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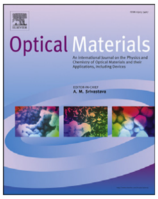

Nonlinear absorption properties of DKDP crystal at 263 nm and 351 nm

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ABSTRACT

At the wavelength of 263 nm and 351 nm, the nonlinear absorption curves of 66% deuterated DKDP crystal were measured in the geometries of beam polarizing along the optics axis (E||Z) and perpendicular to it (E⊥Z). The results indicate that the nonlinear absorption in the E⊥Z geometry is stronger than that in the E||Z geometry. The nonlinear absorptions at 263 nm and 351 nm are identified to two- and three-photon absorption, respectively. The theoretical fits to the experimental data yields the two-photon absorption coefficients of 0.32 ± 0.03 cm/GW (E⊥Z geometry) and 0.17 ± 0.02 cm/GW (E||Z geometry) at 263 nm, and the three-photon absorption coefficients of $(8.1 \pm 1.1) \times 10^{-4}$ cm^3/GW^2 (E⊥Z geometry) and $(2.2 \pm 0.5) \times 10^{-4}$ cm^3/GW^2 (E||Z geometry) at 351 nm.

1. Introduction

In high power large-aperture laser systems, the Nd:glass laser is usually converted to ultraviolet (UV) light by KDP and its deuterated analog DKDP crystals, owning to their high damage threshold, moderate nonlinear-optical coefficient and the ability of growing into large sizes [1,2]. In contrast with KDP crystal, DKDP crystal with the deuterium content of 60%–80% can effectively reduce transverse stimulated Raman scattering, which will induce damage to the optic component [3,4]. Moreover, DKDP crystal is a prominent candidate to realize fourth harmonic generation for achieving non-critical phased-matching to avoid narrow angular acceptance and beam walk-off [5]. Therefore, DKDP crystal is a prospective material for frequency conversion in the UV region.

One of the main problems of the converter crystal is the absorption of UV light increasing nonlinearly with the laser radiation intensity. It is reported that the absorption for 1 cm length KDP crystal at 355 nm increases from 4.4% to 6.3% when energy density increases from 0.1 J/cm^2 to 3 J/cm^2 [6]. The nonlinear absorption (NLA) is usually ascribed to multi-photon absorption. In the process of harmonic generation, the NLA of the converter crystal leads to energy loss limiting the output capability of laser system [7,8]. In addition, the NLA poses a threat to crystal damage. C. W. Carr et al. [9] observed sharp steps in damage threshold of DKDP crystal at 2.55 eV and 3.9 eV associated with multi-photon absorption, while in the region between steps the damage threshold decreases smoothly with decreasing wavelength. The results suggest that the NLA process dominates the mechanism for damage initiation in the component. Therefore, the NLA of converter crystal is of great concern in high power large-aperture laser systems. So far, the NLA of KDP crystal have been investigated at the wavelengths of 211 nm, 216 nm, 248 nm, 264 nm, 266 nm and 355 nm et al., whereas little work has been reported on the NLA of DKDP crystal [10–13]. Besides, the NLA of crystal depends significantly on crystal cut and beam polarization, which has been examined in BBO, GaSe, GaN and CdTe crystals [14–16]. Still, the anisotropy of NLA in DKDP crystal remains ambiguous.

In the present paper, we report the NLA properties of 66% deuterated DKDP crystal at 263 nm and 351 nm. The corresponding NLA coefficients are obtained according to the theory of two- and three-photon absorption. The NLA in the geometries of E||Z and E⊥Z are also compared.

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2. Experimental

2.1. Sample preparation

A large-size DKDP crystal with the deuteration degree of 66% was grown from deuterated aqueous solution by traditional temperature reduction method in the temperature range of 56.4 °C–32.9 °C. DKDP crystal grew along the Z direction with growth rate of 1.2 mm/day. A high-quality sample was cut from the large-size DKDP crystal with the direction at 90° to the crystal Z axis ($\theta = 90^\circ$) and at 45° to the crystal X axis ($\varphi = 45^\circ$), as shown in Fig. 1. The sample size is 15 mm × 15 mm × 8 mm. The (110) planes of the sample were fine polished by using a conventional manual polishing method with nonaqueous slurries.

2.2. Linear transmission

The linear transmittance spectra of the sample were measured by a Lamda-950 spectrophotometer at room temperature. In order to observe the anisotropy of the transmittance, the measurements were operated in the geometries of E||Z and E⊥Z from 220 nm to 800 nm.

