ties. Moreover, the model itself contains one unclear point. It is quite doubtful, for example, that in all the cases a Lorentz distribution of the PSW in the natural frequencies is realized [only then does (3) lead to an exponential damping of $m_z(t)$], whereas in experiment one always observes an exponential behavior after the termination of the transient. It is therefore possible that the observed effects are due not to randomization of the PSW phases, but to entirely different mechanisms, which could not be established in the present study. We note only that these mechanisms cannot be connected with radiative damping due to the reaction of the field in the resonator on m_z and due to the inhomogeneity of the field H_0, inasmuch as these mechanisms were studied in detail and subsequently excluded by choosing small samples and using homogeneous constant magnetic fields. On the other hand, if the $m_z(t)$ relaxation model based on the phase randomization is valid, this means that the presently existing nonlinear theory of parametric excitation of spin waves,[1,7] must be improved, especially in the presence of self-oscillations of the magnetization and of two-magnon scattering of the PSW by inhomogeneities of the sample. Only this will make it possible, by using the procedure proposed here to determine the frequency of the relaxation of the longitudinal high-frequency magnetization, to determine experimentally such important ferrite characteristics as the parameter γ_{ks} of spin-wave damping by the inhomogeneities, the spectrum and amplitude of the inhomogeneous oscillations of the magnetization, and even the value γ_k of the linear damping of the PSW, which is very difficult to determine from the threshold of the instability of imperfect crystals.[7]

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Experimental investigation of the nature of photoelectric phenomena in KDP and DKDP crystals

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A specially developed procedure was used to determine the character of the photoresponse in the dielectric crystals KDP and DKDP to radiation in the transparency band. It is shown that, depending on the experimental conditions on the composition of the crystal, the photoresponse is due mainly to effects of impurity photoconductivity on the nonstationary heating of the crystal lattice, to generation of density oscillations, and to nonlinear optical rectification. It is demonstrated that from the measured photoresponse parameters it is possible to obtain new information on the characteristics of the crystal. In particular, the value of $(\partial\epsilon/\partial T)$, of the KDP crystal was measured at low frequencies.

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1. INTRODUCTION

Dielectric crystals are widely used in nonlinear-optics systems and instruments. However, the study of the structure-sensitive properties, which are responsible for many aspects of the behavior of these materials in laser fields, has not been properly developed as yet. Thus, for most "nonlinear" crystals there is practically no information on the dynamics of the electrons in the allowed bands, on the properties of the defect and impurity states, and others.[1)] This is due to the specific nature of dielectrics, which hinders direct application of procedures developed and verified for the corresponding purposes in semiconductors. However, in analogy with semiconductors, one should expect an investigation of photoelectric phenomena that accompany the interaction between laser radiation and dielectric crystals to be able to fill the existing gaps. Of course, the photoresponse of wide-band crystals to radiation with energy quantum $\hbar\omega$ much less than the band gap[2)] E_g does in fact have a number of specific features connected with the low dark conductivity, low photocarrier concentration,

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and complicated character of the crystal structure. Besides the photoconductivity, which apparently will predominate at $\hbar\omega \gtrsim E_g/2$, the photoresponse may reveal effects due to sample-property changes induced by radiation heating, to nonlinear optical rectification (at the appropriate symmetry) to the onset of sound waves, etc. The complicated character of the photoresponse of dielectrics undoubtedly makes it study difficult, although it does yield additional information on linear and nonlinear properties of crystals, and this information is of independent interest.

In the investigation of the photoresponse of dielectrics, it is convenient and sometimes even necessary to use pulsed lasers of high power. The high laser radiation power makes it possible, despite the high transparency of the crystal, to obtain perfectly measurable values of the photoresponse. The regime of short excitation pulses, which is easily realized with a laser, makes it possible to get around a number of difficulties connected with contact phenomena, which are particularly strong in dielectrics because there is practically no electrostatic screening of the fields. In addition, the pulsed character of the photoexcitation makes it possible to determine in a number of cases the light-induced local changes of the crystal parameters by measuring the integral photocurrent. The purpose of the present work was to investigate the nature of the photoelectric phenomena in dielectrics excited by laser radiation, using as examples the crystals of potassium dihydrophosphate (KDP) and potassium dideuterophosphate (DKDP), which are widely used in nonlinear optics. The last circumstance makes the investigation of the photoresponse particularly timely, in view of the problem of optical endurance of real elements made from these crystals.

