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Second Order Nonlinear Optical Polarization at Different Wavelengths for Zinc-Blende Crystals

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ABSTRACT

Second order nonlinear optical polarization $P^{(2)}$ has been calculated theoretically for several crystals having zinc-blende symmetries. Three distinct wavelengths 457 nm, 488 nm and 514 nm emitted from a continuous wave (cw) Ar-ion laser have been considered for the estimation of the second order nonlinear optical polarization. The study reveals nonlinear dependence of the second order nonlinear optical polarization on the applied electric field intensities at various wavelengths.

Keywords: Second order nonlinear optical polarization, cw Ar-ion laser, zinc-blende crystals.

I. INTRODUCTION

Theoretical estimation of the second order nonlinear optical polarization is vital because of their potential applications in various second order processes like optical phase modulation (OPM) [1, 2], second harmonic generation (SHG) [3-5], sum frequency generation (SFG) [6-8] and difference frequency generation (DFG) processes [9-11]. Second order nonlinear effects are widely being used to convert the output of a fixed frequency laser into a desired spectral region via second harmonic generation, optical sum and difference frequency generations; two photon emission and parametric fluorescence processes [12]. Different noncentrosymmetric crystals are very popular choice to study the second order nonlinear optical processes [12-15].

Geskin *et al* performed quantum-chemical analysis of second order polarization in nonlinear optical chromophores and borato diphenylpolyenes. Their study showed that phenylene groups play the key role in generating a high second order nonlinear response [16]. Wang *et al* applied second harmonic generation to distinguish surface and bulk contribution in isotropic gold films [17]. Aspnes measured zero frequency limit of the second order nonlinear optical susceptibility of crystals of Zinc-Blende symmetry within the framework of energy band theory [18]. They considered both vector and scalar potential representation of the electric field to estimate the second order susceptibility in terms of band theory.

In this research work, second order nonlinear optical polarization has been calculated for GaAs, InSb, GaP, GaSb, InAs, ZnSe, ZnS and ZnTe crystals. Optical power emitted from a continuous wave Ar-ion laser has been converted into corresponding optical (electric) field strengths. Estimated electric field intensities have been used to estimate the second order nonlinear optical polarization at 457 nm, 488 nm and 514 nm wavelengths of the laser radiation using second order nonlinear optical susceptibility from the investigation performed by Aspnese.

II. NONLINEAR OPTICAL POLARIZATION

Dielectric medium exposed under strong optical field, produces induced dipole moment $\boldsymbol{\mu}^{(\sim)}$ in the direction of the applied field as a result of bound charge separation. Microscopic analysis confirms that electrons are accumulated in the opposite direction of the applied field while the nucleus is pushed toward the applied field direction. It is obvious that charge carriers are oscillating since induced by rapidly varying external radiation field. The net average dipole moment per unit volume is termed as polarization and governed by the following relation,

$$P^{(\sim)} = N < \mu^{(\sim)} > .....(1)$$

Superscripted tilde (~) implies that the fields are rapidly varying in time and real quantities. N is the microscopic dipole density and angular brackets has been used to indicate the ensemble average over all of the dipoles in the medium. Existing permanent dipoles within the medium will not radiate since they are not oscillating at frequencies of the external radiation.

For low applied field strength $\mathbf{E}^{(\sim)}$, polarization $\mathbf{P}^{(\sim)}$ varies linearly as function of $\mathbf{E}^{(\sim)}$ and expressed as follows for a homogeneous medium

$$P_L^{(\sim)}(r,t) = \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \chi^{(1)}(r-r',t-t')E^{(\sim)}(r',t')dr'dt'.....(2)$$

Subscript L has been used to indicate linear polarization, ε_0 is the permittivity of free space, $\chi^{(1)}(r-r', t-t')$ is the linear dielectric response tensor. The parenthesis in $\chi^{(1)}$ represents invariance of time and space. The above written equation has been presented in SI unit [12, 13, 19]. Linear polarization P_L can be expressed in terms of atomic polarizability α as shown in the following relationship which is tantamount to equation (2).

$$P_L^{(\sim)}(r,t) = \alpha E^{(\sim)}(r,t).....(3)$$

A straightforward form of equation (2) is can be written as follows which is frequently used for the purpose of numerical estimation.

$$P^{(\sim)}(r,t) = \varepsilon_0 \chi E^{(\sim)}(r,t).....(4)$$

When the incident field intensity is adequate strong to compete with characteristic atomic electric field strength then nonlinear part of the polarization (P_{NL}) plays significant role by acting as a source for the new components of electromagnetic field that are not present in the applied field. Inhomogeneous differential wave equation can describes optical processes in nonlinear regime. Nonlinear polarization P_{NL} can be expressed as a power series expansion in the applied field strength $\mathbf{E}^{(\sim)}$.

