Spectral and spatial characteristics of third-harmonic generation in conical light beams

V. E. Peet and S. V. Shchemeljov
Institute of Physics, University of Tartu, Riia 142, Tartu 51014, Estonia
(Received 16 April 2002; published 7 January 2003)

Generation of resonance-enhanced third harmonic in Bessel and other conical beams is analyzed from a simple picture, where the fundamental light field is decomposed into elementary configurations of crossed plain-wave sub-beams. We show that the overall harmonic output can be derived as a superposition of all partial harmonic components driven by elementary configurations of the fundamental field. Good agreement with experimental observations has been obtained in simulation of spectral and spatial characteristics of the generated third harmonic. Some peculiarities of harmonic generation in conical light fields are discussed.

DOI: 10.1103/PhysRevA.67.013801 PACS number(s): 42.65.Ky, 32.80.Rm

I. INTRODUCTION

Generation of third harmonic (TH) with Bessel beams in negatively dispersive media was the subject of several experimental and theoretical studies. In experiments of Ref. [1] the conical excitation geometry of a focussed ring-shaped beam has shown its ability to support the phase-matched TH in a broad range of variations of refraction index due to the so-called self-phase-matching (SPM) phenomenon [1]. A theory of this SPM phenomenon was developed soon afterwards [2,3] to analyze the results of Ref. [1]. In this theory, the nonlinear response was derived from direct treatment of the scalar wave equation with the conical source field represented as a Bessel beam. Similar approach was used in further studies on generation of low-order harmonics with Bessel beams [4,5].

The excitation wavelength in experiments of Ref. [1] was fixed and the phase matching was controlled by changing the alkali vapor density. Another experimental procedure was used in Refs. [6–8], where the TH generation in Bessel beams was studied through resonance-enhanced multiphoton ionization of the target gas. In those experiments a tunable laser source was used and the TH excitation profiles were measured near the three-photon atomic resonance of xenon. Ionization technique enabled the TH generation to be studied in condition of very strong absorption when no TH photons exit the gas cell. It was found that the phase-matched TH produced in Bessel beams is responsible for the appearance of broad ionization bands in the negatively dispersive side of the atomic resonance [6–8]. The SPM approach was found to be in satisfactory conformity with the observed behavior [7].

Detailed experimental study with Bessel and other conical beams, however, has revealed incrementally several disadvantages of the SPM theory in description of experimental observations. First, this theory fails to explain the appearance of the on-axis TH component in Bessel beams. For a Bessel beam having an inclination angle α the TH is generated along a cone with an inclination angle β. Depending on excitation conditions, the angle β can be tuned from 0 to α. In SPM equations the amplitude of generated TH is proportional to $\tan\beta$ [2,3] and it yields a sharp cutoff of the calculated dependences of the TH output at $\beta\to 0$ [2,3,7]. In other words, the SPM theory predicts the absence of any TH component directed along the propagation axis of a Bessel beam. However, significant on-axis TH was observed in Ref. [1] in conditions near the maximum of conversion efficiency. Similarly, experiments with segmented conical beams [9] have shown efficient production of the on-axis TH, as well. In a full-aperture Bessel beam, the on-axis TH is responsible for a discernible broadening of the spectral TH excitation profiles toward the atomic resonance [8], whereas the calculated TH profiles are always cut to zero at a point where $\beta=0$ [7].

Similar problem arises with the analysis of spatial TH profiles. If the generated TH field is idealized as a Bessel beam [2,3], the far-field TH profile should be a ring as a cross section of the outgoing TH cone. It was the case for conditions far enough from the maximum of conversion efficiency, but near the maximum significant transformations of spatial profiles were observed [1]. Namely, the increase of vapor density was followed by filling in the empty part of the TH ring without significant decrease in the ring diameter. At some pressure the ring was transformed into a disk. In Ref. [1] these observations were explained as due to the onset of the Kerr nonlinearity. However, the presence of an intense on-axis TH seems to be the general feature of the TH generation with Bessel beams independent on the Kerr effect.

The SPM theory is based on an integral representation of both the source field and the generated TH as Bessel beams. This approach breaks down when the conical wave front of the fundamental light field is disturbed and can no longer be approximated by a Bessel profile. Such situation occurs for a family of non-Bessel beams having conical wave front: segmented [9] and Mathieu beams [10,11]. Segmented conical beams are formed when a part of the conical wave front is blocked [9]. Special case of such segmented beams are the Mathieu beams with their amplitude profile described by radial and angular Mathieu functions [10,11]. All these conical light fields are propagation invariant in the same sense as the Bessel beams, but their amplitude profiles differ essentially from the Bessel pattern. This difference results in significant transformations of the TH excitation profiles when the conical wave front of a Bessel beam is segmented by different masks [9]. However, these observations are not readily explained within the framework of the SPM theory.

