TWO-PHOTON CONICAL EMISSION

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Received 15 January 1985

A two-photon resonantly enhanced four-wave mixing (FWM) process leading to the conical emission of two new frequency components has been observed in atomic sodium vapor. A dye laser tuned close to the 3s → 3d two-photon allowed transition produces broad-band emission near the frequencies of the 3d → 3p and 3p → 3s transitions. This radiation is emitted in the forward direction in the form of cones surrounding the transmitted laser beam. The dependence of the cone angle on the emission wavelength and atomic number density is in excellent agreement with the predictions of a model that ascribes the origin of the conical emission to a phase-matched four-wave mixing process.

There has recently been considerable interest in the cone of light that is often observed surrounding a laser beam that has passed through an atomic vapor. Much of the previous work has involved the case of a laser that is tuned close to a one-photon-allowed atomic transition [1–8]. Conical emission under these conditions was first observed by Grischkowsky [1]. This process was studied more extensively by Skinner and Kleiber [2], who proposed several possible explanations for the phenomenon, such as "a hypothetical parametric mixing process" or a form of Cerenkov radiation. Harter et al. [4] have shown that conical emission can result from a phase-matched four-wave mixing process that is resonantly enhanced by ac-Stark-split levels. Several recent papers [5–7] have questioned whether a four-wave mixing model can explain all of the features observed in these experiments. In particular, Burdge and Lee [6] have shown that under some conditions spectral features occur that are not readily explained by a four-wave mixing model. In addition, Golub et al. [7] have suggested that a model based on Cerenkov radiation leads to better agreement with experimental data for the conical emission angle than does a model based on phase matching.

Much of the difficulty in obtaining exact agreement between theory and experiment in these studies [1–8] involving one-photon resonances results from self-focusing effects which usually occur simultaneously with the conical emission. In these experiments, all of the interacting waves have been close to a one-photon resonance, and therefore the refractive index experienced by each of the interacting waves has been subject to modification by the saturation effects caused by the self-focusing of the incident wave. In particular, the conical emission angle can be influenced by refraction at the boundary of a self-trapped filament [4].

In this paper, we describe an experiment that demonstrates conical emission under conditions of two-photon excitation of an atomic vapor in the absence of self-focusing. Conical emission under these conditions has been observed previously [9,10], but has not been studied in detail. We find that the properties of the conical emission are in good agreement with the predictions of a model that ascribes the origin of the effect to a phase-matched, two-photon resonantly enhanced four-wave mixing process.

Our experimental setup included an excimer-laser-pumped dye-laser oscillator followed by two amplifiers. We used a longitudinal pumping arrangement for both the oscillator and the amplifiers. We found that this configuration produced a beam with a good transverse profile and helped to suppress the generation of broadband ASE from the dye cells, thus increasing the spectral purity of the laser output. High spectral puri-

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0 030-4018/85/$03.30 © Elsevier Science Publishers B.V. (North-Holland Physics Publishing Division)

ty proved important in preventing unwanted seeding of the four-wave mixing process and in preventing excitation of sodium molecular levels. Typical pulse energies were 200 μJ in a 1.5 ns pulse. The laser beam was focused using a well-corrected lens of 1 m focal length to a spot size ($2w_0$) of ~30 μm into a heat-pipe vapor cell of interaction length 7 cm containing 1 to 50 $\times 10^{15}$ sodium atoms per cm^3.

Our experiment involved tuning the laser close to the 3s 3d two-photon allowed transition, which led to the emission of an intense cone of light into the forward direction, as shown in fig. 1a. In order to determine the spectral composition of this light, we imaged the cone onto the entrance slit of a 1-m grating spectrograph. The photographs of the output (see fig. 1b) show the angular distribution of the light displayed vertically versus the spectral distribution displayed horizontally. The generated light is seen to consist of spectral regions near the 3d → 3p and 3p → 3s transition frequencies. Note that the two spectra complement each other in that the 3d → 3p emission tails off to higher frequencies whereas the 3p → 3s emission tails off to lower frequencies. Note further that the cone angle increases as the emission frequency approaches that of the intermediate resonance.

