Improved Terahertz Generation Through Heterogenous Multi-Layered Organic Crystal Structures

Aldair Alejandro¹, Daisy J. Ludlow¹, Paige K. Petersen¹, Kayla M. Holland¹, Fatoumata N'diaye¹, Tanner Manwaring¹, David J. Michaelis¹ and Jeremy J. Johnson¹

¹Brigham Young University, Provo, UT, 84602 USA

Abstract—Yellow organic crystals, like BNA, MNA and NMBA can be used to produce terahertz (THz) light via optical rectification of ultrafast laser pulses. In this work, we create new multi-layered "sandwich" structures with these yellow crystals by (1) fusing them to sapphire plates that allow the crystal to withstand higher laser fluences and (2) using MBBA liquid crystal to improve THz output efficiency through refractive-index matching in the layers of the structure. We show that the sapphire plates significantly increase the damage threshold of these yellow organic crystals by a factor of two or more. We also show that the THz output efficiency is further increased by using a multi-layered sandwich structure with an exit liquid crystal layer. In some cases, we show that the sandwich structure increases the THz intensity by more than a factor of two.

I. INTRODUCTION

Nonlinear yellow organic crystals, such as BNA [1,2], MNA [3], and NMBA [4] can generate terahertz (THz) light. These yellow organic crystals have some unique advantages over orange-red organic THz generation crystals, such as DAST, DSTMS, and OH-1, which are optimally pumped with longer wavelengths (1300-1600 nm) from an optical parametric amplifier (OPA). In the visible region of the electromagnetic spectrum, the yellow crystals have lower absorption cutoffs, which allows us to not only pump them with the longer wavelengths derived from an OPA, but we can also generate THz using 800 nm light directly from the laser. However, at 800 nm the yellow crystals cannot be pumped with high laser fluences without being severely damaged due to their low melting points and two-photon absorption-based heating.

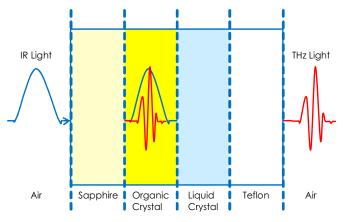


Fig. 1. Diagram of the heterogenous multi-layer organic crystal structure for improved THz output. The IR light travels through sapphire and then into the yellow organic crystal directly fused to the sapphire. THz is generated via optical rectification in the organic crystal, and the newly generated THz light exits the crystal, traveling through a thin layer of liquid crystal MBBA (N-(4-methoxybenzylidene)-4-butylanilinene), then a 1.5 mm Teflon layer before exiting the structure into the air.

In recent work [2], we showed how to circumvent these heating

issues by fusing BNA crystals to sapphire plates with high thermal conductivity, which disperses laser heat and allows the crystal to withstand higher fluences. This increase in damage threshold allows the crystals to attain greater THz electric fields. BNA crystals fused to sapphire were able to attain damage thresholds of ~12 mJ/cm² compared to ~4 mJ/cm² for the bare BNA crystals [2].

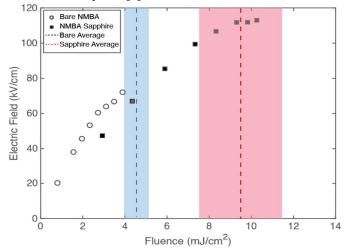


Fig. 2. The electric field strength in kV/cm as a function of the laser fluence in mJ/cm² for NMBA crystals. The blue dotted line shows the average damage threshold for three bare NMBA crystals and the shaded blue region shows its standard deviation. The red dotted line is the average damage threshold for three NMBA crystals fused to sapphire and the shaded red region shows its standard deviation. The open circles show generated peak field strengths for one bare NMBA crystal, while the filled squares show the trend for one NMBA crystal fused to sapphire.

Another way to improve THz output in organic crystals is to reduce reflective losses as the THz exits the generation crystal. We showed that, multi-layered "sandwich" structures with organic DAST crystals minimized the reflective losses usually present when the generated THz exits the organic crystal and propagates back into air [5]. For these structures, a layer of liquid crystal MBBA (N-(4-methoxybenzylidene)-4-butylanilinene), acts as a low THz absorption index-matching fluid between both the organic crystal and a Teflon filter, significantly improving the THz output.

In this work, we created improved sandwich structures by (1) fusing yellow organic crystals (BNA, MNA and NMBA) to sapphire plates and (2) adding a layer of liquid crystal MBBA between the organic crystal and the Teflon filter. These improved structures were designed to both withstand high laser fluences by increasing the damage threshold and to have higher THz output by reducing reflective losses.

II. RESULTS

The new sandwich structures were assembled with the following layers (as shown in Fig. 1): a yellow organic crystal

was fused to sapphire (0.5 mm), and then the liquid crystal MBBA was placed between the THz generator, and a Teflon filter (1.5 mm). The three yellow organic crystals (BNA, MNA and NMBA) were fused to sapphire plates, where the sapphire

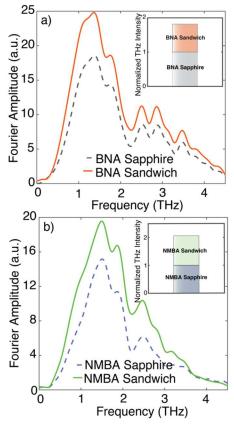


Fig. 3. The Fourier amplitude as a function of frequency of both the presandwich structure (dotted line labeled as "sapphire") and the full sandwich structure (solid line labeled as "sandwich") for the yellow organic crystals BNA (a) and NMBA (b). The insets show the normalized THz intensities that compares the "sandwich" (top bar) to the "sapphire" structure.

acts as the first layer for the incident IR light.