Recent papers [8,9] have shown another way to evaluate the excitation problem for conical light beams. The Bessel and the other conical beams can be viewed as a superposition of plane waves with their wave vectors inclined by a constant angle α toward the propagation axis. It has been shown in Refs. [8,9] that the TH excitation profiles for Bessel beams can be analyzed from a simple picture of a few angled plane waves constituting the Bessel beam. In the TH generation process, every TH photon is made from three photons picked from the fundamental light cone. The source field can thus be decomposed into elementary spatial configurations of three sub-beams having some azimuth angles ϕ_1, ϕ_2, and ϕ_3 on the fundamental cone. For a given spatial arrangement of sub-beams, the nonlinear response builds up according to the phase-matching requirements. Since every partial field configuration contributes the overall TH production, the TH output can be found as a superposition of contributions from all possible elementary configurations of the source field. As will be shown below, such an approach gives simple and reliable way to evaluate the TH generation in conical light beams.

The format of the present paper is as follows. In Sec. II we consider the TH generation in an optically thick medium. This approach corresponds to the experimental conditions of Refs. [7–9], where tightly focussed conical beams were used to generate near-resonance TH. In this case, the TH excitation profiles are monitored through resonance-enhanced multiphoton ionization of gas atoms. In Sec. III we consider the TH generation in an optically thin medium, when the TH absorption is negligible. Such approximation is valid to some extent for experimental conditions of Ref. [1].

II. OPTICALLY THICK MEDIUM

First, we note that the problem of the TH generation with a few plane waves is the well-studied subject of nonlinear optics. In an optically thick medium no TH photons exit the gas cell and the generation of resonance-enhanced TH is monitored through three-photon-resonant excitation and subsequent ionization of gas atoms. In this excitation (ionization) process the generated TH field plays a significant role in determining the total transition probability. For a certain threshold product of number density and oscillator strength, the TH field generated by crossed beams evolves in amplitude and phase to interfere destructively with direct three-photon excitation of dipole-allowed transition everywhere except for a region on the blue side of the resonance position [12–14]. At this region the interference becomes constructive and it produces an ionization profile which is identical to the expected pressure-broadened atomic line [12–15]. This Lorentzian profile is located essentially at the frequency where the phase-matched TH is produced by crossed beams. The shift of the Lorentzian from the resonance position is given by the frequency expression for the so-called cooperative shift [12–14].

Recently, the TH excitation profiles for Bessel beams were shown to be principally describable from a simple picture of a few angled plane waves together with the characteristic feature of the cooperative line shifting associating with noncollinear three-photon excitation [8,9]. At any gas pressure the location of the pressure-dependent TH peak in a Bessel beam with an inclination angle α matches exactly the value of the cooperative shift for two plain waves angled by 2α [8]. In Ref. [9] the concept of the cooperative line shift was applied to analyze the TH excitation spectra in segmented conical beams. It has been shown that the overall TH envelope for a Bessel beam can be viewed as a superposition of ''homogeneous'' Lorentzian profiles variously displaced by the cooperative shift. These Lorentzians correspond to different spatial configurations of two and three plain waves from the conical wave front [8,9]. We will use these results in the present analysis of the TH excitation profiles.

In the description of the wave-mixing process in a full-aperture Bessel beam, all combinations of partial waves having any azimuth angle ϕ from 0 to 2π on the light cone are superimposed. For every configuration of two or three crossing plane waves the concept of the cooperative line shift predicts the excitation (ionization) profile as a Lorentzian peak. The location of this peak in the spectrum is determined by the applicable value of the cooperative shift. Remarkable feature of this elementary excitation profile is that for a fixed interaction length, the magnitude of the Lorentzian remains nearly constant over rather large range of detunings from the atomic resonance position [13,15]. This is because the TH gets stronger as the shift gets larger, while the absorption gets weaker at the same rate. Thus, the product of the TH intensity and the atomic absorption remains nearly constant (see Fig. 3 in Ref. [15]).

The excitation (ionization) signal from any partial TH component depends on the interaction length of the pumping sub-beams (gain length) and the overall excitation volume. In a lowest-order Bessel beam, the light is concentrated along the propagation axis forming beamlike region with maximum light intensity. Any nonlinear process driven by such beam is confined to this narrow and extended core near the beam axis. The interaction length within this high-intensity core is $L = d/\sin\alpha$, where d is the diameter of the central lobe of the Bessel beam. Since all sub-beams from the source field have the same inclination angle α, the interaction length L is the same for any configuration of sub-beams. Additionally, in an optically thick medium the propagation distance of the TH light is very short and all the generated TH photons are absorbed entirely within the excitation region. It means that the excitation volume is also the same for all subbeam combinations. Hence, the TH excitation profile in a Bessel beam can be evaluated from a simple picture of equally weighted Lorentzians, where any partial combination of the fundamental sub-beams yields the same ''brightness'' of the corresponding TH peak. These Lorentzians are variously displaced by the cooperative shift, but all of them have equal amplitudes. In segmented and Mathieu beams the intensity profile differs significantly from the Bessel pattern. In this case, the approach of equally weighted Lorentzians may become incorrect since the gain length and (or) the excitation volume may be different for different sub-beams combinations.

