TABLE II. Spin-orbit parameters for $2p^5 3p^5$ Ar^{+2} in eV.

		$2p$	$3p$	Sum
HF	^1S	1.548	0.026	1.574
	^1D	1.548	0.027	1.575
	^3P	1.547	0.027	1.574
	^3S	1.433	0.141	1.574
	^3D	1.432	0.142	1.574
	^1P	1.432	0.142	1.574
Calc.	(A)	1.432	0.142	1.574

lowest term of a given symmetry and the Hartree-Fock functions have been rotated, thereby changing the spin-orbit parameter. This dependence on LS was first observed by Ridder[7] who has performed an intermediate-coupling calculation for this problem. He obtained good agreement with experimental LSJ energy levels by rather arbitrarily choosing radial functions and spin-orbit parameters for the ^3D term. This happens to be a term where the radial functions are not rotated and the asymptotic form of the wave function is correct for large Z. However, rather than select a particular term energy and use the resulting parameters for a term analysis, I recommend a weighted average of term energies as in calculation (A).

All calculations reported here were performed with a modified version of multiconfiguration Hartree-Fock method.[8]

In summary, Hartree-Fock calculations for configurations with multiple open shells of the same symmetry, and the same occupation number, should only be performed for certain couplings of these shells. The resulting orbitals can be used in a configuration-interaction calculation to improve the accuracy for other couplings, though interactions within the complex and spin-orbit interaction should also be included.

[1]W. Mehlhorn, Z. Phys. 208, 1 (1968).
[2]E. J. McGuire, Phys. Rev. A 11, 1880 (1975).
[3]M. Gavrila and J. E. Hansen, to be published.
[4]J. Bauche and M. Klapisch, J. Phys. B 5, 29 (1972).
[5]C. Froese Fischer, J. Phys. B 6, 1933 (1973).
[6]D. Layzer, Ann. Phys. (N.Y.) 8, 271 (1959).
[7]D. Ridder, private communication.
[8]C. Froese Fischer, Comput. Phys. Commun. 4, 107 (1972), and the code MCHF given therein.

Observation of Amplification in a Strongly Driven Two-Level Atomic System at Optical Frequencies*

F. Y. Wu and S. Ezekiel
Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

and

M. Ducloy
Université Paris–Nord, Villetaneuse 93430, France

and

B. R. Mollow
University of Massachusetts, Boston, Massachusetts 02116
(Received 21 March 1977)

We report the observation of optical amplification in a two-level atomic system driven by a strong, resonant field. By exciting an atomic beam of two-level sodium atoms simultaneously with a strong, fixed-frequency driving field and a weak, tunable, probe field, we have measured the absorption (amplification) line-shape function for several values of driving field strength. In addition, we have verified theoretical predictions that higher amplification is obtained when the strong field is detuned from exact resonance.

We report the observation of optical amplification in a strongly driven two-level system without population inversion. Previously, amplification of amplitude modulation sidebands by a saturated two-level system has been observed with millimeter-wave radiation.[1] Recently, evidence for such amplification at rf[2] and optical frequencies[3] has been demonstrated.

In this Letter, we describe direct measurements of the absorption (amplification) line profile of a strongly saturated two-level optical transition. By simultaneously exciting prepared two-level atoms with a strong, fixed-frequency driving field and a weak, tunable probe field, we have measured the absorption (amplification) line-shape function for several values of driving field strength. In addition, we have demonstrated that higher gain is obtained when the strong field is detuned from exact resonance.

It is well known that a resonant field cannot induce a steady-state population inversion in a two-level system. However, several authors[4-7] have predicted amplification of a weak near-resonant probe field by a strongly saturated resonant medium without population inversion. The energy required for probe amplification is supplied by the saturating field, which experiences increased attenuation due to the presence of the probe.

