# Calculational Study of SHG and LE-O Susceptibilities of InSb Crystal: Linear Absorption Discarded

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***Abstract*:** *A model, involving two coupled anharmonic oscillators (electronic and ionic) is applied here to estimate the Second Harmonic Generation (SHG) and Linear Electro-Optic (LE-O) susceptibilities of InSb crystal. The crystal of InSb belongs to III-V group compounds owing to a cubic zinc-blende-type structure. Linear absorption is discarded for the selected spectral region 1250 nm – 390 nm. So, the contribution of the imaginary part of the involved complex linear ionic susceptibility* $3^{(1)}(\omega)$ *to the resultant SHG and LE-O susceptibilities is neglected in the computation of SHG and LE-O coefficients. All of the four constants (nonlinear strength factors), appearing in the model, are determined with the help of experimental data of SHG susceptibility measured in the selected region of 1076 nm -535 nm. Applying such calculated nonlinear strength factors in the concerned model expressions, SHG and LE-O susceptibility coefficients are computed as a function of frequency to illustrate and hence to study the dispersion in the region of 1250 nm –390 nm.*

***Keywords*:** **Nonlinear Optics, Second Harmonic Generation, LE-O Susceptibility Coefficient**.

## 1. Introduction

Since the invention of the laser (around 1960), extensive advances have been made to understand as well as to apply nonlinear optical interactions. Both experimental, as well as theoretical research in the field of nonlinear optics is represented by the determination of the absolute value of nonlinear susceptibility. C.G. Garrett [1] used a model with two coupled anharmonic oscillators (electronic & ionic) to predict the nonlinear susceptibilities for a simple diatomic, cubic material. With the limitation of 1-D (1-dimensional), the model should give a reasonable description of the behavior of zinc-blende-type materials that are both diatomic and cubic. InSb is one of the III-V group compounds having a zinc-blende-type structure. Formerly, several models are applied by different workers to compute the second-order optical properties of III-V group compounds in the different regions of radiation. Some such models are the bond-charge model [2]-[4] and the charge-transfer model [5]. S.S. Jha and N. Bloembergen [6]; C.L. Tang [7] and C. Flytzanis *et al.* [8] also have calculated the second-order optical susceptibility coefficients such as Second Harmonic Generation (SHG) and Linear Electro-Optic (LE-O) coefficient for III-V group compounds to which InSb belongs. Classically, none of the authors [5]-[8], had obtained a dispersion relation to estimating the second-order optical susceptibilities, involving a simultaneous contribution from linear electronic and linear ionic susceptibilities for InSb along with other III-V group semiconducting compounds. Presently, the author applied a model to the InSb crystal to compute its nonlinear optical properties (SHG and LE-O susceptibility coefficients) in the selected spectral region of 1250 nm – 390 nm. For this, firstly the four Nonlinear Strength Factors (NSF), appearing in our modelling, are computed with the help of existing available experimental data [9]. And then, as per the objectives of the author's present work, by using such calculated parameters; the author estimated the required LE-O and SHG coefficients as a function of the frequency. This way, the dispersion in the near infra-red (NIR) region of 1250 nm- 390 nm, is illustrated.

## 2. Theoretical Aspect

Following a one-dimensional lattice, Garrett had written the equations of motion in terms of configuration co-ordinates $q_e$ and $q_i$ for electronic and ionic oscillation respectively as

$$\ddot{q}_\mathrm{e} + \omega_\mathrm{e}^2 q_e = \text{—}\ E \tag{1}$$

$$q_\mathrm{i} + \omega_\mathrm{i}^2 q_i = \frac{-}{\mu} E. \tag{2}$$

Here, $m_e$ is the electronic mass, μ is the reduced ionic mass, $e_e$ and $e_i$ are the charges of the order of one electronic charge and are defined in terms of cation, anion core and anion shell charges. $\omega_e$ is the resonant frequency associated with the dominant ultraviolet inter-band electronic transition responsible for the dispersion in the visible region and $\omega_i$ is the resonant frequency associated with transverse optical (TO) phonon frequency in the infrared region. $q_e$ is called electronic configuration co-ordinate associated with $\omega_e$ and $q_i$ is ionic configuration co-ordinate associated with $\omega_i$. The polarization, $P = N\ [e_e q_e + e_i q_i]$ and the linear susceptibility is

$$\chi^{(1)} = \frac{N}{0}[e\ q\ + \quad ] = \frac{}{0} + \frac{}{0} = \chi_e^{(1)} + \chi_i^{(1)}$$

