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Data Mining for Terahertz Generation Crystals

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A data mining approach to discover and develop new organic nonlinear optical crystals that produce intense pulses of terahertz radiation is demonstrated. The Cambridge Structural Database is mined for non-centrosymmetric materials and these structural data are used in tandem with density functional theory calculations to predict new materials that efficiently generate terahertz radiation. This enables us to (in a relatively short time) discover, synthesize, and grow large, high-quality crystals of four promising materials and characterize them for intense terahertz generation. In a direct comparison to the current state-of-the-art organic terahertz generation crystals, these new materials excel. The discovery and characterization of these novel terahertz generators validate the approach of combining data mining with density functional theory calculations to predict properties of high-performance organic materials, potentially for a host of exciting applications.

1. Introduction

The discovery and design of advanced solid materials with useful properties and applications are essential for the advancement of many fields, including spectroscopy, catalysis, electron transport, energy storage and release, and air and water purification. As a subset in solid materials research, organic materials provide a powerful advantage in that desired material properties can be finely tuned by editing the structure of molecular building blocks. For organic crystals, the function of the material is governed by the chemical and photophysical properties of the molecular building blocks, along with molecular packing, molecular orientation in the crystal, and specific surface geometries. Therefore, the optimization of organic material properties is complicated by the need to control both molecular properties and solid-state packing preferences. Materials databases, like the Cambridge Structural Database (CSD), compile and make available a wealth of structural information for materials that have been developed for specific applications.[1] One potential use of such databases is to identify already existing materials that may be ideal for applications other than their original intended use. The idea that one can easily mine information about known materials for the development of new and extremely useful purposes will rapidly accelerate the discovery of new materials for many applications. This data mining approach also gives rise to new screening methodologies in the rapidly growing field of materials informatics.[2–7]

Various data mining approaches have been reported that classify material properties for a host of applications.[8–12] In addition, while the data mining and classification of inorganic materials are increasingly common, approaches to identify organic solids are rare.[13–15] Furthermore, most of these reports for organic and inorganic materials give only theoretical predictions and often lack experimental validation. For example, in Ref. [14], Geilhufe et al. used data mining in combination with first-principles calculations of the density of states to identify new candidate high-T_c superconductors. However, no candidate materials were actually tested for superconductivity. In this report, we combine data mining of known organic materials from the CSD with computational analysis of key molecular properties to identify new candidate organic materials for intense terahertz (THz) generation. We then validate our combined data mining and computational approach to materials discovery by synthesizing and fully characterizing four new THz-generating organic materials.

Terahertz radiation, with frequencies from 1 to 10 THz (wavelengths of 300–30 μm), exhibits unique interactions with many materials and thus THz light is able to analyze and control material properties in ways that differ from other forms of radiation. Many emerging and potentially disruptive applications of THz spectroscopy are taking advantage of these unique interactions, including in bioimaging and security,[16] chemical recognition,[17] nondestructive chemical monitoring in industry and food processing,[18,19] and wireless communication and high-speed computational devices.[20] Of the various methods available for generating THz light, optical rectification of

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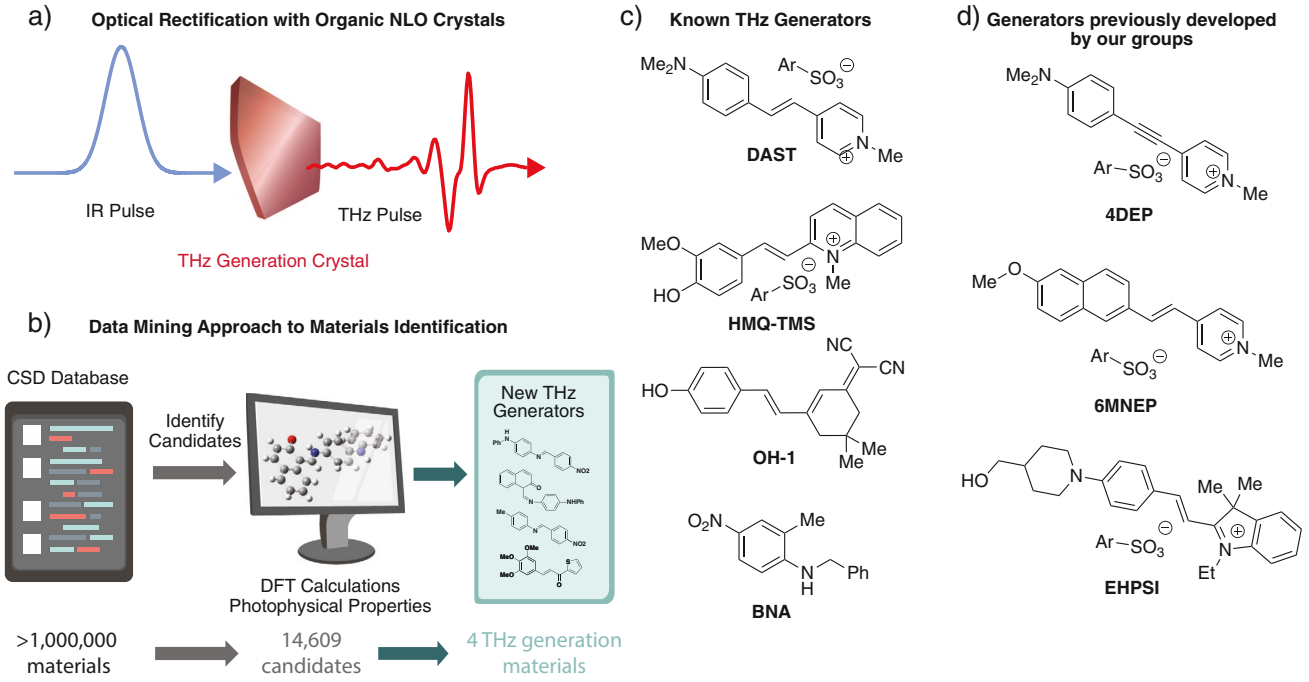

