Four-Wave Parametric Amplification of Rabi Sidebands in Sodium

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Four-wave optical parametric amplification enhanced by the ac Stark effect has been observed, with the use of a dye laser tuned near the D_2 line of sodium. This process leads to the generation of sidebands displaced from the laser frequency by the generalized Rabi frequency. Under certain experimental conditions, the emitted radiation is in the form of a cone surrounding the transmitted laser beam. A theoretical model that describes these effects is presented.

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This Letter reports the observation of four-wave optical parametric amplification of radiation at the Rabi sideband frequencies of an atomic transition that is strongly driven by a near-resonant laser. These sidebands appear as sharp features in the spectrum of a narrow-band laser pulse after traversing a relatively dilute sodium vapor where self-focusing of the laser is negligible. At higher sodium densities, where self-focusing is present, the sidebands broaden spectrally, and radiation on the low-frequency side of resonance is emitted in a conical shell surrounding the transmitted beam. The observed spectral and spatial characteristics of the emission can be explained by a theory of four-wave parametric amplification in a strongly driven two-level system,[1] when the effects of refraction in a self-focused laser beam are taken into account.

The origin of the parametric amplification at the Rabi sideband frequencies is illustrated in Fig. 1. It shows an incident laser wave at frequency ω_1 which drives an atomic transition at frequency ω_{ba}, thereby modifying the absorption spectrum of the atom and also inducing parametric coupling between two waves, nearly copropagating with the laser wave and having frequencies ω_3 and ω_4, where $\omega_3+\omega_4=2\omega_1$. The experiments reported here were conducted by using a laser that was detuned many linewidths from the resonance and had an intensity sufficiently large that the atomic transition frequency was significantly altered by the ac Stark effect.[2] The absorption of two laser photons at ω_1 accompanied by the emission of a photon at ω_3 is known as the three-photon effect, and produces gain at ω_3 even in the absence of parametric coupling.[3] The three-photon effect is maximum at $\omega_3=\omega_1+\Omega'$, where Ω' is the generalized Rabi frequency given by

$$\Omega' = (\Delta/|\Delta|)(\Delta^2+\Omega^2)^{1/2}, \tag{1}$$

where $\Omega = 2\mu A_1/\hbar$ is the Rabi frequency associated with the transition dipole matrix element μ and the electric field amplitude $2A_1$ of the driving laser, and where $\Delta=\omega_1-\omega_{ba}$. While a wave at $\omega_4=\omega_1-\Omega'$ is known to be strongly absorbed by the ac Stark-shifted resonance in the absence of parametric coupling,[3] a recent theoretical treatment[1,4] predicts that such a coupling of waves at $\omega_3=\omega_1+\Omega'$ and $\omega_4=\omega_1-\Omega'$ can cause the wave at ω_4 to be amplified, along with that at ω_3.

This prediction of parametric gain arises from the solution of the coupled propagation equations[1] for the amplitudes A_3 and A_4 of the waves at $\omega_3 \simeq \omega_1+\Omega'$ and $\omega_4\simeq\omega_1-\Omega'$, respectively, with $\omega_3+\omega_4=2\omega_1$:

$$dA_3/dz = -\alpha_3 A_3 + \kappa_3 A_4{}^* e^{i\Delta k z}, \tag{2a}$$

$$dA_4{}^*/dz = -\alpha_4 A_4{}^* + \kappa_4{}^* A_3 e^{-i\Delta k z}, \tag{2b}$$

where Δk is the phase mismatch and where the nonlinear absorption coefficients α_3 and α_4 and coupling coefficients κ_3 and κ_4 are found from

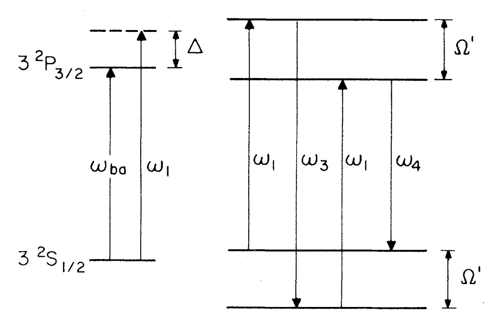

FIG. 1. (a) Laser of frequency ω_1 is tuned near the D_2 line of sodium. (b) Energy levels are split by the ac Stark effect, leading to new resonance frequencies which can enhance the four-wave parametric process shown.

