Dispersion law of photorefractive waves in sillenites

M. P. Petrov, V. V. Bryksin, and V. M. Petrov
Physical Technical Institute of the Russian Academy of Sciences, St. Petersburg 194021, Russia

S. Wevering* and E. Krätzig
Fachbereich Physik, Universität Osnabrück, Barbarastraße 7, D-49069 Osnabrück, Germany

(Received 23 March 1999)

We investigate the dispersion law (relationship between Ω_k and K) of photorefractive waves using a new experimental method based on the excitation of photorefractive waves by a spatially oscillating interference pattern and on the detection of the first non-Bragg diffraction order. The existence of a sharp resonancelike dependence of the output signal on the frequency of excitation allows us to analyze the dispersion law with high precision. $Bi_{12}TiO_{20}$ and $Bi_{12}GeO_{20}$ single crystals are used for the investigations, and the dependences of Ω_k on K, on external field, on index of modulation, and on light intensity are studied. The experimentally observed relation $\Omega_k \propto 1/K$ is in excellent agreement with theory. A contribution of nonlinear effects to the spectrum of photorefractive waves is discussed. [S1050-2947(99)00909-9]

PACS number(s): 42.65.Hw, 73.20.Mf, 42.40.Eq, 42.70.Nq

I. INTRODUCTION

In semiconductor crystals spatial time fluctuations of charge carriers (electrons and holes) can evolve to their equilibrium state either through a simple relaxation process or through dampened waves. In the last case the existence of propagating waves means the existence of corresponding eigenmodes of oscillations which also can be considered as quasiparticles in the material. One well known example of quasiparticles are plasmons — waves in an electron-hole plasma [1]. A relatively high concentration of carriers (10^{17}–10^{18} cm^{-3}) is usually necessary for the existence of plasmons in semiconductors at room temperature. In other cases, for instance for recombination waves [2,3], wave processes and even self-generation of waves can take place if the Maxwell relaxation time τ_M is rather short (less than the lifetime of carriers). However, as it was shown in Ref. [4], wave processes can exist even in semi-insulating crystals with a long Maxwell (dielectric) relaxation time (10^{-1}–10^{-4} s), a relatively low concentration of carriers (10^{12}–10^{14} cm^{-3}), and a low mobility. These are space charge waves or, in other words, trap charge exchanging waves. To our knowledge, the first theoretical analysis of these waves was presented in Ref. [4] and then experimental investigations by measuring impedance oscillations were reported in Ref. [5] for n-type germanium doped with gold.

A very favorable situation for investigations of space charge waves exists in photorefractive materials, such as $Bi_{12}SiO_{20}$ (BSO), $Bi_{12}GeO_{20}$ (BGO), and $Bi_{12}TiO_{20}$ (BTO) — crystals of the sillenite family. In this case an inhomogeneous periodical distribution of space charge can easily be formed by illuminating a sample by any periodical light intensity pattern, for instance, by an interference pattern formed by two coherent plane waves. Then evolution of space charge waves can be detected by observing diffraction of coherent light because in photorefractive crystals a periodical distribution of space charge causes an electric field grating and a corresponding refractive index grating through the linear electrooptic (Pockels) effect. The periodical grating of refractive index is actually a phase holographic grating providing light diffraction.

The theoretical analysis of recording and relaxation processes of holographic gratings in photorefractive crystals is usually based on a set of equations (Kukhtarev equations) which allow wave solutions [6,7]. In the case when the effect of self-diffraction of the incident light beams on the process of recording of the holographic grating is negligible (which means that two-wave mixing inside the crystal can be ignored), the theory [6,7] predicts the same results as given in Ref. [4] for the electric field grating dynamics. In particular, when the drift length of carriers is larger than the grating period, but the grating period is still large enough to neglect diffusion, the eigenmodes of the space charge electric field fluctuations are running waves of the type

$$E_{sc}(x,t) = E_{sc}(t=0)\exp\left(-\frac{t}{\tau_R}\right)\exp[i(\Omega_k t - Kx)], \quad (1)$$

where K is the length of the grating wave vector, Ω_k the frequency of the wave, and τ_R the relaxation time.

