ON THE THEORY OF THIRD HARMONIC GENERATION IN A FABRY–PEROT CAVITY

Sh.Sh. AMIROV[1,2], N.V. KERIMLI[1], N.H. GURBANOVA[1], H.A. ABIYEV[1]

[1]Department of Medical and Biological Physics, Azerbaijan Medical University
167 S.Vurgun str., Az-1022, Baku, Azerbaijan
[2]Department of Physics and Electronics, Khazar University, 41 Mahsati str., Az 1096, Baku, Azerbaijan
Corresponding author :E-mail: phys_med@mail.ru

An analytical expression for the efficiency of conversion into third harmonic in a Fabry-Perot cavity filled with the third order nonlinear medium is obtained in the constant intensity approximation. This approximation takes into account the reverse reaction of excited harmonic wave on the phase of fundamental radiation. It was shown that in order to achieve highest conversion efficiency the optimum values of problem parameters like coefficient of reflection, nonlinear length of medium etc. should be chosen.

Keywords: third harmonic, Fabry-Perot cavity, nonlinear medium, constant intensity approximation.
PACS:78.67. Pt; 42.65-k; 42.62.Hk; 42-70a.

1. INTRODUCTION

Third harmonic generation plays a vital role in optical communications, biological microscopy etc. At Fabry-Perot cavity resonant wavelength in near-infrared regime, the efficiency of THG from a 50 nm thick amorphous GST225 planar film is boosted, by 422 times compared to that of non-resonant conditions [1]. Third harmonic generation in layered or periodically modulated media has been drawing considerable attention [2]. Calculations suggest the possibility of all optical switch at third harmonic frequency controlled by the parameters as intensity and frequency of the fundamental wave [3]. Second harmonic generation occurs at ordered non-centrosymmetric structures that are observed mostly in crystals as wel as metals [4]. Biological materials contain wide-scale non-centrosymmetric buildings with coiled view strucrures and polymeric proteins, for instance, at striated muscle and fibrillar collagen [5]. Generation of third harmonics is due to tripling of the frequency of fundamental wave that means conversion of three photons into one photon of a triplred energy [6]. Third harmonic generation occurs at structural inerfaces, such as local transitions of the refractive index or third-order nonlinear susceptibility [7]. For the Fabry-Perot cavity as wel as V-sahaped cavities authors have derived the rate of conversion of the fundamental frequencies into the second harmonic and the sum frequencies [8]. Third harmonic generation microscopy is a label-free scatter process that is elicited by water-lipid and water protein interfaces, including intra-and extracellular membranes, and extracellular matrix structures [9].

2. THEORETICAL APPROACH AND DISCUSSIONS

In this paper we present the results of analysis of third harmonic generation in the Fabry-Perot cavity filled with medium of third order nonlinear susceptibility. Unlike the constant field approximation , here calculations are carried out in the constant intensity approximation [10] taking into account reverse reaction of generated harmonics on the phase of fundamental. Earlier we have employed this approximation for analysis of frequency conversions in traditional nonlinear crystals [11] as well as metamaterials having negative refractive index in a specific frequency region[12-14].

For the analysis of third harmonic generation in an external cavity we assume that the fundamental wave with frequency ω_1 is normally incident on the nonlinear medium with cubic nonlinearity. As a result of nonlinear interaction between incident wave and nonlinear medium the thir harmonic of frequency $\omega_3 = 3\omega_1$ is generated. The process of generation of third harmonics is described by the following system of truncated equations:

$$\pm\frac{dA_1^{\pm}}{dz} + \delta_1 A_1^{\pm} = -i\gamma_1 A_3^{\pm}(A_1^{\pm})^{*2} e^{\pm i\Delta z}$$
$$\pm\frac{dA_3^{\pm}}{dz} + \delta_3 A_3^{\pm} = -i\gamma_3 (A_1^{\pm})^{3} e^{\mp i\Delta z} \qquad (1)$$

where $A_1^{\pm}$ and $A_3^{\pm}$ are the complex amplitudes at frequencies ω_1 and $\omega_3 = 3\omega_1$ respectively. Here $\Delta = k_3 - k_2 - k_1$ - is the difference of wave numbers; γ_j - are the nonlinear coupling coefficients; δ_j - are linear absorption coefficients (j=1,2,3). In an external cavity the above set of truncated equations is solved according to the following boundary conditions:

$$A_3^-(l) = R_3 A_3^+(l) e^{-i2k_3 l}\ ;\ A_3^+(0) = R_{30} A_3^-(0)\ ;$$
$$A_1^+(0) = A_{10}\ ; \quad A_1^-(l) = 0 \qquad (2)$$

131, H.Javid ave, AZ-1143, Baku
Institute of Physics
E-mail: jophphysics@gmail.com

