Conical emission due to four-wave mixing enhanced by the ac Stark effect in self-trapped filaments of light

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Received July 6, 1982

Experimental evidence is presented that shows that the conical emission often found surrounding a near-resonant laser beam after it passes through an atomic vapor is due to nondegenerate four-wave mixing enhanced by the ac Stark effect. We demonstrate self-trapping of a pulsed dye-laser beam in sodium vapor for wavelengths 0.04 to 10 Å shorter than that of the D_2 resonance and show that the four-wave mixing process leading to conical emission occurs within these self-trapped filaments.

There has recently been great interest in the cone of light that is often found surrounding a near-resonant laser beam after it passes through an atomic vapor. This phenomenon was originally reported by Grischkowsky[1] and has since been studied by Skinner and Kleiber,[2] by Meyer,[3] by Brechignac *et al.*,[4] and by Harter *et al.*[5] Theoretical models[2,6] have had limited success in obtaining detailed agreement between theoretical prediction and observation. In Ref. 5, we presented a brief description of a model of conical emission based on nearly degenerate four-wave mixing enhanced by the ac Stark effect occurring within self-trapped filaments of light. In this Letter, we present a more-detailed description of this model and give experimental and theoretical evidence that self-trapping as described by Javan and Kelley[7] has occurred and that the intensity within the self-trapped filament has the proper value to account for the observed spectrum of the conical emission by the ac Stark effect.

The origin of the resonant enhancement of four-wave mixing by the ac Stark effect is shown in Fig. 1. A strong pump laser of field amplitude E_1 and frequency ω_1 is detuned to the high-frequency side of an atomic resonance by an amount $\Delta = \omega_1 - \omega_{ba}$, where ω_{ba} denotes the unperturbed atomic-resonance frequency. The effect of the strong laser is to split both the upper and lower levels into pairs of levels separated by the generalized Rabi frequency

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

where $\Omega = \mu_{ba}E_1/\hbar$ denotes the Rabi frequency and μ_{ba} denotes the dipole matrix element connecting the atomic levels. As a result of the splitting and shifting of the atomic levels, the four-wave mixing process shown on the right side of Fig. 1 is resonantly enhanced at each of its intermediate levels. A detailed semiclassical calculation that predicts large gain for this four-wave mixing process has been presented by Boyd *et al.*[8] The generation of intense radiation in the forward direction at the Rabi sideband frequencies (i.e., $\omega_1 \pm \Omega'$) attributable to this process has been reported by Harter *et al.*[5] and subsequently by Kleiber *et al.*[9]

The salient observed features of the conical-emission phenomenon are as follows. Conical emission occurs only when self-focusing of the incident laser occurs and hence only when the laser is detuned to the high-frequency side of resonance. The spectral peak of the conical emission is always shifted to the low-frequency side of the laser by an amount that is in the range 1.5Δ to 3.0Δ. The cone half-angle of the conical emission has been found to be well fitted by an empirical relation of the form

$$\theta = K|2\delta n|^{1/2}, \qquad (2)$$

where $1 + \delta n$ denotes the refractive index of the atomic vapor at the *laser frequency*. The multiplicative constant K has the value 1.8 according to the data of Harter *et al.*[5] and the value 2.0 according to the data of Skinner and Kleiber.[2,10]

According to our model of conical emission, incident laser radiation self-focuses on entering the atomic-vapor cell, leading to the formation of a self-trapped filament. Four-wave mixing occurs within the filament, leading to the generation of radiation at the Rabi sideband frequencies. Light at the high-frequency sideband thus can be trapped within the filament by total internal reflection, whereas light at the low-frequency sideband is ejected from the filament at an angle given by Snell's law. The frequency of the conical emission is shifted from that of the incident laser by the value of the gen-

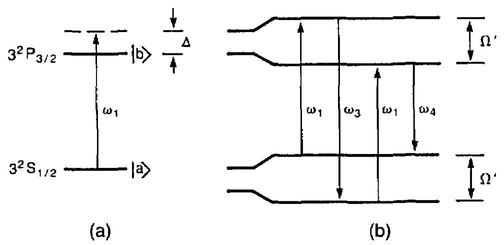

Fig. 1. (a) Laser of frequency ω_1 is tuned near the D_2 line of sodium. (b) Energy levels are split by the ac Stark effect, leading to new resonance frequencies that can enhance the illustrated four-wave mixing process.

