COMPENSATING DOPPLER BROADENING WITH LIGHT-SHIFTS

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We discuss the possibility of a compensation between laser induced light-shifts and linear Doppler shifts. A specific example is considered. We derive the compensation condition, give calculated emission spectra, and discuss some interesting features of this effect, such as its high anisotropy.

1. Introduction

Displacement of atomic energy levels by a quasi-resonant irradiation have been studied for a long time [1]. The interest for these so-called light-shifts has been renewed by the development of laser sources. First, these light-shifts have considerably increased, from a few Hz (corresponding to ordinary light sources such as RF excited discharges) [1,2] to several GHz (pulsed lasers) [3]. Second, in high resolution spectroscopy experiments (optically pumped atomic clocks [4], two-photon Doppler free spectroscopy [5]...), these light-shifts have to be carefully studied and controlled because they could otherwise represent a severe limitation in the precision of determination of atomic frequencies. Finally, light-shifts are closely connected to Autler-Townes effect [6] which has been recently extensively studied in the optical range [7,8].

In this paper, we discuss a possibility of using the perturbations associated with light-shifts for compensating the Doppler broadening of spectral lines. We present an improved and more realistic version of a scheme first proposed in ref. [9].

The basic property used in this paper is that light-shifts depend not only on the laser intensity, but also on the detuning between the laser and atomic frequencies. In the scheme of ref. [9], an atomic vapour (atomic resonance frequency ω_0) is irradiated by two co-propagating laser beams, with frequencies $\omega_0 + \eta$ and $\omega_0 - \eta$ and with the same intensity. For an atom at rest, the two light-shifts balance since the two detunings $\pm\eta$ are opposite. But, for an atom moving with velocity v along the laser axis, the two laser frequencies are Doppler shifted in the atom rest frame, which leads to unequal apparent detunings and consequently to unbalanced light-shifts. It follows that the laser perturbation of atomic frequencies is, to lowest order, proportional to v. By adjusting the laser intensity, one could use this v-dependence of the atomic Bohr frequencies for balancing the emission Doppler factor. In this paper, we consider a scheme where a single laser beam, with frequency ω_0, is coupled to two Zeeman components of an atomic line with frequencies $\omega_0 \pm \delta$. The two light-shifts associated with the excitation of the two Zeeman components balance for $v = 0$ and give rise to a linear v-dependence of the atomic frequency which can compensate the emission Doppler factor. We give an explicit analytical form for the compensation condition, calculated shapes for the emission spectrum and we suggest some interesting application of this effect.

Let's finally emphasize that light-shifts are not the only way to introduce a velocity dependent perturbation of atomic frequencies which could be used for compensating the Doppler broadening. Other schemes using crossed static electric and magnetic fields have been recently proposed [10]. Similar effects giving rise to a "Doppler narrowing" in spin-flip Raman scattering experiments in polar crystals have been also reported [11].

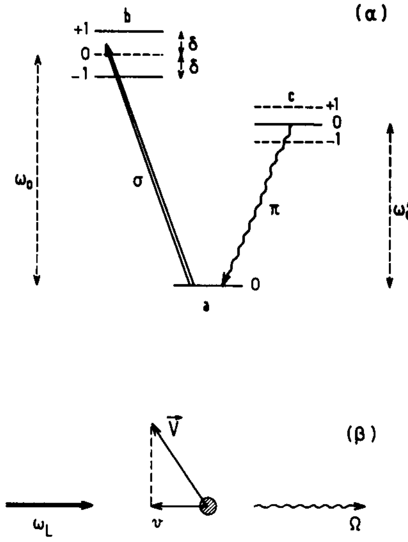

Fig. 1. (α) Energy diagram of the 3-level atom. (β) Relative dispositions of the laser beam, atomic velocity and observation direction.

2. Discussion of a specific example

For the sake of simplicity, we limit ourselves to a specific example although other schemes of energy levels could be considered.

An atomic vapour is irradiated by an intense laser beam with frequency ω_L close to the frequency ω_0 of the atomic transition *ab* (fig. 1). One observes the spectral distribution $L_F(\Omega)$ of the light emitted in the forward direction on the transition *ca* (Bohr frequency ω_0'), the level *c* being populated for example by a weak discharge. The angular momenta of *a, b, c* are respectively 0, 1, 1. A static magnetic field produces a Zeeman splitting δ in level *b*. The laser light (double arrows of fig. 1) is σ polarized so that only sublevels $b_\pm$ of *b* ($m = \pm 1$) are coupled to *a*. The fluorescence (wavy arrows of fig. 1) is observed through a π analyzer so that only sublevel c_0 of *c* ($m = 0$) is involved.