The effect of a strongly driven system on a weak probe field may be calculated by evaluating to first order the weak-field–induced dipole moment, by means of Bloch equations in which the strong field is treated to all orders. In the case of a single, monochromatic probe field, the absorption-amplification line-shape function W' for a radiatively damped two-level system, as given by Eqs. (3.8) and (3.11a) of Ref. 5, is

$$W' = 2\,\mathrm{Re}[\,i\lambda'\epsilon_0'(\delta\alpha_+)^*], \qquad (1)$$

where

$$\delta\alpha_+ = \frac{i\lambda'\epsilon_0'|z|^2}{|z|^2+\frac{1}{2}\Omega^2}\,\frac{(\Gamma - i\Delta\nu)(z - i\Delta\nu) + \frac{1}{2}i\Omega^2\Delta\nu/z}{(\Gamma - i\Delta\nu)(z - i\Delta\nu)(z^* - i\Delta\nu) + \Omega^2(\frac{1}{2}\Gamma - i\Delta\nu)}. \qquad (2)$$

In these equations, Γ is the natural decay rate, the driving field has the form $\vec{E}(t) = (1/\sqrt{2})\hat{e}_0[\epsilon_0 e^{-i\omega t} + \epsilon_0^* e^{i\omega t}]$, and the probe field has the form $\vec{E}'(t) = (1/\sqrt{2})\hat{e}_0'[\epsilon_0' e^{-i\nu t} + \epsilon_0'^* e^{i\nu t}]$. Also, $\lambda' = \hat{e}_0'\cdot\vec{\mu}/\hbar\sqrt{2}$; $\vec{\mu}$ is the dipole matrix element; $z = \frac{1}{2}\Gamma + i\Delta\omega$; $\Delta\omega = \omega - \omega_0$ is the detuning of the driving field from ω_0, the resonance frequency; $\Delta\nu = \nu - \omega$; $\Omega = 2|\lambda\epsilon_0|$ is the Rabi frequency, with $\lambda = \hat{e}_0\cdot\vec{\mu}/\hbar\sqrt{2}$.

Our experiments were performed on an atomic beam of sodium, using the $3^2S_{1/2}(F=2)$-$3^2P_{3/2}(F'=3)$ transition prepared as a two-level system by optical pumping. The atomic beam and its preparation as a two-level system have been described elsewhere.[8]

The sodium atoms are excited by circularly polarized light from two short-term stabilized cw dye lasers[9] at 5890 Å, as shown in Fig. 1. The purpose of the first laser beam, the "polarizing" beam, is to optically pump the $3^2S_{1/2}(F=2)$ ground-state population into the $m_F = +2$ sublevel of that state. The atoms then enter the interaction region, where they are simultaneously excited by a strong, fixed-frequency "driving field" (also from laser 1) and a very weak, tunable "probe" beam from laser 2. For measurement of absorption or amplification, the probe is electro-optically (E/O) intensity modulated at 2 kHz and synchronously detected after it traverses the atomic beam.

All three light beams are aligned orthogonal to the sodium beam and circularly polarized in the same sense by quarter-wave plates. Uniformity of the driving field in the probed region is ensured by proper choice of laser beam dimensions. The $1/e^2$ diameter of the driving field is 2.5 mm, while that of the probe beam is only 0.21 mm. For these experiments, probe beam power has been attenuated to approximately 3 μW.

To obtain the absorption line shapes, dye laser 1 is long-term stabilized such that the polarizing beam is resonant with the $(F=2)$-$(F'=3)$ transition. The driving field can therefore be maintained either at resonance, or at a constant value of detuning when an acousto-optic (A/O) frequency shifter is used as shown in Fig. 1. The probe beam is then tuned across the transition frequen-

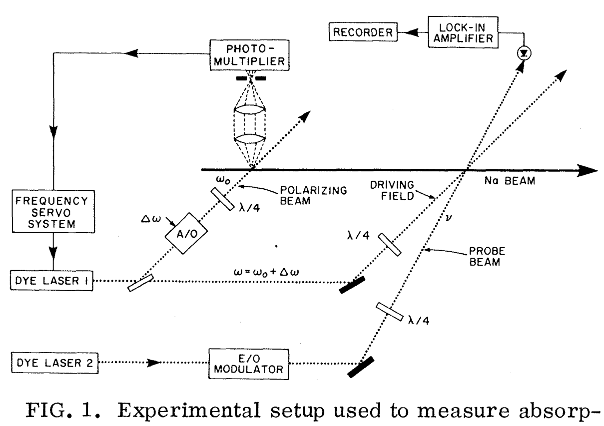

FIG. 1. Experimental setup used to measure absorption or amplification of the probe field. The probe- and driving-field beams propagate at a slight angle to each other, in the same plane orthogonal to the sodium beam. The A/O frequency shifter is removed for the case of a resonant driving field.