steady-state solutions of the density matrix equations which are correct to first order in the amplitudes A_3 and A_4, and to all orders in the amplitude A_1. If the phase mismatch Δk is very large, the amplitudes A_3 and A_4 are not coupled, and thus propagate independently. The absorption coefficients α_3 and α_4 account for the gain at ω_3 by the three-photon effect (for $\alpha_3 < 0$) and loss at ω_4 by absorption (for $\alpha_4 > 0$). On the other hand, when the phase mismatch is small ($\Delta k \simeq 0$), as in the present experiments, parametric coupling arises from the κ_3 and κ_4 terms, and can cause the wave at ω_4 (the *fourth parametric wave*) to be amplified, as well as the *three-photon wave* at ω_3. This situation is analogous to Stokes–anti-Stokes coupling in Raman scattering.[5]

The apparatus used in this experiment included a nitrogen-laser–pumped dye laser that produced 5-ns pulses having an 0.03 cm^{-1} bandwidth tuned near the ($3^2S_{1/2}$-$3^2P_{3/2}$) D_2 line of sodium at 5890 Å. The dye laser power, after spatial filtering, was ~6 kW, and the beam was focused into a cell containing sodium at a density that was varied between 10^{14} and 10^{16} cm^{-3} and an argon buffer gas at ~1 Torr. The length of the sodium vapor region was ~2.5 cm. Under typical experimental conditions the cell contained 5×10^{14} sodium atoms per cm^3 and 1 Torr of argon, and the laser was detuned 2 Å to the short-wavelength side of the D_2 line and was focused to give a power density of 3×10^7 W/cm^2. The atomic decay times under these conditions are $T_1 = 16$ ns and $T_2 = 10$ ns, the Rabi frequency is $\Omega = 2.5\times10^{12}$ rad/s, the three-photon gain coefficient is $\alpha_3 = 4.3\times10^3$ cm^{-1}, and the maximum value of the coupled gain is $g_+ = 1.5\times10^3$ cm^{-1}.

Figure 2 shows a spectrogram of the light surrounding the transmitted laser beam when the laser was detuned 2.4 Å to the short-wavelength side of the D_2 line, with a sodium density of 1×10^{14} cm^{-3} and an argon pressure of 1 Torr.

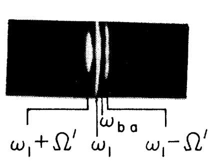

FIG. 2. Spectrum of the light surrounding the transmitted laser beam, showing Rabi sideband frequencies symmetrically displaced from the laser frequency ω_1 by the generalized Rabi frequency Ω'.

This spectrum shows two sidebands, symmetrically displaced from the laser frequency ω_1. We identify these sidebands as the three-photon wave and the fourth parametric wave, in accord with the prediction discussed above. To strengthen this identification we have plotted in Fig. 3 the center wavelength of the observed three-photon wave as a function of laser detuning for several different values of the laser intensity. (The fourth parametric wave was not always present as a narrow line over this range of parameters.) The solid curves show the predicted dependence on detuning and Rabi frequency (laser intensity) calculated from the generalized Rabi frequency in Eq. (1), with the use of the measured value of the detuning and the best-fit value of the Rabi frequency. Each best-fit Rabi frequency is within a factor of 2 of that calculated from the estimated peak laser intensity. Sideband emission occurred only during the 2–3-ns central interval of the 5-ns laser pulse when the laser intensity was nearly constant.

These data indicate the occurrence of four-wave parametric amplification at the Rabi sideband frequencies $\omega_1 \pm \Omega'$. We believe that the source of the low-level radiation that is amplified by the parametric process is either spontaneous scattering or broadband fluorescence from the laser.