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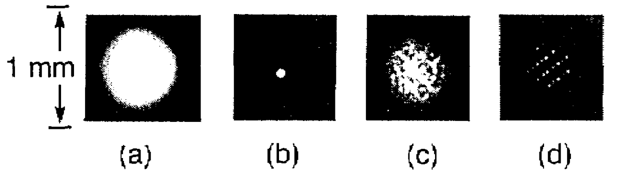

Fig. 2. Magnified images of the laser beam at the exit window of the sodium cell. Beam is shown (a) in the absence of self-focusing and (b) with self-focusing into a single filament. At large input powers, the beam breaks up into many filaments (c), whose positions are fixed (d) by placing spatial structure on the input beam.

eralized Rabi frequency within a self-trapped filament. Equation (1) shows that, in order for Ω' to be in the inferred range 1.5Δ to 3.0Δ, the ratio Ω/Δ within the filament must be in the range 1.1 to 2.8. In fact, the following simple physical argument based on saturation of the refractive index shows that the expected value of Ω/Δ is approximately in this range. As long as the refractive index of the medium increases with increasing field intensity, the medium acts as a lens with positive power, leading to continued self-focusing and an increase in optical intensity. The self-focusing process must terminate when the refractive index saturates, which occurs when Ω is approximately equal to Δ.

In order to test our above-stated model of conical emission, we have performed a study of self-focusing and self-trapping in sodium vapor. We use the word self-trapping to mean the propagation of a self-focused beam with a diameter that is nearly constant in space and time. Our experimental apparatus includes a frequency-doubled Nd:YAG laser, which was used to pump a Littman-style dye laser that produced 2–7-nsec pulses. Each laser was operated in a single longitudinal mode to prevent intensity modulation resulting from mode beating, which would tend to wash out the Rabi sideband structure of the atomic response. Under typical conditions, the laser was tuned 3 Å to the high-frequency side of the ($3^2S_{1/2}$–$3^2P_{3/2}$) D_2 line of sodium and was focused into one of several sodium vapor cells (of lengths varying between 1.25 and 25 cm) containing $\sim 5 \times 10^{15}$ sodium atoms/cm^3. A well-corrected $f/2.5$ lens was used to form a magnified image of the exiting laser beam, whose temporal and spatial properties could thus be monitored.

Figure 2 shows photographs of the emerging laser beam under a variety of circumstances. Figure 2(a) shows the beam transmitted through an empty vapor cell. Figure 2(b) shows the transmitted beam self-focused into a single filament with a sodium density of 5×10^{15} and an input intensity adjusted to the minimum value ($\sim$40 kW/cm^2) that produced self-focusing. As the laser power is increased, the self-focused beam breaks into many filaments, as shown in Fig. 2(c), whose pattern changes randomly from shot to shot. By purposely introducing spatial structure on the unaberrated input beam (i.e., by means of the interference pattern produced by a sapphire window) it is possible to fix the positions of the filaments from shot to shot, as shown in Fig. 2(d). The ability to fix the locations of self-trapped filaments has allowed us to perform accurate measurements of the diameters, intensities, and temporal evolution of individual filaments.

To verify that the phenomenon observed by us was in fact self-trapping, we have compared the measured diameters (at half-intensity points) of our filaments with the theoretical diameter of a self-trapped filament given by Javan and Kelley[7] as

$$d_{\min} = 1.22\lambda(8n\delta n)^{-1/2}, \tag{3}$$

where λ is the vacuum wavelength of the radiation, n is the refractive index of the medium ($\sim$1 for an atomic vapor), and δn is the maximum value of the light-induced change in refractive index, which is taken as $n - 1$ under the assumption that the refractive index is totally saturated within a self-trapped filament. A range of filament diameters was obtained by varying the laser detuning and atomic-number density. Good agreement between the measured and theoretical diameters is obtained, as shown in Fig. 3.

Further evidence that self-trapping has occurred is obtained by comparing the temporal behavior of the intensity of the light leaving a filament with that of the incident laser, as shown in the inset of Fig. 3. The detector used in this measurement is smaller than the magnified image of an individual filament and thus measures the intensity of the self-trapped light. The close tracking of input and output intensities shows that self-trapping has occurred and that the filament diameter does not change appreciably during the laser pulse.