In accordance with Refs. [6,7] and taking into account the aboved mentioned conditions, the eigenfrequency Ω_k of the space charge wave and the grating wave vector K are connected by the dispersion law

$$\Omega_k = \frac{1}{\tau_M K L_0}, \quad (2)$$

where L_0 is the drift length of carriers and τ_M is the Maxwell relaxation time. Here it is assumed that $\Omega_k \tau \ll 1$, where τ is the lifetime of carriers, $L_0 K > 1$ and $L_D K < 1$, with the diffusion length L_D. Expression (2) can be written in the form [8,19]

*FAX: +49(0)541969-2670

$$\Omega_k = \frac{e}{\varepsilon\varepsilon_0 E_0}\frac{\alpha I_0}{\hbar\omega K}. \tag{3}$$

Here e is the electron charge, $\varepsilon\varepsilon_0$ the dielectric permittivity, α the absorption coefficient, I_0 the total light intensity, and $\hbar\omega$ the energy per photon.

The requirement $L_0K>1$ actually means that the relaxation time $\tau_R=\tau_M(L_0K)^2$ is larger than the time period of wave oscillations. In this case we deal with wave processes rather than with a simple relaxation process. The corresponding phase velocity of the propagating wave is

$$v_{\rm ph}=\frac{\Omega_k}{K}=\frac{1}{\tau_M K^2 L_0}. \tag{4}$$

To our knowledge, one of the first unambiguous experimental evidences of the existence of running waves in photorefractive crystals was obtained in Ref. [10] where recording was performed by a moving interference pattern in a thin BSO sample. Because of the small thickness of the sample an amplification of the grating due to two wave mixing could be ignored, but a resonant enhancement of the grating amplitude could be detected if the velocity of the interference pattern coincided with the velocity of the corresponding running grating. This enhancement was observed experimentally in Ref. [10]. Note that the first experiments with a moving interference pattern were reported in Ref. [11] and later the theory was described in Ref. [12]. However, in these papers the main attention was focused on amplification of the moving space charge grating through the two wave mixing mechanism. All these investigations were carried out in the linear regime of recording when a sinusoidal interference pattern results in a similar sinusoidal grating of the space charge and the electric field. A wide spectrum of nonlinear effects has been studied, for instance in Refs. [8,9,13] where a moving interference pattern provides an excitation of higher (or sub) spatial harmonics of the space charge gratings.

Among the main characteristics of space charge waves, one of the most interesting features is the dispersion law (2). The dispersion law is a fundamental relationship for any wave, but in our case it attracts special attention because of its unusual character. In our case Ω_k is inversely proportional to K. This is a very specific dependence which results in some unusual properties of these waves. In particular, a negative group velocity

$$v_g=\mathrm{d}\Omega_k/\mathrm{d}K=-1/(\tau_M K^2 L_0) \tag{5}$$

exists, which means that the phase and the energy of the wave packet move with the same speed in opposite directions. The relationship (2) also predicts a strong group velocity dispersion. The goal of this paper is to present experimental data for the dispersion relation of photorefractive waves in the most unambiguous situation (linear regime of recording, negligibly small diffusion length, and negligibly weak wave mixing processes).

The dispersion law contains three independent parameters. They can be selected as $\tau_M\propto I_0^{-1}$, $K=2\pi\nu$ (ν is the spatial frequency of the grating), and $L_0=\mu\tau E_0$ (here μ is the mobility and τ is the lifetime of carriers). In this paper we investigate experimentally the validity of relationships (2) and (3), respectively, by varying ν, E_0, and I_0.

A few words about the terminology. The majority of authors use the term ''space charge waves.'' In Ref. [14] it was proposed to call these waves ''spastrons'' because they can be regarded as quasiparticles as well. However, in photorefractive materials the light diffracts from a refractive index grating and in the general case the dynamics of this grating may not coincide with those of free space charge waves in nonphotorefractive materials because the processes of formation and propagation of space charge waves in photorefractive crystals include in the general case the processes of self-diffraction, two wave mixing, and enhancement of the propagating light beams [12,15,16]. Thus, a refractive index grating is the result of more complicated processes including space charge dynamics, light diffraction and two wave mixing. That is why we prefer to call these waves ''photorefractive waves'' or ''photorefractons'' in the case when we deal with photorefractive materials.

