Theoretical study of photorefractive effect in reduced potassium niobate crystals

A. Radoua, D. Khatib
Laboratoire de Physique du Solide Théorique, Département de Physique, Faculté des Sciences, BP : 28/S, Université Ibn Zohr, 80000 Agadir, Maroc

Study of photorefractive properties for reduced potassium niobate was performed with using a band model for one impurity center. Which is allowed to study the photoconductivity, to calculate the light induced absorption, to study the space charge field and the exponential gain in the absence of the applied field, as function of the light intensity, the wavelength and the grating spacing. It also allows to study the exponential gain and the phase appear between the space charge field and the illumination as function of the applied field.

I. INTRODUCTION

Photorefractive materials are defined as electro-optic materials in which refractive indices are changed by photoinduced space charge fields via the electro-optic effect. We have already pointed out that the photoinduced space charge fields are due to the generation and the transport of charge carriers. The main three charge transport processes are: the diffusion, the volume photovoltaic effect and the photoconductivity. Since diffusion occurs in any electro-optic material, all such materials which have a photoexcitation mechanism are in principle also photorefractive. In photorefractive crystals a light pattern may be transposed into a refractive index pattern. Inhomogeneous illumination excites charge carriers into conduction or valence band. The charge carriers migrate, currents arise, and finally the carriers are trapped. The excitation rate is smaller in the darker regions and thus charge becomes accumulated in the darker parts of the crystals. Space charge fields build-up and modulates the refractive index via the electro-optic effect. Photorefractive crystals are of special interest for many advanced and promising applications. [1]

II. MODEL DESCRIPTION

The most simple band model allowing to interpret photorefractive phenomenon in reduced $KNbO_3$ crystal with only one type of charge carriers that will be in the occurrence of electrons, is shown in (Fig. 1).

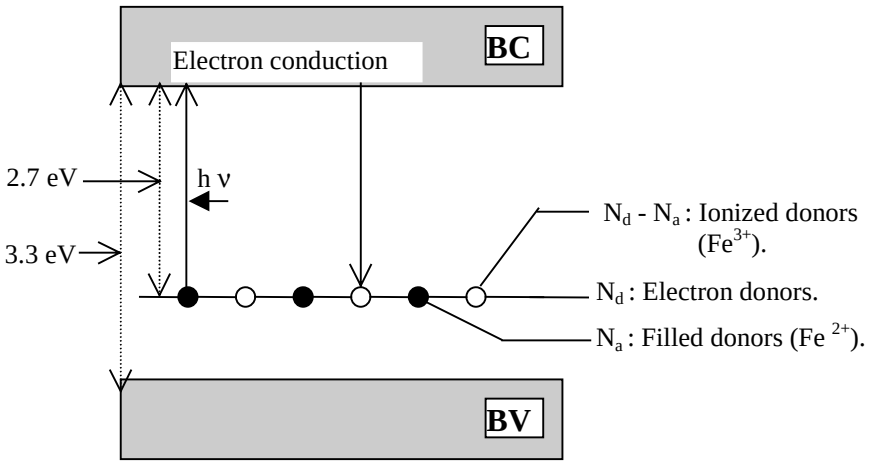

Fig. 1. Simplified band model of $KNbO_3$ for one electron donor center with density N_d. The donors are partially ionized in the dark and at low temperature (density N_a).

The electro-chemical reduction treatment reduces a great majority of impurities to the Fe^{2+} state, which can then act as donor centers. [2, 3]

The band gap energy of 3.3 eV and the position of the donor level at 2.7 eV. [4] Because of the large band gap energy, $KNbO_3$ is in general an insulator with dark conductivities in the range of 10^{-9} - $10^{-14}\ \Omega^{-1}cm^{-1}$. [1, 5] The iron centers are assumed to constitute a donor level with a density N_d. Note that the presence of donor centers is necessary for obtaining photoconductivity and a photorefractive effect, but that it is not compelling to have traps (ionized donors) already in the dark. However, the trap density is most important, since it can limit the maximal space charge field.