Where $\chi_e^{(1)}$ and $\chi_i^{(1)}$ are electronic and ionic susceptibility respectively. As, $q = \frac{}{(\ )}$, one gets, $\chi_e^{(1)} = \frac{Ne_e^2}{0\ \ (\ )}$, where, $(\omega) = \omega_e^2 - \omega^2$. On taking the ionic damping effect on the harmonic oscillatory motion of the ion, into account, an extra damping term is appeared in the equation of motion Eq.(2) as, $q_7 + \tau q_7 + \omega_i^2 q_i = \frac{e_i}{\mu} E$. It gives,

$$= \frac{}{\mu(-\omega^2 - i\tau\omega + \omega_i^2)} E. \text{ And}$$

$$3_i^{(1)}(\ ) = \frac{2}{\epsilon_0 \mu D_i(\omega)}. \tag{3}$$

Where $De\ (\omega) =_i \omega^2 - i\tau\omega - \omega^2$.

Here, the author added a phenomenological damping rate τ in the ionic response only. Cochran [10] introduced the quadratic terms as nonlinear terms in potential as he was interested in centrosymmetric crystals. The noncentrosymmetric 1-D model necessarily possesses a unique polar axis, which will be pyroelectric. So, Garrett [1] added a cubic instead of the quadratic term to potential. So, the potential is

$$U = \frac{m_e q_e^2 \omega_e^2}{2} + \frac{\mu q_i^2 \omega_i^2}{2} + Aq^3 + Bq^2 q_e + \mathrm{C}q\ q^2 + Dq^3 - E(e\ q + e\ q). \tag{4}$$

Where, A, B, C, and D are constants referred as nonlinear strength factors (NSF). So,

$$q = -\omega^2 q + \left(\frac{-}{}\right) E - \left(\frac{3}{}\right) q2 - \left(\frac{2}{}\right) q\ q - \left(\frac{-}{}\right) q2. \tag{5}$$

$$\ddot{q}_7 = -\omega_i^2 q_i - \tau\dot{q}_7 + \left(\frac{}{\mu}\right) E - \left(\frac{}{\mu}\right) q_e^2 - \left(\frac{2}{\mu}\right) q_e q_i - \left(\frac{3}{\mu}\right) q_i^2. \tag{6}$$

An applied electric field E̲ is assumed to be a superposition of two fields as,

$$\underline{E} = \frac{1}{2}\left[\underline{E}_1 e^{-j\omega_1 t} + C.C. + \underline{E}_2 e^{-j\omega_2 t} + C.C.\right] \tag{7}$$

Here, $q_{\underline{e}}$ and $q_{\underline{i}}$ will respond to the applied electric field having components at $\omega_1$ and $\omega_2$ due to linear and at $\omega_1 \pm \omega_2$, $2\omega_1$,$2\omega_2$ due to nonlinear behaviour. Thus,

$$= {}^{(1)} + {}^{(2)}(0)|_{1,1} + {}^{(2)}(0)|_{2,2} + \frac{1}{2}[\,{}^{(2)}(\ _1 - \omega_2)e^{-j(\omega_1-\omega_2)t} + q^{(2)}(2\omega_1)e^{-j(2\omega_1)t} + q^{(2)}(\omega_1 + \omega_2)e^{-j(\omega_1+\omega_2)t} + q^{(2)}(2\omega_2)e^{-j(2\omega_2)t} + C.C.)]$$

(8) $q^{(1)} = \frac{1}{2}[q^{(1)}(\omega_1)e^{-j\omega_1 t} + C.C. + q^{(1)}(\omega_2)e^{-j\omega_2 t} + C.C.]$

(9) $q^{(1)} = \frac{1}{2}[q^{(1)}(\omega_1)e^{-j\omega_1 t} + C.C. + q^{(1)}(\omega_2)e^{-j\omega_2 t} + .\ .]$ (10)

