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Transverse and longitudinal phase-matching in third harmonic generation induced by the Bessel beams

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ABSTRACT

Influence of transverse and longitudinal phase matching on the spectral and intensity properties of third harmonic generation induced by the Bessel pump beams has been investigated both theoretically and experimentally. Transverse phase matching, revealing itself as a ring pattern in the third harmonic intensity angular spectrum was demonstrated in both the normally and anomalously dispersive media. The conditions were found for the optimization of this conical third harmonic signal.

1. Introduction

Third harmonic generation (THG) is an important and well-established technique that can up-convert the coherent output of the laser sources to shorter wavelengths in the visible, UV and VUV spectral regions. However, for the maximum conversion efficiency the process has to be phase matched, i.e., the phase velocities of the fundamental and generated third harmonic waves should be equal. Phase matching in gaseous media can be achieved either by using the mixtures of gasses, or by properly selecting the frequencies of interacting waves [1,2], therefore, it restricts the choice of nonlinear media in which radiation of desired wavelength can be generated. Laser frequency tripling can also be accomplished through the use of higher- order optical nonlinearities [3,4], but in this case the energy conversion efficiency so far has not exceeded 10^{-5}.

The alternative approach is the excitation of nonlinear isotropic media by Bessel pump beams, characterized by specific optical properties [5,6] and allowing different phase matching techniques to be applied. Thus, several reports demonstrating advantages of THG induced by the Bessel pump beams have been published [7–11]. Moreover, it has been shown [12] that the ring-shaped pump beam is capable to provide the self-organized phase matching (SPM) of THG for a wide range of nonlinear media parameters, and thus, can be used for the efficient frequency tripling of broadband and ultrashort laser pulses. Compared to the plane-wave phase matching the SPM provides a high degree of tolerance in the fluctuations of refraction index and pump intensity, but it can take place only in the media with anomalous dispersion of refraction index, i. e., when phase mismatch Δk between the wave vectors of generated radiation and pump is negative ($\Delta k = k_3 - 3k_1 < 0$, where k_1 and k_3 are the fundamental and third harmonic wave vectors, respectively). A more detailed theoretical analysis [13,14] has shown that the power of THG induced by Bessel beams depends mainly on two factors: first, on the longitudinal phase matching (LPM) term, and, second, on the transverse phase matching (TPM). However, the influence of TPM and LPM on the spatial spectra of THG induced by the Bessel beams has not been investigated in detail, especially in the case of normally dispersive nonlinear media, i.e., when the longitudinal phase matching condition cannot be satisfied. Therefore, in this paper we provide the comparison of theoretical and experimental results demonstrating conical TH signal generated in both the normally and anomalously dispersive media.

2. Theoretical background

Here we present an analytical consideration of the third harmonic generation in sodium vapor excited by the Bessel beam. In a paraxial approximation the variation of third harmonic amplitude A_3 with propagation in nonlinear isotropic medium at low conversion efficiency can be described by equation

$$\frac{\partial A_3}{\partial z} = -\frac{i}{2k_3}\left(\frac{1}{r}\frac{\partial}{\partial r} + \frac{\partial^2}{\partial r^2}\right)A_3 + i\sigma A_1^3(r)\exp(i\Delta_z z), \tag{1}$$

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here z and r are the longitudinal and radial coordinates respectively, σ is the coupling coefficient. As a boundary condition of the Eq. (1) at $z=0$ we take the amplitude $A_1(r)$ of the fundamental wave described as an apertured zeroth-order Bessel–Gauss beam

$$A_1(r) = a_0 J_0(k_1 r \sin\theta_1)\exp\left(-r^2/d^2\right), \tag{2}$$

where θ_1 is a half-cone of the Bessel beam and d is the radius of the Bessel–Gauss beam profile, i.e., the radius of the Gaussian function which suppress the Bessel beam oscillations in space. In a direction of propagation the phase mismatch Δ_z for the Bessel beam can be written as

$$\Delta_z = k_3 - 3k_1\cos\theta_1, \tag{3}$$

where $k_3 = 3\omega n_3/c$ and $k_1 = \omega n_1/c$ stand for the wave vectors of the third and fundamental harmonics, respectively. In the paraxial approximation Eq. (3) takes a form

$$\Delta_z = \frac{3\pi}{\lambda_1}\left(2\Delta n + \theta_1^2\right), \tag{4}$$

where λ_1 is a wavelength of the fundamental beam, and $\Delta n = n_3 - n_1$.

A two dimensional Fourier transformation of the Eq. (1) yields

$$\frac{\partial S_3}{\partial z} = \frac{ik_3\theta_3^2}{2}S_3 + i\sigma a_0^3 e^{i\Delta_z z}\int_0^\infty e^{-3r^2/d^2} J_0^3(k_1\theta_1 r) J_0(k_3\theta_3 r) r dr, \tag{5}$$

here $S_3(\theta_3, z)$ denotes an amplitude of the TH angular spectrum. Eq. (5) can be easily solved, and, as a result, an intensity of angular spectrum of the third harmonic $I_3(\theta_3) = |S_3(\theta_3)|^2$ is given by

$$I_3 = S_0^2\left(\frac{l}{L_n}\right)^2 T^2(\theta_3)\mathrm{sinc}^2(\delta l/2), \tag{6}$$

here $S_0 = a_0/(k_1\theta_1)^2$, l is a propagation distance in the nonlinear medium, $L_n = 1/(\sigma a_0^2)$ is the nonlinear length,

$$T(\theta_3) = \int_0^\infty e^{-3\xi^2/m^2} J_0^3(\xi) J_0(3\xi\theta_3/\theta_1)\,\xi d\xi \tag{7}$$

is a transverse phase-matching integral at $n_1 \approx n_3$, $m = k_1\theta_1 d \gg 1$, and

$$\delta = \Delta_z - \frac{k_3}{2}\theta_3^2 = \frac{2\pi}{\lambda_1}\left(2\Delta n + \theta_1^2 - \theta_3^2\right). \tag{8}$$

