

## COMPENSATION OF DOPPLER BROADENING BY LIGHT SHIFTS IN TWO PHOTON ABSORPTION

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We show that the residual Doppler broadening of a two photon absorption line — the two photons having different frequencies — can be compensated by velocity dependent light shifts. We report the results of an experiment performed on  $^{20}\text{Ne}$  atoms and giving evidence for this compensation effect.

### 1. Introduction

The possibility of compensating Doppler broadening of spectral lines by velocity dependent light shifts [1–3] has been experimentally demonstrated by analyzing the spectral profile of fluorescence lines emitted by a neon vapour irradiated by a nearly resonant laser beam [4–6]. We present in this letter an experiment showing that light shifts can also be used to compensate the residual Doppler broadening of a two photon absorption line (the two photons having different frequencies).

### 2. The compensation mechanism

The principle of the experiment is schematized on fig. 1. A vapour of three level atoms is irradiated by two counterpropagating laser beams whose frequencies  $\omega_p$  (pump laser) and  $\omega_d$  (detection laser) are respectively nearly resonant with the frequencies  $\omega_0$  and  $\omega'_0$  of the two allowed transitions  $ab$  and  $bc$ . For an atom having the velocity  $v$  along the laser propagation direction, the two laser frequencies are Doppler shifted (fig. 2) so that the two photon absorption resonance occurs for:

$$\omega_p(1 + v/c) + \omega_d(1 - v/c) = \omega_0 + \omega'_0 \quad (1)$$

(second order Doppler shifts are neglected throughout

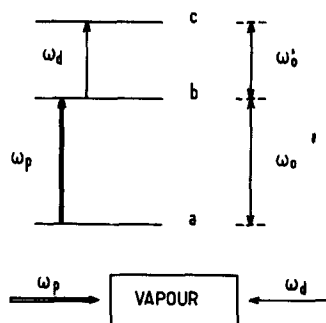


Fig. 1. Energy level scheme. The two counterpropagating pump and detection laser beams have unequal frequencies  $\omega_p$  and  $\omega_d$  close to the atomic frequencies  $\omega_0$  and  $\omega'_0$ .

this paper). When the two laser frequencies are equal, one gets the well known Doppler free two photon absorption [7]. Otherwise, the two photon absorption line remains Doppler broadened (condition (1) is not simultaneously fulfilled for different velocity groups).

Eq. (1) is actually valid only for very low intensity lasers. Otherwise, the laser irradiation produces light shifts [8] of the  $a$  and  $c$  levels which modify the two photon resonance condition [9,10]. We suppose here that the  $c$  level is not shifted (the detection laser remains of weak intensity) so that two photon absorption occurs for:

$$\omega_p(1 + v/c) + \omega_d(1 - v/c) = \omega_0 + \omega'_0 - \epsilon_a(v), \quad (2)$$

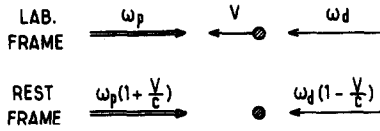


Fig. 2. Apparent frequencies of the pump and detection beams in the laboratory and rest frames.

where  $\epsilon_a$  is the light shift of  $a$ . The important point is that  $\epsilon_a$  depends on the detuning between the apparent pump laser frequency  $\omega_p(1 + v/c)$  and the atomic frequency and is therefore a function of  $v$ . As a consequence, it may be possible to compensate in condition (2) the residual Doppler shift  $(\omega_p - \omega_d)v/c$  by the linear dependence in  $v$  of  $\epsilon_a(v)$ . In such a case, the two photon resonance condition (2) is satisfied for all velocity groups as far as the non linear term in  $v$  of  $\epsilon_a(v)$  remains negligible. The residual Doppler broadening of the two photon absorption line is thus compensated by velocity dependent light shifts.