Using the expressions of , , and _ into (5) and (6), $^{(2)}$ and $^{(2)}$ can be solved in terms of $\underline{1}$, $\underline{E}_2$, $\underline{q}^{(1)}$ and $\underline{q}^{(1)}$. Second-order polarization at ωij, $\underline{P}(\omega_j, \omega, \omega_j) = \frac{1}{2}[\underline{P^{(2)}}(\omega_j, \omega, \omega_j)e^{-j\omega ijt} + .\ .]$. Or,

$$\underline{P}(\omega_j, \omega i, \omega_j) = \frac{1}{2}[3^2(\omega_j, \omega, \omega_j)\underline{E}\,\underline{E_j}\, e^{-j\omega_{ij} t} + C.C]. \tag{11}$$

A detailed tedious calculation results in the general expression for the first-order nonlinear susceptibility,

$$3^{(2)}(\omega_i \pm \omega_j, \omega_i, \omega_j)|i = 1, j = 2| = -\frac{\overset{2}{0}}{2}\{(\frac{3}{2\ 3})[3^{(1)}(\ _1).3^{(1)}(\ _2).3^{(1)}(\omega_1 \pm \omega_2)] +$$
$$(\frac{C}{2})[3^{(1)}(\omega_1).3^{(1)}(\omega_2).3^{(1)}(\omega_1 \pm \omega_2) + 3^{(1)}(\omega_1).3^{(1)}(\omega_2).3^{(1)}(\omega_1 \pm \omega_2)] +$$
$$(\frac{C}{2\ 2})[3^{(1)}(\omega_1).3^{(1)}(\omega_2).3^{(1)}(\omega_1 \pm \omega_2)] + (\frac{B}{2})[3^{(1)}(\omega_1).3^{(1)}(\omega_2).3^{(1)}(\omega_1 \pm \omega_2) +$$
$$3^{(1)}(\omega_1).3^{(1)}(\omega_2).3^{(1)}(\omega_1 \pm \omega_2)] + (\frac{}{2\ 2})[3^{(1)}(\omega_1).3^{(1)}(\omega_2).3^{(1)}(\omega_1 \pm \omega_2)] +$$
$$(\frac{3A}{2\ 3})[3^{(1)}(\omega_1).3^{(1)}(\omega_2).3^{(1)}(\omega_1 \pm \omega_2)]\} \tag{12}$$

## 3. Present Modelling

The author made a realistic approach and modified Garrett's anharmonic model [1] and so the contribution of the imaginary part of the complex linear ionic susceptibility $3^{(1)}(\ )$ , so the real part Re$3^{(1)}(\omega)$ of $\chi_i^{(1)}(\omega)$ is used in place of $\chi_i^{(1)}(\omega)$ in the computation of SHG and LE-O coefficients for the InSb crystal for the selected spectral range.

### 3.1. SHG Susceptibility Coefficient

For sum-frequency mode $_1 + \ _2 = \ + \ = 2$ , Eq. (12) gives the expression for SHG as,

$$3^{(2)}(2\omega, \omega, \omega) = -\frac{\overset{2}{0}}{2}\{(\frac{3}{2\ 3})[3^{(1)}(\ ).3^{(1)}(\ ).3^{(1)}(2\ )] +$$
$$(\frac{2}{2})[3^{(1)}(\ ).\mathrm{Re}3^{(1)}(\ ).3^{(1)}(2\ )] + (\frac{}{2\ 2})[3^{(1)}(\ ).3^{(1)}(\ ).\mathrm{Re}3^{(1)}(2\ )] +$$

$$\left(\frac{2}{\quad \angle}\right)[3^{(1)}(\ ).\mathrm{Re}3^{(1)}(\ ).\mathrm{Re}3^{(1)}(2\ \ )] + \left(\frac{\qquad}{\angle\ \angle}\right)[\mathrm{Re}3^{(1)}(\ ).\mathrm{Re}3^{(1)}(\ ).3^{(1)}(2\ \ )] +$$

$$\left(\frac{3}{2\ 3}\right)[\mathrm{Re}3^{(1)}(\ ).\mathrm{Re}3^{(1)}(\ ).\mathrm{Re}3^{(1)}(2\ \ )]\} \qquad (13)$$

