Observation of Light-Pressure–Induced Dispersion in Yb Vapor

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We report on the observation of a light-pressure–induced dispersion of laser light interacting with a Doppler-broadened atomic medium. This novel dispersion feature displays an even symmetry with respect to the optical Doppler detuning; it is due to the small modification of the atomic velocity distribution caused by the spontaneous scattering force. Our phase-sensitive measurements were performed on the $\lambda = 555.65$ nm ${}^1S_0$-${}^3P_1$ transition in ytterbium vapor by the use of frequency-modulation spectroscopy. The experimental results are in good agreement with theoretical predictions.

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In recent years, resonant-light-pressure effects have attracted increasing attention[1]; here, laser cooling of atoms in a *beam* is one example of special experimental interest.[1,2] In principle, the velocity distribution of an *atomic vapor* also can be modified by resonant light pressure.[3] In this context, recent theoretical investigations have shown that the velocity distribution caused by the spontaneous scattering force can lead to strong modifications of the optical susceptibility of a low-pressure gas, giving rise to new phenomena in its optical response.[4,5] For the very basic situation of a single laser beam interacting with a Doppler-broadened medium, the occurrence of a nonlinear contribution to the dispersion of the light field has been predicted as the main consequence of radiation pressure. This *light-pressure-induced nonlinear dispersion phenomenon* can have drastic consequences for the total dispersion curve of the medium, affecting, e.g., its symmetry and line center.[5] In this contribution, we report the first experiments on atomic ytterbium vapor that clearly demonstrate the existence of this novel dispersion phenomenon.

Before we present our measurements, let us first give a brief theoretical outline of the phenomenon. We consider the interaction of an atomic vapor with a monochromatic laser field of frequency ω and wave number k. The medium is assumed to consist of two-level atoms with transition frequency Ω, mass M, and a Doppler distribution $N_0(v)$. For simplicity, let us assume here a velocity distribution of Lorentzian shape $N_0(v) = u/\pi(v^2 + u^2)$.[6] For small light-pressure–induced modifications of the velocity distribution, the total dispersion D can be written in the form[5]

$$D = \frac{\kappa}{\pi u}\left[-\frac{\delta}{\delta^2+1} + \epsilon_r \tau \frac{r^2(1+2r^2)^{-3/2}}{\delta^2+1}\right]. \qquad (1)$$

Here D is related to the refractive index n of the medium by $D = (n-1)kL$, where L is the interaction length; κ denotes a proportionality constant. The dimensionless optical detuning $\delta = (\omega - \Omega)/ku$ is expressed in units of the Doppler width ku. $2\epsilon_r = \hbar k^2/M$ is the Doppler shift that an atom experiences as a result of one photon momentum transfer, and τ denotes an effective interaction time of single atoms with the light field. $r = \chi/2\Gamma$ is the optical Rabi frequency χ normalized to the full natural optical linewidth 2Γ. While the first term in Eq. (1) is the ordinary linear dispersion, the second term describes the light-pressure–induced nonlinear dispersion. A representative plot of the corresponding dispersion curves for $\epsilon_r \tau = 1$ and $r = 1$ is shown in Fig. 1: The light-pressure–induced dispersion (dotted curve) displays an *even symmetry* with respect to the laser detuning δ. This remarkable property is due to the fact that the nonlinear dispersion arises from a local antisymmetry of the velocity distribution (see inset in Fig. 1). The antisymmetry is induced by the light itself via its radiation pressure and occurs in a narrow interval corresponding to the homogeneous optical linewidth.[7] Let us point out that neither the shape nor the sign of the antisymmetric modification depends on the Doppler detuning δ; δ only determines the velocity $v_0 = \delta u$ where the Doppler distribution is redistributed. As a consequence of the unusual

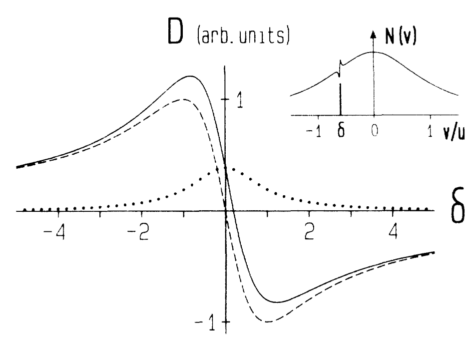

FIG. 1. Dispersion curves for $\epsilon_r \tau = 1$ and $r = 1$ according to Eq. (1): The total dispersion (solid curve) is the sum of the light-pressure–induced contribution (dotted curve) and the ordinary linear dispersion (dashed curve). $\delta = (\omega - \Omega)/ku$ denotes the laser detuning in units of the Doppler width. The inset shows the atomic velocity distribution $N(v)$ with a typical modification caused by light pressure.

