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Saturation of an atomic transition by a phase-diffusing laser field

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We have studied the effect of well-characterized laser frequency fluctuations on the saturation of an atomic resonance in a double optical resonance experiment. The peak-height asymmetry of the observed Autler-Townes signal was reversed at small detunings when the shape of the saturating laser power spectrum was changed from nearly Gaussian to nearly Lorentzian. This behavior agrees qualitatively with theoretical calculations and also with previous observations using lasers without such well-characterized fluctuations.

We report here an experimental investigation of the effect of optical fluctuations on the saturation of an atomic transition, examined by means of the Autler-Townes effect. In this double optical resonance (DOR) process (see Fig. 1) two states $|1\rangle$ and $|2\rangle$ are coupled by an intense laser field. When one probes the system with a *weak* laser field which couples $|2\rangle$ to a third level $|3\rangle$, one sees a double-peaked structure in the absorption spectrum of the weak field. This is called the (optical) Autler-Townes effect. The absorption peaks correspond to excitation from the ac Stark-split components of $|2\rangle$.

Early investigations of this effect which used relatively broad-band pulsed lasers showed a reversal in the peak-height asymmetry[1] in comparison to that predicted by theories which considered the laser fields to be monochromatic.[2] This asymmetry reversal was observed at small detunings of the saturating laser from the unperturbed transition frequency; at larger detunings the asymmetry reverted to "normal," i.e., that expected from the monochromatic theory. Furthermore, an experiment[3] done with a narrow-band cw saturating laser showed only normal asymmetry. Laser bandwidth effects were further demonstrated in a DOR experiment using a pulsed dye laser which could be made either relatively narrow or broad band.[4] Narrow-band excitation gave normal asymmetry in contrast to the reversed asymmetry seen with broad-band excitation. Subsequently Dixit, Zoller, and Lambropoulos[5] employed a generalization of the phase-diffusion model (PDM) to calculate DOR signals and found that the same behavior could occur, i.e., reversed asymmetry for small saturating laser detunings and normal for large detunings. Although these experimental and theoretical results were in good qualitative agreement, quantitative comparisons were not possible because of the differences between the field models on which theories are based and the laser fields normally produced in the laboratory. For example, pulsed lasers have amplitude fluctuations as well as frequency fluctuations which cannot be completely characterized statistically, and can only be controlled to a limited degree. We have constructed a laser field[6] which is described by the generalized PDM used by Dixit *et al.* and have repeated the DOR experiment using this idealized broadened laser spectrum for direct comparison with theoretical calculations.

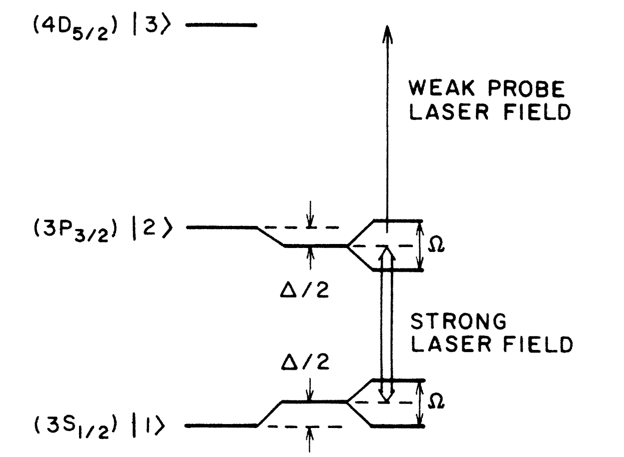

FIG. 1. The energy levels involved in our double optical resonance experiment. The ac Stark-split components of $|1\rangle$ and $|2\rangle$ are separated by the generalized Rabi frequency Ω. The corresponding sodium levels are in parentheses.

The properties of the generalized PDM and its production have been presented elsewhere,[5,6] but for clarity of presentation we will review the main points here. The field is characterized by

$$E(T)=\tfrac{1}{2}(E_0e^{-i[\omega_0t+\phi(t)]}+\text{c.c.}) \ , \qquad (1)$$

where E_0 is the constant field amplitude and ω_0 the mean (angular) frequency. The phase $\phi(t)$ is a Gaussian variable related to the frequency fluctuation $\omega(t)$, by $\omega(t)=d\phi/dt$. The frequency is stationary in the statistical sense. The correlation function of the frequency fluctuations is

$$\langle\omega(t)\omega(t+\tau)\rangle=b\beta e^{-\beta|\tau|} \ , \qquad (2)$$

where b is the spectral density of the fluctuations and $1/\beta$ is the correlation time of the fluctuations. The angular brackets denote an ensemble average. The two parameters, b and β, and the Gaussian nature of $\omega(t)$ completely define the statistical properties of the laser field.

