

# In-Plane Optical and Electrical Anisotropy of 2D Black-Arsenic

Mianzeng Zhong<sup>1</sup>, Haotong Meng<sup>1</sup>, Sijie Liu<sup>1</sup>, Huai Yang<sup>2</sup>, Wanfu Shen<sup>3</sup>, Chunguang

Hu<sup>3</sup>, Juehan Yang<sup>2</sup>, Zhihui Ren<sup>2</sup>, Bo Li<sup>4</sup>, Yunyan Liu<sup>5</sup>, Jun He<sup>1\*</sup>, Qinglin Xia<sup>1\*</sup>, Jingbo

Li<sup>6</sup>, and Zhongming Wei<sup>2\*</sup>

<sup>1</sup>Hunan Key Laboratory of Super-microstructure and Ultrafast Process, School of

Physics and Electronics, Central South University, Changsha 410083, Hunan, China

<sup>2</sup>State Key Laboratory of Superlattices and Microstructures, Institute of Semiconductors,

Chinese Academy of Sciences & Center of Materials Science and Optoelectronics

Engineering, University of Chinese Academy of Sciences, Beijing 100083, China

<sup>3</sup>State Key Laboratory of Precision Measuring Technology and Instruments, Tianjin

University, Tianjin 300072, China

<sup>4</sup>Department of Applied Physics, School of Physics and Electronics, Hunan University,

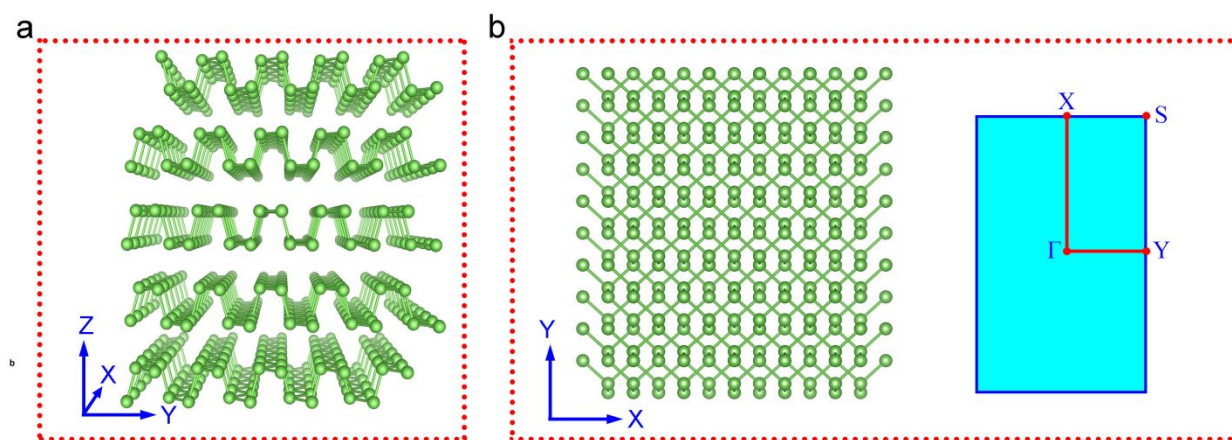
Changsha 410082, Hunan, China

<sup>5</sup>School of Physics and Optoelectronic Engineering, Shandong University of