The most straightforward way to investigate the dynamics of photorefractive waves is to record a refractive index grating by a moving interference pattern and to analyze the light diffraction as a function of the velocity of motion and other parameters [17,18]. However, in this paper we use a simple, but very sensitive and efficient technique: the detection of non-Bragg orders diffracted from the grating that is recorded by a spatially oscillating interference pattern. The oscillations of the interference pattern occur when one of the recording beams is phase modulated with the frequency Ω and the amplitude Θ. As it was shown in Refs. [17,18], the intensity of the first non-Bragg diffraction order is described by the relationship

$$I_{\rm NB}=I_{\rm const}+\frac{2I_S\eta_{\rm NB}\Theta g d}{D(d)}\cos(\Omega t+\gamma), \tag{6}$$

where

$$D(d)=[1+2g^2(1-d^2)+g^4(1+d^2)^2]^{1/2}, \tag{7}$$

$$\gamma=-\arctan\left[\frac{g^2(1+d^2)-1}{2g}\right], \tag{8}$$

$$g=\Omega\tau_M, \tag{9}$$

$$d=KL_0. \tag{10}$$

The diffraction efficiency of a thin hologram for $\Theta=0$ is $\eta_{\rm NB}=(mE_0q)^2/4$. Here q is a coefficient depending on the electrooptic parameters of the material [19,20], $m=2\sqrt{I_RI_S}/(I_R+I_S)$ is the contrast ratio of the interference pattern, and I_R and I_S are the intensities of the recording beams. The assumptions $\Theta,m\ll 1$ were used to derive Eq. (6).

The dependence of $I_{\rm NB}$ on modulation frequency Ω shows a maximum at the resonance frequency Ω_r. The value of this frequency can be written as [20]

$$\Omega_r=\frac{1}{\tau_M(d^2+1)^{1/2}}. \tag{11}$$

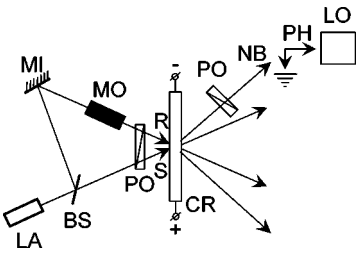

FIG. 1. Experimental setup showing the incident beams R and S and the beam NB diffracted from a thin hologram to the first non-Bragg order: LA: laser; BS: beam splitter; MI: mirror; MO: electrooptic modulator; PO: polarizer; CR: crystal with externally applied electric field; PH: photodiode; LO: lock-in amplifier.

As one can see, for the condition $d=KL_0>1$ we can neglect one compared to d^2 and relationship (11) coincides with the dispersion law (2). Thus, measurements of Ω_r as a function of L_0, K, and τ_M allow us to verify relationship (2). To neglect all diffusion processes and to observe the non-Bragg diffraction order, we take the value of $K=2\pi\nu$ in the interval $2\pi(10–130)$ mm^{-1} for which the diffusion field is less than 250 V/cm and so it can be neglected compared to the used external electric fields of 1–10 kV/cm.

II. EXPERIMENTAL RESULTS

Figure 1 shows the experimental setup. For the measurements the second harmonic of a Nd:YAG laser ($\lambda=532$ nm) is used (laser: Coherent DPSS, 532-400). The intensitiy of the light incident on the crystal surface is in the interval 25–400 mW/cm^2. The signal corresponding to the first non-Bragg diffraction order is detected. One of the incident beams is phase modulated by an electrooptic modulator and the amplitude of modulation is $\Theta=0.5$ rad. The signal is detected by a photodiode and a lock-in amplifier.

Thin plates (0.7 mm thick) of BTO and BGO single crystals are investigated. The used cut is (110) where the electric field and the grating wave vector are chosen to be parallel to the $[1\bar{1}0]$ axis. The polarization of the incident light is directed along [001]. This configuration provides diffraction with rotation of the polarization plane and prevents practically any effects of two beam coupling inside the crystals.

