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Theoretical Study of SHG and LE-O Susceptibilities of InAs Crystal: Linear Absorption Discarded

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Abstract

A model involving two coupled anharmonic oscillators (electronic and ionic) is applied to estimate the Second Harmonic Generation (SHG) and Linear Electro-Optic (LE-O) susceptibilities of InAs crystal. The crystal of InAs 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 to the resultant SHG and LE-O susceptibilities is not taken into account and hence the value of the real part of the complex-linear ionic susceptibility i.e. $\mathrm{Re}\chi_i^{(1)}(\omega)$ is used in place of $\chi_i^{(1)}(\omega)$ 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 the dispersion in the region of 1250 nm –390 nm.

Keywords

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

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1. Introduction

Extensive advances have been made in the understanding and application of nonlinear optical interactions since the invention of the laser (around 1960). 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 behaviour of zinc-blende-type materials that are both diatomic and cubic. InAs 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 InAs 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 InAs along with other III-V group semiconducting compounds. Presently, the author applied a model to the InAs 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

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

$$\ddot{q}_e + \omega_e^2 q_e = \frac{e_e}{m_e} E \quad (1)$$

$$\ddot{q}_i + \omega_i^2 q_i = \frac{e_i}{\mu} E. \quad (2)$$

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. ω_e is the resonant frequency associated with the dominant ultraviolet inter-band electronic transition responsible for the dispersion in the visible region and ω_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 ω_e and q_i is ionic configuration co-ordinate associated with ω_i. The polarization, $P = N[e_e q_e + e_i q_i]$ and the linear susceptibility is

$$\chi^{(1)} = \frac{N}{E\epsilon_0}[e_e q_e + e_i q_i] = \frac{N}{E\epsilon_0} e_e q_e + \frac{N}{E\epsilon_0} e_i q_i = \chi_e^{(1)} + \chi_i^{(1)}$$

Where $\chi_e^{(1)}$ and $\chi_i^{(1)}$ are electronic and ionic susceptibility respectively.

As, $q_e = \frac{e_e E}{m_e D_e(\omega)}$, one gets, $\chi_e^{(1)} = \frac{N e_e^2}{\epsilon_0 m_e D_e(\omega)}$, where, $D_e(\omega) = \omega_e^2 - \omega^2$.

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, $\ddot{q}_i + \tau\dot{q}_i + \omega_i^2 q_i = \frac{e_i}{\mu} E$. It gives,

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

$$\chi_i^{(1)}(\omega) = \frac{N e_i^2}{\epsilon_0 \mu D_i(\omega)}. \quad (3)$$

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

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

$$U = \frac{m_e q_e^2 \omega_e^2}{2} + \frac{\mu q_i^2 \omega_i^2}{2} + Aq_i^3 + Bq_i^2 q_e + Cq_i q_e^2 + Dq_e^3 - E(e_e q_e + e_i q_i). \quad (4)$$

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

$$\ddot{q_e} = -\omega_e^2 q_e + \left(\frac{e_e}{m_e}\right) E - \left(\frac{3D}{m_e}\right) q_e^2 - \left(\frac{2C}{m_e}\right) q_e q_i - \left(\frac{B}{m_e}\right) q_i^2. \quad (5)$$

$$\ddot{q}_i = -\omega_i^2 q_i - \tau\dot{q}_i + \left(\frac{e_i}{\mu}\right) E - \left(\frac{C}{\mu}\right) q_e^2 - \left(\frac{2B}{\mu}\right) q_e q_i - \left(\frac{3A}{\mu}\right) q_i^2. \quad (6)$$

An applied electric field $\underline{E}$ is assumed to be a superposition of two fields as,

$$\underline{E} = \frac{1}{2}\left[\underline{E_1} e^{-j\omega_1 t} + C.C. + \underline{E_2} e^{-j\omega_2 t} + C.C.\right] \quad (7)$$

