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Changes in the nonlinear optical properties of a DKDP crystal under the influence of high-temperature photorefraction

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Abstract. The change in phase-matching conditions in second-harmonic generation in DKDP crystals under the influence of the photorefraction appearing because of additional illumination of a sample was investigated. Comparison of the results of a calculation carried out on the basis of a phenomenological model with experimental data revealed their fundamental disagreement. Direct measurements were made of the internal field in the sample induced on photorefraction. A relationship was observed between the phase-matching conditions and the spectroscopic properties of the photorefraction, because irradiation of the sample ensured the excitation of both the photorefraction and of nonlinear optical effects. The internal electric field strength up to 0.5 kV cm^{-1}, obtained for an energy density of the pulsed irradiation of the medium of 10 – 20 J cm^{-2} indicates the necessity to take into account the internal field in the majority of situations when nonlinear optical effects are excited.

1. Introduction

The photorefractive effect (PRE) in DKDP crystals was investigated earlier [1] at temperatures below the Curie point and comparatively recently above it [2 – 4]. According to model ideas, it is associated in both cases with the appearance of electric fields of photorefractive origin. An external electric field may induce the corresponding change in the nonlinear properties of the medium, which should be reflected in the phase-matching conditions in second-harmonic generation [5]. This fact may be described in a phenomenological model of the process, which yields a monotonic dependence of the phase-matching angle on the strength of the electric field generated.

The PRE in hydrogen-containing ferroelectrics is due to elementary quantum processes [6]. The accompanying generation of internal fields with the corresponding alteration of the nonlinear properties of the medium was not discussed previously. We describe below the determination in the experiments of the internal field arising in the crystal from the change in the phase-matching angle and compare this change with the change in the angle when the corresponding external field is applied. It is expected that the efficiency of the nonlinear optical processes in the medium should depend significantly both on the energy parameters of the light pulse at the fundamental frequency and on the properties of the medium. The latter, considered as a set, also depend in their turn on the energy parameters of the pulses [7]. In other words, in the given instance the excitation of the second harmonic should be regarded as parametric, in contrast to its interpretation based on the phenomenological model [6].

The aim of the present study was to demonstrate the dependence of the efficiency of the nonlinear frequency conversion and of its angular characteristics on the PRE intensity. The experiments were performed under the standard conditions for the excitation of the PRE at room temperature (see, for example, Ref. [4]). The results not only revealed for the first time a definite change in the phase-matching angle as a result of the PRE but also made it possible to obtain quantitative data concerning the electric field of photorefractive origin arising in the medium. Under these conditions, the relationship between the efficiency of the second-harmonic generation and the angular and energy characteristics of the fundamental-radiation pulse is significantly nonmonotonic. The latter actually indicates the parametric character of the second-harmonic generation under PRE conditions.

2. Experimental apparatus

The PRE was excited in a DKDP crystal at room temperature under the influence of the radiation from a free-running laser [1]. At the same time, the radiation of another laser, operating in the giant-pulse generation regime, was converted into the second harmonic inside the sample. The phase-matching angle, corresponding to the maximum in the second-harmonic intensity, was determined from measurements of the dependence of the power of the second-harmonic radiation on the angle of incidence of the fundamental radiation on the crystal. Being related to the excitation of electric polarisation in the medium, the PRE should lead to the corresponding changes in the angular dependences of the power and the phase-matching angle itself. By following the behaviour of the phase-matching angle when a nonlinear medium is subjected to two different influences (the photorefractive field and the external electric field), we determined the changes in the phase-matching angle directly as a result of the PRE.

The total experimental setup is presented in Fig. 1. The PRE in a Nd^{3+} : YAP crystal was excited by a free-running laser (21). The lasing energy density was varied in the range 3 – 15 J cm^{-2} with the aid of plane-parallel plates (24), which introduced additional losses. The energy density was monitored with an IMO-2N power meter (6) after passing through a calibrated plate (9). The polarisation plane of

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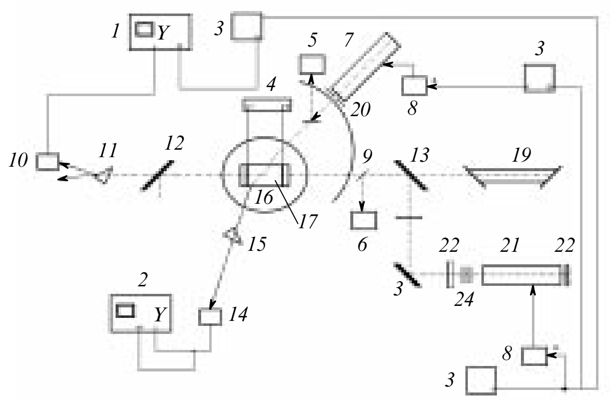

