# Light-Pressure-Induced Line-Shape Asymmetry of the Saturation Dip in an Atomic Gas

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We report on the observation of a light-pressure-induced asymmetry of the well-known adsorption dip occurring in optical saturation spectroscopy. This asymmetry is due to the small modification of the atomic velocity distribution that is caused by the spontaneous scattering force of the saturating light field. Our measurements were performed on the $\lambda = 555.65$ nm $^1S_0$-$^3P_1$ transition in atomic ytterbium vapor by the use of a frequency-modulated test field propagating collinearly with the saturating field. The experimental results clearly confirm theoretical predictions and demonstrate the significance of the phenomenon for high-resolution laser spectroscopy.

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The study of resonant light forces on the motion of free atoms is of strong current interest.[1] So far, most of the experiments on light forces have been performed with the use of atomic beams; it has been shown that atoms can be strongly deflected, decelerated, and even trapped by laser light. On the other hand, it seems obvious that the photon momentum that is transferred to the resonant atoms in a *gas* also leads to a redistribution of the atomic velocities.[2] In this context, recent investigations[3,4] showed that the well-known spontaneous light pressure can have substantial effects on the *optical response* of an atomic gas to a laser field, even if the modification of the total atomic velocity distribution seems to be insignificantly small. For a single beam interacting with a Doppler-broadened medium, the occurrence of a strong nonlinear contribution to the dispersion of the light field was observed experimentally.[4] In contrast to this dispersion effect, light-pressure-induced modifications of the absorption of a light field in a gas can only play a significant role when two or more laser beams are involved in an experiment.[3]

As an important example, a light-pressure-induced asymmetry of the well-known Doppler-free saturation dip occurring in saturated absorption spectroscopy was predicted by Kazantsev, Surdutovich, and Yakovlev.[3] Surprisingly, this phenomenon has not yet been studied experimentally, although saturation spectroscopy represents a basic and widely explored technique in high-resolution optical spectroscopy. Here, we report on the first experiments that clearly demonstrate the influence of spontaneous light pressure on the line shape of the saturation dip in an atomic gas. The observed line-shape asymmetry is of fundamental significance for high-precision measurements using absorption cells.

Let us start with a simple qualitative explanation of the phenomenon. As is well known, saturation spectroscopy is based on the velocity-selective excitation of a certain velocity subgroup in an atomic or molecular gas by a relatively strong optical pump field.[5] The induced hole occurring at the resonant velocity $v_0$ in the population difference of ground and excited states, known as the Bennett hole, is probed by a weak tunable test field: As a consequence, a corresponding saturation dip occurs in the absorption curve of the test field, displaying a minimum width given by the natural linewidth of the optical transition. Usually, this saturation dip is considered as being completely attributed to a perturbation of the internal atomic degrees of freedom, namely the saturation of the optical transition. But, in addition to this, the external atomic degrees of freedom can also experience a perturbation: the redistribution of atomic velocities caused by the spontaneous scattering force of the pump field [see Fig. 1(a)]. The corresponding distortion of the atomic velocity distribution $N(v)$ leads to a modification of the Bennett hole [see Fig. 1(b)] in the population difference of ground and excited states, $n_g(v) - n_e(v) = N(v)[1-2p(v)]$; here $p(v)$ denotes the steady-state excitation probability. Thus, obviously, modifications of the saturation dip in the absorption profile of the test field can also occur as a result of resonant light pressure.

