PHYSICAL REVIEW A VOLUME 51, NUMBER 4 APRIL 1995

Theory of self-phase-matching

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(Received 17 November 1994)

A theory of the recently discussed and demonstrated novel phenomenon of self-phase-matching [B. Glushko, B. Kryzhanovsky, and D. Sarkisyan, Phys. Rev. Lett. **71**, 243 (1993)] is given by representing the conical beam, produced in the experiments, by a Bessel beam and as a superposition of Bessel beams. The predictions of the theory with the superposition of Bessel beams are in conformity with the observed behavior.

PACS number(s): 42.65.Ky, 32.80.Wr, 33.80.Wz

Phase matching is crucial in harmonic generation. In atomic vapors, for example, it influences the operating pressures, and through Kerr susceptibility it influences the intensities of the fundamental radiation that can be used to produce the third harmonic (TH) radiation [1]. Recently Glushko, Kryzhanosky, and Sarkisyan [2] discussed and demonstrated the phenomenon of self-phase-matching (SPM) for TH generation. Compared to the plane-wave-phase-matching the SPM provides a high degree of tolerance in the fluctuations of the vapor pressure, and in the fluctuations of the intensity of the fundamental. In SPM geometry the fundamental radiation is focused into the nonlinear atomic vapor in the form of a conical distribution of wave vectors [2]—henceforth called the fundamental conical beam. The cone of wave vectors in it makes a semiangle α with the direction of symmetry that is the principal direction of propagation. Every generated photon of the TH is made from three photons of the fundamental. These are picked from the available wave vectors in the fundamental conical beam. The generated TH radiation comes out as a conical beam of semiangle β. The most probable value of β is governed by the basic conservation law of nonlinear optics, viz.,

$$\vec{k}_{\rm TH}=\vec{k}_1+\vec{k}_2+\vec{k}_3. \qquad (1)$$

Here $\vec{k}_{\rm TH}$ is the wave vector of a TH wave generated by the medium and $\vec{k}_1,\vec{k}_2,\vec{k}_3$ are the three arbitrarily chosen wave vectors out of the fundamental conical beam. Equation (1) implies a three-dimensional quadrilateral, which becomes possible if and only if

$$|\vec{k}_{\rm TH}|\leq 3|\vec{k}_f|, \quad k_1=k_2=k_3=k_f=|\vec{k}|. \qquad (2)$$

Equation (2) requires the medium to be negatively dispersive in that

$$\chi^{(1)}(\omega_3)-\chi^{(1)}(\omega_1)<0\ ; \qquad (3)$$

here $\chi^{(1)}(\omega_m)$ is the linear susceptibility of the medium for the radiation of frequency ω_m (ω_1 is the fundamental, ω_3 is the TH). Equation (3) is satisfied in the experiments of Ref. [2].

It must now be noted that several combinations of the azimuths ϕ_1,ϕ_2,ϕ_3 of the vectors $\vec{k}_1,\vec{k}_2,\vec{k}_3$ of the fundamental conical beam can subscribe to the same azimuth ϕ_β of $\vec{k}_{\rm TH}$. The answer to the question, How much is the total subscription of such combinations to the amplitude of the generated TH? has to be obtained. One has thus to examine the growth of the TH under the conditions of SPM. Equation (7) of Ref. [2] does not contain an answer to such a question, as it is based on the plane-wave approximation. The major hurdle in analyzing the generation of the TH from SPM comes from the fact that at the outset one does not have a suitable description of the fundamental conical beam.

A description of the conical beam in the experiments of Glushko, Kryzhanovsky, and Sarkisyan has been obtained as a solution of the suitable diffraction integral by Tewari and Ashoka [3]. The resultant field is described in terms of Lommel functions [4]. The analytical expression, though accurate, is far too complicated for a simplified description of the conical beams. It shall be published elsewhere. The purpose of this Rapid Communication is to report our choice of a simple description of the conical beam. The present analysis is an improved approximation over Eq. (7) of Ref. [2] and can answer the question raised above. The important results of the choice are presented and the details are relegated to a separate paper [5].

