here additional experimental results obtained on ^{20}Ne and which exhibit important polarization effects.

Let us finally mention that other mechanisms for producing velocity dependent energy shifts, which could lead to a compensation of the Doppler broadening, have been recently suggested. They use quadratic Stark shifts in crossed static electric and magnetic fields [9], or motional interactions of spins with static electric fields in polar crystals [10].

2. Discussion of the narrowing mechanism

Consider an atom moving, in the laboratory frame, with a velocity v towards a laser beam with frequency ω_L (Fig.1).

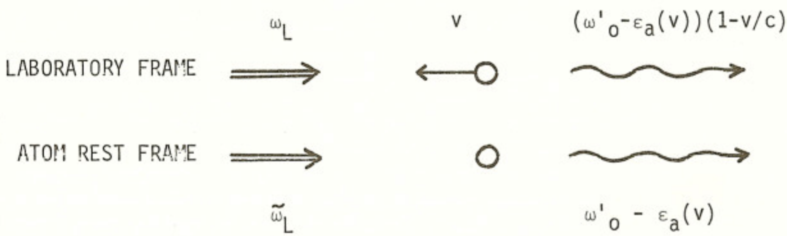


Fig.1 - *Laser and emission frequencies in laboratory and atom rest frames.*

The light-shifts induced by such a laser irradiation have to be evaluated in the rest frame of the atom where the laser frequency is Doppler shifted from ω_L to $\tilde{\omega}_L = \omega_L\ (1 + \frac{v}{c})$. The v-dependence of $\tilde{\omega}_L(v)$ results in a v-dependent detuning $\tilde{\omega}_L(v) - \omega_0$ between the apparent laser frequency $\tilde{\omega}_L(v)$ and the frequency ω_0 of the transition a-b. It follows that the light-shift ε_a of a which depends on this detuning is also v-dependent.

Suppose now that we observe, in the same direction as the laser, the light spontaneously emitted from a third level c to a by an atom excited in c (for example by a discharge). The frequency ω'_0 of transition a-c is completely off-resonance with ω_L, so that level c is not perturbed by the laser. In the rest frame, the emitted frequency is equal to ω'_0 corrected by the light shift $\varepsilon_a(v)$ of a, i.e to $\omega'_0 - \varepsilon_a(v)$. Coming back to the laboratory frame introduces the well-known Doppler factor $(1 - \frac{v}{c})$ since the atom is moving away with a velocity v.

Now, the basic idea discussed in this paper is to try to achieve a compensation between the v-dependence of the light-shifted internal frequency $\omega'_0 - \varepsilon_a(v)$ and the v-dependence of the emission Doppler factor $(1 - \frac{v}{c})$, in order to have, in the laboratory frame, all atoms emitting at the same frequency in the forward direction.

Such a compensation condition is easily found to be

$$\varepsilon_a(v) = - \omega'_0\ v/c \qquad (2)$$

Before describing how it can be achieved, let's first discuss some important characteristics of this new scheme.

First, the narrowing is not due to a population effect. We are looking at a spontaneous emission signal which is independent of the population of the final state a and which originates from a level c which is not perturbed by the laser. Levels a and b could even be empty.

Second, there is no power broadening of the Doppler free line. When the compensation condition is fulfilled, all atoms emit at the same frequency. The width of the line is just the homogeneous width γ. These features clearly distinguish this effect from others, such as fluorescence line narrowing [11] [12] where the laser irradiation creates in the velocity distribution of atoms a population hole or a population peak which is power broadened.

Finally, and this is perhaps the most interesting point, this effect is highly anisotropic. If one looks at the light emitted not in the forward but in the backward direction, the Doppler emission factor changes from $(1 - \frac{v}{c})$ to $(1 + \frac{v}{c})$. If the Doppler broadening is compensated for one direction of emission, it is clearly doubled for the opposite one. Such a scheme could therefore provide an atomic medium emitting with the homogeneous width γ in one direction and with twice the Doppler width Δ in the opposite one. Forward-backward asymmetries are well known effects in laser spectroscopy of 3-level systems [12] but, here, such an asymmetry could be particularly important because of the large difference between γ and Δ.

