Nonlinear selective reflection of bichromatic light from interface between transparent dielectric and resonant gas

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The differential reflection spectrum of one component of intense bichromatic light at an interface between glass and cesium vapor has been studied experimentally. Resonances have been detected in the nonlinear variation of the reflection coefficient. An explanation proposed for the formation of these resonances incorporates four mechanisms: saturation of an optical transition, a collision with a resonant transfer of excitation, a velocity-selective optical pumping of atoms, and a coherent capture of populations.

Selective specular reflection has recently been attracting increasing interest. In particular, there have been theoretical and experimental studies of the shape of the sub-Doppler reflection resonances which stem from depolarizing collisions of an atomic gas with a gas-glass interface at low[1-3] and high[4-7] light intensities. Resonances of the saturated reflection have been detected in a layout involving two counterpropagating surface waves.[3,8] Self-diffraction resonances at a nonlinear gas mirror have been detected.[5]

In this letter we are reporting an experimental study of the spectrum near the D_2 line ($\lambda = 852$ nm) of the nonlinear variation of the reflection coefficient of one component of intense bichromatic light as it is reflected at an interface between glass and ^{133}Cs vapor. The hyperfine splitting of the corresponding excited states, $6P_{3/2}(F')$, is smaller than the Doppler width $\Delta\nu_D \approx 0.4$ GHz, and the spectral interval ($\Delta\nu_0 \approx 9.2$ GHz) between the sublevels of the ground states, $6S_{1/2}$ ($F = 3$) and $6S_{1/2}$ ($F = 4$), is much larger than $\Delta\nu_D$. The experiments were carried out at a cesium-atom density $N \approx 3.3 \times 10^{14}$ cm^{-3}, at which the sum of the radiation width, $\gamma_r \approx 5.3$ MHz, and the collisional width, $\gamma_c \approx 1.1 \times 10^{-7} N$ Hz ≈ 36 MHz (N[cm^{-3}]), of the line is smaller than[1] $\Delta\nu_D$. The homogeneous linewidth, $\gamma = \gamma_r + \gamma_c$, was found by the method developed in Ref. 1. Specifically, the frequency interval between the inflection points of the sub-Doppler resonances of the linear selective specular reflection was measured as light was incident normally. The bichromatic light was formed by two independent injection lasers with external cavities, L1 (the saturating light) and L2 (the probe light). The frequencies of L1 and L2 were stabilized with the help of confocal interferometers. One of these interferometers operated in a scanning mode, so the frequency of L2 was tuned near either the long-wavelength component (Fig. 1) or the short-wave component (Fig. 2) of the cesium D_2 line. The frequency of the saturating laser was fixed within ± 25 MHz at the value corresponding to the $6S_{1/2}$ ($F = 4$) $-$ $6P_{3/2}$ ($F' = 5$) (ν_{45}) transition (Figs. 1b and 2b) or the $6S_{1/2}$ ($F = 4$) $-$ $6P_{3/2}$ ($F' = 4$) (ν_{44}) transition (Figs. 1c and 2c) on the basis of the sub-