The field in the focal region is represented by a Bessel beam (J_0) solution of the scalar wave equation. This is an idealization that suits the purpose. Mainly because, as per the need, the Bessel beam can be thought of as a superposition of plane waves with their wave vectors inclined, by constant angle α, toward the z axis and having all possible values of the azimuth ϕ [6], so that

$$\psi_1^+=A\ e^{i\omega_1 t}e^{-ik_f\cos(\alpha)z}\int_0^{2\pi}e^{-ik_f\sin\alpha[x\cos\phi+y\sin\phi]}\frac{d\phi}{2\pi},$$

$$\psi_1^+=A\ e^{i\omega_1 t}e^{-ik_f\cos(\alpha)z}J_0(k_f\sin(\alpha)\rho), \qquad (4)$$

$$\rho^2=x^2+y^2.$$

A is a constant. Now, using (4), the slowly varying envelope approximation, and disregarding the nonlinear susceptibilities other than the one for TH generation, one finds from the

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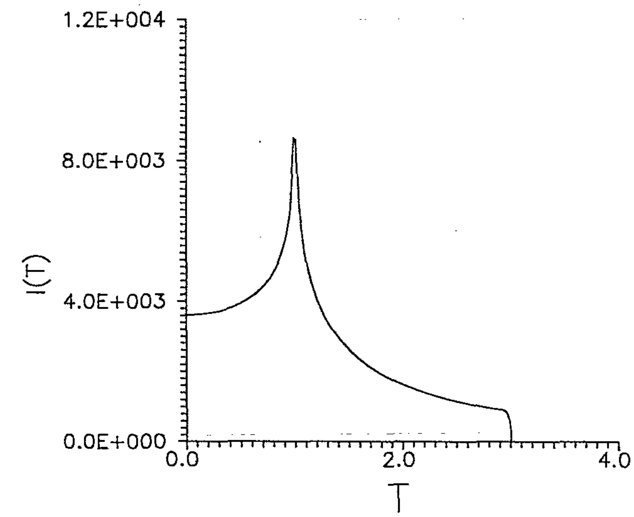

FIG. 1. Transverse phase-matching integral $I(T)$ as a function of variable $T=k_{\mathrm{TH}}\sin\beta/k_f\sin\alpha$. The ordinates are normalized to $I(0)$. The integral is infinite at $T=1$.

scalar inhomogeneous wave equation of nonlinear optics, apart from some phase terms, the amplitude of the generated TH to be

$$\psi_3^{(+)}=2\pi\frac{\omega_3^2A^3}{ik_{\mathrm{TH}}c^2}N\chi^{(3)}(\omega_1,\omega_1,\omega_1)\times\tan\beta\left(\frac{\sin\theta}{\theta}\right)IJ_0(k_{\mathrm{TH}}\sin\beta\rho), \tag{5}$$

$$\theta=\frac{L}{2}(k_{\mathrm{TH}}\cos\beta-3k_f\cos\alpha), \tag{6}$$

$$I=2\pi\int_0^\infty\rho J_0(k_{\mathrm{TH}}\sin\beta\rho)[J_0(k_f\sin\alpha\rho)]^3d\rho. \tag{7}$$

Here L is the length of the medium along the z axis, and for simplicity the medium is assumed to be extended to infinity in transverse directions; N is the number of atoms per unit volume; c is the velocity of light in vacuum; $\chi^{(3)}(\omega_1,\omega_1,\omega_1)$ is the nonlinear atomic susceptibility for TH generation. Unlike Eq. (7) of Ref. [2], Eq. (5) has separate factors for the longitudinal and transverse phase-matching terms, respectively, as $(\sin\theta/\theta)$ and I. The $(\sin\theta/\theta)\rightarrow1$, as $\theta\rightarrow0$, asserts only the longitudinal phase-matching requirement. This is quite different from the usual plane-wave situation whereas $\sin\theta/\theta$ implies the full phase-matching requirement. The integral I has been shown to represent, subject to the conditions (1) and (2), the sum of the subscriptions of all the combinations of azimuths ϕ_1,ϕ_2,ϕ_3 towards an azimuth ϕ_β of $\vec{k}_{\mathrm{TH}}$. This sum is determined by the overlap of the Bessel beam of the generated TH with the cube of the Bessel beam of the fundamental. The behavior of the integral I—henceforth called the transverse phase-matching integral (TPMI)—as a function of the ratio $T=k_{\mathrm{TH}}\sin\beta/k_f\sin\alpha$ is shown in Fig. 1. The transverse coordinate ρ is taken in optical units in the numerical work. From Fig. 1 note that the TPMI is infinite at $T=1$ and is nonzero for $0\leqslant T\leqslant3$.