The power spectrum of the laser field is given by

$$P(\Delta\omega)=\tfrac{1}{2}|E_0|^2\int_0^\infty d\tau\cos(\Delta\omega\tau)\times\exp\{-b[\tau+1/\beta(e^{-\beta\tau}-1)]\} \ . \qquad (3)$$

When the fluctuations of the frequency are allowed to be very fast, $\beta>>b$, $P(\Delta\omega)$ tends to a Lorentzian of half-width at half maximum (HWHM) b, but falls off more rapidly than a Lorentzian in the wings. In the limit as $\beta \rightarrow \infty$, $P(\Delta\omega)$ becomes exactly Lorentzian. If the fluctuations are limited to being very slow, $b >> \beta$, $P(\Delta\omega)$ is similar to a Gaussian function of HWHM $[(2\ln 2)b\beta]^{1/2}$. Strictly speaking, the phase does not diffuse in this case, but we will still consider this to be part of the PDM. The method of producing a phase-diffusing field is to use a statistically well-characterized random voltage to modulate the frequency and phase with wide bandwidth acousto-optic (AOM) and electro-optic (EOM) modulators.

We chose the sodium $3S_{1/2} \rightarrow 3P_{3/2}$ and $3P_{3/2} \rightarrow 4D_{5/2}$ transitions with which to do the experiment (see Fig. 1). The first transition was saturated with the randomly modulated laser and the weak probe-laser beam was tuned through the second transition. Figure 2 shows a schematic of our experiment. We measured the width of the $3S_{1/2} \rightarrow 3P_{3/2}$ transition to be 12.0 ± 0.5 MHz where the natural width is 10 MHz.

Before entering the experimental region the atoms were optically pumped by a circularly polarized laser beam tuned to the $F=2 \rightarrow F=3$ hyperfine component of the $S_{1/2} \rightarrow P_{3/2}$ transition. The atoms were thus prepared in the $3S_{1/2}(F=2, m_F=2)$ state. Because subsequent excitation is with purely σ^+ circularly polarized light to the $3P_{3/2}(F=3, M_F=3)$ state, we have an effective two-state transition.[7] In fact, the optical pumping is done with a frequency sideband which is imposed on an unmodulated beam from the same dye laser which is input to the noise-modulation apparatus. By dithering the frequency of the sideband and monitoring the intensity of the fluorescence from the optical pumping region, we lock the sideband to the transition frequency using a lock-in amplifier. The feedback signal went to the external scan input of the dye laser, thus stabilizing the saturating frequency at a given detuning. The frequency of the signal which produced the optical pumping sideband was varied about the value of the frequency offset produced by the AOM in the random modulation in order to scan the saturating laser frequency. The average intensity of light in the sideband which did the optical pumping was about 100 mW/cm^2. A magnetic field of 650 mG, parallel to the direction of the laser beams, was present to maintain the orientation of the atoms.

The saturating and probe beams were combined using an uncoated beam splitter and were collinear in the interaction region. The radius of the saturating beam was 0.6 mm and the probe beam was focused to a 0.13-mm radius. The Rabi frequency of the atoms was thus nearly constant across the diameter of the probe beam. Both probe and saturating beams were circularly polarized by a linear polarizer and a Fresnel rhomb. The absorption spectrum of the probe beam was measured by scanning the probe frequency and detecting fluorescence at 330 nm which resulted from decay of the $4D$ level by way of the $4P$ level. The intensity of this fluorescence was proportional to the population of the $4D$ level. A fused silica lens placed near the interaction region collected fluorescence and directed it to a photomultiplier through a bandpass (330 nm) interference filter.

The peak intensity at the center of the saturating beam was ~ 1 W/cm^2 and the (average) probe intensity was ~ 10 mW/cm^2. The scan of the probe beam frequency was calibrated by sending a portion of this light through a near-confocal Fabry-Perot which gave frequency marker peaks at

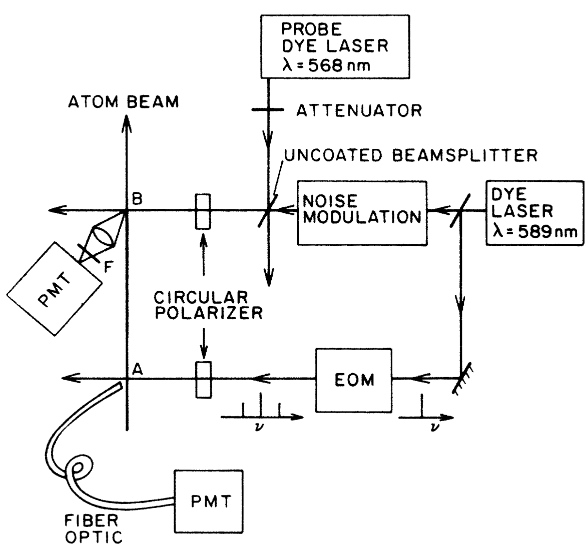

FIG. 2. Schematic of the experiment. A denotes the optical-pumping region and B the saturation region. The EOM produces the sideband used for optical pumping. F denotes a bandpass (330 nm) interference filter.