### 3.2. LE-O Susceptibility Coefficient

For $_1 = 0,\ \ _2 =\ $, Eq.(12) gives the LE-O coefficient,

$$3^{(2)}(0+\omega,0,\omega) = -\frac{\overset{2}{0}}{2}\{\left(\frac{3}{2\ 3}\right)[3^{(1)}(0).3^{(1)}(\ ).3^{(1)}(\ )] +$$

$$\left(\frac{\qquad}{2}\right)[3^{(1)}(0).\mathrm{Re}3^{(1)}(\ ).3^{(1)}(\ ) + 3^{(1)}(\ ).\mathrm{Re}3^{(1)}(0).3^{(1)}(\ )] +$$

$$\left(\frac{\qquad}{2\ 2}\right)[3^{(1)}(0).3^{(1)}(\ ).\mathrm{Re}3^{(1)}(\ )\ ] + \left(\frac{\qquad}{\angle}\right)[3^{(1)}(0).\mathrm{Re}3^{(1)}(\ ).\mathrm{Re}3^{(1)}(\ ) +$$

$$3^{(1)}(\ ).\mathrm{Re}3^{(1)}(0).\mathrm{Re}3^{(1)}(\ )] + \left(\frac{\qquad}{\angle\ \angle}\right)[\mathrm{Re}3^{(1)}(0).\mathrm{Re}3^{(1)}(\ ).3^{(1)}(\ )] +$$

$$\left(\frac{3}{2\ 3}\right)[\mathrm{Re}3^{(1)}(0).\mathrm{Re}3^{(1)}(\ ).\mathrm{Re}3^{(1)}(\ )]\}\ . \qquad (14)$$

Where, Linear electronic susceptibility,

$$3^{(1)}(\ ) = \frac{e\ \overset{2}{e}}{m_e\epsilon_0(\omega^2-\omega^2)}, \twoheadrightarrow \textit{Real.}$$

Linear ionic susceptibility, $3_i^{(1)}(\ ) = \frac{\overset{2}{\ }\ \ {}_{\iota}^{2}}{\mu\epsilon_0(\omega_i - \omega^2 - i\tau\omega)}, \twoheadrightarrow \textit{Complex.}$

And $\mathrm{Re}3^{(1)}(\ )$ = Real part of $3^{(1)}(\ )$.

## 4. Applications and Numerical Computations

The input parameters are listed in Table 1a. and the SHG experimental data are given in Table 1b.

### 4.1. Nonlinear Strength Factors (NSF)

Using the input parameters (Table 1a.) and the experimental data [9] (Table 1b.), in Eq.(13), A, B, C, and D are calculated for further applications (Table 2.).

**Table 1a. Input Parameters [11] for Calculation of Nonlinear Strength Factors A, B, C and D for InSb.**

| Parameter | Sym. | Value | Unit |
|---|---|---|---|
| Electronic Oscillator Density | Ne | 2.230000E*+28 | $m^{-3}$ |
| Electronic Charge ** | ee | -1.600000E-19 | C |
| Electronic Mass** | me | 9.109999E-31 | Kg |
| Electronic Resonant Frequency | ωe | 8.510000E+15 | rad/s |
| Ionic Oscillator Density | Ni | 2.230000E*+28 | $m^{-3}$ |
| Reduced Mass of Electronic & Ionic Oscillator *** | μ | 6.000000E-26 | Kg |

| | | | |
|---|---|---|---|
| TO Phonon Frequency | $\omega_i$ | 5.070000E+13 | rad/s |
| Ionic Charge | $e_i$ | 1.600000E-19 | C |
| Damping Rate | $\tau$ | 5.070000E+11 | rad/s |
| The permittivity of Free Space ** | $\varepsilon_0$ | 8.854000E-12 | $C^2/N.m^2$ |
| **E* $\underline{+}$ n = x $10^{\pm n}$, ** Standard data, * ** Calculated data.** | | | |

**Table 1b. Input Data (Experimental) [9] for InSb.**

| **Photon Energy,[eV]** | **Frequency,[xE+15 rad/s]** | **Normalized SHG Susc. Coefficient** |
|---|---|---|
| 1.152 | 1.7515 | (5.83±0.79)E+2 |
| 1.604 | 2.4378 | (9.95±0.34)E+2 |
| 1.789 | 2.7194 | (4.91±0.40)E+2 |
| 2.325 | 3.5343 | (8.14±1.09)E+2 |

**Table 2. Calculated NSF A, B, C, and D for InSb.**

| **N S F** | **Value, [$kg/ms^2$]** |
|---|---|
| A | 1.37916152E+25 |
| B | 3.47905682E+21 |
| C | 3.26093047E+17 |
| D | 1.10257586E+13 |

### 4.2. SHG and LE-O Coefficients

So calculated factors A, B, C, and D (Table 2), are applied in Eq. (13) and Eq. (14) to compute SHG and LE-O susceptibility coefficients respectively, at several different frequencies in the selected spectral region of 1.000 eV – 3.200 eV. The normalization of SHG results is done with $\chi_{KDP}^{36}$ = 0.39 pm/v (at 1064 nm) [12]. Here, the author did the computations in double precision to record the changes in the results of the dependent function.

