Reprinted from Journal of the Optical Society of America B, Vol. 1, page 73, March 1984
Copyright © 1984 by the Optical Society of America and reprinted by permission of the copyright owner.

Saturation and inverse-saturation absorption line shapes in alexandrite

Michelle S. Malcuit, Robert W. Boyd, Lloyd W. Hillman, Jerzy Krasinski,* and C. R. Stroud, Jr.

The Institute of Optics, University of Rochester, Rochester, New York 14627

Received June 15, 1983; accepted November 9, 1983

Through the use of modulation spectroscopy we have measured the absorption spectrum experienced by a weak probe beam in the presence of a saturating pump beam at two different wavelengths within the broad homogeneous absorption band of alexandrite. At 584 nm the absorption saturates at high pump intensities, and a hole (of width 612 Hz) appears at frequencies near that of the pump laser. However, at 457 nm the absorption demonstrates the inverse of normal saturation because of strong excited-state absorption, and an antihole appears at the pump frequency. These results are in good quantitative agreement with the predictions of a model that ascribes the origin of these spectral features to the temporal modulation of the ground-state population at the beat frequency between the pump and the probe beams. These population oscillations are treated properly within a rate-equation approximation in which the amplitude modulation of the total optical intensity caused by the beating of the pump and the probe beams is taken into account.

Pump–probe techniques are commonly used in nonlinear optics to monitor the level of saturation induced in a material system by a strong optical field. A common example of the use of this technique is spectral-hole-burning studies of inhomogeneously broadened optical absorption lines. In such experiments, a strong, narrow-band laser field saturates one segment of the inhomogeneous absorption profile (e.g., one velocity group for the case of Doppler broadening), leading to decreased absorption of the probe beam when its frequency is near that of the pump.

A spectral hole that is due to this mechanism cannot exist for homogeneously broadened media since all the atoms experience the same level of saturation. However, it has been pointed out by several authors[1–3] that another mechanism exists that can also lead to a dip in the probe absorption profile at the pump frequency, even for homogeneously broadened media. This mechanism involves the periodic modulation of the ground-state population at the beat frequency between the pump and the probe fields. It has been shown[3] that these population oscillations give rise to a dip in the probe absorption profile whose width in the limit of low saturation is equal to the inverse of the ground-state recovery time T_1.

The first experimental observation of such a spectral hole in an essentially homogeneously broadened medium was recently reported by Hillman *et al.*[4] In this experiment, an argon-ion laser was used to saturate partially the broad green absorption band of ruby. A narrow hole of breadth ~50 Hz (corresponding to the inverse of the fluorescent decay time of the ^2E levels) was observed at the laser frequency. Even for an inhomogeneously broadened transition, the probe absorption line shape will be altered by the modulation of the ground-state population. Probe-beam absorption line shapes have been measured under such conditions by Keilmann[5] in p-type germanium.

In this paper we present the results of an experimental study of saturated absorption line shapes for the case of alexandrite. These results are qualitatively different from those obtained in ruby because of the influence of strong excited-state absorption in alexandrite. Figure 1 shows an energy-level diagram showing the alexandrite levels relevant to our experiment. Following optical excitation to the phonon-broadened $^4T_1$ or $^4T_2$ levels, population decays rapidly to the ^2E level, where it is trapped because of the long (260-μsec) fluorescent lifetime of these levels. Shand *et al.*[6] have shown that, for excitation wavelengths in the approximate range 450–510 nm, the cross section σ_2 for absorption out of the ^2E excited state exceeds that of the ground state σ_1. The absorption rate for excitation at these wavelengths thus shows the inverse of normal saturation in that the *total* absorption coefficient increases rather than decreases with laser intensity. A consequence of this inverse saturation is that an antihole rather than a hole is produced in the probe absorption spectrum at the frequency of the pump laser.

