PHYSICAL REVIEW A, VOLUME 63, 034501

Saturated absorption signals for the Cs D_2 line

Kang-Bin Im, Hye-Yun Jung, Cha-Hwan Oh, Seok-Ho Song, and Pill-Soo Kim
Department of Physics, Hanyang University, Seoul 133-791, Korea

Ho-Seong Lee
Division of Electromagnetic Metrology, Korea Research Institute of Standards and Science, P.O. Box 102,
Yusong, Taejon 305-600, Korea

(Received 8 October 1999; published 13 February 2001)

The change of saturated absorption spectra of the Cs D_2 line were measured with the pump beam intensity. The line shapes of the resonance signals, especially for the cyclic transition $F=4 \rightarrow F'=5$, were very sensitive to the pump beam intensity. We suggested a simple model that phenomenologically describes the change of the line shape of the resonance signal with the pump beam intensity.

DOI: 10.1103/PhysRevA.63.034501 PACS number(s): 32.70.Jz, 32.80.Bx

Doppler-free saturation spectroscopy [1] is one of the simplest forms of high-resolution laser spectroscopy. The saturated absorption signal is widely used as a reference signal for frequency stabilization of the laser source [2,3]. The cyclic transition $F=4 \rightarrow F'=5$ of the Cs D_2 line is especially important in laser cooling and atomic clock experimentation [4–6]. Many investigations of the saturated absorption spectra of alkali atoms have been carried out [7–14].

In order to explain the saturated absorption spectra of alkali atoms, several approximate theories for the atomic susceptibility based on the rate equation of atomic states have been suggested. The saturation theory of a two-level atom [1] is applicable to the case where the pump beam intensity is higher than the saturation intensity of the atomic transition line. The velocity selective optical pumping theory [7–9] reasonably explains the saturated absorption spectra of alkali-metal atoms with two hyperfine levels in the ground state when the pump beam intensity is sufficiently lower than the saturation intensity of the atomic transition line. Grimm *et al.* [10] reported that the line shape of the resonance signal can be modified by resonant light pressure.

Recently, Schmidt *et al.* [11] and Oh and Ohshima [12] reported that the dip as well as sign reversal in the resonance for the transition $F=4 \rightarrow F'=5$ of the Cs D_2 line arose as the pump beam intensity increased. The dip shape in the resonance signal cannot be explained by only the one approximate theory mentioned above. For the exact description of the resonance signal, the density-matrix equation (considering the optical coherence and the detuning of the laser frequency from the center frequency of the resonance line) should be calculated. This calculation will be an enormous and tedious job.

In this paper, we suggested a simple model that phenomenologically describes the change of the saturated absorption signal of the Cs D_2 line with the pump beam intensity. The saturated absorption spectra were obtained by measuring the absorption of probe beams that passed through a counter-propagating pump beam in a Cs cell that was maintained at room temperature. The experiment setup was typically used in the saturated absorption spectroscopy [11,12,14]. The linearly polarized laser beam [New Focus, 6226, linewidth $(50\,\mathrm{ms}) < 300\,\mathrm{kHz}$] was divided into three beams by a 10-mm thick BK-7 bare glass. Two beams reflected at both surfaces passed through the Cs cell (10 mm length) to be used for probe beams, and one transmitted beam entered backward into the cell to be used for a pump beam. By taking the difference of the intensities of the two probe beams, the Doppler background could be eliminated from the spectra. The diameters of the pump and probe beams were 2.2 and 1.4 mm, respectively. The cell was located at the center of three orthogonal Helmholtz coil pairs. These coils compensated the geomagnetic field. Residual magnetic field at the position of the cell was below 2 mG.