In order to analyze (5) we concentrate on the function $y(\beta)$,

$$y(\beta)=\tan\beta\left(\frac{\sin\theta}{\theta}\right)I.$$

Note that the limits of variation of the angle β, as fixed by $\sin\theta/\theta$, are given by (λ_1 is the wavelength of the fundamental wave)

$$\frac{\cos\alpha}{\cos\mu}\leqslant\cos\beta\leqslant\frac{\lambda_1}{3L}+\frac{\cos\alpha}{\cos\mu}, \tag{8}$$

$$\cos\mu=\frac{k_{\mathrm{TH}}}{3k_f}. \tag{9}$$

Similarly the limits of variation of β, as fixed by the domain of T for nonzero values of TPMI, are given by

$$0\leqslant\sin\beta\leqslant\frac{\sin\alpha}{\cos\mu}.$$

Now, considering $0<\alpha<\pi/2$ and $\sin\beta\leqslant1$, this gives $0\leqslant\mu\leqslant(\pi/2-\alpha)$ which also implies $0\leqslant\beta\leqslant\pi/2$.

However, for experiments [2], $\lambda_1\ll L$. One notices then that the three sides of the inequality (8) are nearly equal. Thus

$$\cos\beta=\frac{\cos\alpha}{\cos\mu}. \tag{10}$$

For $\cos\beta\leqslant1$, this gives $0\leqslant\mu\leqslant\alpha$ and

$$0\leqslant\beta\leqslant\alpha. \tag{11}$$

Equation (11) agrees well with the range of the angle β observed in Ref. [2]. The behavior of the amplitude ψ_3^+ of TH with pressure (proportional to N) is determined as follows. For a given N, the μ is determined from (9). Using (9) and (10) and a well-known trigonometric identity in the definition of T, one can write

$$T=\frac{3\sqrt{\cos^2\mu-\cos^2\alpha}}{\sin\alpha}.$$

This determines for a μ, a T, which in turn determines from Fig. 1 the value of the TPMI. This is used in $y(\beta)$ and in (5).

The maximum of TPMI occurs at $T=1$, which happens at $\mu=\mu_0$ given by

$$\cos\mu_0=\frac{\sqrt{1+8\cos^2\alpha}}{3}, \tag{12}$$

from which, using trigonometric relations, one finds

$$\tan\beta_0=\tfrac{1}{3}\tan\alpha. \tag{13}$$

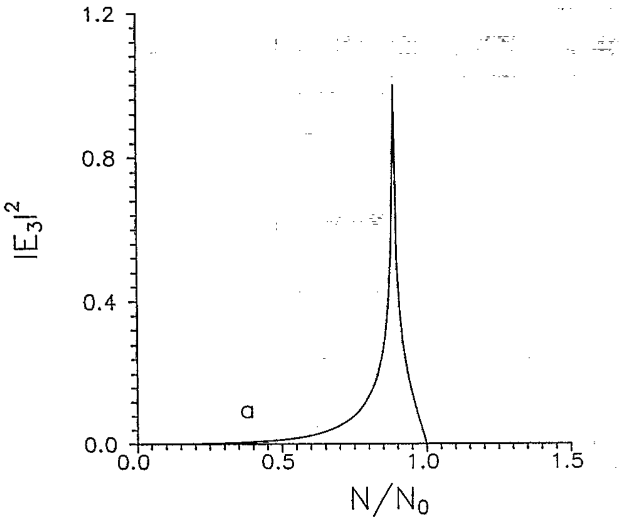

FIG. 2. Detected power [Eq. (14)] of the third harmonic generated by a Bessel beam of angle $\alpha=45$ mrad. $L=10$ cm, $a=5$ $(k_{\mathrm{TH}}\sin\alpha)^{-1}$, $\lambda_1=1.064$ μm. The ordinate is normalized to the maximum for a given A and susceptibility $\chi^{(3)}$.

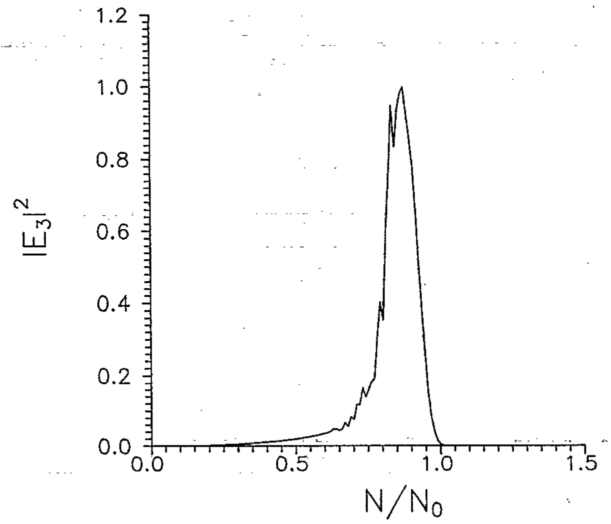

FIG. 3. Detected power (in normalized units) for the case of 11 Bessel beams representing a distribution of Bessel beams in the focal region. Observe the wide width compared to that in Fig. 2. $\alpha_{\min}=44.5$ mrad; $\alpha_{\max}=54.5$ mrad; $d\alpha=1$ mrad. Normalization used is similar to that for Fig. 2.