Figure 2(a) shows examples of the frequency dependence of I_{NB} for the BTO sample for two different values of spatial frequency ν, for $E_0=9.4$ kV/cm, $I_0=225$ mW/cm^2, and the contrast ratio $m=0.2$. The solid curves correspond to calculations using Eq. (3) and the values $d=1$ and 3.3, respectively, and $\tau_M=0.35$ ms. One can see that there is a quite good agreement between experiment and theory for $d=1$ except for the low-frequency region. For $d=3.3$ the theoretical curve is narrower than the experimental curve. The possible origin of this discrepancy will be discussed at the end of this paper. Figure 2(b) shows the dependence of the phase on the frequency for the same crystal and the same experimental conditions. One can see that the sign of the phase changes at Ω_r and that the slope of the phase dependence near Ω_r increases with increasing d which is consistent with Eq. (8). Note, that in accordance with Eq. (8), $\gamma\approx-(1+d^2)\Delta\Omega\tau_M$ near Ω_r. Here $\Delta\Omega$ is the magnitude of detuning. Using the last relationship it is possible to check the value of d obtained from other measurements.

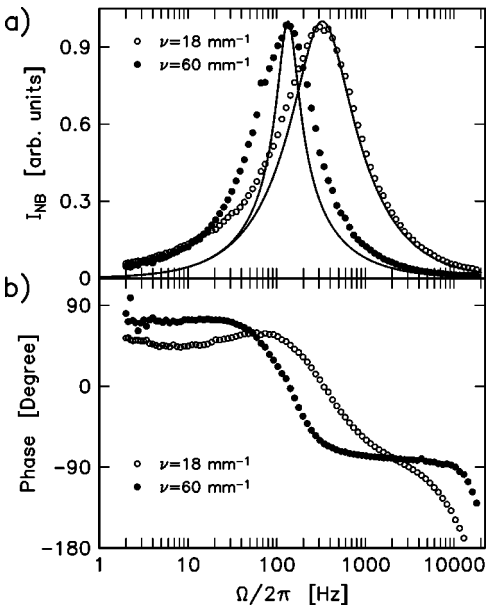

FIG. 2. Frequency dependence of the amplitude I_{NB} (a) and the phase (b) of the intensity diffracted from the BTO sample into the first non-Bragg order for two values of spatial frequency ν. The total light intensity is $I_0=225$ mW/cm^2 with a contrast ratio $m=0.2$ and the applied electric field is $E_0=9.4$ kV/cm. The solid lines correspond to calculations (see text).

Figure 3 shows the experimental dependence of Ω_r on ν for the BTO and the BGO sample for $m=0.2$ and $I_0=225$ mW/cm^2 and 340 mW/cm^2, respectively. The solid and dashed curves are theoretical dependences calculated with Eq. (11) where the fit parameters are $\mu\tau=9.3\times10^{-12}$ m^2/V and $\tau_M=3.5\times10^{-4}$ s for BTO and $\mu\tau\tau_M=6.5\times10^{-15}$ m^{2}s/V for BGO. Note that the data for the BGO sample can be described by $\Omega_r\propto1/\nu$ which means that in Eq. (11) one can be neglected compared to d^2 for the whole investigated range of spatial frequencies. Therefore only the product $\mu\tau\tau_M$ can be determined accurately for the BGO crystal. However, this product equals $\epsilon\epsilon_0\hbar\omega/(e\alpha I_0)$ according to Eq. (3) which means that it does not depend on $\mu\tau$ at all. Taking $\alpha=1.5$ cm^{-1} one obtains $\mu\tau\tau_M=3.6\times10^{-15}$ m^{2}s/V which is in a qualitative agreement with the

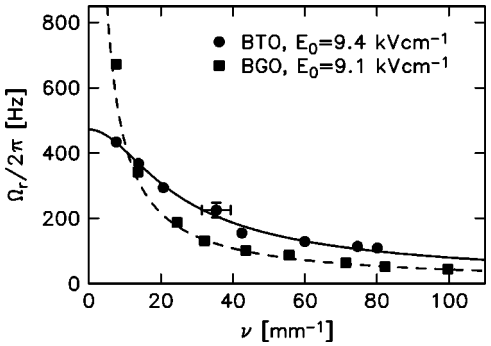

FIG. 3. Resonance frequency $\Omega_r/2\pi$ in dependence of the spatial frequency ν for the BTO and BGO sample. The solid and dashed curves are fits of the theoretical expression for the resonance frequency to the experimental data. The light intensity is $I_0=225$ mW/cm^2 for BTO and $I_0=340$ mW/cm^2 for BGO, and the contrast ratio is $m=0.2$ for both crystals.