For the general case of three interacting waves from a conical wave front, the location of the cooperative line is given by the cooperative shift δ_c written as [9]

$$\delta_c = \Delta_0\left(-1.11 + \frac{18}{\sin^2\alpha[3-\cos(\phi_1-\phi_2)-\cos(\phi_1-\phi_3)-\cos(\phi_2-\phi_3)]}\right), \tag{1}$$

where $\Delta_0 = \pi N F_{01} e^2/2m\omega$, N is the gas density, F_{01} is the oscillator strength, ω is the resonance angular frequency, and ϕ_1, ϕ_2, and ϕ_3 are the azimuth angles of three fundamental waves on the light cone. The angles ϕ_i have all possible choices from 0 to 2π. Neglecting the small constant term, Eq. (1) can be written as

$$\delta_c = \frac{4\delta_0}{3-\cos(\phi_1-\phi_2)-\cos(\phi_1-\phi_3)-\cos(\phi_2-\phi_3)}, \tag{2}$$

where $\delta_0 = 9\Delta_0/2\sin^2\alpha$ determines the location of the ''main'' TH peak. This peak corresponds to the excitation geometry when $\phi_2 = \phi_1 \pm 180°$ and ϕ_3 is arbitrary [8,9]. The minimum value of the cooperative shift is $\delta_c = \frac{8}{9}\delta_0$ and it is realized for symmetrical configuration of three fundamental sub-beams separated by 120°. In this case, $\beta = 0$ and the generated TH is directed along the propagation axis of the Bessel beam.

The inclination angle β of the TH wave vector is given by

$$\tan\beta = \frac{1}{3}(\tan\alpha)\sqrt{3+2(\cos(\phi_1-\phi_2)+\cos(\phi_1-\phi_3)+\cos(\phi_2-\phi_3))}. \tag{3}$$

Thus, Eq. (2) can be written as

$$\delta_c = \frac{8\delta_0}{9\left(1-\frac{\tan^2\beta}{\tan^2\alpha}\right)}. \tag{4}$$

Equation (4) couples the spectral variable δ_c and the angular variable β. The angle β changes from 0 to α when δ_c changes from its minimum value of $\delta_c = \frac{8}{9}\delta_0$ to infinity.

The overall TH excitation profile in a Bessel beam builds up from all elementary TH components. This profile is to be found as a squared sum of all partial TH amplitudes. For an optically thick medium, however, all the generated TH photons are absorbed within the excitation volume. One can suppose that the integral excitation in such conditions can be derived as simple incoherent sum of intensities rather than coherent sum of amplitudes. It simplifies the calculation procedure very much and, as will be shown below, gives very good agreement with experimental observations.

Every elementary TH profile, monitored in an optically thick medium through atomic excitation and subsequent ionization, is a Lorentzian cooperative line with the pressure-dependent width γ and the cooperative shift δ_c. For an excitation frequency ω, the contribution of a Lorentzian line into the overall TH excitation envelope is given by

$$I(\omega) = \frac{\gamma/\pi}{(\omega-\omega_0-\delta_c)^2+\gamma^2}, \tag{5}$$

where ω_0 is the resonance frequency, $\omega_0+\delta_c$ is the central frequency of the Lorentzian line, and δ_c is given by Eq. (2). Cylindrical symmetry of the Bessel beam allows one to fix $\phi_1 = 0$ and the overall signal $S(\omega)$ from the TH generation is then given by an integral over azimuth angles ϕ_2 and ϕ_3:

$$S(\omega) = \int_0^{2\pi}\int_0^{2\pi} I(\omega, \delta_c(\phi_2,\phi_3))\,d\phi_2\,d\phi_3. \tag{6}$$

Figure 1 shows the TH excitation profile calculated using Eq. (6) for $\delta_0 = 18\gamma$. Such δ_0 corresponds to the experimental conditions of Refs. [8,9], where the Bessel beam with $\alpha = 17°$ was used to generate TH near the 6s resonance of xenon. Calculated profile in Fig. 1 shows sharp near-resonance peak followed by a tail toward the blue side of the spectrum. Such shape of the profile has an excellent agreement with the TH excitation bands registered at a moderate

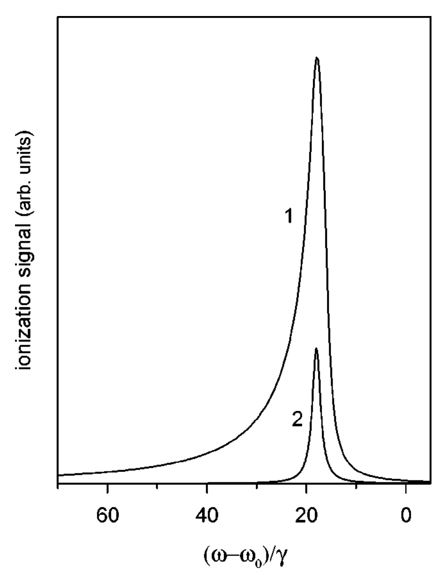

FIG. 1. Near-resonance excitation profiles associated with the phase-matched TH in an optically thick medium: (1) superposition of multiple Lorentzians in a Bessel beam; (2) single Lorentzian peak (not scaled to 1) located at δ_0.