Figure 1. a) Optical rectification scheme for conversion of IR light to THz frequency radiation. b) Combined data mining/computational approach to identification of new organic NLO crystals for intense THz generation. c) Structure of known, state-of-the-art organic NLO crystals. d) Organic NLO crystals previously developed by our groups via modification of known generator structures.

infrared (IR) light with organic nonlinear optical (NLO) crystals is the most efficient method to produce high-intensity THz fields with extremely broad bandwidths (**Figure 1**a).[21–28] NLO crystals made up of organic molecular building blocks hold significant promise for THz generation due to the ease of editing and creating new organic chromophores with custom properties. However, very few efficient organic NLO crystals for THz generation have been reported and even fewer crystals are commercially available.[24,27,29,30] This currently limits the potential applications of high-power THz spectroscopy. The discovery of novel, high-performing organic NLO materials will impact the efficiency of intense THz generation and will open and expand avenues for NLO applications that utilize harmonic generation, optical parametric amplification, optical phase conjugation, and electro- and acousto-optic modulation.

The classical approach to develop new organic materials for intense THz generation is a tedious process that involves both molecular design, crystal structure determination, and crystal growth optimization. The first step involves the identification of new organic chromophores with ideal photophysical properties. Promising molecules must then form large single crystals with a suitable molecular packing that aligns chromophores in a parallel fashion. In this work, we greatly accelerate this development process by using structural data mining tools to identify known organic materials from the CSD with ideal non-centrosymmetric crystal properties for THz generation (Figure 1b). We then perform first-principles calculations of the core molecular property that influences nonlinear optical properties (hyperpolarizability), and we inspect crystal packing to rank the molecular crystals based on their potential for THz generation. Through these efforts, we identify several promising organic materials with previously unknown THz generation properties and report the synthesis and crystal growth of large, high-quality crystals for four candidates that demonstrate the usefulness of this strategy. We also characterize these four new materials for their THz generation capabilities and compare with benchmark organic THz generators. Our new organic material (E)-4-((4-nitrobenzylidene)amino)-*N*-phenylaniline (PNPA) shows exceptional THz generation properties that exceed current state-of-the-art red-orange organic THz generators such as DAST and OH-1 (Figure 1c).[24] We also discover a yellow THz generator (E)-4-((4-methylbenzylidene)amino)-*N*-phenylaniline (NMBA) that compares favorably with a yellow THz generator BNA, which has recently gained attention as a new standard for THz generation.[29,31–33]

2. Results and Discussion

Previous approaches to develop organic NLO crystals from the authors[25–28,34] and others[21,35–44] have shown that the process to identify, crystalize, and develop new organic THz generators can take years. Even when these efforts are successful in accessing new NLO crystals, the improvements in THz output are often small compared to benchmark organic crystals like DAST, HMQ-TMS, OH-1, and BNA (Figure 1c). Our previous efforts in organic NLO design have resulted in the development of new THz generation crystals, including 4DEP,[26] P-BI,[25] EHPSI-4NBS,[27,30] and 6MNEP[28] (Figure 1d). The new chromophores 4DEP, EHPSI, and 6MNEP resulted from modifications to the known structure of NLO crystals P-BI and DAST. EHPSI, the highest performing of the three, outperforms state-of-the-art

NLO crystal OH-1 by generating stronger peak THz electric fields with a broader spectrum.[30] However, EHPSI required years of development and only resulted in a ≈1.2 × improvement in THz output. In addition, our previous work, as with the most recent work in THz crystal development, relied on modification of previously known chromophores for THz generation and did not lead to the discovery of new classes of THz generating chromophores.[25–28,30] In contrast, our data mining efforts reported here have led to the rapid discovery and development of four new THz generators with unique chromophores in a short amount of time (<1 year) that rival the efficiency of previously reported generators.

