Optical-pumping dips in a homogeneously broadened fluorescence line

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Several authors have recently discussed and observed optical-pumping dips in inhomogeneously broadened spectral lines. We have observed a qualitatively similar, but fundamentally distinct phenomenon: optical-pumping dips in a *homogeneously* broadened D_1 line of ^{87}Rb. To our knowledge this represents the first observation of homogeneous-optical-pumping dips.

I. INTRODUCTION

Recently, there has been considerable interest in the distortion of spectral line shapes by optical pumping.[1–5] This distortion occurs not because of any light-induced alteration of the transition matrix element, as can occur in the Autler-Townes effect and similar phenomena,[6,7] but as a result of a redistribution of the atomic population density.[8] Of particular relevance to the present work are the dips which have been observed in Doppler-broadened line shapes when velocity-selective optical pumping[9] essentially "burns" a hole in the velocity distribution of a probed atomic state. In the present work we observed a qualitatively similar, but fundamentally distinct phenomenon: optical-pumping dips in a *homogeneously* broadened fluorescence line shape. To our knowledge, these experiments represent the first observations of such an effect.

II. EXPERIMENT AND RESULTS

The apparatus used in the present experiment has been described previously,[10] so that only a brief outline will be given here. Basically, the emission from a Mitusubishi TJS single-mode diode laser was split into two collimated beams having a 100:1 intensity ratio. The diode laser's output power was ~ 1 mW, and its single-mode linewidth was determined to be ~ 100 MHz. The two laser beams then intersected a rubidium atomic beam in two spatially distinct regions as illustrated in Fig. 1(a); the residual Doppler broadening of the atomic beam was ~ 40 MHz; and as illustrated in Figs. 1(b) and 1(c) ^{87}Rb atoms were optically pumped out of the $5^2S_{1/2}(F=1)$ state. The laser was tuned across the $5^2P_{1/2}(F=2)-5^2S_{1/2}(F=1)$ D_1 transition at 794.7 nm by varying the diode-laser injection current.

The residual population in the optically depleted $5^2S_{1/2}(F=1)$ state was excited downstream by the probe beam, and the induced fluorescence as a function of laser tuning was observed with a cooled RCA 31034 photomultiplier and photon-counting instrumentation. A narrow-band interference filter (1.0 nm) was used to reduce scattered room light. Since the laser linewidth was much broader than the residual Doppler broadening, the induced fluorescence spectrum was homogeneously broadened. Thus, the dip observed in the probe-induced fluorescence, illustrated in Fig. 2(a), cannot be explained by any phenomenon associated with velocity-selective optical pumping.[3–5]

An interesting feature of this homogeneous-optical-pumping dip is the asymmetry which is produced when the pump beam is angled with respect to the atomic beam. As shown in Fig. 2(b) and 2(c), the sign of the asymmetry is dependent on whether the projection of the laser-propagation direction is antiparallel or parallel to the atomic-beam velocity. When the projection is antiparallel,

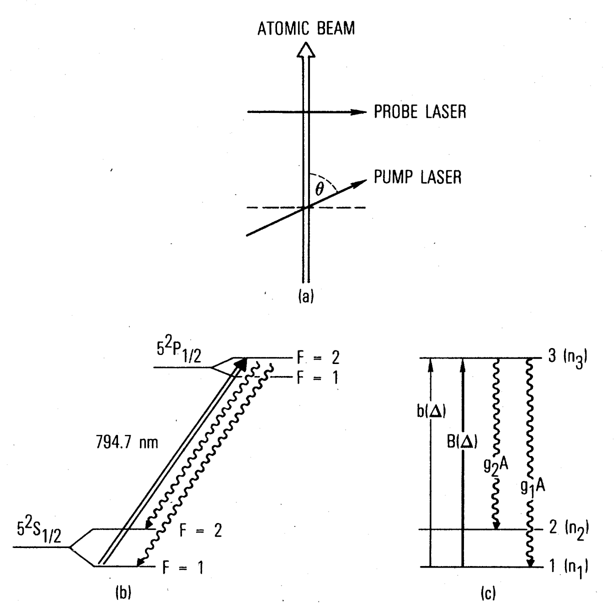

FIG. 1. (a) Schematic diagram of the experimental configuration. The pump and probe beams are spatially separated, and θ is the angle between the atomic beam and pump laser beam. (b) Energy-level diagram of ^{87}Rb showing the radiative processes of interest. The laser excites the $5^2P_{1/2}(F=2)$-$5^2S_{1/2}(F=1)$ transition; optical pumping occurs because of the $5^2S_{1/2}(F=2)$ fluorescence decay channel. (c) Simplified three-level system of ^{87}Rb; complete mixing of the Zeeman sublevels within the individual hyperfine states is assumed. A, $B(\Delta)$, and $b(\Delta)$ are the fluorescent decay rate, pump photon-absorption rate, and probe photon-absorption rate, respectively; the g_i are branching ratios.