Figure 1. Schematic illustration of the experimental apparatus for the study of the influence of the photorefractive effect in a DKDP crystal on the phase-matching conditions: (1, 2) S9-8 storage oscilloscopes; (3) G5-63 pulse generator; (4) IVN-5 constant-voltage source; (5, 6) IMO-2N power meters; (7) Nd^{3+} : YAG laser; (8) laser pump units; (9) beam splitter; (10) FEU-14B photodetector; (11) polarising KDP prism; (12, 13) interference mirrors; (14) LFDP-2 photodetector; (15) direct-viewing dispersive prism; (16) object stage of the goniometer; (17) DKDP crystal; (18) goniometer alidade; (19) He – Ne laser; (20) aperture; (21) Nd : YAP laser; (22) output and nontransmitting laser mirrors; (23) converging lens; (24) plane-parallel glass plates.

the radiation of this laser was oriented in the XY plane of the crystallographic coordinate system at an angle of 45° relative to the direction of the polariser (11) crossed with the polarisation of an He – Ne laser (19), which corresponds to the maximum probability of the excitation of the PRE [8]. A mechanically free DKDP crystal (17), consisting of a 3.6 cm × 2 cm × 8.5 cm parallelepiped, was placed within the mounting of a G5 goniometer located on the object stage (16) and permitting both additional irradiation of the crystal by a fundamental-harmonic pulse under controlled conditions and the application of a regulated electric field to the sample along the optical axis of the crystal (the Z axis). This made it possible to measure in exactly the same way the changes in the phase-matching angle owing to the external field in the medium in the absence of the PRE and as a result of the internal field in the presence of the PRE.

The photoinduced field in the apparatus was monitored by means of an optical polarisation method which is standard for this case [9]. The radiation exciting the PRE via the mirrors (13) was formed by a long-focus lens into a beam 4 mm in diameter and was directed along the optical axis of the crystal. The testing channel in this system consisted of a low-noise LGN-214 laser (21) and an analyser (11). Its radiation was polarised at an angle of 45° relative to the crystallographic X or Y axis. The beam diameter (2 mm) guaranteed its interaction with the central zone of the photorefractive changes in the medium.

The photorefractive response was recorded by an oscilloscope (1) with an FEU-14B-based detection system (10) having a matching stage based on a high-frequency KT973A transistor. The proportionality of the output electric signal to the signal from the photodetector input was ensured in the detection process. Monitoring of the input signal makes it possible to measure the photorefractive field $E_3^{\rm pr}(t)$ in the crystal. The use in the pump and testing channels of radiations with different wavelengths together with an interference mirror (12) ensured the effective optical decoupling of the channels.

An Nd^{3+} : YAG laser (7), operating in the giant-pulse regime, was used as the source of converting radiation. The energy of an individual pulse was measured with the aid of an IMO-2N meter (5). The laser (7) was attached to the alidade of the G5 goniometer (18) in a mounting permitting the rotation of the polarisation plane of the radiation. After passage through the aperture (20), restricting the beam almost as far as the diffraction limit, the radiation passed into the sample (17), which was rigidly attached to the object stage of the goniometer (16). After passing through the crystal, the first- and second-harmonic radiations were separated with the aid of a direct-viewing dispersive prism (15).

An LFDP-2 photodetector (14) together with the storage oscilloscope (2) were used to record the energy of the second-harmonic radiation. The chosen dynamic range of the signals guaranteed the direct proportionality of the electric-pulse amplitude to its power. In order to investigate the influence of the PRE on the phase-matching conditions, the operation of both lasers was synchronised taking into account the delay time of the photorefractive response relative to the pump pulse.

Fig. 2 presents a typical dependence of the lasing efficiency at a frequency 2ω on the angle of orientation of the polarisation α and the first-harmonic radiation when phase-matching conditions are fulfilled. This dependence, the maximum of which corresponds to 48°, agrees fully with the direction of the PRE maximum obtained previously [8]. Thus it becomes clear that, for a constant pump power, the beam power $P_{2\omega}$ is explicitly related to the direction of polarisation, whereas the $P_{2\omega}$ maximum is explicitly related to the PRE maximum. Apart from other features, the determination of the phase-matching angle whilst varying the parameters of the influence applied to a nonlinear medium may be recognised as an extremely efficient method for the determination of the quantitative characteristics of such influences when a priori information about them is lacking. In the present study, this was achieved in respect of photorefractive electric fields.