We first characterized these structures by performing damage threshold measurements at 800 nm on MNA and NMBA to confirm that the same advantages of fusing BNA to sapphire hold for MNA and NMBA as well. To measure any improvement, we first determined the damage threshold on bare crystals and then on crystals fused to sapphire. In Figure 2, we show the THz electric field strength measured as a function of laser fluence for bare NMBA crystals and NMBA crystals fused to sapphire. The damage threshold of the bare NMBA crystals was ~4.5 mJ/cm² (blue region), while it was ~9.5 mJ/cm² (red region) for the crystals fused to sapphire. This shows that the NMBA crystal fused to sapphire can withstand double the laser fluences compared to its bare counterpart, which leads to higher output THz electric fields (~1.6× higher field-strengths).

To demonstrate the improved THz generation efficiency of the complete new sandwich structures with index-matching exit layers, we measured the THz output with the yellow crystals only fused to sapphire, and then again when they were made into a sandwich structure. The crystals were pumped with the same laser fluence and 1250 nm light, as this is the wavelength where the yellow crystals are more efficient at converting IR into THz light.

Figures 3a and 3b show a comparison of the THz frequency spectra of the crystals fused to sapphire (labeled as "sapphire") and of the sandwich structure (labeled as "sandwich") for BNA and NMBA, respectively. In either of these structures, we observe that the Fourier amplitude of the sandwich structure (solid line) is higher than the crystal fused to sapphire (dotted line) between 0.5 and 4.5 THz. In the insets of Figures 3.a and 3.b, the area under the Fourier amplitudes was integrated, converted into THz intensity and normalized to the crystalsapphire value, allowing us to quantify the THz output efficiency of the full sandwich structure compared to that of the crystal only fused to sapphire. In Figure 3.a, we observe that the BNA sandwich structure is ~1.9 times more efficient than the BNA crystal fused to sapphire. In Figure 3.b, we see that the NMBA sandwich doubles the THz intensity compared to the NMBA crystal fused to sapphire. Although not shown, improvements in the THz intensity profile for MNA sandwich structures were also observed.

III. SUMMARY

In conclusion, we show that we can improve the THz output efficiency of yellow organic crystals (MNA, BNA and NMBA) by creating a multi-layer structure that reduces reflective losses by matching the refractive indices. The refractive index of the MBBA liquid crystal aids in this because it is a good THz-index matching fluid with minimal THz absorption. These structures also have the advantage of having higher damage threshold due to the yellow crystals being fused to sapphire plates, enabling the use of higher 800-nm laser fluences for maximum THz output.

REFERENCES

[1]. I.C. Tangen, G.A. Valdivia-Berroeta, L.K. Heki, Z.B. Zaccardi, E.W. Jackson, C.B Bahr, S Ho, D.J. Michaelis and J.A. Johnson, "Comprehensive characterization of terahertz generation with the organic crystal BNA," Journal of the Optical Society of America B, vol. 38, pp. 2780-2785, Sept., 2021 [2]. Z.B. Zaccardi, I.C. Tangen, G.A. Valdivia-Berroeta, C.B Bahr, K.C. Kenney, C. Rader, M.J. Lutz, B.P. Hunter, D.J. Michaelis and J.A. Johnson, "Enabling high-power, broadband THz generation with 800-nm pump wavelength," Optics Express, vol. 29, pp. 38084-38094, Nov., 2021 [3]. B.W.H. Palmer, C. Rader, E.S. Ho, Z.B. Zaccardi, D.J.H. Ludlow, N.K. Green, M.J. Lutz, A. Alejandro, M.F. Nielson, G.A. Valdivia-Berroeta, C.C. Chartrand, K.M. Holland, S.J. Smith, J.A. Johnson and D.J. Michaelis, "Large Crystal Growth and THz Generation Properties of 2-Amino-5-Nitrotoluene (MNA)," ACS Applied Electronic Materials, vol. 4, pp. 4316-4321, Aug., 2022 [4]. G.A. Valdivia-Berroeta, Z.B. Zaccardi, S.F. Pettit, S. Ho, B.W. Palmer, M.J. Lutz, C. Rader, B.P. Hunter, N.K. Green, C. Barlow, C.Z. Wayment, D.J. Ludlow, P. Peterson, S.J. Smith, D.J. Michaelis and J.A. Johnson, "Data Mining for Terahertz Generation Crystals." Advanced Materials, vol. 34. pp. 2107900, March, 2022

[5]. C.B. Bahr, N.K. Green, L.K. Heki, E. McMurray, I.C. Tangen, G.A. Valdivia-Berroeta, E.W. Jackson, D.J. Michaelis and J.A. Johnson, "Heterogeneous layered structures for improved terahertz generation," *Optics Letters*, vo. 45, pp. 2054-2057, April, 2020