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Wide-Bandgap Organic Crystals: Enhanced Optical-to-Terahertz Nonlinear Frequency Conversion at Near-Infrared Pumping

Deokjoong Kim, Won Tae Kim, Jae-Hyun Han, Ji-Ah Lee, Seung-Heon Lee, Bong Joo Kang, Mojca Jazbinsek, Woojin Yoon, Hoseop Yun, Dongwook Kim, Stein van Bezouw, Jochen Campo, Wim Wenseleers, Fabian Rotermund, and O-Pil Kwon**

Enhanced terahertz (THz) wave generation is demonstrated in nonlinear organic crystals through refractive index engineering, which improves phase matching characteristics substantially. Unlike conventional low-bandgap nonlinear organic crystals, the newly designed benzimidazolium-based HMI (2-(4-hydroxy-3-methoxystyryl)-1,3-dimethyl-1H-benzoimidazol-3-ium) chromophore possesses a relatively wide bandgap. This reduces the optical group index in the near-infrared, allowing better phase matching with the generated THz waves, and leads to high optical-to-THz conversion. A unique feature of the HMI-based crystals, compared to conventional wide-bandgap aniline-based crystals, is their remarkably larger macroscopic optical nonlinearity, a one order of magnitude higher diagonal component in macroscopic nonlinear susceptibility than NPP ((1-(4-nitrophenyl)pyrrolidin-2-yl)methanol) crystals. The HMI-based crystals also exhibit much higher thermal stability, with a melting temperature T_m above 250 °C, versus aniline-based crystals (116 °C for NPP). With pumping at the technologically important wavelength of 800 nm, the proposed HMI-based crystals boost high optical-to-THz conversion efficiency, comparable to benchmark low-bandgap quinolinium crystals with state-of-the-art macroscopic nonlinearity. This performance is due to the excellent phase matching enabled by decreasing optical group indices in the near-infrared through wide-bandgap chromophores. The proposed wide-bandgap design is a promising way to control the refractive index of various nonlinear organic materials for enhanced frequency conversion processes.

1. Introduction

Research interest in broadband coherent sources in the terahertz (THz) frequency range is growing rapidly due to advanced applications in THz spectroscopy and imaging, as well as fundamental studies of the ultrafast dynamics of diverse materials, and nonlinear THz photonics.[1–5] To date, designing and developing highly efficient THz wave generators remains challenging.[4,6–10] Nonlinear optical organic crystals which exploit either optical rectification (OR) or difference frequency generation (DFG) processes have been reported to provide excellent optical-to-THz conversion.[7,11–18] For example, benchmark nonlinear organic crystals with state-of-the-art macroscopic optical nonlinearity have demonstrated high THz wave generation efficiencies of up to a few percent when pumped by infrared (IR) pulses in the wavelength range of 1200–1600 nm.[13–17] An alternative to employing pump sources in this range is to pump at a near-IR wavelength near 800 nm. This is a very important practical wavelength and is available with widespread femtosecond Ti:sapphire

D. Kim, J.-H. Han, J.-A. Lee, Dr. S.-H. Lee, Prof. O-P. Kwon
Department of Molecular Science and Technology
Ajou University
Suwon 443-749, Korea
E-mail: opilkwon@ajou.ac.kr

W. T. Kim, Dr. B. J. Kang, Prof. F. Rotermund
Department of Physics
Korea Advanced Institute of Science and Technology (KAIST)
Daejeon 34141, Korea
E-mail: rotermund@kaist.ac.kr

Dr. M. Jazbinsek
Institute of Computational Physics
Zurich University of Applied Sciences (ZHAW)
Winterthur 8401, Switzerland

W. Yoon, Prof. H. Yun
Department of Chemistry and Department of Energy Systems Research
Ajou University
Suwon 443-749, Korea

Prof. D. Kim
Department of Chemistry
Kyonggi University
San 94-6, Iui-dong, Yeongtong-gu, Suwonsi, Gyeonggi 443-760, Korea

S. van Bezouw, Dr. J. Campo, Prof. W. Wenseleers
Department of Physics
University of Antwerp
Campus Drie Eiken (CDE)
Universiteitsplein 1, Wilrijk B-2610, Belgium

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