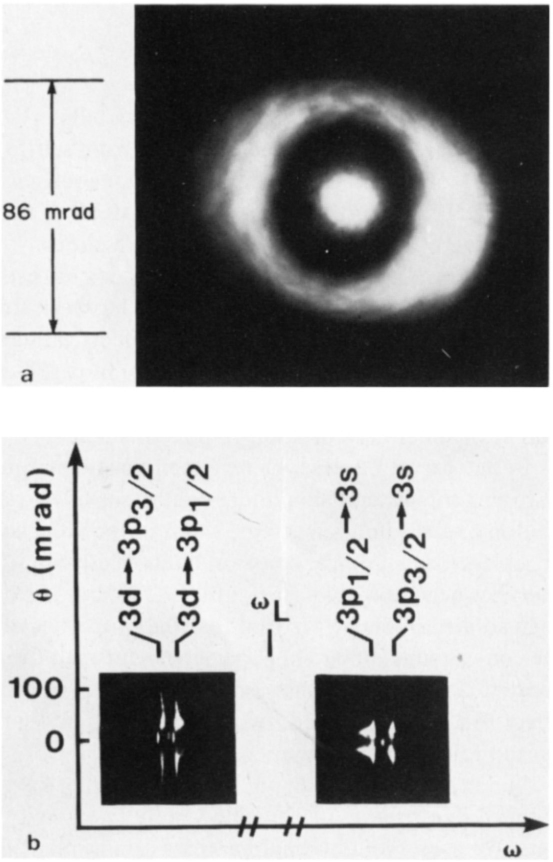

Fig. 1. (a) Photograph of the emission in the forward direction near the 3p → 3s transition frequency, at an atomic number density of ~4 $\times 10^{16}$ cm^{-3}. (b) Spatially resolved spectrographs of the conical emission at an atomic number density of 1.8 $\times 10^{16}$ cm^{-3}.

These observations can be explained in terms of a four-wave parametric mixing process that is resonantly enhanced by the two-photon resonant 3d level and the intermediate 3p level, as shown in fig. 2. We assume that ω_1 denotes the frequency of the applied laser field and that ω_2 and ω_3 denote the frequencies of the generated fields. The generated fields are assumed to grow from noise, and thus the only restriction on their frequencies is that $\omega_2 + \omega_3 = 2\omega_1$. The amplitudes of the generated fields are coupled to one another through nonlinear susceptibilities whose resonant contributions are given by

$$\chi^{(3)} \equiv \chi^{(3)}(\omega_3 = 2\omega_1 - \omega_2) = \chi^{(3)}(\omega_2 = 2\omega_1 - \omega_3)$$
$$= N|\mu_{3s\to3p}|^2 |\mu_{3p\to3d}|^2 / \hbar^3(\omega_1 - \omega_{3p\to3s})$$
$$\times (2\omega_1 - \omega_{3d\to3s} + i\Gamma_{3d})(\omega_3 - \omega_{3p\to3s} + i\Gamma_{3p}), \quad (1)$$

where N denotes the number density of atoms, μ_{ij} the electric dipole matrix element connecting states i and j, ω_{ij} the transition frequency between levels i and j, and Γ_i the width of level i. Due to this coupling, the two generated fields experience exponential growth

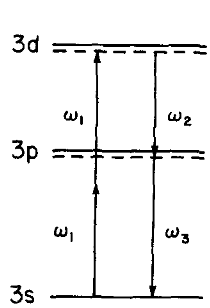

Fig. 2. Energy level diagram showing the resonances of the four-wave mixing process. The ω_1, ω_2, and ω_3 fields correspond to wavelengths of ~6856, ~8183, and ~5890 Å, respectively.

with a gain per unit length given by [11]

$$g = \tfrac{1}{2}[(\alpha_2 - \alpha_3 - \mathrm{i}\Delta k)^2 + 4G^2]^{1/2} - \tfrac{1}{2}(\alpha_2 + \alpha_3), \quad (2)$$

where

$$G = (2\pi/c)(\omega_2\omega_3)^{1/2}|\chi^{(3)}||A_1|^2. \quad (3)$$

Here α_i denotes the absorption coefficient for wave i, Δk denotes the longitudinal component of the propagation vector mismatch, and A_1 denotes the amplitude of the laser field. For the optimum case of negligible absorption and perfect phase matching (i.e., $\Delta k = 0$), the coupled gain g is equal to G. Our experiments were typically carried out using a number density N equal to 1×10^{16} cm^{-3}, a laser field strength of 3000 esu (corresponding to a power density of ~1 GW cm^{-2}), and a detuning from the two-photon resonance of 3 GHz. For an ω_3 wave that is 1 cm^{-1} from the third intermediate resonance, evaluation of eqs. (1) and (3) gives a value of the maximum gain G equal to 4×10^4 cm^{-1}. In our experiment, absorption was negligible except for emission precisely at the frequencies of the D lines. Thus, we expect efficient conversion of the laser radiation into the new frequency components whenever the phase-matching requirements are met.