For an atom moving with a velocity v along the laser beam direction (fig. 1-β), the laser frequency is Doppler shifted from ω_L to ‡

$$\tilde{\omega}_L = \omega_L(1 + v/c) \simeq \omega_L + \omega_0\, v/c. \qquad (1)$$

‡ In the Doppler correction term, the difference between atomic and laser frequencies can be neglected as well as recoil and higher order Doppler effects.

The laser irradiation perturbs such an atom, mixing and shifting levels *a* and *b* (the perturbation of level *c* can be neglected since the laser ω_L is completely off resonance for transition *ca*). This introduces a shift and a splitting of the line emitted at ω_0' by the atom in its rest frame. The important point to realize is that the new emission frequencies depend not only on the laser intensity, but also on the apparent frequency $\tilde{\omega}_L$, and are therefore v-dependent. We will note $\tilde{\Omega}(\tilde{\omega}_L)$ these frequencies. It follows that, in the laboratory frame, the frequencies Ω of the emitted light, which are given by

$$\Omega(v) = \tilde{\Omega}(\tilde{\omega}_L)\,[1 - v/c] \simeq \tilde{\Omega}(\tilde{\omega}_L) - \omega_0'\, v/c, \qquad (2)$$

depend on v through two different physical effects: the v-dependence of the laser induced light-shifts [contained in the first term of (2)] and the well known emission Doppler shift [last term of (2)].

The basic effect discussed in this paper lies in the possibility of a compensation between these two v-dependences leading to a Doppler free emission on *ca*. From (1) and (2), one easily derives a mathematical condition for such a compensation:

$$\mathrm{d}\Omega/\mathrm{d}v = 0 \leftrightarrow \mathrm{d}\tilde{\Omega}/\mathrm{d}\tilde{\omega}_L = \omega_0'/\omega_0. \qquad (3)$$

This leads us to consider "frequency diagrams" giving in the atom rest frame the laser perturbed emission frequencies $\tilde{\Omega}$ versus the apparent laser frequency $\tilde{\omega}_L$. Condition (3) appears simply as a condition on the slope p of the corresponding curve which must be equal to the ratio $s = \omega_0'/\omega_0$. Since the curves of the frequency diagram are not straight lines, condition (3) is satisfied only for a discrete set of points. But the Doppler effect remains compensated around these points as long as the curve remains sufficiently close to its tangent.

Actually, for atoms moving in a cell, the frequency diagram has to be considered only over an interval of width Δ (Doppler width) around ω_L (see eq. (1)). If condition (3) is realized for $\tilde{\omega}_L = \omega_L$ and if the curvature of the lines of the frequency diagram may be neglected over an interval Δ, a compensation of Doppler effect occurs for all atoms. Otherwise, we will have only a local compensation around the points solutions of (3), lying in the Doppler distribution.

We have represented on fig. 2 such a frequency diagram for the atom of fig. 1. The horizontal dotted line corresponds to the unperturbed atomic frequency ω_0'

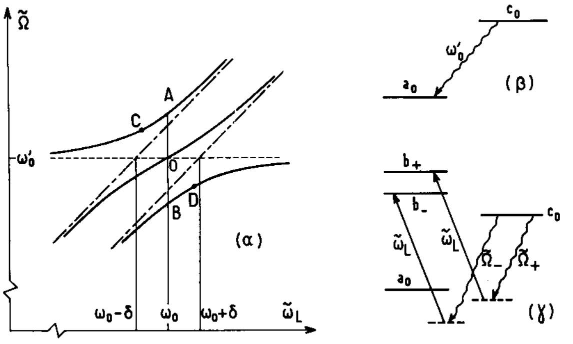

Fig. 2. (α) Frequency diagram giving, in the atom rest frame, the frequencies $\tilde{\Omega}$ emitted around ω_0' by an atom perturbed by a laser with apparent frequency $\tilde{\omega}_L$ close to ω_0. (β) Zeroth order process (in the laser intensity) corresponding to the dotted line of fig. 2-α. (γ) First order inverse Raman processes corresponding to the interrupted lines of fig. 2-α.