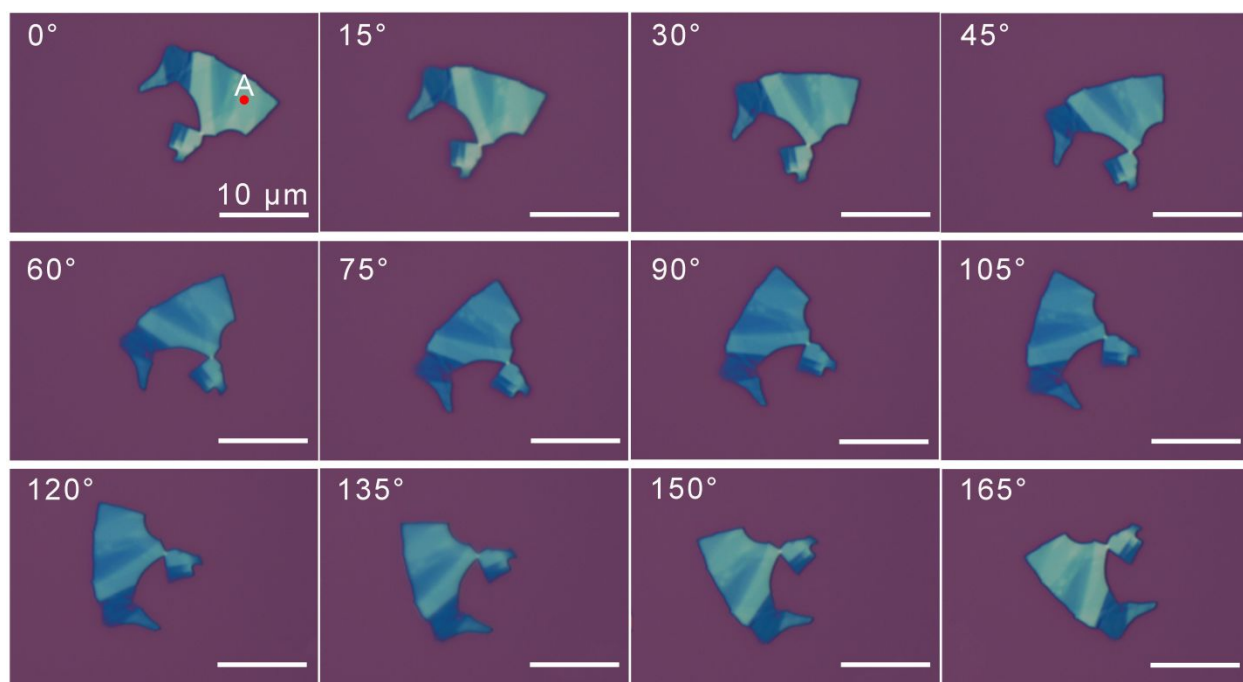
Technology, Zibo 255049, Shandong, China

<sup>6</sup>Institute of Semiconductors, South China Normal University, Guangzhou 510631,