In order to achieve a linear  $v$  dependence of the light shift,  $\epsilon_a$ , the atomic level configuration sketched in fig. 3 is particularly suitable: the Zeeman sublevels  $b_+$ ,  $b_0$ ,  $b_-$  of  $b$  ( $J = 1$ ) are split by a static magnetic field; the two lasers have a  $\sigma$  polarization so that the levels  $a$  and  $c$  (both  $J = 0$ ) are coupled to  $b_+$  and  $b_-$  ( $b_0$  can be ignored). The pump laser is tuned to the atomic frequency in zero magnetic field ( $\omega_p = \omega_0$ ) so that, for an atom at rest, the apparent pump laser frequency is symmetrically detuned from the two Zeeman components  $\omega_0 \pm \delta$  ( $\delta$  is the Zeeman splitting). The two light shifts of  $a$  associated with the transitions

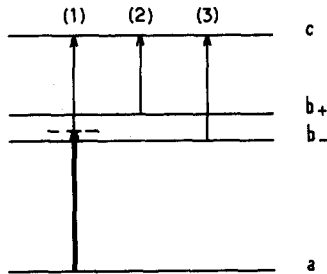


Fig. 3. The two Zeeman sublevels  $b_+$  and  $b_-$  of  $b$  are split by a magnetic field. Two photon absorption process  $a \rightarrow c$  occurs via  $b_+$  or  $b_-$  (1). Level  $c$  can also be excited by the detection beam from  $b_+$  (2) or  $b_-$  (3), the two levels  $b_{\pm}$  being populated by the pump beam (stepwise processes).

$ab_+$  and  $ab_-$  therefore balance ( $\epsilon_a(v=0) = 0$ ). For a moving atom, the two detunings now differ which leads to a velocity dependent shift of  $a$ . From symmetrically considerations, this dependence contains only odd powers in  $v$  so that the range of velocities over which the compensation is effective is limited only by cubic terms (and not by quadratic ones).

Actually, the compensation of Doppler broadening by the linear term of  $\epsilon_a(v)$  requires such a high laser intensity that the previous perturbative analysis is not sufficient. As a matter of fact, the two photon absorption process is mixed with the stepwise processes  $a \rightarrow b_+$  and  $b_+ \rightarrow c$ ,  $a \rightarrow b_-$  and  $b_- \rightarrow c$  (see fig. 3). The dressed atom approach [11,12] provides a simple non perturbative treatment. In such an approach, one studies the absorption of the detection laser, considered as a probe beam, by the compound system "atom plus pump laser photons" (the so called dressed atom). Absorption resonance occurs when the Doppler shifted detection frequency is equal to a Bohr frequency of the dressed atom (these Bohr frequencies taking into account the Doppler shift of the pump frequency; see fig. 2). The effect discussed here thus appears, in the dressed atom approach, as closely related to the compensation of Doppler broadening of fluorescence lines [4-6,12], the only difference being that one probes the dressed atom Bohr frequencies by an absorption study rather than by a fluorescence analysis. One gets in this way the linear term in  $v$  of  $\epsilon_a$ , calculated non perturbatively, i.e. without any restrictions on the value of  $\omega_1/\delta$  where  $\delta$  is the Zeeman splitting in the  $b$  level (fig. 3) and  $\omega_1$  the Rabi frequency characterizing the strength of the coupling of the pump laser field with each of the two transitions  $ab_+$  and  $ab_-$ . Comparing this term with the residual Doppler shift in (2) leads to the following compensation condition:

$$\frac{\omega_1^2}{\omega_1^2 + 2\delta^2} = \frac{\omega_p - \omega_d}{\omega_p} = \frac{\omega_0 - \omega'_0}{\omega_0} \quad (3)$$

If  $\omega'_0$  is smaller than  $\omega_0$ , there exists a value of the ratio  $\omega_1/\delta$  for which this condition is achieved. The compensation effect is effective for atomic velocities such that the cubic term in  $v$  of  $\epsilon_a(v)$  does not exceed the homogeneous width  $\gamma$  which leads to the condition:

$$|v| < v_{\max},$$

with

$$v_{\max} = v_D (\gamma \omega_1^2 / \Delta^3)^{1/3} \quad (4)$$

( $v_D$  is the width of the Maxwell velocity distribution and  $\Delta$  the Doppler width). If  $v_{\max} > v_D$ , all atoms present in the vapour contribute to the absorption (complete compensation of Doppler broadening). Otherwise, the compensation occurs only for a fraction of them (partial compensation).