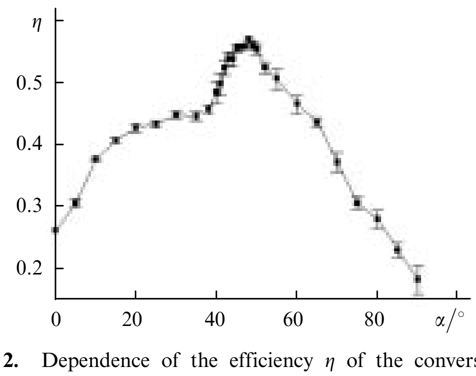

Figure 2. Dependence of the efficiency η of the conversion of the Nd^{3+} : YAG laser radiation into the second harmonic on the angle α of the converted-radiation polarisation.

3. The influence of photorefractive perturbations in the medium on the phase-matching conditions

The dependence of the shift of the phase-matching angle on the direction of the field applied to the crystal and the induced internal photorefractive field was calculated on the basis of a phenomenological model [5]. The efficiency η of the conversion of the radiation into the second harmonic is defined as the ratio of the power $P(2\omega)$ of the converted radiation to the power $P(\omega)$ of the incident radiation:

$$\eta = \frac{P(2\omega)}{P(\omega)} = \frac{2\pi^2 d^2 L^2 P(\omega)}{\varepsilon\, n_{1o} c n_{2e}^2 \lambda_2^2 A}\left(\operatorname{sinc}\frac{|\delta k|L}{2}\right)^2 , \qquad (1)$$

where ε is the permittivity of the crystal; n_{1o} is the refractive index of the ordinary beam at the fundamental frequency; L is the length of the path traversed by the converted radiation in the crystal; n_{2e} is the refractive index of the extraordinary beam at the frequency 2ω; λ_2 is the wavelength of the second-harmonic radiation; A is the cross-sectional area of the beam; $d = d_{36}\sin\theta_m$ is the effective nonlinear parameter; $d_{36} = 4.02\times10^{-12}$ mV^{-1} is a component of the nonlinear susceptibility tensor of the DKDP crystal [10].

When the crystal is irradiated by powerful laser radiation, the refractive index for the extraordinary beam changes as a consequence of the Pockels effect under the influence of the external photorefractive field. The refractive index for the extraordinary beam also changes when the converted radiation propagates at an angle θ relative to the optical axis of the crystal:

$$n_o(E) = n_o(0) + n_o^3 r_{63} E , \qquad (2)$$

$$n_e(E,\theta) = n_o(E)\left[\frac{1+\tan^2\theta}{1+[n_o(E)\tan\theta/n_e]^2}\right]^{1/2} , \qquad (3)$$

where E is the electric field strength of the crystal; $r_{63} = 26.4\times10^{-12}$ mV^{-1} is a component of the linear electro-optical effect tensor in a DKDP crystal. A change in temperature also alters the birefringence of the crystal, which may be calculated from the relationships

$$\Delta n_o = (298\text{K} - T)(n_o^2 - 1.047)2.28\times10^{-5} ,$$
$$\Delta n_e = (298\text{K} - T)n_e^2 9.55\times10^{-6} , \qquad (4)$$

where T is the temperature; Δn_o and Δn_e are the changes in the ordinary and extraordinary refractive indices.

All this indicates a phase mismatch:

$$\Delta k = \frac{4\pi}{\lambda_1}[n_{1o}(T,E) + n_{1e}(T,E,\theta) - n_{2e}(T,E,\theta)] . \qquad (5)$$

Relationship (1) makes it possible to estimate the changes in the dependences of η on the angle θ between the wave vector of the converted radiation and the optical axis of the crystal when the latter is placed in an external field on the assumption that d_{36} = const. The angle corresponding to the maxima in the relationships was in fact adopted as the phase-matching angle θ_{pm}. Fig. 3a presents a plot of the $\eta(\theta)$ relationship, which is smooth over a fairly wide range of angles and demonstrates a monotonic shift of the maximum as the field strength varies. The $\delta\theta_{pm}(E) = \theta_{pm}(E) - \theta_{pm}(0)$ relationship was plotted accordingly. The $\theta_{pm}(E)$ relationship plotted on the basis of the plots in Fig. 3a is linear (Fig. 3b). The calculation was carried out within the framework of the phenomenological model of the nonlinear effect described above; the contribution of the energy of the converted pulse to the photorefractive state was not then taken into account.