At sodium densities above 10^{15} cm^{-3}, that is, 10 times greater than those used in Figs. 2 and 3, the laser beam self-focused (or defocused) and other qualitatively different phenomena occurred: The transmitted spectrum broadened

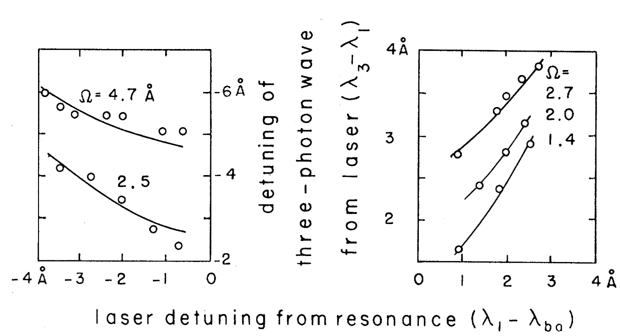

FIG. 3. Detuning of the three-photon wave from the laser wavelength plotted against the laser detuning from resonance for several values of the laser intensity (given in terms of the Rabi frequency Ω in wavelength units). The solid curves are theoretical predictions given by the expression for the generalized Rabi frequency in Eq. (1).

considerably and part of the radiation was emitted in a conical shell surrounding the transmitted laser beam. While these phenomena have been observed previously their origin is under debate.[6,7]

Figure 4(a) is a photograph of the output beam with the central laser spot partially blocked and with conical emission forming a halo around the central spot. The laser detuning was 2.4 Å to the short-wavelength side of resonance. In order to determine the spectral composition of the various spatial regions of the output beam, a slice of the pattern in Fig. 4(a) was focused onto the entrance slit of the monochromator. The resulting spectrum in Fig. 4(b) shows that the halo is made up of radiation at the low-frequency side of the resonance, while the central spot has a broad spectrum extending from the incident laser frequency ω_1 to higher frequencies. As the sodium density is varied, the spectra in Figs. 2 and 4(b) evolve continuously into one another. This suggests that a four-wave parametric process is responsible for the conical emission, but complicated by the presence of self-focusing and self-phase-modulation.

The cone angle of the halo can be predicted by a model that includes the effects of phase matching and of refraction. The laser beam creates an approximately cylindrical region of saturated atoms, having refractive index approximately equal to unity, which is surrounded by ground-state atoms with the unsaturated anomalous dispersion profile centered around the resonance frequency.[8] Thus radiation with frequencies above the resonance will be trapped inside the saturated region, while radiation below the resonance traveling inside the saturated region with a small cone half-angle θ_0 (perhaps due to phase-matching) will be refracted out of the region with a cone half-angle θ given by Snell's law as

$$\theta \simeq (\theta_0{}^2 + 2\delta n)^{1/2}, \tag{3}$$

where δn is the difference between the refractive indices of the outer and inner regions. This angle θ is independent of the spatial profile of the saturating beam. Thus, if $\delta n \gg \theta_0$, the cone angle θ will be largely independent of the mechanism responsible for the origin of the radiation comprising the cone.

In Fig. 5 we have plotted the observed half-angle of the conical emission as a function of the laser detuning and of atomic number density. The data shown in Fig. 5(a) were taken with a number density of 5.0×10^{15} cm^{-3}, and the data in Fig. 5(b) were taken with a laser detuning of 2 Å. The solid curves are given by Eq. (3) under the assumption that the contribution of the internal angle θ_0 to the half-angle is negligible. In order to obtain the good agreement shown in Fig. 5, we found it necessary in evaluating Eq. (3) to use atomic number densities that were consistently a factor of 3.2 larger than those inferred from the temperature of the vapor measured with a thermocouple. We do not know the origin of this small discrepancy. However, our model exactly predicts the cone angles reported by Skinner and Kleiber.[6] It is possible that a nonzero value of θ_0 is responsible for the discrepancy in our case. As a further test of our model of conical emis-

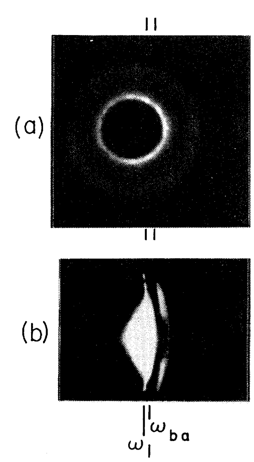

FIG. 4. (a) Halo surrounding the transmitted laser beam. The intense central portion of the laser beam is blocked by a circular beam stop. The laser is detuned 2.4 Å to the short-wavelength side of resonance. (b) Spectrum of the vertical slice indicated in (a).