### 3. Connection with previous theoretical works

The possibility of observing Doppler free structures by a high intensity mixing between two photon and stepwise processes in three level systems has been first mentioned by Popova et al. [13]. The connection between velocity dependent light shifts and narrow structures appearing in laser spectroscopy of three level systems has been discussed by Toschek [14]. Finally, Liao and Bjorkholm have suggested the possibility of line narrowing by light shifts in two photon absorption [9].

### 4. The experimental set-up

The atomic levels  $a$ ,  $b$ ,  $c$  are respectively the levels  $1s_3$  ( $J = 0$ ),  $2p_2$  ( $J = 1$ ),  $2s_3$  ( $J = 0$ ) of Ne (Paschen notations). The pump laser is a single mode dye laser (CR 599 pumped by a SP 171 argon laser) tuned on the  $ab$  transition ( $\lambda_0 = 2\pi c/\omega_0 = 616.3$  nm). The detection laser is a home made He-Ne laser. The active element is a dc discharge cell (length 1 m, internal diameter 3 mm, current 7 mA) containing a mixture of natural helium and neon (total pressure 3 to 6 Torr, Ne proportion 15%). Single mode operation at the wavelength of the  $bc$  transition ( $\lambda'_0 = 2\pi c/\omega'_0 = 1.198$   $\mu$ m) is obtained by inserting, inside the near confocal cavity, a prism which prevents the easier oscillation at 1.152  $\mu$ m and a second dc discharge cell (length 0.4 m, internal diameter 4 mm, current 15 to 30 mA) containing natural neon (pressure 0.6 to 1 Torr) and behaving as a saturable absorber [15]. This laser has an output power of 0.3 mW (with an output mirror transmission equal to 1%) so that it can actually be considered as a probe laser (no light shift of the  $c$  level). The scan range of this laser is limited to 150 MHz.

The two counterpropagating lasers are focused onto a Ne dc discharge cell (length 4 cm, internal diameter 3 mm, pressure 0.3 Torr). The detection laser spot size must be smaller than the pump one; the atoms which are in the detection volume associated with the probe laser must indeed experience the same pump laser electric field, so that they undergo the same light shifts. On the other hand, the velocity range over which compensation is effective becomes wider when the pump spot size is decreased (in eq. (4),  $\omega_1$  and therefore  $v_{\max}$  increase). These considerations lead to choose the minimum value for the detection spot size, i.e. the value such that the diffraction length of the detection beam is equal to the length of the Ne discharge ( $l = 4$  cm gives a detection beam waist  $w_d = 90$   $\mu$ m). The pump beam waist is then chosen sufficiently large ( $w_p = 330$   $\mu$ m) so that the spatial non uniformity of the pump electric field does not broaden too much the compensation peak. The value thus obtained for the Rabi frequency ( $\omega_1 \approx 250$  MHz for a dye laser output power of the order of 150 mW) is much greater than the homogeneous width ( $\gamma \approx 20$  MHz) but much smaller than the Doppler width. As a consequence, one can get only a partial compensation of Doppler broadening (see eq. (4)).

The Ne cell is placed inside a solenoid which produces a static magnetic field parallel to the laser propagation axis. The compensation condition (3) is achieved by varying the magnetic field (and therefore the Zeeman splitting  $\delta$ ). The polarizations of the two lasers are both linear  $\sigma$  polarizations. They can be either parallel ( $\sigma_{\parallel}$  configuration) or perpendicular ( $\sigma_{\perp}$ ).

