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THEORETICAL ESTIMATION OF THE THIRD ORDER NONLINEAR OPTICAL POLARIZATION AT DIFFERENT WAVELENGTHS

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ABSTRACT

Third order nonlinear optical polarization ($P^{(3)}$) plays key role for the characterization of the material systems possessing third order nonlinear optical susceptibility ($\chi^{(3)}$). A theoretical calculation has been performed to estimate the third order nonlinear optical polarization $P^{(3)}$ for alpha quartz, BK-7 glass and fused silica. Values of the estimated third order nonlinear optical polarization have been calculated at 457 nm, 488 nm and 514 nm wavelengths emitted from a continuous wave Argon ion laser. Optical powers of the laser beam were converted into corresponding electric field strengths. Estimated values of the electric field intensity were used to estimate the magnitude of third order polarization for alpha quartz, BK-7 glass and fused silica. It was found that the third order nonlinear optical polarization increases linearly as the beam power is increased. Consequently, third order optical polarization varies nonlinearly with the applied field strength. These observations hold good for alpha quartz, BK-7 glass and fused silica at all three available wavelengths. BK-7 glass exhibits maximum third order nonlinear polarization among three samples. Otherwise fused silica has least third order nonlinear optical polarization. The estimated values of the third order nonlinear optical polarization were found to be of the order of 10^3 FVm^{-2}. Values of the third order nonlinear optical polarizations are strongly affected by the strength of the applied optical field since third order polarization varies cubically to the applied optical field. Third order nonlinear optical polarization can be estimated for other materials if values of third order nonlinear optical susceptibility are known.

Keywords*: Third order nonlinear optical polarization, Continuous wave laser, Third order susceptibility.*

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1. INTRODUCTION

Materials possessing third order nonlinear optical properties are crucial because they are vastly being used in optical information processing, data storing, optical computing, optical sensor manufacturing applications [1-2]. In recent decades different crystals as well as glass mediums are drawing attention of the researchers in the field of nonlinear optics. These materials are capable to ignite stable nonlinear response to the applied optical field. Different phenomena that occur due to the third order optical susceptibility of the nonlinear optical materials are used to characterize the nonlinear refractive index [3-6].

M. Sheikh-bahae used highly sensitive single-beam Z-scan technique for the determination of both the sign and magnitude of the third order optical nonlinearity for solids and liquids [7, 8]. Hong *et al* proposed a new model Mach-Zehnder interferometer, to study the interfered intensity modulation under external magnetic fields [9]. A pump-probe system was implemented in the Mach-Zehnder interferometer by Boudebs *et al* to characterize the third order nonlinear optical polarization of chalcogenide glasses and poor optical quality samples [10-12]. P. D. Maker and R. W. Terhune performed series of experiments to study induced third order optical polarization in the electric field strength using giant pulsed ruby laser [13]. D. A. Kleinman investigated both second and third order dielectric polarization in quartz crystals [14]. A. Tokmakoff calculated tensor components of the third order nonlinear optical polarization for different isotropic media [15]. Geskin at all performed quantum-chemical analysis of the molecular structure to investigate the third order nonlinear characteristics in nonlinear optical (NLO) chromophores, zwitterionic ammonio/borato diphenylpolyenes [16]. Bjorklund studied effects of focusing of laser beam on third order optical materials for several third order optical processes [17].

In this research work numerical values of the third order nonlinear optical polarization $P^{(3)}(t)$ have been estimated theoretically. Values of $P^{(3)}(t)$ has been estimated for alpha quartz, BK-7 glass and fused silica for different wavelengths. Applied electric field intensity has been considered for the case of laser radiation emitted from a continuous wave Argon-ion laser. The laser is capable to emit radiation at 457 nm, 488 nm and 514 nm wavelengths starting from 3 mW to 33 mW optical powers.

2.1 NONLINEAR POLARIZATION

Under the influence of external electric field, the nucleus of an atom is pushed toward the applied field direction whereas the electrons accumulate in the opposite direction. The situation is commonly known as *polarization* and a dipole moment is set up in the direction of the applied field. The induced space charge densities are the source of the induced dipole moment. The induced dipole moment p(r, t) is related to the applied electric field strength by the following equation,

$$p(r,t) = \alpha E(r,t)\text{...............}(1)$$

α is the atomic polarizability that characterizes the capability of an atom to be polarized for applied field [18].