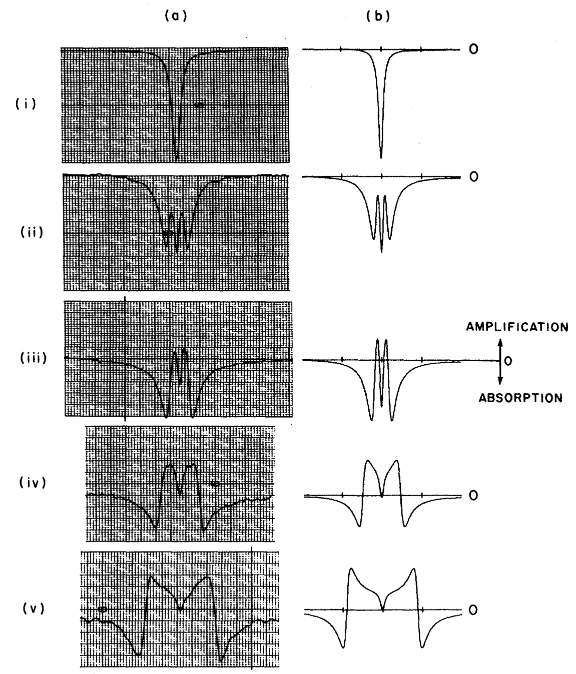

FIG. 2. (a) Experimentally measured line shapes with resonant driving field showing percent change of probe power vs frequency ($\Delta\nu$). Horizontal scale 6 MHz per small division. Peak driving-field intensity and vertical scale per large division: (i) no driving field, 1%; (ii) 26 mW/cm^2, 0.2%; (iii) 47 mW/cm^2, 0.15%; (iv) 130 mW/cm^2, 0.15%; (v) 560 mW/cm^2, 0.06%. Scan rate 2.5 MHz/sec. (b) Theoretical line shapes roughly corresponding to measured line shapes in (a), plotted with $\Gamma/2\pi = 10$ MHz, $\Delta\omega = 0$, and $\Omega/2\pi =$ (i) 0, (ii) 11 MHz, (iii) 14 MHz, (iv) 36 MHz, and (v) 67 MHz. Horizontal scale 75 MHz per division.

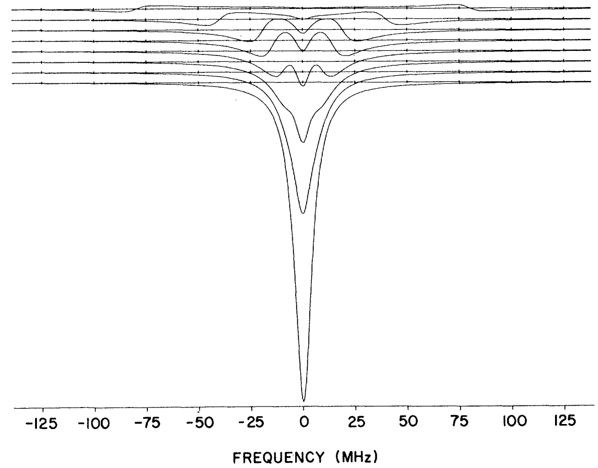

FIG. 3. Progression of theoretical line shapes for resonant driving field of increasing strength, all on same vertical scale. $\Omega/\Gamma = 0$ (bottom curve), 0.5, 0.7, 1, 1.5, 2, 4, 8 (top curve).

cy, and its absorption or amplification recorded.

