Spatial Rectification of the Electric Field of Space Charge Waves

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A new phenomenon associated with a nonlinear interaction of optically excited space charge waves has been discovered in photorefractive crystals of the sillenite family. This interaction provides a spatial rectification of the electric field of the space charge waves and leads to a temporally oscillating, but spatially homogeneous electric field of the order of 50–60 V/cm which is detected by a probe beam via the electro-optic effect. Taking into account that the displacement current plays an important role, we demonstrate a very good agreement between the experimental data and the suggested theoretical model.

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In semi-insulating materials like photorefractive crystals space charge waves exist [1–3]. They can be regarded as quasiparticles in solids and are of great interest not only in the case of photorefractive materials but also in the case of many semiconductors. Space charge waves determine the dynamic behavior of the space charge distribution in the crystal and can be excited optically by various methods. A powerful technique is to use an interference pattern created by two coherent laser beams which is spatially oscillating with a small amplitude around an average position. Then, if the frequency of oscillation and the grating spacing of the interference pattern coincide with the frequency and the wavelength of the corresponding space charge wave, a resonance excitation of the space charge wave occurs.

In studying excitations of space charge waves two different approaches can be considered: linear and nonlinear ones. In the linear approach only contributions proportional to the contrast ratio m of the interference pattern are taken into account within the buildup and decay of space charge gratings, whereas in the nonlinear approach contributions that are proportional to m^2 become of importance. Among the nonlinear effects the appearance of higher spatial harmonics of the static grating [4] and the excitation of subharmonics and higher harmonics of space charge waves [5–7] are of special importance.

In this paper we describe a new nonlinear phenomenon which is—in our opinion—associated with a spatial rectification of the periodic space charge electric field due to an interaction between running and standing components of the space charge grating. Such an interaction leads to a space charge electric field $E_{\mathrm{sc},h}$ that is oscillating in time but homogeneous in space (homogeneous compared to distances of the order of the grating spacing of about 50 μm). This oscillating field appears although a constant electric field is applied to the crystal and it is completely determined by the dynamics of the space charge waves. In our experiments we register the oscillating field via the electro-optic effect which modulates the polarization state of a probe laser beam propagating through the crystal. The occurrence of the observed effect is not expected because the traditional theoretical approach shows that a temporally oscillating but spatially homogeneous component of the space charge field is prohibited by the boundary conditions [8,9]. The investigation of the effect of a spatial rectification of the electric field is useful not only for a better understanding of the nature of space charge waves, which are a rather new subject in studying solids, but also for practical applications, for instance, for the detection of weakly phase-modulated laser beams, for remote laser testing such as laser vibrosonic diagnostics, etc.

For recording holographic gratings a frequency-doubled Nd:YAG laser (wavelength $\lambda = 532$ nm, output power 100 mW) is used. Because the phase of one recording beam is modulated sinusoidally with an amplitude Θ and a frequency Ω by an electro-optic modulator, the interference pattern spatially oscillates around an average position. A HeNe laser ($\lambda = 633$ nm, output power 1 mW) is used for probing the crystal, and the polarization state of the transmitted probe beam is analyzed with the help of a polarizer, a photoreceiver, and a lock-in amplifier. Two crystals are used for experiments. One is a $Bi_{12}SiO_{20}$ (BSO) sample of dimensions $4 \times 3 \times 2.5$ mm^3 and the second is a $Bi_{12}GeO_{20}$ (BGO) crystal of the same size. The long edges of both crystals are oriented along the crystallographic [001] axis. The light beams are incident onto the (110) plane, and an external electric field is applied along the [001] direction which is also the direction of the grating wave vector. The probe beam (diameter approximately 0.5 mm) is positioned at the central part of the crystals. Measurements are performed for different input polarizations of the probe beam and for different orientations of the output polarizer. The obtained results qualitatively coincide for both crystals, and, therefore, the presented data for one crystal are qualitatively valid for the other crystal as well.

Figure 1 shows the dependence of the output signal U_{out} on the frequency $\Omega/2\pi$ of phase modulation for the BGO sample. The dependence shown has a resonance character

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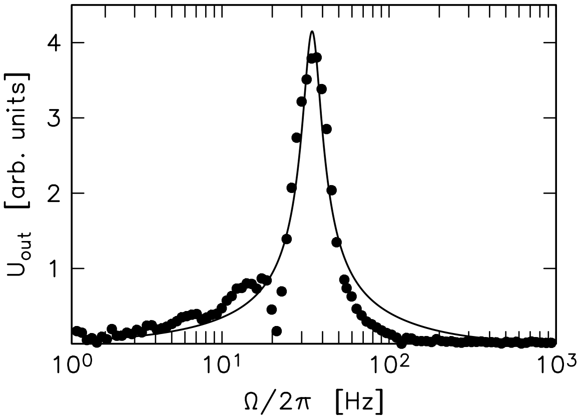