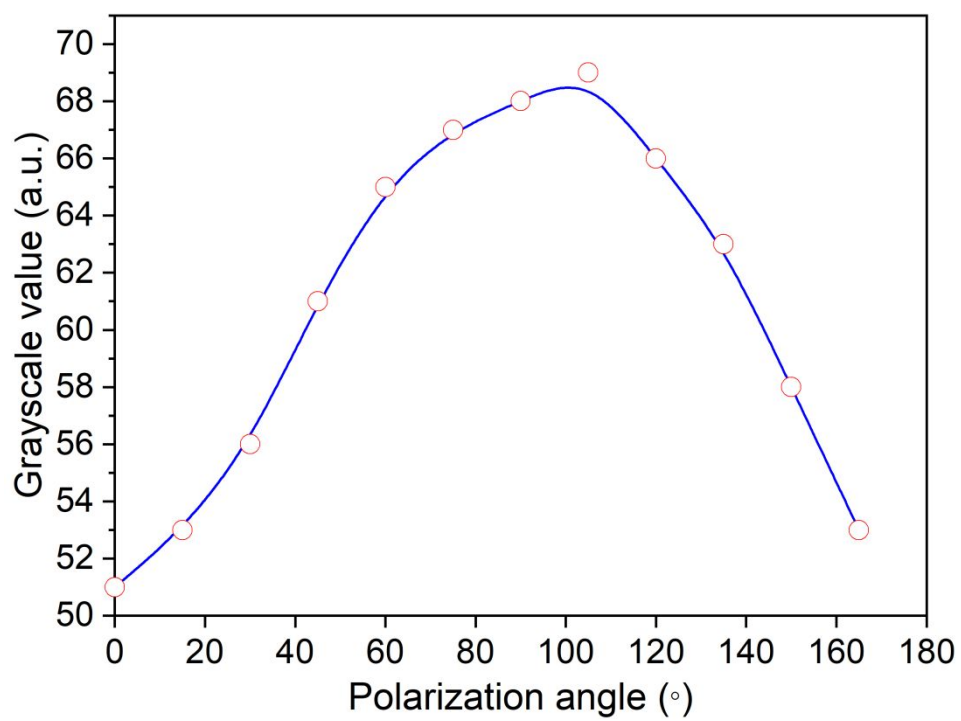
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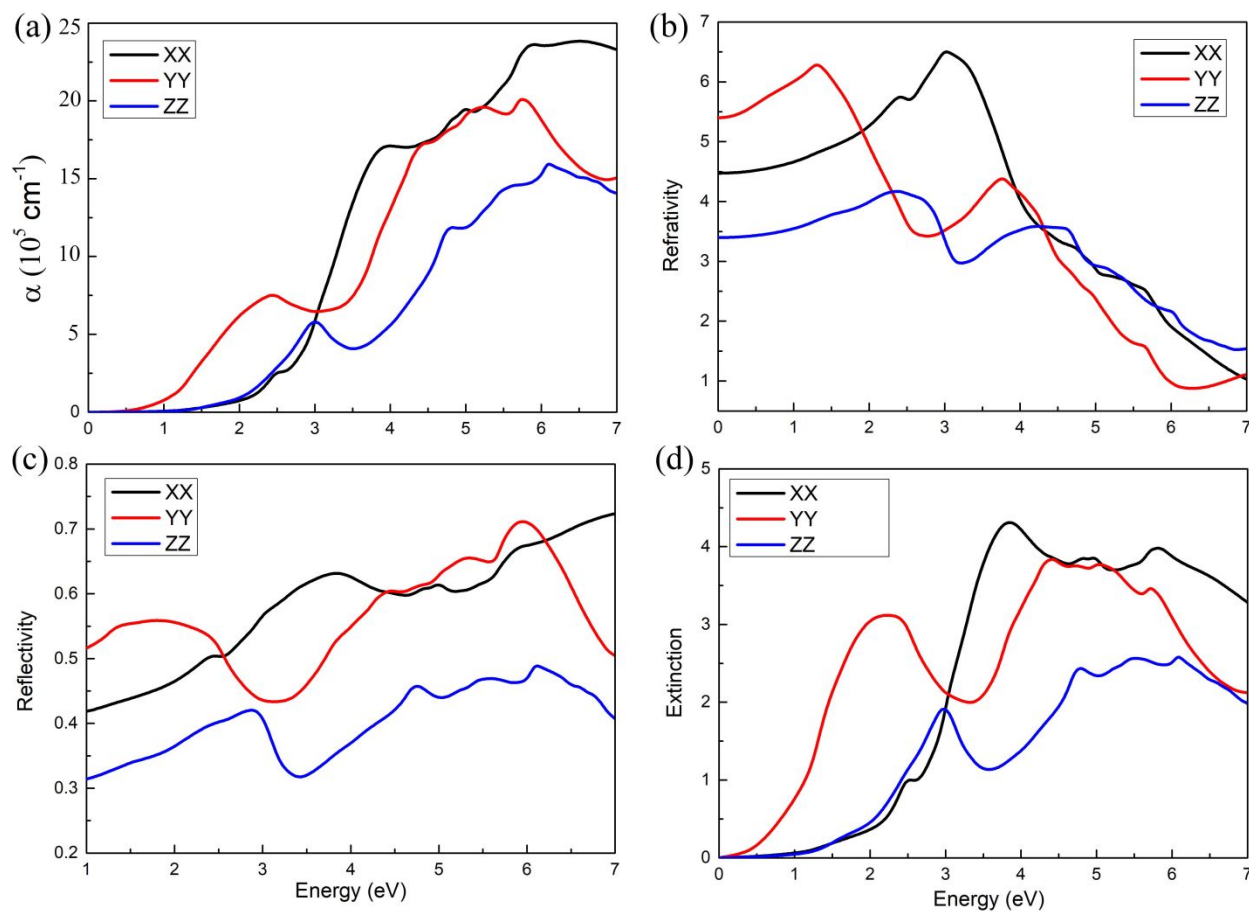
**Figure S1.** Crystal structures of orthorhombic b-As from (a) side view and (b) top view, showing its highly anisotropic nature.