3. How to get a light-shift proportional to v ?

Two possibilities are represented on Fig.2. One of them (Fig.2a) considers a 2-level system with frequency ω_0 irradiated by two co-propagating laser beams with frequencies $\omega_0 + \delta$ and $\omega_0 - \delta$. In the second scheme (Fig.2b), which is simpler experimentally, we have a single laser beam ω_0, which is σ-polarized and which excites a J = 0 to J = 1 transition in a static field, so that the two Zeeman sublevels b_+ and b_-, which are coupled to the laser, are detuned by $+\delta$ and $-\delta$ (δ is the Zeeman splitting).

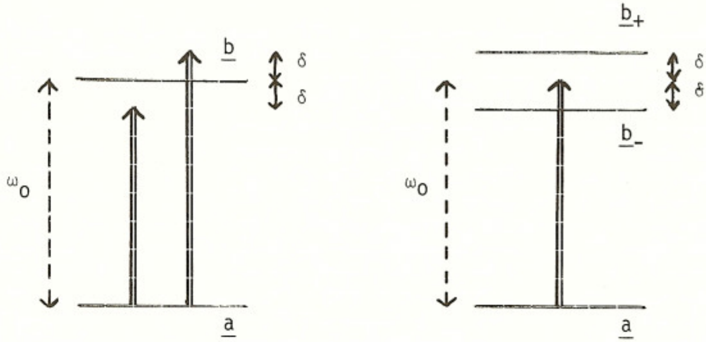


Fig.2a Fig.2b

For a v = 0 atom, we have in both cases two opposite detunings, so that the two light-shifts produced by the two lasers in the first case, by the virtual excitation of the two Zeeman components in the second one, balance. It follows that $\varepsilon_a(v=0) = 0$.

On the other hand, when v is different from zero, the Doppler shift of the laser frequencies introduce unequal detunings and, consequently, unbalanced light-shifts. This shows that $\varepsilon_a(v)$ is an odd function of v which depends on the laser intensity I and on the detuning δ.

From now on, we will consider only the second scheme (Fig.2b). The laser intensity I being fixed (generally at its highest possible value), we have to find if it is possible to adjust δ so that the linear term in the expansion of $\varepsilon_a(v)$ coincides with $-\omega'_0\ v/c$. We have also to understand the effect of higher order terms (in v^3, v^5, ...) and to determine under what conditions they can be neglected.

4. Calculation of the compensation condition and of the emission spectra

It turns out that the light intensities required for achieving the compensation condition may be quite large,so that a non-perturbative treatment of the atom-laser coupling is necessary [13].

There are actually three relevant states of the atom-laser system which are strongly coupled. Let $|\underline{a}, n\rangle$ be the state corresponding to the atom in $\underline{a}$ in presence of n laser photons. We take its unperturbed energy equal to 0 so that the corresponding perturbed energy will represent the light-shift ε_a. The atom in $\underline{a}$ can absorb one laser photon and jump into one of the two Zeeman sublevels $\underline{b}_+$ or $\underline{b}_-$ of $\underline{b}$, so that we have also to consider the states $|\underline{b}_+, n-1\rangle$ and $|\underline{b}_-, n-1\rangle$ with unperturbed energies respectively equal to $\delta - u$ and $-\delta - u$ where δ is the Zeeman splitting and $u = \tilde{\omega}_L - \omega_0 = \omega_0\ v/c$ the detuning of the laser with respect to ω_0 in the rest frame, proportional to v. The atom-laser coupling V is characterized by the matrix elements

$$\langle \underline{a}, n|\ V\ |\ \underline{b}_\pm, n-1\rangle = \omega_1/2 \tag{3}$$

where ω_1 is a Rabi frequency equal to the product of the atomic dipole by the laser electric field. We suppose $\omega_1 \gg \gamma$ so that we can neglect the damping processes in a first step and diagonalize the matrix

$$\begin{pmatrix} 0 & \omega_1/2 & \omega_1/2 \\ \omega_1/2 & \delta-u & 0 \\ \omega_1/2 & 0 & -\delta-u \end{pmatrix} \tag{4}$$

The eigenvalues of (4) give the three perturbed energies versus u, i.e. versus v (in particular, the light-shift of $\underline{a}$ is not just the sum of the two light-shifts associated with the coupling to $|\underline{b}_+, n-1\rangle$ alone, and $|\underline{b}_-, n-1\rangle$ alone, as it would be in a perturbative treatment). One gets in this way an exact expression for the linear term (in v) of $\varepsilon_a(v)$ leading, for the compensation condition (2), to the following exact relation

$$\omega_1^2/(\omega_1^2 + 2\delta^2) = \omega'_0/\omega_0 = s \tag{5}$$

between ω_1^2 (proportional to the laser intensity), δ, and ω'_0/ω_0.