$$P^{(\sim)(2)}(r,t) = \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \chi^{(2)}(t-t',t-t'')E^{(\sim)}(r,t')E^{(\sim)}(r,t'')dt'dt''....(5)$$

$$P^{(\sim)(3)}(r,t) = \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \chi^{(3)}(t-t',t-t'',t-t''')E^{(\sim)}(r,t')E^{(\sim)}(r,t'')E^{(\sim)}(r,t''')dt'dt''dt'''....(6)$$

$\mathbf{P}^{(\sim)(2)}$ and $\mathbf{P}^{(\sim)(3)}$ define second and third order nonlinear optical polarization respectively which are characterized by second ($\boldsymbol{\chi}^{(2)}$) and third ($\boldsymbol{\chi}^{(3)}$) order nonlinear optical susceptibilities. Equations (5) and (6) describe local response and thus spatial dependence of $\boldsymbol{\chi}^{(2)}$ and $\boldsymbol{\chi}^{(3)}$ has been ignored. Expansion of polarization $\mathbf{P}^{(\sim)}(\mathbf{r}, t)$ as a power series in the applied electric field strength $\mathbf{E}^{(\sim)}(\mathbf{r}, t)$ can be generalized as follows,

$$P^{(\sim)}(r,t) = \varepsilon_0 \chi^{(1)} E^{(\sim)}(r,t) + \varepsilon_0 \chi^{(2)} E^{2(\sim)}(r,t) + \varepsilon_0 \chi^{(3)} E^{3(\sim)}(r,t) + ...(7)$$

$$P^{(\sim)}(r,t) = P^{(\sim)(1)}(r,t) + P^{(\sim)(2)}(r,t) + P^{(\sim)(3)}(r,t).....(8)$$

Second order nonlinear polarization processes like second harmonic generation (SHG), sum (SFG) and difference (DFG) frequency generation, optical parametric amplification (OPA) are characterized by the second term $\mathbf{P}^{(\sim)(2)}(\mathbf{r},t)$ in equation (8). All the second order processes are forbidden in centrosymmetric materials as they possess inversion symmetry. Materials having nonzero second order nonlinear optical susceptibility, exhibit dependence of the polarization on the applied field strength qaudratically. In consequence, several second order processes can readily occur in noncentrosymmetric medium if certain phase matching condition is achieved. Material having nonzero second order nonlinear optical susceptibility exposed under strong optical field strength $\mathbf{E}^{(\sim)}(\mathbf{r},t)$ and frequency ω can be described by the following equation.

$$P^{(\sim)(2)}(r,t) = \varepsilon_0 \chi^{(2)} E^{2(\sim)}(r,t) e^{-i2\omega t} + 2\varepsilon_0 \chi^{(2)} \mid E^{(\sim)}(r,t) \mid^2 + c.c....(9)$$

Notation c.c. stands for complex conjugate. First term in equation (9) describes second harmonic generation process where input field is converted into second harmonic at the output. Obviously wavelength of the output field becomes half of the input field. This process is efficiently being used to convert the output of a fixed frequency laser to a different spectral region. The second term describes a process called optical Rectification (OR) where a static (DC) electric field is generated inside the nonlinear material which in turn produces a DC potential difference across the medium. Unlike second harmonic generation, optical rectification does not lead to generation of any new frequency component. The process can efficiently convert optical pulse of MW peak power into μV DC voltage depending on the nonlinear medium.

III. CALCULATION

Gaussian beam emerged from laser is sufficiently intense to produce optical nonlinearity in second order materials. Complex amplitude of a finite TEM_{00} mode of a circular Gaussian beam is given by,

$$E^{(\sim)}(r,t) = \hat{e}\mathrm{A}(z,t)\frac{W_0}{W(z)}\exp\left(-\frac{\rho^2}{W^2(z)}\right)\exp\left(-ikz - ik\frac{\rho^2}{2R(z)} + i\zeta(z)\right) + c.c...(10)$$

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 and z_0, which are determined from the boundary conditions. The remaining parameters are dependent on z_0 and wavelength λ [20-22].

Experimental values of the optical beam powers at three available wavelengths (457 nm, 488 nm and 514 nm) from a continuous wave Ar-ion laser have been converted into intensity using the following equation,

$$P = \frac{1}{2} I(\pi W_0)^2 .....(11)$$

For available optical intensities, corresponding electric fields have been calculated to estimate the second order nonlinear optical polarization ($P^{(2)}$) for GaAs, InSb, GaP, GaSb, InAs, ZnSe, ZnS and ZnTe crystals using values of the second order nonlinear optical susceptibility $\chi^{(2)}$ from the literature [18].

IV. GRAPHS AND DISCUSSION

This section represents the estimated second order nonlinear optical polarization $P^{(\sim)(2)}$ for GaAs, InSb, GaP, GaSb, InAs, ZnSe, ZnS and ZnTe crystals which exhibit zinc-blende symmetries. The calculation was performed at three wavelengths which are 457 nm, 488 nm and 514 nm emerged from a continuous wave Ar-ion laser.

