

Resonant excitation of space-charge and photoconductivity waves in semiconductors with shallow energy levels

Mikhail A. Bryushinin and Igor A. Sokolov*

*A. F. Ioffe Physical Technical Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia
and Institute of Problems of Mechanical Engineering, Russian Academy of Sciences, 199178 St. Petersburg, Russia*

(Received 27 December 2000; published 27 March 2001)

We report resonant excitation of space-charge and photoconductivity waves in semiconductors with shallow energy levels. We predict the appearance of several resonance maxima in the frequency dependence of non-steady-state photocurrent excited in a semiconductor material. We show that both the effective and actual mobilities of photocarriers can be estimated from the characteristic resonant frequencies. Experimental measurements of the effective mobility are carried out in photorefractive n -type $\text{Bi}_{12}\text{SiO}_{20}$ grown in an argon atmosphere.

DOI: 10.1103/PhysRevB.63.153203

PACS number(s): 72.20.Jv, 42.70.Nq

The subject of space-charge waves covers a wide range of physics from plasmas, acoustics, and semiconductors to the relatively new area of photorefractive optics.¹⁻³ Space-charge waves 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. They determine the dynamic behavior of the space-charge distribution in the crystal and can be excited optically by various methods.⁴⁻⁷ A powerful technique is to use the spatially oscillating interference pattern created by two coherent laser beams. 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. Space-charge and photoconductivity waves play an important role in inducing non-steady-state photocurrents in photorefractive crystals.^{8,9} This effect provides a unique opportunity for direct measurement of the photoelectron's drift mobility from the position of the corresponding resonance maximum in the frequency transfer function of the photocurrent.^{9,10}

The effect of the non-steady-state photo-EMF and most holographic techniques are usually analyzed using the one-level model of a semiconductor. However, space-charge and photoconductivity relaxation in $\text{Bi}_{12}\text{SiO}_{20}$ crystals occur in two steps: fast and slow.¹¹ This behavior is well explained by means of a band transport model with deep and shallow traps.¹¹⁻¹⁴

In this paper we analyze the resonant excitation of space-charge and photoconductivity waves and their detection using the non-steady-state photoelectromotive force effect. We consider the model of a semiconductor with a shallow energy level and present numerical calculations for the commonly used $\text{Bi}_{12}\text{SiO}_{20}$ crystal.^{4-6,8,9,11-16} It is important to point out that application of an external electric field completely changes the physics of the phenomenon and here we deal with the *resonant* excitation of the *eigenwaves* in the material and not with the *forced* excitation of the effect as in Refs. 8 and 14. The investigation of photocurrent generation is useful not only for a better understanding of space-charge waves, but also for practical applications, for instance, for the detection of weakly phase-modulated laser beams for re-

mote laser testing including laser ultrasonic diagnostics.

Let us consider the model of a semiconductor with two levels from which photoelectrons are generated [Fig. 1(a)]. We confine ourselves to photoexcitation from the deep donor level (with total density N_D) and both light and thermal excitation from the shallow level (with total density M_T). The shallow level is assumed to be empty in the dark and the deep one is partially filled (with density of vacant states N_A). The balance equations for the model of photoconductor discussed can be written as¹⁴

$$\partial N / \partial t = S_D I (N_D - N_A - N) - \gamma_D n (N_A + N), \quad (1)$$

$$\partial M / \partial t = -(\beta + S_T I) M + \gamma_T n (M_T - M). \quad (2)$$

Here N is the density of deep centers emptied by light ($N_D^+ = N_A + N$ is the total concentration of ionized donors), M is the density of shallow centers filled with electrons, n is the density of free electrons, S_D and S_T are the light excitation cross sections from deep and shallow levels, β is the thermal excitation rate from the shallow level, γ_D and γ_T are the capture constants for deep and shallow traps, respectively, and I is the light intensity.

