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Linearity and Non-linearity of Photorefractive effect in Materials using the Band transport Model.

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Abstract

In this paper we have studied the Linearity and Non-linearity of Photorefractive effect in materials using the band transport model. For low light beam intensities the change in the refractive index is proportional to the electric field for linear optics while for non- linear optics the change in refractive index is directly proportional to the square of the electric field. From our studies, it can be seen that the diffusion field is inversely proportional to period of interference pattern while the space charge field is directly proportional to the period of interference pattern. In the absence of the external electric field the space-charge field is spatially phase shifted by π/2 relative to the interference fringe pattern, which plays a key role in the energy exchange in two-wave mixing. In the presence of the external electric field the scaling factor is a complex quantity indicating that the externally applied electric field does not only change the magnitude of the space-charge field but also alters its spatial phase.

1.0 Introduction

The refractive index of a medium, in general, depends on the arrangement of atoms and the distribution of electrons. For low light beam intensities the index of refraction is a constant depending on the frequency (or wavelength) of the light. Under the illumination of light, the charges (mostly electrons) are driven up and down by the electric field of the light beam and these moving electrons then radiate and generate a field which is proportional to the incident field which yields a phase shift in the transmitted field. If the intensity of the incident beam (sunlight or ordinary light) is small (~10V/cm) as compared to the intra-atomic field (10^7 V/cm), the phase shift introduced is independent of the intensity. In other words, the refractive index is independent of the intensity of the light beam. This is the regime of linear optics. On the other hand when the intensity of the light beam is comparable to the intra-atomic electric field, the distribution of electrons in the medium can be modified by the radiation field [1]. This leads to intensity dependence of the refractive index. The reflection and refraction at an interface and the propagation through a medium will now depend on the intensity of the incident beam. This is the regime of non-linear optics. Non-linear optics is the study of the interaction of light beams with the matter when the materials respond non-linearly to the amplitude of the electric field of the light beam.

Photorefractive effect is a phenomenon in which the local refractive index of the medium is changed by the spatial variation of light intensity. This effect was discovered in 1966 when researchers were studying the transmission of laser beams through electro-optical crystals. There are a number of photorefractive materials, e.g., lithium niobate ($LiNbO_3$), lithium tantalite ($LiTaO_3$), barium titanate ($BaTiO_3$), potassium tantalate niobate (KTN), barium sodium niobate (SBN). In general the electro-optic effect arises in photorefractive materials in two forms: (i) linear electro-optic (LEO) effect or Pockels effect and (ii) quadratic electro-optic (QEO) effect or Kerr effect.

Photorefractive (PR) materials have attracted interest in recent years because of their potential applications in the fields of holographic storage, phase conjugation, optical image processing, and pattern recognition, among others [1-2]. The PRE that occurs in systems of liquid crystals (LCs) with dopants is especially interesting and important because such systems exhibit large optical nonlinearity. During the past decade, several researchers have examined this effect in LCs [3–9]. Briefly, spatial charges are generated in response to the establishment of an interference pattern with a spatial intensity modulation created by two coherent light beams with identical polarization. Applying a dc voltage establishes a space-charge field by charge separation (diffusion and drift); then a nonlocal sinusoidal index-modulated pattern is formed, which leads to a PRE.

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Azo dyes exhibit super photoanisotropic characteristics, including anisotropic absorption (dichroism) and birefringence [10]. These characteristics make LC films doped with azo dyes appropriate for polarization holographic studies. Studies of the PRE of dye-doped LC (DDLC) systems have been performed in a holographic setup because the spatial intensity-modulation pattern formed by two coherent beams is required for inducing the PRE. We verify that a PRE can be observed and studied in a polarization grating in a DDLC film. By use of a suitable polarization-modulated interference pattern, the anisotropy characteristics of an azo dye can induce a spatially absorption-modulated pattern and then produce a space-charge field, which, in turn, has a PRE. This nonlocal effect is verified by measurement of the two-beam coupling (TBC) of the two pump beams.