The polarization P(r, t) describes the response of the medium to the applied field and provides information on the induced dipole density. When E is not too strong, the polarization is related to E(r, t) by the following equation,

$$P(r,t) = \varepsilon_0 \chi_e E(r,t)\text{..............}(2)$$

The constant of proportionality χ_e is the electric susceptibility of the medium which describes the response of the medium and depends on the microscopic structure of the medium. ε_0 is known as the permittivity of free space. For a symmetrical molecule, general form of equation (2) is given by,

$$p_x = \varepsilon_0 \chi_{exx} E_x + \varepsilon_0 \chi_{exy} E_y + \varepsilon_0 \chi_{exz} E_z \text{...............}(3a)$$
$$p_y = \varepsilon_0 \chi_{eyx} E_x + \varepsilon_0 \chi_{eyy} E_y + \varepsilon_0 \chi_{eyz} E_z \text{...............}(3b)$$
$$p_z = \varepsilon_0 \chi_{ezx} E_x + \varepsilon_0 \chi_{ezy} E_y + \varepsilon_0 \chi_{ezz} E_z \text{...............}(3c)$$

The set of the nine co-efficients $\chi_{e_{xx}}, \chi_{e_{xy}} \dots\dots \chi_{e_{zz}}$ constitute the susceptibility tensor [19-20]. Equation (3) is the basis of dielectric theory, which is based on the assumption that the material system responds to the external field E(r, t) instantaneously. Any physical system requires a finite time to respond to the applied field. If time lag is considered, then in the real situation, the measured dipole moment at any time t at position r is the outcome of the applied field E(r, t) which was applied on the system at some preceding time t′. The system seizes the characteristic time τ = t - t′ to produce the effect of the external field. Equation (3) takes the following form,

$$P(r,t) = \varepsilon_0 \int_{-\infty}^{+\infty} dt' \chi(t-t') E(r,t')\text{...............}(4)$$

Since the material system always responds to the past behaviour of an optical field,

$$\chi(t - t') = 0 \text{ for } t' > t\text{...............}(5)$$

While the material system is subjected to an intense impulsive field strength at time t_0, then $E(r, t') = E\,\delta(t - t_0)$, where δ is the Dirac delta function. For $t > t_0$ the polarization becomes,

$$P(r,t) = \varepsilon_0 \int_{0}^{\infty} dt'' \chi(t'') E(r,t-t'')\text{..............}(6)$$

Extremely localized electric field in space not only polarizes atoms and molecules within the area where it is applied, but also produces dipole moments in the near vicinity. Dipole moment P(r, t) at point r depends on the electric field at other points near r in addition to the local electric field strength at r. Relationship between electric field and polarization is nonlocal in space. Equation (6) can be updated as follows,

$$P(r,t)=\varepsilon_0\int_{-\infty}^{+\infty}dt'dr'\chi(r-r',t-t')E(r',t')..............(7)$$

For homogeneous medium χ depends only on the difference r - r′ not on r and r′ separately. The intense optical field induces a refractive index gradient when passing through the medium. This might lead to the beam focusing, beam trapping and beam defocusing effect which can be explained precisely if an intensity dependent refractive index is considered. Moreover the induced refractive index gradient inside the material can lead to the modification of the original beam. It is quite possible to obtain new frequency components at the output field which were not present in the input field. For example, output frequency becomes double and triple of the input frequency at second and third harmonic generation respectively. Nonlinear absorption coefficient is found to decrease with the increasing input intensity, which is completely opposite to the behaviour of linear absorption coefficient. None of these phenomena are observed if the input intensity is low [5, 21].
When a material is exposed to such intense optical field (usually obtained from lasers) then electronic polarization of the material system can be expressed as,

$$\begin{aligned}P(r,t)&=\varepsilon_0\int_{-\infty}^{+\infty}\chi^{(1)}(r-r',t-t')E(r',t')dr'dt'+\varepsilon_0\int_{-\infty}^{+\infty}\chi^{(2)}(r-r_1,t-t_1;r-r_2,t-t_2)E(r_1,t_1)E(r_2,t_2)dr_1dr_2dt_1dt_2+\\&\varepsilon_0\int_{-\infty}^{+\infty}\chi^{(3)}(r-r_1,t-t_1;r-r_2,t-t_2,r-r_3,t-t_3)E(r_1,t_1)E(r_2,t_2)E(r_3,t_3)dr_1dr_2dr_3dt_1dt_2dt_3+................\\&=\varepsilon_0\chi^{(1)}E(r,t)+\varepsilon_0\chi^{(2)}E^2(r,t)+\varepsilon_0\chi^{(3)}E^3(r,t)+...........\\&=P^{(1)}(r,t)+P^{(2)}(r,t)+P^{(3)}(r,t)+...............(8)\end{aligned}$$

First term in equation (8) is the usual linear polarization and rest of the terms describe the nonlinear optical polarizations. Here $\chi^{(1)}$ is the linear optical (dielectric) susceptibility. $\chi^{(2)}$ and $\chi^{(3)}$ are the second and third order nonlinear optical susceptibilities respectively [5, 21, 22].