2. EXPERIMENTAL PROCEDURE AND CONDITIONS

The general experimental setup used to investigate the photoresponse, with capacitive coupling of the electrodes to the crystal, was close to that cited in Ref. 3. The exciting radiation was the first ($\hbar\omega = 1.17$ eV) or second ($\hbar\omega = 2.34$ eV) harmonic of a neodymium laser. The laser generated single giant pulses with one longitudinal and one transverse (zeroth) mode. The radiation pulse waveform was close to Gaussian with duration $\tau_p \sim 4\times 10^{-8}$ sec. The radiation power varied in a wide range ($P_p \lesssim 25$ MW), but was always lower than the threshold of optical damage to the investigated samples.

It must be pointed out that when a laser is used the illumination of the crystal is essentially inhomogeneous, and this can hinder the interpretation of the experimental results. This difficulty can be circumvented, however, by investigating sufficiently rapid processes under conditions when the local changes of the crystal properties are small. Thus, if the radiation causes the low-frequency dielectric constant ε to change by an amount $\Delta\varepsilon$ and (or) it produces conductivity σ, then it is necessary to satisfy the condition

$$\Delta\varepsilon, 4\pi\tau\sigma \ll \varepsilon, \qquad (1)$$

where τ is the characteristic duration of the photoresponse pulse. The inequality (1) was always satisfied under the experimental conditions, so that the contributions to the individual sections of the samples to the photoresponse were additive.

To prevent possible injection of photocarriers from the metallic contacts, the dielectric liners between the electrodes and the sample were made of teflon, whose ionization potential is ~10 eV. To exclude the influence of surface photoconductivity, the sample and contact dimensions were so chosen[3] that there was no external electric field on the illuminated surfaces. The measures taken to suppress the surface phenomena turned out to be quite effective, as was verified in special experiments.

The relatively high ion conductivity of the crystals at room temperature ($\sigma_0 \sim 3\cdot 10^{-11}\ \Omega^{-1}\text{cm}^{-1}$), with the characteristic dielectric relaxation time $\tau_r \sim 10^{-1}$ sec (Ref. 4) excluded the possibility of using dc sources. High field intensities in the crystal volume can be produced only with alternating fields of frequency $F \gtrsim (2\pi\tau_r)^{-1}$. In experiments we used an alternating voltage $U_0 \lesssim 2\cdot 10^3$ V at $F = 50$ Hz, synchronized with the laser in such a way that the lasing pulse coincided with the maximum voltage on the crystal. Obviously, for processes with duration $\tau \ll F^{-1}$, which we in fact investigated, the voltage supplied to the samples can be regarded as constant.

The change of the effective capacitance of the sample under the influence of the light led to the appearance of a current pulse in the load resistance R connected in series with the crystal resistance. The photocurrent pulse was amplified with a broadband amplifier and registered on an S1-11 oscilloscope. The sensitivity of the registration system was $J_{\min} \sim 6\times 10^{-8}$A at a time resolution $\tau_{s.s.} \lesssim 10^{-8}$ sec. In the experiment we used the photoelectric response in KDP and DKDP (with 93% deuteration), predominantly $X(Y)$- and Z-cut, crystals.[4]

3. EXPERIMENTAL RESULTS AND DISCUSSION

The photoelectric response in KDP and DKDP crystals had a rather complicated character. Typical oscillograms of the photocurrent are shown in the figure. The shapes of the oscillograms dependend substantially both on the radiation wavelength and on the crystal type. Photocurrent could flow also at $U_0 = 0$ (see Figs. a and b), and at $U_0 \neq 0$ the sign of the photocurrent corresponded, depending on the experimental conditions, either to an increase of the effective capacitance of the crystal (c)—positive photoresponse—or conversely to a decrease (d)—negative photoresponse. Besides the photocurrent produced simultaneously with the action of the laser radiation, we observed also current pulses (e) due to the aftereffect. The aggregate of the experimentally observed phenomena can be explained as being due to effects of nonlinear optical rectification, generation of free carriers, and heating of the crystal the latter causing a change of ε and the appearance of acoustic waves.

