We are glad to acknowledge the contribution of B. Preedom for making the programs FITPI and DWPI available at SIN and for lending us the <sup>90</sup>Zr target, and we thank G. Miller for enlightening discussions.

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## Experimental Evidence for Compensation of Doppler Broadening by Light Shifts

S. Reynaud, M. Himbert, J. Dupont-Roc, H. H. Stroke, (a) and C. Cohen-Tannoudji Ecole Normale Superieure and Collège de France, 75231 Paris Cedex 05, France (Received 7 December 1978)

Velocity-dependent light shifts may be used to suppress the Doppler broadening of an atomic spectral line observed along a given direction. We present emission spectra of <sup>20</sup>Ne demonstrating the existence of such an effect for an appreciable portion of the atoms. Various possible applications taking advantage of the high anisotropy of this effect are suggested.

The possibility of compensating Doppler broadening of spectral lines by velocity-dependent light shifts<sup>1-3</sup> is illustrated with the following example of a three-level system a-b-c [Fig. 1(a)]. Suppose that one analyzes the spectral profile of the light emitted by atoms excited in level c (for example, by a discharge) around the frequency  $\omega_0$ of the transition c-a. Simultaneously, the atomic vapor is irradiated by an intense single-mode laser beam, with frequency  $\omega_L$  close to the frequency  $\omega_0$  of the transition b-a. For an atom moving with velocity v in the laboratory frame [Fig. 1(b)], the laser frequency in its rest frame [Fig. 1(c)] is Doppler shifted from  $\omega_L$  to  $\tilde{\omega}_L$  $=\omega_L(1+v/c)$ . The laser radiation perturbs the energy levels of such an atom, inducing "light shifts"4 which depend not only on the laser intensity, but also on the detuning of the apparent laser frequency  $\tilde{\omega}_L$ , seen by the atom in its rest frame, from the atomic frequency. Since  $\tilde{\omega}_L$  is

v dependent, it follows that the light shifts, and consequently the frequency  $\tilde{\Omega}$  emitted by the atom in its rest frame [Fig. 1(c)], are also v dependent. Coming back to the laboratory frame [Fig. 1(b)], we get for the frequency  $\Omega_{\rm fw}$  of the light emitted in the forward direction  $\Omega_{\rm fw}(v) = \tilde{\Omega}(v)(1-v/c)$ . One can then try to choose the experimental parameters in such a way that the v dependence of  $\tilde{\Omega}(v)$  compensates the emission Doppler factor.

The most interesting feature of this effect is its high anisotropy. An observation of the light emitted in the backward direction would lead to  $\Omega_{\rm bw}(v) = \tilde{\Omega}(v)(1+v/c)$ . If the v dependence of  $\tilde{\Omega}(v)$  compensates the emission Doppler shift in the forward direction, it doubles it in the backward one. It should also be emphasized that the emitting level c is not coupled to the laser ( $\omega_L$  is not in resonance with  $\omega_0'$ ) and that the perturbed levels a and b could even be empty. The narrowing

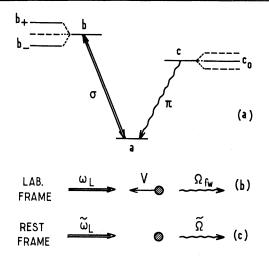


FIG. 1. (a) Energy level scheme for Doppler-broadening compensation by light shifts. The splittings in levels b and c are produced by a magnetic field b. A  $\sigma$ -polarized laser excites the a-b<sub>+</sub> and a-b<sub>-</sub> transitions. One detects the  $\pi$ -polarized emission from c<sub>0</sub> to a. (b) Laser frequency  $\omega_L$  and forward emission frequency  $\Omega_{\rm fw}$  in the laboratory frame. v is the velocity of the atom. (c) The same frequencies seen in the atom rest frame.

mechanism is not due to a population effect but to a velocity-dependent shift of the final state a. An important consequence is the absence of power broadening of the narrow line. Other mechanisms for producing velocity-dependent internal frequencies, which could lead to a compensation of the Doppler broadening, have been suggested recently. They use quadratic Stark shifts in crossed static electric and magnetic fields or motional interaction of spins with static electric fields in polar crystals.