## 5. Results and Discussion

### 5.1. SHG Results

Following the present model, the computed results of normalized SHG (absolute values) are plotted as a function of photon energy, in Fig.1. InSb shows large absolute values of SHG susceptibility for the range 1.532 eV– 1.678 eV that belongs to the NIR region. The first two dips result from the sum of +ive and –ive values of the cross terms involved in the concerned expression.

Absolute SHG susceptibility goes to infinite at $\omega_e/2$ = 4.2550E+15 rad/s ( ≈ 2.799588 eV) which is caused by the doubling of the applied field (fundamental) frequency (SHG process) equal to $\omega_e/2$, at which the electronic oscillators get in their resonance-mode and causing the maximum (infinite) absolute value of the SHG susceptibility. Near $\omega_e/2$, InSb show large SHG susceptibility but it falls exponentially in the region near $\omega_e/2$. For 2.895 eV – 3.200 eV, InSb crystal shows a very small variation in the dispersion and hence indicates an almost constant SHG response regarding this special band of frequencies.

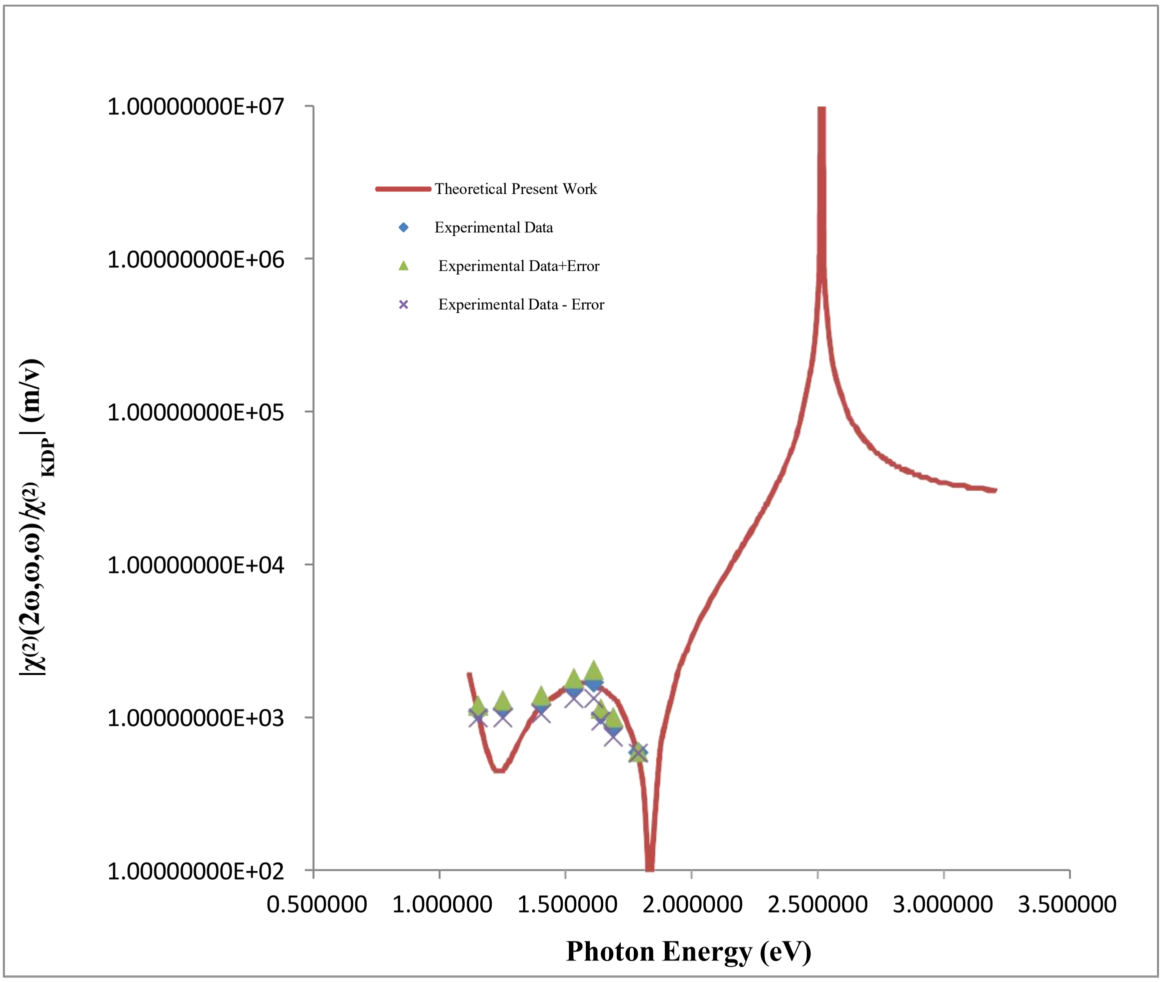

**Figure1: Normalized SHG Susceptibility Coefficient (Absolute value) [$|\chi^{(2)}(\omega+\omega,\omega,\omega)/\ \chi^{(2)}KDP^{36}|$] shows its variation with Photon Energy of radiation hitting the crystal of InSb. Normalization is done with $\chi^{(2)}KDP^{36}$ =0.39 pm/V(at1064 nm).**

### 5.2. LE-O Results

Computed results of LE-O susceptibility coefficients are illustrated in Fig.2. InSb shows large values of absolute LE-O susceptibility coefficient in the region 1.119 eV – 1.382 eV of NIR region. Around 1.873 eV, LE-O susceptibility has lower absolute values (including the lowest at 1.873 eV). InSb shows large absolute values of LE-O coefficient in the NIR region (1.150 eV – 1.480 eV). Thus it is found that for this region, InSb can be potentially useful to fabricate the SHG based NLO devices than other NLO materials.