1. Negative photoresponse (see Fig. d) is observed when the KDP and DKDP crystals are acted upon by neodymium laser light ($\lambda = 1.06\ \mu$m). The current pulse waveform duplicates the oscillogram of the laser pulse, and the amplitude depended linearly on the voltage U_0

applied to the sample and on the incident radiation power P_p. A typical value of the effect over one centimeter along the beam was 5.2×10^{-7} for 2.6×10^{-8} MW in KDP and DKDP, respectively, at $U_0=2\times10^3$ V.

The negative photoresponse cannot be due to the negative photoconductivity observed in semiconductors,[5] since the investigated crystals have practically no dark conductivity. It is natural to attribute the decrease of the effective capacitance of the crystals to the decrease of the dielectric constant. The corresponding photocurrent, when condition (1) is satisfied, can be represented in the form[5)]

$$J=\frac{U_0}{4\pi h^2}\frac{C^2}{C_0^2}\int_{V_0}\frac{d\varepsilon}{dt}dV. \qquad (2)$$

Here C_0, V_0, and h are respectively the capacitance, volume of the sample, and its dimension in the direction between the contacts, and C are the series connected capacitances of the contacts and of the sample.

Turning to the evaluation of $\Delta\varepsilon$, by substituting in (2) the experimental values of the corresponding quantities (the maximum intensity of the light field $E\approx1\times10^6$ V/cm, beam diameter in the sample $d=0.1$ cm, $J_\varepsilon=1.3\times10^{-5}$ A, $U_0=2\times10^3$ V, $h=0.5$ cm, and $C^2/C_0^2=0.55$, we obtain $\Delta\varepsilon=0.2$. This leads to two conclusions: first, the inequality (1) is in fact satisfied ($\varepsilon=50$, $\Delta\varepsilon/\varepsilon<10^{-2}$), second, changes of ε due to the cubic nonlinearity can be disregarded, since the cubic susceptibility in KDP and DKDP is of the order of 10^{-15} cgs esu.[6] The second conclusion is confirmed by the fact that J_ε in KDP turned out to be much larger than in DKDP, whereas the nonlinear characteristics of these crystals are practically the same.

It is well known that in the region of the neodymium-laser frequency KDP has much stronger absorption than DKDP, and this absorption is due to overtones of the hydrogen vibrations in the crystal lattice.[7] Naturally, the large absorption leads to a large heating, and a comparison of the responses of KDP and DKDP points to a thermal character of the effect. In this case

$$\int_{V_0}\frac{d\varepsilon}{dt}dV\approx\frac{\alpha lP_p}{C_v}\left(\frac{d\varepsilon}{dT}\right), \qquad (3)$$

where α is an absorption coefficient ($\alpha=5.5\times10^{-2}$ cm^{-1} for KDP samples used in the experiment) and C_v is the heat capacity.

It must be pointed out here that all the thermodynamic quantities are taken here at constant volume, since the characteristic dimensions are such that the change in the sample volume during the photoresponse time can be neglected. Using (2) and (3) and taking into consideration the experimental values $J_\varepsilon/P_p=5,2.10^{-7}$ A/MW and $C_v\approx C_p=2$ J/cm^{-3} deg^{-1}, we get for KDP $(d\varepsilon_\parallel/dT)_v=5\cdot10^{-2}$ deg^{-1}. The obtained value of $(d\varepsilon_\parallel/dT)_v$ is very close to the value $(d\varepsilon_\parallel/dT)_p=7\cdot10^{-2}$ deg^{-1}.[8]

It is appropriate to emphasize here that the use of pulsed lasers makes possible measurements of thermodynamic characteristics at constant volume, this being of direct interest for crystal physics. We note also that the proximity of the corresponding characteristics of the KDP and DKDP makes it possible, by comparing the photoresponses, to estimate the absorption coefficient in DKDP. The corresponding value is $\alpha_{DKDP}\sim7.10^{-2}$ α_{KDP} The ratio of the absorption coefficients agrees with the relative concentration of the hydrogen in DKDP although this may be accidental.

