Supplementary information for

Determination of intrinsic defects of functional KDP crystal with flawed surfaces and their effect on the optical properties

Wenyu Ding,[a] Jian Cheng,*[a] Linjie Zhao,[a] Zhenhua Wang,[a] Hao Yang,[a] Zhichao Liu,[b] Qiao Xu,[b] Jian Wang, [b] Feng Geng [b] and Mingjun Chen *[a]

[a] *State Key Laboratory of Robotics and System, Harbin Institute of Technology, Harbin 150001, China*

[b] *Research Center of Laser Fusion, China Academy of Engineering Physics, Mianyang 621900, China*

* Email: chenmj@hit.edu.cn (M.J. Chen)

* Email: cheng.826@hit.edu.cn (J. Cheng)

Contents

1. Pre-set of lateral cracks Ⅲ

Fig. S1 Laser-induced damage threshold (LIDT) for KDP crystal with different kinds of manufacturing-induced structural defects.

Fig. S2 Schematic diagram of preset cracks. (a) Schematic diagram of indenter loading, (b) Front view of schematic diagram of crack formation and (c) Morphology of preset cracks on the crystal surface.

2. Information of vibration spectra of lateral cracks Ⅴ

Table S1 The specific vibration spectrum information of lateral cracks: FTIR spectrum and Raman spectrum.

3. Electronic structure and optical properties of perfect KDP crystal Ⅵ

Fig. S3 Distribution of energy level structure and density of states of perfect KDP crystal. (a) Energy level structure, (b) Total density of states.

Fig. S4 Imaginary part of dielectric function of perfect KDP crystal.

4. The laser damage experiments Ⅶ

Fig. S5 The diagram of laser damage light path.

1. Presets of lateral cracks

Extensive laser damage threshold experiments were conducted for the KDP crystal surface with different kinds of manufacturing-induced structural defects. The results in Fig. S1 showed that presence of lateral cracks greatly reduce the laser-induced damage threshold (LIDT) of the KDP crystal. In addition, the previous research results of laser damage tests [1,2] also showed that the LIDT of crystals with lateral cracks is the lowest. Therefore, the effect of lateral cracks on the laser damage of crystals is more representative. The intrinsic defects introduced by lateral cracks were explored in this work.

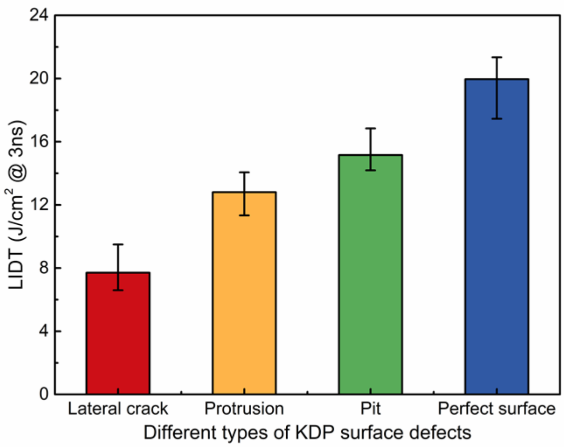


Fig. S1 Laser-induced damage threshold (LIDT) for KDP crystal with different kinds of manufacturing-induced structural defects.

The sample size of the KDP crystal is 50×50×10mm. A standard Vickers indenter with a cross-sectional angle of 136° was applied to the crystal surface with a load of 100g to achieve the pre-setting of lateral cracks. Fig. S2(a) and (b) are the schematic diagram of artificial cracks prepared by Vickers indenter. Fig. S2(c) is the preset cracks on the crystal surface. There would be crescent-shaped lateral cracks on both sides of the indentation on the surface due to the extrusion of the indenter.

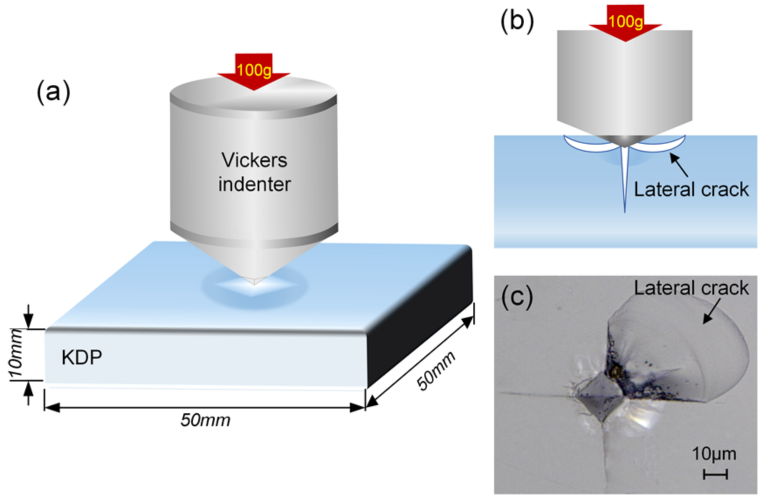


Fig. S2 Schematic diagram of preset cracks. (a) Schematic diagram of indenter loading, (b) Front view of schematic diagram of crack formation and (c) Morphology of preset cracks on the crystal surface.

Reference:

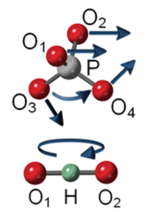
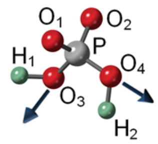
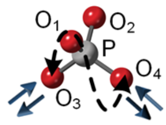
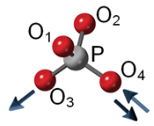
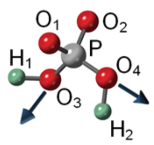
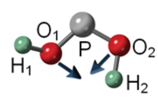
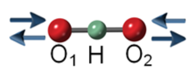
(1) J. Cheng, M. Chen, W. Liao, H. Wang, J. Wang, Y. Xiao and M. Li, *Opt. Express*, 2014, **22**, 28740.

