

## OBSERVATION OF A SPECTRAL HOLE DUE TO POPULATION OSCILLATIONS IN A HOMOGENEOUSLY BROADENED OPTICAL ABSORPTION LINE

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Received 7 February 1983

We have observed a dip in the absorption profile of the homogeneously broadened green absorption band of ruby for frequencies near that of a saturating, cw laser beam. The shape and depth of the hole thus produced have been measured using weak amplitude modulation sidebands as a probe, and are found to be in good agreement with theoretical predictions. These predictions are based on a model that ascribes the origin of the hole to the periodic modulation of the ground state population at the beat frequency between the pump and probe beams. The theory includes the effect of pump depletion. Holes as narrow as 37 Hz (HWHM) have been observed.

An intense, monochromatic laser beam can partially saturate an inhomogeneously broadened absorption line, leading to a well-known dip in the absorption profile at the laser frequency [1]. It is commonly believed that the existence of such a dip is not possible for the case of a homogeneously broadened absorption line [2]. However, on the basis of solutions of the density matrix equations of motion, Schwartz and Tan [3] predicted in 1967 that a strong pump wave can cause a spectral "hole" to appear when probed by a second wave, even though the broadening mechanism is conventionally homogeneous. In addition, a number of more recent papers [4–6] have shown that this hole is produced as the result of the periodic modulation of the ground state population at the beat frequency between the pump and probe beams. The population can follow the oscillations of the optical intensity only if the beat frequency is less than or approximately equal to the inverse of the population relaxation time, and in this case population oscillations can lead to a decreased absorption of the probe beam. This effect can occur either for a two-level system or under appropriate circumstances for more complicated multilevel systems. The relation between this effect and the dynamic Stark effect has been discussed by Sargent [4]. Furthermore, it has been suggested that the existence of such a hole might be the reason why single-mode homogeneously broadened lasers

are unstable far above threshold [3,7]. It has also been suggested that this effect provides the gain necessary for optical bistability and might also produce further instabilities and chaos [8].

We have carried out an experiment in ruby at room temperature that displays a spectral hole in a homogeneously broadened optical absorption line. An argon ion laser operating at 514.5 nm is resonant from the ground state to the broad  ${}^4F_2$  absorption band of ruby. The population in this band decays very rapidly to the  $2A$  and  $E$  levels where it is trapped due to the long lifetimes of these levels before it decays back to the ground state via the  $R_1$  and  $R_2$  lines and via vibronic transitions [9]. The homogeneously broadened absorption band has a width of  $\sim 100$  nm which corresponds to a  $T_2$  dephasing time of a few fs but a  $T_1$  time for return to the ground state of a few ms. Theory predicts that a weak probe beam should see a hole in the absorption band of width  $\sim (T_1)^{-1}$  centered at the pump laser frequency. We observed a 37 Hz wide (HWHM) hole in the probe absorption profile. To our knowledge it is the narrowest spectral feature observed using laser spectroscopy [10]. Usually optical hole burning experiments are conducted using separate pump and probe lasers. However, in our case the expected line width of the hole was too narrow to allow the use of this approach, and hence we created the probe field as weak

frequency sidebands on the pump laser using amplitude modulation. The spectral hole phenomenon reported here shares a common origin, (namely, population oscillations) with the radio frequency experiments reported by Senitzky et al. [11] and with the sharp resonances in the four-wave mixing susceptibility observed by Song et al. [12] and by Bogdan et al. [13].

In a recent paper we have developed a theory that is capable of treating the propagation of a weak probe beam in a two-level medium that is saturated by a strong resonant pump beam [6]. It was shown that the probe field is properly described by a set of bichromatic natural modes. The natural modes of the probe are phased combinations of two sidebands symmetrically displaced from the saturating field. In the case of a pump beam tuned to resonance, one natural mode is purely amplitude modulated (AM) while the other is purely frequency modulated (FM). If the dipole dephasing time,  $T_2$ , is much less than the population decay time,  $T_1$ , then the absorption profile seen by the weak probe field is identical to that seen by the saturating field except when the frequency difference between the two fields is small. In this case, there is a decrease in absorption for the AM natural mode. This spectral hole was a width of the order of  $(T_1)^{-1}$  which is much narrower than  $(T_2)^{-1}$ .

