Structural distributions of induced electrostatic field upon volumetric all-optical poling

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In work the calculations of structural distributions of induced electrostatic field upon volumetric all-optical poling have been made. There are some corresponding experimental results which coincide with calculations. As known, the all-optical poling process is a result of the phase-matched nonlinear interaction ω + ω - 2ω → 0 that can leads to appearance of the non-zero space-periodical photocurrent $\mathbf{J} = \mathbf{e}_J A_1^2 A_2 \cos(\Delta\mathbf{k}\mathbf{r})$ (1) by means of coherent photo-galvanic (CPG) effect with accumula-- tion of the periodic electrostatic field $\mathbf{E}_0(\mathbf{r},t) = I_1\sqrt{I_2}\mathbf{A}_0(\mathbf{r})\cos(\Delta\mathbf{k}\mathbf{r})\left[1-\exp(-4\pi\sigma t/\varepsilon)\right]/\sigma(I_1,I_2)$ (2) . Here $\sigma(I_1,I_2)$ is the effective conductivity which depends on intensities of light harmonics, $\Delta\mathbf{k} = 2\mathbf{k}_1 - \mathbf{k}_2$, ε-permittivity. Separation of charges by means of coherent photocurrent (1) results in formation of the spatially periodic electrostatic field (2) inside sample. Let consider that the Gaussian light beams of the basic and doubled frequencies during the all-optical poling process of isotropic medium are intersected at angle α inside volume of sample, see setup in Fig. 1. We show here that the geometry of the intersecting Gaussian beams with changing their polarizations gives the different variations of properties of induced field lattices. At the small change of angle α, the angle θ between the direction of the basic light beam and the induced lattice planes and also the period of the induced lattice can be varied in a very large interval of their values. Indeed, at the change of angle α in interval from 0 to $\pi/2$, the period of lattice changes from the hundredth parts of the wavelength to the value $\lambda_2/\sqrt{2}$. The angle θ can be received from condition $\Delta\mathbf{k} = 2\mathbf{k}_1 - \mathbf{k}_2$ and it equal

$$\sin^2\theta = (1-\tfrac{n_2}{n_1}\cos\alpha)^2/(1-\tfrac{2n_2}{n_1}\cos\alpha+\tfrac{n_2^2}{n_1^2}),\quad \delta n=(n_2-n_1)/n_1 \ll 1 \qquad (3)$$

where $n_{1,2}$ are the refractive indices for light beams of the basic and doubled frequencies, correspondingly. As can see, only in region of small angles α, the sharp dependence $\theta(\alpha)$ exists. So, at variation of $\cos\alpha$ from 1 to $1+\delta n$ the angle θ is changed from $\pi/2$ to very small value $2\delta n$, and then for $\cos\alpha > 1+\delta n$ the slow monotonous growth of angle θ is observed. So, the more experimental interest has the region of small optical poling angles α and we will concentrate the attention below on such case.

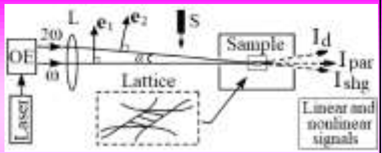

Fig. 1. Setup for volumetric all-optical poling

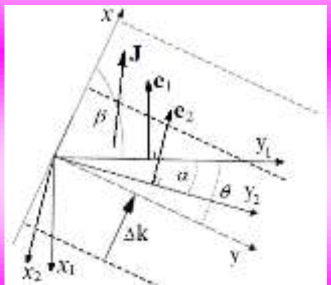

Fig. 2. The light beams interaction.