Although the photorefractive effect (PRE) in LiNbO3 crystals has recently been in the focus of interest mostly due to its usefulness for optical memories [11- 12] or construction of elements for optical circuits [13-14], there is still space for studying its nature, after more than forty years from the first reference to the effect, [15 -16]. Today, there are several models known describing the photorefractivity of crystals but as default they all consider the electric field built up by redistribution of the charge carriers and electro optic effect (EOE) as dominant mechanisms responsible for the light-induced refractive index inhomogeneities [17–19]. For very long time, investigations of the kinetics of the recording process by means of illumination with periodic (harmonic) spatial distribution of intensity were carried out in order to come to these conclusions but, to the best of our knowledge, only rarely was the attention paid to application of the aperiodic illumination which gives more complex information on the origin of PRE. However, also in the cases where aperiodic illumination is used one has to take care of the proper choice of the spatial distribution of the illumination in order to get as unambiguous and convincing results as possible. For example, the illumination used in [20-21] was aperiodic and 2-D spatially distributed in its nature. Thus, the observed results can be due to a combination of several effects which arise in 2-D case but do not occur in 1-D case.

The photorefractive effect occurs in several stages:

A photorefractive material is illuminated by coherent beams of light. (In holography, these would be the signal and reference beams). Interference between the beams results in a pattern of dark and light fringes throughout the crystal.

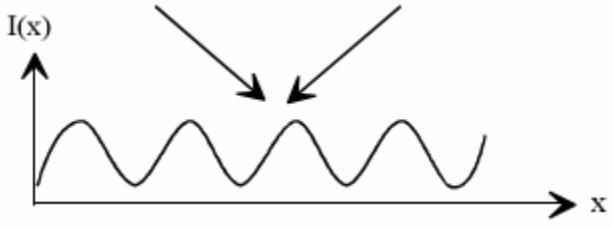

Figure 1: [24] shows a photorefractive material illuminated by a coherent beam of light.

In regions where a bright fringe is present, electrons can absorb the light and be photo excited from an impurity level into the conduction band of the material, leaving an electron hole (a net positive charge). Impurity levels have an energy intermediate between the energies of the valence band and conduction band of the material. Once in the conduction band, the electrons are free to move and diffuse throughout the crystal. Since the electrons are being excited preferentially in the bright fringes, the net electron drift is towards the dark-fringe regions of the material. The electrons migrate due to diffusion and the photovoltaic effect to the dark regions. Thus a space charge field is formed.

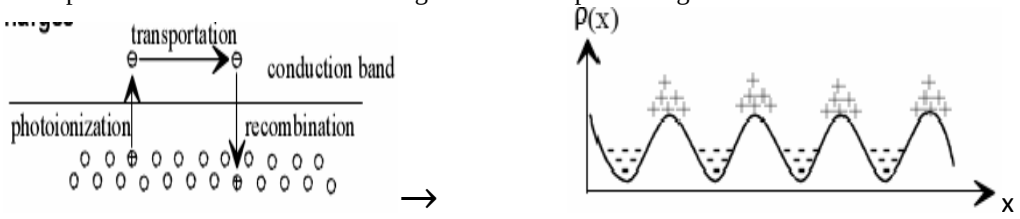

Figure 2: [24] shows the photoionization of electron to the conduction band of the photorefractive material and the formation of a space charge field.

To explain the phenomenon of photorefractive effect a number of models have been introduced. In order to understand the PRE, we have used the band transport model.

2.0 Band Transport Model

The band transport model [2] is one of models used to explain PRE. In this model it is assumed that the photorefractive materials contain certain types of impurities, namely, donor and acceptor impurities. All the donor

impurities are identical and have exactly the same energy state which lies in the middle of the band gap. These donor impurities can be ionized by absorbing photons. As a result of the ionization electrons are generated in the conduction band leaving an empty state behind. Such ionized impurities are capable of capturing electrons. The density of donors is often much larger than that of the acceptor impurities. It is assumed here that all the acceptor impurities are also identical. The neutral donor impurities are capable of donating electrons due to photo- excitation and ionized ones are capable of capturing these photo-electrons. The presence of acceptor impurities is to maintain the charge neutrality only and these do not participate in the photorefractive effect.