(2) S. Wang, J. Wang, Q. Xu, X. Lei, Z. Liu and J. Zhang, *Appl. Opt*., 2018, **57**, 2638.

2. Information of vibration spectra of lateral cracks

The Raman spectroscopy and Fourier transformed infrared spectroscopy (FTIR) complement each other to achieve the double check of the molecular structure information of the manufacturing-induced lateral cracks.

Table S1 The specific vibration spectrum information of lateral cracks: FTIR spectrum and Raman spectrum

Frequency (cm^{-1}) / Intensity		Assignment	Description
Raman spectra	Infrared spectra		
367/ w, b	–	v_2	
391 / m	–	v_2	
475 / w	–	v_2	
530/ m	530 / s	v_2 / O-H wag	
556 / w, b	–	out-of-plane bend	
–	825 / vs	P-O(H) str.	
914 / vs	–	v_1	
1010 / mb	–	v_3	
–	1024 / m	v_3	
–	1274 / m	P=O str.	
–	1524 / w, b	P-OH bend	
1664 / w, b	–	P-OH bend	
–	2351 / w	O-H str.	
2372 / w	–	O-H str.	
2695/ w	2695 / w	O-H str.	
2950 / w	–	O-H str.	

[1] *s=strong, m=medium, w=weak, v=very, b=broad, –=zero;*

[2] *v_1: symmetric stretching vibration, v_2: Bending vibration, v_3: asymmetric stretching vibration, str: stretch.*

3. Electronic structure and optical properties of perfect KDP crystal

Based on local density functional theory (DFT), the band gap of the perfectly configured KDP crystal was calculated to be 7.23 eV, as shown in Fig. S3. This information is used as the basis for discussing the effect of different intrinsic point defects on the energy level structure of KDP crystals in the main text.

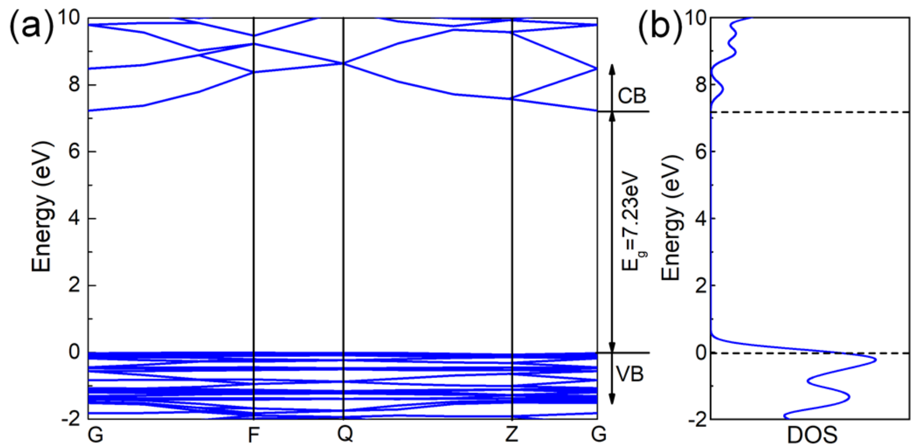


Fig. S3 Distribution of energy level structure and density of states (DOS) of perfect KDP crystal. (a) Energy level structure, (b) Total DOS.

To obtain the changes in optical property, the imaginary part peaks of the dielectric function of KDP crystals with different kinds of intrinsic defects was compared with the imaginary part of the perfectly configured KDP crystals shown in Fig. S4.

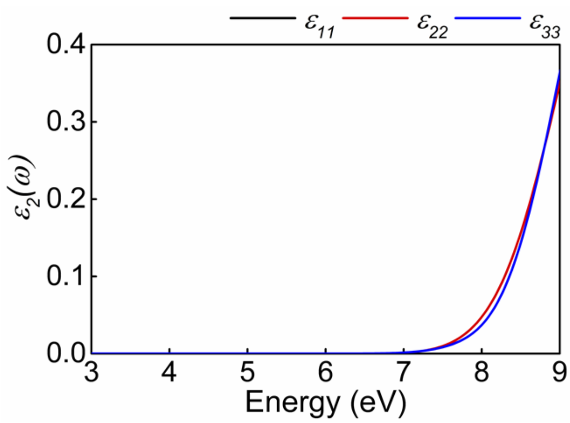


Fig. S4 Imaginary part of dielectric function of perfect KDP crystal.

4. The laser damage experiments

A Gaussian temporal shaped strong pulsed beam with a wavelength of 355nm and FWHM=3ns is output by the Nd: YAG laser. It transmits through the attenuator, polarizer, focusing lens, and finally acts on the front surface of the KDP crystals. The diagram of the laser damage light path is shown in Fig. S5. CCD is used to observe the surface damage morphology of the crystal in real time. An energy meter is used to measure the output energy.

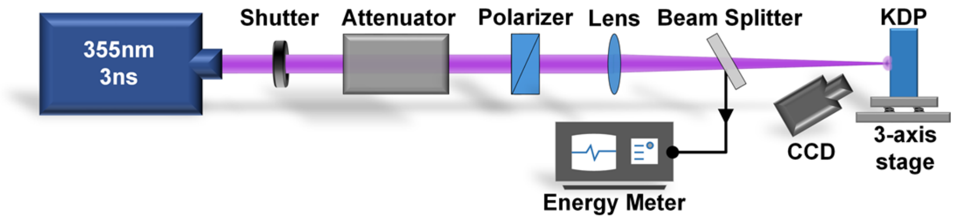


Fig. S5 The diagram of laser damage light path.