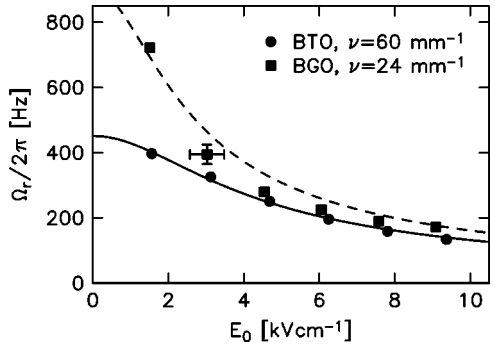

FIG. 4. Resonance frequency $\Omega_r/2\pi$ in dependence of the externally applied field E_0 for BTO and BGO. The solid and dashed curves are calculations using the same crystal parameters as in Fig. 3. Light intensity and contrast ratio are equal to those used in Fig. 3.

value obtained from the experimental data. From an estimation it follows that the mobility lifetime product is about $\mu\tau = 2-4\times10^{-11}$ m^2/V which means that the parameter d is about 6 to 12 for an electric field $E_0 = 9.1$ kV/cm^2 and a spatial frequency $\nu = 50$ mm^{-1}.

Figure 4 shows the experimental dependence of Ω_r on E_0 for both crystals, where the light intensity is $I_0 = 225$ mW/cm^2 for BTO and $I_0 = 340$ mW/cm^2 for BGO and the contrast ratio is $m = 0.2$. The solid and dashed curves are theoretical dependences calculated from Eq. (11) with the same set of fit parameters as in Fig. 3. The data presented in Figs. 3 and 4 show a very good agreement between theory and experiment. A reasonable agreement is also found if we analyze the dependence of Ω_r on the product KL_0.

Figure 5 shows the experimental dependence of Ω_r and of the maximum signal intensity $I_{\mathrm{NB}}(\Omega_r)$ on the total light intensity I_0 incident on the BTO crystal. The solid curves are shown for clarity. It can be seen that Ω_r grows linearly with increasing light intensity. This means that τ_M is determined mostly by the photoconductivity σ_{ph}, thus [9]

$$\tau_M^{-1} = \frac{\sigma_{\mathrm{ph}}}{\varepsilon\varepsilon_0} = \frac{e\alpha\mu\tau I_0}{\varepsilon\varepsilon_0\hbar\omega}. \tag{12}$$

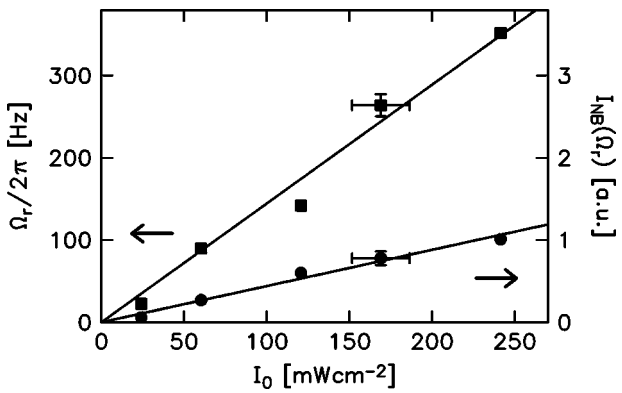

FIG. 5. Intensity dependence of the resonance frequency Ω_r and of the amplitude of the non-Bragg intensity $I_{\mathrm{NB}}(\Omega_r)$ at the resonance frequency for the BTO crystal. An electric field of $E_0 = 6.25$ kV/cm is applied and the contrast ratio is $m = 0.5$. The solid curves are shown for clarity.