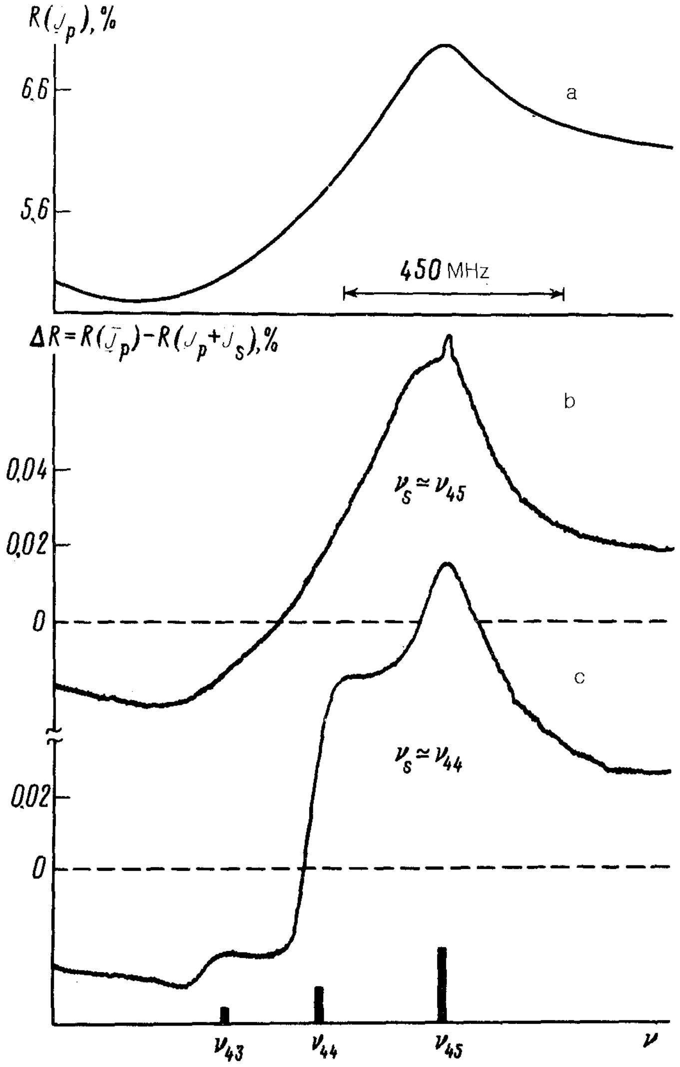

FIG. 1.

Doppler saturated absorption spectrum. The latter spectrum was measured with the help of an auxiliary cell with a low density ($\sim 3 \times 10^{10}$ cm^{-3}) of cesium atoms. The spectral width of the output of each laser was less than 1 MHz; the output power was about 2 mW. The laser is described in more detail in Ref. 9. The laser light was focused on the window of a glass cell holding saturated cesium vapor, to increase the intensity. The convergence angle of the light beams was less than 2×10^{-2} rad, as was the deviation of these beams from a parallel orientation. The cross section of the light beams at the glass-(cesium vapor) interface was 0.2×0.5 mm^2; the two beams were carefully brought into coincidence. The saturating beam was chopped at a frequency of 500 Hz before it reached the cell with the cesium vapor. After the probe beam was reflected from the glass-(cesium vapor) interface, it was sent to a photodiode. The output signal from the photodiode was fed to a tuned amplifier and then subjected to synchronous detection at a frequency of 500 Hz. This signal was then fed to an x, y chart recorder. The intensity of the saturating light, $J_s \approx 2.8$ W/cm^2, was four times that of the probe light, J_p. To eliminate the sub-Doppler resonances of the selective specular reflection which are observed during normal incidence, and also to simplify the spectrum of the nonlinear variation of the reflection coefficient, the bichromatic light was applied to the gas-vapor interface at the comparatively large angle $\Theta \approx 16°$.

We also detected the linear profile of the selective specular reflection (Figs. 1a and 2a). In this case the saturating beam was cut off, and the intensity of the probe light was attenuated to $J_p \approx 3$ mW/cm^2, at which the Rabi frequency is considerably smaller than the homogeneous linewidth $\gamma \approx 41$ MHz. Figures 1b and 2b show spectra of the nonlinear variation of the reflection coefficient, $\Delta R = R(J_p) - R(J_p + J_s)$, as

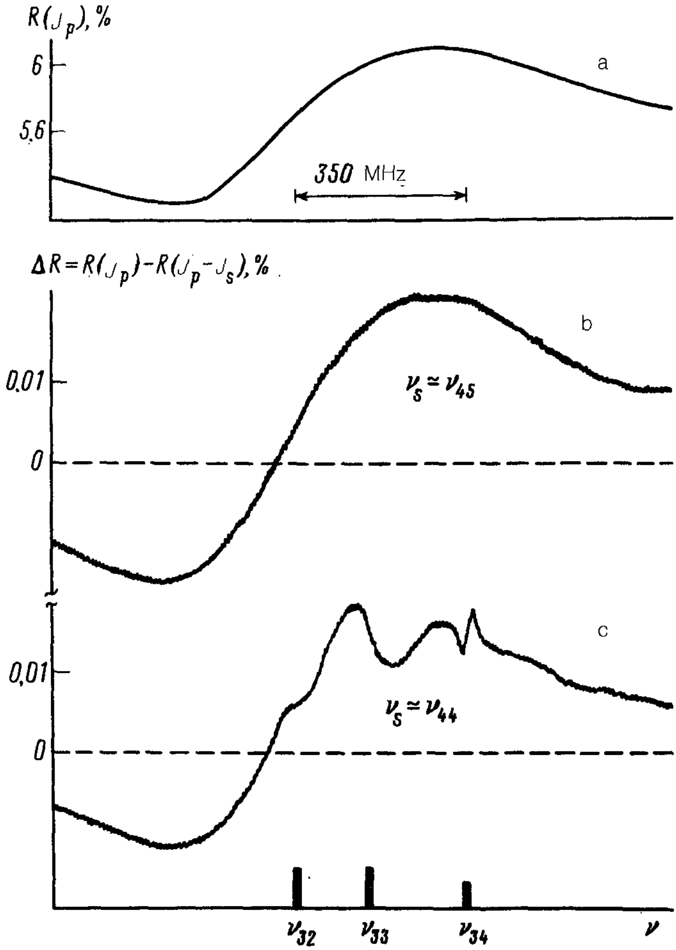

FIG. 2.