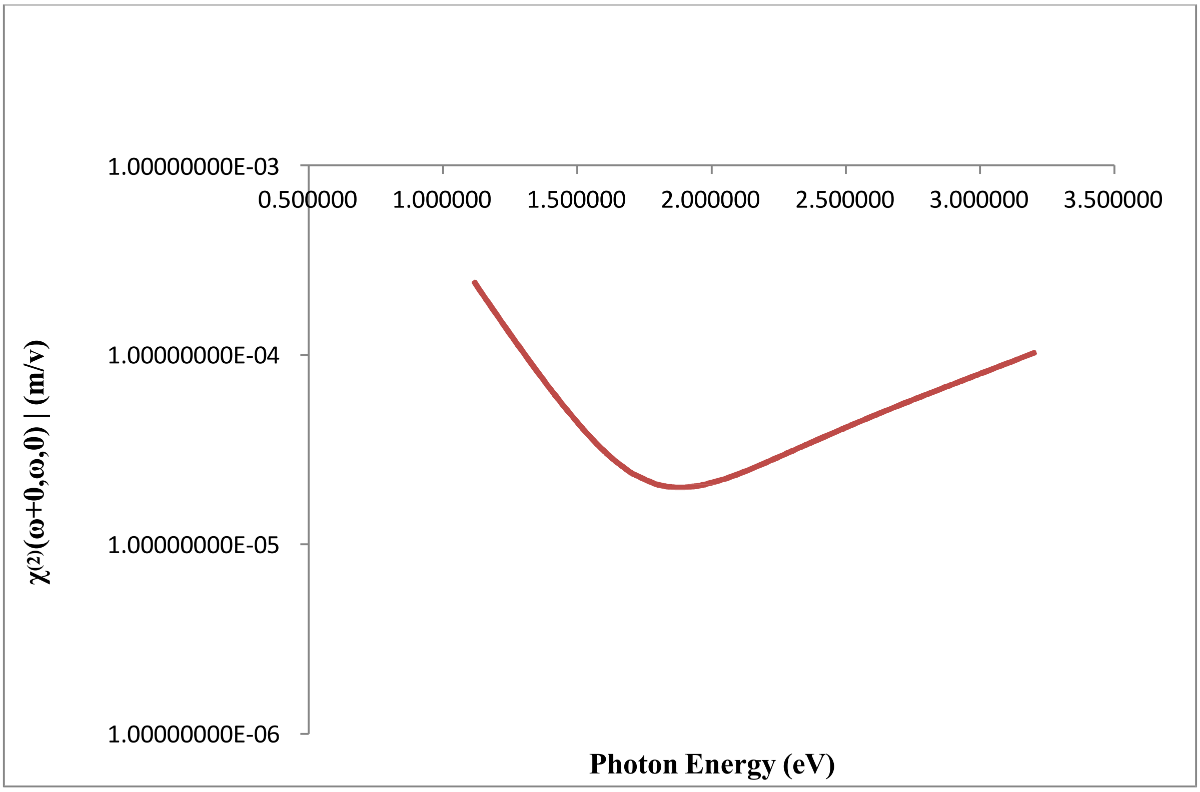

**Figure 2: Linear Electro-Optic (LE-O) Susceptibility Coefficient (Absolute value) [$|\chi^{(2)}(0+\omega,0,\omega)|$(m/V)] shows its Variation with Photon Energy of Radiation hitting the Crystal of InSb.**

## 6. Conclusion

As the results, obtained in the present work, are in good agreement with the experimental [9] ones, the modelling applied here, can be justified for the considered region of radiation. And hence it can be concluded that the theoretical consideration to discard the contribution of the imaginary part of the complex linear ionic susceptibility in the calculated results of SHG coefficients is quite right. This implies that in the selected spectral region (1250 nm – 390 nm) InSb shows no linear absorption of radiation as found through the present modelling which is already applied for the other III-V group compounds like InAs [13] successfully and so is highly acceptable in the spectral region selected here. Also, the LE-O coefficient, estimated in the present work, shows a large absolute value in the NIR region, which indicates the potential application of InSb crystal in the fabrication of nonlinear electro-optic devices such as electro-optic modulators and electro-op rectifiers for the concerned spectral regions.