Here, $\underline{q_e}$ and $\underline{q_i}$ will respond to the applied electric field having components at ω_1 and ω_2 due to linear and at $\omega_1 \pm \omega_2$, $2\omega_1$,$2\omega_2$ due to nonlinear behaviour. Thus,

$$q_e = q_e^{(1)} + q_e^{(2)}(0)|_{\omega_1,\omega_1} + q_e^{(2)}(0)|_{\omega_2,\omega_2} + \frac{1}{2}\Big[q_e^{(2)}(\omega_1 - \omega_2) e^{-j(\omega_1-\omega_2)t} + q_e^{(2)}(2\omega_1) e^{-j(2\omega_1)t} + q_e^{(2)}(\omega_1 + \omega_2) e^{-j(\omega_1+\omega_2)t} + q_e^{(2)}(2\omega_2) e^{-j(2\omega_2)t} + C.C.)\Big] \quad (8)$$

$$q_e^{(1)} = \frac{1}{2}\left[q_e^{(1)}(\omega_1)e^{-j\omega_1 t} + C.C. + q_e^{(1)}(\omega_2)e^{-j\omega_2 t} + C.C.\right] \quad (9)$$

$$q_i^{(1)} = \frac{1}{2}\left[q_i^{(1)}(\omega_1)e^{-j\omega_1 t} + C.C. + q_i^{(1)}(\omega_2)e^{-j\omega_2 t} + C.C.\right] \quad (10)$$

Using the expressions of $\underline{q_e}, \underline{q_i}$, and $\underline{E}$ into (5) and (6), $\underline{q_e^{(2)}}$ and $\underline{q_i^{(2)}}$ can be solved in terms of $\underline{E_1}$, $\underline{E_2}$, $\underline{q_e^{(1)}}$ and $\underline{q_i^{(1)}}$. Second-order polarization at ω_{ij}, $\underline{P}(\omega_{ij}, \omega_i, \omega_j) = \frac{1}{2}\left[\underline{P^{(2)}}(\omega_{ij}, \omega_i, \omega_j)e^{-j\omega_{ij}t} + C.C.\right]$. Or,

$$\underline{P}(\omega_{ij}, \omega i, \omega_j) = \frac{1}{2}\left[\chi^2(\omega_{ij}, \omega_i, \omega_j)\underline{E_i}\,\underline{E_j}\,e^{-j\omega_{ij}t} + C.C\right]. \quad (11)$$

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

$$\chi^{(2)}(\omega_i \pm \omega_j, \omega_i, \omega_j)|i=1, j=2| = -\frac{\epsilon_0^2}{2}\left\{\left(\frac{3D}{N_e^2 e_e^3}\right)\left[\chi_e^{(1)}(\omega_1).\chi_e^{(1)}(\omega_2).\chi_e^{(1)}(\omega_1 \pm \omega_2)\right] + \left(\frac{C}{N_e N_i e_e^2 e_i}\right)\left[\chi_e^{(1)}(\omega_1).\chi_i^{(1)}(\omega_2).\chi_e^{(1)}(\omega_1 \pm \omega_2) + \chi_i^{(1)}(\omega_1).\chi_e^{(1)}(\omega_2).\chi_e^{(1)}(\omega_1 \pm \omega_2)\right] + \left(\frac{C}{N_e^2 e_e^2 e_i}\right)\left[\chi_e^{(1)}(\omega_1).\chi_e^{(1)}(\omega_2).\chi_i^{(1)}(\omega_1 \pm \omega_2)\right] + \left(\frac{B}{N_e N_i e_i^2 e_e}\right)\left[\chi_e^{(1)}(\omega_1).\chi_i^{(1)}(\omega_2).\chi_i^{(1)}(\omega_1 \pm \omega_2) + \chi_i^{(1)}(\omega_1).\chi_e^{(1)}(\omega_2).\chi_i^{(1)}(\omega_1 \pm \omega_2)\right] + \left(\frac{B}{N_i^2 e_i^2 e_e}\right)\left[\chi_i^{(1)}(\omega_1).\chi_i^{(1)}(\omega_2).\chi_e^{(1)}(\omega_1 \pm \omega_2)\right] + \left(\frac{3A}{N_i^2 e_i^3}\right)\left[\chi_i^{(1)}(\omega_1).\chi_i^{(1)}(\omega_2).\chi_i^{(1)}(\omega_1 \pm \omega_2)\right]\right\} \quad (12)$$