For the photocurrent amplitude calculation we have to add to the balance equations (1) and (2) the continuity and Poisson's equations and the expression for the current density:⁹

$$\partial (N - M - n) / \partial t + e^{-1} \operatorname{div} \mathbf{j} = 0, \quad (3)$$

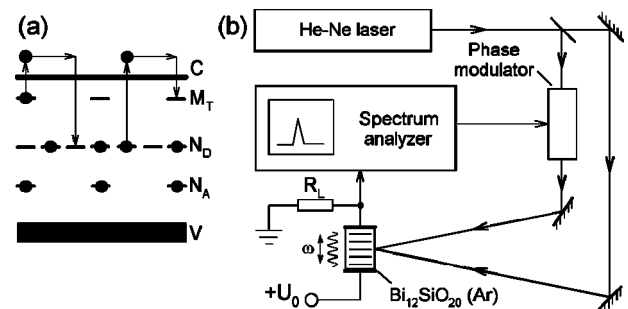


FIG. 1. Model of photoconductor with a shallow level (a) and experimental setup for measurements of the non-steady-state photocurrent (b).

$$\text{div } \mathbf{E} = e(N - M - n)/\epsilon\epsilon_0, \quad (4)$$

$$\mathbf{j} = e\mu n\mathbf{E} + eD \text{ grad } n. \quad (5)$$

Here \mathbf{j} is the electric current density, \mathbf{E} is the electric field, ϵ is the dielectric constant, ϵ_0 is the free space permittivity, μ is the electron mobility, $D = \mu k_B T / e$ is the diffusion coefficient, e is the elementary charge, k_B is the Boltzmann constant, and T is the temperature.

Suppose a crystal is illuminated with the oscillating interference pattern $I(x, t) = I_0 \{1 + m \cos[Kx + \Delta \cos(\omega t)]\}$, where I_0 is the average light intensity, K is the spatial frequency of the interference pattern, m is the contrast, and Δ is the amplitude of phase modulation. Theoretical analysis of the non-steady-state photocurrent generation was performed with the following simplifying assumptions: a negligible dark conductivity, a low-contrast light pattern ($m \ll 1$), and low-amplitude vibrations of the interference pattern ($\Delta \ll 1$). No assumptions were made on the occupancy of the two levels and the prevalence of any activation mechanism for the shallow level.

We have obtained a general expression for the photocurrent amplitude. It is too bulky, however, so we shall discuss the most interesting case of linear generation from the deep level, low filling degree, and negligible light excitation from the shallow level:

$$N_0 \ll N_D - N_A, \quad M_0 \ll M_T, \quad S_T I_0 \ll \beta, \quad (6)$$

$$S_T M_0 \ll S_D (N_D - N_A), \quad \beta T_D \gg 1.$$

Here N_0 , M_0 , and n_0 are the stationary and spatially uniform densities [defined from Eqs. (1) and (2) and the neutrality condition $N_0 = M_0 + n_0$], and $T_D = (S_D I_0 + \gamma_D n_0)^{-1} \approx (\gamma_D n_0)^{-1}$ is the inverse of the sum of the generation and ion recombination rates for the deep level. The fifth condition ($\beta T_D \gg 1$) states that the number of free carriers excited from the shallow level during the lifetime of the ionized deep level is larger than the number of carriers captured at this level.

Under these assumptions the expression for the photocurrent amplitude is simplified to

$$j^\omega = \frac{0.25m^2 \Delta \sigma_0 (Z^*)^{-1} [i2E_0 - \omega \tau_M (E_0 + iE_d)]}{Z - \omega^2 \tau' \tau_M + i\omega [\tau' + \tau_M (1 + \Theta + K^2 L_D^2 + iKL_0)]} - \frac{0.25m^2 \Delta \sigma_0 Z^{-1} [i2E_0 - \omega \tau_M (E_0 - iE_d)]}{Z^* - \omega^2 \tau' \tau_M + i\omega [\tau' + \tau_M (1 + \Theta + K^2 L_D^2 - iKL_0)]}. \quad (7)$$

Here $\sigma_0 = e\mu n_0$ is the average photoconductivity of the crystal, E_0 is the external electric field, $E_d = Kk_B T / e$ is the diffusion field, $\tau_M = \epsilon\epsilon_0 / \sigma_0$ is the Maxwell relaxation time,

$$\tau' = \tau'(\omega) = \tau_D \left(1 + \frac{\tau_T^{-1} \beta^{-1}}{1 + i\omega \beta^{-1}} \right) \quad (8)$$

is the complex and frequency-dependent effective electron lifetime, $\tau_D = [\gamma_D (N_A + N_0)]^{-1}$, $\tau_T = [\gamma_T (M_T - M_0)]^{-1}$ are