## Appendix-A

Following Sugie and Tada's three-dimensional model, we have the anharmonic potential, for i$^{th}$ location,

$$U = \sum_{klm}[A_{klm}q_{ik}qilq_{im}+B_{klm}q_{ik}q_{il}q_{em}+C_{klm}q_{ik}q_{el}q_{em}+D_{klm}q_{ek}q_{el}q_{em}] \quad \text{(A-1)}$$

For cubic crystals like CdTe, we have the tensorial form given by Bhagvantum,

$$\begin{array}{cccccc} 0 & 0 & 0 & _{123} & 0 & 0 \\ 0 & 0 & 0 & 0 & _{123} & 0 \\ 0 & 0 & 0 & 0 & 0 & _{123} \end{array}$$

So, U gets the form,

$$U = A_{123}q_{i1}q_{i2}q_{i3}+ A_{123}q_{i2}q_{i3}q_{i1}+ A_{123}q_{i3}q_{i1}q_{i2} +B_{123}q_{i1}q_{i2}q_{e3}+B_{123}q_{i2}q_{i3}q_{e1} + B_{123}q_{i3}q_{i1}q_{e2}+ C_{123}q_{i1}q_{e2}q_{e3} + C_{123}q_{i2}q_{e3}q_{e1} + C_{123}q_{i3}q_{e1}q_{e2} + D_{123}q_{e1}q_{e2}q_{e3} + D_{123}q_{e2}q_{e3}q_{e1} + D_{123}q_{e3}q_{e1}q_{e2}. \quad \text{(A-2)}$$

For a cubic crystal, $q_{i1} = q_{i2} = q_{i3}$ and $q_{e1} = q_{e2} = q_{e3}$ .

Taking,

$3A_{123} = A$ (A-3.1)

$3B_{123} = B$ (A-3.2)

$3C_{123} = C$ (A-3.3)

and, $3D_{123}=D$ (A-3.4)

Now, U may take the form,

$$U = Aq_i^3+Bq_i^2qe+Cq_iq_e^2+Dq_e^3, \quad \text{(A- 4)}$$

which is the same as given by Garrett. Therefore for CdTe, instead of the 3-dimensional Sugie and Tada model, we can take the 1-dimensional Garrett model for calculating the nonlinear susceptibilities.

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