The absorption signal, measured by a modulation technique, is the difference between the absorption of the detection beam in presence or in absence of the pump one. This signal is monitored versus either the pump or the detection laser frequency.

### 5. Discussion of the results

The spectra, represented in fig. 4, give the variation of the absorption signal when the pump laser frequency is scanned, for various magnetic fields and for the two polarization configurations. In a zero magnetic field, one gets the well known optical Autler-Townes

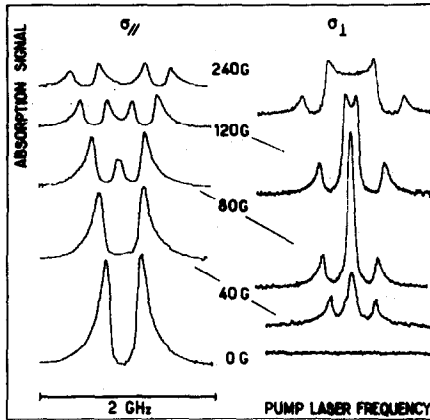


Fig. 4. Experimental absorption signal plotted versus the pump laser frequency for various magnetic fields and in the two polarization configurations. The detection laser frequency is fixed to  $\omega_d = \omega'_0$ . The compensation peak (80 G in the  $\sigma_\perp$  polarization) is centered at  $\omega_p = \omega_0$ .

doublet [16–18] in the  $\sigma_\parallel$  configuration. There is no signal in the  $\sigma_\perp$  configuration since the coherent superposition of  $b_+$  and  $b_-$  coupled to  $c$  is thus orthogonal to the one coupled to  $a$ . When the magnetic field is applied, a narrow peak appears in the  $\sigma_\perp$  configuration, reaches a maximum when the compensation condition is achieved (around  $B = 80$  Gauss) and then broadens. The arrow peak observed for  $B = 80$  Gauss gives an experimental evidence for the possibility of compensating the residual broadening of a two photon absorption line by velocity dependent light shifts.

The weakness of the structure appearing in the  $\sigma_\parallel$  configuration can be related to the following perturbative feature (see fig. 3): when the splitting of  $b_+$  and  $b_-$  is much larger than the homogeneous width, the interference between the two paths contributing to the two photon absorption (from  $a$  to  $c$  via  $b_+$  or  $b_-$ ) is constructive in the  $\sigma_\perp$  configuration and destructive in the  $\sigma_\parallel$  one.

On fig. 5 is represented the variation of the signal in the  $\sigma_\perp$  configuration when the detection laser frequency is scanned through the narrow structure (pump laser frequency fixed to  $\omega_p = \omega_0$ ). In zero magnetic field, the absorption signal is flat. When the magnetic field is adjusted to the compensation condition (here  $B = 70$  G), a narrow absorption peak appears (width 20 MHz). The scan range of the He-Ne

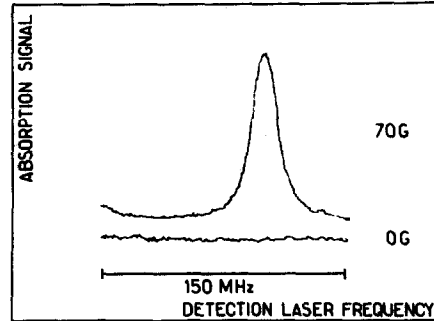


Fig. 5. Experimental absorption signal plotted versus the detection laser frequency for  $B = 0$  G and  $B = 70$  G in the  $\sigma_\perp$  configuration. The pump laser frequency is fixed to  $\omega_p = \omega_0$ . The compensation peak is centered at  $\omega_d = \omega'_0$ .

laser (150 MHz) does not allow to explore the whole absorption spectrum.

