$Ca_3(TeO_3)_2(MO_4)$ (M = Mo, W): Mid-Infrared Nonlinear Optical Tellurates with Ultrawide Transparency Ranges and Super-high Laser Induced Damage Thresholds

Qian Wu[1]*, Mingjun Xia[1], Rukang Li[1].
*lead presenter: wuqian910230@mail.ipc.ac.cn
1 Beijing Center for Crystal Research and Development, Key Laboratory of Functional Crystals and Laser Technology, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100190, China

Intense interests in mid-infrared (MIR) nonlinear optical (NLO) crystals have erupted in recent years due to the development of optoelectronic applications ranging from remote monitoring to molecular spectroscopy [1-5]. In recent years, a series of mid-infrared nonlinear optical crystals represented by barium tellurium molybdate [6] have been reported, most of which exhibit excellent photoelectric properties. Here, two polar crystals $Ca_3(TeO_3)_2(MO_4)$ (M = Mo, W) were grown from TeO_2-MO_3 flux by high temperature solution methods. $Ca_3(TeO_3)_2(MoO_4)$ and $Ca_3(TeO_3)_2(WO_4)$ are isostructural, which feature novel structures consisting of asymmetric MO4 tetrahedra and TeO3 trigonal pyramids. Optical characterizations show that both crystals display ultrawide transparency ranges (279 nm – 5.78 μm and 290 nm – 5.62 μm), especially high optical transmittance over 80% in the important atmospheric transparent window of 3-5 μm, and superhigh laser dam-age thresholds (1.63 GW/cm^2 and 1.50 GW/cm^2), 54.3 and 50 times larger than that of state-of-the-art MIR NLO $AgGaS_2$, respectively. Notably, they exhibit the widest transparency ranges and the loftiest laser induced threshold damages amongst the reported tellurates so far. Moreover, $Ca_3(TeO_3)_2(MO_4)$ exhibit type I phase-matching at two working wavelengths owing to their large birefringence and strong second-harmonic generation responses from the distorted anions, as further elucidated by the first-principles calculations. Above characteristics indicate that $Ca_3(TeO_3)_2(MO_4)$ crystals are high performance MIR NLO materials, especially applying in high power MIR laser operations.

References

[1] Petrov V. The coverage of the mid-infrared spectral range by solid-state laser sources. Opt. Mater. 2012;34:536-554.
[2] Pushkarsky MB et al. High-sensitivity detection of TNT. Proc. Natl. Acad. Sci. USA. 2006;103:19630.
[3] Ikuta R et al. Wide-band quantum interface for visible-to-telecommunication wavelength conversion. Nat. Commun. 2011, 2, 1544.
[4] Petrov V. Frequency down-conversion of solid-state laser sources to the mid-infrared spectral range using non-oxide nonlinear crystals. Prog. Quantum Electron. 2015; 42:1.
[5] Kang L et al. Frequency down-conversion of solid-state laser sources to the mid-infrared spectral range using non-oxide nonlinear crystals. J. Am. Chem. Soc. 2015;137:13049.
[6] Guo XJ et al. Recent Advances in Tellurite Molybdates/Tungstates Crystals. CrystEngComm 2022;24:7516-7529.