For small values of α, (13) gives the observed relationship [2] between angles β and α. The behavior of the detected power

$$|E_3|^2=2\pi\int_0^a|\psi_3^+|^2\rho\,d\rho, \tag{14}$$

as a function of N/N_0 (see below for N_0), is shown in Fig. 2. a is the radius of the detector. The curve in Fig. 2 is remarkably similar to that in Fig. 2 of Ref. [2], particularly in the rise of $|E_3|^2$ with an increase of N before the peak is reached and in the nature of the decline of $|E_3|^2$ after the peak is passed. The widths of the two peaks are significantly different.

The reasons for the difference in widths do not lie in the Kerr nonlinearity. This may be understood heuristically in the following way. Let it be assumed that Eq. (5) can, as assumed in Ref. [2] for Eq. (7) (there), be used even in the presence of Kerr nonlinearity with the modification of linear susceptibility to

$$\chi^{(1)}(\omega_3)+\chi^{(S)}(\omega_3)|\psi_1^+|^2=\bar{\chi}^{(1)}(\omega_3). \tag{15}$$

$\chi^{(S)}(\omega_3)$ is the Kerr susceptibility at frequency ω_3. Let it also be assumed that $|\psi_1^+|^2$ is constant. Then the peak in Fig. 2 does not change in shape but merely shifts along the x axis, depending on the change in the value of μ_0 to $\bar{\mu}_0$ obtained by using (15) in (9). We therefore ignore the Kerr susceptibility effects in the following.

There are other differences, too. For example, in the detailed variation with pressure of the angle β of the TH Bessel beam. The maximum in Fig. 2 (here) occurs at β_0 given by (13), where, in the terminology of Ref. [2], $N=\frac{8}{9}N_0$. N_0 is given by $\mu=\alpha$, where one also has from (10) $\beta=0$. The value $N=N_0$ occurs at the right-hand foot of the peak in Fig. 2. Thus β changes from β_0 to 0 as N changes from $\frac{8}{9}N_0$ to N_0, along with the right-hand-side sharp fall of $|E_3|^2$. This is very different from the expectation in Ref. [2], wherein the flatness of the curve in Fig. 2 (there) is attributed to the constructive interference between the two ways the SPM is achieved. In the present calculations the sum of all possible ways in which the SPM can be achieved leads to a sharp drop in intensity for $N>\frac{8}{9}N_0$.

The flatness of the experimental curve may be explained in the following way. We may relax the assumption that a Bessel beam of cone angle α represents the field in the focal region of the experiments. Instead, assume that the focal field is a superposition of Bessel beams lying within minimum $\alpha_{\min}$ and maximum $\alpha_{\max}$ values of the cone angle α. Then, for each α, one has a curve similar to that in Fig. 2. The curve for $\alpha_{\min}$ starts the left leg, and the curve for $\alpha_{\max}$ gives the right falling-off edge of the broad peak. The intermediate region is filled by the coherent superpositions of the apex of all peaks. This situation gives a broad top. The result of the superpositions of 11 different values of α is shown in Fig. 3, displaying a broader peak compared to Fig. 2, and having the similar rise and fall behavior of the intensity on the two legs, as in Fig. 2 of Ref. [2].

We have thus shown that treating the conical beam of Ref. [2] as a superposition of Bessel beams is useful in developing a theory of SPM. A major result of the theory is that the width of tolerance in pressure (proporational to N) observed in the experiments is due to the spread in the cone angle of the Bessel beams. This result can be verified experimentally by changing the annular width of the ring field and the lens position. These changes can produce different values of mean α, and a different spread in α, in the setup of Ref. [2].

Note that in obtaining Fig. 3 all 11 beams are coherent and the generated ψ_3^+ field is determined by a coherent sum of the fields generated by a coherent sum of all incident Bessel beams. Thus the interference effect of the fundamental and the generated TH components has been taken into account in Fig. 3.

Finally it may be added that a high tolerance in the fluctuations of the vapor pressure, in the intensity of the fundamental and in the spectral bandwidth acceptance for efficient TH generation, is a consequence of the broad width of Fig. 3, and is predicted by the present theory as well as by the discussion in Ref. [2], for the self-phase-matching geometry.

The visit of S.P.T. to the Institute of Optics, Rochester, NY, was supported by NSF Grant No. INT 9100 685 to Professor G. S. Agarwal.

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