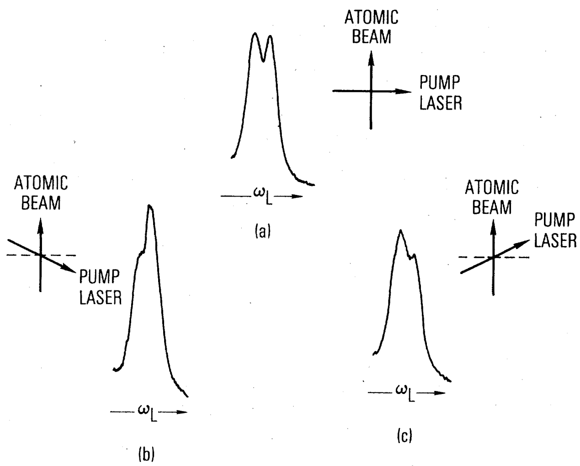

FIG. 2. Probe-induced fluorescence excitation spectrum from the depleted $5^2S_{1/2}(F=1)$ state. (a) Both pump and probe beams perpendicular to the atomic beam. (b) Pump beam angled so that the projection of its propagation direction is antiparallel to the atomic-beam velocity. (c) Pump beam angled so that the projection of its propagation direction is parallel to the atomic-beam velocity.

the dip is shifted to lower laser frequency; when the projection is parallel, the dip is shifted to higher laser frequency. This effect is very suggestive of a Doppler shift; and as we will discuss below, it is perfectly consistent with the model for the homogeneous-optical-pumping dips without velocity-selective optical pumping.

III. THEORY

The optical-pumping dips in our experiment can be adequately described by a theory of depopulation pumping by frequency-selected light.[8] Consider the three-level system illustrated in Fig. 1(c), where the n_i are normalized population densities. We define $B(\Delta)$ $[b(\Delta)]$ as the photon-absorption rate of the pump (probe) beam for a particular detuning Δ of the laser from the atomic resonance, and A as the fluorescent decay rate of level 3; the g_i are branching ratios: $g_1+g_2=1$. The fluorescent signal induced by the probe beam is proportional both to the probe photon-absorption rate and the population density in the probed state. Thus, as the pump and probe scan together across resonance, the fluorescence signal depends on the laser's detuning in two distinct ways: (1) as the probe approaches resonance the probe photon-absorption rate resonantly increases, but (2) as the pump approaches resonance the population density of the probed state can either resonantly increase or decrease depending on the state being probed. The net effect will be a distorted fluorescence line shape which can show dips if the probe couples to the optically emptied state.

In the model of the homogeneous-optical-pumping dips to be discussed, stimulated emission and coherence are ignored. The purpose of this omission does not derive from any real complexity in the calculation; rather, it is meant to clearly distinguish the homogeneous-optical-pumping dips from other types of line-shape distortion associated with high-intensity fields (for example, those of Ref. 7). Thus, from Fig. 1(c) the rate equations describing the redistribution of population due to depopulation pumping are immediately obtained:

$$\dot{n}_1(t)=-B(\Delta)n_1(t)+g_1An_3(t)\ , \tag{1a}$$

$$\dot{n}_2(t)=g_2An_3(t)\ , \tag{1b}$$

$$\dot{n}_3(t)=B(\Delta)n_1(t)-An_3(t)\ , \tag{1c}$$

where we have assumed that the probe has no effect on the redistribution of population. Taking the derivative of Eq. 1(a) and making substitutions for $\dot{n}_3(t)$ and $n_3(t)$, one arrives at a second-order differential equation with constant coefficients for $n_1(t)$. In the limit that $B(\Delta)/A \ll 1$ the solution of this equation takes on a particularly simple form,

$$n_1(t)=\tfrac{1}{2}\exp[-g_2B(\Delta)t]\ , \tag{2}$$

where $t=0$ corresponds to the time that the atom enters the pump beam.

As previously discussed, the optical-pumping dips are intimately related to the frequency dependence of the photon-absorption rate and this rate takes the form,[11]

$$B(\Delta)=\int_0^\infty \Phi(\omega_L-\omega)\sigma(\omega_0'-\omega)d\omega\ , \tag{3}$$

where $\Phi(\omega_L-\omega)$ is the spectral density of the incident light for the laser centered at ω_L, and $\sigma(\omega_0'-\omega)$ is the absorption cross section centered at ω_0' for a photon of frequency ω. In the present experiment the residual Doppler broadening of the atomic beam is much less than the homogeneous laser linewidth. Thus, for simplicity we will assume that the absorption cross section is a δ function centered on $\omega_0'=\omega_0+\vec{k}\cdot\vec{v}$, where we explicitly include a term for the Doppler shift when the laser-propagation direction is not perpendicular to the atomic beam. For a Lorentzian laser spectral profile of half-width γ,[12]

$$B(\Delta)=\frac{B_0\gamma^2}{(\Delta_0-kv\cos\theta)^2+\gamma^2}\ , \tag{4}$$

where B_0 and Δ_0 are the peak photon-absorption rate and non-Doppler-shifted detuning, respectively ($\Delta_0=\omega_L-\omega_0$).