The diagonalization of (4) gives also three perturbed states $|i, n\rangle$ (i=1,2,3), which all contain admixtures of $|\underline{a}, n\rangle$, so that the emission spectrum $\underline{c} \rightarrow \underline{a}$

is actually a triplet corresponding to the three transitions $|\underline{c}, n\rangle \to |i, n\rangle$. These three lines have a simple physical interpretation in the perturbative regime. We have first the Doppler free line corresponding to the spontaneous emission from $\underline{c}$ to the light-shifted level $\underline{a}$ (Fig.3a), and then two Raman sidebands corresponding to inverse Raman processes where the atom starting from $\underline{c}$ spontaneously emits one photon and absorbs one laser photon to end in sublevels $\underline{b}_+$ or $\underline{b}_-$ which are also light-shifted (Fig.3b).

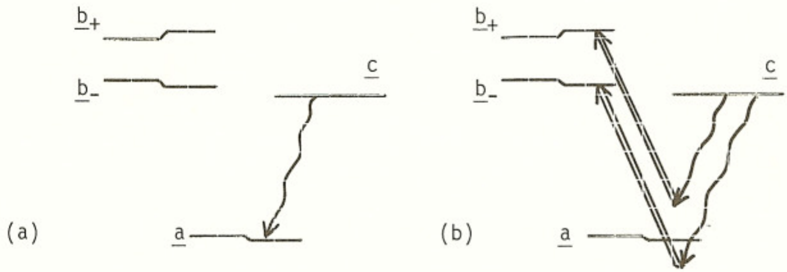


Fig. 3 - *Perturbative interpretation of the three lines emitted from* $\underline{c}$

Because of the Maxwellian distribution of velocities, the diagonalization of (4) must be done for a range of values of v corresponding to the Doppler width Δ of $\underline{a}$ - $\underline{b}$.

If the light intensity is very large, so that

$$\omega_1 \gg \Delta \tag{6}$$

we can neglect the curvature of the perturbed energy levels associated with (4) when v varies within the Doppler width. In such a case, we have, when condition (5) is fulfilled, a complete compensation of Doppler broadening : all atoms emit the central line at the same frequency ω'_0. The theoretical spectrum obtained in these conditions (Fig.4) exhibits a central narrow peak at ω'_0 with the homogeneous width γ. The compensation of Doppler broadening does not occur for the two Raman sidebands which are located at a distance $\pm\, \omega_1/\sqrt{2s}$ and which remain Doppler broadened. The weights of the lines are respectively equal to s/2, 1-s, s/2. They are comparable, so that the height of the narrow peak is much larger.

On the other hand, if

$$\Delta \gg \omega_1 \gg \gamma \tag{7}$$

we cannot neglect the curvature of the energy levels versus v within the Doppler width. In such a case, we have only a partial compensation of Doppler broadening, occurring for the fraction of the velocity distribution where the linear approximation is valid. The spontaneously emitted frequencies are redistributed into a central symmetric narrow peak (which represents the contribution of atoms having a light shift linear in v), and two sharp-edged sidebands (see for exemple the theoretical curve B = 42 G of Fig.5).

If one increases the light intensity and, simultaneously, changes the Zeeman splitting δ in order to maintain (5), the height of the central peak would increase without any broadening, whereas the two sidebands would move away and become Doppler broadened.

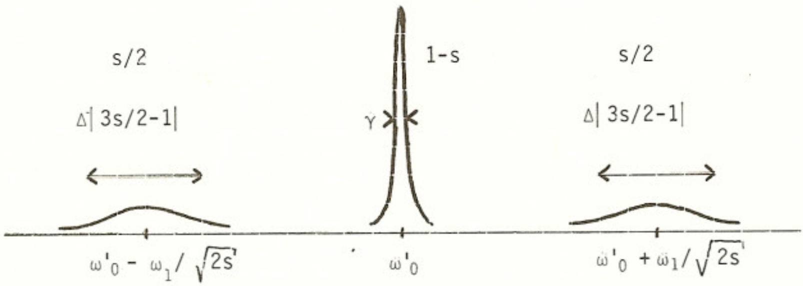


Fig.4 - *Theoretical emission spectrum in the case of a complete compensation of Doppler broadening.*

5. Experimental results

The first experimental evidence for the compensation of Doppler broadening by velocity-dependent light-shifts has been obtained on ^{20}Ne atoms [8].