A theoretical prediction of the probe-beam absorption line shape can be obtained by treating the alexandrite system in the rate-equation approximation. This approximation is expected to be reliable since the dipole dephasing time T_2 is much smaller than the ground-state recovery time T_1 or the inverse of the pump–probe detuning Δ. By assuming the relaxation scheme shown in Fig. 1, the rate equation for the ground-state population density n is given by

$$\frac{\mathrm{d}n}{\mathrm{d}t} = -\frac{n\sigma_1 I_{\mathrm{tot}}(t)}{\hbar\omega} + \frac{(\bar{n} - n)}{T_1}, \tag{1}$$

where $\bar{n}$ is the total density of absorbers and $\hbar\omega$ is the energy of a pump photon. The optical field is assumed to be in the $+z$ direction, and the z dependence of the various quantities has been supressed in our notation. The instantaneous intensity of the total optical field is taken to be

$$I_{\mathrm{tot}}(t) = I_0 + [I(\Delta)e^{-i\Delta t} + \mathrm{c.c.}]. \tag{2}$$

The time-varying part $I(\Delta)$ can either be due to the beating of distinct pump and probe fields detuned by a frequency Δ or can be created by passing an amplitude-stable laser beam through an amplitude modulator operating at frequency Δ.

0740-3224/84/010073-03$02.00 © 1984 Optical Society of America

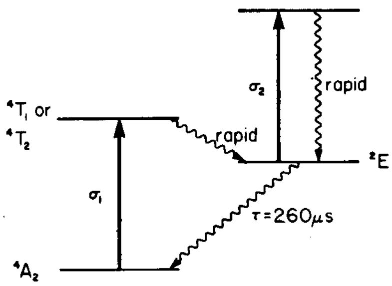

Fig. 1. Energy-level diagram showing relevant decay routes in alexandrite. The ground-state and excited-state absorption cross sections are denoted σ_1 and σ_2, respectively.

The steady-state (i.e., stationary) solution of Eq. (1) in the presence of such a field is given for $I(\Delta) \ll I_0$ by

$$n(t) = \frac{\bar{n}}{1 + I_0/I_s} - \left[\frac{\bar{n}}{1 + I_0/I_s}\left(\frac{I(\Delta)/I_s}{1 + I_0/I_s - i\Delta T_1}\right) e^{-i\Delta t} + \text{c.c.}\right], \quad (3)$$

where $I_s \equiv \hbar\omega/\sigma_1 T_1$ denotes the saturation intensity. The spatial variation of the total intensity that is due to ground- and excited-state absorption is described by

$$\frac{\partial}{\partial z} I_{\text{tot}}(t) = -\{\sigma_1 n(t) + \sigma_2[\bar{n} - n(t)]\} I_{\text{tot}}(t). \quad (4)$$

It is convenient to express this absorption rate in terms of separate absorption coefficients for the unmodulated and modulated components, which are thus given by using Eqs. (2)–(4) as

$$\alpha_0 \equiv -\frac{1}{I_0}\frac{\partial I_0}{\partial z} = \sigma_1\left(\frac{\bar{n}}{1 + I_0/I_s}\right) + \sigma_2\left(\frac{I_0/I_s}{1 + I_0/I_s}\right) = \frac{\bar{n}}{1 + I_0/I_s}(\sigma_1 - \sigma_2) + \sigma_2\bar{n} \quad (5)$$

and

$$\alpha(\Delta) \equiv -\frac{1}{I(\Delta)}\frac{\partial I(\Delta)}{\partial z} = \alpha_0 - \left[(\sigma_1 - \sigma_2)\bar{n}\left(\frac{I_0/I_s}{1 + I_0/I_s}\right)\frac{1 + I_0/I_s + i\Delta T_1}{(1 + I_0/I_s)^2 + (\Delta T_1)^2}\right]. \quad (6)$$

Comparison of Eqs. (5) and (6) shows that the modulated component (and hence the probe beam) experiences appreciably different absorption from the pump beam when the modulation frequency Δ is comparable to or less than $(1 + I_0/I_s)/T_1$. The probe beam experiences less or more absorption depending on whether the absorption coefficient for the excited state is less or greater than that of the ground state.