Figure 1 shows the spectra of the Cs D_2 line when the polarizations of pump and probe beams were linear (lin∥lin and lin⊥lin configurations) and circular ($\sigma^+\sigma^+$ and $\sigma^+\sigma^-$ configurations). lin∥lin and lin⊥lin indicate the relatively parallel and perpendicular configurations of linearly polarized pump and probe beams. $\sigma^+\sigma^-$ indicates that the pump beam is left-circularly polarized and the probe beam is right-circularly polarized. There are three principal and three crossover resonances in the Cs D_2 line. The first, third, and sixth peaks ($k=1$, 3, and 6) are the principal resonances by

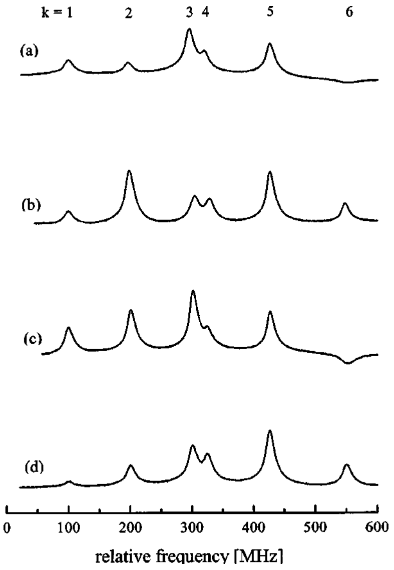

FIG. 1. Saturated absorption spectra for the transitions $F=4 \rightarrow F'=3,4,5$: (a) lin∥lin; (b) lin⊥lin; (c) $\sigma^+\sigma^+$; and (d) $\sigma^+\sigma^-$ configurations. Pump beam intensity: 1.7 μW/mm^2, probe beam intensity: 1.1 μW/mm^2.

the transitions $F=4\rightarrow F'=3$, $F=4\rightarrow F'=4$, and $F=4\rightarrow F'=5$, respectively. The second, fourth, and fifth peaks ($k=2$, 4, and 5) are the crossover resonances by the transitions $F=4\rightarrow F'=3, 4$, $F=4\rightarrow F'=3, 5$, and $F=4\rightarrow F'=4, 5$, respectively. A positive peak means the enhanced transmission and a negative peak the enhanced absorption of the probe beams. When the pump beam intensity was as weak as in the case of the figure, the spectra—especially the signs of each resonance signals—were reasonably explained by the calculated results in Ref. [9].

Figure 2 shows the saturated absorption signals for lin‖lin and $\sigma^+\sigma^+$ configurations at various pump beam intensities. As shown in the figure, the amplitude and sign of the resonance signal for the transition $F=4\rightarrow F'=5$ sensitively depended on the pump beam intensity. In both configurations, the resonance signal showed a dip [(b), (b′)] and sign reversal [(a), (a′)] as the pump beam intensity increased. These phenomena were reported [11,12] but any concrete expression has not been given, as far as we know.

Figures 3(a), 3(a′), 3(b), and 3(b′) show the experimental results compared with the calculated results by the four-level model [9] considering only the velocity-selective optical pumping effect. As shown in the figure, the resonance signals for the transition, $F=4\rightarrow F'=5$ are quite different from the calculated results.

Therefore, we know that the saturation effect as well as the optical pumping effect must be considered. Note that the saturation effect causes the enhanced transmission of the probe beams, that is, a positive peak in the resonance signal. In the saturation theory of a two-level atom, the pump beam must have a high intensity to saturate the atoms. However, the saturation effect cannot be neglected even if the pump beam intensity is lower than the saturation intensity of the Cs D_2 line in a two-level model, 10 μW/mm^2. From this consideration, these behaviors can be understood by the combined effect of the saturation, the velocity-selective optical pumping, and the resonant light pressure. As can be seen in Figs. 4(a), 4(a′), 4(b), and 4(b′), the linewidth of negative peak for the transition, $F=4\rightarrow F'=5$, was broader than that of a positive peak. This means that the saturation intensities for the velocity-selective optical pumping and saturation effects should be different because the linewidth of a resonance signal explicitly depends on the saturation intensity. The narrow saturation peak is not centered on the broad optical pumping dip. This shift is induced by resonant light pressure, which becomes important for the $F=4\rightarrow F'=5$ closed system [10,11].