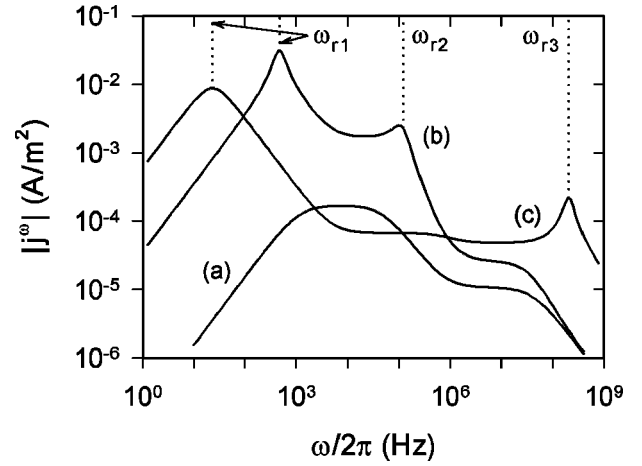


FIG. 2. Theoretical dependencies of the photocurrent amplitude $|j^\omega|$ on the oscillation frequency ω ($I_0 = 10^4$ W/m², $m = 0.2$, $\Delta = 0.1$): (a) $E_0 = 0$, $K = 1.00 \times 10^6$ m⁻¹, (b) $E_0 = 6 \times 10^5$ V/m, $K = 1.89 \times 10^5$ m⁻¹, (c) $E_0 = 2 \times 10^6$ V/m, $K = 1.89 \times 10^6$ m⁻¹.

the recombination times of electrons to the deep and shallow traps, $L_D = \sqrt{D\tau_D}$, $L_0 = \mu \tau_D E_0$ are the average diffusion and drift lengths of photocarriers, $L_S = [\epsilon\epsilon_0 k_B T / e^2 (N_A + N_0) (1 - N_A / N_D)]^{1/2}$ is the Debye screening length,¹² and $Z = 1 + \Theta + K^2 L_S^2 (1 + iE_0 / E_d)$ and $\Theta = [(1 + N_A / N_0) (1 - N_A / N_D)]^{-1}$ are dimensionless parameters ($0 < \Theta < 1$).

The frequency dependence of the photovoltage amplitude is shown in Fig. 2. The material parameters used for numerical calculations are taken from Refs. 5, 11, and 16. Depending on the amplitude of the applied electric field E_0 , spatial frequency K , and material parameters, the frequency response of the photocurrent can have from one to three maxima.

Let us first consider the low-frequency range of photocurrent excitation when $\omega \ll \beta$. For such modulation frequencies the effective electron lifetime is the real value: $\tau' \approx \tau'(0) = \tau_D (1 + \tau_T^{-1} \beta^{-1})$. It can be shown that for $KL_0 > 1 + \Theta + K^2 L_D^2$ there is a maximum in the frequency dependence of the non-steady-state photocurrent at $\omega_{r1} \approx |Z| / |\tau'(0) + \tau_M (1 + \Theta + K^2 L_D^2 + iKL_0)|$. The existence of such a peak is associated with the excitation of space-charge waves.^{4,6} For $L_S < L_D$ the resonant frequency ω_{r1} is a decreasing function of the applied electric field E_0 .^{4,9} However, if the finite screening length L_S is taken into account, ω_{r1} tends to a nonzero limit for high electric fields $E_0 \rightarrow \infty$: $\omega_{r1} \rightarrow T_D^{-1}$ [compare to Eq. (16) in Ref. 9].