3. Present Modelling

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

3.1. SHG Susceptibility Coefficient

For SHG, under sum-frequency mode $\omega_1 + \omega_2 = \omega + \omega = 2\omega$, Eq. (12) gives-

$$\chi^{(2)}(2\omega, \omega, \omega) = -\frac{\epsilon_0^2}{2}\left\{\left(\frac{3D}{N_e^2 e_e^3}\right)\left[\chi_e^{(1)}(\omega).\chi_e^{(1)}(\omega).\chi_e^{(1)}(2\omega)\right] + \left(\frac{2C}{N_e N_i e_e^2 e_i}\right)\left[\chi_e^{(1)}(\omega).Re\chi_i^{(1)}(\omega).\chi_e^{(1)}(2\omega)\right] + \left(\frac{C}{N_e^2 e_e^2 e_i}\right)\left[\chi_e^{(1)}(\omega).\chi_e^{(1)}(\omega).Re\chi_i^{(1)}(2\omega)\right] + \left(\frac{2B}{N_e N_i e_i^2 e_e}\right)\left[\chi_e^{(1)}(\omega).Re\chi_i^{(1)}(\omega).Re\chi_i^{(1)}(2\omega)\right] + \left(\frac{B}{N_i^2 e_i^2 e_e}\right)\left[Re\chi_i^{(1)}(\omega).Re\chi_i^{(1)}(\omega).\chi_e^{(1)}(2\omega)\right] + \left(\frac{3A}{N_i^2 e_i^3}\right)\left[Re\chi_i^{(1)}(\omega).Re\chi_i^{(1)}(\omega).Re\chi_i^{(1)}(2\omega)\right]\right\} \quad (13)$$

3.2. LE-O Susceptibility Coefficient: For $\omega_1 = 0$, $\omega_2 = \omega$, Eq.(12) gives the LE-O coeffict.,

$$\chi^{(2)}(0+\omega, 0, \omega) = -\frac{\epsilon_0^2}{2}\left\{\left(\frac{3D}{N_e^2 e_e^3}\right)\left[\chi_e^{(1)}(0).\chi_e^{(1)}(\omega).\chi_e^{(1)}(\omega)\right] + \left(\frac{C}{N_e N_i e_e^2 e_i}\right)\left[\chi_e^{(1)}(0).Re\chi_i^{(1)}(\omega).\chi_e^{(1)}(\omega) + \chi_e^{(1)}(\omega).Re\chi_i^{(1)}(0).\chi_e^{(1)}(\omega)\right] + \left(\frac{C}{N_e^2 e_e^2 e_i}\right)\left[\chi_e^{(1)}(0).\chi_e^{(1)}(\omega).Re\chi_i^{(1)}(\omega)\right] + \left(\frac{B}{N_e N_i e_i^2 e_e}\right)\left[\chi_e^{(1)}(0).Re\chi_i^{(1)}(\omega).Re\chi_i^{(1)}(\omega) + \chi_e^{(1)}(\omega).Re\chi_i^{(1)}(0).Re\chi_i^{(1)}(\omega)\right] + \left(\frac{B}{N_i^2 e_i^2 e_e}\right)\left[Re\chi_i^{(1)}(0)Re\chi_i^{(1)}(\omega)\chi_e^{(1)}(\omega)\right] + \left(\frac{3A}{N_i^2 e_i^3}\right)\left[Re\chi_i^{(1)}(0)Re\chi_i^{(1)}(\omega)Re\chi_i^{(1)}(\omega)\right]\right\} \quad (14)$$

Where Linear electronic susceptibility, $\chi_e^{(1)}(\omega) = \frac{N_e e_e^2}{m_e \epsilon_0 (\omega_e^2 - \omega^2)}$, $\leftarrow\leftarrow Real$

Linear ionic susceptibility, $\chi_i^{(1)}(\omega) = \frac{N_i e_i^2}{\mu \epsilon_0 (\omega_i^2 - \omega^2 - i\tau\omega)}$, $\leftarrow\leftarrow Complex.$

Where $Re\chi_i^{(1)}(\omega)$ = Real part of $\chi_i^{(1)}(\omega)$.

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 InAs.

Parameter	Sym.	Value	Unit
Electronic Oscillator Density	N_e	1.798800E*+28	m^{-3}
Electronic Charge **	e_e	-1.600000E-19	C
Electronic Mass**	m_e	9.109999E-31	Kg
Electronic Resonant Frequency	ω_e	7.644800E+15	rad/s
Ionic Oscillator Density	N_i	1.798800E*+28	m^{-3}
Reduced Mass of Electronic & Ionic Oscillator ***	μ	7.526100E-26	Kg
TO Phonon Frequency	ω_i	3.950000E+13	rad/s
Ionic Charge	e_i	1.600000E-19	C
Damping Rate	τ	3.950000E+11	rad/s
The permittivity of Free Space **	ε_0	8.854000E-12	$C^2/N.m^2$
E* $\pm$n = x $10^{\pm n}$, ** Standard data, * ** Calculated data.			