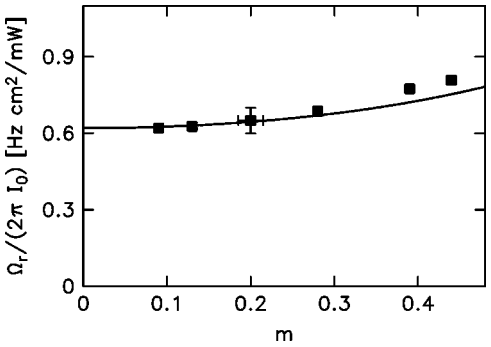

FIG. 6. Dependence of the resonance frequency normalized to the light intensity $\Omega_r/(2\pi I_0)$ on the contrast ratio m of the light interference pattern for BTO where an electric field $E_0 = 9.4$ kV/cm is applied. The solid curve is a calculation using the theoretical formula for this dependence and the crystal parameters obtained from the fits in Figs. 3 and 4.

The linear dependence of Ω_r on I_0 is in excellent agreement with Eq. (3) and with previously published data [17]. The linear dependence of the maximum signal intensity $I_{\mathrm{NB}}(\Omega_r)$ on I_0 is consistent with Eq. (6) because I_S is proportional to I_0 in our case. The obtained data allow us to conclude that formula (11) describes the experimental data quite well. Because of the identity between Eq. (2) and Eq. (11) for the condition $KL_0 = d > 1$, this means that the dispersion law (2) is also in good agreement with the experimental data.

We also investigated a BSO crystal. All data are in a reasonable agreement with the data published earlier Ref. [7] and quite similar to the results for BTO and BGO presented above.

III. DISCUSSION

The conclusion about the agreement between the theory and experiment was made on the basis of measurements of the resonance frequency. One of the serious advantages of this method is that the resonance frequency is only weakly sensitive to a nonlinearity of the recording process at small m. But nevertheless, the role of the nonlinearity of the recording process can be important in some specific cases. Therefore we will discuss this point in more detail.

Generally speaking, a number of mechanisms can contribute to nonlinear processes in photorefractive materials. Some of them, such as a nonlinearity of the photoconductivity in the dependence of light intensity or a nonlinearity of the current dependence on the applied electric field, are connected to specific material properties and very often they can be ignored.

On the other hand, the nonlinearity due to the finite value of the contrast ratio m is a fundamental intrinsic property of the mechanism of holographic recording in photorefractive materials. The origin of this nonlinearity is the influence of the arising space charge field on the recording process which is described in the Kukhtarev equations by the product of space charge field and density of photoinduced charge carriers.

This product is proportional to m^2 and usually it is ignored for small m. However, just this product produces a