The experimental setup used to observe these effects (see Fig. 2) is similar to that used by Szabo[7] in his studies of saturated absorption line shapes in ruby. A cw laser beam is passed through an amplitude modulator that creates the probe field as weak frequency sidebands to the pump field. The modulated beam is focused by using a 5× microscope objective to a spot size of approximately 10 μm into a 3.5-mm-long alexandrite crystal. The crystal was oriented so that the c axis was parallel to the electric field direction of the incident light. The transmitted beam was detected by using a silicon photodiode, and the amplitude of the modulated component was determined by using a lock-in amplifier. Our measurement procedure involves determining how the depth of modulation of the transmitted component varies as a function of frequency resulting from the nonlinear response of the alexandrite crystal.

In Fig. 3 the attenuation of the modulated component of the laser beam [i.e., the real part of the complex absorption coefficient given by Eq. (6)] is plotted as a function of modulation frequency. An amplitude-stabilized argon-ion laser producing 110 mW of power at a wavelength of 457 nm was used. At this wavelength, the excited-state absorption cross section is greater than that of the ground state, and therefore an antihole is observed at low modulation frequencies. To compare the experimental line shape with the predicted line shape, the data were fitted to a Lorentzian curve whose width is the inverse of the known value of the ground-state recovery time (T_1 = 260 μsec).[6]

We have also measured the attenuation of the modulated component at a wavelength of 584 nm using the output of a tunable dye laser. At this wavelength, the ground-state absorption cross section is greater than that of the excited state,[6] and hence a hole is predicted at the laser frequency. The

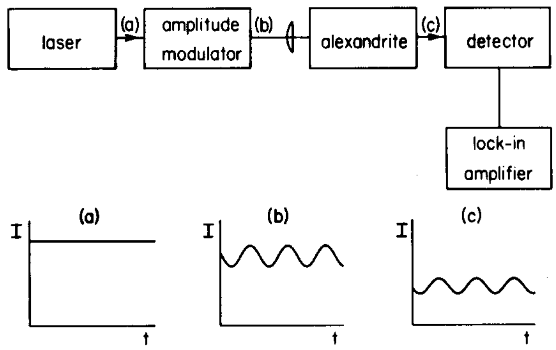

Fig. 2. Experimental setup. A cw laser beam is passed through an amplitude modulator to create a probe beam as weak-frequency sidebands about the laser frequency. The absorption spectrum of this probe beam is determined by measuring, as a function of modulation frequency, how the amplitude of the modulation is changed by the nonlinear absorption of the alexandrite crystal. Curves (a)–(c) show schematically the intensity of the laser beam at representative points in the system.

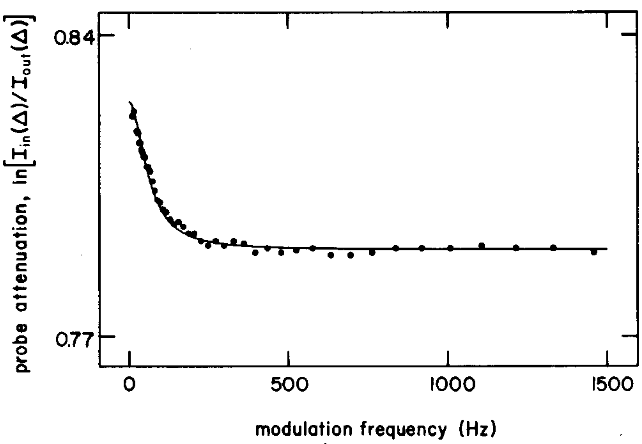

Fig. 3. Attenuation of the modulated component of the laser beam as a function of modulation frequency at a wavelength of 457 nm. An antihole is observed at the laser frequency, as predicted by Eq. (6) for $\sigma_2 > \sigma_1$. The solid line is a Lorentzian whose width is the inverse of the 260-μsec ground-state recovery time of alexandrite.