The intensity of the amplitude modulated applied field may be written as

$$\begin{aligned} \mathcal{G}(t) &= (c/4\pi) [\mathcal{E}_0^2 + 2\mathcal{E}_0 \delta \mathcal{E}_1 \cos \delta\omega t] \\ &= \mathcal{G}_0 + \delta \mathcal{G}_1 \cos \delta\omega t, \end{aligned} \quad (1)$$

where  $\mathcal{E}_0$  is the amplitude of the strong pump beam,  $\delta \mathcal{E}_1$  is the amplitude of the AM natural mode probe beam, and  $\delta\omega$  is the detuning of the probe frequency from the pump frequency. The response of the ruby to the applied field is calculated by solving the density matrix equations of motion for the three-level ruby system. The spatial variation of the pump field due to absorption in the ruby is then given by

$$(\partial/\partial z)I_0(z) = -\alpha I_0(z)[1 + I_0(z)]^{-1}, \quad (2)$$

where we have introduced the dimensionless intensity of the pump field

$$I_0 = \mathcal{G}/\mathcal{G}_{\text{sat}} = \kappa^2 T_1^2 T_2 \mathcal{E}_0^2.$$

The constant  $\alpha$  is the unsaturated reciprocal absorption length and  $\kappa = 2d/\hbar$  where  $d$  is the transition dipole moment. The two decay times in the three-level cascade system of ruby satisfy the inequality  $T_2 \ll T_1$  where  $T_1 = 2T_1'$  is the relaxation time of the population from the intermediate level to the ground state<sup>‡</sup>. The saturation intensity  $\mathcal{G}_{\text{sat}}$  has previously been determined to have the value  $1.5 \text{ kW/cm}^2$  for excitation at  $5145 \text{ \AA}$  [14]. For a ruby crystal of length  $l$ , eq. (2) may be integrated exactly giving the intensity  $I_0(l)$  at the exit face in terms of the intensity  $I_0(0)$  at the entrance face:

$$\ln(I_0(l)/I_0(0)) + I_0(l) - I_0(0) = -\alpha l. \quad (3)$$

The spatial variation of the probe amplitude in the ruby is similarly obtained and is given by

$$\begin{aligned} \frac{\partial}{\partial z} \delta \mathcal{E}_1 &= -\frac{1}{2} \frac{\alpha \delta \mathcal{E}_1}{1 + I_0(z)} \\ &\times \left[ 1 - \frac{2I_0(z)[1 + I_0(z)]}{[1 + I_0(z)]^2 + (\delta\omega T_1)^2} \right]. \end{aligned} \quad (4)$$

Eq. (4) may be integrated exactly using eq. (2) yielding a relation between the entering and exiting probe amplitudes in terms of the entering and exiting pump intensities determined by eq. (3):

$$\begin{aligned} \ln(\delta \mathcal{E}_1(l)/\delta \mathcal{E}_1(0)) &= \frac{1}{2} \ln(I_0(l)/I_0(0)) \\ &- \frac{1}{2} \ln \left[ \frac{[1 + I_0(l)]^2 + (\delta\omega T_1)^2}{[1 + I_0(0)]^2 + (\delta\omega T_1)^2} \right]. \end{aligned} \quad (5)$$

In our experiment the output of an amplitude stabilized, multimode argon ion laser operating at  $514.5 \text{ nm}$  was amplitude modulated and sent through a  $10 \text{ mm}$  long ruby crystal as shown in fig. 1. Amplitude modulation was achieved by passing the laser beam through an electro-optic crystal followed by a Glan-Thompson polarizer. The modulation depth

<sup>‡</sup> The dynamics of the three-level cascade system of ruby may be reduced to the dynamics of a two-level system, because of the rapid decay from the absorption band to the intermediate level. The time  $T_1$  associated with the saturation intensity is no longer the population relaxation time  $T_1$ . This difference has previously been discussed by M. Sargent III (see. ref. [4]).