From the above consideration, we can suggest a simple model for the signal of the principal resonances ($k=1,3,6$ in Fig. 1) as follows [7,9,10]:

$$
\begin{aligned}
L_k(I_p) \propto{}& S_k \frac{I_p/I_{\mathrm{sat}}}{\sqrt{1+I_p/I_{\mathrm{sat}}}\,(1+\sqrt{1+I_p/I_{\mathrm{sat}}})}\,\frac{(\Delta\nu/2)^2}{(\nu-\nu_k)^2+(\Delta\nu/2)^2}\\
&+P_k \frac{I_p/I'_{\mathrm{sat},k}}{\sqrt{1+I_p/I'_{\mathrm{sat},k}}\,(1+\sqrt{1+I_p/I'_{\mathrm{sat},k}})}\\
&\times\frac{(\Delta\nu'_k/2)^2}{(\nu-\nu_k)^2+(\Delta\nu'_k/2)^2}\\
&+2\varepsilon_r\tau_{\mathrm{tr}}S_k \frac{I_p/I_{\mathrm{sat}}(\nu-\nu_k)}{\sqrt{1+I_p/I_{\mathrm{sat}}}\,(1+\sqrt{1+I_p/I_{\mathrm{sat}}})}\\
&\times\frac{(\Delta\nu/2)^3}{[(\nu-\nu_k)^2+(\Delta\nu/2)^2]^2},
\end{aligned}
\tag{1}
$$

where ν and ν_k are the relative frequency of the laser and the kth resonance, respectively. $\Delta\nu$ and $\Delta\nu'_k$ are the linewidths for the saturation and the velocity selective optical pumping effect, respectively. I_p is the pump beam intensity.

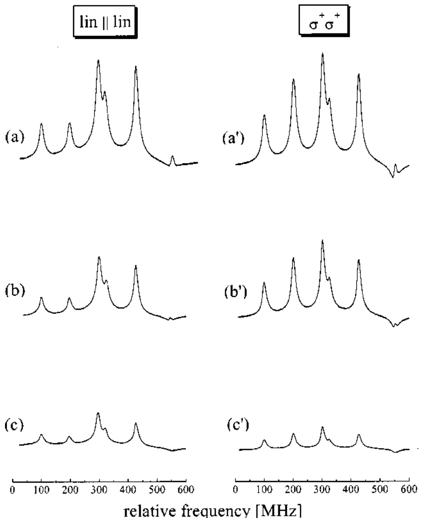

FIG. 2. Saturated absorption spectra for the transition $F=4\rightarrow F'=3,4,5$ at various pump beam intensities. (a)–(c) lin‖lin configuration, (a′)–(c′) $\sigma^+\sigma^+$ configuration. Pump beam intensity: (a) and (a′) 5.8 μW/mm^2; (b) and (b′) 3.2 μW/mm^2; (c) 1.7 μW/mm^2; and (c′) 0.7 μW/mm^2.

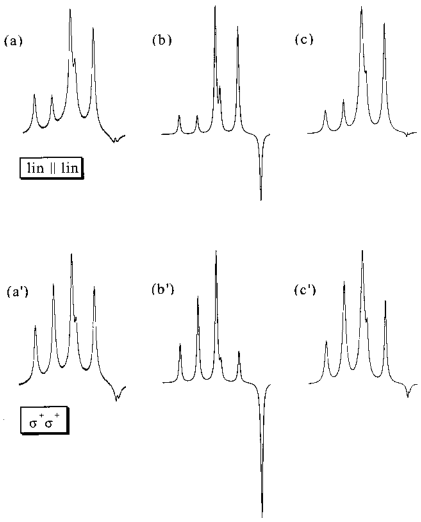

FIG. 3. Saturated absorption spectra for the transition $F=4\rightarrow F'=3,4,5$. (a), (b), (c) lin‖lin; (a′), (b′), (c′) $\sigma^+\sigma^+$ configuration. (a) and (a′) measured; (b) and (b′) calculated by the four-level model considering only the velocity-selective optical pumping effect; (c) and (c′) calculated by Eq. (1). Pump beam intensity: 3.4 μW/mm^2.

TABLE I. Parameters used in the calculation of the resonance signals. The Doppler factor was calculated for the room temperature, 21 °C.