The concentration of Fe^{3+} centers in the dark and at low temperature is denoted by the acceptor density N_a. The electrons can be excited from the field donors either by photons (absorption) or by phonons (thermal excitation). They migrate in the conduction band under the applied and the space charge electric field and due to the diffusion. The photovoltaic effect is small in reduced $KNbO_3$ crystals and is neglected.[6] The electrons are retrapped by empty donors.

A description of photorefractive phenomenon was given by Kukhtarev and *al*. for a light-intensity pattern resulting from the interference of two coherent beams I_s and I_p.[7,8]

$$I(z,t) = I_0(t)(1 + m\cos(K_g z)) \tag{1}$$

where m is the modulation index given by $m = 2(I_s I_p)^{1/2} / I_0$; $I_0 = I_s + I_p$ and K_g ($K_g = \frac{2\pi}{\Lambda}$, Λ is the fringe spacing) is the grating wave number.

The basic equations to describe the photorefractive effect are :

$$j = j_{pv} + e\mu nE + D_s\mu\frac{\partial n}{\partial z}, \tag{2}$$

$$\frac{\partial N_d^+}{\partial t} = (N_d - N_d^+)(sI + \beta) - \gamma_r N_d^+ n, \tag{3}$$

$$\frac{\partial (N_d^+ - n)}{\partial t} = -\frac{1}{e}\frac{\partial j}{\partial z}, \tag{4}$$

and

$$\frac{\partial E}{\partial z} = \frac{e}{\varepsilon\varepsilon_0}(N_a + N_d^+ - n) \tag{5}$$

where j is the current density; j_{pv} the photovoltaic current density (it will be able to be neglected ahead the other terms for the case of reduced $KNbO_3$ [6]); e the elementary electric charge; μ the mobility; n the electron density in conduction band; E is the sum of applied and space-charge field; $D_S = k_B T$ the diffusion constant; k_B the Boltzmann constant; T the absolute temperature; N_d the donors density; N_d^+ the ionized donors density; s the electron excitation cross section per photon energy; β the thermal excitation rate; γ_r the electron recombination constant and ε the dielectric constant.

For a small modulation ($m << 1$), it is adequate to use linearized solutions for the free electron density n, the ionized donor density N_d^+ and the electric field E, only the first Fourier component is taken into account.[4]

$$n(z,t) = n_0(t)(1 + Re(M(t)e^{iK_g z}). \tag{6}$$

$$N_d^+(z,t) = N_0(t) + Re(N(t)e^{iK_g z}). \tag{7}$$

$$E(z,t) = E_0(t) + Re(E_{sc}(t)e^{iK_g z}). \tag{8}$$

where n_0 denotes the average free electron density and N_0 the average ionized donor density. The terms *M(t), N(t)* and E_{sc} *(t)* in Eqs. (6)-(8) are complex functions to allow for relative phase shifts to the intensity pattern.

When the Eqs. (1, 6-8) are inserted into equations (2-5), a first order differential equation for the average free electron density

$$\frac{\partial}{\partial t}n_0(t) = (N_d - N_a)(sI_0(t) + \beta) - n_0(t)(sI_0(t) + \beta + \gamma_r N_a) - \gamma_r n_0^{\,2}(t) \tag{9}$$

is found.

III. PHOTOCONDUCTIVITY

The homogeneous stationary illumination of the crystal leads to a constant free electron density and a constant photoconductivity $\sigma_{ph} = e\ \mu\ n_0$ respectively. The free electron density n_0 is the positive solution of the second-order algebraic equation obtained by omitting the time derivative of n_0 in Eq. (9). It is given by

$$n_0(I_0) = \frac{N_a}{2}\left[-(1 + a(I_0)) + \sqrt{(1 + a(I_0))^2 + 4a(I_0)\left(\frac{N_d}{N_a} - 1\right)}\right] \tag{10}$$

with $a(I_0) = \frac{sI_0 + \beta}{\gamma_r N_a}$ (11)

The photoconductivity is obtained by replacing n_0 by the expression (10), then

$$\sigma_{ph}(I_0) = e\mu\frac{N_a}{2}\left[-(1 + a(I_0)) + \sqrt{(1 + a(I_0))^2 + 4a(I_0)\left(\frac{N_d}{N_a} - 1\right)}\right] \tag{12}$$

We reported in Fig. 2, the variation of the photoconductivity as a function of the average illumination intensity I_0, calculated by using the equation (12) and parameters of materials derived from the work of Amrhein.[4] This figure indicates that the photoconductivity depends sublinearly on the illumination intensity, which is a direct consequence of the variation of the trap density due to the photoexcitation of free charge carriers and leads to saturation effects for high intensities.