2.3. Nonlinear transmission

The transmission-intensity dependence is an intuitive method to characterize the NLA. The experimental setup for measuring the transmission-intensity dependence is depicted in Fig. 2. We used a Nd:YLF laser system to provide the fundamental light with a wavelength of 1053 nm at the repetition rate of 1 Hz. The fundamental light is a temporal and spatial Gaussian beam. 263 nm and 351 nm laser were obtained through the harmonic generation system consisting of KDP and DKDP crystals and the pulse width is estimated to be 50 ps (FWHM). The incident beam for NLA measurement was selected by a prism and focused into the DKDP crystal sample by a lens with a focal length of 1.3 m, so that the beam size could be regarded as constant within even the longest crystal of 10 mm. A homogeneous part was selected using a circular aperture. The position of the sample remained 0.3 m in front of lens focal point to avoid self-focusing. The incident beam propagated along the [110] direction of DKDP crystal and was monitored with the sampling system made of two splitters (fused silica plates), attenuator and CCD camera. The CCD camera pixels have 4095 Gy levels and each pixel is 13 μm × 13 μm. Attenuator was used to keep the camera within its linear response range. To ensure the beam size on the sample is imaged onto the CCD camera, the propagating length from the lens to the input plane of CCD camera is equal to that from the lens to the sample center. The energy of the transmitted pulse was measured by an energy meter. For each measurement point, we measured three positions in the sample.

The beam profile obtained by the CCD camera, as shown in Fig. 3a, reveals almost Gaussian intensity distributions in space and the beam radius at the e^{-1} level can be therefore obtained. Within the linear response range of CCD camera, the gray of the beam profile is approximately proportional to the input energy. With an appropriate calibration, the mean gray of the beam profile could be used to characterize the input energy. The calibration curve was measured with no crystal sample, as shown in Fig. 3b. As a result, both size and energy of the incident beam can be evaluated from the beam profile. This new method has the advantage of simple manipulation and higher sensitivity.

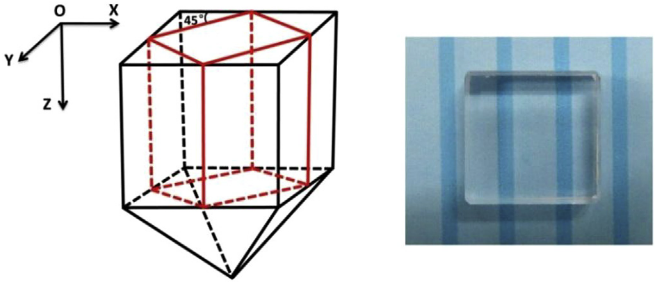

Fig. 1. Cutting schematic diagram and photograph of the crystal sample.

3. Theory

The incident intensity for generated UV pulse with a Gaussian profile in time and space is defined as [17].

$$I_{in} = I_0 \exp\left[-(2t/\tau_P)^2\right] exp\left[-(r/\omega_0)^2\right] \tag{1}$$

where I_0 is the maximum on-axis intensity, τ_p is the pulse width at the e^{-1} level, and ω_0 is the beam radius at the e^{-1} level (at FWHM $\tau = \sqrt{\ln 2}\tau_P$, $\omega = 2\sqrt{\ln 2}\omega_0$).

It is known from literature that the band gap E_g of DKDP crystal is between 7.6 eV and 8.8 eV at room temperature [9,18]. According to the theory of multi-photon absorption, two-photon absorption (2 PA) dominates the NLA when the photon energy $h\nu$ is in the spectral region of $E_g/2 < h\nu < E_g$, whereas, three-photon absorption (3 PA) manifests itself primarily in the region $E_g/3 < h\nu \le E_g/2$ [19]. Theoretically, the NLAs in DKDP crystal at 263 nm (4.72 eV) and 351 nm (3.54 eV) are ascribed to 2 PA and 3 PA, respectively. Now we consider a Gaussian laser beam traveling in the x direction within crystal sample exhibiting linear and nonlinear (2 PA or 3 PA) absorptions. The light beam propagation through the sample is governed by the following equation [19].

$$2PA : dI/dx = -\alpha I - \beta I^2 \tag{2}$$

$$3PA : dI/dx = -\alpha I - \gamma I^3 \tag{3}$$

where I is the instantaneous intensity, α is the linear absorption coefficient, β and γ represent the 2 PA and 3 PA absorption coefficients, respectively. The intensity I_{out} at the output of a crystal can be derived according to Eqs. (1)–(3). The pulse energy should be the integration of intensity over space and time, namely.

$$E_{in} = \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} I_{in}(r,t) 2\pi r dr \tag{4a}$$

$$E_{out} = \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} I_{out}(r,t) 2\pi r dr \tag{4b}$$

To determine the NLA coefficients, we should measure the intensity-dependent energy transmission $T = E_{out}/E_{in}$ in a thick sample. For 2 PA and 3 PA, the expressions of intensity-dependent energy transmission can be expressed as [12,20].

$$T_{2PA}(I_0) = \frac{(1-R)^2 \exp(-\alpha L)}{\sqrt{\pi} q_0} \times \int_{-\infty}^{\infty} \ln\left[1 + q_0 exp\left(-x^2\right)\right] dx \tag{5a}$$