Transition	S_k		P_k			
(k=1)	lin‖lin	$\sigma^+\sigma^+$	lin‖lin	$\sigma^+\sigma^+$	$I'_{\mathrm{sat}.k}$	D_k
1	0.250	0.250	0.072	0.091	0.0378	1
2	0.833	0.833	0.084	0.240	0.0378 0.0678	0.8213
3	0.583	0.583	0.480	0.300	0.0678	1
4	1.250	1.250	0.410	0.110	0.0378 0.910	0.3697
5	1.583	1.583	0.550	0.090	0.0678 0.910	0.7358
6	1	1	-0.254		0.910	1

The first term represents the contribution to the signal by the saturation effect, which can be described by a two-level atomic model and obtained from the rate equation of atomic states [1]. In the two-level atomic model, the saturation intensity I_{sat} for the Cs D_2 line is calculated to be 10 μW/mm^2 [11]. The linewidth $\Delta\nu$ is given by $\Delta\nu = R_D + \Gamma(1 + \sqrt{1 + I_p/I_{\mathrm{sat}}})/2$. The natural linewidth of the Cs D_2 line Γ is known to be 4.9 MHz. S_k is the relative signal magnitude for the saturation effect, which is proportional to the transition rate between the corresponding hyperfine lines (see Table I).

The second term represents the contribution to the signal by the velocity-selective optical pumping effect. P_k is the relative signal magnitude for the velocity-selective optical pumping effect, which can be calculated by Nakayama's four-level model [9]. Contrary to the two-level model, the population in one of the ground states of alkali atoms can be bleached by even a weak pump beam because the lifetime of the excited state is much shorter than that of the ground state. This bleaching of the population in the hyperfine level gives the effect of a decrease in the saturation intensity [8], $I'_{\mathrm{sat},k}$ is given by [7,8] $I'_{\mathrm{sat},k} = (\tau_e/\tau_{\mathrm{eff}}) I_{\mathrm{sat}}$, where τ_e is the excited lifetime (32.7 ns) and τ_{eff} the effective lifetime, which is given by $\tau_{\mathrm{eff}} = 1/\gamma_e + 1/\gamma_g - \Gamma_{eg}/\gamma_e\gamma_g$. Neglecting atomic collisions, $1/\gamma_g$ is equal to the transit time τ_{tr} of the atom across the laser beam. Considering the pump beam diameter and the most probable speed of the Cs atom, τ_{tr} was estimated to be 11.5 μs in our experiment. γ_e is the total spontaneous emission rate of the upper level and Γ_{eg} the partial transition rate. $I'_{\mathrm{sat},k}$ is calculated to be $I'_{\mathrm{sat},1} = 0.0378\,\mu\mathrm{W/mm}^2$ for $F = 4 \rightarrow F' = 3$ and $I'_{\mathrm{sat},3} = 0.0678\,\mu\mathrm{W/mm}^2$ for $F = 4 \rightarrow F' = 4$. As for the cyclic transition, $F = 4 \rightarrow F' = 5$, there is no hyperfine pumping effect to another ground state, $F = 3$ because this transition is theoretically cyclic. However, the effect of atomic population reduction in the ground state has to be considered. This can be caused by the population leakage to $F = 3$. Avila *et al.* reported that the effective reduction of population density of $F = 4$ level arose even by the cyclic transition, $F = 4 \rightarrow F' = 5$ [5]. The effective reduction of population density of $F = 4$ by the cyclic transition, $F = 4 \rightarrow F' = 5$ was found to be about 3% at the pump beam intensity of 10 μW/mm^2, then $I'_{\mathrm{sat},6}$ was determined to be 0.91 μW/mm^2 from Eq. (2). The decrease of saturation intensity leads to a pronounced increase in linewidth for the optical pumping effect $\Delta\nu'_k$,

$$\Delta\nu'_k = R_D + \Gamma/2\,(1 + \sqrt{1 + I_p/I'_{\mathrm{sat},k}}), \tag{2}$$

where R_D is the residual Doppler linewidth, which equals 6 MHz in our experimental condition. The residual Doppler linewidth is caused by the slight tilt of the propagation direction of the pump beam intended to prevent the laser light from returning to the laser diode cavity. The P_6 value for the resonance signal $F = 4 \rightarrow F' = 5$, relative to $S_6(=1)$, was adjusted by fitting the measured peak of the resonance signal with the pump beam intensity to Eq. (1). P_6 was determined to be -0.254 for lin‖lin configuration and -0.312 for $\sigma^+\sigma^+$ configuration. In Table I, the parameters used in the calculation of the resonance signals are summarized.