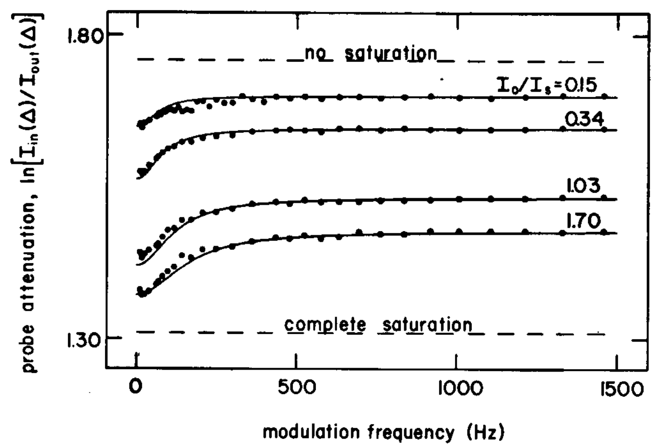

Fig. 4. Attenuation of the modulated component of the laser beam as a function of modulation frequency at a wavelength of 584 nm. The theoretical curves are obtained from Eq. (6) by using the known value of T_1 and the value of the saturation parameter listed by each curve. Also shown are the limiting values of the attenuation that would be experienced by weak or strong beams.

results of this measurement are shown in Fig. 4 for several different values of the laser power ranging from 15 to 200 mW. At large laser powers, significant broadening of the hole is evident. The solid lines are theoretical predictions of the probe line shape and are proportional to the absorption coefficient $\alpha(\Delta)$ of Eq. (6) using the known value of the ground-state recovery time ($T_1 = 260\ \mu$sec). The curves are labeled by the value of the saturation parameter I_0/I_s, which was selected to give the best fit to the data. The small discrepancy between theory and experiment is likely to result from a heating of the crystal at high input powers leading to a variation in the ground-state recovery time T_1.[8]

In conclusion, we have shown, through the use of amplitude-modulation spectroscopy, that we can produce either a hole or an antihole in the probe-beam absorption spectrum depending on the wavelength of the incident radiation. An antihole is produced only if the excited-state absorption cross section exceeds that of the ground state. In either case, the spectral feature has the same width (T_1^{-1}) in the limit of low pump intensity. Our results are in good agreement with a model that ascribes the origin of this effect to the temporal modulation of the ground-state population. This technique can be used to measure the ground-state recovery time even for a system with complicated relaxation processes.

ACKNOWLEDGMENTS

The authors gratefully acknowledge discussions of saturation effects in alexandrite with D. J. Harter and M. L. Shand. This work was supported in part by the National Science Foundation and the U.S. Army Research Office.

* Permanent address, Institute of Experimental Physics, University of Warsaw, Warsaw, Poland.

REFERENCES

1. S. E. Schwartz and T. Y. Tan, "Wave interactions in saturable absorbers," Appl. Phys. Lett. **10,** 4–7 (1967).
2. M. Sargent III, "Spectroscopic techniques based on Lamb's laser theory," Phys. Rep. **43,** 223–265 (1978).
3. R. W. Boyd, M. G. Raymer, P. Narum, and D. J. Harter, "Four-wave parametric interactions in a strongly driven two-level system," Phys. Rev. A **24,** 411–423 (1981).
4. L. W. Hillman, R. W. Boyd, J. Krasinski, and C. R. Stroud, Jr., "Observation of a spectral hole due to population oscillations in a homogeneously broadened optical absorption line," Opt. Commun. **45,** 416–419 (1983).
5. F. Keilman, "Tunable laser-induced grating dip for measuring sub-picosecond relaxation," Appl. Phys. **14,** 29–43 (1977).
6. M. L. Shand, J. C. Walling, and R. C. Morris, "Excited-state absorption in the pump region of alexandrite," J. Appl. Phys. **52,** 953–955 (1981).
7. A. Szabo, "Observation of hole burning and cross relaxation effects in ruby," Phys. Rev. B **11,** 4512–4517 (1975).
8. J. C. Walling, O. G. Peterson, H. P. Jenssen, R. C. Morris, and E. W. O'Dell, "Tunable alexandrite lasers," IEEE J. Quantum Electron. **QE-16,** 1302–1314 (1980).