Figure 2(a) is a set of absorption line shapes measured with resonant driving fields of various intensities. Figure 2(a), part (i) is the unsaturated absorption profile of the optically pumped two-level system without the driving field. The peak absorption was 9.4%, corresponding to a linear atomic density of about 6×10^5 mm^{-2}. Increasing the driving-field intensity [Fig. 2(a), part (ii)] decreases the peak absorption and burns "holes" on both sides of resonance. As the transition becomes strongly saturated, the holes become regions of negative absorption, representing amplification of the probe field [Fig. 2(a), part (iii)]. Maximum gain of approximately 0.4% was measured with a resonant driving-field intensity of 130 mW/cm^2 [Fig. 2(a), part (iv)]. At higher intensities the peak gain decreased, as in Fig. 2(a), part (v).

A set of theoretical line shapes calculated from Eq. (1) are plotted in Fig. 2(b), with $\Gamma/2\pi = 10$ MHz, the natural decay rate of the sodium excited state, and values of Ω roughly corresponding to the intensities used in the experiment. The data and the theoretical curves clearly exhibit similar qualitative features.[10] In Fig. 3 we show the theoretical line shapes for various driving-field intensities plotted on the same scale for comparison purposes. In the limit of high intensities, the absorption curve crosses zero at frequencies shifted from resonance by the Rabi frequency, i.e., at $\omega_0 \pm \Omega$.

The absorption line shape was also studied when the transition was driven by a strong field detuned from resonance. Figure 4(a) is the line shape measured when the driving field was detuned 28 MHz above resonance and had an intensity of 560 mW/cm^2 ($\Omega/2\pi = 66$ MHz). Figure 4(b) is a set of theoretical curves corresponding to various detunings with constant field strength ($\Omega = 6.6\ \Gamma$); the origin of the horizontal axis represents the transition resonance frequency. The curve labeled $\Delta\omega/2\pi = 28$ MHz is in good qualitative agreement with the data of Fig. 4(a). Comparison of Fig. 4(a) with Fig. 2(a), part (v) shows that at the same driving-field intensity, the detuned line shape has higher peak amplification than the on-resonance line shape (0.7%, compared with 0.17%). This is in good agreement with theory, which predicts that peak gain first increases with detuning, reaches a maximum, and then decreases [Fig. 4(b)]. In the limit of

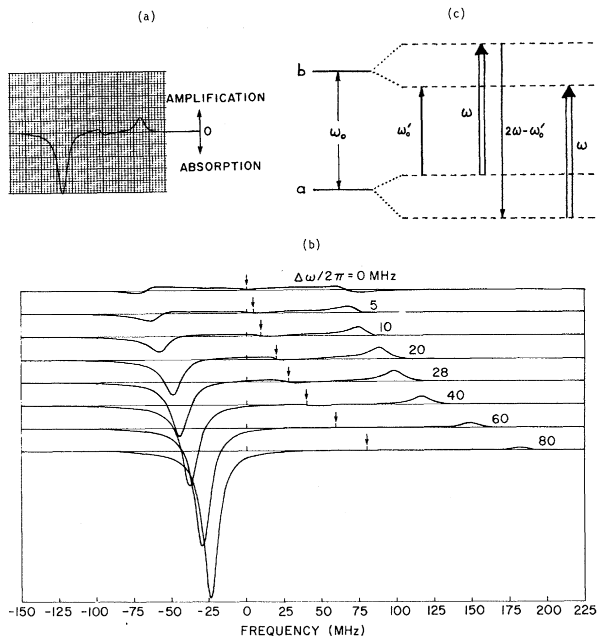

FIG. 4. (a) Measured line shape with driving field detuned by 28 MHz above resonance frequency ($\Delta\omega/2\pi = 28$ MHz). Intensity 560 mW/cm^2 ($\Omega/2\pi = 66$ MHz). Vertical scale 0.6% per large division. (b) Progression of theoretical line shapes with constant field strength ($\Omega/2\pi = 66$ MHz) and increasing detuning $\Delta\omega/2\pi = 0$, 5, 10, 20, 28, 40, 60, and 80 MHz. Origin of horizontal axis is transition resonance frequency. Arrows indicate frequency of driving field. (c) Energy-level diagram illustrating the interaction of a two-level atom with a strong off-resonance field. The frequency of maximum probe absorption is shifted from ω_0 to ω_0'; the probe is amplified at $2\omega-\omega_0'$ due to a three-photon process, involving absorption of two driving-field photons at ω (double arrows) and stimulated emission at $2\omega-\omega_0'$.