The third term represents the contribution to the signal by the resonant light pressure effect. The light pressure effect leads to the asymmetric lines. The modification of the line shape occurs as a result of the small modification of the atomic velocity distribution caused by the spontaneous scattering force of the pump field. τ is the transit time and $\hbar\varepsilon_r$ the recoil energy [10], $\varepsilon_r = (\hbar k_w^2/2M)$, where M is the atom mass and k_w the wave number, respectively, $\varepsilon_r\tau_{\mathrm{tr}} = 0.155$ under our experimental condition. Then, the light pressure effect was too small to induce the sign reversal. The light pressure effect could describe only the asymmetric dip shape in the resonance signal [10,11].

Note that the signal for the crossover lines ($k = 2$, 4, and 5 in Fig. 1) is the sum of the associated principal lines. Therefore, the saturation intensity for the crossover resonance cannot be determined to a single value. For example, the signal of the transition $F = 4 \rightarrow F' = 3,4$ ($k = 2$) can be given by

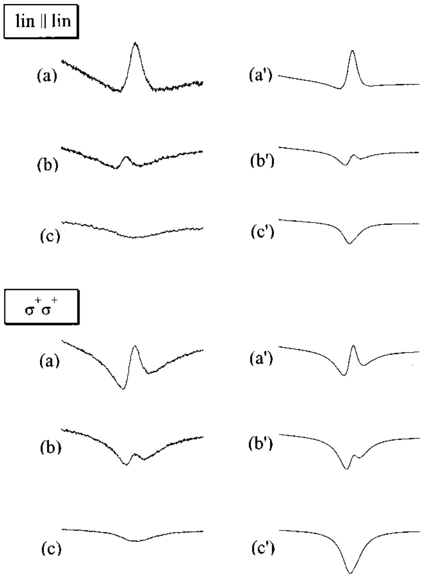

FIG. 4. Saturated absorption spectra for the transition $F = 4 \rightarrow F' = 5$ compared with the calculated results (a)–(c) measured; (a′)–(c′) calculated. Pump beam intensity: (a) and (a′) 5.2 μW/mm^2; (b) and (b′) 3.4 μW/mm^2; (c) and (c′) 1.7 μW/mm^2.

$$
\begin{aligned}
L'_{k=2}(I_p) \propto\, & (S_1+S_3) D_2 \frac{I_p/I_{\mathrm{sat}}}{\sqrt{1+I_p/I_{\mathrm{sat}}}\,(1+\sqrt{1+I_p/I_{\mathrm{sat}}})} \frac{(\Delta\nu/2)^2}{(\nu-\nu_2)^2+(\Delta\nu/2)^2} + \frac{P_2}{2} D_2 \frac{I_p/I'_{\mathrm{sat},1}}{\sqrt{1+I_p/I'_{\mathrm{sat},1}}\,(1+\sqrt{1+I_p/I'_{\mathrm{sat},1}})} \\
& \times \frac{(\Delta\nu'_1/2)^2}{(\nu-\nu_2)^2+(\Delta\nu'_1/2)^2} + \frac{P_2}{2} D_2 \frac{I_p/I'_{\mathrm{sat},3}}{\sqrt{1+I_p/I'_{\mathrm{sat},3}}\,(1+\sqrt{1+I_p/I'_{\mathrm{sat},3}})} \frac{(\Delta\nu'_3/2)^2}{(\nu-\nu_2)^2+(\Delta\nu'_3/2)^2} \\
& + 2\varepsilon_r \tau_{\mathrm{tr}} (S_1+S_3) D_2 \frac{I_p/I_{\mathrm{sat}}(\nu-\nu_2)}{\sqrt{1+I_p/I_{\mathrm{sat}}}\,(1+\sqrt{1+I_p/I_{\mathrm{sat}}})} \frac{(\Delta\nu/2)^3}{[(\nu-\nu_2)^2+(\Delta\nu/2)^2]^2},
\end{aligned}
\tag{3}
$$

where $D_k(=\exp[-(\Delta\omega_{ij}/2k_w u)^2])$ is the Doppler factor [9], which implies the reduction of the relative magnitude of a crossover resonance between principal resonances (see Table I). $\Delta\omega_{ij}=\omega_i-\omega_j$, where ω_i and ω_j are the relative angular frequency of the associating principal resonances. k_w is the wave number of the laser light, and the most probable velocity u is given by $(2k_BT/M)^{1/2}$, where T is the gas temperature, k_B is Boltzmann's constant, and M is the atomic mass. In the second and third terms of Eq. (3), the factor $\frac{1}{2}$ is multiplied by the signal for the crossover lines because each magnitude of the crossover lines can be calculated.