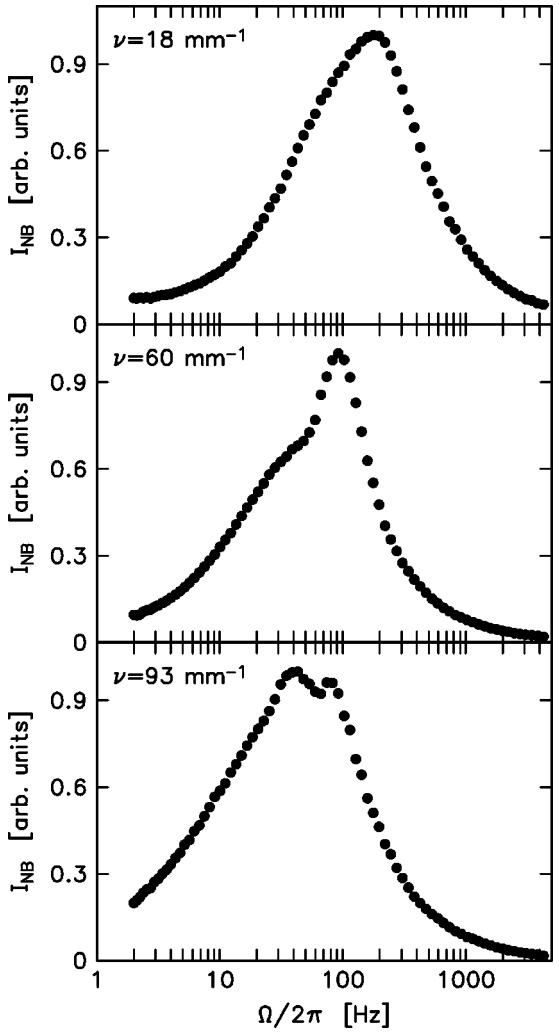

FIG. 7. Frequency dependence of the amplitude of the non-Bragg intensity for different values of spatial frequency ν for the BTO sample. The applied electric field is $E_0=9.4$ kV/cm and the light intensity is $I_0=250$ mW/cm^2 with a contrast ratio of $m=0.5$.

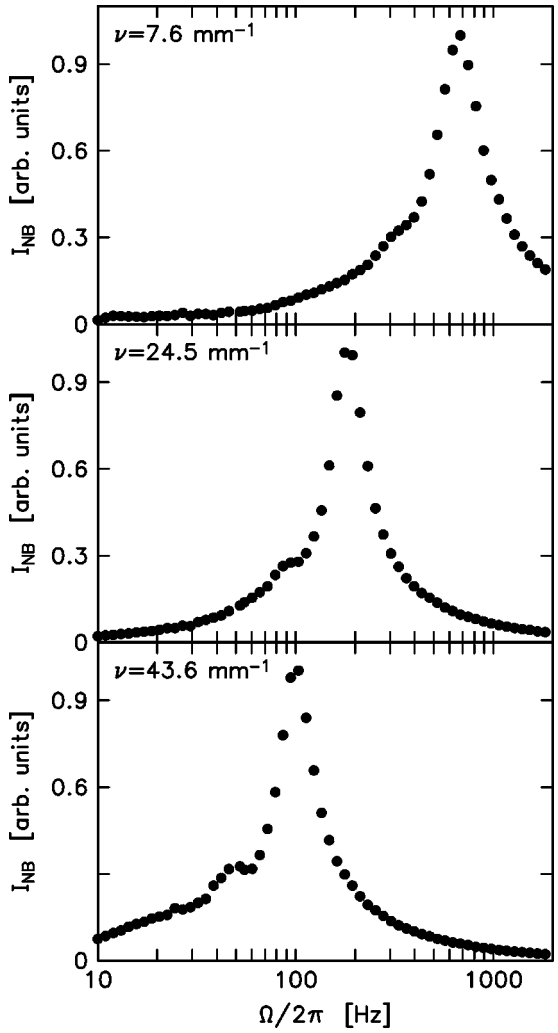

FIG. 8. Frequency dependence of the amplitude of the non-Bragg intensity for different values of spatial frequency ν for the BGO sample. The applied electric field is $E_0=9.1$ kV/cm and the light intensity is $I_0=340$ mW/cm^2 with a contrast ratio of $m=0.2$.

nonsinusoidal profile of the space charge field even if the interference pattern has an ideal sinusoidal shape. Therefore higher spatial harmonics of the fundamental electric field grating arise in this case. This means that during illumination of the crystal with an interference pattern with the wave number K we record a number of gratings with wave numbers $K_p=pK$ and amplitudes proportional to m^p, where p is an integer. Then, if the frequency of phase modulation Ω is changed, the corresponding photorefractive waves with resonance frequencies $\Omega_r^{(p)}=\Omega_r/p$ will be excited. From the first glance it seems that the registration of the corresponding pth resonance requires detection of the pth order diffraction. However, all oscillations of the electric field gratings are coupled through the mechanism of formation of the space charge field and as it was shown theoretically in Refs. [20,21], any (even the first order) diffraction peak can exhibit additional resonance maxima at the frequency $\Omega_r^{(p)}$.

Moreover, this nonlinearity results in a shift of the main resonance Ω_r with respect to the contrast ratio in accordance with the relationship

$$\Omega_r=\frac{1+m^2/2}{\tau_M[(KL_0)^2(1-m^2)+1]^{1/2}}. \tag{13}$$

Note that formula (13) was obtained for relatively small m and for the condition $(KL_0)^2(1-m^2)\gg 1$. So one should be careful in using it for $m\to 1$.