Let us now present the results of our theoretical approach in a more quantitative way: We consider the in-

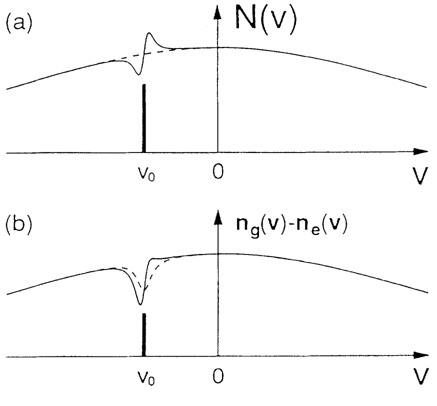

FIG. 1. (a) Typical velocity distribution $N(v)$ of the number density and (b) corresponding velocity distribution $n_g(v) - n_e(v)$ of the population difference of an ensemble of two-level atoms interacting with a monochromatic laser field at velocity $v_0$. Both curves are shown with (solid lines) and without (dashed lines) modifications due to resonant light pressure.

teraction of an atomic vapor with two monochromatic plane laser fields, a strong pump field with frequency $\omega_0$ and wave number $k_0$ and a weak test field with $\omega_1$ and $k_1$. The medium is assumed to consist of two-level atoms with transition frequency $\Omega$, mass $M$, and a Doppler distribution $N_0(v)$, where $v$ denotes the atomic velocity component in the propagation direction of the test field. We calculate the absorption coefficient of the test field under the following conditions: The pump field intensity $I_0$ is assumed to be small compared with the saturation intensity $I_s$ of the transition ($I_0 \ll I_s$); in this case, the total effect of the pump field on the medium can be treated as a perturbation in first order of the saturation parameter $G = I_0/I_s$. The test field with intensity $I_1$ is considered sufficiently weak that it leads to no significant saturation effects. For the optical transition, spontaneous emission is regarded as the dominating relaxation mechanism with a corresponding natural linewidth $\Gamma$ (HWHM). The medium is considered in the limit of a large Doppler width $\sigma$ ($\sigma \gg \Gamma$). The effect of light pressure is treated in the quasiclassical limit by introducing the spontaneous scattering force; this approach is valid if the Doppler shift $2\epsilon_r = \hbar k_0^2/M$ of an atom due to one-photon momentum transfer is small compared with the natural linewidth ($\epsilon_r \ll \Gamma$). Under these assumptions, the absorption coefficient $\alpha(\omega_1)$ of the test beam interacting with the optically thin sample can be written in the form

$$\alpha(\omega_1) = \alpha_0(\omega_1) - \alpha_0(\omega_0) G X_\pm \,, \tag{1}$$

with

$$X_- = \frac{1}{2}\frac{1}{\delta_-^2+1} + \epsilon_r \tau \frac{\delta_-}{(\delta_-^2+1)^2} \,, \tag{2a}$$

$$X_+ = \frac{1}{(\delta_+^2+1)^2} - \epsilon_r \tau \frac{\delta_+}{(\delta_+^2+1)^2} \,, \tag{2b}$$

and

$$\delta_- = (\omega_1 + \omega_0 - 2\Omega)/2\Gamma \,, \tag{3a}$$

$$\delta_+ = (\omega_1 - \omega_0)/2\Gamma \,. \tag{3b}$$

The first term in Eq. (1) is the ordinary Doppler-broadened absorption profile $\alpha_0(\omega_1) \propto N_0((\omega_1 - \Omega)/k_1)$. The saturation dip is described by the second term in Eq. (1); its shape is given by $X_-$ and $X_+$ as functions of the dimensionless detuning parameters $\delta_-$ and $\delta_+$ defined in Eq. (3). $X_-$ is valid for the case of a test field propagating counter to the pump field, and $X_+$ describes the situation of copropagating fields.

The first terms in Eqs. (2a) and (2b) are due to the ordinary saturation of the transition, and thus represent the signals usually considered in saturation spectroscopy. Their different shape for counterpropagating and copropagating fields is due to a different effect of coherence phenomena of the four-wave mixing type.[6] The second terms in Eqs. (2a) and (2b) appear as a result of the modification of the atomic velocity distribution that is caused by the resonant light pressure of the pump beam. Most importantly, the light-pressure-induced contributions display an *odd symmetry*, which stands in contrast to the *even symmetry* of the ordinary saturation contributions [first terms in Eqs. (2a) and (2b)]. As a consequence, the total Doppler-free signal becomes *asymmetric.* The relative strength of the light-pressure-induced contribution is determined by the dimensionless parameter $\epsilon_r\tau$; here $\tau$ denotes an effective time where a free interaction of the atoms with the light field takes place. Our result is valid if the modification of the velocity distribution remains weak [$|N(v) - N_0(v)| \ll N_0(v_0)$]; this condition is equivalent to the restriction that the total Doppler shift $2G\epsilon_r\tau\Gamma$ a resonant atom experiences has to be small compared with the linewidth $\Gamma$ ($2G\epsilon_r\tau \ll 1$).