the saturating light is tuned to the frequency of the closed transition ν_{45}. The positive value of ΔR in Figs. 1b, 1c, 2b, and 2c corresponds to a decrease in the reflection coefficient for the light from the probe laser. The probable reason for the presence of a signal over a broad spectral band, including the $6S_{1/2}$ ($F=3$) $-$ $6P_{3/2}$ ($F'=2,3,4$) transitions (Fig. 2b), is a resonant transfer of excitation from the $6P_{3/2}$ ($F'=5$) sublevel to other sublevels of the excited state in the course of interatomic collisions. Support for this hypothesis comes from the qualitative agreement of the ΔR spectrum in Fig. 2 and the linear profile of the selective specular reflection in Fig. 2a. This situation corresponds to an independence of the cross section of resonant collisions from the velocity, as predicted by the theory of atomic collisions.[10] The nonlinear variation of the reflection coefficient for the probe wave, as its frequency is scanned over the spectral band $6S_{1/2}$ ($F=4$) $-$ $6P_{3/2}$ ($F'=3,4,5$) (Fig. 1b), stems primarily from a direct saturation of the ν_{45} optical transition by the second laser. Processes of a resonant transfer of excitation from the $6P_{3/2}$ ($F'=5$) level to the $6P_{3/2}$ ($F'=4,3,2$) levels and an optical pumping of the atoms from the $S_{1/2}$ ($F=4$) level to the $6S_{1/2}$ ($F=3$) level apparently also play a part in the formation of the ΔR signal in Fig. 1b. The ΔR spectrum in Fig. 1b agrees qualitatively with the linear profile of the selective specular reflection (Fig. 1a). Only at the ν_{45} frequency is there a structural feature in the spectrum. Near the frequency of the ν_{45} transition, the probe light and the saturating light interact with cesium atoms in the same velocity group, and a narrow saturation resonance appears. This resonance is probably associated with coherent effects in the population distribution.[11] The width of this saturation peak, $\Gamma_1 \approx 20$ MHz, is smaller than the homogeneous width of the atomic transition, γ. A sharp change in the ΔR profile is observed when the frequency of the saturating light is shifted to the unclosed transition ν_{44} (Figs. 1c and 2c). In this case, there are increases in the efficiency of (first) the saturation of the ν_{44} and ν_{43} transitions and (second) the optical pumping of atoms from the $6S_{1/2}$ ($F=4$) sublevel to the $6S_{1/2}$ ($F=3$) sublevel. The result is an increase in ΔR near the frequencies ν_{44} and ν_{43} (Fig. 1c). In Fig. 2c there are some comparatively broad resonances near the frequencies ν_{32}, ν_{33}, and ν_{34} (a consequence of optical pumping), with a width ~ 100 MHz. The appearance of these resonances leads to a decrease in ΔR. In addition, at the frequency ν_{34} we see a narrow resonance with a dispersive shape (in contrast with the shape of the narrow resonance in Fig. 1b), with a width $\Gamma_2 \approx 20$ MHz. This resonance corresponds to a Λ configuration for the excitation of the ν_{44} and ν_{34} atomic transitions. The apparent reason for the appearance of the narrow resonances in Figs. 1b and 2c is that coherence effects occur in the distribution of the sublevel populations on the ground and excited states as a result of the exposure to the bichromatic light.[11] According to Ref. 11, the width of the resonances due to the coherent trapping of populations is determined by the relaxation rate of the atomic ground state and field-induced broadening. For cesium vapor, the cross sections for the collisional broadening of the ground state and the excited state differ by a factor of 10^3 (Ref. 12). In other words, the width of the narrow resonances in Figs. 1b and 2c is limited by the field-induced broadening. This interpretation is based on the relatively small broadening (<10 MHz) of the resonances, $\Gamma \approx 20$ MHz, as the collisional width γ_c of the atoms increases from 36 MHz to 160 MHz. By reducing the intensity of the bichromatic light, we can apparently approach a width of the resonances which is comparable to the relaxation rate of the ground state.

In summary, this new procedure for studying the selective specular reflection by means of bichromatic light, has proved to be highly informative: Effects stemming from optical pumping, a coherent trapping of population, and a resonant exchange have been observed. The qualitative explanation of the observed spectra of the nonlinear variation of the reflection coefficient offered here is not an exhaustive explanation. A detailed theoretical analysis is required. A further study of these effects hold promise for furnishing new information about the processes that occur in optically dense gaseous media.

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