which is the only one to be spontaneously emitted in the absence of laser irradiation (fig. 2-β). For weak laser intensities, two new frequencies $\tilde{\Omega}_\pm = \omega_0' + \tilde{\omega}_L - (\omega_0 \pm \delta)$ appear (interrupted lines of fig. 2-α), due to inverse Raman processes where the atom starting from c_0 emits one of these two frequencies and then absorbs one laser photon ending to b_+ or b_- (fig. 2-γ). When the laser intensity is large enough to saturate the atomic transition, i.e. when the Rabi frequency ω_1 becomes of the order of the radiative and collisional rates γ, a non-perturbative treatment of the coupling is necessary. The three processes of figs. 2-β,γ mix up and give rise to new frequencies which can be easily computed from a dressed atom approach [12] and which are represented by the full lines of fig. 2-α. The two crossings between the dotted and interrupted lines, which correspond to a resonance between $\tilde{\omega}_L$ and the frequencies $\omega_0 \pm \delta$ of one of the two Zeeman components ab, transform into "anticrossings".

Point 0, with coordinates $\tilde{\omega}_L = \omega_0$, $\tilde{\Omega} = \omega_0'$, is obviously a center of symmetry of the frequency diagram. It does not move when the laser intensity is increased. The slope p_0 in 0 can be easily computed and is found to be:

$$p_0 = \omega_1^2/(\omega_1^2 + 2\delta^2). \tag{4}$$

When the ratio ω_1/δ between the Rabi frequency ω_1 and the Zeeman detuning δ increases from 0 to $+\infty$, p_0 varies from 0 to 1 ... One can therefore compensate the Doppler effect for a certain value of the laser intensity given, according to (3) and (4), by:

$$\omega_1^2/(\omega_1^2 + 2\delta^2) = \omega_0'/\omega_0 \leftrightarrow p_0 = s, \tag{5}$$

which requires $s = \omega_0'/\omega_0$ to be smaller than 1. Furthermore, since point 0 is, by symmetry, an inflexion point, the curve remains close to its tangent over a large interval (limited only by the third order derivative), which is a favourable condition for our purpose.

3. Complete compensation of Doppler broadening

We consider first the very simple case where the laser is tuned on the atomic frequency ($\omega_L = \omega_0$) and where ω_1 and δ are large compared to the Doppler width Δ. In such a case, the frequency diagram has to be used around the points 0, A, B of abscissa ω_0 (fig. 2-α) over such a small interval that the three curves may be assimilated to their tangents. If, in addition, the laser intensity is adjusted so that condition (5) is fulfilled, all atoms will emit at the same frequency ω_0' giving rise to the Doppler free line of fig. 3 having a pure homogeneous width γ (determined by collisional and radiative dampings). They will also emit two other lines centered at the ordinates of A and B which are found to be $\omega_0' \pm \omega_1/\sqrt{2s}$. Since the slopes in A and B, p_A and p_B (equal by symmetry), are not equal to s, these two lines are not Doppler free. One easily shows from (2) that they have a width $(1 - p_A/s)\,\Delta'$ (Doppler width Δ' on the transition ca modified by the light-shifts). The population of level c being not perturbed by the laser, the weights Π_0, Π_A and Π_B of the three

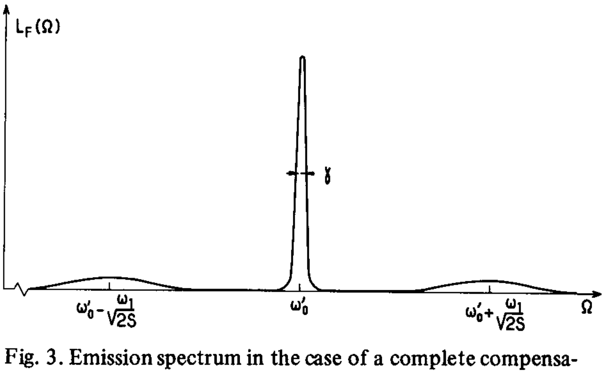

Fig. 3. Emission spectrum in the case of a complete compensation of Doppler broadening. The central narrow line (corresponding to point 0 of fig. 2-α) has a width γ (homogeneous width) and a weight $1-s$. The two sidebands at $\omega_0' \pm \omega_1/\sqrt{2s}$ (corresponding to points A and B of fig. 2-α) have an inhomogeneous width $\Delta'|3/2 - 1/s|$ (Δ': Doppler width of transition ca) and a weight $s/2$.

lines of fig. 3 are just proportional to the transition rates between the corresponding dressed atom energy levels (square of the dipole matrix elements) [12]. From the diagonalization of the dressed atom hamiltonian, one derives simple results such as:

$$p_0 + p_A + p_B = 2 \quad (6)$$

$$\Pi_0 \sim (1 - p_0); \quad \Pi_A \sim (1 - p_A); \quad \Pi_B \sim (1 - p_B).$$

For $p_0 = s$, one gets the results given in fig. 3.