We have interpreted above the experiment by assuming that the action of radiation with $\lambda=1.06$ μm reduces mainly to heating of the crystal. Naturally, fast inhomogeneous heating leads to the appearance of mechanical stress in the sample and these in turn are sources of compression waves. These waves manifest themselves in a change of the capacitance of the sample, owing to the change of its dimensions. Indeed, a photoresponse (see Fig. e) was produced in the crystal with a substantial delay after the passage of the laser pulse through the crystal, and the duration of the current pulses and their repetition period are determined by the speed of sound, by the diameter of the light beam, and by the transverse dimensions of the crystal. A detailed analysis of the acoustic effects is the subject of a special investigation, which is most pressing because of the role attributed to sound in optical breakdown (see, e.g., Ref. 9). Without dwelling here on these questions, we note only that the employed procedure has made it possible to reveal acoustic effects at illumination intensities greatly lower than the threshold of the optical endurance of the crystals, whereas usually the methods employed make it possible to register sound waves only by destroying the sample.[10]

2. The positive photoresponse, corresponding to an increase of the sample conductivity (see Fig. c) was observed when the KDP and DKDP crystals were acted upon by the second harmonic of a neodymium laser ($\lambda=0.53$ μm, $\tau_p\sim3\times10^{-8}$ sec). The photocurrent pulse duration did not exceed the laser pulse duration, and the photocurrent amplitude was proportional to the amplitude of the applied voltage U_0 and increased with in-

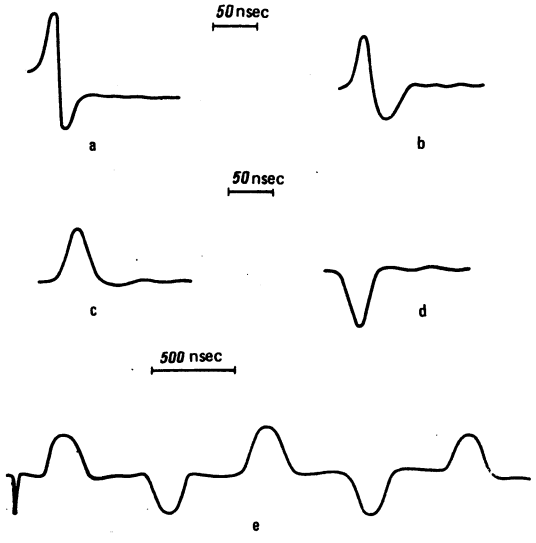

FIG. 1. Typical oscillograms of photoresponses: a,b—KDP and DKDP oriented relative to the radiation polarization $\theta=90°$, $\varphi=45°$; $\lambda=1.06$ μm, $U_0=0$; c—KDP and DKDP, $\lambda=0.53$ μm, $U_0=2\cdot10^3$ V; d—KDP and DKDP, $\lambda=1.06$ μm, $U_0=2\cdot10^3$ V; e—KDP, $\lambda=1.06$ and, $U_0=2\cdot10^3$ V.

creasing radiation power P_p in nearly linear fashion. As to the magnitude of the photoresponse, at a radiation energy $W = 0.1$ J in the pulse and at $U_0 = 2 \times 10^3$ V, the photocurrent in crystals measuring $5 \times 10 \times 40$ mm and at a contact length 10 mm reached $J_\sigma = 2 \times 10^{-6}$ A, and there was no substantial difference between the response in KDP and DKDP.