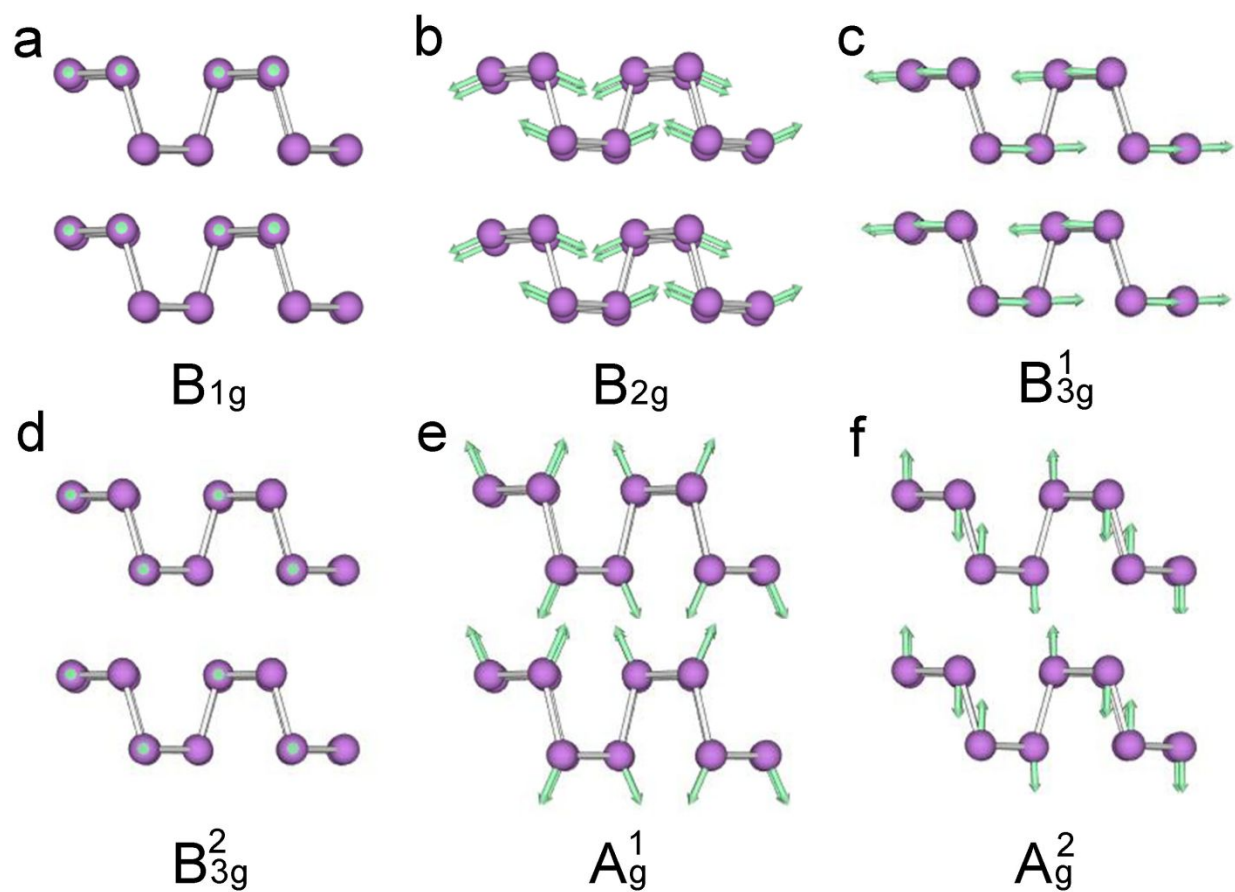
**Figure S2.** Polarization-resolved optical images of b-As flake.



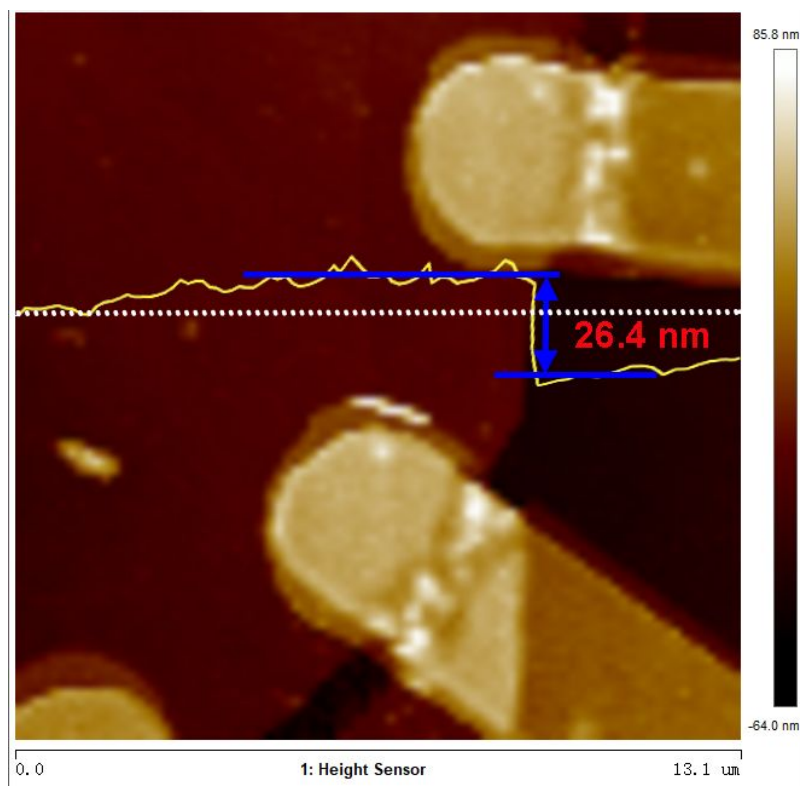
**Figure S3.** Grayscale values obtained from the A point in Figure S1 under different polarization angles as a function of the polarization angle.