By small angles of α the CPG-current has direction which is practically parallel to vector $\Delta\mathbf{k}$ and $\sin\theta \approx 1-[\sin^2(\alpha/2)+\delta n\cos^2(\alpha/2)]/2,\quad |\delta n| \ll (\alpha/2) \ll 1$ (4).
For solution we choose the coordinate system which is related with induced field lattice (see Fig. 2). The light beams of basic and doubled frequencies in all-optical poling propagate along directions y_1 and y_2, accordingly, the point of intersection is the origin of coordinates and it is the center of the beam's waists. There are formulas

$$y_1 = -x\sin\theta + y\cos\theta,\quad y_2 = -x\sin(\alpha-\theta)+y\cos(\alpha-\theta),$$
$$x_1 = x\cos\theta + y\sin\theta,\quad x_2 = x\cos(\alpha-\theta)+y\sin(\alpha-\theta) \qquad (5)$$

Expression for CPG-current in coordinate system of induced lattice has view

$$\mathbf{J}(\mathbf{r}) = \boldsymbol{e}_j A_1^2 A_2 \exp(-(ax^2+by^2+cxy+dz^2))\cos(\eta x),\quad \eta = \Delta k a_{ef}, \qquad (6)$$
$$a=\cos^2(\theta-\alpha)-2a_{ef}^2 a_1^{-2}\sin(2\theta-\alpha)\sin\alpha,\quad b=\sin^2(\theta-\alpha)+2a_{ef}^2 a_1^{-2}\sin(2\theta-\alpha)\sin\alpha,$$
$$c=\sin 2(\theta-\alpha)+4a_{ef}^2 a_1^{-2}\sin(2\theta-\alpha)\cos\alpha,\quad d=1/(2a_{ef}^2),\quad a_{ef}^2 = a_2^2 a_1^2/(2a_2^2+a_1^2)$$

The a_1 and a_2 are radii of incident laser beams in waist ($2a_2^2 \approx a_1^2$). Inside con-cerned interval angles θ the condition $b \ll c \ll a_1$ is fulfilled and non-zero CPG-currents are $\mathbf{j}_x = c_1\cos\alpha\cos\theta + c_2\sin\alpha\sin\theta$, $\mathbf{j}_y = c_1\sin\theta\cos\alpha + c_2\cos\theta\sin\alpha$, and their ratio is $s_y = \mathbf{j}_y/\mathbf{j}_x = c_1\tan\theta + c_2\tan\alpha \ll 1$, $s_y b/c$ (c_1 and c_2 are the photo-galvanic constants of an isotropic medium).

The equation for the potential $\Phi(\mathbf{r})$ in the coordinate system of the induced lattice has the view

$$\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}+\frac{\partial^2}{\partial z^2}\right)\Phi(\mathbf{r}) = -j_x I(z)\left(s_y\frac{\partial}{\partial y}+\frac{\partial}{\partial x}\right)I(x,y),\quad I(x,y)=\exp\left(-\frac{1}{2}(ax^2+by^2+cxy)\right)\cos\eta x,\quad I(z)=\exp\left(-\frac{z^2}{2}\right),\quad \eta=\Delta k a_{ef} \gg 1 \qquad (7)$$

For solution we use the Fourier transformation on the coordinates (x,y) and then we integrate by z and carry out the reverse Fourier transformation. In resulting expressions, we neglect the values which are ~1 in comparison with values $\sim\eta$ and also we neglect the small values $\sin\theta$ and $\sin\alpha$. Note, that, by carrying out the reverse Fourier transformation, we take into consideration only the small regions of the values $k_y=0$ and $k_y=\pm\eta$ which give the basic contribution to the integral. The solution for potential is

$$\Phi(x,y,z) = -\frac{j_x}{2}W(\eta,z)W(x,y)\left[\left(2ax+(2s_y b+c)y\right)\cos x\eta + \eta\sin x\eta\right],\quad W(x,y)=\exp\left[-\frac{1}{2}(x^2a+y^2b+yxc)\right],$$
$$W(\eta,z) = -\frac{\sqrt{\pi}}{2\sqrt{2}\eta}\exp\left(\frac{\eta^2}{2}\right)\left(\mathrm{erfc}\left(\left(\frac{\eta+z}{\sqrt{2}}\right)\right)\exp\eta z + \mathrm{erfc}\left(\left(\frac{\eta-z}{\sqrt{2}}\right)\right)\exp(-\eta z)\right) \qquad (8)$$

