Second Order Difference-Frequency Generation and Linear Electro-Optic Susceptibilities of GaP Crystal: Linear Absorption Discarded

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ABSTRACT: *A model involving two coupled anharmonic oscillators (electronic and ionic) is applied here for theoretical computation of the second order Difference-Frequency Generation (DFG) and Linear Electro-Optic (LEO) susceptibilities of GaP crystal. The crystal of GaP belongs to III-V group compounds owing to a cubic zinc-blende type structure. Linear absorption is discarded for the selected spectral region of 0.112 eV - 0.332 eV. So, the contribution of the imaginary part of the involved complex linear ionic susceptibility $_i^{(1)}(\ )$ to the resultant DFG and LE-O susceptibilities is neglected in the computation of DFG 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 DFG susceptibility observed in the region of 0.112 eV - 0.326 eV. Applying such calculated nonlinear strength factors in the concerned model expressions, DFG and LE-O susceptibility coefficients are computed as a function of frequency to illustrate and hence to study the dispersion in the region of 0.112 eV - 0.332 eV.*

Keywords: *Nonlinear Optics, Difference-Frequency Generation, LE-O Susceptibility Coefficient.*

1. INTRODUCTION

Extensive advances have been made to understand as well as to apply nonlinear optical interactions since the invention of the laser in 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 behavior of zinc-blende-type materials that are both diatomic and cubic. GaP 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 Difference Frequency Generation (DFG) and Linear Electro-Optic (LE-O) coefficient for III-V group compounds to which GaP 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 GaP along with other III-V group semiconducting compounds. Presently, the author applied a model to the GaP crystal to compute its nonlinear optical properties (DFG and LE-O susceptibility coefficients) in the selected spectral region of 0.112 eV - 0.332 eV. 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 DFG coefficients as a function of the frequency. This way, the dispersion in the Far Infrared (FIR) and near- visible (NV) region of 0.112 eV - 0.332 eV 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

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

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

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

$$\chi^{(1)} = \frac{N}{\epsilon_0}[e\ q\ + e\ q\ ] = \frac{N}{\epsilon_0} e\ q\ + \frac{N}{\epsilon_0} e\ q\ = \chi_e^{(1)} + \chi_i^{(1)}$$

Where $\chi_e^{(1)}$ and $\chi_i^{(1)}$ are electronic and ionic susceptibility respectively. As, $q = \frac{e_e E}{(\ )}$, one gets, $\chi_e^{(1)} = \frac{Ne_e^2}{\epsilon_0 (\ )}$, where, $D\ (\omega) = \omega_e^2 - \omega^2$. 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, $\ddot{q}_7 + \tau \dot{q}_7 + \omega_i^2 q_i = \frac{e_i}{\mu}$ E. It gives,

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

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

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

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

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

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

$$\ddot{q}\ = -\omega^2 q\ + (\ )E - (\ )q_2 - (\ )q\ q\ - (\ )q_2. \tag{5}$$

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

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

$$E = \frac{1}{2}[E_1\ e^{-j\omega_1 t} + C.C. + E_2\ e^{-j\omega_2 t} + C.C.] \tag{7}$$

Here, q_e and 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,

$$= {}^{(1)} + {}^{(2)}(0)| + {}^{(2)}(0)| + \frac{1}{2}[{}^{(2)}(\ - \omega_2)e^{-j(\omega_1-\omega_2)t} + q^{(2)}(2\omega_1)e^{-j(2\omega_1)t} + q^{(2)}(\omega_1 + \omega_2)e^{-j(\omega_1+\omega_2)t} + q^{(2)}(2\omega_2)e^{-j(2\omega_2)t} + C.C.)] \tag{8}$$

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

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

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

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

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

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

(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 $\mathrm{Re}3^{(1)}(\omega)$ of $\chi_i^{(1)}(\omega)$ is used in place of $\chi_i^{(1)}(\omega)$ in the computation of DFG and LE-O coefficients for the GaP crystal for the selected spectral range.

3.1. DFG Susceptibility Coefficient

For 2nd order Difference-frequency mode, $\omega_1 - \omega_2 = \omega - \omega_L$, where ω_L is visible laser frequency. So, the Eq. (12) gives the expression for DFG as,