For the intermediate range of excitation frequencies when $\omega_{r1} \ll \omega \ll \beta$ and for the case of a relaxation type semiconductor [$\tau'(0) \ll \tau_M (1 + \Theta + K^2 L_D^2 + iKL_0)$] a second maximum is observed in the frequency transfer function. Two types of this maximum can be considered in this case. The first one corresponds to the resonant excitation of a photoconductivity wave and is detected at $\omega_{r2} \approx KL_0 / \tau'(0) = \mu' E_0 K$, where $\mu' = \mu / (1 + \tau_T^{-1} \beta^{-1})$ is the effective mobility of electrons. Such a resonant maximum exists in the frequency range $(1 + \Theta + K^2 L_D^2) / \tau'(0) < \omega < \beta$. This characteristic frequency ω_{r2} is an increasing function of E_0 and K , but when it approaches the thermal excitation rate β , the

corresponding peak disappears. Thus there is an optimum value of the external electric field that maximizes the amplitude of the resonant peak:

$$E_0 \approx \frac{\beta}{\mu' K} \sqrt{\frac{\tau_T}{\tau_D} (1 + \Theta + K^2 L_D^2)}, \quad (9)$$

$$j^\omega \approx \frac{-im^2 \Delta \sigma_0 \beta}{8Z \mu' K} \sqrt{\frac{\tau_T}{\tau_D (1 + \Theta + K^2 L_D^2)}}. \quad (10)$$

The second type of maximum can be observed at another characteristic frequency: $\omega_{m2} \approx (1 + \Theta + K^2 L_D^2) / \tau'(0)$. This peak in the frequency response of the photocurrent is detected only for specific relations between the amplitude of the applied electric field E_0 and the spatial frequency of the interference pattern K : $KL_0, KL_D, KL_S \leq 1$, $L_0 \approx [(1 + \Theta)(L_D^2 + L_S^2)]^{0.5}$. It is important to point out that the origin of this maximum is not associated with the resonant excitation of photoconductivity waves. The characteristic frequency ω_{m2} depends weakly on spatial frequency K , and the amplitude of this maximum increases with increasing K value.

For high excitation frequencies ($\omega > \tau_T^{-1}$) the effective lifetime of electrons is the real value as well: $\tau' \approx \tau_D$. The third resonant peak is observed in this case at the following frequency:

$$\omega_{r3} \approx KL_0 / \tau_D = \mu E_0 K. \quad (11)$$

The presence of such a peak is a characteristic feature of the model discussed. This maximum can be observed in the frequency region $\omega_{r3} > (1 + \Theta + K^2 L_D^2) \tau_D^{-1} + \tau_T^{-1}$. In other words, this condition means that photoconductivity waves are resonantly excited if an electron in the conduction band is captured by neither deep nor shallow traps during the period of interference pattern oscillation. The values of effective μ' and actual μ mobilities can easily be obtained from the characteristic resonant frequencies ω_{r2} and ω_{r3} in the frequency dependence of the non-steady-state photocurrent.

The non-steady-state photocurrent amplitude for the second and third resonances can be estimated from the expression

$$j_{r2,r3}^\omega \approx \frac{-i0.25m^2 \Delta \sigma_0 E_0 Z^{-1}}{1 + \Theta + K^2 L_D^2 + \tau_D / \tau_T (1 + \beta^2 / \omega_{r2,r3}^2)}. \quad (12)$$

We have assumed here that $\omega_{r2}, \omega_{r3} \gg \omega_{r1}$, $|\tau'| \ll |\tau_M(1 + \Theta + K^2 L_D^2 + iKL_0)|$, and $E_0 \gg E_d$.

The origin of the resonance maxima in the frequency transfer function can be explained as follows. The light intensity distribution $I(x, t)$, which is an external periodic driving force in our case, contains waves running in the opposite direction with respect to the applied electric field ($E_0 > 0$): $I^{\pm \pm} \exp[\pm i(Kx + \omega t)]$. These light waves give rise to analogous waves of free carrier concentration: $n^{\pm \pm} \exp[\pm i(Kx + \omega t)]$. The phase velocities of these waves are $v_{ph} = -\omega / K$. For $E_0 \gg E_d$ and $m \ll 1$ each separate electron migrates in the semiconductor crystal with the average drift

velocity $v_{dr} = -\mu E_0$ [during the actual electron lifetime $(\tau_D^{-1} + \tau_T^{-1})^{-1}$] and $v_{dr} = -\mu' E_0$ [during the effective electron lifetime $\tau'(0)$]. If the phase and drift velocities are not matched, i.e., $v_{ph} \neq v_{dr}$, this mentally selected electron outruns or lags behind the packet whose phase velocity v_{ph} is determined by the external influence $I(x, t)$. As a result, the concentration density distribution $n^{\pm \pm} \exp[\pm i(Kx + \omega t)]$ becomes blurred. At resonant frequencies ω_{r2} and ω_{r3} the photoconductivity wave and all the photocarriers migrate synchronously ($v_{ph} = v_{dr}$) giving rise to the appearance of corresponding resonant peaks. For waves traveling in the direction of the applied electric field, i.e., for $n^{\pm \mp} \exp[\pm i(Kx - \omega t)]$, the phase and drift velocities have opposite signs. This means, in particular, that "smoothing" of the photoconductivity wave takes place for any modulation frequency ω and this component does not contribute to the resonant excitation of the external photocurrent.