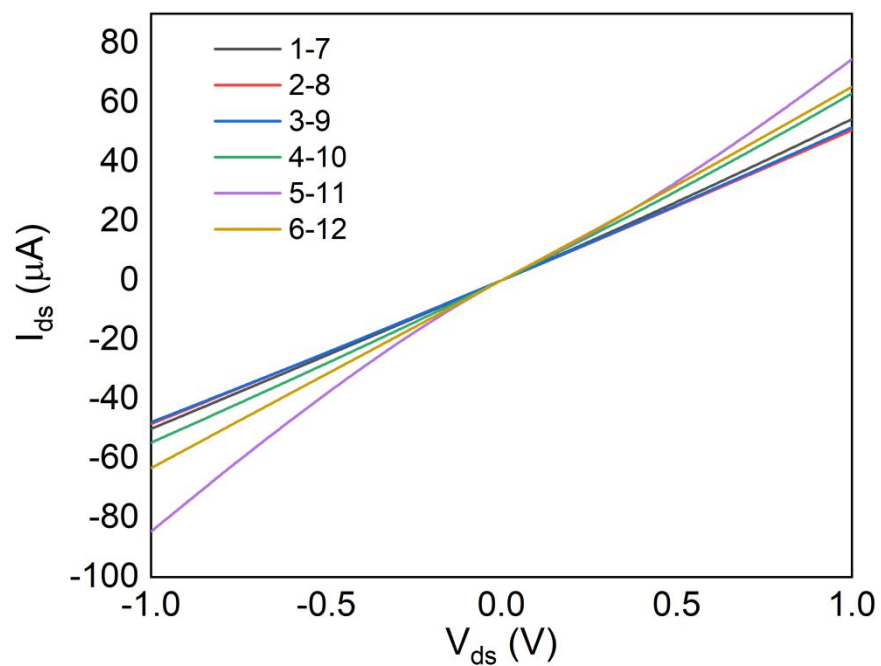
**Figure S4.** The absorption(a), reflectivity(b), refractive index (c), and extinction (d) for b-As considering XX, YY and ZZ directions.



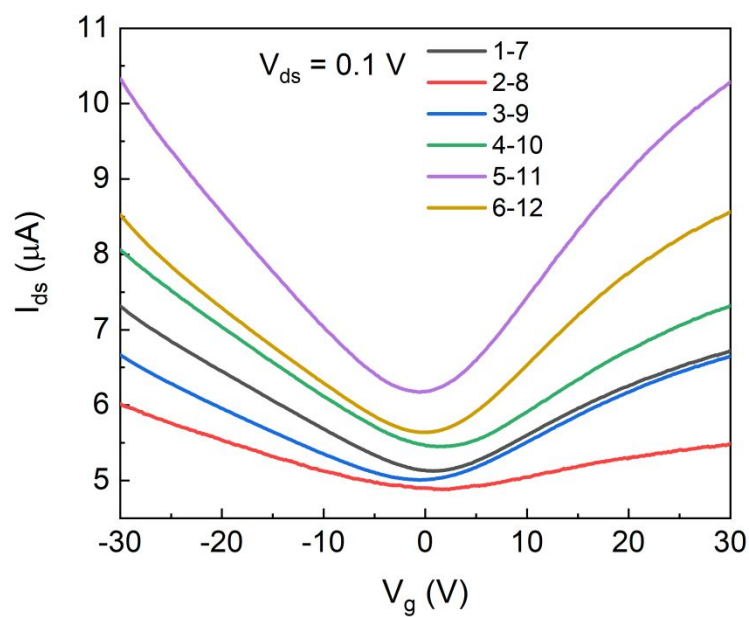
**Figure S5.** Schematic diagrams for six Raman-active modes in b-As. Green arrows indicate atomic displacements.