The three levels a, b, c are respectively the $1s_3$, $2p_2$ and $2p_{10}$ levels (Paschen notations). The output of a c.w. dye laser is tuned to the wavelength 6163 Å of the a - b transition and is focussed after a σ polarizer inside a d.c. discharge cell containing 1.5 torr of ^{20}Ne and put in a static magnetic field. The π-polarized fluorescence which is emitted from the $m = 0$ sublevel of $2p_{10}$, at 7439 Å, is isolated by color and interference filters and spectrally analyzed with a confocal Fabry-Perot interferometer. The laser power was of the order of 150 mW and the waist w in the discharge cell of the order of 400μ, which leads to values of ω_1 of the order of 200 MHz, small compared to Δ ($\Delta \sim 1100$ MHz), but large compared to γ ($\gamma \sim 30$ MHz). We are therefore in a regime of partial compensation of Doppler broadening.

Fig. 5 shows, on the left part, the recorded emission spectral profiles for increasing values of the static magnetic field. They are in good agreement with the corresponding computed curves represented on the right part. In zero magnetic field, we have actually a 2-level system since only a superposition of b_+ and b_- is coupled to the laser. We get the well known Autler-Townes doublet [14] observed on the spontaneous emission from c. When the magnetic field is increased, a narrow structure appears in the center of the spectrum, reaches a maximum around B = 42 G, and then broadens. For B = 42 G, ω_1 and δ satisfy the compensation condition (5) and one gets the Doppler free line discussed above with the two sharp-edged sidebands.

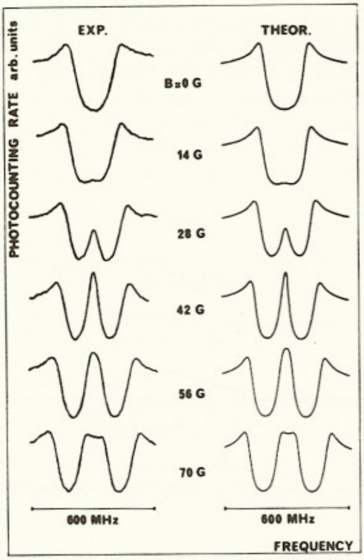


Fig.5 - *Experimental and theoretical emission spectral profiles on the $2p_{10} \rightarrow 1s_3$ transition of ^{20}Ne (laser excitation of the $2p_2$ - $1s_3$ transition)*

More recently, we have studied other transitions of ^{20}Ne (Fig.6) for which the Zeeman structure appears now in the lower level a ($1s_4$) of the a - b transition ($1s_4$ - $2p_3$) which is coupled to the laser. With this new energy level scheme, it is now the upper state b which undergoes a light shift ε_b linear in v, so that the compensation of Doppler broadening can be achieved on the inverse Raman spontaneous line starting from c ($2p_{10}$, m =0), whereas the two spontaneous transitions c - a_+ and c - a_- give rise to two sidebands which remain Doppler broadened or sharp-edged.