even symmetry of the nonlinear contribution, the total dispersion curve (solid line in Fig. 1) is asymmetric, and its line center displays a significant shift towards higher frequencies; for the parameter values of Fig. 1, this shift amounts to approximately 20% of the Doppler width. Equation (1) also predicts a pronounced nonlinear intensity dependence of the light-pressure–induced dispersion; furthermore, for a given laser intensity, its strength is closely related to the laser beam diameter via the effective atom-field interaction time τ.

Our experiment to measure the light-pressure–induced dispersion phenomenon is based on frequency-modulation spectroscopy[8]; the experimental scheme is shown in Fig. 2. In this simple and sensitive technique the interaction of pure phase-modulated light with the atomic sample gives rise to amplitude-modulation components in the transmitted laser light; here an amplitude modulation occurs not only because of a different absorption of the frequency-modulation sidebands in the sample but also because of a phase shift of the carrier field with respect to the sideband fields. Let us assume a small modulation phase shift $\Delta\Phi = m\cos(\omega_M t)$ of the incident light with $m \ll 1$. Under appropriate conditions $[\Gamma(1+2r^2)^{1/2} \ll \omega_M \ll ku]$, the modulated part $I_{\rm mod}$ of the intensity of the transmitted light can be written in the form[5]

$$I_{\rm mod} = I[q\sin(\omega_M t) - p\cos(\omega_M t)]\,, \tag{2}$$

with

$$q = 2m\kappa\frac{\omega_M}{ku}u\frac{\partial N_0(v_0)}{\partial v_0}\,, \tag{3a}$$

$$p = 2m\kappa N_0(v_0)\epsilon_r\tau r^2(1+2r^2)^{-3/2}\,; \tag{3b}$$

here I is the incident light intensity. The resonant velocity $v_0 = (\omega - \Omega)/k$ and the normalized Rabi frequency r are defined with respect to the carrier field.

The signal components p and q clearly contain different information. The modulation component q oscillates in quadrature to the applied phase modulation; this signal arises from the different absorption of the frequency-modulation sidebands. Its dependence on the

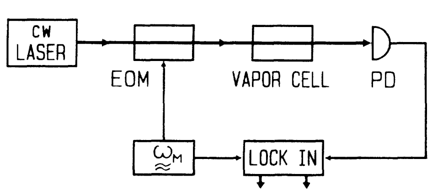

FIG. 2. Experimental scheme to observe the light-pressure–induced nonlinear dispersion in an atomic vapor: EOM, electro-optic phase modulator; PD, photodetector; p and q, in-phase and quadrature component of the detected amplitude modulation, respectively. For further details see text.

optical detuning $kv_0 = \omega - \Omega$ simply displays the derivative of the Doppler distribution $N_0(v)$, which corresponds to the absorption profile for weak light intensities. The in-phase modulation component p results directly from the slight phase shift of the strong carrier with respect to the weak sidebands; this phase shift is caused by the light-pressure–induced dispersion effect. Thus, the modulation component p allows a direct measurement of the nonlinear dispersion phenomenon.

Our measurements were performed on the ytterbium line $\lambda = 555.65$ nm ($4f^{14}6s^2\,{}^1S_0$–$4f^{14}6s6p\,{}^3P_1$). The ytterbium vapor was contained in a heated ceramic tube of 1.8-cm diameter; the temperature of the vapor cell was about 700 K with the length of the heated zone being approximately 6 cm. Natural ytterbium consists of a mixture of seven isotopes with mass numbers 168, 170, 171, 172, 173, 174, and 176. The total Yb number density was estimated to be 4×10^{11} cm^{-3} with a corresponding vapor pressure of roughly 5×10^{-5} mbar. The natural linewidth of the transition is $\Gamma = 2\pi\times95$ kHz[9]; its Doppler width ku amounts to $2\pi\times460$ MHz. We note that the width of the total isotopic structure[10] of this transition exceeds the Doppler width of its single components; thus, a separate optical excitation of even as well as odd isotopes is possible. Let us point out that only the even isotopes represent a Doppler-broadened two-level system corresponding to the theory presented above.