Before proceeding to a theoretical analysis of the present experiment, and the subsequent prediction of optical-pumping dips, it is instructive to first calculate the expected signal from a different experimental design where the pump and probe beams overlap. Consider both the probe and pump beams to have the same diameter d and a monoenergetic beam of atoms traveling at a speed $\bar{v}$. For simplicity we will assume that both laser beams are perpendicular to the atomic beam. The observed fluorescence signal is then

$$S(\Delta_0)=b(\Delta_0)\int_0^\tau n_1(\Delta_0,t)dt\ , \tag{5}$$

where τ is the amount of time an atom spends in the laser beam: $\tau=d/\bar{v}$. Using Eqs. (2) and (4), we have

$$S(\Delta_0)=\frac{1}{2g_2}\left[\frac{\eta_b}{\eta_B}\right]\left[1-\exp\left[-\frac{g_2\eta_B\gamma^2}{\Delta_0^2+\gamma^2}\right]\right]\ , \tag{6}$$

where η_b and η_B are the maximum number of probe and pump photon absorptions possible: $\eta_b = b_0\tau$ and $\eta_B = B_0\tau$. It is clear from Eq. (6) that as the pump and probe scan across resonance, there will be *no* dips in the observed fluorescence signal.

The present experiment is fundamentally different from the one just described, because the pump and probe beams are spatially separated. Thus, the atoms which are probed are optically pumped atoms and not atoms in the process of being optically pumped. In the present case the observed fluorescence signal is

$$S(\Delta) = b(\Delta_0) \int_0^{\tau'} n_1(\Delta,\tau)dt' , \tag{7}$$

where the integral is over the time the atom spends in the probe beam. Again using Eqs. (2) and (4), but now allowing for the possibility that the pump beam is not perpendicular to the atomic beam, we have

$$S(\Delta) = \frac{1}{2}\frac{\eta_b\gamma^2}{\Delta_0^2+\gamma^2}\exp\left[-\frac{g_2\eta_B\gamma^2}{(\Delta_0 - kv\cos\theta)^2+\gamma^2}\right] , \tag{8}$$

where η_b is now equal to $b_0\tau'$. This signal is shown in Fig. 3 as a function of normalized detuning Δ_0/γ for various values of η_B ($\eta_b = 0.01$, $g_2 = \frac{1}{2}$, and $\cos\theta = 0$), and as can be seen, optical-pumping dips are readily obtained.

The preceding comparison of the line shapes observed for different experimental configurations illustrates quite clearly that the optical-pumping dips under discussion are not solely the result of optical pumping. Rather, their appearance is intimately related to when in the optical-pumping process observations are made. In the first type of experimental design the entire optical-pumping process is observed, from the moment the atom enters the laser beam until it exits. In the present experimental design only the end result of optical pumping is observed. This conclusion, that the optical-pumping dips are a temporal phenomenon, would seem to imply a critical value of η_B: when η_B is greater than this critical value, the atoms in the course of their flight through the pump laser beam are efficiently optically pumped; and dips are observed in the probe-induced fluorescence. For the parameters of Fig. 3 this critical value would seem to be: $1 < \eta_B^c < 3$.

The critical value of η_B can be determined quite easily by computing the "width" of the dip (i.e., the frequency separation of the double peaks). Setting the first derivative of Eq. (8) equal to zero and solving for the points of extremum, the dip width δ is found:

$$\delta = 2\gamma\sqrt{g_2\eta_B - 1} . \tag{9}$$

This dip-width formula then indicates that $\eta_B^c = 1/g_2$, so that for the parameters of Fig. 3 optical-pumping dips only occur when $\eta_B > 2$. Furthermore, the critical value of η_B determined by Eq. (9) is consistent with the critical value of η_B necessary for efficient optical pumping, as can be seen by inspection of Eq. (2) and Fig. 1(c).

If the pump beam is not exactly perpendicular to the atomic beam, or more to the point, if the probe and pump laser beams are not parallel, because of the Doppler shift the atoms will respond as if there was a frequency offset between the pump and probe. Thus, when the optical pumping is most efficient, the probe-photon-absorption rate will not be at its resonant peak. This Doppler-shift effect is responsible for the asymmetry observed in the experimental line shapes when the pump laser beam is angled slightly. The theoretical predictions of this asymmetry are shown in Figs. 4(a) and 4(b) for $\theta = 95°$ and 85°, respectively, and it should be noticed that the sign of the asymmetry is consistent with the experimental observations. This predicted asymmetry lends strong support to the validity of the preceding theory.