gas pressure in ionization experiments [7,8]. For a high gas pressure, the experimental profiles show similar shape but with less pronounced peak and more intense tail [6,8] because of additional molecular absorption in a dense gas. In agreement with experimental data [8,9], the location of the TH peak coincides the location of the cooperative line at δ_0 (curve 2 in Fig. 1). Note distinct red wing of the calculated profile, where according to the SPM theory the TH should vanish. Red wing covers the region toward and beyond the resonance position at ω_0. Again, it agrees very well with experimental observations [6–8].

The peak of the TH profile occurs at a wavelength where $\delta_c = \delta_0$ and the angle β satisfies the well-known condition [2,3]

$$\tan\beta = \frac{1}{3}\tan\alpha. \tag{7}$$

In the case here, Eq. (7) follows from Eq. (4) at $\delta_c = \delta_0$.

Segmented and Mathieu beams share the propagation properties of a Bessel beam, but their amplitude profiles differ significantly from the Bessel pattern. Thus, the Bessel-beam representation of the fundamental and the TH fields is not valid for such beams. In segmented and Mathieu beams the light energy also propagates along a cone surface, but the light field is comprised of sub-beams from a limited range of azimuth angles [9–11]. Thus, some subbeam combinations and the correspondent TH components of a full-aperture Bessel beam disappear in segmented and Mathieu beams. In this sense, a Bessel beam is simply a particular case of the full-aperture conical excitation geometry when the azimuth angles ϕ_i of sub-beams have any value from 0 to 2π. With the present approach, the source field of a non-Bessel conical filed can again be decomposed into elementary subbeam combinations and the overall TH excitation profile is then constructed from partial TH profiles. The general formalism is the same, except for a restriction of the azimuth angles to a limited extent.

Figure 2 shows the TH excitation profile calculated for a conical beam segmented by symmetrical three-slit mask. Such mask was used in experiments with segmented beams [9] and the experimental TH profile is also shown in Fig. 2. Theoretical profile in Fig. 2 was obtained by numerical evaluation of the integral

$$S(\omega) = \int_{\phi_1}\int_{\phi_2}\int_{\phi_3} I(\omega,\delta_c)\,d\phi_1 d\phi_2 d\phi_3, \tag{8}$$

where $I(\omega,\delta_c)$ is given by Eq. (5) and δ_c is given by Eq. (2). Being segmented by a mask, the range of possible azimuth angles is determined by two factors. First, this range is given by geometry of the used mask. For the case shown in Fig. 2, the mask selected three sub-beams separated by 120°. Second, a finite width of mask slits and diffraction on mask edges give some spreading $\pm\Delta\phi$ of the azimuth angles for every selected subbeam. Thus, for the case considered all the azimuth angles ϕ_i in Eq. (8) run over three ranges of $\pm\Delta\phi$, $120°\pm\Delta\phi$, and $240°\pm\Delta\phi$. Best calculation results were

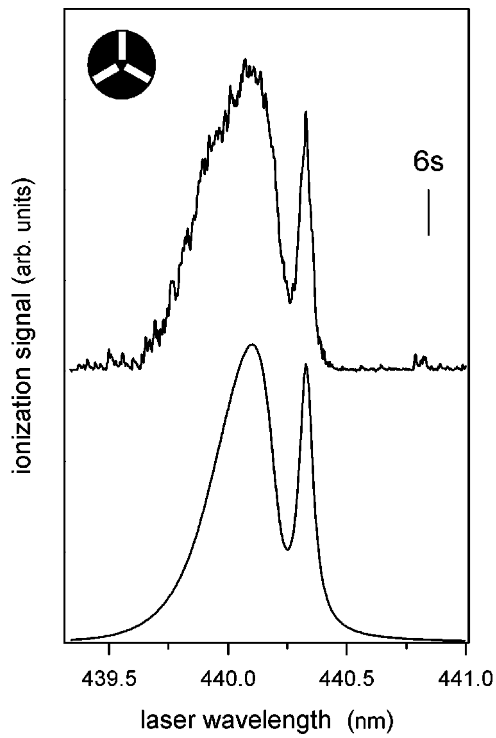

FIG. 2. Near-resonance TH excitation profiles for segmented conical beam. Upper trace, experiment; lower trace, numerical simulation. The inset shows the used slit mask.

obtained for $\Delta\phi = 15° - 20°$, which agrees well with the value of spreading of up to $\pm 20°$ estimated in experiment [9].