If N_D is the donor density of which N^i_D are ionized, the rate of electron generation is (sI+β)(N_D - N^i_D) while the rate of trap capture is $\gamma_R N N^i_{D,}$ where N is the electron density, s is the photo-excitation cross-section, I is the light intensity, β is the rate of thermal generation of electrons and γ_R is the electron-ionized trap recombination rate. Neglecting the thermal generation ($sI >>$ β) the rate equation for the electron density in a photorefractive material is written as [25-26],

$$\frac{\partial N_D^i}{\partial t} = sI\left(N_D - N_D^i\right) - \gamma_R N N_D^i \tag{1}$$

For each ionized impurity an electron is generated while for each capture impurity, an electron is eliminated. The rate of the electron generation is the same as the rate of the impurity ionization, except that the electrons are mobile whereas the impurities are immobile which is essential for the photo-refractive effect. The rate equation for the electron density can be written as,

$$\frac{\partial N}{\partial t} - \frac{\partial N_D^i}{\partial t} = \frac{1}{q}\vec{\nabla}.\vec{J} \tag{2}$$

where, $\vec{J}$ is the current density and q is the electronic charge. Presence of these charge carriers (electrons) leads to a space-charge field which in turn affects the transport of the charge carriers. The current density comprises of contributions from the drift of the charge carriers due to the applied electric field and the diffusion due to the gradient of carrier density and it is given by,

$$\vec{J} = qN\mu\rho\vec{E} + k_B T\vec{\nabla}N \tag{3}$$

where, μ is the mobility tensor, $\vec{E}$ is the applied electric field, k_BT is the product of the Boltzman constant and temperature. The electric field $\vec{E}$ obeys the Poisson equation given by,

$$\vec{\nabla}.\varepsilon\vec{E} = \rho(\vec{r}) = -q\left(N + N_A - N_D^i\right) \tag{4}$$

where, ε is the dielectric tensor, $\rho(\vec{r})$ is the charge density and N_A is the acceptor impurity density. The presence of acceptor impurities is to maintain the charge neutrality. In the absence of light illumination the charge neutrality is written as,

$$N + N_A - N_D^i = 0 \tag{5}$$

In the absence of light, $N_A = N_D^i$ for small electron densities. On the RHS of the equation (3), the first term represents contribution due to the drift current and the second term represents the contribution due to the diffusion current, here the photovoltaic current contribution has been neglected. Consider incidence of two laser beams of fields $\vec{E}_a$ and $\vec{E}_b$ giving rise to the resultant field $\vec{E}$ given by,

$$\vec{E} = \overrightarrow{E_a} exp\left(iwt - i\overrightarrow{k_a}.\vec{r}\right) + \overrightarrow{E_b} exp\left(iwt - i\overrightarrow{k_b}.\vec{r}\right) \tag{6}$$

If the states of polarizations of the two beams are not orthogonal the two beams form an interference pattern with the resultant intensity I given by,

$$I(\vec{r}) = I_0 + Re\left(I_1 e^{-\vec{k}.\vec{r}}\right) \tag{7}$$

where,

$$I_0 = \left|\overrightarrow{E_a}\right|^2 + \left|\overrightarrow{E_b}\right|^2 \tag{8}$$

$$I_1 = 2\overrightarrow{E_b}.\overrightarrow{E_b^*} \tag{9}$$

and $\vec{K}$ is the grating wave-vector defined by,

$$\vec{K} = \overrightarrow{k_b} - \overrightarrow{k_a} \tag{10}$$

The magnitude of the grating wave-vector is related to the period (Λ) of the interference pattern by,

$$K = \frac{2\pi}{\Lambda} \tag{11}$$

The maximum fringe visibility is obtained for $\vec{E}_a = \vec{E}_b$ and in such a case one has,

The equation (4.14) represents an array of interference fringes with the dark fringe occurring for $\vec{k}\cdot\vec{r} = (2m+1)\pi$. For $\vec{k}\cdot\vec{r} = 2m\pi$ one has the bright fringes with the maximum intensity $I = 2I_0$. In the bright regions photo-ionized charges are generated by the absorption of light which diffuse away from the bright regions leaving behind positively charged ionized donors. When these charge carriers are trapped in the dark regions, these remain there as there is no light to re-excite them, which leads to a charge separation. Consequently, due to the illumination with periodic intensity in the photorefractive medium, the dark regions are negatively charged and the bright regions are positively charged. The buildup of the space charge continues unless the diffusion current is counterbalanced by the drift current. The space-charge density is given by,

$$\rho = \rho_0 \cos \vec{k}\,.\,\vec{r} \tag{12}$$

where, ρ_0 is a constant. On integrating the equation (4) yields,

$$\vec{E} = \rho_0 \left(\frac{\vec{K}}{\vec{K}.\varepsilon\vec{K}}\right) \sin\left(\vec{K}.\vec{r}\right) \tag{13}$$

It can be noted that the space-charge field is shifted in phase by π / 2 relative to the intensity pattern and induces a change in the refractive index via the Pockels effect. To summarize, the photorefractive effect basically consists of the following five processes occurring in the electro optic crystals:

- Photo-ionization of impurities and the generation of the charge carriers.
- Transport of these charge carriers
- Trapping of the charge carriers and the formation of the space-charge density.
- Formation of the photo-induced space-charge electric field.
- the formation of the index grating via the LEO (Pockels) effect .