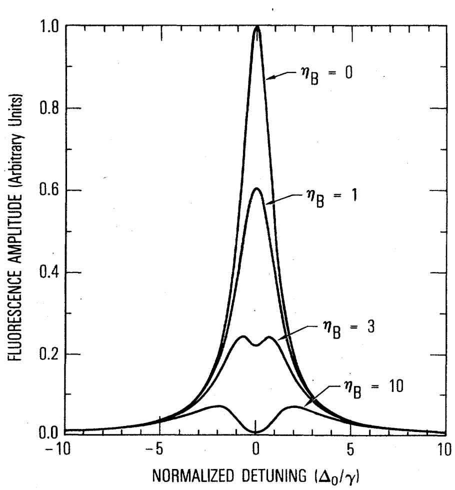

FIG. 3. Predicted probe-induced fluorescence line shapes as a function of normalized detuning for various values of η_B; the probe and pump have the same detuning Δ_0. ($\eta_b = 0.01$ and $g_2 = \frac{1}{2}$.)

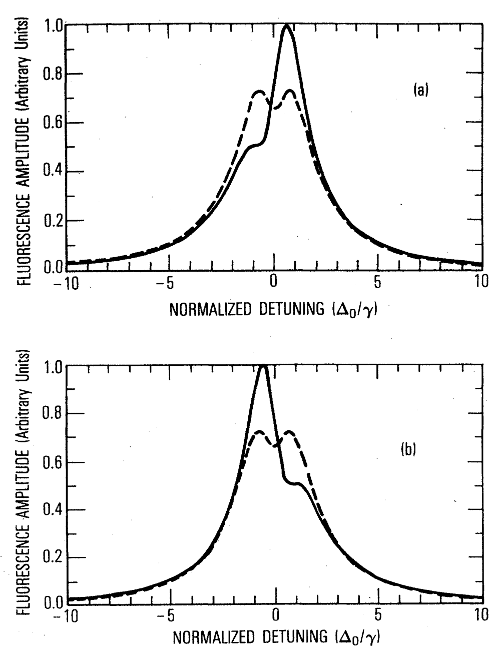

FIG. 4. Predicted asymmetry of the dip with the pump beam angled slightly: (a) $\theta = 95°$ and (b) $\theta = 85°$. Dashed curve corresponds to the line shape obtained when $\cos\theta = 0$. ($\eta_B = 3$ and $\eta_b = 0.01$.)

It should not, however, be assumed that the prediction of the dips is necessarily associated with a transient description of optical pumping. In the present experiment one could just as easily obtain the same results by solving the optical-pumping equations in steady state, using a phenomenological relaxation rate to account for the finite interaction time of the atoms with the laser. However, in this description it is necessary to realize that there will be a population gradient along the optical-pumping beam diameter; and that this implies a z dependence for the phenomenological relaxation rate, where z denotes the radial position in the pump beam: $0 \le z \le d$. In this model the optical-pumping equations become

$$\dot{n}_1(z)=0$$

$$=-B(\Delta)n_1(z)+g_1An_3(z)-\bar{v}\frac{dn_1(z)}{dz}\ , \tag{10a}$$

$$\dot{n}_2(z)=0=g_2An_3(z)-\bar{v}\frac{dn_2(z)}{dz}\ , \tag{10b}$$

$$\dot{n}_3(z)=0=B(\Delta)n_1(z)-An_3(z)-\bar{v}\frac{dn_3(z)}{dz}\ , \tag{10c}$$

which are formally equivalent to Eqs. (1a)–(1c).

IV. DISCUSSION

In conclusion, we note that the dip width governed by Eq. (9) can in principle go to zero, which suggests the possibility of observing very narrow spectral features. In practice, however, the minimum attainable width will be limited by the minimum observable dip amplitude. Thus, the observation of extremely narrow dips is essentially a problem of sensitivity and not instrument resolution. As an example, consider a laser linewidth narrower than the absorption line's natural width, and a highly collimated atomic beam. Under these conditions the resonance width of the photon-absorption rate would be determined by the natural absorption linewidth. In order to obtain a dip width 30% narrower than the natural linewidth, Eqs. (8) and (9) indicate that the dip to peak amplitude ratio would be 0.7%. This sensitivity is certainly within the realm of possibilities; and thus implies that the homogeneous-optical-pumping dips could find an application in high resolution, possibly even subnatural, spectroscopy.

ACKNOWLEDGMENT

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