The dependences of the phase-matching angle on the energy W of the Nd^{3+} : YAG laser (7) pulse and on the field strength E were determined from the measured $\eta(\theta)$: $\delta\theta_{pm}(W) = \theta_{pm}(E,W) - \theta_{pm}(E, W=0)$ and $\delta\theta_{pm}(E) = \theta_{pm}(E, W=0) - \theta_{pm}(E=0, W=0)$. All the relationships

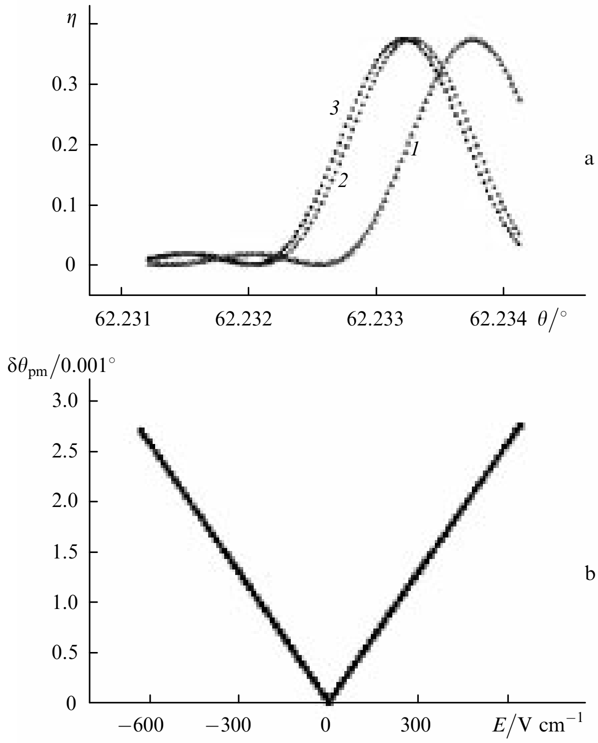

Figure 3. Calculated dependences of the efficiency η of the conversion of the Nd^{3+} : YAG laser radiation into the second harmonic on the angle θ relative to the optical axis of the crystal for internal-field strengths $E = 0$ (1), 110 V cm^{-1} (2), and 125 V cm^{-1} (3) (a) and of the changes in the phase-matching angle $\delta\theta_{pm}$ on the electric field strength E in the crystal (b).

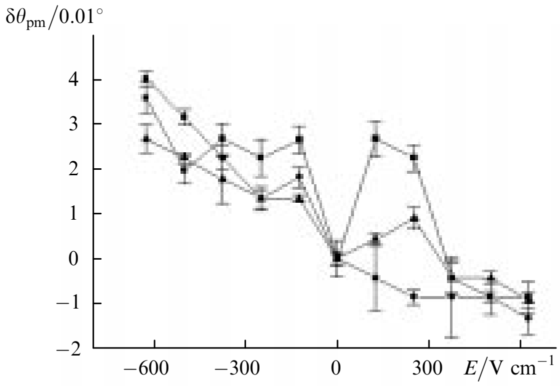

Figure 4. Dependence of $\delta\theta_{pm}$ on the internal field strength E in the crystal for energy densities of the radiation being converted of 20.5 J cm^{-2} (■), 17.5 mJ cm^{-3} (▲), and 15.5 mJ cm^{-2} (●).

obtained here and subsequently were calculated from the results of 5–8 measurements. The plots in Fig. 4 exhibit the nonlinear response of the medium, associated with the pulse energy of the laser (7), to the complex influence of the pulsed pumping.

The main and striking feature is the existence in the region of $E \approx \pm150$ V cm^{-1} of a pseudoparabolic relationship centred on $E = 0$. This result agrees with data obtained earlier on the electric hysteresis for the medium under consideration [3]. These data made it possible to establish the fact that the external field imposes the sign of the internal photorefractive field starting from precisely such field strengths (cf the relationship in Fig. 2 with the relationships in Fig. 3 of Ref. [3]).