Due to the dispersion of the D lines, the phase-matching condition cannot in general be satisfied for collinear propagation, and therefore the generated waves are usually emitted at some nonzero angle with respect to the laser propagation direction. These directions can be calculated with the help of the phase-matching diagram shown in fig. 3a, with propagation vector magnitudes given by $k_2 = \omega_2/c$, $k_3 = n_3\omega_3/c$, and $k_1 = (\omega_1/c)\cos\theta_1$, with θ_1 denoting the convergence half angle of the incident laser beam. The $\cos\theta_1$ factor is included to account for the modification of the phase-matching condition due to the effects of focusing on the laser beams [12]. The requirement that $k_2 + k_3 = 2k_1$ implies the two simultaneous equations

$$k_2 \sin\theta_2 = k_3 \sin\theta_3 \quad (4a)$$

and

$$2k_1 \cos\theta_1 = k_2 \cos\theta_2 + k_3 \cos\theta_3, \quad (4b)$$

which can be solved to yield

$$\theta_3^2 = \frac{(n_3 - 1) + (\omega_1/\omega_3)\theta_1^2}{n_3(1 + n_3\omega_3/\omega_2)/2}, \quad (5)$$

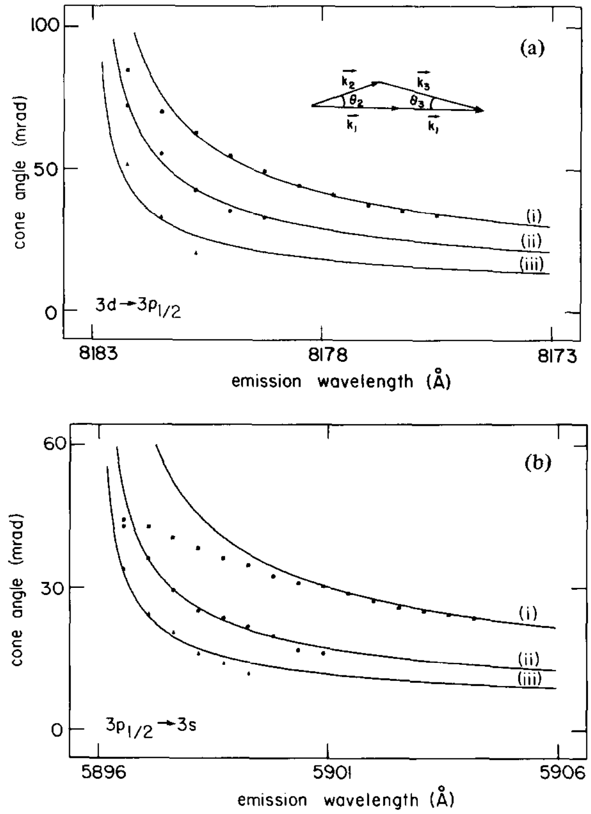

Fig. 3. Cone angle versus emission wavelength for (a) the 3d → 3p$_{1/2}$ and (b) 3p$_{1/2}$ → 3s transitions, for atomic number densities of (i) 3.8×10^{16} cm^{-3}, (ii) 1.8×10^{16} cm^{-3}, and (iii) 7.4×10^{15} cm^{-3}.

and

$$\theta_2 = n_3\theta_3\omega_3/\omega_2. \quad (6)$$

Fig. 3a shows a plot of the measured cone angle versus emission wavelength for several different values of the sodium number density for the 3d → 3p$_{1/2}$ transition. The data are in excellent agreement with the theoretical curves, which were obtained from eqs. (5) and (6) using $n_3 = 1 + (1.4 \times 10^{-19})N/\Delta\lambda$ with N in cm^{-3} and $\Delta\lambda$ in Å. A small roll-off of the experimental data is evident for large cone angles due to the decreased interaction length at these angles. Fig. 3b shows similar results for the 3p$_{1/2}$ → 3s transition. There is now a visible discrepancy between theory and experiment for high sodium density close to the resonance. These discrepancies result from the effects of absorption of the emitted radiation by ground state atoms. This discrepancy occurs only under conditions where the sodium cell is optically thick.