$$3^{(2)}(\omega - \omega_L, \omega, \omega_L) = -\frac{0^2}{2}\{(\frac{3}{2\ 3})[3^{(1)}(\ ).3^{(1)}(\ ).3^{(1)}(\omega - \omega_L)] +$$
$$(\frac{C}{2})[3^{(1)}(\omega).\mathrm{Re}3^{(1)}(\omega\ ).3^{(1)}(\omega - \omega\ ) + \mathrm{Re}3^{(1)}(\omega).3^{(1)}(\omega\ ).3^{(1)}(\omega - \omega\ )] +$$
$$(\frac{C}{2\ 2})[3^{(1)}(\omega).3^{(1)}(\omega\ ).\mathrm{Re}3^{(1)}(\omega - \omega\ )] + (\frac{B}{2})[3^{(1)}(\omega).\mathrm{Re}3^{(1)}(\omega\ ).\mathrm{Re}3^{(1)}(\omega - \omega\ ) +$$
$$3^{(1)}(\omega\ ).\mathrm{Re}3^{(1)}(\omega).\mathrm{Re}3^{(1)}(\omega - \omega\ )] + (\frac{B}{2\ 2})[\mathrm{Re}3^{(1)}(\omega).\mathrm{Re}3^{(1)}(\omega\ ).3^{(1)}(\omega - \omega\ )] +$$
$$(\frac{3A}{2\ 3})[\mathrm{Re}3^{(1)}(\omega).\mathrm{Re}3^{(1)}(\omega\ ).\mathrm{Re}3^{(1)}(\omega - \omega\ )]\} \quad (13)$$

3.2. LE-O Susceptibility Coefficient

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

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

$$(\quad)[3^{(1)}(\ ).\mathrm{Re}3^{(1)}(0).3^{(1)}(\ ) + 3^{(1)}(0).\mathrm{Re}3^{(1)}(\ ).3^{(1)}(\ )] +$$

$$(\quad)[3^{(1)}(\ ).3^{(1)}(0).\mathrm{Re}3^{(1)}(\ )\ ] + (\quad)[3^{(1)}(\ ).\mathrm{Re}3^{(1)}(0).\mathrm{Re}3^{(1)}(\ ) +$$

$$3^{(1)}(0).\mathrm{Re}3^{(1)}(\ ).\mathrm{Re}3^{(1)}(\ )] + (\quad)[\mathrm{Re}3^{(1)}(\ ).\mathrm{Re}3^{(1)}(0).3^{(1)}(\ )] +$$

$$(\quad)[\mathrm{Re}3^{(1)}(\ ).\mathrm{Re}3^{(1)}(0).\mathrm{Re}3^{(1)}(\ )]\}. \qquad (14)$$

Where, Linear electronic susceptibility, $3^{(1)}(\ ) = \frac{e^2 e}{m_e\epsilon_0(\omega^2-\omega^2)}, \rightarrow\rightarrow Real.$

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

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 DFG 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 GaP.

Parameter	Sym.	Value	Unit
Electronic Oscillator Density	N_e	2.490000E*+28	m^{-3}
Electronic Charge **	e_e	-1.600000E-19	C
Electronic Mass**	m_e	9.109999E-31	Kg
Electronic Resonant Frequency	ω_e	8.880000E+15	rad/s
Ionic Oscillator Density	N_i	2.490000E*+28	m^{-3}
Reduced Mass of Electronic & Ionic Oscillator ***	μ	3.560000E-26	Kg
TO Phonon Frequency	ω_i	6.700000E+13	rad/s
Ionic Charge	e_i	1.600000E-19	C
Damping Rate	τ	6.900000E+11	rad/s
The permittivity of Free Space **	ε_0	8.854000E-12	$C^2/N.m^2$
Visible Laser Frequency	ω_L	7.539800E+13	rad/s

E* ± n = x $10^{\pm n}$, ** Standard data, * ** Calculated data.

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

Photon Energy,[eV]	Frequency,[xE+15 rad/s]	Normalized DFG Susc. Coefficient
0.018	2.7090	-(1.96±0.70)E-01
0.029	4.4280	(5.32±1.50)E-01
0.040	6.0300	(1.16±0.36)E+01
0.052	7.8750	(3.10±0.92)E+00

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

N S F	Value, [kg/ms^2]
A	6.33700963E+29
B	-3.20162963E+30
C	5.39177568E+30
D	-3.02672167E+30

4.2. DFG and LE-O Coefficients

So calculated factors A, B, C, and D (Table 2), are applied in Eq. (13) and Eq. (14) to compute DFG and LE-O susceptibility coefficients respectively, at several different frequencies in the selected spectral region of 0.112 eV - 0.332 eV. 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. DFG Results

Following the present model, the computed results of DFG (absolute values) are plotted as a function of photon energy, in Fig.1. GaP shows large absolute values of DFG susceptibility for the range 0.112 eV - 0.332 eV that belongs to the FIR and near- visible (NV) region. The dips resulted from the sum of some +ive and –ive values of the cross terms involved in the concerned expression.