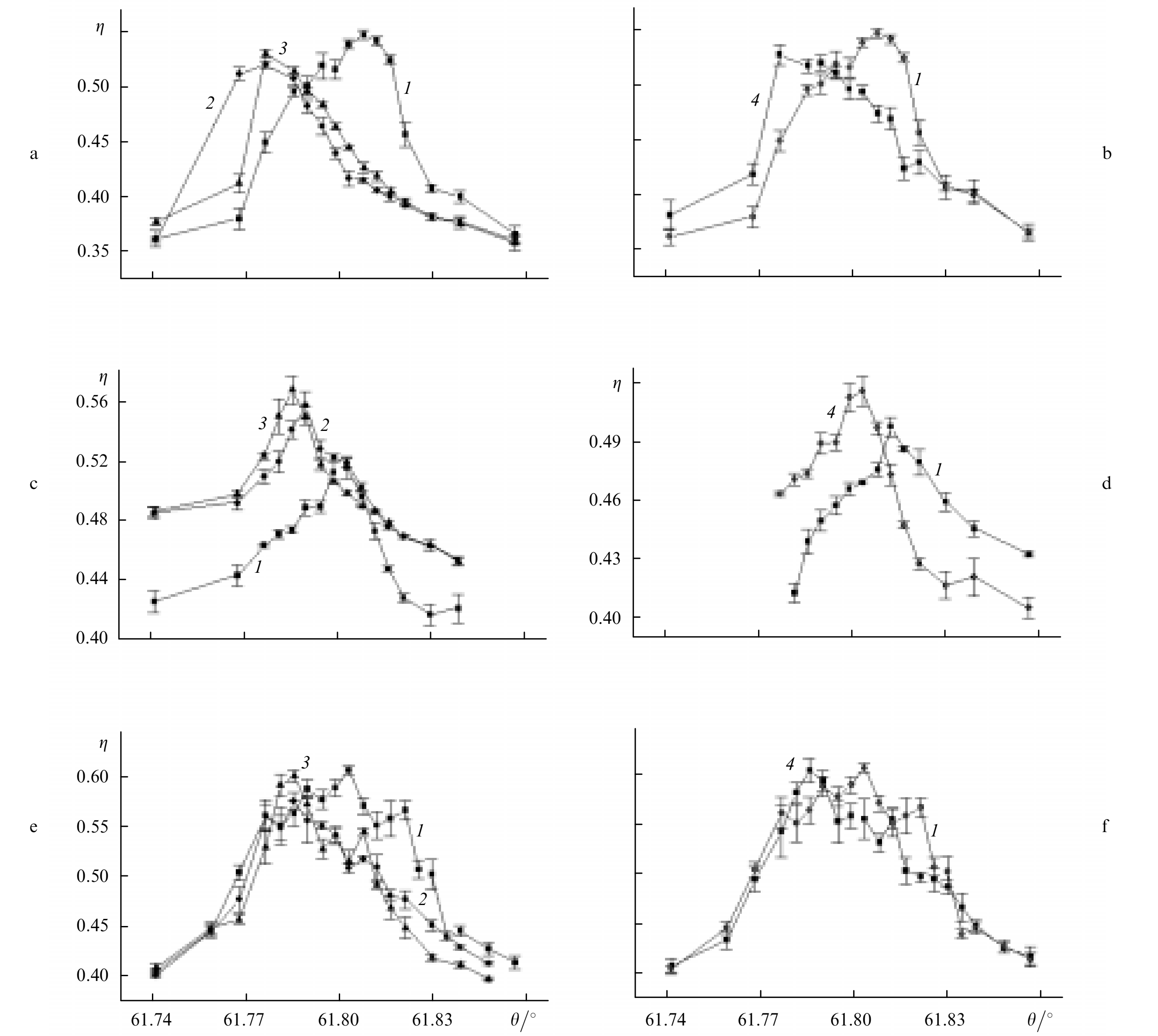

Figure 5. Eaperimental dependences of the conversion efficiency η on the angle θ relative to the optical axis of the crystal at $T = 18.5$ °C and $W = 0$ [$E_c = 0$ (1), -125 V cm^{-1} (2), and 125 V cm^{-1} (3)] (a), $T = 18.5$ °C and $E_c = 0$ [$W = 0$ (1) and 15 J cm^{-2} (4)] (b), $T = 21.5$ °C and $W = 0$[$E_c = 0$ (1), -110 V cm^{-1} (2), and 110 V cm^{-1} (3)] (c), $T = 21.5$ °C and $E_c = 0$ [$W = 0$ (1) and 10 J cm^{-2} (4)] (d), $T = 22.5$ °C and $W = 0$ [$E_c = 0$ (1), -125 V cm^{-1} (2), and 125 V cm^{-1} (3)] (e), and $T = 22.5$ °C and $E_c = 0$ [$W = 0$ (1) and 15 J cm^{-2} (4)] (f).