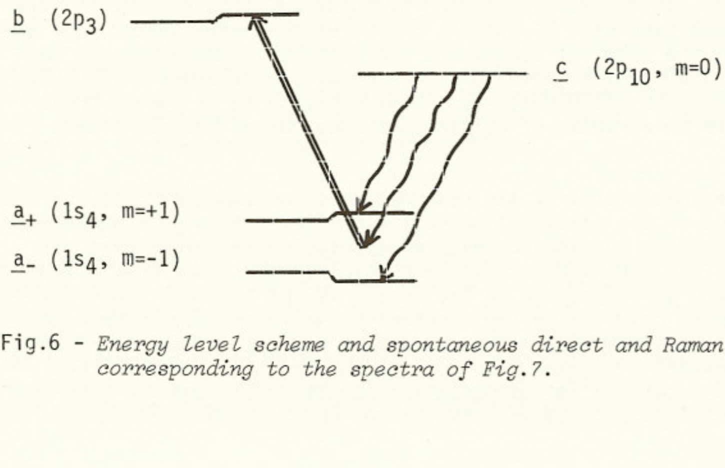
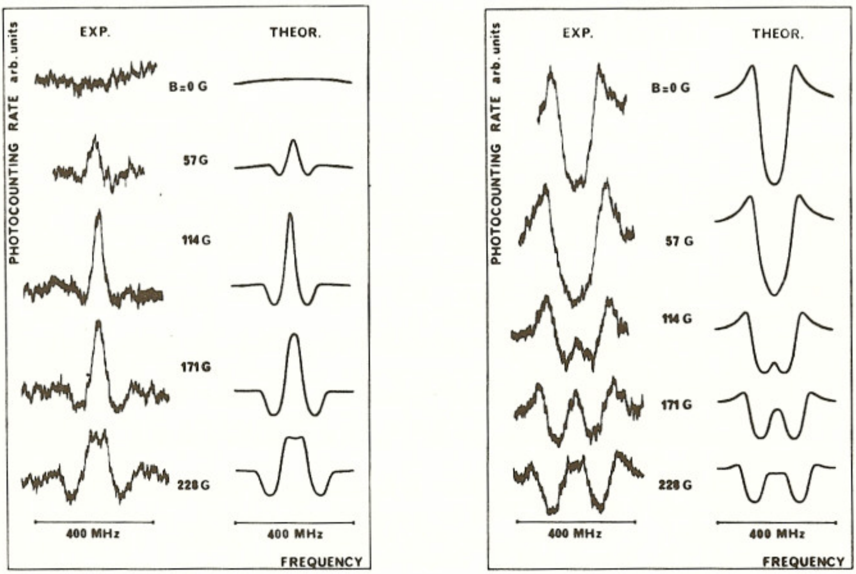


Fig.6 - *Energy level scheme and spontaneous direct and Raman transitions corresponding to the spectra of Fig.7.*

Note that we have now the possibility of choosing at the detection a σ polarization either parallel or perpendicular to the one of the laser beam. The emission rate on the three lines $|c, n\rangle \to |i, n\rangle$ is proportional to $|\langle i, n| \vec{\varepsilon}^{*}.\vec{D}\, |c, n\rangle|^2$ ($\vec{\varepsilon}$ is the detection polarization, $\vec{D}$ the atomic dipole operator). The emission spectra are therefore different on these two polarizations.

Fig.7a shows the comparison between experimental [15] and theoretical spectra when the detection polarization is parallel to the laser one. In zero magnetic field, no structure appears. The reason is that this detection polarization connects c to a linear superposition of a_+ and a_- which is just orthogonal to the one coupled to the laser. The central Doppler free line appears, when the field increases, with a better contrast than on Fig.5. The compensation condition corresponds to a field around 114 G [16]. The results obtained when the detection polarization is orthogonal to the laser one are represented on Fig.7b. The zero field spectrum exhibits now the Autler-Townes doublet. But, for this polarization, a perturbative treatment shows that the inverse Raman process vanishes for $v = 0$, because of a destructive interference between the two possible paths via a_+ and a_- (Fig.6) This is why the height of the central narrow peak is now smaller than the one of the two sidebands.



(a) (b)

Fig.7 - *Experimental and emission spectral profiles on the $2p_{10} \to 1s_4$ transition of ^{20}Ne (laser excitation of the $2p_3 - 1s_4$ transition). The σ detection polarization is either parallel (a) or perpendicular (b) to the laser one.*

6. Conclusion

In conclusion, we have proposed a new scheme for compensating the Doppler broadening by velocity dependent light-shifts and we have obtained experimental results giving good confidence in the theoretical analysis of such a scheme.

Up to now, we have used moderate laser intensities (150 mW) and moderate focalisations ($w = 400\mu$). Commercial ring lasers can give now powers higher by a factor 10, even still higher if one works inside the laser cavity. The laser beam could be also focussed to smaller waists if one uses a second laser beam for probing the $\underline{c}$ - $\underline{a}$ transition rather than a spontaneous emission signal.

Consequently, it does not seem hopeless to obtain, even with c.w. lasers, values of ω_1 of the order of, or larger than Δ. In such a case, one could achieve a nearly complete compensation of Doppler broadening leading to forward-backward asymmetries as large as 10 or 100. This would open the way to various interesting applications such as reduction of threshold in laser media, ring lasers, directed Doppler free superradiance, Doppler free coherent transients, non-reciprocal devices...

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[16] When the Zeeman structure exists in the lower level a (scheme of Fig.6) calculations similar to the ones of section 4 lead to a slightly different compensation condition $2\delta^2/(\omega_1^2 + 2\delta^2) = s$.