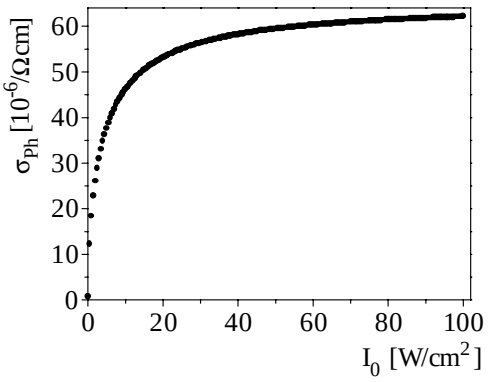

Fig. 2. Variation of the photoconductivity as a function of the illumination intensity I_0.

IV. ABSORPTION COEFFICIENT

If we assume that the absorption of photons is completely due to photoexcitation of electrons from filled donor sites to the conduction band, we get an absorption coefficient [4]

$$\alpha = S_p(N_d - N_a - n_0) \tag{13}$$

where $h\nu$ is the photon energy and $S_p = h\nu s$ the photoexcitation cross section. For photon energies close to or above the band gap energy this assumption is not valid.

The absorption coefficient calculated, for an illumination intensity fixed to 50 mW/cm^2 is, $\alpha = 0.84cm^{-1}$, $\alpha = 0.80cm^{-1}$ and $\alpha = 0.65cm^{-1}$ for the three wavelength, $488\mu m$, $514.5\mu m$ and $632.8\mu m$ respectively.

V. STEADY STATE OF SPACE CHARGE FILED

The space charge filed can be calculated in absence and in the presence of an applied electric field.

A. Space charge field without applied field E_0

The space charge field is purely imaginary; it is given by [9]

$$E_{sc} = im\Phi E_q \frac{E_d}{(E_d + E_q)}, \tag{14}$$

with

$$E_d = -\frac{k_B T}{e} K_g, \tag{15}$$

$$E_q = -\frac{e}{\varepsilon\varepsilon_0 K_g} N(I_0), \tag{16}$$

$$N(I_0) = N_e(I_0) + n_0(I_0) \tag{17}$$

$$N_e(I_0) = \frac{N_a + n_0(I_0)}{N_d}\left(N_d - N_a - n_0(I_0)\right) \tag{18}$$

$$\Phi = \xi\left(1 + \frac{\beta}{sI_0}\right)^{-1} \text{ and } \quad \xi = \left(1 + \frac{n_0(I_0)}{N_e(I_0)}\right)^{-1}. \tag{19}$$

The Eqs. (14-19) illucidate several interesting properties of the photorefractive effect. First, the space charge field or the two-beam coupling gain coefficient, respectively, is diminished by the intensity dependent factor ξ, which can take values between zero and one. Second, the charge limited field E_q is the sum of the trap density limited field, which is proportional to the effective trap density N, and a free carrier density limited field, which is proportional to the free electron density N_0. Therefore, the density N, usually determined from the fringe spacing dependence of the gain coefficient and identified with N_e, differs from the density N_e by a factor η, which can take values between one and N_d/N_a and may thus differ appreciably from unity. Third, the density N changes with the intensity. Therefore, the grating vector dependence of the space charge field as well as the gain coefficient change for different writing intensities.

The formula (14) show that the space charge field is phase shifted by $\frac{\pi}{2}$ with respect to the interference pattern.

On the Fig. 3, variation as a function of fringe spacing, of space charge field is represented for different light intensities. This field happens by a maximum for a fringe spacing of $2.4\mu m$, and then he decreases when the fringe spacing become of more in big addition, it is very important for the small light intensities.