In the experiment described here, the atomic levels a, b, and c are, respectively, the levels  $1s_3$  (J=0),  $2p_2$ , and  $2p_{10}$  (J=1) of <sup>20</sup>Ne (Paschen notation). A static magnetic field produces Zeeman splittings  $\delta$  and  $\delta'$  in b and c, and the  $\pi$ -polarized emission from  $c_0$  to a is detected. The laser light is  $\sigma$  polarized, coupling a to  $b_+$  and b. with a strength characterized by the Rabi nutation frequency  $\omega_1 = \mathfrak{D}\mathcal{E}_L$  ( $\mathfrak{D}$  dipole moment of  $a-b_+$ and a-b-;  $\mathcal{E}_L$  laser electric field). The laser frequency is tuned to  $\omega_L = \omega_0$  so that, for an atom at rest (v=0), the detuning of the apparent laser frequency  $\tilde{\omega}_L$  from the frequencies  $\omega_0 \pm \delta$  of the two Zeeman components is equal and opposite; the two light shifts of a associated with the transitions  $a-b_+$  and  $a-b_-$  therefore balance. For a moving atom, the two detunings now differ, leading to a velocity-dependent net shift of a. From symmetry considerations, this dependence only contains odd powers of v. The compensation of the Doppler shift by the linear term of  $\tilde{\Omega}(v)$  requires laser intensities so high that the previous perturbative analysis is not sufficient. A nonperturbative treatment<sup>2</sup>, (in  $\omega_1/\delta$ ) using a dressedatom approach and frequency diagrams provides the compensation condition

$$\omega_1^2 / (\omega_1^2 + 2\delta^2) = \omega_0' / \omega_{0^*}$$
 (1)

The range of velocities over which the emission Doppler shift is compensated (within the homogeneous width  $\gamma$ ) is limited by the cubic term of  $\tilde{\Omega}(v)$  to  $|v| < v_{\max}$ , where  $v_{\max} = v_D (\gamma \omega_1^2/\Delta^3)^{1/3} \left[ v_D \right]$  is the width of the Maxwell velocity distribution and  $\Delta$  the Doppler width. If  $v_{\max} > v_D$ , all atoms will emit at the same frequency (complete compensation of Doppler broadening). If  $v_{\max} < v_D$ , the compensation will occur only for a fraction of them (partial compensation): A part of the emission Doppler profile is concentrated in a narrow peak.

The experimental setup is simple. The beam of a single mode dye laser<sup>10</sup> operating at 6163 Å is focused onto a <sup>20</sup>Ne cell (length 10 cm, internal diameter 3 mm, pressure 1.5 Torr, dc discharge current 16 mA). The power at the entrance of the cell is 140 mW, the waist of the beam 0.4 mm (which leads to  $\omega_1 \sim 250$  MHz). Two coils produce an homogeneous static field perpendicular to the light beam. The spectral profile of the light emitted at 7439 Å in the forward direction by a volume of the discharge (length 10 cm, diameter 0.4 mm) within the irradiation volume is analyzed by a piezoelectrically scanned confocal Fabry-Perot interferometer (length 10 cm, finesse 50). The detection device comprises a low-noise photomultiplier<sup>11</sup> and standard photon-counting electronics. Filters eliminate the laser light. The photocounting rate is 104 per second which corresponds to about 108 atoms per cubic centimeter excited in the level c by the discharge.

The experimental curves, represented on the left-hand side of Fig. 2, give the recorded emission spectral profile for increasing values of the static magnetic field B. They are in good agreement with the corresponding computed curves, <sup>12</sup> represented on the right-hand side of Fig. 2. For zero magnetic field, one gets the well-known Autler-Townes doublet which has been recently extensively studied in the optical range either with atomic beams <sup>13</sup> or in vapors. <sup>14</sup> When the magnetic field is increased, a narrow structure

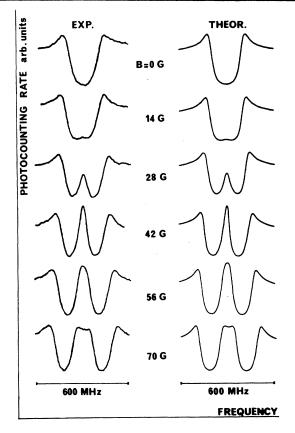


FIG. 2. Experimental and theoretical emission spectral profiles for the c-a transition ( $^{20}$ Ne,  $\lambda = 7439$  Å), for a fixed laser intensity and various magnetic fields B. The compensation of the Doppler effect occurs for B=42 G. The Doppler width is 1100 MHz.

appears in the center of the spectrum, reaches a maximum (around B=42 G), and then broadens. For the optimum value (B=42 G),  $\omega_1$  and  $\delta$  satisfy the compensation condition (1), and one gets the Doppler-free line  $c_0 - a$  discussed above, exhibiting a homogeneous width (50 MHz). The Doppler width is 1100 MHz.