The connection of the photocurrent J_σ with the light-induced sample conductivity $\sigma(t, r)$ can be represented in the approximation (1) in a form close to (2):

$$J_\sigma = \frac{U_0}{h^2} \frac{C^2}{C_0^2} \int_{V_0} \sigma(t, r) dV. \quad (4)$$

The maximum value of the conductivity at the center of the beam (beam diameter in the sample $d = 1.5$ mm) at $J_\sigma = 2 \times 10^{-6}$ A was, according to (4), 1.5×10^4 sec^{-1}. Recognizing that the duration of the photoresponse is $\tau \sim 3 \times 10^{-8}$ sec, it is easy to verify that the inequality (1) is well satisfied also in this case. The experimentally established fact that the role of the effects due to photocarriers increases with increasing radiation frequency in comparison with the thermal effects, is natural. Indeed, the dominating loss mechanism in the near infrared is due to multiphonon transitions both in the case of KDP and, apparently, in the case of DKDP.[7] We have in mind here, at least for KDP, transitions with excitation of three or four photons of the optical mode of the hydrogen-atom vibrations. Naturally, transitions in the region of one micron are strongly suppressed ($10^{-4} - 10^{-5}$) compared with single-phonon transitions. Doubling the transition frequency enhances this suppression strongly.[6)] Thus, at the neodymium second harmonic the lattice absorption is practically nonexistent and could not be measured to this day in either KDP or DKDP. We note that in the experiments described here where the second harmonic of the neodymium laser was used the compression waves, whose origin is mainly thermal, were not observed. It follows therefore that, within the limits of experimental accuracy, there was likewise no thermal current, and that the absorption did not exceed 10^{-4} cm^{-1}.

On the other hand, an increase of the frequency is accompanied by a sharp increase of the concentration N_n of the forbidden-band states from which transitions with excitation of free carriers at room temperature are possible provided, of course, that the energy of the absorbed quantum is much less than the distance from the Fermi level E_F to the edges of the allowed bands. In fact,

$$N_n = \int_0^{\hbar\omega} N(E) f(E, E_F, kT) dE, \quad (5)$$

where $N(E)$ is the state density in the band, $f(E, E_F, kT)$ is the Fermi distribution function. The energy E is reckoned from the bottom of the conduction band in the case when electrons are excited (or from the top of the valence band in the case of Hall excitation).

At $kT \ll E \leqslant \hbar\omega < E_F$ the second factor in (5) increases very rapidly (like $e^{E/kT}$) with increasing E, whereas the first factor decreases. Little is known concerning the law governing the decrease of $N(E)$, but if, for example, we are dealing with fluctuation states connected with the random distribution of defects, then $N(E)$ can be approximated by $N_{n_0} \exp(-E/kT)$, where T^* is the effective temperature and, as a rule, $T^* > T$ (for crystals grown from the melt, T^* is close to the melting temperature). Under these assumptions we have

$$N_n = N_{n_0} \exp\left[\frac{\hbar\omega}{kT}\left(1 - \frac{T}{T^*}\right)\right]. \quad (6)$$

Thus, a transition from the first to the second harmonic of the neodymium laser ($\hbar\omega/kT \sim 30$) increases strongly the carrier photoexcitation probability.

Of course, the photoexcitation is accompanied by absorption of a light quantum, but the relative magnitude of the γ effects connected with heating and with the change of conductivity amounts in this case, as follows from (2) and (4), to

$$\gamma = \frac{\hbar\omega}{4\pi C_v e \mu \tau_{1t}} \frac{d\varepsilon}{dT}. \quad (7)$$

Numerical estimates at $\mu = 1$ cm^2/V-sec yield $\gamma < 10^{-5}$.

As to the information concerning the nonequilibrium carriers themselves, it follows from the experimental data that their lifetime is $\tau_{1t} \lesssim 10^{-8}$ sec, and the product of the parameters is $n\mu\tau_{1t} = 10^6$ cgs esu. The short lifetime of the nonequilibrium carriers points to a relatively high concentration of the recombination centers N_r. If it is recognized that the upper limit of the cross section for the capture of carriers by a recombination center (at $\varepsilon = 50$ and $T = 300$ K) is $S \approx 10^{-14}$ cm^2 (Ref. 5), then $N_r \gtrsim 10^{15}$ cm^{-3}.