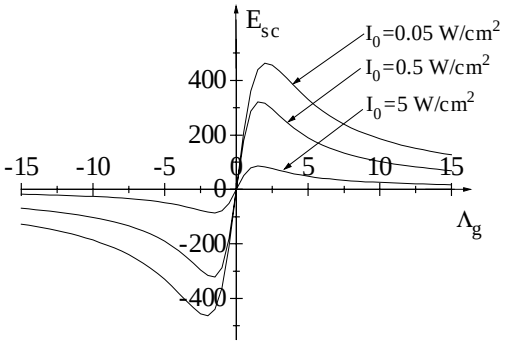

FIG. 3. Variation of space charge field as a function of fringe spacing for different light intensities I_0.

B. The exponential gain

In two beams coupling, two coherent waves interfere in a crystal under an angle 2θ and may exchange energy in the region where the beams overlap. Commonly the beams are plane waves with polarization either perpendicular or parallel to the plane of incidence. In $KNbO_3$ the grating vector of the phase grating is chosen parallel to the polar z axis and the interacting waves are polarized parallel to the plane of incidence. In this case the electro-optic coefficient r_{33} is responsible for beam coupling.

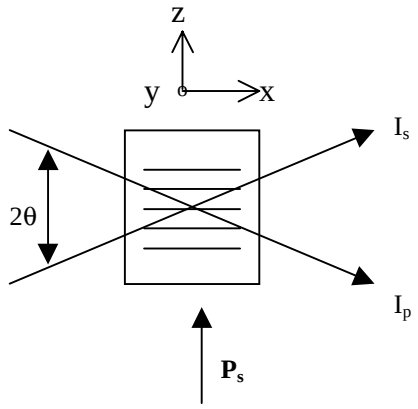

FIG. 4. Two beam coupling in $KNbO_3$ crystals with the grating vector parallel to the z axis and beam polarizations in the x-z plane.

The exponential gain can be calculated by solving coupled wave equations for the two beams, [7, 8, 10-13] The coupling strength is given by the refractive index variation, which is derived from the imaginary part of the space charge field and the electro-optic tensor r_{mn}, giving rise to a modulation of the dielectric tensor. For the situation described in Fig.4, when the incident beams are polarized perpendicular to the y-axis the exponential gain is given by [14]

$$\Gamma = \frac{2\pi\cos\theta}{\lambda}\frac{n_3^{\,4} r_{33} - n_1^{\,4} r_{13}\tan^2\theta}{n(\theta)}\frac{\mathrm{Im}(E_{sc})}{m}\cos(2\theta) \qquad (20)$$

$$\text{with} \quad n(\theta) = \frac{n_1 n_3}{\sqrt{n_1^{\,2}\cos^2\theta + n_3^{\,2}\sin^2\theta}} \qquad (21)$$

In Fig. 5, Variation for a light intensity I_0 = 70 mW/cm^2 and tree different wavelength, of the exponential gain as a function of fringe spacing, is represented by using equations (14) and (20), the electro-optiqc coefficients r_{13} = 28 pm/V, r_{33} = 64 pm/V [15, 16] and the refractive indices regrouped in Table I.

The exponential gain has the same appearance for the three wavelengths, it passes by a maximum for a fringe spacing of $2\mu m$, and then it decreases when the fringe spacing increases. It is very important for the small wavelength.

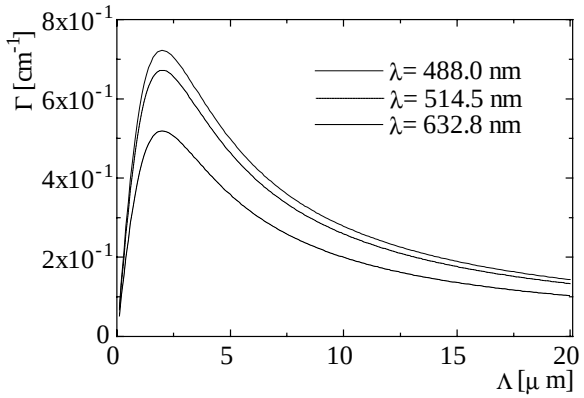

FIG. 5. The coupling gain Γ as a function of fringe spacing Λ for a light intensity I_0 = 70 mW/cm^2 and tree different wavelength.