Since, in our experiment, the Rabi frequency  $\omega_1$  (250 MHz) is smaller than the Doppler width, one gets only a partial compensation of the Doppler broadening. By using more intense and more focused laser beams, and eventually by putting the cell inside a ring cavity, one could achieve nearly complete compensation of Doppler broadening  $(\omega_1/\Delta \sim 10)$ . In such a situation, one would have a very interesting medium. First, all atoms contribute to the central narrow line (homogeneous width), well separated from the two sidebands which can be interpreted as caused by inverse Raman processes and which remain Doppler broadened. Second, the peak of the

narrow line can be much higher than that of the original Doppler line so that the absorption or the amplification of a weak probe beam can be considerably enhanced. Finally, the forward-backward asymmetry for such a probe beam becomes spectacular (of the order of  $\Delta/\gamma$ ). This opens the way to various interesting applications: reduction of the threshold for laser media, ring laser, directed Doppler-free superradiance, Doppler-free coherent transients, and enhancement of Faraday rotation in one direction.

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 $^{16}$  This homogeneous width is due to spontaneous emission (10 MHz) and collisions (~30 MHz) and is increased by the Fabry-Perot spectral width (~15 MHz).

<sup>17</sup>One can show that about 50% of the atoms contribute to the central narrow line, and each of these contributes 20% of its total emission rate to the central peak.

## Excitation of Atomic Hydrogen to the n = 2 State by Helium Ions

## Victor Franco

Physics Department, Brooklyn College of the City University of New York, Brooklyn, New York 11210 (Received 8 January 1979)

Differential and total cross sections for  $\operatorname{He}^+(1s)+\operatorname{H}(1s)\to\operatorname{He}^+(1s)+\operatorname{H}(n=2)$  are calculated using a formalism developed for ion-atom and atom-atom collisions. Results are obtained in terms of amplitudes for electron-hydrogen (eH) and proton-hydrogen (pH) n=2 excitations and elastic scattering of electrons by a (p+e)-"atom," the  $\operatorname{He}^+(1s)$  form factor, and the n=2 form factors of hydrogen. Agreement with recent data is good. A remarkably simple approximation,  $\sigma \approx \sigma_{\rm pH}(1s\to n=2)+3\sigma_{\rm pH}(1s\to 2s)$ , is obtained and agrees with recent data.

One of the simplest and most basic of the collisions involving two composite atomic systems is that between helium ions (He<sup>+</sup>) and hydrogen atoms (H). The wave functions for these systems are known exactly. Consequently predictions of cross sections involving these systems depend solely on the scattering theory. Measurements of such cross sections thus become stringent tests of the theory.

I present here the first application of a theory developed for describing collisions between arbitrary ions or atoms, by calculating both differential and total cross sections for the excitation process

$$He^{+}(1s) + H(1s) - He^{+}(1s) + H(n = 2).$$
 (1)

I restrict the calculation to medium and high incident energies ( $v \ge 0.5$  a.u.). For these energies

the four-state impact-paremeter method<sup>2</sup> yields a rather marked improvement in the cross sections for process (1) over those obtained from the Born approximation.<sup>3</sup> Nevertheless the results of Ref. 2 have been recently reported<sup>4</sup> to be in disagreement with the total-cross-section measurements and with the first differential-cross-section measurements of (1).<sup>4</sup> To my knowledge there are no previously published calculations which yield agreement with these measurements. I also obtain a remarkably simple approximate formula for the cross section for (1), which agrees extraordinarily well with recent measurements.<sup>4</sup>

The amplitude for arbitrary ion-atom or atomatom collisions in which the target (T) undergoes a transition from initial state  $i_T$  to final state  $f_T$  and the projectile (P) undergoes a transition from state  $i_P$  to state  $f_P$  is given approximately by  $i_P$ 

$$F_{fi}(\vec{\mathbf{q}},k) = k \left[ \delta_{f_P i_P} (Z_P - \frac{1}{2}N) M f_{pH,fi}^T (\vec{\mathbf{q}},k_p) + \delta_{f_T i_T} (Z_T - \frac{1}{2}M) N f_{pH,fi}^P (\vec{\mathbf{q}},k_p) \right] / k_p$$

$$+ \frac{1}{2} M N k \left[ S_{fi}^P (-\vec{\mathbf{q}}) f_{eH,fi}^T (\vec{\mathbf{q}},k_e) + S_{fi}^T (\vec{\mathbf{q}}) f_{eH,fi}^P (\vec{\mathbf{q}},k_e) \right] / k_e,$$
(2)

where  $\hbar k$  is the incident momentum (with corresponding relative velocity v) and  $\hbar \bar{\mathbf{q}}$  is the momentum transferred. Here  $Z_P$  and  $Z_T$  are the atomic numbers of the projectile and target, and N and M are the number of electrons in the projectile and target, respectively. The function  $S_{fi}^{P}(\bar{\mathbf{q}})$  is the transition form factor for the projectile,

$$S_{fi}{}^{P}(\vec{\mathbf{q}}) \equiv \int e^{i\vec{\mathbf{q}} \cdot \vec{\mathbf{r}}} \rho_{fi}{}^{P}(\vec{\mathbf{r}}) d^{3}r, \tag{3}$$