The photorefractive effect is a macroscopic phenomenon and it requires the generation and transportation of a large number of charge carriers (~ 10^{15}/cm^{-3}).

For a given beam intensity one needs to solve for the space-charge field as a function of both $\vec{r}$ and t using the equations (1–4). However, closed form solutions of these equations are not available. In the case of the steady state the time derivatives are zero and the equations (1-4) gives,

$$sI\left(N_D - N_D^i\right) - \gamma N N_D^i = 0 \tag{14}$$

$$\vec{\nabla}.\vec{J} = 0 \tag{15}$$

$$\vec{J} = qN\mu\,\vec{E} + k_B\,T\mu\,\vec{\nabla}N \tag{16}$$

$$\vec{\nabla}.\varepsilon\vec{E} = \rho\left(\vec{r}\right) = -q\left(N + N_A - N_D^i\right) \tag{17}$$

Using these equations one can solve for $\vec{E}$ in terms of the intensity. For small intensity modulation ($|I_1| << I_0$) closed form solutions are available. Under such condition one can ignore the higher spatial harmonic terms and the steady state solutions can be written as,

$$N(\vec{r}) = N_0 + Re\left(N_1 e^{-\vec{k}.\vec{r}}\right) \tag{18}$$

$$N_D^i(\vec{r}) = N_{D0}^i + Re\left(N_{D1}^i e^{-\vec{k}.\vec{r}}\right) \tag{19}$$

$$\vec{J}(\vec{r}) = \vec{J}_0 + Re\left(\vec{J}_1 e^{-\vec{k}.\vec{r}}\right) \tag{20}$$

$$\vec{E}(\vec{r}) = \vec{E}_0 + Re\left(\vec{E}_1 e^{-\vec{k}.\vec{r}}\right) \tag{21}$$

Where $N_0, N_1, N_{D0}^i, N_{D1}^i, \vec{J}_0, \vec{J}_1, \vec{E}_0,$ and $\vec{E}_1$ are constants. Now the problem is to solve for these constants in terms of the intensity constants $\vec{I}_0$ and $\vec{I}_1$. We consider the limiting case when $\vec{I}_1 = 0$ which occurs when either one beam is absent or the two beams have crossed polarizations ($I_1 = 2E_b.E_a^* = 0$)

2.1 Uniform Illumination ($\vec{I}_1 = 0$)

In this case there is no spatial variation of all the physical quantities. The charge carrier densities are related by,

$$sI\left(N_D - N_D^i\right) = \gamma_{R\,N\,N_D^i} \tag{22}$$

$$N + N_A - N_D^i = 0 \tag{23}$$

The equations (22)-(23) can be used to find N and N_D^i . From the equation (22) one gets,

$$N_D^i = \frac{N_D\, sI}{sI + \gamma_R N} \tag{24}$$

Equations (23) and (24) yield the relation,

$$\gamma_{R N^2} + (sI + \gamma_R N_A)\, N + sI\,(N_A - N_D) = 0 \tag{25}$$

For the conditions, $N << N_A$ and $sI << \gamma_R N_A$ one gets the value for N as,

$$N = \frac{N_D - N_A}{\gamma_R N} sI \tag{26}$$

The equation (24) can also be written as,

$$N_D^i = N_A \frac{N_D - N_A}{\gamma_R N} sI \tag{27}$$

2.2 Periodic Illumination ($I_1 \neq 0$)

This is the case when the two beams intersect inside the medium. Substituting the equations (18-21) and neglecting the higher order terms one has,

$$sI_1 (N_D - N_{D0}^i) + sI_0 (-N_{D1}^i) - \gamma_R \gamma_R N_1 N_{D0}^i - \gamma_R N_0 N_{D1}^i = 0 \tag{28}$$

$$sI_0 \left(N_D - N_{D0}^i\right) - \gamma_R N_0 N_{D0}^i = 0 \tag{29}$$

$$\vec{K}.\left(qN_1\mu\,\vec{E}_0 + qN_1\mu\,\vec{E}_1 - ik_b\,T\mu\vec{K}N_1\right) = 0 \tag{30}$$

$$-i\vec{K}.\varepsilon\vec{E}_1 = -q\left(N_1 - N_{D1}^i\right) \tag{31}$$

$$N_0 + N_A - N_{D0}^i = 0 \tag{32}$$

In the equations (28-32) N_A, N_D and E_0 are constants and N_0 and N_{D0}^i can be obtained by solving the equations (28) and (32); the results are the same as the equations (24) and (26). The other three equations can be employed to solve for N_1, N_{D1}^i and E_1.

