

Two-beam-excited conical emission

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We describe a conical emission process that occurs when two beams of near-resonant light intersect as they pass through sodium vapor. The light is emitted on the surface of a circular cone that is centered on the bisector of the two applied beams and has an angular extent equal to the crossing angle of the two applied beams. We ascribe the origin of this effect to a perfectly phase-matched four-wave mixing process.

Transverse effects¹ are important in nonlinear optics because processes such as optical phase conjugation² and two-beam coupling³ can have their fidelity severely degraded by the undesired modification of the spatial profiles of the interacting beams. An important example of a transverse instability is the emission of radiation in the form of a cone when a single laser beam is passed through a resonant nonlinear medium.^{4,5} The cone of emission is centered on the transmitted laser beam and occurs when the frequency of the applied beam is tuned to the blue side of an atomic resonance. For the case of two applied counterpropagating beams, conical emission has also been observed when the applied beams are tuned to either the red or blue side of the resonance.^{5,6} Many of the physical properties of conical emission have been found to be adequately explained by an analysis based on four-wave mixing.⁴⁻⁷ There have also been observations of somewhat different types of conical emission that occur when photorefractive crystals are used as the nonlinear material.⁸

In this Letter, we describe a conical emission process that occurs when two beams of the same frequency intersect in a nonlinear medium.⁹ Unlike as in other types of conical emission, where the cone angle depends on material parameters, the cone angle of the two-beam-excited (TBE) conical emission that we observe is defined by the geometry of the interaction. The cone is found to be centered on the bisector of the two applied beams with a cone angle that is equal to the crossing angle of the beams (see Fig. 1). Observation of TBE conical emission has been reported earlier by Kirilenko *et al.*⁹ in connection with their studies of a noncoplanar four-wave mixing process in sodium vapor. In their paper the authors speculate that the TBE conical emission process occurs as a result of a nonstationary nonlinearity that is higher than third order. In contrast, we show theoretically that the TBE conical emission process can occur in the steady state and results from a perfectly phase-matched four-wave mixing process that arises from the third-order nonlinearity of the medium.

The geometry of our experimental study of TBE conical emission in atomic sodium vapor is shown in Fig. 1. The vapor was contained in a cell with an interaction length of 5 cm. The density of the vapor

was approximately $3 \times 10^{14} \text{ cm}^{-3}$. The two beams intersecting in the cell were obtained from a pulsed dye laser that had a bandwidth of 1.2 GHz and a pulse length of 10 ns. The output of the laser was spatially filtered to give a smooth spatial profile and a 7-mm beam diameter. The unfocused beams were passed through the cell, and the transmitted light was observed on a screen positioned 2 m from the cell.

An example of the TBE conical emission, as viewed on the screen, is shown in Fig. 2. For this observation the laser was tuned to the red side of the *D2* line by 40 GHz. The pulse energy of each beam was $50 \mu\text{J}$, and the angle between the two intersecting beams was equal to 5° . The two bright spots on the screen correspond to the direct transmission of the applied beams. When either of the applied beams was blocked, the ring vanished and the other beam was transmitted with only a small modification of its spatial profile as a result of self-defocusing. The conical emission was at the same frequency as the two applied fields, and the polarization of the cone was the same as that of the applied fields. We found that TBE conical emission occurred for a wide range of experimental conditions, such as when the laser was tuned to either side of the *D1* or *D2* resonance lines. However, when the laser was tuned too close to one of the resonances, we no longer observed the cone. We believe that this effect is due to strong self-focusing or self-defocusing as well as to absorption in the sodium vapor. The size of the cone is determined by the crossing angle as illustrated

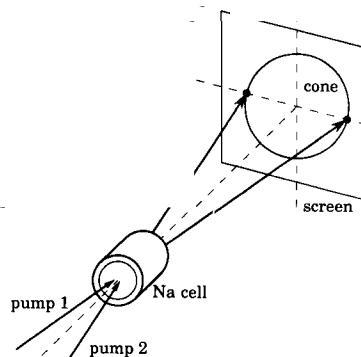


Fig. 1. Schematic of the experimental configuration.

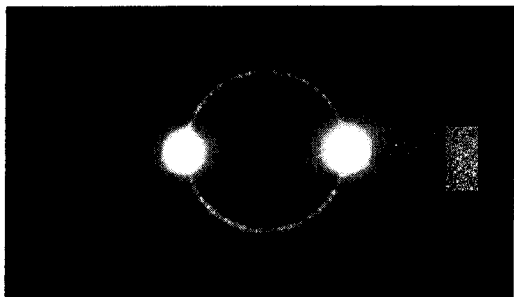


Fig. 2. Photograph of the TBE conical emission as observed on a screen located 2 m from the atomic sodium vapor cell. For this photograph the laser was tuned to the red side of the D2 line by 40 GHz.

