Supplementary Information:

## Anisotropic Raman spectral analysis and laser output based on large-sized Scheelite bismuth molybdate tungstate mixed crystals

Xiangxin Tian,<sup>\*,a</sup> Lijuan Chen,<sup>\*,b</sup> Peng Zhao,<sup>c</sup> Youxuan Sun,<sup>d</sup> and Zeliang Gao<sup>\*,d</sup>

<sup>a.</sup> School of Materials and Engineering, Linyi University, Linyi 276000, China.

<sup>b.</sup> School of Physics and Technology, University of Jinan, Jinan 250100, China.

<sup>c.</sup> Sinoma Synthetic Crystals Co., Ltd., Beijing 100018, China.

<sup>d</sup> State Key Laboratory of Crystal Materials, Shandong University, Jinan 250100, China.

Figure S1. EDS composition analysis based on the as-grown single crystal of BMO.

**Figure S2.** Polarized spontaneous Raman spectra of BMO crystal under different geometric configurations with the incident light propagating along the optical X principal axis. (a)-(d) represent the polarized Raman spectra under the X(YY)X, X(ZZ)X, X(YZ)X, and X(ZY)X configurations, respectively.

**Figure S3.** Polarized spontaneous Raman spectra of BMO crystal under different geometric configurations with the incident light propagating along the optical Y principal axis. (a)-(d) represent the polarized Raman spectra under the Y(XX)Y, Y(ZZ)Y, Y(XZ)Y, and Y(ZX)Y configurations, respectively.

**Figure S4.** Polarized spontaneous Raman spectra of BMO crystal under different geometric configurations with the incident light propagating along the optical Z principal axis. (a)-(d) represent the polarized Raman spectra under the Z(XX)Z, Z(YY)Z, Z(XY)Z, and Z(YX)Z configurations, respectively.

Table S1. EDS analysis results of the BMO mixed crystals

**Table S2.** XRF analysis results of the BMO mixed crystals.



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**Figure S4.** Polarized spontaneous Raman spectra of BMO crystal under different geometric configurations with the incident light propagating along the optical Z principal axis. (a)-(d) represent the polarized Raman spectra under the Z(XX)Z, Z(YY)Z, Z(XY)Z, and Z(YX)Z configurations, respectively.

Element	Weight%	Atomic%
Bi	37.00	8.29
Мо	28.03	13.68
W	9.09	2.32
Ο	25.87	75.71
Total	100	100

 Table S1. EDS analysis results of the BMO mixed crystals

**Table S2.** XRF analysis results of the BMO mixed crystals.

Element	Weight%	Atomic%
Bi	56.6	40.5
Мо	32.7	50.9
W	10.6	8.6
Total	100	100