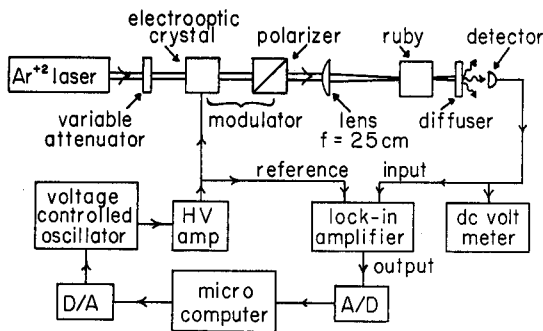


Fig. 1. The experimental setup.

was about 5%. This modulated beam was focused into the ruby crystal by a lens with a focal length of 25 cm producing a confocal parameter greater than the length of the ruby. The transmitted light was diffused by a piece of ground glass and detected by a photodiode. The detected signal was sent to a digital dc voltmeter and to a lock-in amplifier, the reference signal for which was supplied by the amplifier that drove the electro-optic crystal. The output of the lock-in amplifier was proportional to the modulated component of the total intensity.

The experiment was controlled by a microcomputer that set the modulation frequency and monitored the output of the lock-in amplifier. This output signal was sampled  $\sim 300$  times at each frequency and these values were subsequently averaged. Input laser powers ranged from 25 mW to 1.5 W which correspond to intensities in the crystal from 0.1 kW/cm<sup>2</sup> to 6 kW/cm<sup>2</sup> for our 0.18 mm diameter laser beam. In addition, calibration scans were taken without the ruby to determine the frequency response and zero-offsets of the system.

In fig. 2, we plot the attenuation experienced by both the unmodulated and (2 kHz) modulated components of the laser beam as a function of the power of the incident beam. Here attenuation is defined as the natural logarithm of the ratio of incident intensity  $\mathcal{I}(\text{in})$  to transmitted intensity  $\mathcal{I}(\text{out})$ , including both nonlinear absorption and linear losses such as surface reflections and scattering. The attenuation for the unmodulated and (2 kHz) modulated components is found to be the same, as expected from theory since the modulation frequency is much greater than  $(T_1)^{-1}$ . The data were fit by the functional form,

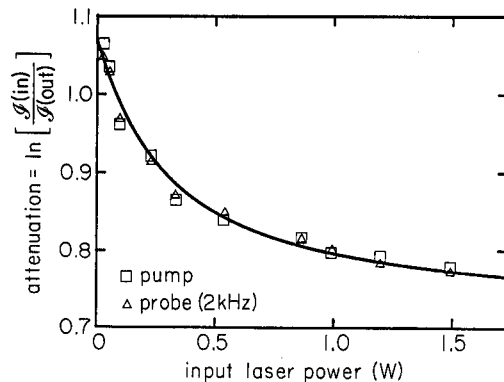


Fig. 2. Attenuation of the unmodulated (pump) beam and 2-kHz-modulated (probe) beam is plotted as a function of laser power.

$$\ln(\mathcal{I}(\text{out})/\mathcal{I}(\text{in})) + a\mathcal{I}(\text{out}) - b\mathcal{I}(\text{in}) = c, \quad (6)$$

where the three parameters,  $a$ ,  $b$  and  $c$  were obtained by a least squares analysis. For our data,  $c = -1.07 \pm 0.02$  is the total unsaturated loss coefficient from the crystal faces. The unsaturated absorption coefficient of the ruby crystal is found to be  $aI = \ln(b/a) - c = 0.355 \pm 0.03$ .