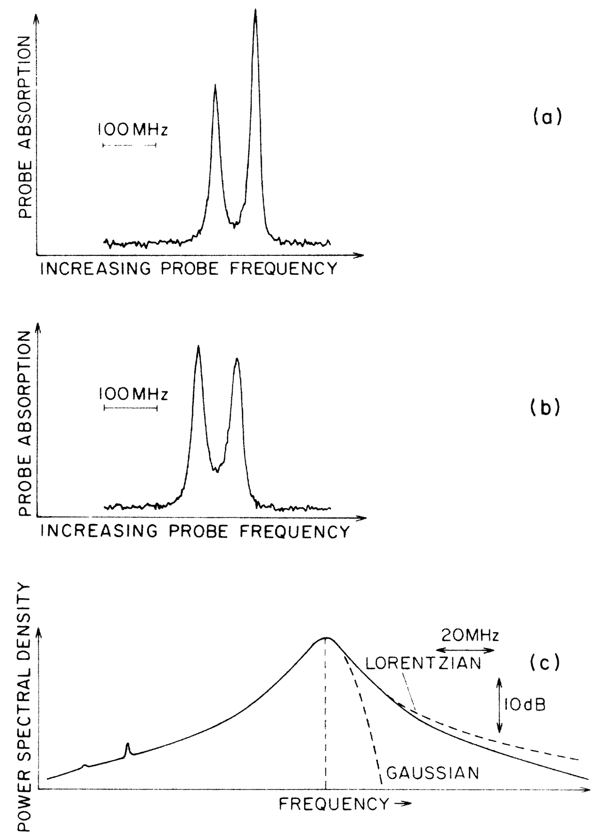

FIG. 3. The probe laser absorption spectrum for (a) a monochromatic saturating laser and (b) a saturating laser with a nearly Lorentzian-shaped power spectrum (c) with HWHM of 7 MHz. In (c) the subsidiary peaks on the left are frequency components on the local oscillator beam used in the heterodyne detection of this spectrum. Thus they can be ignored. Note that the vertical scale is logarithmic. The Lorentzian and Gaussian curves drawn for comparison also have HWHM of 7 MHz.

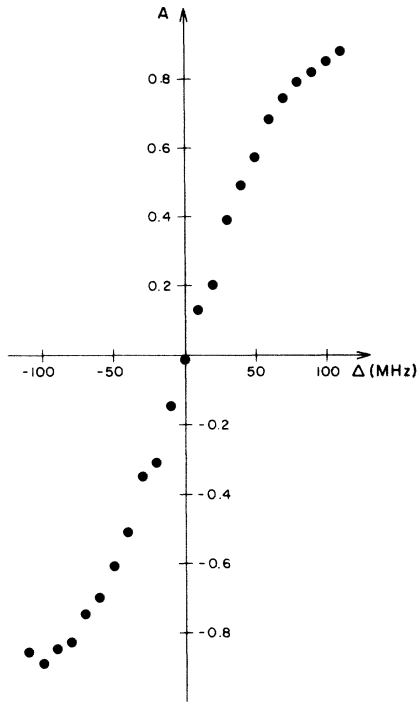

FIG. 4. A plot of asymmetry parameter A vs saturating laser detuning. The Rabi frequency (at zero detuning) is 67 MHz.

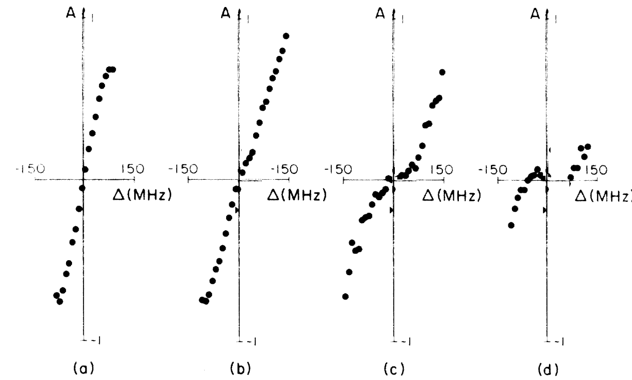

FIG. 5. Graphs of asymmetry parameter vs saturating laser detuning for four nonmonochromatic laser power spectra. The Rabi frequency is as for Fig. 4. (a) $\beta/2\pi = 10$ MHz; (b) $\beta/2\pi = 30$ MHz; (c) $\beta/2\pi = 60$ MHz; (d) $\beta/2\pi = 80$ MHz.