The dependence of $T(\theta_3)$ is presented in Fig. 1. The transverse phase-matching integral depends on the overlap of the fundamental and third harmonic beams. It clearly peaks at $\theta_3/\theta_1 = 1/3$, and in this case the simultaneous longitudinal phase-matching ($\delta = 0$) is possible at $2\Delta n + \theta_1^2 = \theta_3^2$ [12].

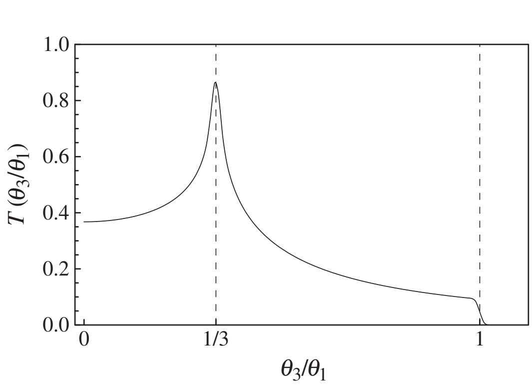

Fig. 1. Transverse phase matching integral, $m = 100$.

Further we analyze THG in a low pressure sodium vapor for the two wavelengths of the fundamental beam (1.62 and 1.86 μm, which correspond to the anomalously and normally dispersive media, respectively).

The refraction indices of the fundamental and third harmonic waves can be calculated approximately by using the relation [15]:

$$n = 1 + \frac{4.3694 \cdot 10^{-6} N}{2.8796 - 1/\lambda^2}, \tag{9}$$

where N is a density of sodium vapor taken in 10^{16} cm^{-3}. In the further calculations we take $N = 0.3$ and $\theta_1 = 7.25 \cdot 10^{-3}$. Note, that for the typical experimental conditions (pump peak intensity of up to 20 GW/cm^2) the change of the refraction index due to the Kerr effect is negligible ($\delta n < 10^{-9}$ for $\chi^{(3)}(\omega) \approx 10^{-33}$ (esu) [15]).

2.1. Anomalously dispersive medium

For $\lambda_1 = 1.62\mu$m we obtain $\Delta n = n_3 - n_1 = -2.91 \cdot 10^{-6}$. In this case the longitudinal phase-matching ($\delta = 0$) is possible for the third harmonic cone with an angle $\theta_3 = \sqrt{\theta_1^2 - 2|\Delta n|} \approx 0.94\theta_1$. The third harmonic generation at a cone angle $\theta_3 = \theta_1/3$ takes place at some phase-mismatch with a fundamental wave, and for this process the coherence length is

$$L_{\mathrm{coh}}(\theta_1/3) = \frac{\pi}{\delta} = \frac{\lambda_1}{3(8/9\,\theta_1^2 - 2|\Delta n|)} \approx 13.2 \text{ mm}. \tag{10}$$

The normalized angular spectrum of third harmonic intensity $I_{3n} = I_3 L_n^2/S_0^2$ is presented in Fig. 2. The spectrum consists of two rings with the angles $\theta_3 \approx \theta_1$ and $\theta_3 \approx \theta_1/3$. The intensity ratio η_a of these two rings is

$$\eta_a = \frac{I_3(\theta_1/3)}{I_3(\theta_1)} = \frac{4}{\pi^2}\frac{L_{\mathrm{coh}}^2(\theta_1/3)}{l^2}\frac{T^2(\theta_1/3)}{T^2(\theta_1)}. \tag{11}$$

As a result, the ring with an angle $\theta_3 = \theta_1/3$ generated at a phase-mismatch should be detected simultaneously with the ring $\theta_3 \approx \theta_1$ if the nonlinear medium length is approximately equal to the uneven number of the coherent length $L_{\mathrm{coh}}(\theta_1/3)$.

2.2. Normally dispersive medium

For $\lambda_1 = 1.86\mu$m we obtain $\Delta n = 4.23 \cdot 10^{-6}$. In this case the generation of two third harmonic rings with the angles $\theta_3 \approx \theta_1$ and

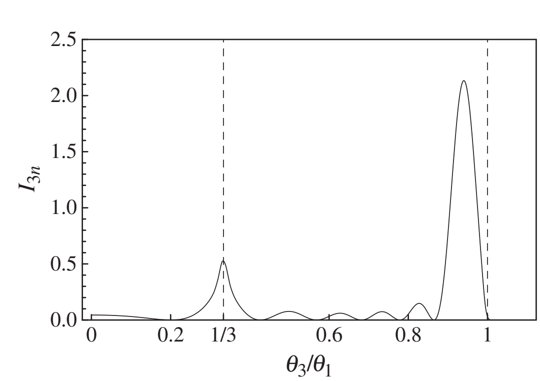

Fig. 2. Angular spectrum of the third harmonic intensity excited in sodium vapor at $\lambda_1 = 1.62\mu$m. Anomalously dispersive medium, $l = 11L_{\mathrm{coh}}(\theta_1/3)$.