The TH profiles in Fig. 2 have two distinct spectral components. Angular spreading of individual sub-beams yields some spectral broadening of the corresponding components in the TH profile. This broadening increases rapidly with an increased δ_c [9]. Sharp near-resonance peak results from three-beam excitation, when all three sub-beams enter the excitation zone from different slits. Spectral spreading of individual Lorentzians in this case is small and the envelope is formed as a sharp and narrow peak located at $\delta_c = \frac{8}{9}\delta_0$. If any pair of sub-beams enter the excitation zone from the same slit (two-beam excitation), the range of δ_c is larger and individual Lorentzians are spread over a more broad spectral range. It yields a second component of the profile as a broad band located at the blue side of the peak. Finally, when all three sub-beams enter the excitation zone from the same slit (single-beam excitation), the range of shift is $\delta_c > 15\delta_0$ and the corresponding TH components are located off the range of interest. Such far wing of the TH envelope is very weak since the spectral density of Lorentzians is reduced rapidly with an increased δ_c.

Our simulation of the TH profile for segmented beam in Fig. 2 was again based on the approach of equally weighted Lorentzians. For the beam considered, such an assumption is correct since the interaction length and the excitation volume are nearly equal for all two- and three-beam combinations of the source field. For other segmented conical beams such an assumption may be incorrect. However, an accurate account for the gain length and the excitation volume will allow the present approach to be applied to other conical beams, as well.

III. OPTICALLY THIN MEDIUM

In conditions of very strong absorption, the TH generation is monitored as an intrinsic process when no TH photons exit the gas cell. The observable quantity in this case is the ionization yield due to atomic excitation and subsequent ionization of the target gas. More interesting for practical applications is the case of weak absorption, when the generated TH photons can leave the cell. In this section we consider the TH generation with conical beams in conditions, when the TH absorption can be neglected.

In experiments of Ref. [1] on the TH generation with conical beams the excitation wavelength was fixed. Thus, the detuning of the excitation frequency from the resonance position was fixed and the phase matching was controlled by changing the vapor density. To avoid the TH absorption, the detuning from the atomic resonance should be large enough. It suggests the use of beams with relatively small inclination angle α. In experiments of Ref. [1] this angle was 10 mrad or about 0.6°, being thus significantly smaller than $\alpha > 10°$ in ionization experiments [7–9]. For large detuning the resonance TH absorption can be neglected and the approach of an optically thin medium can be used. Note, that the medium is negatively dispersive at the TH frequency.

In order to analyze experimental observations of Ref. [1] we again decompose the source field into elementary sub-beam combinations and construct the overall response from partial TH contributions. Observable quantity now is the TH emission and we evaluate the spatial TH profiles and the pressure dependence of the TH output.

Generation of resonance-enhanced TH with crossed plane waves has been analyzed in Refs. [15,16]. It has been shown that for two beams crossed at some angle, both the maximum of TH radiation and the atomic excitation show frequency shift $\Delta\omega$. This shift is written in the present notations as $\Delta\omega = 4\Delta_0/(1-\mu^2)$, where $\mu = \kappa c/\omega$. Parameter κ is determined as $\kappa = (\frac{1}{3}\omega/c)(2\hat{\mathbf{n}}_1 + \hat{\mathbf{n}}_2)$ so that $\kappa^2 = (\omega^2/c^2)[1 - \frac{8}{9}\sin^2\alpha]$, where 2α is the crossing angle. Frequency shift $\Delta\omega$ can be written as $\Delta\omega = 9\Delta_0/2\sin^2\alpha$ and it is just the value of the cooperative shift δ_0 (see above). Thus, for any pair of sub-beams from conical wave front the generated TH emission can be characterized by parameter μ and corresponding frequency shift $\Delta\omega = \delta_c$.

The general case of the TH generation with conical beams involves interaction of three pumping waves. For many of the three-beam combinations of the source field, however, an equivalent two-beam configuration can be found on the conical wave front if $\tan\beta \geq \frac{1}{3}\tan\alpha$. Such three- and two-beam combinations yield the same TH component. No equivalent two-beam combinations exist if $\tan\beta < \frac{1}{3}\tan\alpha$ and all such TH components originate from a three-beam pumping. For these components we generalize κ as $\kappa = (\frac{1}{3}\omega/c)(\hat{\mathbf{n}}_1 + \hat{\mathbf{n}}_2 + \hat{\mathbf{n}}_3)$ and, using Eqs. (3) and (4), we have

$$\mu = \frac{1}{3}\sqrt{9 - 8\frac{\delta_0}{\delta_c}\sin^2\alpha} = \sqrt{1 - \sin^2\alpha(1-\rho^2)}$$
$$\simeq 1 - \frac{\sin^2\alpha}{2}(1-\rho^2), \tag{9}$$

where $\rho = \tan\beta/\tan\alpha$. For a Bessel beam, μ changes from its minimum value of $\cos\alpha$ for the on-axis TH ($\beta = 0$, $\rho = 0$) to 1 for collinear TH ($\beta = \alpha$, $\rho = 1$). Any TH component, regardless of its origin in one-, two- or three-beam pumping process, can be characterized by parameter μ.