The experimental relationships (Fig. 5) analogous to the calculated relationships (Fig. 3a) exhibited significant differences from the relationships plotted on the basis of the phenomenological model even for the small temperature variation in the range 18.5 – 22.5 °C. The form of the relationships changes significantly: from asymmetric at the lower limit [this happens in both the external field E_c and photorefractive field E (Figs 5a and 5b)] to symmetrical with an expanded profile at somewhat higher temperatures (Figs 5e and 5f). At the middle temperature, the relationships acquire a distinct central peak (Figs 5c and 5d). The experimental confirmation of the repeatability of the relationships in Fig. 3b for different field polarities is striking.

The values of W for the relationships in Figs 5b, 5d, and 5f were chosen so that $\delta\theta_{pm}$ was equal to the same change for a particular field E applied to the crystal. This yielded a temperature-dependent change of the phase-matching angle $\delta\theta_{pm}$ and the corresponding photorefractive field (Table 1). Thus, an increase in temperature induces an overall decrease in the photorefractive field.

Finally, the relationships of the type presented in Fig. 5 made it possible to obtain the dependence of the photorefractive field on the pump pulse energy, which, according to the results of optical polarisation measurements, is smooth outside the limits of the photorefractive threshold [2]. We superimposed on this dependence the results of measurements

Table 1.

T/°C	W/J cm^{-2}	$\delta\theta_{pm}$/′	E/V cm^{-1}
18.5	15	3.5	90
21.5	10	1.5	65
22.5	15	2	72

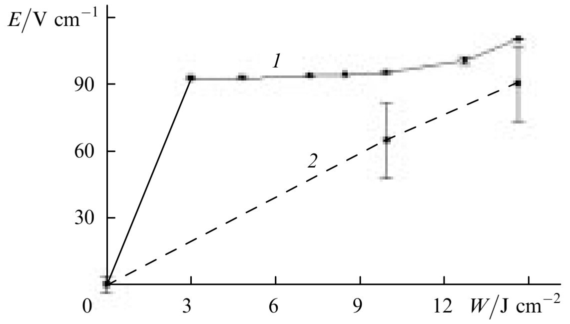

Figure 6. Dependence of the internal field E on the energy density of the exciting radiation W obtained by the polarisation-optical method (1) and from the change in the phase-matching angle (2).

by the method developed here — by recording the phase-matching angles. Evidently, these two dependences (Fig. 6) do not differ to an unduly great extent.

4. Discussion of the results

The results presented above demonstrate the possibility of direct almost local measurements of the photorefractive fields in crystals of the KDP group. The usual optical polarisation method is indirect and yields a result integrated both over the cross section of the testing beam (2 mm in diameter) and over the entire length of the crystal (about 8 cm) (curve 1, Fig. 6). Our measurements of the photorefractive field based on the change in the phase-matching angle have an almost local character (the length of the nonlinear interaction of the radiation with the photorefractive zone is of the order of 5 mm for an aperture of about 1 mm) an agree quite well with the results of optical polarisation measurements (Fig. 6). Here, we begin with the fact that the nonlinear susceptibility determining the parameter d remains unchanged when n_o and n_e are varied. Further studies on these lines will make possible to find with the aid of experimental data the nonlinear susceptibility of photorefractive origin and the changes in it.

Analysis of Fig. 5 shows that the experimental width of the angular dependence of the conversion efficiency exceeds almost by an order of magnitude the calculated width (Fig. 3a) and that a slight variation in the sample temperature induces significant changes in the structure of the angular dependence of the conversion efficiency with appearance of its fine details (Figs 5e and 5f). An increase in the range of second-harmonic angles following a slight increase in T in the region of room temperatures indicates the dominant role of secondary processes in the presence of hydrogen bonds, in conformity with the PRE model adopted earlier [6]. Since the typical hydrogen-bond dissociation temperature is approximately 100 °C, the data presented may be regarded as arising precisely from the processes in the proton subsystem of the ferroelectric.

The observed relationship between the PRE and the nonlinear optical process partly requires an additional analysis in specific situations in relation to the operating regime of the device. This is important, because such a task should include both ensuring the required conversion efficiency and allowance for the temporal and angular aspects of the interaction; analysis based solely on simplified models of the processes is in this case clearly insufficient. This question will be the subject of the next publication, which will be based on a model taking into account the influence of the converted radiation and the changes in the effective nonlinearity of the crystal on the phase-matching conditions.

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