For the steady state case the Maxwell's equation $\vec{\nabla} \times \vec{E} = 0$ implies that $\vec{K} \times \vec{E}_1 = 0$, which suggests that $\vec{E}_1$ is parallel to $\vec{K}$. After some manipulation one obtains the relation for the space charge field, E_1 as,

$$E_1 = \frac{iK\,\frac{k_B T}{q} - \frac{\vec{K}.\mu\vec{E}_0}{(\mu)K}}{1 + \frac{K^2}{k_D^2} + i\frac{q\,\vec{K}.\mu\vec{E}_0}{(\mu)k_B T k_D^2}} \frac{I_1}{I_0} \tag{33}$$

Where,

$$k_D^2 = \frac{q^2}{(\varepsilon)k_B\,T}\left(\frac{N_A}{N_D}\right)(N_D - N_A) \tag{34}$$

$$(\varepsilon) = \frac{\vec{K}.\varepsilon\vec{K}}{K^2} \tag{35}$$

$$(\mu) = \frac{\vec{K}.\mu\vec{K}}{K^2} \tag{36}$$

In arriving at the equation (33) it is assumed that (i) $\gamma_R N >> sI_0$ and (ii) $N_D\, sI_0 << \gamma_R\, N^2$, in addition to the small modulation approximation ($I_1 << I_0$). When the dc field is also parallel to $\vec{K}$ the equation (33) reduces to,

$$E_1 = \frac{iK\ \frac{k_B T}{q} - \frac{\mu \vec{E}_0}{(\mu)}}{1+\frac{K^2}{k_D^2}+i\frac{q\,\mu \vec{E}_0}{(\mu)Tk_D^2}}\ \frac{I_1}{I_0} \tag{37}$$

For the case of pure diffusion $\vec{E}_0 = 0$ and hence, the above equation becomes,

$$E_1 = \frac{iK\ \frac{k_B T}{q}}{1+\frac{K^2}{k_D^2}}\ \frac{I_1}{I_0} \tag{38}$$

The equations (36) and (38) for the amplitude of the space charge field are key results of the band transport model. The parameter k_D is known as the Debye wave-number. In the theory of electrodynamics in plasmas the electrons move in such a way as to screen out the Coulomb field of the test charge in a distance of $\Lambda_D = 2\pi/K_D$. Such a distance is determined by the valence between the thermal kinetic energy and electrostatic energy of these charge carriers and is also known as the Debye screening radius. For $N_D >> N_A$, K_D is given by,

$$k_D^2 = \frac{q^2 N_A}{\langle\varepsilon\rangle k_B T} = \frac{q^2 N_{D0}^i}{\langle\varepsilon\rangle k_B T} \tag{39}$$

3.0 Space-charge field

3.1 No applied electric field ($E_0=0$)

In the absence of an applied electric field the photoelectrons are transported by diffusion only, which is as a result of the electron density gradient. When the electrons are generated by light, a current is produced by the diffusion and negative and positive charges are separated. The static electric field produced by the buildup of the space charges causes to move these electrons in the opposite direction. In the steady state there is no net current and the electric field is given by the equation (3) which for $N_0 = 0$ becomes ,

$$E = iK\ \frac{k_B T}{q}\ \equiv\ iE_d \tag{40}$$

In the above equation E_d is known as the diffusion field and it is independent of the dopant densities. A given charge separation with the space-charge density
$qN_{D0}^i\ exp(-i\vec{K}\ z)$ produces an electric field known as the space charge field with the magnitude given by,

$$E = i\ \frac{qN_{D0}^i}{K\langle\varepsilon\rangle} = i\frac{qN_A}{\langle\varepsilon\rangle K}\ \equiv iE_q \tag{41}$$

where, it is assumed that $N_A >>\ N_0$. The field E_q also known as the saturation field gives the maximum achievable electric field for a given charge density N_{D0}^i . Using the equations (38), (40) and (41) we can write the space charge field E_1 as,

$$E_1 = \frac{iK\ \frac{k_B T}{q}}{1+\frac{K^2}{k_D^2}}\ \frac{I_1}{I_0}\ = \frac{iE_d}{1+\frac{E_d}{E_q}}\ \frac{I_1}{I_0} \tag{42}$$