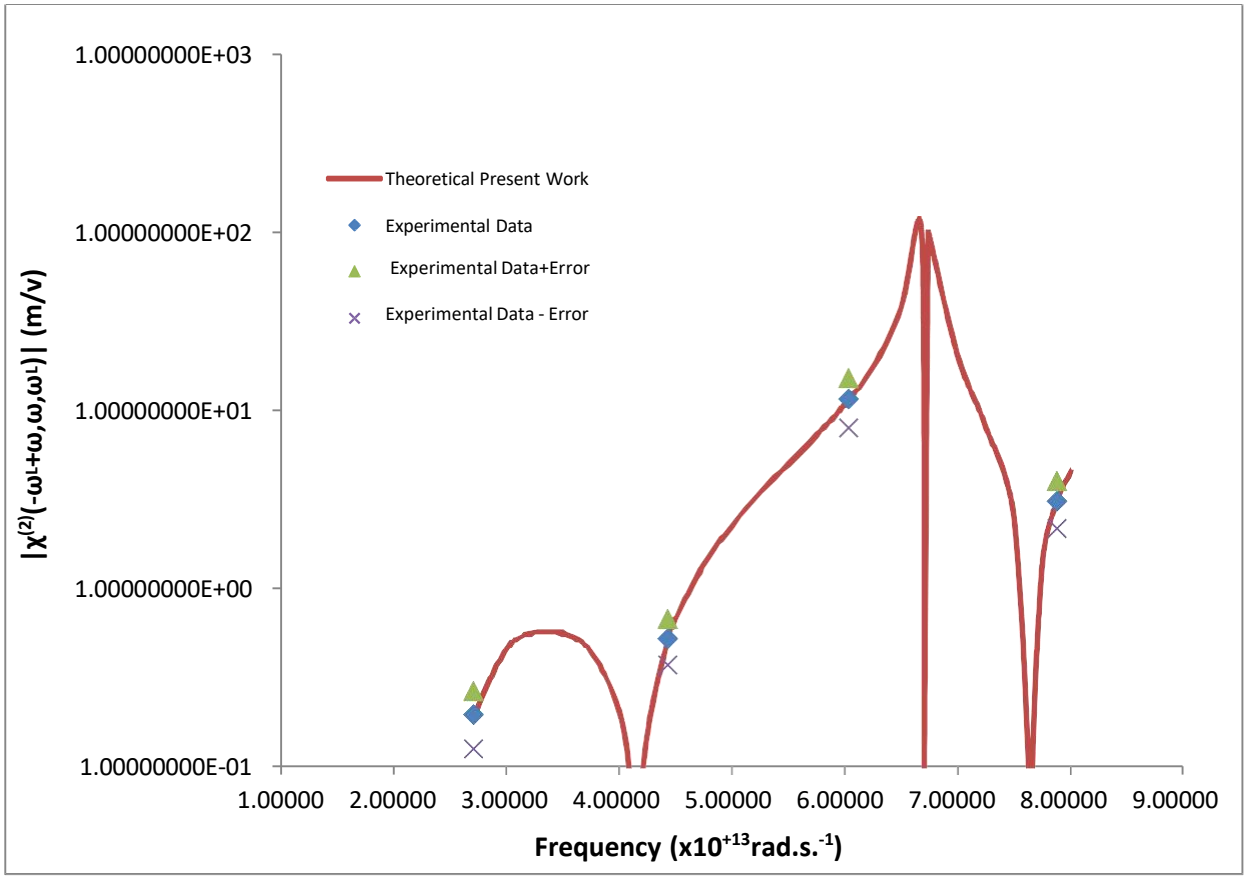

Figure 1: Normalized DFG Susceptibility Coefficient (Absolute value) [$|\chi^{(2)}(\omega+\omega,\omega,\omega)/\ \chi^{(2)}KDP^{36}|$] shows its variation with Photon Energy of radiation hitting the crystal of GaP. Normalization is done with $\chi^{(2)}KDP^{36}$ =0.39 pm/V(at1064 nm).

The peak (absolute) value of DFG susceptibility occurs at 6.6669E+13 rad/sec and not at the TO phonon(ionic resonant) frequency ωi (= 6.7000E+13 rad/sec). It is due to the fact that as we towards the ionic resonant frequency ωi (= 6.7000E+13 rad/sec), the ionic contribution to the absolute value of DFG susceptibility goes to decrease and becomes zero at ωi .The results obtained through the current model, reasonably, are not in close agreement with the results experimentally observed by Faust and Henry[9].

5.2. LE-O Results

Computed results of LE-O susceptibility coefficients are illustrated in Fig.2. GaP shows a very gradual (nearly uniform) increase in absolute LE-O susceptibility coefficient in the FIR region with frequencies ranging from 2.7090E+13 rad/sec to 6.0301E+13 rad/sec. The absolute LE-O coefficient attains its peak value at frequency 6.06669E+13 rad/sec. In the close vicinity of ionic resonant frequency ωi, GaP shows large absolute values of LE-O susceptibility while at ωi, there is a dip corresponding to completely zero contribution from the linear ionic susceptibility. Beyond this, there is a fall in absolute LE-O values up to the frequency of 7.5000E+13 rad/sec. Obviously, in the considered spectral region 0.112 eV - 0.332 eV, the obtained LE-O results are of some oscillatory nature.