Intensity of the TH emission in crossed beams is given by a *sync* function [15]

$$I(\omega) \sim \frac{\sin^2\left(\frac{1}{2}u_\omega\right)}{u_\omega^2}, \tag{10}$$

where $u_\omega = (\omega L/c)(n_3 - \mu)$, and n_3 is the refraction index at the TH frequency (for the fundamental frequency the refraction index $n_1 = 1$ is assumed). Spectral dependence of the TH output in crossed beams has a sideband structure with the principal maximum at $n_3 = \mu$ (see Fig. 4 in Ref. [15]).

For a two-level system the refraction index n_3 can be approximated as $n_3 = 1 - 2\Delta_0/(\omega - \omega_0)$ [8]. Following the terminology of Ref. [1], we determine the gas density N_0, when the TH is peaked along the propagation axis, i.e., $\beta = 0$, $\rho = 0$, and $\Delta_0(N_0) = [(\omega - \omega_0)\sin^2\alpha]/4$. It is easy to see that the density N_0 corresponds to the frequency shift $\delta_c = \frac{8}{9}\delta_0$.

The TH output is then written as

$$I(\omega) \sim (N/N_0)^2 \frac{\sin^2\left(\frac{1}{2}u_\omega\right)}{u_\omega^2}, \tag{11}$$

where $u_\omega = (\omega L/2c)(1 - \rho^2 - N/N_0)\sin^2\alpha$. Circular symmetry of the pumping beam results in a ring-shaped far-field profile of emerging TH. If the far-field radii of the fundamental and the TH lights are R and r, respectively, then $r/R = \tan\beta/\tan\alpha = \rho$. The expression (11) determines the TH intensity as a function of ρ and N/N_0 and, being integrated over radial coordinate ρ, Eq. (11) gives the overall TH output as a function of N/N_0.

Figure 3 shows spatial profiles of the TH output calculated for different values of N/N_0. These numerical results were obtained with the use of Eq. (11) and experimental parameters of Ref. [1], namely, $L = 4$ cm, $\alpha = 10$ mrad, and $\lambda = 1.064\ \mu$m. At a low pressure, the TH is generated along a cone surface close to the fundamental beam. In this case, there are fewer atoms in the cell and the TH intensity is small. With the increased vapor density the TH is increased gradually in intensity and decreased in cone angle. This evolution is followed by filling in the inner part of the TH cone. At $N = N_0$ the far-field TH ring is transformed into a disk. Further, the TH becomes peaked along the beam axis and decreases rapidly. All these transformations follow exactly the experimental observations of Ref. [1]. According to the

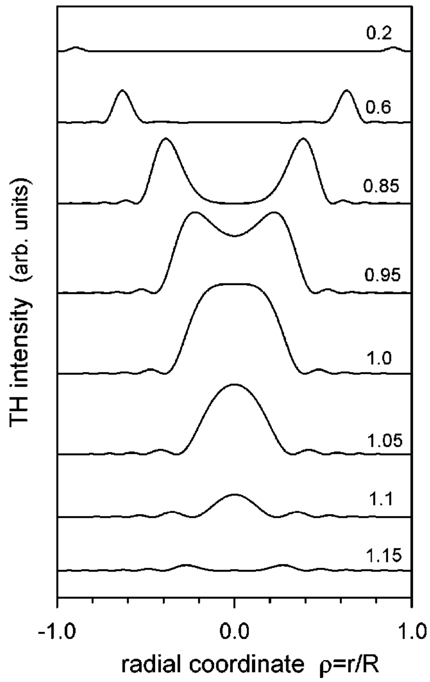

FIG. 3. Spatial evolution of the TH emission with pressure (labeled in units of N/N_0).

SPM theory, however, no on-axis TH should be present in the output beam and the overall TH emission should vanish at $N=N_0$ [2,3]. Note, that in the SPM theory both the source field and the generated TH are represented as Bessel beams. This approximation, together with some window function (e.g., a Gaussian) to describe physically realizable beams, is correct for the fundamental field but becomes questionable for TH. Any Bessel beam can be viewed as a superposition of two conical beams or Hankel waves [17–19]. One of these cones is slanting inwards toward the axis (ingoing cone) and the other away from the axis (outgoing cone). For the fundamental beam the ingoing cone is created by some optical element (e.g., by an axicon lens) and the outgoing counterpart appears on the intersection of the beam axis. Overlap of these cones results in a Bessel profile of the fundamental field. For the TH emission, however, the situation is changed. The source of the TH light is the central lobe of the fundamental beam. It gives the outgoing TH cone, but its ingoing counterpart is absent or very weak. Thus, the Bessel pattern for the generated TH is not established. Being approximated by a Bessel profile, the generated TH is expanded artificially over the whole source field. Such approximation may be good enough to evaluate the TH generation for relatively large β, but it breaks down when $\beta\rightarrow 0$ and the generated TH has much of a character of a plane wave driven along the beam axis. In our approach the source field enters as a set of different combinations of sub-beams and the TH output does not vanish at $\beta=0$ since some particular TH components are driven along the beam axis by symmetrical three-beam combinations of the source field.