It is appropriate to cite here the experimental data on surface photoconductivity. The photoresponse due to ionization of surface states and corresponding to the increased conductivity of the samples turned out to be significant or even dominant when the external electric field near the illuminated surfaces was of the same order as in the volume of the crystal. The characteristic lifetime of the nonequilibrium carriers near the surafce is $\tau_{sur} \sim 10^{-7}$ sec, i.e., much longer than in the volume, while the amplitude of the "surface photoresponse" J_{sur} depends on the power density, thus indicating a steplike character of excitation of the surface states. At a given power density J_{sur} decreases monotonically upon successive action of a series of pulses. Attention is called to the fact that the first pulse of the series leads to a photoresponse that is substantially (by several orders of magnitude) larger than in the case of volume photoconductivity. It appears that the observed depletion of the photocurrent is due to cleaning of the surface (photodesorption), analogous to that observed earlier for other materials.[12] An additional argument favoring this explanation is the recovery of the surface properties after several days.

3. Nonlinear optical rectification in KDP and DKDP crystals was investigated in rather great detail in Ref. 13. In our experiments at $U_0 = 0$ we also observed a photoresponse due to nonlinear rectification, both at $\lambda = 1.06$ μm and at $\lambda = 0/53$ μm. Figures a and b show oscillograms of this photocurrent in KDP and DKDP at a crystal orientation relative to the radiation polariza-

tion $\theta = 90°$, $\varphi = 45°$.[7)] Inasmuch as under the experimental conditions the inequality $\tau_{s.s.} < \tau_p$ was satisfied, the observed photocurrent was proportional to the derivative of the laser-pulse power. The susceptibilities $\chi^0_{36} = \chi^0_{zxy} + \chi^0_{zyx}$ were close to those measured in Ref. 13.

In an investigation of the photoresponse J_ε and J_σ (in the presence of an external field) the crystals were oriented in such a way that the effect of the nonlinear optical rectification was minimal.

4. CONCLUSION

We have demonstrated above the possibility of observing a photoresponse in dielectric acted upon by radiation with $\hbar\omega \ll E_g$ and with intensity much lower than the optical endurance threshold. The described methods have made it possible to establish the role of various mechanisms responsible for the onset of the photocurrent. In particular, they revealed experimentally the mechanisms connected with the heating of the crystal lattice, generation of sound, and nonequilibrium carriers in the volume and on the surface of the crystals. This yields extensive information on the properties of crystals, for example on their thermodynamic characteristics at constant volume.

One should expect the study of the photoresponse by the procedure described above, at radiation intensities close to the threshold of the optical breakdown, to be able to determine the mechanisms that are responsible for the destruction of real crystals. In addition, the results give grounds for hoping to develop nondestructive methods of monitoring the endurance and the surface states of elements used in nonlinear optics.

[1)] The problem is so pressing that it is proposed to use for its solution, for example, [1] the phenomenon of optical breakdown, which itself has not yet been sufficiently well studied.

[2)] The band gaps of the dielectric crystals KDP and DKDP are apparently ~10 eV.[2]

[3)] The crystal length along the light beam was 40 mm, the length of the contacts was 10 mm, and the distance between contacts was ~5 mm.

[4)] This crystal orientation prevented the second harmonic from being generated in them.

[5)] The signal voltage drop across the load resistance R was neglected in the derivation of (2).

[6)] This was demonstrated earlier [11] with hydrogen-containing molecules as the example.

[7)] The long tail in the photoresponse in KDP at $\lambda = 1.06\ \mu m$ was apparently due to the induced pyroelectric effect (see also Ref. 13) when the crystal was heated by laser radiation. There is no tail in DKDP, nor at $\lambda = 0.53\ \mu m$ in KDP.

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