Representative plots of the line-shape functions $X_-$ and $X_+$ for $\epsilon_r\tau = 1$ are shown in Figs. 2(a) and 2(b), respectively. In both cases, the light-pressure-induced contributions (dashed lines) lead to a pronounced asymmetry and shift of the center of the total Doppler-free resonances (solid lines). In the case depicted in Fig. 2(a), the maximum transmission of the test beam occurs for $\delta_- \approx 0.5$; this corresponds to a frequency shift of approximately 25% of the full width of the resonance.

The quantity $\tau$, which we introduced as an effective interaction time, deserves some discussion: For a low-pressure gas in the linear regime of a low laser intensity ($I_0, I_1 \ll I_s$) and a weak light-pressure-induced modification of the velocity distribution as discussed above, $\tau$ is determined by the transit time of the atoms through the laser beam of diameter $d$ by $\tau = d\pi k_0/4\sigma$: Here both the distribution of different transit times and the variation of the laser intensity due to the Gaussian beam profile are

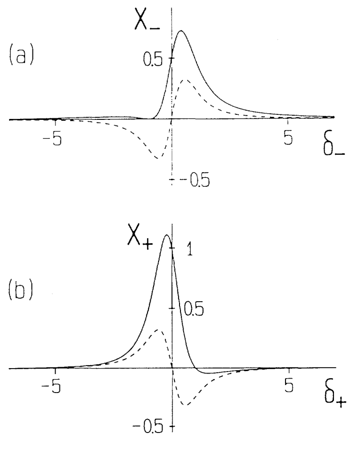

FIG. 2. Representative plots of the functions $X_-$ and $X_+$ describing the line shapes of the saturation dip for (a) counterpropagating and (b) copropagating pump and test fields, respectively; here $\epsilon_r\tau = 1$ is assumed. The light-pressure-induced signal contributions are shown by the dashed curves.

properly taken into account in $\tau$. As a consequence, $\tau$ and the strength of the light-pressure-induced phenomenon reported here depend linearly on the laser beam diameter.[3]

Our experiment to observe the light-pressure-induced asymmetry of the saturation dip is based on frequency-modulated saturation spectroscopy[7] [see Fig. 3(a)]. In this technique, one weak modulation sideband (frequency $\omega_C - \omega_M$ or $\omega_C + \omega_M$) of a frequency-modulated test beam (carrier frequency $\omega_C$) can be used as a probe field for the saturation of the medium caused by the pump field. Absorption and dispersion of this test sideband give rise to different amplitude-modulation components in the transmitted test beam, which can be detected easily with high sensitivity and a good signal-to-noise ratio. Here the modulation component ($\omega_M$) that oscillates in quadrature to the applied phase modulation directly reflects the absorption of the test field. Under appropriate conditions, the corresponding modulation amplitude $q$ can be described by

$$q \propto \omega_M \alpha_0'(\omega_C) + \tfrac{1}{2}\, \alpha_0(\omega_0) G X_{\pm} \; ; \qquad (4)$$

here it is assumed that the lower sideband acts as a test field with frequency $\omega_1 = \omega_C - \omega_M$. The first term in Eq. (4) displays the frequency derivative $\alpha_0'$ of the Doppler-broadened absorption profile as a result of the different absorption of the sidebands in the slope of the Doppler profile. The second term of Eq. (4) directly shows the narrow saturation dip.