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

Photon Energy,[eV]	Frequency,[xE+15 rad/s]	Normalized SHG Susc. Coefficient
1.293	1.9653	(8.20±1.27)E+2
1.552	2.3584	(7.23±0.99)E+2
1.707	2.5943	(3.10±0.40)E+2
2.353	3.5769	(8.09+0.87)E+2

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

N S F	Value, [kg/ms²]
A	3.37956805E+25
B	4.84953698E+21
C	2.84497189E+17
D	5.49731433E+12

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. InAs shows large absolute values of SHG susceptibility for the range 1.283 eV– 1.600 eV that belongs to the NIR region.

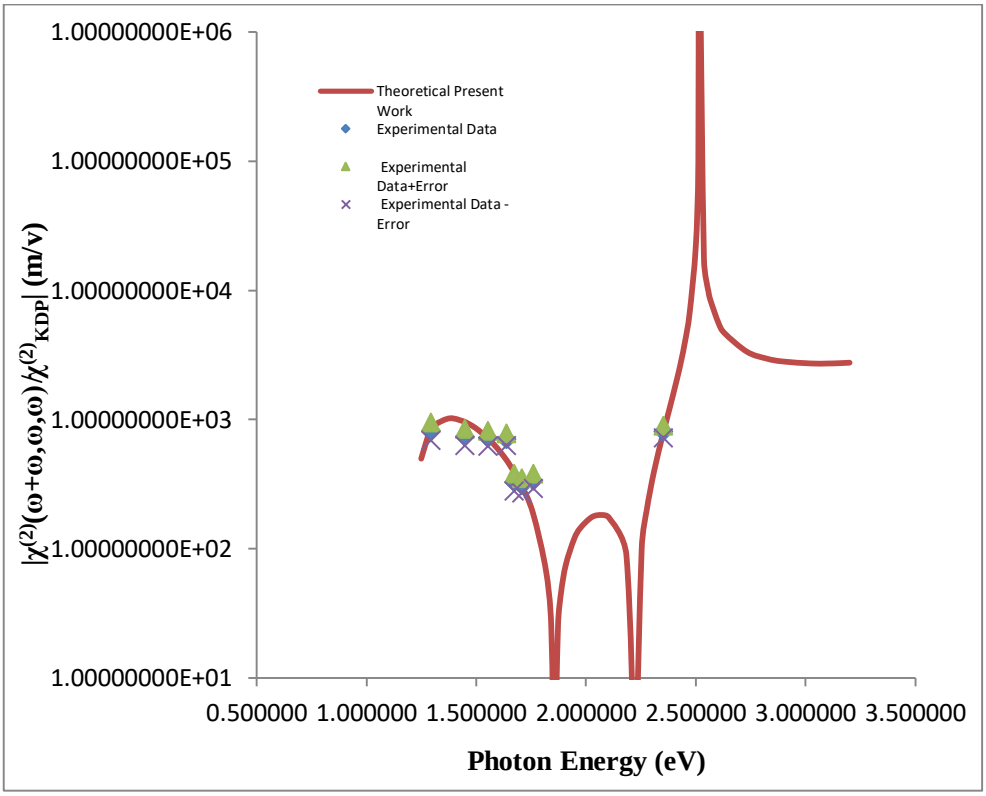

Figure 1. Normalized SHG Susceptibility Coefficient (Absolute value) [$|\chi^{(2)}(\omega+\omega,\omega,\omega)/\chi^{(2)}KDP^{36}|$] shows its variation with Photon Energy of radiation hitting the crystal of InAs. Normalization is done with $\chi^{(2)}KDP^{36}$ =0.39 pm/V(at1064 nm).

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 ωe/2 = 3.8224E+15 rad/s (≈ 2.514957 eV) which is caused by the doubling of the applied field (fundamental) frequency (SHG process) equal to ωe/2, at which the electronic oscillators get in their resonance-mode and causing the maximum (infinite) absolute value of the SHG susceptibility. Near ωe/2, InAs show large SHG susceptibility but it falls exponentially in the region near ωe/2. For 2.822 eV – 3.200 eV, InAs crystal shows a very small variation in the dispersion and hence refers to an almost constant SHG response concerning this special band of frequencies.

5.2. LE-O Results

Computed results of LE-O susceptibility coefficients are illustrated in Fig.2. InAs show large values of absolute LE-O susceptibility coefficient in the region 1.250 eV – 1.645 eV of NIR region. Around 2.223400 eV, LE-O susceptibility has lower absolute values (including the lowest at 2.223353 eV).