To optimally generate THz light, organic crystals must possess two key parameters: the molecular units of the crystal must have high hyperpolarizability and the molecules must align in the crystalline state in a non-centrosymmetric head-to-tail arrangement. Hyperpolarizability is a molecular property that relates to how the dipole moment of a molecule responds or is polarized by the electric field of incoming pump light. Molecules that contain highly conjugated systems of electrons often have high hyperpolarizability because of the large size of the polarizable electron cloud. In this study, we use DFT calculations to quantify the hyperpolarizability of candidate molecules as an additional screening method for identifying the most promising candidates for THz generation. The second requirement is that molecules pack in a perfectly parallel head-to-tail arrangement in the crystal state that aligns their hyperpolarizability vectors. Because the CSD contains crystal structures of organic materials, we can screen candidate organic molecules for THz generation based on large hyperpolarizabilities and their crystalline alignment. The combination of these two properties allows us to identify highly promising new organic materials for THz generation.

Our initial data-mining efforts to identify new organic materials involved the use of an automated search of the CSD for known organic crystals. As noted above, our search focused on compounds that contain conjugated π-systems, and thus likely possess high molecular hyperpolarizability. A custom python program was developed to search the CSD 2020.0 database, which includes over 1 million materials; the python script included several filters to find organic materials with optimal properties. First, due to the ease of processing neutral organic crystals (such as BNA) compared to ionic organic crystals (like DAST), we only searched for neutral molecules with C, N, S, O, P, F, Cl, or Br atoms. Only materials with r refinement values < 15% were considered, indicating a reliable X-ray structure. These two filters resulted in 241 219 candidate entries. Non-centrosymmetric crystals were selected in the next stage because of the asymmetric molecular packing required for NLO applications (giving 66 661 entries). Relatively small molecules (<600 g mol^{-1}) were considered for the following stage to keep the number density in the crystal packing as high as possible (63 067 entries). Following this same consideration, crystal structures with only one molecular species were considered, ruling out cocrystals (52 382 entries). We also removed chiral compounds because many non-centrosymmetric chiral structures have poor alignment of THz generating chromophores (16 679 remaining candidates). One final round of filtering removed molecules with more than one structure file in the database, and we kept the most recent entry.

The chromophores from the resulting 14 609 compounds were submitted for DFT hyperpolarizability calculations and ranked accordingly (see Supporting Information for details). Previous data mining approaches for materials discovery that combined calculations of materials properties used calculations of electronic properties such as bandgap.[45] To understand THz generation potential, we needed to calculate advanced (nonlinear) electronic properties, in particular, the hyperpolarizability. This ability to combine calculations of advanced properties with data mining will benefit every realm of materials development because it demonstrates that any computationally accessible molecular property can be used in materials screening. Finally, together with our molecular calculations of hyperpolarizability, we calculated the molecular packing order parameter $\cos^3(\theta_p)$, where θ_p is the angle between the polar axis and the molecular hyperpolarizability vector in the crystalline state. Crystal structures with order parameter of 0 have no NLO activity and structures with order parameter 1 exhibit the ideal head-to-tail alignment of acentric molecular building blocks. Crystal packing with an order parameter close to one can lead to the largest nonlinear susceptibility and highest THz generation efficiency.

Molecular crystals with good crystal packing in tandem with molecular hyperpolarizability (β_{tot}) greater than 50×10^{-30} esu were identified as promising for organic synthesis. **Figure 2** shows the most promising molecules identified via our data mining approach (see .csv file in Supporting Information for a complete list of CSD IDs, calculated hyperpolarizabilities, and order parameters). Many of these chromophores have dramatically higher calculated hyperpolarizabilities than the commercial state-of-the-art crystal OH-1 ($\beta_{tot} = 93 \times 10^{-30}$ esu)[35], and the crystal structures exhibit good molecular alignment. In addition, many of these molecules contain unique chromophores for THz generation that have not previously been explored. While some of the materials identified were first developed for nonlinear optical applications, many were developed for completely different purposes. This latter result highlights the importance of our data mining approach in identifying completely new molecular structures and chromophores for the discovery of THz generating materials.