It is to be noted that the space-charge field is proportional to I_1 / I_0 . The factor $i(=\sqrt{-1}\ )$ in the above equation represents a spatial phase shift of $\pi/2$; hence the electric field is spatially shifted by ¼ of a period relative to the interference fringe pattern. Such a spatial phase-shift play a key role in the energy exchange in two-wave mixing. According to the above equation (42) E_1 is always less than the smaller of the two fields E_d and E_q . The space-charge field increases linearly with increasing Λ and a reaches a maximum at $\Lambda = \Lambda_D$ when $E_d = E_q$. For grating periods $\Lambda >> \Lambda_1$, the space-charge field decreases like $1/\Lambda$. The maximum field is given by,

$$E_{1max}\ = \frac{1}{2}\left(\frac{N_A K_B T}{\langle\varepsilon\rangle}\right)\frac{1}{2}\frac{I_1}{I_0} \tag{43}$$

The magnitude and location of the maximum depend upon dopant density N_A (N_{D0}^i) . The increase in the maximum space-charge field is significant in the regime of small grating periods. For larger grating spacing the space-charge field is dominated by the diffusion
($E_d >>\ E_q$) and one has ,

$$E_1\ \cong\ iE_d\ \frac{I_1}{I_0} \tag{44}$$

Contrary to this for small grating spacing the space-charge field is given by,

$$E_1 \cong iE_a \frac{I_1}{I_0} \tag{45}$$

3.2 Non-zero applied electric field ($E_0 \neq 0$)

With non-zero electric field the amplitude of the space-charge field is given by the equation (41) which can be written as,

$$E_1 = \frac{iE_q - E_0}{1+\frac{E_d}{E_q}+\frac{E_0}{E_q}} \frac{I_1}{I_0} \tag{46}$$

In the above discussion applied field is taken parallel to the grating wave vector. In this case the space- charge field E_1 is no more limited by the smallest of the two fields E_d and E_q . The space-charge field can also be written as,

$$E_1 = \frac{iE_d}{\left(1+\frac{E_d}{E_q}\right)}\left\{\frac{1+\frac{E_d}{E_q}}{1+i\left(\frac{E_0}{E_d+E_q}\right)}\right\}\frac{I_1}{I_0} \tag{47}$$

The first term on the R.H.S of the above equation is the space-charge field in the absence of the external field while the second term represents the scaling factor due to the presence of the external field. The scaling factor is a complex quantity indicating that the applied electric field not only changes the magnitude of the space-charge field but also alters its spatial phase. From the equation (47) it is clear that when $E_q << E_d$, which is true at large wave-numbers ($k_D^2 << k^2$) or small grating periods, the effect of applied field E_0 is insignificant. The externally applied electric field E_0 has a large effect on the space-charge field when either $E_q >> E_d$ or $E_0 \geq E_d$ .The diffusion and space-charge field can be written in terms of the grating period Λ as,

$$E_d = \frac{2\pi}{\Lambda}\frac{k_B T}{q} \tag{48}$$

$$E_q = \frac{qN_A}{2\pi\langle\varepsilon\rangle}\Lambda \tag{49}$$

From the equations (48) and (49) it can be seen that the diffusion field is inversely proportional to Λ while the space charge field is directly proportional to Λ.

Conclusion

In conclusion, in the absence of the external electric field the space-charge field is spatially phase shifted by $\pi/2$ relative to the interference fringe pattern, which plays a key role in the energy exchange in two-wave mixing. The space-charge field E_1 is always less than the smaller of the two fields E_d and E_q. The space-charge field increases linearly with increasing Λ and reaches a maximum at $\Lambda = \Lambda_D$ when $E_d = E_q$. For grating period's $\Lambda >> \Lambda_D$, the space-charge field decreases like $1/\Lambda$. In the presence of the external electric field the scaling factor is a complex quantity indicating that the externally applied electric field does not only change the magnitude of the space-charge field but also alters its spatial phase. At large wave-numbers or small grating periods, the effect of the externally electric applied field is insignificant. The externally applied electric field E_0 has a large effect on the space-charge field when either $E_q >> E_d$ *or* $E_0 \geq E_d$.

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