InAs shows large absolute values of LE-O coefficient in the region 1.250 eV – 1.645 eV of the NIR region. Thus it is found that for these regions, InAs can be more useful to fabricate the SHG based NLO devices than other NLO materials.

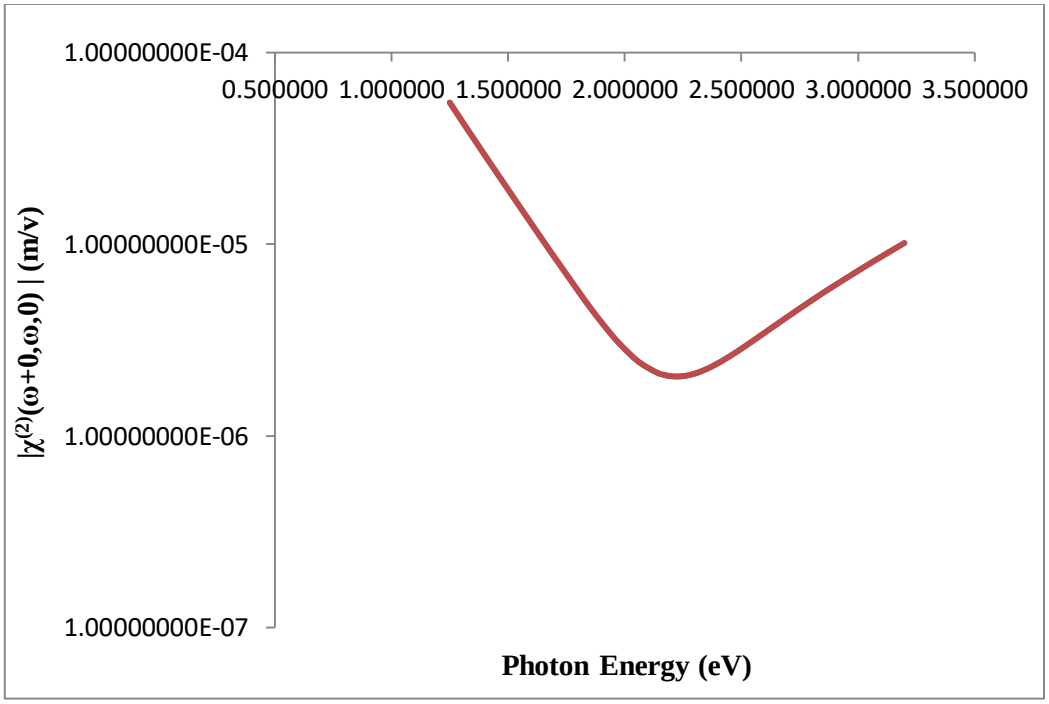

Figure 2. Linear Electro-Optic (LE-O) Susceptibility Coefficient (Absolute value) [$|\chi^{(2)}(0+\omega,0,\omega)|$(m/V)] shows its Variation with Photon Energy of Radiation hitting the Crystal of InAs.

6. Conclusion

As 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 to the resultant SHG coefficients, gives results very closely true to the

experimental results. So, discarding the linear absorption corresponding to $Im\chi_i^{(1)}(\omega)$ susceptibility 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 InAs crystal in the fabrication of nonlinear electro-optic devices such as electro-optic modulators and electro-op rectifiers. Further, a comparative study of the findings obtained here can be done with results [13] calculated through the modelling that accounts for the linear absorption of radiation.

Appendix-A

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

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

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

$$\begin{matrix} 0 & 0 & 0 & d_{123} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{123} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{123} \end{matrix}$$

So, U gets the form,

$U = A_{123}q_{i1}q_{i2}q_{i3} + A_{123}q_{i2}q_{i3}q_{i1} + A_{123}q_{i3}q_{i1}q_{i2} + B_{123}q_{i1}q_{i2}q_{e3} + B_{123}q_{i2}q_{i3}q_{e1} + B_{123}q_{i3}q_{i1}q_{e2} + C_{123}q_{i1}q_{e2}q_{e3} + C_{123}q_{i2}q_{e3}q_{e1} + C_{123}q_{i3}q_{e1}q_{e2} + D_{123}q_{e1}q_{e2}q_{e3} + D_{123}q_{e2}q_{e3}q_{e1} + D_{123}q_{e3}q_{e1}q_{e2}$. (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)

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

Now, U may take the form,

$U = A\, q_i^3 + B\, q_i^2\, qe + C\, q_i\, q_e^2 + D\, q_e^3$, (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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