We have also studied the absorption signal on the  $2p_2-2s_2$  transition when the  $1s_3-2p_2$  transition is saturated. The detection laser ( $\lambda_0 = 1.177 \mu\text{m}$ ) is now coupled to a  $J = 1$  to  $J = 1$  transition (the pump transition remains the same as above). One thus observes that the roles of the  $\sigma_\parallel$  and  $\sigma_\perp$  configurations are exchanged since the symmetry of the Clebsch–Gordan coefficients is modified.

## 6. More complex atomic configurations

The compensation effect is not limited to the simple atomic configuration of fig. 3. This is demonstrated by the spectra of fig. 6 which corresponds to the more complex configuration  $a = 1s_4$  ( $J = 1$ );  $b = 2p_3$

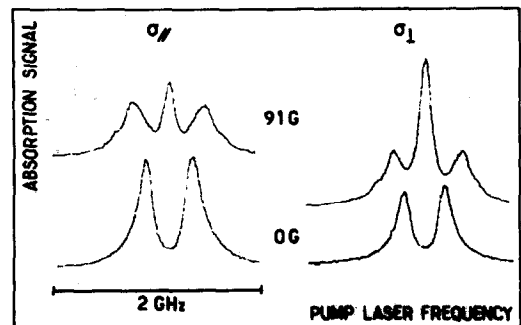


Fig. 6. Same experiment as in fig. 4 but for a more complex level configuration:  $J_a = 1$ ,  $J_b = 2$ ,  $J_c = 1$ .

( $J = 2$ );  $c = 2s_2$  ( $J = 1$ ) (pump laser wavelength  $\lambda_0 = 609.6$  nm; detection laser wavelength  $\lambda'_0 = 1.152$   $\mu\text{m}$ ). The optical Autler–Townes doublet in zero magnetic field now appears in the two polarization configurations. When the magnetic field is adjusted to the compensation condition, one gets a Doppler free structure more intense in the  $\sigma_\perp$  configuration than in the  $\sigma_\parallel$  one.

## 7. Comparison with previous experiments

We have yet emphasized the close connection between this experiment and the compensation of Doppler broadening of fluorescence lines by light shifts [4–6]. An absorption probing presents two main experimental advantages when compared to a fluorescence analysis. First, the spectral resolution is now determined by the detection laser (jitter 1 MHz) instead of the Fabry–Perot interferometer (bandwidth 15 MHz). Then, the two photon absorption signal is related to the population of the metastable  $a$  level. Since this population is greater, and varies more slowly, than populations of the more excited levels contributing to fluorescence signals [4], it has been possible to reduce the Ne pressure in the discharge cell to 0.3 Torr (1.5 Torr for fluorescence analysis). These two improvements pull the width of the compensation peak down to 20 MHz (from 60 MHz for fluorescence analysis; see ref. [4]).

To conclude this comparison, we can remark that the populations contributing to the absorption signal are modified by the pump laser irradiation, contrary to what occurs in fluorescence experiments. These population effects are superposed upon the effect of the light shifts described above. But it must be emphasized that the appearance of narrow structures is still entirely due to the compensation effect. In particular, there is no power broadening of the narrow peak. This clearly distinguishes the present work from previous experiments, such as absorption line narrowing, where narrowing results from a velocity selection produced by a “hole burning” type effect. [19–22]. This difference has an interesting consequence: the forward backward anisotropy here takes considerable values (of the order of  $\Delta/\gamma$ ) since, when light shifts compensate the Doppler broadening for one direction, they double it in the opposite one. We

have verified that no Doppler free structures are observed when the probe beam has the same direction as the pump one.

## 8. Conclusion

We have observed the compensation of Doppler broadening of a two photon absorption line by velocity dependent light shifts.

This compensation effect can have interesting applications, specially when the detection laser is coupled to an amplifying atomic transition. It would lead, in this case, to a unidirectional enhancement of the gain, controlled by the pump laser. It could, for example, be used to get a reduction of threshold in laser media or a unidirectional operation of a ring laser. Recently, such a compensation effect has been applied to the unidirectional triggering of a superradiance signal [23,24].

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