Figures 3(c) and 3(c′) show the results calculated by Eqs. (1) and (3). As shown in the figure, the relative magnitudes and the linewidths of the resonance signals were fitted to the experimental data better than the case considering only the velocity selective optical pumping effect [Figs. 3(b) and 3(b′)]. In addition, the dip shape in the resonance signal, $F=4\rightarrow F'=5$, could be obtained, which was impossible using only the velocity-selective optical pumping theory.

The detailed shapes of the resonance signal for the transition, $F=4\rightarrow F'=5$, are shown in Fig. 4. The calculated results reasonably agree with the experimental data. Especially, the asymmetry of the dip also can be described by the resonant light pressure effect. As the pump beam intensity increases, the saturation effect dominates the optical pumping effect. As shown in the figure, the saturation effect should be considered even though the pump beam intensity is lower than that of the two-level atomic model 10 μW/mm^2. In the region of pump beam intensity below 2 μW/mm^2, the calculated results were not matched precisely to the measured results. This may be attributed to the fact that the pump beam intensity in this region was lower than or as much as the probe beam intensity. We neglected the optical pumping by the probe beam in Eq. (1). However, the effect of the probe beam cannot be neglected in this region. Another reason may be the residual magnetic field at the Cs cell, since the resonance signal at low-pump beam intensity is more sensitive to the magnetic field than at high-pump beam intensities, which we observed easily in our previous experiment [14] by increasing the magnetic-field value.

In conclusion, we measured the saturated absorption spectra of Cs D_2 line as a function of the pump beam intensity. As the pump beam intensity increased, we observed the dip in the resonance signal for the cyclic transitions, $F=4\rightarrow F'=5$ in the cases of lin∥lin and $\sigma^+\sigma^+$ configurations. Measured signals could be explained by the simple model considering the combined effects of the saturation, the velocity-selective optical pumping, and the resonant light pressure. We confirmed that the saturation effect is an important factor in sign reversal and dip even though the pump beam intensity is lower than the saturation intensity of the two-level atomic system, 10 μW/mm^2. The effect of atomic population reduction has to be considered even in the cyclic transition $F=4\rightarrow F'=5$. The reduction of effective atomic population in the cyclic transition might arise through the leakage effect by laser frequency noise.

This work was supported by the star project program, Korea Research Institute of Standards and Science, 1998. Dr. C. Oh wishes to acknowledge the financial support of Hanyang University, Korea, made in the program year of 1999.

[1] V. S. Letokov and V. P. Chebotayev, *Nonlinear Laser Spectroscopy*, Springer Series in Optics Sciences Vol. 4 (Springer-Verlag, Berlin, 1977).
[2] T. Ikegami *et al.*, Jpn. J. Appl. Phys., Part 2 **28**, L1839 (1989).
[3] T. P. Dinneen *et al.*, Opt. Commun. **92**, 277 (1992).
[4] A. Clairon *et al.*, Proc. SPIE **1837**, 306 (1992).
[5] G. Avila *et al.*, Phys. Rev. A **36**, 3719 (1987).
[6] H. S. Lee *et al.*, Metrologia **35**, 25 (1998).
[7] P. G. Pappas *et al.*, Phys. Rev. A **21**, 1955 (1980).
[8] H. Rinneberg *et al.*, Z. Phys. A **295**, 17 (1980).
[9] S. Nakayama, J. Appl. Phys. **24**, 1 (1985).
[10] R. Grimm and J. Mlynek, Appl. Phys. B: Photophys. Laser Chem. **49**, 179 (1989).
[11] O. Schmidt *et al.*, Appl. Phys. B: Lasers Opt. **59**, 167 (1994).
[12] C. H. Oh and S. Ohshima, Jpn. J. Appl. Phys., Part 1 **33**, 6350 (1994).
[13] H. S. Lee *et al.*, J. Opt. Soc. Am. B **11**, 558 (1994).
[14] H. Y. Jung *et al.*, J. Korean Phys. Soc. **1**, 3 (1998).