Thus it is found that for this region, GaP can be potentially useful to fabricate the DFG -based NLO devices than other NLO materials.

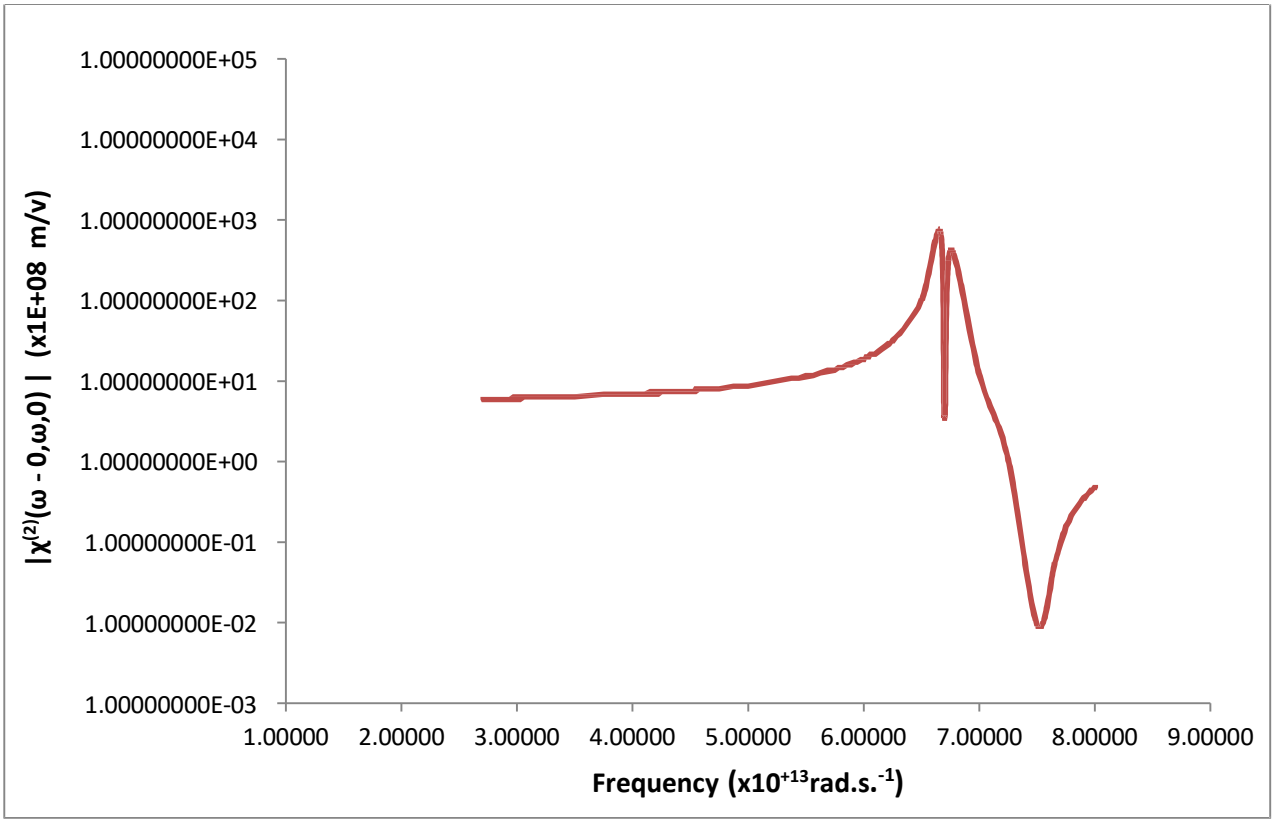

Figure 2: Linear Electro-Optic (LE-O) Susceptibility Coefficient (Absolute value) [$|\chi^{(2)}(\omega - 0,\omega,0)|$(m/V)] shows its Variation with Photon Energy of Radiation hitting the Crystal of GaP.

6. CONCLUSION

As the results, obtained in the present work, are in good agreement with the experimental9 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 DFG coefficients is quite right. This implies that in the selected spectral region (0.112 eV - 0.332 applied for the other III-V group compounds like InAs [12] successfully for the estimation of its SHG (second harmonic generation) and LE-O susceptibilities 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, w h i c h indicates the potential application of GaP crystal in the fabrication of nonlinear electro-optic devices such as electro-optic modulators and electro-optic 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,

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

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

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

So, U gets the form,

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

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,

$$U = A\, q_i^3 + B\, q_i^2\, qe + C\, q_i\, q_e^2 + D\, q_e^3, \quad (A\text{-}4)$$

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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