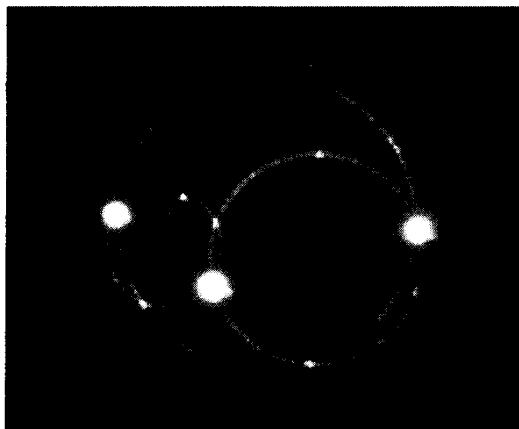


Fig. 3. Photograph of TBE conical emission when three beams are applied. The three possible pairings of the applied beams lead to the generation of three rings of different diameters.

in Fig. 3. For this figure, three beams of equal input intensity were incident upon the cell in such a manner that the crossing angle of each pair of beams was distinct. Three rings of unequal diameter were observed on the screen; each ring is the result of the interaction of two of the input beams.

We now present a brief theoretical description of TBE conical emission. We assume that the nonlinear medium can be treated as a Kerr medium, i.e., that the total third-order nonlinear polarization at frequency ω is

$$P^{NL} = 3\chi^{(3)}|E|^2E, \quad (1)$$

where $\chi^{(3)} = \chi^{(3)}(\omega, \omega, \omega, -\omega)$ is the third-order nonlinear susceptibility and E is the complex electric-field amplitude at frequency ω . We express the total field as the sum of four components as

$$E(\mathbf{r}) = \sum_{j=1}^4 A_j(\mathbf{r})\exp(i\mathbf{k}_j \cdot \mathbf{r}), \quad |\mathbf{k}_j| = \omega/c, \quad (2)$$

where the amplitudes A_j vary slowly in space. We take A_1 and A_2 to be the amplitudes of the two applied beams and A_3 and A_4 to be the amplitudes of two of the fields that grow from noise. The slowly varying polarizations driving the fields A_3 and A_4 will each contain several terms of which the ones that lead to TBE conical emission are given by

$$P_3 = 6\chi^{(3)}A_1A_2A_4^* \exp[i(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3 - \mathbf{k}_4) \cdot \mathbf{r}],$$

$$P_4 = 6\chi^{(3)}A_1A_2A_3^* \exp[i(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3 - \mathbf{k}_4) \cdot \mathbf{r}]. \quad (3b)$$

We assume that the wave vectors of the two applied waves lie in the x - z plane with components k_x and k_z such that

$$\mathbf{k}_1 = k_x\hat{x} + k_z\hat{z}, \quad \mathbf{k}_2 = -k_x\hat{x} + k_z\hat{z}, \quad (4)$$

where \hat{x} , \hat{y} , and \hat{z} are the Cartesian unit vectors. We express the wave vectors for the two generated waves in the general form

$$\mathbf{k}_j = k_{jx}\hat{x} + k_{jy}\hat{y} + k_{jz}\hat{z} \quad (5)$$

for $j = 3$ and 4 . The phase-matching condition $\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3 - \mathbf{k}_4 = 0$ leads to the requirement that the three Cartesian components of the wave vectors of the generated waves must obey the following relations:

$$k_{3x}^2 + k_{3y}^2 = k_{4x}^2 + k_{4y}^2 = k_x^2, \quad (6a)$$

$$k_{3z} = k_{4z} = k_z, \quad (6b)$$

$$k_{3x} = -k_{4x}, \quad k_{3y} = -k_{4y}. \quad (6c)$$

Equations (6a) and (6b) show that the generated fields are emitted in the forward direction in the form of a cone centered on the z axis (see Fig. 4) with the cone angle determined solely by the angle between the two applied fields. Equations (6c) show that the diametrically opposite parts of the conical emission are coupled through the four-wave mixing process described by Eqs. (3).¹⁰ This coupling is especially evident in Fig. 3. We see that wherever two rings cross there is a bright spot on the screen. We expect that the points on the cones that are coupled to the bright spots will also be more intense than other parts of the cones. This effect can be seen by the appearance of bright spots directly opposite the point where any two cones cross.