2.2 Third Order Nonlinear Optical Polarization

Third term in equation (8) describes third order nonlinear optical polarization. Both centrosymmetric and non-centrosymmetric materials exhibit third order nonlinear optical effects if necessary experimental conditions are satisfied. Most general form of the third order nonlinear optical polarization is given by,

$$P^{(3)}(r,t)=\varepsilon_0\chi^{(3)}\left(E_0^3(r,t)e^{-i3\omega t}+3|E_0(r,t)|^3e^{-i\omega t}+E_0^{*3}(r,t)e^{i3\omega t}+3|E_0^*(r,t)|^3e^{i\omega t}\right)..............(9)$$

The first term in (9) describes contribution at 3ω which is known as the optical third harmonic generation (THG) and the second term at the fundamental frequency ω. In THG process, output wave is generated at the third harmonic of the incident radiation field. Consequently the wavelength of the output field is reduced by one-third of the input field. According to the photon energy consideration, three photons at frequency ω are destroyed and a photon at frequency 3ω is produced in a single quantum mechanical process [5].

3. CALCULATION

Third order nonlinear optical polarization is described by,

$$P^{(3)}(t)=\varepsilon_0\chi^{(3)}E^3(t)..............................(10)$$

Intense optical beam emitted from laser has complex amplitude (Gaussian beam) give by,

$$U(r) = A_0 \frac{W_0}{W(z)} \exp\left(-\frac{\rho^2}{W^2(z)}\right) \exp\left(-jkz - jk\frac{\rho^2}{2R(z)} + j\zeta(z)\right) \ldots\ldots\ldots\ldots(11)$$

Constant A_0 has been defined for convenience. W_0 is the waist radius and W(z) is the beam waist as function of the axial distance z. Radial distance (ρ) is defined as $\rho^2 = x^2 + y^2$ and k is the wave vector. R(z) is Rayleigh range and ς(z) is the phase retardation of the Gaussian beam. The complex amplitude of the Gaussian beam depends only on two independent parameters A_0 and z_0, which are determined from the boundary conditions. The remaining parameters are dependent on z_0 and wavelength λ [23, 24].

Experimental values of the optical beam powers at three available wavelengths (457 nm, 488 nm and 514 nm) have been converted into intensity using the following equation,

$$P = \frac{1}{2} I (\pi W_0)^2 \ldots\ldots\ldots\ldots(12)$$

From the optical intensities, corresponding electric fields have been calculated to estimate the third order nonlinear optical polarization for alpha quartz, BK-7 glass and fused silica. Numerical values of the third order susceptibility $\chi^{(3)}$ of alpha quartz, BK-7 glass and fused silica has been used from the literature [25].

4. GRAPHS AND DISCUSSION

In this research work, third order nonlinear optical polarization $P^{(3)}$ has been estimated for alpha quartz, BK-7 glass and fused silica. Values of $P^{(3)}$ have been calculated as function of different optical powers for three distinct wavelengths (457 nm, 488 nm and 514 nm) emitted from a continuous wave Argon ion laser.

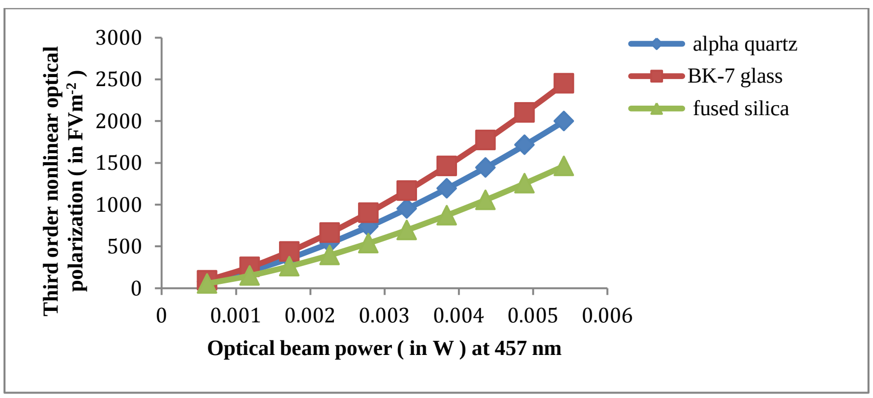

Fig-1: Variation of third order nonlinear optical polarization $P^{(3)}$ as function of the optical beam powers at 457 nm wavelength. The plot shows that alpha quartz, BK-7 glass and fused silica follow similar trends.