By using the same equations, we also calculated the exponential gain (Fig. 6) for a small modulation m = 0.06, with varying the light intensity and in fixing the fringes spacing to 1.14 μm. The exponential gain increases very quickly in order to reach the saturation effect.

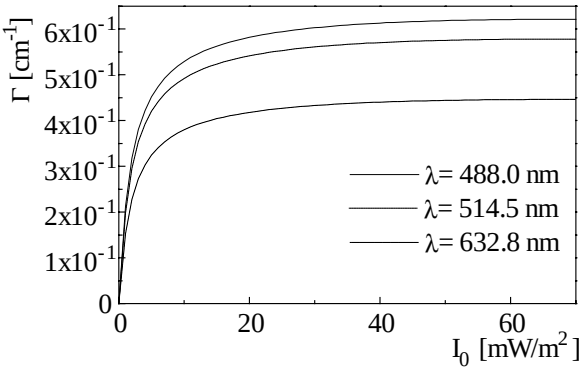

FIG. 6. The coupling gain Γ as a function of the light intensity I_0 (modulation m = 0.06) for a fringe spacing Λ=1.14 μm and tree different wavelength.

Table I. Refractive indices of $KNbO_3$ at room temperature for selected wavelengths λ.

	λ= 488.0 nm	λ= 514.5 nm	λ= 632.8 nm
n_1	2.3527	2.3334	2.2801
n_2	2.4187	2.3947	2.3269
n_3	2.2274	2.2116	2.1687

C. Space charge field with an applied electric field E_0

The application of an electric field along the grating vector K_g influences the charge transport during grating build-up, leads to an increased space charge field and introduces an additional shift of the phase grating with respect to the interference pattern that causes the grating. In this case the space charge field has two components, a real component and an imaginary. They are given by[9]

$$Re(E_{sc}) = -m\Phi E_q \frac{E_d E_0}{(E_d + E_q)^2 + E_0^2} \tag{22}$$

$$\mathrm{Im}(E_{sc}) = m\Phi E_q \frac{E_d (E_d + E_q) + E_0^2}{(E_d + E_q)^2 + E_0^2} \tag{23}$$

In the presence of an applied field, the exponential gain is proportional to the imaginary part of space charge field. Its formula is given by the insertion of the equation (23) into the equation (20).

Looking at the electric field dependence of Γ, Fig. 7 shows the two wave mixing gain Γ as a function of E_0 for different light intensities and a grating spacing of Λ=1 μm, whereas in Fig. 8 the grating spacing is Λ=10 μm. It is found that the gain saturates at a field of 1.5 kV/cm for a grating spacing of 1 μm. In this case E_{sc} is given by the trap limited field E_q (eq. 10). Since E_q is proportional to Λ, the gain can be larger for larger grating spacing Fig. 5.

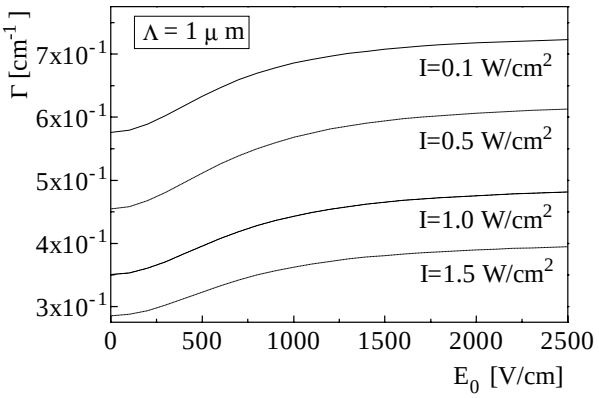

FIG. 7. The coupling gain Γ as a function of the applied electric field E_0 for a grating spacing Λ=1 μm and four different light intensities I_0.