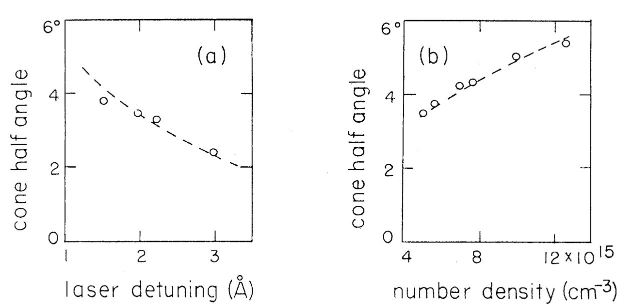

FIG. 5. Cone half-angle plotted against (a) laser detuning and (b) atomic number density. The curves are theoretical predictions given by Eq. (3).

sion, we tuned the laser to the low-frequency side of resonance and again observed conical emission, as was also observed in Ref. 7. The center frequency of this emission was again on the low-frequency side of resonance, in agreement with the model. Because of self-defocusing of the laser beam in this case, experimental conditions under which conical emission occurred were difficult to achieve. The occurrence of conical emission always on the low-frequency side of resonance and the agreement between Eq. (3) and the cone-angle data indicate that the creation of the cone can be explained as largely due to the model discussed above.

Finally, we have measured the temporal pulse shape of the off-axis emission, using a fast silicon photodiode. The pulse duration of this emission was found to be 2 to 3 ns, with no measurable time delay with respect to the incident laser pulse of duration 5 ns. This time dependence, which is independent of the laser detuning and sodium density, is consistent with the above-mentioned model based on four-wave parametric amplification.

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[1]R. W. Boyd, M. G. Raymer, P. Narum, and D. J. Harter, Phys. Rev. A, to be published.

[2]F. Schuda, C. R. Stroud, Jr., and M. Hercher, J. Phys. B 7, L198 (1974).

[3]V. M. Arutyunyan, N. N. Badalyan, V. A. Iradyan, and M. E. Movsesyan, Zh. Eksp. Teor. Fiz. 60, 66 (1971) [Sov. Phys. JETP 33, 34 (1971)]; F. Y. Wu, S. Ezekial, M. Ducloy, and B. R. Mollow, Phys. Rev. Lett. 38, 1077 (1977).

[4]D. J. Harter and R. W. Boyd, IEEE J. Quantum Electron. 16, 1126 (1980).

[5]N. Bloembergen, *Nonlinear Optics* (Benjamin, New York, 1965), pp. 110–119.

[6]C. H. Skinner and P. D. Kleiber, Phys. Rev. A 21, 151 (1980); G. Brechignac, Ph. Cahuzac, and A. Debarre, Opt. Commun. 35, 87 (1980); A. C. Tam, Phys. Rev. A 19, 1971 (1979).

[7]Y. H. Meyer, Opt. Commun. 34, 439 (1980).

[8]A. M. Bonch-Bruevich, V. A. Khodovoi, and V. V. Khromov, Pis'ma Zh. Eksp. Teor. Fiz. 11, 431 (1970) [JETP Lett. 11, 290 (1970)].

Oscillator Strength for Principal Series Transitions to the High Rydberg States of Potassium

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This Letter reports accurate values for the oscillator strength of potassium, and possible resolution of some outstanding discrepancies concerning the oscillator strengths and photoionization cross sections of this atom. Our results establish a value of $(5.2 \pm 0.5) \times 10^{-21}$ cm^2 for the photoionization cross section at the series limit.

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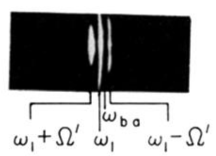
The oscillator strength of alkali atoms deviates significantly from that of the hydrogen atom because of deviations from a Coulomb potential. The effect of core polarization,[1,2] which was thought to be particularly important for transitions to higher Rydberg states and the continuum, has been dealt with in various semiempirical computations,[3-6] and apparent agreement with experimental values of the oscillator strength has been achieved. On the other hand, the quantum defect method[7] (or Coulomb approximation[8]), which has proved to be very powerful in predicting general features in the spectrum of photoionization, does not appear to be always accurate in calculating the value of oscillator strength for transitions between bound states. This difficulty is compounded further by lack of reliable experimental data, and many discrepancies exist concerning the oscilla-



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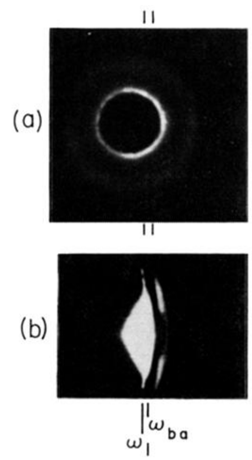

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