**Figure S6.** Atomic force microscopy (AFM) image of this typical b-As FET, the thickness of the b-As crystal is about 26.4nm.



**Figure S7.** Room-temperature output characteristics ( $I_{ds}$ - $V_{ds}$ ) of this FET with the different directions (different symmetrical source-drain electrodes), the gate voltage is 0V.



**Figure S8.** Room-temperature transfer characteristics ( $I_{ds}$ - $V_g$ ) of this FET with the different directions (different symmetrical source-drain electrodes), the drain-source voltage is 1V.

### Note S1

For b-As, the Raman tensors of the phonons with  $A_g$ , and  $B_{2g}$  symmetry are

$$\vec{R}_{A_g} = \begin{pmatrix} a & 0 & 0 \\ 0 & b & 0 \\ 0 & 0 & c \end{pmatrix} = \begin{pmatrix} |a|e^{i\phi_a} & 0 & 0 \\ 0 & |b|e^{i\phi_b} & 0 \\ 0 & 0 & |c|e^{i\phi_c} \end{pmatrix} \quad (S1)$$

and

$$\vec{R}_{B_{2g}} = \begin{pmatrix} a & 0 & f \\ 0 & 0 & 0 \\ f & 0 & 0 \end{pmatrix} = \begin{pmatrix} 0 & 0 & |f|e^{i\phi_f} \\ 0 & 0 & 0 \\ |f|e^{i\phi_f} & 0 & 0 \end{pmatrix} \quad (S2)$$

where  $|a|$ ,  $\phi_a$ ,  $|b|$ ,  $\phi_b$ ,  $|c|$ ,  $\phi_c$ ,  $|f|$ , and  $\phi_f$  are the magnitude and complex phase of the independent components of these two tensors, respectively. In our experimental configuration, the incident laser beam along the y direction (perpendicular to the layer plane of our arsenic sample). So, the polarization vector of the incident beam ( $\hat{e}_i$ ) can be expressed as  $\hat{e}_i = (\sin \theta \ 0 \ \cos \theta)$  ( $\theta$  is the sample rotation angle), and the polarization



vector of the scattered light ( $\hat{e}_s$ ) can be expressed as  $\hat{e}_s = (\sin \theta \ 0 \ \cos \theta)$  and  $\hat{e}_s = (\cos \theta \ 0 \ -\sin \theta)$  for the parallel and cross-polarization configurations, respectively. The Raman scattering intensity, which is related to the Raman tensor and the polarization vectors of incident laser beam and scattered light, is expressed as  $S \propto |\hat{e}_i \cdot \vec{R} \cdot \hat{e}_s|^2$ . As a consequence, the Raman intensities of  $A_{1g}$  and  $B_{2g}$  modes under parallel and cross-polarization configurations are written as following

$$S_{A_g}^{//} = (|a| \sin^2 \theta + |c| \cos \phi_{ca} \cos^2 \theta)^2 + |c|^2 \sin^2 \phi_{ca} \cos^4 \theta \quad (S3)$$

$$S_{A_g}^{\perp} = [(|a| - |c| \cos \phi_{ca})^2 + |c|^2 \sin^2 \phi_{ca}] \cos^2 \theta \sin^2 \theta \quad (S4)$$

$$S_{B_{2g}}^{//} = |f|^2 \sin^2 2\theta \quad (S5)$$

$$S_{B_{2g}}^{\perp} = |f|^2 \cos^2 2\theta \quad (S6)$$

where  $\phi_{ca}$  is the phase difference  $\phi_c - \phi_a$ . As described, the Raman intensity of  $A_{1g}$  and  $B_{2g}$  modes shows different periodic variation with the sample rotation angle under parallel and cross-polarization configurations of the scattered light. According to (3) and (5), under the parallel configuration, when the crystal orientation parallel to the scattered light, the Raman intensity of  $A_{1g}$  mode has maximum or minimum value, while  $B_{2g}$  mode

is forbidden. In contrast, under the cross-polarization configuration, the  $A_{1g}$  mode is forbidden, and Raman intensity of  $B_{2g}$  mode has the maximum value.