Here R_{30}, R_3 –are the complex reflective coefficients of third harmonic for the left and right mirrors respectively, when the wave is incident from nonlinear crystal. In the case of consideration fundamental wave freely passes through cavity ($R_{10}, = R_1 = 0$) and third harmonic wave undergo multiple reflections ($R_{30},\ R_3 \neq 0$).
Substituting $A_3^{\pm}(z) = a_3^{\pm}(z)e^{[-(\delta_3+3\delta_1+i\Delta)z/2]}$ in above set of equations (1) yiedlds

$$\frac{d^2 a_3^{\pm}}{dz^2} + \mu^2 a_3^{\pm} = 0 \tag{3}$$

where $\mu = \left[3\Gamma_3^2 + \left(\frac{\delta_3-3\delta_1-i\Delta}{2}\right)^2\right]^{1/2}$
Solving the equation (3) for the complex amplitude of third harfmonic in the constant intensity approximation gives :

$$A_3^{+}(l) = \frac{-i\gamma_3 A_{10}^2 l sinc\mu l\cdot e^{[-(\delta_3+3\delta_1+i\Delta)l/2]}}{1-r_{30}r_3 K\cdot e^{[-\frac{(\delta_3+3\delta_1)l+i(\varphi+2k_3 l+\Delta l)}{2}]}} \tag{4}$$

where $K = cos\mu l + [(\delta_3 + 3\delta_1 + i\Delta)/2\mu]sin\mu l$, $\Gamma_3^2 = \gamma_1\gamma_3 I_{10}^2$, $r_{30}, r_3 -$ are absolute values of coefficients of reflection and $\varphi = \varphi_{30} + \varphi_3$ are the phase shifts for harmonic wave at mirrors.
From (4) we get expression for the intensity of third harmonics at the output mirror ($r_{30} = 1$) :

$$I_3^{+}(l) = \frac{\gamma_3^2 I_{10}^3 \chi^{-1}(sin^2 x ch^2 y + sh^2 y cos^2 x)exp[-(\delta_3+3\delta_1)l]}{1-2r_3\chi_1 cos\Psi\cdot exp[-(\delta_3+3\delta_1)l/2] + r_3^2\chi_1^2\cdot exp[-(\delta_3+3\delta_1)l]} \tag{5}$$

here $x = \chi^{1/2} l cos\theta/2$, $y = \chi^{1/2} l sin\theta/2$, $\chi = \left[3\Gamma^{\ 2} + \frac{\Delta^2}{4} - \frac{(\delta_3-3\delta_1)^2}{4}\right]^2 +$
$+\frac{\Delta^2(\delta_3-3\delta_1)^2}{4}$; $\theta = arctg\left[\frac{\Delta(\delta_3-3\delta_1)/4}{3\Gamma^{\ 2}+\frac{\Delta^2}{4}-\frac{(\delta_3-3\delta_1)^2}{4}}\right]$;

$$\chi_1^2 = (cosxchy + asinxchy - bcosxchy)^2 + (bsinxchy + acosxshy - sinxshy)^2$$

$$a = \frac{\Delta y+(\delta_3+3\delta_1)x}{2(x^2+y^2)} l\ ;\ b = \frac{\Delta x-(\delta_3-3\delta_1)y}{2(x^2+y^2)} l\ ,\ \Psi = \varphi + 2k_3 l + \frac{\Delta l}{2} - \theta_1$$

$$\theta_1 = arctg\frac{bsinxchy + acosxshy - sinxchy}{cosxchy + asinxchy - bcosxshy}$$

From the obtained expression for the intensity of third harmonics we can get efficiency of frequency conversion into third harmonic: $\eta = I_{3,out..}^{+}/I_{10}$

$$\eta = \frac{\gamma_3^2 I_0^2 \chi^{-2}(sin^2 x + sh^2 y)exp[-(\delta_3+3\delta_1)l]}{1-2r_3\chi_1 cos\Psi\cdot exp[-(\delta_3+3\delta_1)l/2] + r_3^2\chi_1^2\cdot exp[-(\delta_3+3\delta_1)l]} \tag{6}$$

where $I_{3,out.}^{+} = I_3^{+}(l)(1 - r_3^2)$
When $r_3 = 0$ from the formula (6) we obtain

$$\eta_0 = \gamma_3^2 I_0^2 \chi^{-2}(sin^2 x + sh^2 y)exp[-(\delta_3 + 3\delta_1)l] \tag{7}$$

Using last two formulas gives us gain ($\tilde{\eta} = \eta/\eta_0$) due to use the resonator

$$\tilde{\eta} = \frac{(1-r_3^2)}{1-2r_3\chi_1 cos\Psi\cdot exp[-(\delta_3+3\delta_1)l/2] + r_3^2\chi_1^2\cdot exp[-(\delta_3+3\delta_1)l]} \tag{8}$$

As can be seen from above expression one of the ways ofto acheive the maximum gain is fulfilling the resonance phase condition ($\Psi = 2n\pi$):

$$\varphi + 2k_3 l + \frac{\Delta l}{2} - \theta = 2n\pi \tag{9}$$

Thus in the constant intensity approximation resonance phase condition depends on the pump intensity. At definite values of other parameters of problem condition $\tilde{\eta} > 1$ implies limitation to the absolute value of coefficient of reflection r_3 of harmonic wave: $r_3 < 2\mu cos\,\Psi/(1+\mu^2)$. Other condition for obtaining maximum frequency conversion serves choosing optimum value of reflective coefficient.