When an electric field is applied, the phase shift ϕ between light and refractive index pattern, vary of its initial value $\frac{\pi}{2}$ that it has if the applied field is void. It is given by

$$\tan\phi = \frac{E_d}{E_0} + \frac{E_0}{E_q} + \frac{E_d^2}{E_q E_0} \tag{24}$$

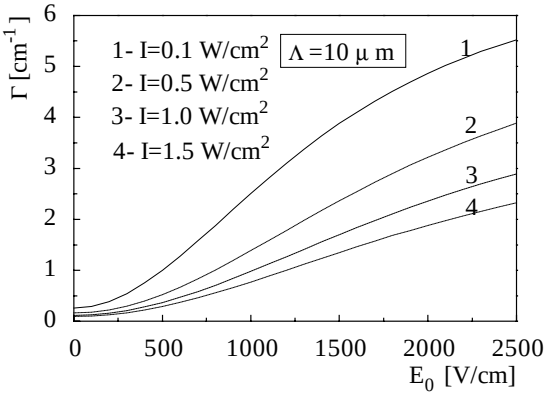

FIG. 8. The coupling gain Γ as a function of the applied electric field E_0 for a grating spacing Λ=10 μm and four different light intensities I_0.

The origin of the phases is being the value for which the illumination is maximum. The phase ϕ decreases continuously from $\frac{\pi}{2}$ to a minimum when E_0 increasing from 0 to $\sqrt{E_q E_d + E_d^2}$, but if E_0 becoming enough larger than E_q, then the phase shift increasing to $\frac{\pi}{2}$.

The variation of the phase shift between light and refractive index, as function of the applied electric field for different intensities is reported in Fig.9.

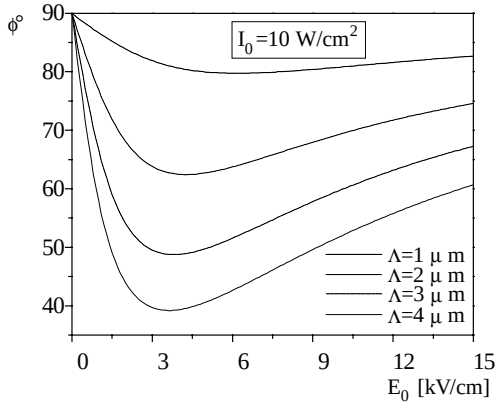

Fig. 9 Phase shift ϕ between light and refractive index as a function of an applied electric field E_0 for different fringe spacing Λ.

VI. DISCUSSION AND CONCLUSION

In the case of the ferroelectric compounds $KNbO_3$, mainly iron impurities are assumed to play the role of donor and trapping centers. The iron ions are introduced in the crystal at the sites of the Nb^{5+} ions. Since the

preferred valence states of iron are Fe^{3+} (acting as electron trap and Fe^{2+} acting as electron donor, there must be a charge compensation mechanism, which is attributed to oxygen vacancies. The oxygen vacancies may be located in the direct neighborhood of the iron ion. The concentration of iron, introduces voluntarily or not, has an influence on the different parameters physical like that the concentration of impurity levels, the photoexcitation cross section and the recombination coefficient. The increase of the concentration of iron in these compounds is accompanied by the increase of the oxygen vacancies, this who renders difficult the determination of these physical parameters.

The calculated results show that the evolution, in function of light intensity, of the photoconductivity and the exponential gain has the same behavior. These result also shows that the saturation of the exponential gain is obtained for a field applied in the order 1.5 kV/cm^2 when the fringe spacing is 1μm, on the other hand, it increases very fast if the fringe spacing is $10\mu m$. More if an external field is applied, the phase between light and refractive index, distances of its initial value $\frac{\pi}{2}$ (if no electric field is applied) more and more when the fringe spacing increases. This phase pass by a minimum for an applied field in the order of 3.6 kV/cm^2, then it increases in function of the applied field to reach its initial value for an external field very great.

The different obtained results can help to the comprehension of photorefractive mechanisms in general and are therefore interesting in the framework of the nonlinear-optic.

Acknowledgment

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