Our measurements were performed on the line $\lambda = 555.65$ nm ($4f^{14}6s^2\,{}^1S_0$–$4f^{14}6s6p\,{}^3P_1$) of isotopic pure ytterbium with mass number 172. The Yb vapor was contained in a heated ceramic tube of 1.8 cm diam; the temperature of the vapor cell was about 800 K with the length of the heated zone being approximately 6 cm. The total Yb number density was estimated to be $10^{11}$ cm$^{-3}$ with a corresponding vapor pressure of roughly $10^{-5}$ mbar. The natural linewidth of the transition is $\Gamma = 2\pi \times 95$ kHz;[4] its Doppler width amounts to $\sigma = 2\pi \times 510$ MHz. We note that this ${}^{172}$Yb transition represents a *closed two-level system* as considered in our theoretical approach.

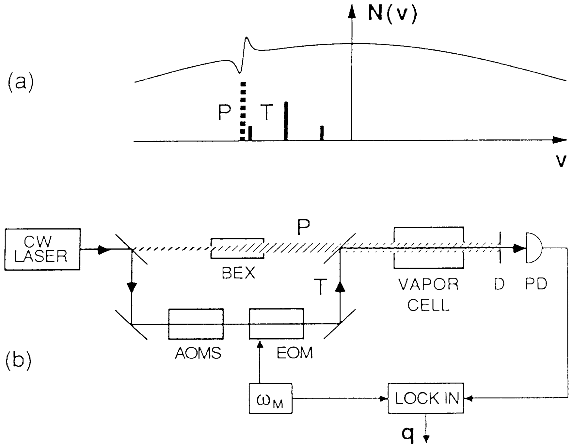

FIG. 3. (a) Scheme of pump beam (P) and frequency-modulated test beam (T) interacting with the Doppler-broadened medium. (b) Experimental scheme: P, pump beam; BEX, beam expander; T, test beam; AOMS, acousto-optic modulation system; EOM, electro-optical modulator; D, diaphragm to block pump beam; PD, photodetector.

In our experiment, to demonstrate the light-pressure-induced asymmetry of the saturation dip we used copropagating laser beams;[8] the experimental scheme is shown in Fig. 3(b). The light of a continuous-wave single-mode dye ring laser was tuned close to the center of the ytterbium line. The beam was divided into pump and test beams. The test beam was directed through an acousto-optical modulation system, which generated a variable frequency upshift in the range of 5–15 MHz. Thereafter, the test beam was modulated by an electro-optical modulator with a fixed modulation frequency $\omega_M \approx 2\pi \times 10$ MHz and a modulation depth of $\sim \frac{1}{5}$. This total modulation scheme allowed the generation of a frequency-modulation sideband of the test beam with a variable frequency shift of $-5$ to $+5$ MHz with respect to the pump field. In front of the vapor cell, the test beam and expanded pump beam were recombined and directed through the Yb vapor. The beams were carefully collimated and aligned in order to avoid possible line-shape asymmetries due to the curvature of the wavefronts.[9] In the sample, the $1/e^2$ diameter of the pump beam was $d \approx 1.5$ cm; for this value, the effective interaction time $\tau$ is about 40 $\mu$s, corresponding to $\epsilon_r \tau \approx 1$. The $1/e^2$ diameter of the test beam was $\sim 1.5$ mm. Low laser intensities ($I_0, I_1 \ll I_s = 150\ \mu$W/cm$^2$) were used for both pump and test fields and the maximum small-signal absorption of the laser light in the optically thin medium was less than 10%; thus, in our experiment, propagation effects within the sample (beam focusing, etc.) cannot be of importance in the signal formation.