Intensity of generated TH as a function of vapor density is shown in Fig. 4. The calculated TH output increases gradually with pressure and maximum conversion efficiency is achieved near $N=\frac{8}{9}N_0$, when $\rho=\frac{1}{3}$ and $\tan\beta=\frac{1}{3}\tan\alpha$. The last again is the known relationship between angles α and β [1–3]. After the peak is passed, the TH output is reduced rapidly till some residual level caused by the presence of weak sidelobes in the TH output.

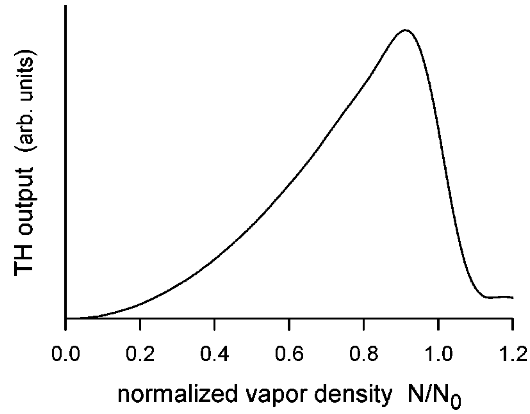

FIG. 4. TH output as a function of vapor density.

The curve in Fig. 4 is remarkably similar to the experimental dependence [1]. The use of the SPM theory results in a much narrow profile peaked at $N=\frac{8}{9}N_0$ and with a sharp cutoff at $N=N_0$ [2,3]. Our calculations give more broad and smooth dependence which simulates the experimental curve rather well. Further improvement of the present approach needs the TH absorption to be taken into account. This absorption is increased with pressure and it will reduce progressively the TH output. The peak of the calculated curve will then be flattened and the residual TH at high vapor density will be suppressed.

We add a few concluding remarks on the TH generation with conical beams. Noncollinear interaction of individual waves in such beams induces nonlinear polarization wave which travels with the phase velocity $v_{\mathrm{exc}}=c/n_1\cos\alpha$ along the beam axis. Momentum conservation imposes the axial phase-matching condition for the wave vectors k_1 of the fundamental and k_3 of the generated TH as

$$k_3\cos\beta=3k_1\cos\alpha, \tag{12}$$

or

$$\cos\beta=\frac{n_1\cos\alpha}{n_3}=\frac{c}{v_{\mathrm{exc}}\, n_3}, \tag{13}$$

which is the known Cherenkov condition, where β is the cone angle of the Cherenkov emission. In other words, the generation of conical TH proceeds as a Cherenkov-type process [20–22] when phase velocity of the driven polarization $v_{\mathrm{exc}}=c/n_1\cos\alpha$ exceeds the phase velocity c/n_3 of the generated radiation. At $\beta\rightarrow 0$ the Cherenkov emission disappears. In the same manner, the TH emission vanishes at $\beta=0$ in the SPM approach.

The threshold condition $\beta=0$ is achieved when $n_3=n_1\cos\alpha\simeq\cos\alpha$. It is easy to see that it occurs when $N=N_0$ and $\delta_c=\frac{8}{9}\delta_0$. If $\delta_c>\frac{8}{9}\delta_0$ $(N<N_0)$, the phase velocity of the driven nonlinear polarization v_{exc} exceeds the phase

velocity c/n_3 of the TH emission. In this case, a Cherenkov-type process is established and the TH output is identical to the Cherenkov cone. When $\delta_c < \frac{8}{9}\delta_0$ ($N > N_0$), the phase velocity of TH exceeds v_{exc}. Phase-matched TH can still be generated and resulting TH emission is directed along the propagation axis. These two regimes of the TH generation may be termed as superluminal ($v_{\mathrm{exc}} > c/n_3$) and subluminal ($v_{\mathrm{exc}} < c/n_3$) ones. Note that in all cases $c/n_3 > c$ since the medium is negatively dispersive. The evolution of spatial TH profiles in Fig. 3 can then be interpreted as a gradual transition from superluminal to subluminal regimes of the TH generation. When gas pressure is low, the TH is produced in a Cherenkov-type superluminal process and the TH output is identical to the Cherenkov cone. This TH cone is squeezed toward the beam axis when pressure is increased. At $N > N_0$ the cone disappears and the TH output, driven by a subluminal excitation process, becomes peaked along the beam axis.