Let us write down the corresponding complex amplitude of the space-charge electric field formed in the crystal bulk for $E_0 \gg E_d$, $\Theta \ll 1$:

$$E^{++} = \frac{-i0.25m \Delta E_0}{Z^* - \omega^2 \tau_M \tau' + i\omega[\tau' + \tau_M(1 - iKL_0)]}. \quad (13)$$

It is seen from Eq. (13) that the time-dependent electric field grating amplitude is strongly decreased for high excitation frequencies $\omega \gg \omega_{r1}$. For example, this amplitude calculated at ω_{r3} and for the parameters used in Fig. 2, line (c), is only 3×10^{-3} V/m. This fact may greatly complicate the observation of the effects discussed using conventional holographic techniques.

The influence of the space-charge field on the transient response of the photocurrent in semiconductor photodetectors used in astronomy applications was studied in Ref. 17. In our opinion, alternative methods can be based on nonresonant measurements of the non-steady-state photocurrent^{8,14} as well as the concept presented in this paper.

The conventional experimental setup for observation of the non-steady-state photocurrents is presented in Fig. 1(b). A standard single-mode He-Ne laser with an average power of $P_0 = 40$ mW ($\lambda = 633$ nm) was used as the basic source of coherent radiation for formation of a recording interference pattern. To form the interference pattern with a specified fringe spacing and to cause its sinusoidal vibrations within the sample volume, we used a conventional Twyman-Green interferometer. An electro-optic phase modulator ML-102A produced phase modulation of the laser beam. The non-steady-state photocurrent was measured using a standard SK4-56 spectrum analyzer. Figure 3 presents the frequency dependence of the non-steady-state photocurrent flowing in n -type photorefractive $\text{Bi}_{12}\text{SiO}_{20}$ grown in an argon atmosphere. These crystals have several distinct features with respect to sillenite crystals grown in an air atmosphere.¹⁸ First, these crystals have rather high photoconductivity in the red region of the spectrum, comparable with the photoconductivity of the usual sillenite crystals in the blue-green region of the spectrum.⁸ Second, in all investigated crystals for moderate light intensity of the red light we observed an unusual relation between the lifetime of the photocarriers τ' and the

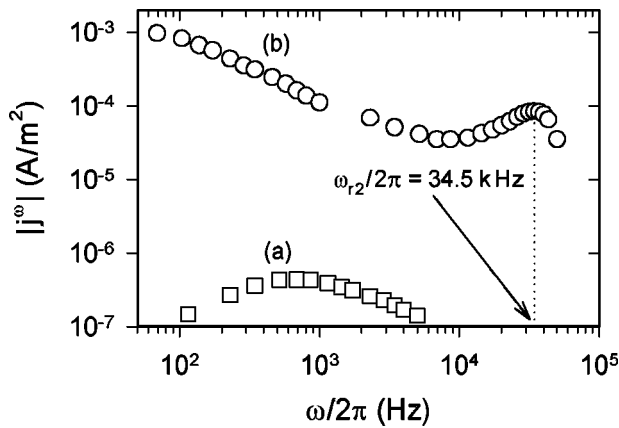


FIG. 3. Frequency dependencies of photocurrent measured in $\text{Bi}_{12}\text{SiO}_{20}$ grown in argon atmosphere ($\lambda = 633$ nm, $K = 5.05 \times 10^4$ m $^{-1}$, $I_0 = 60$ W/m 2 , $m = 0.2$, $\Delta = 0.2$): (a) $E_0 = 0$, (b) $E_0 = 10^6$ V/m.