A typical measured saturation signal is shown by the solid curve in Fig. 4: The observed line shape clearly displays an asymmetry in good qualitative agreement with the corresponding theoretical line shape [see Fig. 2(b)]; here less than three cycles of absorption and spontaneous reemission took place for an excited atom. We note that the small difference between experimental and theoretical line shapes can be explained in a satisfying way by the short-term frequency jitter of our dye laser; during the atomic time of flight through the laser beam this frequency jitter has a bandwidth of roughly 250 kHz and spreads the modification of the velocity distribution among a wider velocity subgroup. We also note that the small narrow feature observed at the very center of the resonance is an artifact of our phase-sensitive experimental method; it is due to the direct beat signal of the pump field ($\omega_0$) and the carrier field ($\omega_C$) of the test beam exactly on resonance ($|\omega_C - \omega_0| = \omega_M$).

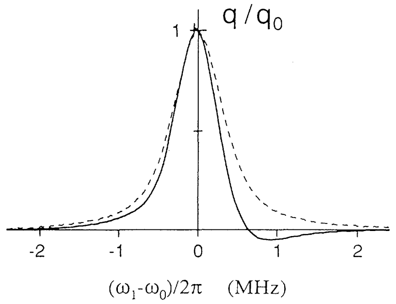

FIG. 4. Typical measured modulation signals $q$ for a pump beam intensity of $I_0 \approx 20\ \mu\mathrm{W/cm^2}$, corresponding to a saturation parameter $G \approx \frac{1}{8}$; the intensity of the test sideband was $I_1 \approx 10\ \mu\mathrm{W/cm^2}$. The solid curve was observed on pure Yb vapor and the dashed curve with 0.02 mbar argon added. Both curves are normalized to the signal strength $q_0$ observed exactly on resonance ($\omega_1 = \omega_0$).

In a variation of our experiment, we added small amounts of rare-gas perturber atoms (Ar) to the ytterbium vapor. For a sufficiently high buffer-gas pressure, velocity-changing collisions lead to a rapid thermalization of the velocity distribution. As a consequence, the light-pressure-induced signal contribution vanishes and the even symmetry of the saturation signal is restored; this is demonstrated by the dashed curve in Fig. 4. We note that, for high-precision experiments, it may be of importance to carefully choose a proper buffer-gas pressure in order to eliminate the light-pressure-induced line-shape asymmetries without introducing unwanted effects due to collisions. In another variation of our experiment, we used a pump and a test field which did not spatially overlap in the sample. In this way, the light-pressure-induced signal contributions (dashed curves in Fig. 2) can be observed separately from the ordinary saturation contributions since, under suitable conditions, the perturbation of all internal atomic degrees of freedom is completely relaxed when the atoms enter the probe zone. The signals observed in this way yield an unambiguous proof for a perturbation of the *external* atomic degrees of freedom. Corresponding experimental results are in good agreement with theory; details will be reported elsewhere.

In laser spectroscopy, line-shape modifications related to photon momentum transfer have already been observed in some other experimental situations. The well-known recoil splitting of the Lamb dip,[10] e.g., occurs for very weak optical transitions as a consequence of the *single*-photon momentum transfer that is inherently connected with an absorption process. In contrast to this, the line-shape modifications considered in our work are a result of photon momentum transfer that is connected with *cumulative cycles* of absorption and spontaneous reemission, leading to a well-known spontaneous scattering force; as a consequence, the effect reported here can also be present for strong optical transitions. Various other line-shape asymmetries induced by spontaneous[11] and stimulated light forces[12] have been studied for the transverse excitation of a well-collimated atomic beam with traveling- and standing-wave laser fields. We point out that the asymmetries reported there vanish for low laser intensities; in contrast to this, the light-pressure-induced asymmetry of the saturation dip in a low-pressure gas, which we study here, is also present for the case of low laser intensities, where the asymmetric line shape is *independent* of the laser intensity.

In conclusion, our experiments have demonstrated that the spontaneous scattering force can lead to a substantial modification of the line shape of Doppler-free resonances obtained by saturated absorption spectroscopy in low-pressure gases; our experimental results are in satisfying agreement with theory. On one hand, light-pressure-induced asymmetries and line shifts as reported here contain basic information on the interaction of light and matter; on the other hand, measurements in high-resolution optical spectroscopy may be affected with possible consequences, e.g., for high-precision frequency measurements using absorption cells.

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