The light of a continuous-wave single-mode dye ring laser was phase modulated by an electro-optical modulator with a modulation frequency $\omega_M = 2\pi\times9.8$ MHz. In most of our experiments, we chose a modulation depth of $m \approx \frac{1}{5}$; in this case, about 1% of the total light intensity is transferred to each of the two sidebands. Behind the modulator the laser beam was expanded to a diameter of approximately 1.3 cm; for this value, the effective interaction time τ is about 35 μs, corresponding to $\epsilon_r\tau \approx 0.85$. The expanded beam was carefully collimated, in order to obtain plane wave fronts within the optically thin sample. After the interaction with the medium, the resulting amplitude modulation of the light was detected by a fast photodiode. The detector output was demodulated in a high-frequency lock-in amplifier, yielding both the in-phase and quadrature amplitude-modulation components p and q, respectively.

Experimental curves for the modulation components were recorded while the laser frequency was scanned over the entire ytterbium line including all isotopes. Typical measured curves for the quadrature and in-phase modulation components are shown in Figs. 3(a) and 3(b), respectively. In order to facilitate an interpretation of these curves, center frequencies and relative weights of the lines corresponding to the various even and odd isotopes are indicated in Fig. 3(b). As expected [see Eq. (3a)], the quadrature component q shows the derivative of the linear absorption profile; here *all* isotopes contribute according to their relative weights. In contrast, in

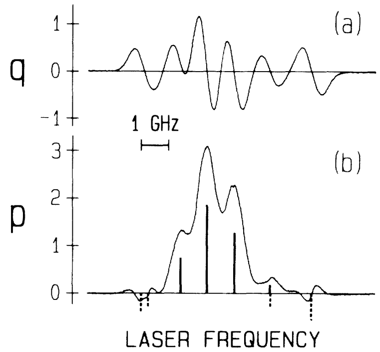

FIG. 3. Typical measured (a) quadrature and (b) in-phase amplitude-modulation components q and p of the transmitted laser light as a function of the laser frequency; q and p are given in arbitrary units with the same scale in (a) and (b). Here, the laser intensity was 2.8 mW/cm^2 and a beam diameter of about 1.3 cm was used. The p component directly reflects the light-pressure–induced dispersion, which occurs only for the even ytterbium isotopes. Center frequencies and relative weights of the various even and odd isotopes are illustrated by the solid and dashed bars in (b), respectively.

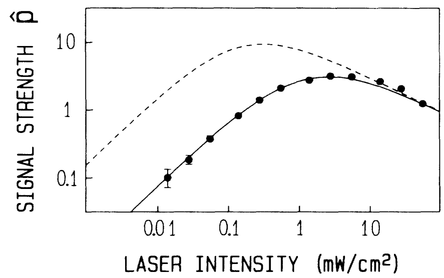

FIG. 4. The measured strength $\hat{p}$ of the light-pressure–induced dispersion signal as a function of the laser intensity. For comparison, the dashed curve shows the calculated signal strength according to Eq. (3); the solid curve results from a theory that takes into account a laser frequency jitter with a bandwidth of 360 kHz.

the in-phase component p, the strong light-pressure–induced signal shows up only for the *even* isotopes; their Doppler distribution is directly reflected in this signal, in full agreement with our theoretical expectations [see Eq. (3b)]. For the odd isotopes no strong in-phase signal occurs; here resonant light pressure cannot act effectively on the atoms because of optical pumping between the Zeeman sublevels of the ground state.[11]

The modulation signals were studied for laser intensities from 10 $\mu W/cm^2$ to 50 mW/cm^2, covering a range from far below ($r \approx \frac{1}{5}$) to far above saturation ($r \approx 10$). To measure the maximum strength of the light-pressure–induced dispersion signal in the p component, the laser frequency was tuned to the line center of the most abundant isotope ^{174}Yb. The observed strength $\hat{p}$ of the light-pressure–induced dispersion signal is shown in Fig. 4 as a function of the laser intensity; this signal strength $\hat{p}$ is normalized to the corresponding maximum strength of the signal component q, which is related to the absorption of the light field. A full quantitative agreement with the theory presented above only exists for high laser intensities; for low intensities, the observed signal is about 10 times weaker than predicted (see dashed line in Fig. 4). Without going into details here, we note that this discrepancy can be explained by the short-term frequency jitter of our dye laser.