For $\eta \gg 1$, that corresponds to the experimental parameters, the expressions for the corresponding components of photo-induced field have the view

$$E_x = -\frac{j_x}{4}W(x,y,z)\sqrt{\left((2ax^2-4yxac-y^2c^2)\frac{1}{2\eta^2}+1\right)^2+\frac{(2xa+yc)^2}{\eta^2}}\cos(x\eta+\varphi_x(x,y)),$$
$$E_y = \frac{j_x}{4\eta}(cx+2by)W(x,y,z)\sin x\eta,\quad E_z = j_x\frac{z}{2\eta}W(x,y,z)\sqrt{1+\eta^{-2}\left(2ax+y(2s_y|b|+|c|)\right)^2}\sin(x\eta+\varphi_z(x,y)),$$
$$W(x,y,z)=\exp\left(-\frac{1}{2}\left(x^2|a|+y^2|b|+yx|c|+z^2\right)\right),\quad \arctan\varphi_x(x,y)=\frac{2xa+yc}{\eta},\quad \arctan\varphi_z(x,y)=\frac{2ax+y(2s_y b+c)}{2\eta}$$

As can see in obtained formulas, the amplitude of the component E_x is not depend practically on η. It seems that the optical piling in this geometry can be most effective and there is the possibility in this case to creating the structures with homogeneous distribution inside all photo-induced area. In contrast, both of other components of the induced field (E_y and E_z) decrease as η^{-1} and for this case the creation of the homogeneous nonlinear-optical micro-periodic structure with big size is not be realized.

In other case if the polarizations of both light beams are directed along axis Z then $\mathrm{div}\mathbf{J}(\mathbf{r}) = -j_z I(x,y)\frac{d}{dz}I(z)$ (10)
and potential is determined by the expression (11)

$$\Phi(x,y,z) = -j_z W_2(\eta,z)\cos(x\eta)\exp\left(-\frac{1}{2}\left(x^2|a|+y^2|b|+yx|c|\right)\right)\cos(x\eta),$$
$$W_2(\eta,z)=\frac{\eta\sqrt{\pi}}{2\sqrt{2}}\exp\left(\frac{\eta^2}{2}\right)\left(-\mathrm{erfc}\left(\frac{\eta+z}{\sqrt{2}}\right)\exp\eta z+\mathrm{erfc}\left(\frac{\eta-z}{\sqrt{2}}\right)\exp(-\eta z)\right)$$

and the field components have the view

$$E_x = -\frac{j_z}{\eta}zW(x,y,z)(\sin\eta x),\quad E_z = j_z\left(\frac{1-z^2}{\eta^2}\right)W(x,y,z)\cos\eta x \qquad (12)$$

In this case of the optical poling geometry the induced electric field component E_x, which is orthogonal to the planes of the induced lattice, decreases as $\sim\eta^{-1}$, and the electric field component E_z, which is perpendicular to the beam axis, decreases more sharply as $\sim\eta^{-2}$. And this variant, correspondingly, is not also perspective for the creation of the structures with sufficiently big sizes.

So, it means that the efficiency of the volumetric optical poling of an isotropic medium depends fundamentally on the configuration of the CPG-current as well as on the relation η of the value of the beam envelope to the period of the induced field lattice during the optical poling of a medium. The various components of induced electric field have different dependences: so that may be the situation where it not depend on η, where it decreases as $\sim\eta^{-1}$, and where it decreases more sharply as $\sim\eta^{-2}$. The most effective optical poling in volumetric material can be observed only in the special used geometry of the interaction of intersecting Gaussian beams. The performed initial experiment in silicate glass by scanning of the form of the induced lattice, created with use the special geometry is in agreement with theoretical prediction. So, the calculated results can be useful for various estimations and in realization of other experiments in future.

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