In this paper we study experimentally both effects, namely the appearance of additional maxima in the resonance curves for first order diffraction and the shift of the main resonance. Figure 6 shows the dependence of the resonance frequency normalized to light intensity on the contrast ratio m for BTO with an applied field of $E_0=9.1$ kV/cm. The normalization is necessary because if we change m in our experiments we also change the total light intensity I_0. The solid line is a calculation using Eq. (13) with the same crystal parameters as in the Figs. 3 and 4.

Figure 7 shows the frequency dependence of I_{NB} for different spatial frequencies at $m=0.5$ for BTO and Fig. 8 at $m=0.2$ for BGO. As one can see from both figures there are

some anomalies or even clear maxima at $\Omega=\Omega_r/2$. For other experimental conditions these anomalies can be stronger or weaker, but they can be found almost always for high values of m. We studied also the dependence of the phase of the detected signals on frequency and clear anomalies are found as well for frequencies near $\Omega_r/2$. This is in agreement with the model of nonlinear excitations of photorefractive waves with doubled wave vectors. Thus the experimental data shown above can be interpreted as an illustration of the role of the nonlinearity in the spectral properties of photorefractive waves. A more detailed comparison of the experimental results with theory is difficult because no exact analytical formula for the frequency dependence of I_{NB} for the nonlinear case is available.

It is worth to mention some other factors which can contribute to a deformation of the frequency dependence of I_{NB}. As one can see from Fig. 2 there exists a definite discrepancy between the calculated and the measured width of the resonance curve for $\nu=60\ \mathrm{mm}^{-1}$. To our opinion a reasonable explanation for this discrepancy is an inhomogeneous broadening due to a nonuniformity of the electric field inside the crystal. It is well known that such a nonuniformity appears rather often because of non-Ohmic contacts and due to a spatial inhomogeneous distribution of light intensity [7]. Furthermore, a inhomogeneity of light intensity leads to a Maxwell-relaxation time that varies over the crystal surface and thickness. This also results in a broadening of the frequency dependence of I_{NB}. Both of these sources of broadening are not in contradiction with our experimental results.

A next factor is a possible broadening at high values of d [22]. If d is high enough to have $\Theta md>1$, some deformation of the resonance curve may exist because the last condition means that the amplitude of the grating oscillations is higher than the applied electric field [18], which has definitely no physical meaning. However, more detailed experiments where Θ was changed to reduce the product Θmd reveal no indications that the width of the resonance peak depends on this product in our case.

Another factor is the contribution of second order diffraction of the beam with intensity I_R. This factor may be important for the case when I_R is higher than or comparable with I_S. However, in our experiments $I_R \ll I_S$ is valid even for $m=0.5$ and we do not expect a strong influence of this factor on our experimental results.

The considered origins of the observed shift of the position of the resonance curves and of their deformation are caused mainly by the nonlinearity of holographic recording. However, for the small value of m used in the experiments to verify the dispersion law, these effects can be neglected.

IV. CONCLUSIONS

In this paper the dispersion law (the dependence of Ω_k on K) for the photorefractive waves (or waves of space charge in photorefractive crystals) was investigated in detail. The technique of detecting the non-Bragg self-diffraction of phase-modulated laser beams was used. The experimental data concerning the position of the resonance peak which is associated with an excitation of photorefractive waves is used to check the theoretical expression of the dispersion law. An excellent agreement between theory and experiment is found if the necessary requirement for the linear regime of recording ($m \ll 1$, $\Theta \ll 1$) and the condition of the absence of two beam coupling are satisfied.

The contribution of nonlinearity in holographic recording to position and shape of the resonance peak is analyzed. It is found that the shift and deformation of the resonance curves observed experimentally at rather high values of modulation index can be explained (at least qualitatively) by the theory that describes nonlinear mechanisms of generation of photorefractive waves.

ACKNOWLEDGMENTS

Financial support by the Volkswagen-Stiftung (I/72919) and partially by the Russian Foundation for Fundamental Investigations (Grant No. 98-02-18254) is gratefully acknowledged.

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