Let us now emphasize some important characteristics of this method of compensation of Doppler broadening.

First, the narrow structure of fig. 3 is not due to a population effect, as in the fluorescence line narrowing (F.L.N.) method [13,14] where one detects population changes induced by the laser beam. Here, the detection signal is proportional to the population of level c which is not perturbed by the laser. Any excitation mechanism can be used for putting atoms in this emitting level c. The perturbed levels a and b could even be empty. The narrowing mechanism is entirely due to an energy perturbation of the final state a. Furthermore, the width of the narrow line of fig. 3 is of the order of the homogeneous width γ whereas, in the same high intensity regime ($\omega_1 \gg \gamma$), a F.L.N. resonance would be considerably power broadened.

Second, the effect we are discussing is highly anisotropic. Coming back to fig. 1-β, suppose that one observes the fluorescence light not in the forward, but in the backward direction. The Doppler emission factor $(1 - v/c)$ of (2) has to be replaced by $(1 + v/c)$. If the v-dependence of the laser induced light shifts balances the forward Doppler shift, it clearly doubles the backward one. It follows that, when condition (5) is fulfilled, the atomic medium emits 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 three level systems [8,14], but here such an asymmetry is particularly important because of the great difference between Δ' and γ.

4. Partial compensation of Doppler broadening

Let us consider now the situation: $\Delta \gg \omega_1 \gg \gamma$. The dressed atom energy levels as well as the transition rates between them have now to be calculated for every abscissa of the frequency diagram of fig. 3. Such a calculation only requires the diagonalization of a 3 X 3 matrix. The corresponding emission spectrum [15] is represented in fig. 4 in the case $\omega_L = \omega_0$; $\omega_1 = 10\gamma$; $\Delta \gg \omega_1$; $s = \frac{1}{2}$; δ is such that condition (5) is fulfilled. Three narrow structures appear which correspond to the three point C, 0 and D of the frequency diagram (fig. 2-α) where the slope of the tangent has the correct value s. The contrast is quite sufficient even though ω_1/γ is only equal to 10. Similar calculations for the backward direction give a simple Doppler line without any structure.

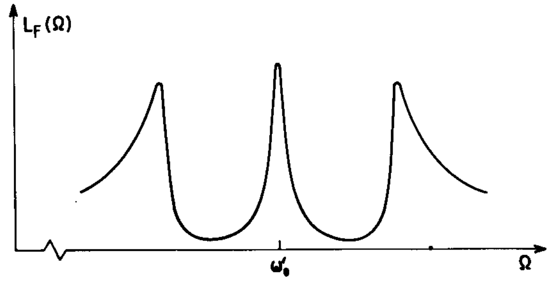

Fig. 4. Calculated spectrum in the case of a partial compensation of Doppler broadening ($\omega_L = \omega_0$, $\omega_1 = 10\gamma$, $\Delta \gg \omega_1$, $s = 1/2$, δ such that $p_0 = s$). The three narrow structures correspond to the three points 0, C, D of fig. 2-α where the slope has the value s.

5. Conclusion

From an experimental point of view, a single mode laser beam is needed with a very good frequency stability and a power as high as possible. With a power of the order of 200 mW, an oscillator strength of the order of 1 and a waist focalization of 0.2 mm, one could achieve values of ω_1 of the order of the Doppler width, leading to a nearly complete compensation of the Doppler broadening. Actually, focussing too much the laser beam decreases drastically the volume of observation and makes difficult the detection of the emission spectrum. A solution to this problem would be to detect the compensation of Doppler broadening not on the emission from c but on the absorption of a weak laser beam probing the transition ac (the previous calculations can be easily extended to such a case by replacing the population of the emitting level c by a difference of populations). A particularly interesting situation occurs when, in the absence of laser, the atomic medium is amplifying on *ca* (for example, levels *a* and *b* empty). Compensation of the inhomogeneous Doppler broadening by the previous method would drastically enhance the amplification in the forward direction. This could have interesting applications: Doppler free directed superradiance, ring lasers, Doppler free coherent transients etc.

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