FIG. 1. Dependence of the output signal U_{out} on the frequency $\Omega/2\pi$ of phase modulation for the BGO sample. The total intensity of the recording beams is $I_0 = 60$ mW/cm^2 with a contrast ratio $m = 0.23$, the intensity of the probe beam is $I_p = 12$ mW/cm^2, the applied electric field is $E_0 = 7.5$ kV/cm, and the grating spacing is $\Lambda = 2\pi/K = 50$ μm. The solid line shows the theoretical frequency dependence according to Eq. (5).

similar to that observed for the resonance excitation of space charge waves published in [3]. The resonance frequency Ω_r depends on the electric field E_0 and is shifted to lower frequencies with increasing E_0. The input polarization of the probe beam is oriented along the $[1\bar{1}0]$ direction and the position of the output polarizer is at 56° with respect to the $[001]$ direction. In Fig. 2 the dependence of the maximum output signal $U_{\text{out}}(\Omega_r)$ is plotted versus the applied electric field E_0 (lower curve) and versus the contrast ratio m (upper curve). A proportionality between $U_{\text{out}}(\Omega_r)$ and E_0^3 is observed and for small values of m, $U_{\text{out}}(\Omega_r)$ increases quadratically with m, as indicated by the solid lines. Figure 3 shows the dependence of $U_{\text{out}}(\Omega_r)$ on the angle α' between the $[001]$ direction and the orientation of the output polarizer for a fixed input polarization along $[1\bar{1}0]$ using a polar plot.

Because our model for the interpretation of the observed effect is based on the existence of an oscillating homogeneous space charge field, we have performed a further experiment to estimate experimentally the magnitude of this field. In addition to the constant applied electric field, we used an alternating electric field coupled into the electrical circuit by a transformer which is in series with our high voltage source. In this case we observe the usual electro-optic effect under an oscillating electric field, in the absence of any phase modulation of the laser beam by the electro-optic modulator. Then we tuned the amplitude of the alternating electric field to have the same output signal which was observed at the presence of phase modulation without an external alternating field. In other words, this experiment imitates the induced effect by an ordinary one and as a result we can estimate the amplitude of the oscillating space charge field for BGO as 50–60 V/cm for $E_0 = 7.5$ kV/cm, $m = 0.23$, $\Theta = 0.5$, and $\Lambda = 50$ μm.

To analyze theoretically our experimental results two problems have to be solved. The first one is the calculation of $E_{\text{sc},h}$, and the second one is the calculation of the influence of $E_{\text{sc},h}$ on the output signal. To solve the first problem we use the ordinary set of equations describing the formation of the space charge field, the so-called Kukhtarev equations [8]. In our case we can neglect for the sake of simplicity diffusion processes because our results are obtained for high values of grating spacing and

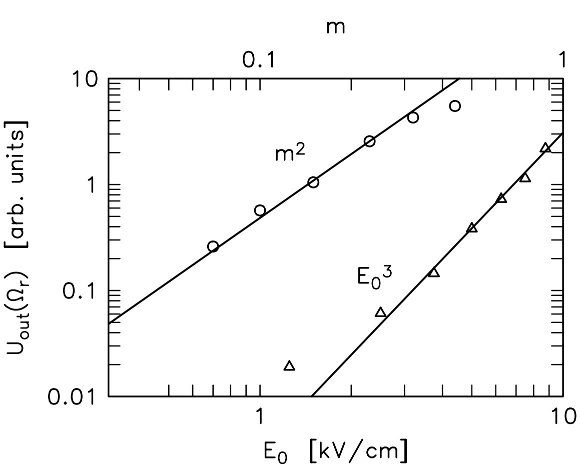

FIG. 2. Maximum output signal $U_{\text{out}}(\Omega_{\text{r}})$ versus applied electric field E_0 (lower curve) and contrast ratio m (upper curve) in a double-logarithmic plot for the BGO crystal. A proportionality between $U_{\text{out}}(\Omega_{\text{r}})$ and m^2 for small values of m and between $U_{\text{out}}(\Omega_{\text{r}})$ and E_0^3 can be observed as indicated by the solid lines. For the dependence on m the electric field is $E_0 = 7.5$ kV/cm and for the dependence on E_0 the contrast ratio is $m = 0.23$. The light intensities and the grating spacing are the same as in Fig. 1.