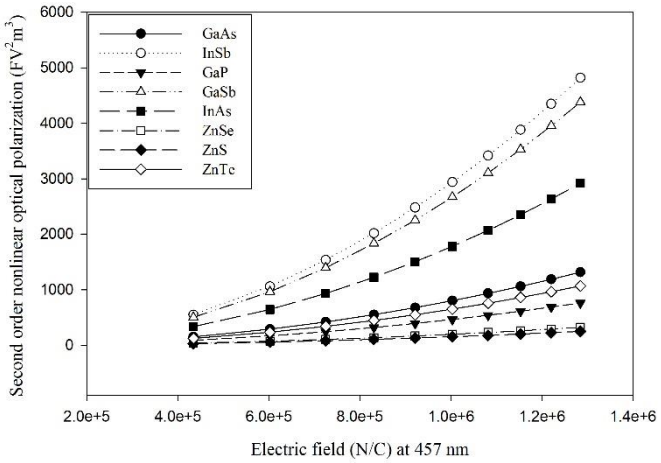

Figure 1: A Plot of the second order nonlinear optical polarization of GaAs, InSb, GaP, GaSb, InAs, ZnSe, ZnS and ZnTe crystals for different electric fields at 457 nm wavelength.

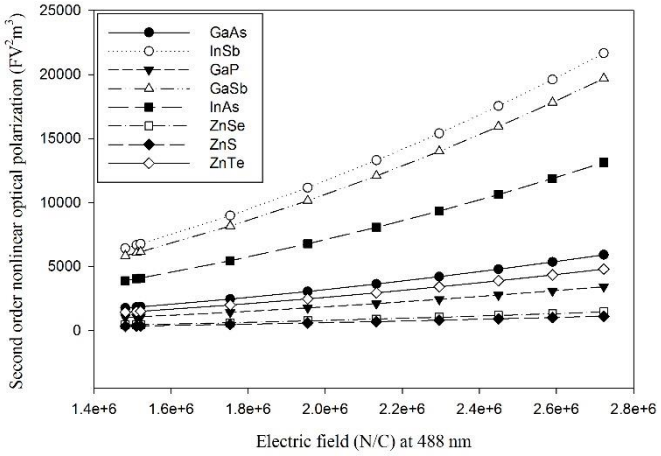

Figure 2: Second order polarization of GaAs, InSb, GaP, GaSb, InAs, ZnSe, ZnS and ZnTe for several electric fields at 488 nm wavelength.

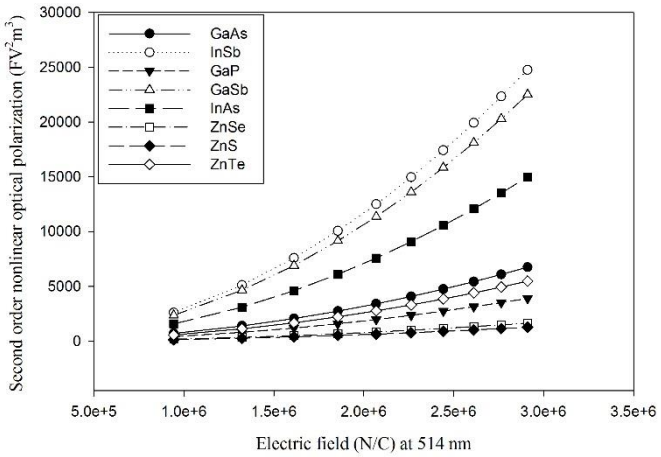

Figure 3: Second order nonlinear optical polarization of GaAs, InSb, GaP, GaSb, InAs, ZnSe, ZnS and ZnTe crystals for different electric fields at 514 nm wavelength.

Fig. 1 to Fig. 3 show the variation of the second order nonlinear optical polarization of GaAs, InSb, GaP, GaSb, InAs, ZnSe, ZnS and ZnTe crystals at available wavelengths. All three graphs show that the second order polarization varies nonlinearly with the increasing electric field intensities. As the square of the electric field strength is proportional to the optical beam power, second order nonlinear optical polarization also increases with increasing beam powers. Since beam power is comparatively low at 457 nm wavelength, the initially second order polarization shows very insignificant nonlinearity with increasing optical field strength or beam power. As the optical field intensity is increased, the second order optical polarization shows noticeable nonlinear variation as function of the applied electric field strength (Fig. 1). For other two wavelengths (488 nm and 514 nm) the second order nonlinear optical polarization $P^{(2)}$ varies significantly as the applied optical field is increased up to several milliwatt (Fig. 2 and Fig. 3).

Graphs show that variation of second order nonlinear optical polarization $P^{(2)}$ is prominent for InSb and GaSb followed by InAs. GaAs, ZnTe and GaP exhibit moderate dependence of second order nonlinear optical polarization on external field intensity. On the contrary, ZnSe and ZnS demonstrate least variation of the second order nonlinear optical polarization for different incident electric field strengths. These observations are found to be similar at all available wavelengths and in good agreement with theoretical concepts.

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