high intensities ($\Omega \gg \Gamma$), the predicted maximum gain for a given field strength occurs at a detuning $\Delta\omega$ equal to $\Omega/3$ and is about 5% of the probe field absorption in the absence of the saturating field.[7]

The absorption and amplification peaks in the off-resonance curves may be interpreted with the aid of Fig. 4(c). The strong nonresonant driving field causes a shift of the energy levels, the light shift,[11] resulting in the absorption peak at ω_0'. A three-photon process, involving absorption of two driving-field photons at ω and stimulated emission at $2\omega - \omega_0'$ is responsible for the amplification resonance at $2\omega - \omega_0'$.

In performing the on-resonance measurements, we found it necessary to compensate for atomic recoil due to intense driving fields. This compensation was accomplished by adjusting the angle between the *polarizing beam* and the atomic beam away from orthogonality by a small angle (of the order of 1 mrad); as a result, the frequency of laser 1 (and also the driving field) was locked to a Doppler-shifted resonance frequency, which could be made equal to that of the recoil-shifted resonance. Line shapes measured without compensation for recoil closely resembled theoretical line shapes corresponding to driving fields tuned below the resonance frequency by 0–2 MHz depending on the intensity.

In conclusion, we have directly observed gain at optical frequencies from a strongly driven two-level system without population inversion. The absorption-amplification line shape is seen to be quite different from the emission spectrum.[8,12] The method of probing two-level atoms in an atomic beam as described in this Letter may also be used for highly precise measurements of level shifts. These measurements, as well as quantitative comparison of the absorption-amplification line shapes with theory, are in progress.

*Work supported by the Joint Services Electronics Program, Contract No. DAAB07-76-C-1400.

[1]B. Senitzky, G. Gould, and S. Cutler, Phys. Rev. 130, 1460 (1963); B. Senitzky and S. Cutler, Microwave J. 7, 62 (1964).

[2]A. M. Bonch-Bruevich, V. A. Khodovoi, and N. A. Chigir', Zh. Eksp. Teor. Fiz. 67, 2069 (1974) [Sov. Phys. JETP 40, 1027 (1975)].

[3]S. L. McCall, Phys. Rev. A 9, 1515 (1974); H. M. Gibbs, S. L. McCall, and T. N. C. Venkatesan, Phys. Rev. Lett. 36, 1135 (1976).

[4]S. G. Rautian and I. I. Sobel'man, Zh. Eksp. Teor. Fiz. 41, 456 (1961) [Sov. Phys. JETP 14, 328 (1962)].

[5]B. R. Mollow, Phys. Rev. A 5, 2217 (1972).

[6]S. Haroche and F. Hartmann, Phys. Rev. A 6, 1280 (1972); M. Sargent III and P. E. Toschek, Appl. Phys. 11, 107 (1976).

[7]M. Ducloy, in Proceedings of the Symposium on Resonant Light Scattering, Massachusetts Institute of Technology, Cambridge, Massachusetts, April, 1976 (unpublished); C. Cohen-Tannoudji and S. Reynaud, J. Phys. B 10, 345 (1977).

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

[9]The dye lasers are Spectra Physics Model 580A, stabilized as described in F. Y. Wu and S. Ezekiel, Laser Focus 13, No. 3 (1977).

[10]Quantitative comparison with theory will be the subject of a future publication.

[11]J. P. Barrat and C. Cohen-Tannoudji, J. Phys. Rad. 22, 329, 443 (1961); W. Happer, Rev. Mod. Phys. 44, 169 (1972).

[12]B. R. Mollow, Phys. Rev. 188, 1969 (1969).