25.1 MHz (transverse mode) and 339.1 MHz (free spectral range). To improve the signal-to-noise ratio the probe beam was chopped at about 400 Hz and a lock-in amplifier was used to measure the photocurrent from the photomultiplier tube monitoring the 330-nm fluorescence. A Keithley model 610 electrometer served as a preamplifier for the photocurrent.

Figure 3 shows two examples of DOR signals that were obtained: In Fig. 3(a) the saturating laser was not modulated with noise and had residual frequency fluctuations of about 150 kHz rms. For Fig. 3(b) the saturating laser had the power spectrum shown in 3(c). This example clearly shows the reversal of asymmetry for a saturating power spectrum which approaches a Lorentzian, i.e., $\beta >> b$. The Rabi frequency for both 3(a) and 3(b) is 67 MHz, as determined from the minimum separation of the Autler-Townes peaks and independently from measurement of the laser power and intensity profile of the beam. The saturating laser frequency is, in both cases, 10 MHz less than the unperturbed atomic transition frequency. What is of interest to us in this work is the asymmetry in the peak heights. If we define an asymmetry parameter $A = (h_u - h_l)/(h_u + h_l)$, where h_u (h_l) is the height of the peak at the higher (lower) probe frequency, we can plot A versus the saturating laser detuning $\Delta = \nu_{atom} - \nu_{laser}$. In Fig. 4 we show experimentally measured asymmetry parameters as a function of the (monochromatic) saturating laser detuning. The sideband separation in the optical pumping beam is not, in general, exactly equal to the frequency offset produced by the noise modulation when $A = 0$. This is due to errors ($\sim \pm 2$ mrad) in the alignment of the laser beams with respect to each other and with respect to the atomic beam. For this reason the zero detuning points in Figs. 4 and 5 are determined from the $A = 0$ point in the DOR signals taken with the "monochromatic" saturating laser.

Figure 5 shows a series of four graphs of the asymmetry parameter measured using nonmonochromatic saturating laser power spectra. The only parameters that differ between 5(a)–(d) are b and β. We have adjusted b in each case to give a constant HWHM of the laser power spectrum, i.e., 7 MHz. This figure shows clearly the development of asymmetry reversal as one increases β for a constant HWHM and the shape of the power spectrum changes from Gaussian to Lorentzian. Indeed the data for $\beta/2\pi = 10$ MHz are indistinguishable from that for the monochromatic laser. These data agree qualitatively with the published theory of Dixit *et al.*[5] The same authors are currently modifying their theory to take our experimental conditions into account. This will allow a proper quantitative comparison between theory and experiment.

Analysis of further experimental results is currently under way and will be reported in a subsequent publication. For more extreme experimental parameters some anomalies appear, which may imply that the simple two-level atom model is beginning to break down.

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[1]S. E. Moody, Ph.D. thesis, University of Colorado, 1971 (unpublished); S. E. Moody and M. Lambropoulos, Phys. Rev. A 15, 1497 (1977); P. B. Hogan, S. J. Smith, A. T. Georges, and P. Lambropoulos, Phys. Rev. Lett. 41, 229 (1978).

[2]For example, see P. L. Knight and P. W. Milonni, Phys. Rep. **66**, 21 (1980).

[3]H. R. Gray and C. R. Stroud, Jr., Opt. Commun. **25**, 359 (1978).

[4]D. E. Nitz, A. V. Smith, M. D. Levenson, and S. J. Smith, Phys. Rev. A **24**, 288 (1981).

[5]S. N. Dixit, P. Zoller, and P. Lambropoulos, Phys. Rev. A **21**, 1289 (1980).

[6]D. S. Elliott, M. W. Hamilton, K. Arnett, and S. J. Smith, Phys. Rev. A **32**, 887 (1985); D. S. Elliott, R. Roy, and S. J. Smith, *ibid.* **26**, 12 (1982); in *Spectral Line Shapes,* edited by K. Burnett (de Gruyter, Berlin, 1983), Vol. 2, p. 989.

[7]R. E. Grove, F. Y. Wu, and S. Ezekiel, Phys. Rev. A **15**, 227 (1977).