We also measured the dependence of the light-pressure–induced signal on the laser beam diameter. For the low vapor pressure in our experiments, the laser beam diameter directly determines the effective interaction time τ: As collisions can be neglected here, τ is simply given by the transit time of the atoms through the laser beam. As a consequence, τ depends linearly on the laser beam diameter, and, according to Eq. (3b), also a linear dependence of the strength of the light-pressure–induced dispersion phenomenon on the beam diameter is expected. In order to measure the signal strength as a function of the laser beam diameter for a constant light intensity, the fully expanded beam with a $1/e^2$ diameter of 1.3 cm was reduced to various diameters by the use of an iris diaphragm. The observed linear dependence (see Fig. 5) is in full accordance with our theoretical expectations.

In another series of experiments, we added small amounts of rare-gas perturber atoms (Ar, He) to the ytterbium vapor. For both argon and helium, the absorptive signal component q is essentially not affected. In contrast, we observe a clear decrease of the light-pressure–induced signal in the p component for pressures larger than about 10^{-3} mbar; above this value, the ytterbium atoms undergo a substantial amount of velocity-changing collisions, which lead to a rapid thermalization of their velocity distribution. Thus, our measurements clearly show that both a two-level system *and* a collision-free regime are conditions for a strong occurrence of light-pressure–induced dispersion. This confirms that the phenomenon is, in fact, related to *accumulated* photon momentum transfer. In this respect, it stands in con-

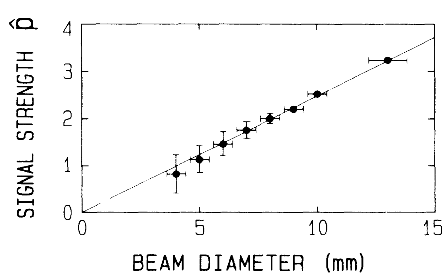

FIG. 5. The observed strength $\hat{p}$ of the light-pressure–induced dispersion signal as a function of the laser beam diameter; the laser intensity is kept constant. The solid line shows a linear fit to the measured values.

trast with previous work on the recoil splitting of the Lamb dip for very weak optical transitions,[12] which is related to the *single* photon momentum transfer that is inherently connected with an absorption process.

In conclusion, our experiments have demonstrated that resonant light pressure can lead to a substantial contribution to the dispersion of a light field interacting with a Doppler-broadened medium. All our experimental results are in good agreement with theoretical predictions[4,5]: We have verified the even symmetry of the dispersion phenomenon with respect to the optical Doppler detuning; moreover, we have observed the pronounced saturation behavior of the dispersion feature and the direct relation of its strength to the laser beam diameter. Finally we note that this effect may play a role whenever optical phase-sensitive methods are applied to investigate low-pressure gases. In this respect, various nonlinear spectroscopic techniques using optical heterodyne detection may be mentioned as examples.[13]

[1]See, e.g., *The Mechanical Effects of Light,* edited by P. Meystre and S. Stenholm [J. Opt. Soc. Am. B **2**, 1705–1860 (1985)], and references therein.

[2]See, e.g., S. V. Andreev *et al.*, Zh. Eksp. Teor. Fiz. **82**, 1429 (1982) [Sov. Phys. JETP **55**, 828 (1982)]; J. Prodan, W. Phillips, and H. Metcalf, Phys. Rev. Lett. **49**, 1149 (1982).

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[5]R. Grimm and J. Mlynek, J. Opt. Soc. Am. B. **5**, 1655 (1988); in this reference, a factor $1/u$ is missing on the right-hand side of Eq. (14).

[6]For a Gaussian velocity distribution, the mathematical formulation of the total dispersion and thus its interpretation are much more complicated than for a Lorentzian distribution; the results, however, show only an insignificant quantitative difference.

[7]As a consequence of the fact that a small light-pressure–induced modification of the velocity distribution is essentially antisymmetric, the absorption profile of the vapor is not modified significantly.

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[13]See, e.g., M. Gehrtz, G. C. Bjorklund, and E. A. Whittaker, J. Opt. Soc. Am. B **2**, 1510 (1985), and references therein.