A close inspection of fig. 1b shows several narrow wavelength regions in which radiation is emitted on axis. There are three wavelengths near the 3s → 3p transition wavelength for which the refractive index is exactly equal to unity. At each of these wavelengths, the four-wave mixing process is perfectly phase matched for emission in the forward direction. Each of these wavelengths has a complement near the 3d → 3p transition. These three components are seen on the left of fig. 1b. The right side of the figure, which displays the spectrum near the 3p → 3s transition, shows only one of the expected components because the other two correspond to the wavelengths of the D_1 and D_2 lines and are therefore absorbed.

In this paper, we have interpreted our experimental results in terms of a phase-matched, parametric four-wave mixing process. It has been proposed [2,7] that conical emission from atomic vapors can be interpreted as a form of Cerenkov radiation [13] Cerenkov-like emission is expected to occur whenever the nonlinear polarization wave at frequency ω_c travels in the forward direction with a phase velocity v_p that exceeds the phase velocity v_c of the emitted radiation. The longitudinal component of the phase velocity of the emitted radiation can be equal to v_p if the radiation is emitted at the Cerenkov angle θ_c, given by

$$\cos\theta_c = v_c/v_p . \tag{7}$$

For nonlinear optical processes (such as second-harmonic generation [14]) in which the nonlinear polarization is determined solely by the applied fields, the predictions of the Cerenkov model for the conical emission angle are completely consistent with the predictions of a phase-matching model that assumes that only the longitudinal components of the propagation vectors of the interacting fields are conserved. However, the Cerenkov model cannot be applied directly to experiments, such as those reported in ref. [4] and in the present paper, in which two different fields are generated from noise by the nonlinear process. The complication in applying the Cerenkov model arises from the fact that the direction of propagation of the nonlinear polarization wave at frequency ω_2 depends on the direction of propagation of the ω_3 electromagnetic wave (and vice versa), which is not known a priori since

$$P(\omega_2) \propto E(\omega_1)^2 E^*(\omega_3) \exp[i(2\boldsymbol{k}_1 - \boldsymbol{k}_3)\cdot\boldsymbol{r}] .$$

In our interpretation, all vector components of the propagating waves are conserved, as shown in the inset to fig. 3a. This condition is that of perfect phase matching and is possible whenever the ω_3 wave is generated on the low-frequency side of the 3p → 3s resonance, since in this case n_3 is greater than unity and real angles θ_2 and θ_3 satisfying eqs. (5) and (6) can always be found. Under these conditions, the polarization wave giving rise to emission in some particular direction propagates precisely in that direction, in contrast to the Cerenkov model in which the polarization wave propagates always on axis.

Golub et al. [7] predict on the basis of a Cerenkov model that the cone angle θ_c will be given by

$$\cos\theta_c = n_L/n_c , \tag{8}$$

where n_L and n_c are the refractive indices seen by the laser and the conical emission, respectively. They hence implicitly assume (see eq. (7)) that the polarization wave travels at the phase velocity of the laser, which in general is not true. In accordance with eq. (8), their model also predicts that the cone angle of the emitted radiation at ω_3 depends on the refractive index at ω_3, but not on the refractive index at ω_2 (and vice versa). In particular, their model predicts that the cone angle will be zero whenever the refractive index of the emitted radiation is equal to that of the laser. When applied to our experiment, their model predicts that the ω_3 radiation is emitted in a cone, as observed, but that the ω_2 radiation is emitted only on axis, in contrast with our experimental observations. Our model, based on phase-matching considerations, predicts that both generated fields are emitted in cones, as predicted by eqs. (5) and (6).

In conclusion, we have studied two-photon conical emission under conditions such that the complicating effects of self-focusing are not present. The emission angle of both generated waves is found to be in excellent agreement with a theoretical model based on a phase-matched four-wave mixing process.

This work was supported by the Joint Services Optics Program.

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