The attenuation of the modulated component of the incident beam is plotted in fig. 3 as a function of the modulation frequency. A pronounced dip is observed at low modulation frequency, as expected. To compare our measured line shapes with theoretical predictions, we make use of the fact that the intensity of the modulated component satisfies the equation

$$\ln(\delta\mathcal{I}(\text{out})/\delta\mathcal{I}(\text{in})) = \ln(\mathcal{I}(\text{out})/\mathcal{I}(\text{in})) - \frac{1}{2} \ln \left[ \frac{[1 + a\mathcal{I}(\text{out})]^2 + (\delta\omega T_1)^2}{[1 + b\mathcal{I}(\text{in})]^2 + (\delta\omega T_1)^2} \right]. \quad (7)$$

The theoretical curves shown in fig. 3 were obtained from this equation using the previously determined values of  $a$ ,  $b$  and  $c$  and using the value  $T_1 = 4.3$  ms which gave the best fit to the data. No free parameters other than  $T_1$  were used.

The measured line shape is in good agreement with our theoretical model over a broad range of laser intensities. At low laser powers, the linewidth (HWHM) is 37 Hz. At large laser intensity, significant power broadening of the spectral hole is observed, as predict-

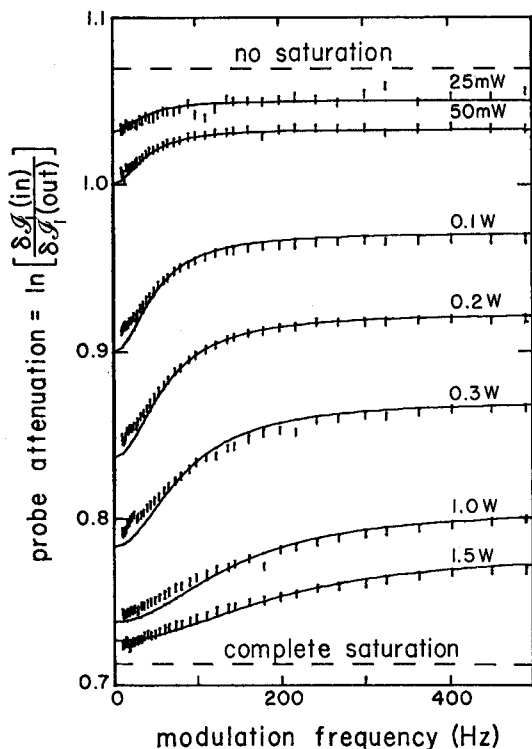


Fig. 3. Attenuation of the modulated component (probe beam) is plotted as a function of modulation frequency. The probe beam experiences decreased absorption at low modulation frequencies. The width of this hole is 37 Hz for low laser powers. The spectral hole is power broadened at high laser powers.

ed by eq. (7). The small discrepancies between theory and experiment are perhaps due to the plane-wave approximation used in our theory as well as experimental difficulties involved in cancelling the noise at low frequencies. The measured value of the fluorescent decay time for our ruby sample is  $3.4 \pm 0.1$  ms. However, our experiment measures the effective population relaxation rate to the ground state, whose inverse can differ from the radiative decay time of the fluorescing level due to the possibility of complicated dynamics of the decay involving more than two levels. Such an effect is not unlikely since the room temperature quantum efficiency of the R line is about 50% [9].

In our experiment, the laser was operating multi-mode with a total bandwidth spanning several GHz. A hole is produced at each mode frequency, and amplitude modulation of the laser produces sidebands about each such mode. A bandwidth is associated with each laser mode due to frequency modulation noise. Our technique is sensitive only to amplitude modulation and hence can achieve resolution not only better than the laser bandwidth but even better than the bandwidth of each laser mode.

The authors gratefully acknowledge useful discussions of these effects with M.A. Kramer, M.G. Raymer, M. Sargent, and A. Szabo, and the technical assistance of R.J. Smith. R.W. Boyd acknowledges partial support by the National Science Foundation, and C.R. Stroud by the U.S. Army Research Office.

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