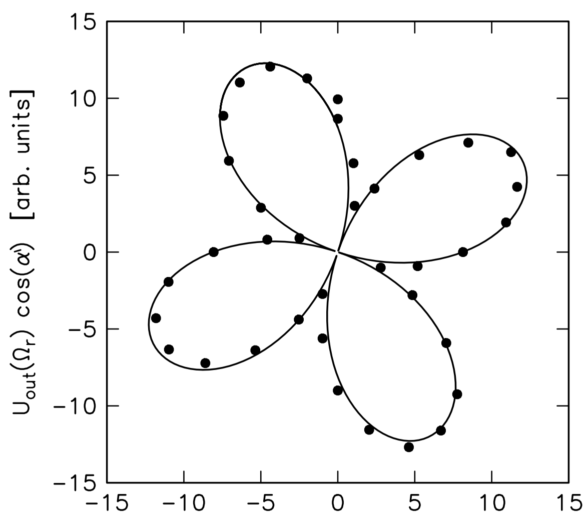

FIG. 3. Maximum output signal $U_{\text{out}}(\Omega_{\text{r}})$ as a function of the angle α' between the $[001]$ axis and the orientation of the output polarizer for the BSO sample using a polar plot. The applied field is $E_0 = 12.5$ kV/cm and the grating spacing is $\Lambda = 25$ μm. The solid curve is a fit of Eq. (7) to the experimental data.

high applied electric fields. It is assumed that the electrons are excited with a generation rate

$$g(x,t) = wI(x,t) = g_0[1 + m\cos(Kx + \Theta\cos\Omega t)], \quad (1)$$

where $I(x,t)$ is the light intensity, w is the product of the light absorption coefficient and the quantum efficiency for electron excitation, and $K = 2\pi/\Lambda$. A further condition has to be imposed to solve the problem. Usually the boundary condition is applied that the homogeneous part of the space charge field grating is equal to zero, i.e.,

$$\int_0^L E_{\mathrm{sc}}(x,t)\,dx = 0, \quad (2)$$

where L is the length of the crystal. The physical interpretation of this condition is clear: If the crystal is connected to an ideal voltage source, no additional voltage can exist between the crystal electrodes. However, in this approach our experimentally registered effect cannot exist. That is why we use another approach which takes into account an additional condition (instead of the boundary condition) relative to the total current density, namely,

$$J = j(x,t) + \varepsilon\varepsilon_0 \frac{\partial E_{\mathrm{sc}}(x,t)}{\partial t} = \mathrm{const.} \quad (3)$$

That means that the total current density J is constant—the crystal is connected to a current source rather than to a voltage source. Here $j(x,t) = e\mu n(x,t)E_{\mathrm{sc}}(x,t)$ denotes the electronic current density with the density $n(x,t)$ of optically excited carriers in the conduction band and their mobility μ. The term $\varepsilon\varepsilon_0\partial E_{\mathrm{sc}}(x,t)/\partial t$ is the displacement current density with the dielectric permittivity $\varepsilon\varepsilon_0$.

We are studying nonlinear effects proportional to m^2. The terms $\propto m^2$ appear in the theory because the expression for the electronic current density $j(x,t)$ contains the product of $n(x,t)$ and $E_{\mathrm{sc}}(x,t)$. For the recording technique used, both of these multipliers contain three terms. The first term is a static grating which is proportional to $m\exp(iKx)$. The other two terms are dynamic. One of them is proportional to $m\exp[i(Kx + \Omega t)]$, and the other one to $m\exp[i(-Kx + \Omega t)]$. These terms describe two forced waves propagating in opposite directions where the propagation direction of the latter wave coincides with that of the eigen space charge wave. In calculating the product $n(x,t)E_{\mathrm{sc}}(x,t)$, a term appears which is proportional to $m\exp(iKx)m\exp[i(-Kx + \Omega t)] = m^2\exp(i\Omega t)$. In other words, the product of two (in space) cosinusoidal functions results in a spatially homogeneous term. This is just the origin of the temporally oscillating but spatially homogeneous component of the current and electric field, i.e., the reason for the spatial rectification. At resonance this effect is a result of the resonance excitation of space charge waves and an interaction of these waves with the static space charge grating. Note that the photoinduced current in the presence of a sufficiently strong external electric field [9,10] is also caused by a rectification of the current density. However, no homogeneous space charge field (field rectification) was predicted in these papers because the boundary condition given in Eq. (2) was utilized.

Using the condition (3) the problem can be solved in the same manner as described in [7–9,11] for small m in a nonlinear approximation. Then we find that the oscillating homogeneous space charge field can be written as

$$E_{\mathrm{sc},h}(t) = E_0\Theta m^2 d\, f(\omega)\cos(\Omega t + \gamma), \quad (4)$$

with

$$f(\omega) = \left[\frac{\omega^2(1 + \omega^2/4)/(1 + \omega^2)}{1 + 2\omega^2(1 - d^2) + \omega^4(1 + d^2)^2}\right]^{1/2}, \quad (5)$$

and the phase shift γ. Here $d = KL_0$ and $\omega = \Omega\tau_{\mathrm{M}}$ hold with the Maxwell relaxation time τ_{M} and the drift length L_0.