Theoretical analysis showed that the gain due to use of cavity has pronounced maxima depending on the coefficient of reflection r_3 and reduced length of nonlinear crystal has pronounced maxima. Gain reaches its maxima at optimum values of coefficient of reflection depending on other parameters of problem. Also it was found that in order to obtain effective frequency conversion it is reasonable to choose optimum length of nonlinear crystal. Optimum length of crystal decreases with increase in difference of wave numbers of interacting waves.

3. CONCLUSIONS

On the basis of above stated one can conclude that resonance phase condition determines the effective frequency conversion in third order nonlinear medium. This condition is a function of intensiy of fundamental wave. When reflective coefficient does no agree with inequality obtained in this paper the employment of resonator is not reasonable. An optimum value of coefficient of reflection depends on nonlinear length of medium, intensity of pump wave , phase mismatch etc.

---

[1] *C. Tun, L. Kuan, T. Yutao* et al. Giant enhancement of third harmonic generation from based Fabry-Perot cavity. Archiv, 2019, physics,1901.08795.

[2] *N. Hashizume, M. Ohashi, R. Ito* Opical harmonic generation in multilayered structures: a comprenhensive analysis. J. Opt. Soc. Am., 1995, B12, pp.1894-1904.

[3] *J. Jose, S.D. Gupta* . Third harmonic generation in layered media in presence of optical bistability of the fundamental. Pramana, journal of Physics, 1998, v. 50, No.3, pp.239-251]

[4] *S.Y. Chen, C.Y. Chen, and C.K. Sun.* Epi-third and second harmonic generation microscopic imaging of abnormal enamel. Opt.Express, 2008, v.16, pp. 11670-11679.

[5] *P. Campagnola, A. Millard, M. Terasaki M.* Three dimensional high resolution second-harmonic generation imaging of endogeneous structural proteins in biological tissues. Biophysics 2002, J.82483-50. https.//doi.org/10.1016/50006-3495(02)75414-3 2002.

[6] *Y. Barad, H. Eisenberg, M. Horowitz, and Y. Silberberg.* Nonlinear scanning laser microscopy by third harmonic generation. Appl.Phys. Lett., 1997, 70, pp.922-931.

[7] *M. Muller, J. Squier, K. Wilson, and G. Brakenhoff G.* 3D microscopy ofd transparent objects using third-harmonic generation. J. Microscopy,1998, v.191, pp.266-274.

[8] *L. Fri , P. Mandel, E. Viktorov* . Intracavity second harmonic generation in a Fabry-Perot resonator: Polarization effects.Quantum and Semiclassical optics: Joiurnal of the European optical Society,1998, part B, v.10, No.1, pp.1355-1363.

[9] *B. Weigelin, G.J. Bakker and P. Friedl*. Third harmonic generation microscopy of cells and tissue organization. Journal of Cell Science, 2016, v. 1 No. 11, pp.32-43.

[10] *Z.H. Tagiev, A.S. Chirkin*. Fixed intensity approximation in the theory of nonlinear waves Zh.Eksp. Teor. Fiz.1977, 73, pp.1271-1282.

[11] *Sh.Sh. Amirov, Z.H. Tagiyev*. Conversion of frequencies of ultrashort pulses in external cavity. Optics and Spectroscopy, v.69, No. 3. pp.678-686.

[12] *R.J. Kasumova, Sh.Sh. Amirov*. Nonstationary sum frequency generation in inhomogeneous optical fiber. AJP "Fizika"(EN), 2022, Vol.XXVIII, No. 2, p.24-30.

[13] *R.J. Kasumova, Sh.Sh. Amirov, Sh.A. Shamilova.* Parametric ineraction of optical waves in metamaterials under low-frequency pumping. Quantum Electronics.2017. v.47, No.7,pp.655-660.

[14] *R.J. Kasumova, Z.H. Tagiev, Sh.Sh. Amirov, Sh.A. Shamilova, G.A. Safarova.* Phase effects at parametrical interaction in metamaterials. Journal of Russian Laser Research. 2017. V.38.p.211-218.

Received: 19.09.2023