How To Identify Nonlinear Optical Crystals for Thz Generation

Sin Hang (Enoch) Ho[1], Gabriel A. Valdivia-Berroeta[2], Zachary B. Zaccardi[1], Sydney K. F. Pettit[1], Bruce Wayne Palmer[1], Matthew J. Lutz[1], Claire Rader[1], Brittan P. Hunter[1], Natalie K. Green[1], Connor Barlow[1], Coriantumr Z. Wayment[1], Daisy J. Harmon[1], Paige Petersen[1], Dr. Stacey J. Smith[1], Dr. David J. Michaelis[1], Jeremy A. Johnson[1]

[1]Brigham Young University, [2]Boehringer Ingelheim Inc

enochho201315@gmail.com

Nonlinear optical (NLO) crystals have the ability to convert light into different frequencies through nonlinear processes such as second harmonic generation and optical rectification. One of the fastest-growing applications of NLO materials is in generating terahertz (THz) radiation. Yet while the need for THz generating NLO materials is growing, only a few such NLO crystals have been developed. This is because the molecular building blocks of THz- generating NLO crystals must have strong hyperpolarizability yet also pack into a noncentrosymmetric space group so that the hyperpolarizability vectors do not cancel.
Predicting and achieving successful crystal packing for molecules with high hyperpolarizability has proven to be exceedingly difficult.
We are developing a new approach for identifying THz-generating organic NLO crystals which combines data mining with DFT calculation. Using the CSD API, we have written a custom code to search for noncentrosymmetric materials from the Cambridge Structural Database (CSD) that might exhibit the desired optical properties. From over 1 million materials in the CSD, 15,782 compounds were isolated based on our selection criteria. All the resulting compounds were submitted for DFT hyperpolarizability calculation. In addition, a crystal packing order parameter was calculated for each structure to indicate the degree to which packing may be optimal for NLO applications. We selected four of the most promising candidates to synthesize in the lab: PNPA, ZPAN, NMBA and TMOAT (as shown in fig. 1). Each were characterized using single-crystal X-ray diffraction to identify the structures and main crystal faces. All four proved to be amenable to large crystal growth, and the main crystal faces for three of the four proved optimal for NLO applications. The developed crystals were all tested for NLO activity via standard electro-optic measurement using a Ti:sapphire laser system with optical parametric amplifier. All exhibit THz generation ability (as shown in fig.2), thus our novel approach for identifying new THz-generating NLO materials seems to be highly effective.

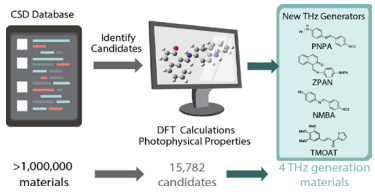

Figure 1

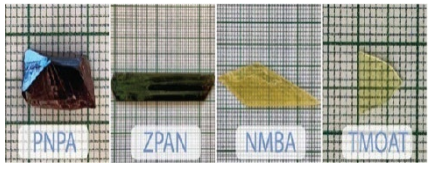

Figure 2

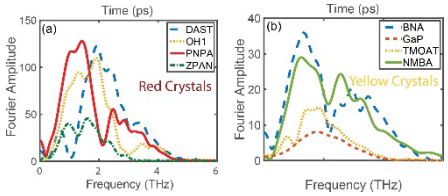

Figure 3