Now we can estimate numerically the amplitude of $E_{\mathrm{sc},h}$. For typical experimental conditions ($E_0 = 7.5$ kV/cm, $d = 7$, $m = 0.23$, and $\Theta = 0.5$) we obtain $E_{\mathrm{sc},h} = 700$ V/cm, i.e., a value approximately 1 order of magnitude higher than the experimentally observed value. This difference is reasonable because the assumption of an ideal current source has to give a higher value of the oscillating field than the real situation with a nonideal current source. The origin of nonideality of the source can be a finite internal resistance, a possible load resistance in the circuit, or non-Ohmic electrode contacts (which is the most important factor in our case). We have performed calculations for a nonideal source (neither ideal voltage source nor ideal current source), and we have found that the fundamental results described above remain the same—only the absolute value of $E_{\mathrm{sc},h}$ depends on the degree of nonideality of the source. An agreement with the experimental data can be achieved if we use a proper value of the resistance of the contact area in the crystal. Since the calculations for the general case are rather cumbersome, they will be published elsewhere.

Now we comment on the characteristics of the electro-optic effect. Our crystals exhibit both the electro-optical effect and the natural optical activity. When an external electric field is applied to the crystal (along the [001] axis), the crystal becomes birefringent where one axis of the optical indicatrix is along $[1\bar{1}0]$ and the other one along [001]. Furthermore, the additional alternating field $E_{\mathrm{sc},h}$ modulates the initially static electro-optic effect. Then the standard calculation results in the output light intensity

$$I_{\mathrm{out},\Sigma} = \{[\cos\Delta\varphi\cos(\alpha - \alpha') - 2q/(1 + q^2)\sin\Delta\varphi\sin(\alpha - \alpha')]^2 + [(1 - q^2)/(1 + q^2)\sin\Delta\varphi\cos(\alpha - \alpha')]^2\}I_{\mathrm{in}}. \quad (6)$$

Here q is the ellipticity of the eigenmodes, α and α' are the angles between the $[001]$ direction and the input polarization of the probe beam and the orientation of the output polarizer, respectively, $\Delta\varphi = 1/2L\sqrt{\delta^2 + 4\varrho^2}$, with $\delta = \pi n^3 r_{41}[E_0 + E_{sc,h}(t)]/\lambda$, where ϱ denotes the rotatory power, n the refractive index, r_{41} the electro-optic coefficient, and λ the wavelength. For BGO and BSO we have $\varrho \approx 22-23$ deg/mm for $\lambda = 633$ nm [12] and $r_{41} = 3.2$ pm/V (BGO) and $r_{41} = 5.0$ pm/V (BSO) [13]. Finally, the dependence of the alternating part ($I_{out,\sim}$) of $I_{out,\Sigma}$ on α' can be written as

$$I_{out,\sim} = I(E)\,|\cos(2\alpha' - \Psi)|\,, \tag{7}$$

where $I(E)$ and Ψ depend in the general case on E_0 and α. For the experimental condition $\alpha = 90°$ and $\alpha' = 56°$, it can be shown with an uncertainty of less than 10% that

$$I_{out,\sim} \propto I(E) \approx 0.55 I_{in} \cos\Delta\varphi \, \frac{L(\pi n^3 r_{41})^2}{2\lambda^2 \varrho} E_0 E_{sc,h}\,. \tag{8}$$

The obtained formulas allow us to explain all experimental data. The solid curve in Fig. 1 is the function $f(\omega)$ for $d = 7.4$ and $\tau_M = 6.1 \times 10^{-4}$ s. One can see a good agreement between theory and experiment. The dependence of the maximum amplitude of the output signal $U_{out}(\Omega_r)$ on the contrast ratio m (Fig. 2) is quadratic as the theory predicts for small values of m, and the dependence of $U_{out}(\Omega_r)$ on E_0 (Fig. 2) is also in agreement with theory: Formula (4) contains E_0 directly, then it contains the parameter d which is proportional to E_0, and finally, $I_{out,\sim}$ is also proportional to E_0 for the used experimental configuration. Altogether we then have a proportionality to E_0^3 clearly observed experimentally. At last the angular dependence in Fig. 3 is in reasonable agreement with theory (solid curve) as well. We mention that even the absolute value of our registered output signal coincides with the one obtained from Eq. (8) very well (accuracy 30%) if we use for the calculation of $I(E)$ the value $E_{sc,h} = 50-60$ V/cm.

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