IV. CONCLUSION

Several spectral and spatial characteristics of the TH generation in conical light fields can be evaluated from a simple picture, where the whole excitation problem is decomposed into elementary processes of interaction of a few waves. With this approach, the source field enters a set of different spatial configurations of sub-beams and the overall TH output is derived as a superposition of all partial TH components. This approach does not require any integral representation of the generated TH field and can be applied for any conical beam.

In many cases, which correspond to real experimental conditions, the superposition of TH components can be found as a simple sum of intensities rather than a coherent sum of amplitudes. We have used this approach to analyze the TH production in conditions of strong TH absorption (optically thick medium) and weak absorption (optically thin medium). The former corresponds to the generation of near-resonance TH, when no TH photons exit the gas cell. The elementary TH profile in this case is a pressure-broadened Lorentzian peak (cooperative line) located on the blue side of the atomic resonance. The overall TH envelope is formed as a superposition of such Lorentzians variously displaced by the cooperative shift. Simulation of the TH excitation profiles for Bessel and segmented conical beams has shown very good agreement with experimental observations.

For an optically thin medium, the TH output can similarly be found as a superposition of elementary TH components. Again, good agreement has been obtained in the simulation of spatial evolution and intensity of the generated TH as functions of vapor density. Simple analysis of the TH generation in conical beams has shown that the phase-matched TH in a negatively dispersive medium can be produced by two excitation regimes. When phase velocity of the driven nonlinear polarization exceeds the phase velocity of the generated TH, a Cherenkov-type superluminal process is established and the TH output is similar to the Cherenkov cone. If phase velocity of the TH light exceeds that for the nonlinear polarization, the TH generation is driven as a subluminal process. The TH output in this case is peaked along the propagation axis of the fundamental beam. For a given gas pressure, the subluminal regime is realized on the red wing of the spectral TH excitation profile. If the excitation wavelength is fixed, the subluminal process is established when the gas pressure exceeds optimum for given excitation conditions. In any case, the maximum conversion efficiency is achieved with the realization of the Cherenkov-type superluminal process.

ACKNOWLEDGMENTS

The authors thank W. R. Garrett, V. Hizhnyakov, and A. Sherman for helpful discussions. This work was supported by the Estonian Science Foundation under Grant No. 5030.

[1] B. Glushko, B. Kryzhanovsky, and D. Sarkisyan, Phys. Rev. Lett. **71**, 243 (1993).
[2] Surya P. Tewari, H. Huang, and R.W. Boyd, Phys. Rev. A **51**, R2707 (1995).
[3] Surya P. Tewari, H. Huang, and R.W. Boyd, Phys. Rev. A **54**, 2314 (1996).
[4] C.F.R. Caron and R.M. Potvliege, J. Opt. Soc. Am. B **15**, 1096 (1998).
[5] C.F.R. Caron and R.M. Potvliege, J. Opt. Soc. Am. B **16**, 1377 (1999).
[6] V.E. Peet, Phys. Rev. A **53**, 3679 (1996).
[7] V.E. Peet and R.V. Tsubin, Phys. Rev. A **56**, 1613 (1997).
[8] W.R. Garrett and V.E. Peet, Phys. Rev. A **61**, 063804 (2000).
[9] V.E. Peet, W.R. Garrett, and S.V. Shchemeljov, Phys. Rev. A **63**, 023804 (2001).
[10] J.C. Gutiérrez-Vega, M.D. Iturbe-Castillo, and S. Chávez-Cerda, Opt. Lett. **25**, 1493 (2000).
[11] J.C. Gutiérrez-Vega, M.D. Iturbe-Castillo, G.A. Ramírez, E. Tepichin, R.M. Rodríguez-Dagnino, S. Chávez-Cerda, and G.H.C. New, Opt. Commun. **195**, 35 (2001).
[12] W.R. Garrett, R.C. Hart, J.E. Wray, I. Datskou, and M.G. Payne, Phys. Rev. Lett. **64**, 1717 (1990).
[13] M.G. Payne and W.R. Garrett, Phys. Rev. A **42**, 1434 (1990).
[14] M.G. Payne, W.R. Garrett, R.C. Hart, and I. Datskou, Phys. Rev. A **42**, 2756 (1990).
[15] R. Friedberg, S.R. Hartmann, and J.T. Manassah, J. Phys. B **22**, 2211 (1989).
[16] R. Friedberg, S.R. Hartmann, and J.T. Manassah, Phys. Rev. A **39**, 93 (1989).
[17] S. Chávez-Cerda, G.S. McDonald, and G.H.C. New, Opt. Commun. **123**, 225 (1996).
[18] S. Chávez-Cerda, M.A. Maneses-Nava, and J.M. Hickmann, Opt. Lett. **23**, 1871 (1998).
[19] S. Chávez-Cerda and G.H.C. New, Opt. Commun. **181**, 369 (2000).
[20] I. Golub, Opt. Lett. **15**, 305 (1990).
[21] I. Golub, Opt. Lett. **20**, 1847 (1995).
[22] V. Vaičaitis, Opt. Commun. **185**, 197 (2000).