Under conditions of perfect phase matching and assuming that all fields propagate almost parallel to the z axis, the spatial evolution of the fields A_3 and A_4 is governed, in the steady state, by the equations

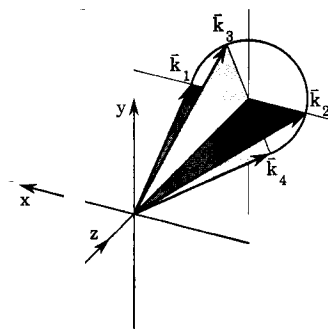


Fig. 4. Illustration of the phase-matching condition $\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3 + \mathbf{k}_4$ for TBE conical emission, where \mathbf{k}_1 and \mathbf{k}_2 are the wave vectors of the applied waves and \mathbf{k}_3 and \mathbf{k}_4 are the wave vectors of the generated waves.

$$\frac{dA_3}{dz} = i2\beta(|A_1|^2 + |A_2|^2)A_3 + i2\beta A_1 A_2 A_4^*, \quad (7a)$$

$$\frac{dA_4^*}{dz} = -i2\beta(|A_1|^2 + |A_2|^2)A_4^* - i2\beta A_1^* A_2^* A_3,$$

where $\beta = 6\pi k\chi^{(3)}$. Equations (7) include only those contributions that are phase matched for TBE conical emission. In each equation, the first term describes a nonlinear phase shift, and the second term describes the forward four-wave mixing process that produces TBE conical emission. We assume that the input fields are undepleted but that their field amplitudes A_1 and A_2 include the nonlinear phase shifts due to their mutual interaction, i.e.,

$$A_1(z) = A_1(0)\exp[i\beta(|A_1|^2 + 2|A_2|^2)z], \quad (8a)$$

$$A_2(z) = A_2(0)\exp[i\beta(2|A_1|^2 + |A_2|^2)z]. \quad (8b)$$

The solution to Eqs. (7) for the fields A_3 and A_4 can then be readily shown to be of the form

$$A_j(z) = B_j \exp(G_- z) + C_j \exp(G_+ z) \quad \text{for } j = 3 \text{ and } 4, \quad (9)$$

where B_j and C_j are constants that represent the spontaneous initiation of the TBE conical emission process. In Eq. (9) the gain coefficients G_{\pm} are given by

$$G_{\pm} = i \frac{3\pi}{c} \beta (I_1 + I_2) \pm \frac{\pi}{c} |\beta| (14I_1 I_2 - I_1^2 - I_2^2)^{1/2},$$

where $I_j = c/2\pi |A_j|^2$.

In the case of equal pump-wave intensities (i.e., $I_1 = I_2 = I$) and large gain [i.e., $(\text{Re } G_{\pm})z \gg 1$], the solutions for the intensities I_3 and I_4 of the generated fields are

$$I_j(z) = |C_j|^2 \exp(gIz) \quad \text{for } j = 3 \text{ and } 4, \quad (11)$$

where $g = 24\sqrt{3}\pi^2 k |\chi^{(3)}|/c$. As can be seen from these expressions, both fields experience exponential spatial growth.

In order to estimate the value of the gain factor g under our experimental conditions, we calculate the nonlinear susceptibility $\chi^{(3)}$ using the predictions of the adiabatic-following model,¹¹ which gives $\chi^{(3)} = 2N|\mu|^4/3\hbar^3\Delta^3$. For our experimental conditions ($N = 3 \times 10^{14} \text{ cm}^{-3}$, $\Delta/2\pi = 40 \text{ GHz}$, $I = 2 \times 10^4 \text{ W cm}^{-2}$, $L = 5 \text{ cm}$, $k = 10^5 \text{ cm}^{-1}$, and $\mu = 6 \times 10^{-18} \text{ esu}$), gIL is approximately equal to 20, which is the typical value of the gain at threshold for many types of stimulated scattering processes.¹²

Equation (10) shows that exponential growth of the TBE conical emission process occurs only when the intensities of the two pump waves are within a factor of ~ 14 from each other. We have observed experimentally that TBE conical emission can be suppressed by introducing a pump imbalance of ~ 20 or greater. The small discrepancy between the theoretical value of 14 and the experimental value of 20 may be due to the fact that for this experiment only it was necessary to use pump intensities that exceeded the saturation intensity and thus the $\chi^{(3)}$ limit no longer applies.

TBE conical emission can form a severe limitation for several other wave-mixing processes. In fact, we have found that phase conjugation by degenerate four-wave mixing in an atomic vapor can be degraded through the generation of TBE conical emission in the forward and backward directions. In this case the TBE conical emission is driven by the forward pump wave and the probe wave (in the forward direction) or by the backward pump wave and the conjugate wave (in the backward direction). Hence, the generated backward traveling wave does not have the correct transverse spatial structure to be the phase conjugate of the probe wave. However, this problem can be avoided through the use of probe and conjugate waves that are much weaker than the pump waves, as mentioned above in our discussion of Eq. (10).

In conclusion, we have described a conical emission process that occurs when two beams intersect in a nonlinear medium and whose origin is a perfectly phase-matched forward four-wave mixing process.

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