Maxwell relaxation time $\tau_M < \tau'(0)$. This means, in particular, that photocurrent measurements are done in the lifetime regime without an external electric field. Nevertheless, the appropriate choice of the external electric field makes the

observation of the second resonance possible and photocurrent measurements are done in the relaxation time regime [$\tau'(0) < \tau_M |1 + \Theta + K^2 L_D^2 + iKL_0|$]. The drift mobility value was found to be $\mu' = (4.3 \pm 0.4) \times 10^{-6}$ m 2 /V s ($I_0 = 60$ W/m 2) and corresponds to the values of effective mobilities observed in conventional sillenites.⁹ Thus we think that crystal growth in an oxygen-free atmosphere affects mainly the deep levels, changing the properties of the shallow levels only slightly. The mobility turned out to be dependent on the light intensity, which can be associated with both filling of shallow levels and heating of the crystal in the external electric field.

In conclusion, we have reported resonant excitation of space-charge and photoconductivity waves in semiconductors with shallow energy levels. We predicted the appearance of several resonance maxima in the frequency transfer function of the photocurrent and showed that observation of a photoconductivity wave with conventional holographic techniques is unlikely.

This work was supported by RFBR Grant No. 98-03-32791. I.S. thanks the AvH Foundation for financial support. M.B. acknowledges support from the Grant Center for Natural Sciences.

*Present address: Institute of Physical Chemistry, University of Heidelberg, Im Neuenheimer Feld 253, D-69120 Heidelberg, Germany.

¹R.F. Kazarinov, R.A. Suris, and B.I. Fuks, *Fiz. Tekh. Poluprovodn.* **7**, 149 (1973) [*Sov. Phys. Semicond.* **7**, 102 (1973)].

²A.R. Hutson, J.H. McFee, and D.L. White, *Phys. Rev. Lett.* **7**, 237 (1961).

³A.S. Furman, *Fiz. Tverd. Tela (Leningrad)* **29**, 1076 (1987) [*Sov. Phys. Solid State* **29**, 617 (1987)].

⁴S.I. Stepanov, V.V. Kulikov, and M.P. Petrov, *Opt. Commun.* **44**, 19 (1982).

⁵Ph. Refregier, L. Solymar, H. Rajbenbach, and J.P. Huignard, *J. Appl. Phys.* **58**, 45 (1985).

⁶S.F. Lyuksyutov, P. Buchhave, and M.V. Vasnetsov, *Phys. Rev. Lett.* **79**, 67 (1997).

⁷M. Segev, B. Collings, and D. Abraham, *Phys. Rev. Lett.* **76**, 3798 (1996).

⁸M.P. Petrov, I.A. Sokolov, S.I. Stepanov, and G.S. Trofimov, *J. Appl. Phys.* **68**, 2216 (1990).

⁹I.A. Sokolov and S.I. Stepanov, *J. Opt. Soc. Am. B* **10**, 1483 (1993).

¹⁰M. Hundhausen, *J. Non-Cryst. Solids* **198-200**, 146 (1996).

¹¹G. Pauliat and G. Roosen, *J. Opt. Soc. Am. B* **7**, 2259 (1990).

¹²P. Tayebati and D. Mahgerefteh, *J. Opt. Soc. Am. B* **8**, 1053 (1991).

¹³L. Solymar, D.J. Webb, and A. Grunnet-Jepsen, *The Physics and Applications of Photorefractive Materials* (Oxford University Press, New York, 1996), pp. 117–151.

¹⁴M.A. Bryushinin and I.A. Sokolov, *Phys. Rev. B* **62**, 7186 (2000).

¹⁵E. Soergel and W. Krieger, *Phys. Rev. Lett.* **83**, 2336 (1999).

¹⁶I. Biaggio, R.W. Hellwarth, and J.P. Partanen, *Phys. Rev. Lett.* **78**, 891 (1997).

¹⁷N.M. Haegel, C.R. Brennan, and A. Michael White, *J. Appl. Phys.* **80**, 1510 (1996); N.M. Haegel, J.C. Simoes, A.M. White, and J.W. Beeman, *Appl. Opt.* **38**, 1910 (1999).

¹⁸E.V. Mokrushina, M.A. Bryushinin, V.V. Kulikov, A.A. Petrov, and I.A. Sokolov, *J. Opt. Soc. Am. B* **16**, 57 (1999).