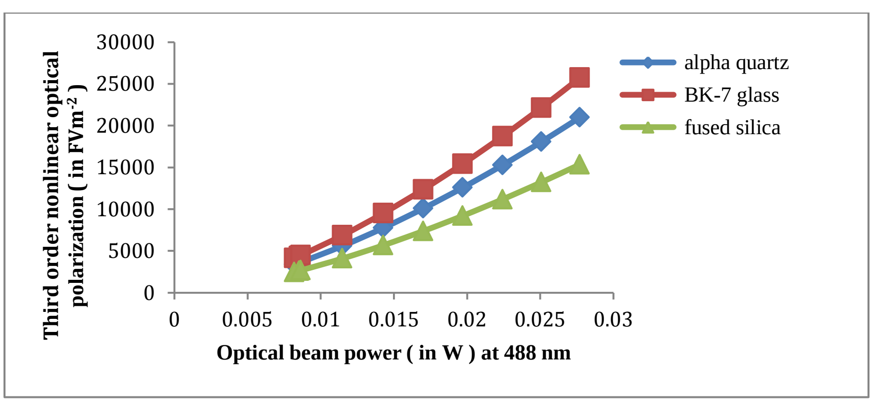

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Fig-2: Variation of third order nonlinear optical polarization $P^{(3)}$ as function of the optical beam powers at 488 nm wavelength of the laser radiation for alpha quartz, BK-7 glass and fused silica.

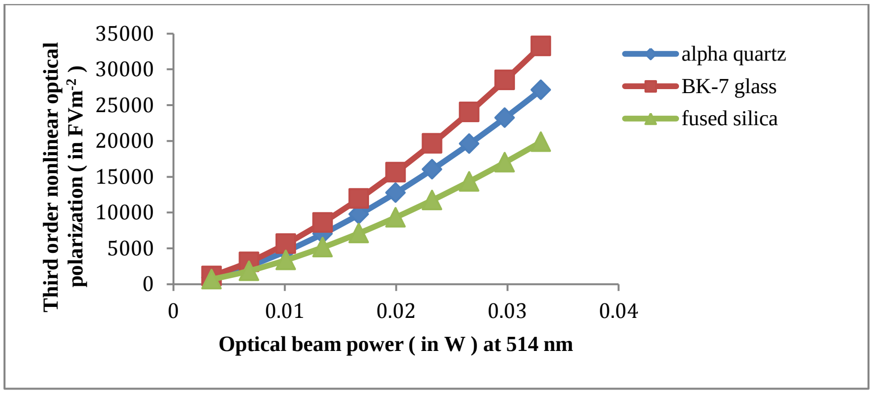

Fig-3: Plot between third order nonlinear optical polarization $P^{(3)}$ for different optical beam powers at 514 nm wavelength of laser. The plot shows similar trend for alpha quartz, BK-7 glass and fused silica.

Variation of third order nonlinear optical polarization $P^{(3)}$ as function of the incident optical beam powers has been shown in Fig-1 to Fig-3. Plots are drawn for alpha quartz, BK-7 glass and fused silica at 457 nm, 488 nm and 514 nm wavelengths. All three plots show that, values of the third order nonlinear optical polarization $P^{(3)}$ increases linearly as the optical beam power is increased. It is obvious that the values of $P^{(3)}$ will vary nonlinearly with the applied field strength. The result is in good agreement with the equation describing third order nonlinear optical polarization.

Estimated values of the third order nonlinear polarization $P^{(3)}$ for alpha quartz, BK-7 glass and fused silica have been presented in table-1. Calculated values of $P^{(3)}$ have been found of the order of 10^3 FVm^{-2} at all three available wavelengths.

alpha quartz $P^{(3)}$ x10^3 (in FVm^{-2})			**BK-7 glass $P^{(3)}$ x10^3 (in FVm^{-2})**			**fused silica $P^{(3)}$ x10^3 (in FVm^{-2})**		
457 nm	488 nm	514 nm	457 nm	488 nm	514 nm	457 nm	488 nm	514 nm
1.19	12.59	16.03	1.46	15.43	19.65	0.87	9.21	11.73
1.45	15.29	19.60	1.77	18.75	24.03	1.06	11.19	14.35
1.71	18.07	23.25	2.10	22.16	28.51	1.25	13.23	17.02
1.99	20.99	27.14	2.45	25.75	33.27	1.46	15.37	19.86

Table-1: Values of $P^{(3)}$ for alpha quartz, BK-7 glass and fused silica

5. CONCLUSION

Third order nonlinear optical polarization $P^{(3)}$ has been estimated theoretically for alpha quartz, BK-7 glass and fused silica. Applied optical field was taken relevant to a continuous wave Argon ion laser to produce the optical nonlinearity. Calculated values of $P^{(3)}$ was found to be of the order of 10^3 FVm^{-2} at all available wavelengths. Similar calculations can be done for other third order nonlinear optical materials.

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