Self-trapping of light is considered to be a steady-state effect and hence might not be expected from a laser pulse whose duration τ_p ($\sim$3 nsec) is much less than the atomic upper-state lifetime T_1 (16 nsec). In fact, under our experimental conditions and those of Refs. 2–5 (that is, $\Delta^{-1} < T_2 \ll \tau_p < T_1$, where T_2 is the dipole relaxation time), the atomic response is nearly in the steady state for most of the duration of the laser pulse. To demonstrate this fact, we model the laser

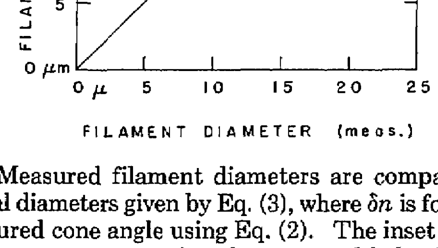

Fig. 3. Measured filament diameters are compared with theoretical diameters given by Eq. (3), where δn is found from the measured cone angle using Eq. (2). The inset shows an oscilloscope trace comparing the temporal behavior of the light leaving the self-trapped filament with a delayed portion of the input laser pulse.

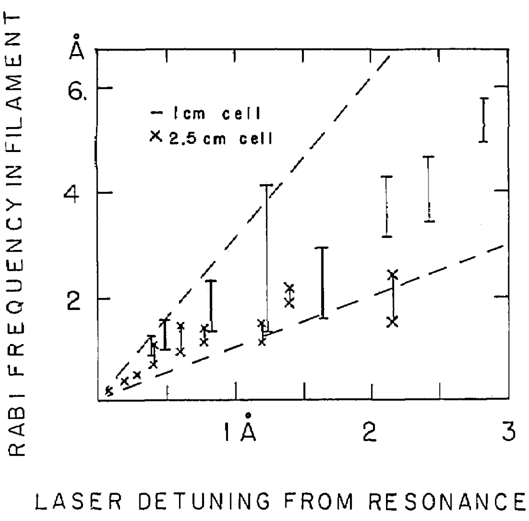

Fig. 4. The measured intensity within a self-trapped filament (expressed as a Rabi frequency in angstroms) is compared with the laser detuning from resonance for two different cell lengths.

radiation as a square pulse with a turn-on at time $t = 0$ and calculate the atomic response using the density-matrix equations of motion in the rate-equation limit, which for $\Delta T_2 \gg 1$ is valid whenever $\Omega/\Delta \ll 1$. We find that, for times $t \gg T_2$ (i.e., after any coherent transients have died away), the refractive index of the medium is given by $n = 1 + (\rho_{bb} - \rho_{aa})\delta n_{\max}$, where $\delta n_{\max} = -2\pi N|\mu_{ab}|^2\hbar^{-1}(\Delta^2 + T_2^{-2})^{-1}$, with N denoting the atomic-number density and where

$$(\rho_{bb} - \rho_{aa}) = (\rho_{bb} - \rho_{aa})^0 e^{-t/\tau_R} + (\rho_{bb} - \rho_{aa})^{\mathrm{dc}}(1 - e^{-t/\tau_R}), \quad (4)$$

$(\rho_{\mathrm{bb}} - \rho_{\mathrm{aa}})^0$ is the initial population inversion, $(\rho_{bb} - \rho_{aa})^{\mathrm{dc}} = (\rho_{bb} - \rho_{aa})^0(1 + \Delta^2 T_2^2)/(1 + \Delta^2 T_2^2 + \Omega^2 T_1 T_2)$ is the dc component of the steady-state population inversion, and $\tau_R = [T_1^{-1} + T_2^{-1}\Omega^2/(\Delta^2 + T_2^{-2})]^{-1}$ is the effective population relaxation time, which under appropriate conditions can be much shorter than T_1. Under our conditions $T_2 \ll T_1$, and thus as Ω increases because of self-focusing, the population inversion $(\rho_{bb} - \rho_{aa})$ becomes nearly saturated even before Ω increases to a value comparable to Δ. The fact that the atomic response quickly reaches the steady state within the filament indicates that the steady-state model of four-wave mixing given in Ref. 8 is applicable under the present experimental circumstances.

In order to establish that the ac Stark effect is the origin of the frequency shift observed in the conical emission, we have measured the intensity of the light contained within individual self-trapped filaments. We find that, at a fixed value of the laser detuning, the observed Rabi frequencies within self-trapped filaments span a remarkably narrow range as parameters such as the atomic-number density, buffer-gas pressure, and incident laser-power density are varied. The results of this study are shown in Fig. 4, in which the vertical lines denote the range of observed Rabi frequencies and the two dashed lines delineate the range of values of Ω/Δ (1.1 to 2.8) required to account for the observed spectra of conical emission as a Rabi sideband phenomenon. As mentioned previously, the observed Rabi frequencies are in the range where strong saturation of the refractive index would limit the self-focusing process.

We gratefully acknowledge useful discussions with M. G. Raymer and the technical assistance of L. W. Hillman and J. W. Kuper. This work was partially supported by the U.S. Army Research Office.

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