From our list of potential THz generators, we selected molecules with a range of different hyperpolarizabilities (all greater than 50×10^{-30} esu) and order parameters to determine how closely the actual THz generation properties matched our calculated values. The molecules in Figure 2 were specifically chosen based on their ease of synthesis (1–2 chemical steps from commercial materials). Nine of these molecules were synthesized based on known literature procedures and attempts were made to grow large single crystals of each molecule. Through these efforts, we were able to grow large (>5 mm × 5 mm) single crystals of four different molecules with a range of expected THz generation efficiencies, including PNPA, ZPAN, NMBA, and TMOAT (see Supporting Information for full synthesis and crystallization details). Determining optimal conditions for crystallization of large, high-quality crystals is still one of the longest steps in the process, taking potentially 2–3 months for each material. Our combined data mining and computational approach help us isolate extremely promising materials so that

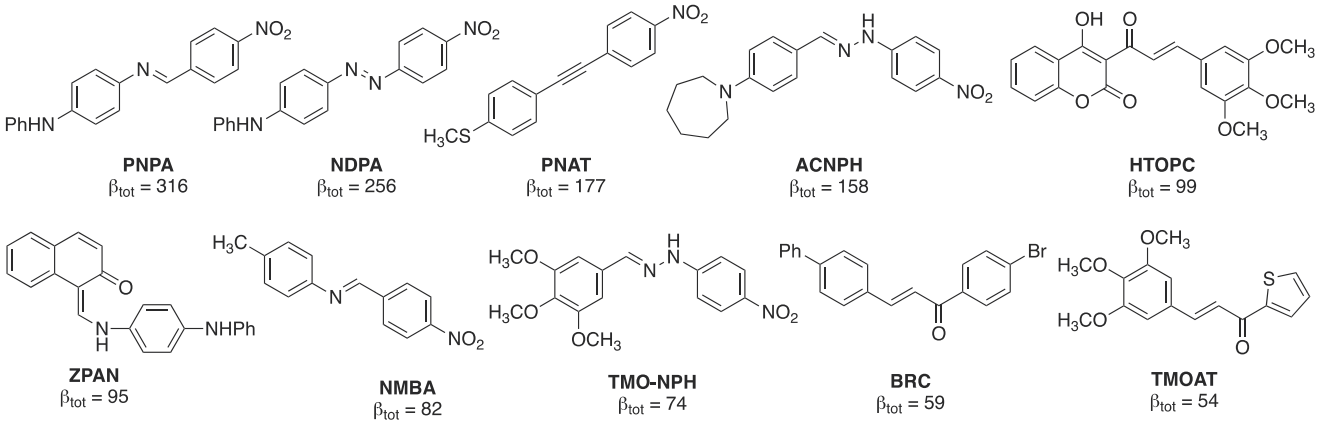

Figure 2. Newly identified THz generation molecules with calculated molecular hyperpolarizabilities (β_{tot}).

time is not wasted developing crystallization growth procedures for low-performing THz generators.

Large crystals were grown via slow evaporation protocols and representative examples of crystals grown for each molecule are shown in **Figure 3**a. X-ray crystallographic analysis was performed on each crystal to verify that the structures of the grown crystals matched the previously reported structures in the database. See Table S1 in the Supporting Information for details on X-ray analysis. The structures of all four crystals are consistent with the reported structures in the database. Figure 3b shows the crystal unit cell for each of the four crystals with the blue arrows indicating the direction of hyperpolarizability. It is important to note that PNPA has perfect alignment of hyperpolarizability vectors (order parameter of ~1.0), which maximizes its potential for THz generation. We then measured the THz generation spectrum for each crystal and compared their THz generation efficiency. The unique Fourier spectrum (normalized) for each crystal is shown in Figure 3c. Importantly, each new crystal shows THz generation at 1450 nm irradiation and can generate intense THz pulses.

Our final goal was to compare the THz efficiency and spectra of our four new THz generators to known, state-of-the-art organic NLO crystals. We separated the crystals into two different groups based on color (red crystals and yellow crystals). In the red crystal category, we compared THz generation from an 850 µm thick PNPA crystal and a 600 µm thick ZPAN with industry-standard DAST (470 µm) and OH-1 (300 µm) by irradiation at 1450 nm pump wavelength. **Figure 4**a shows the comparison of the time traces and Figure 4b shows the Fourier transforms of the four red crystals when irradiated under the exact same conditions and using the same parameters for each measurement. The THz peak-to-peak electric field

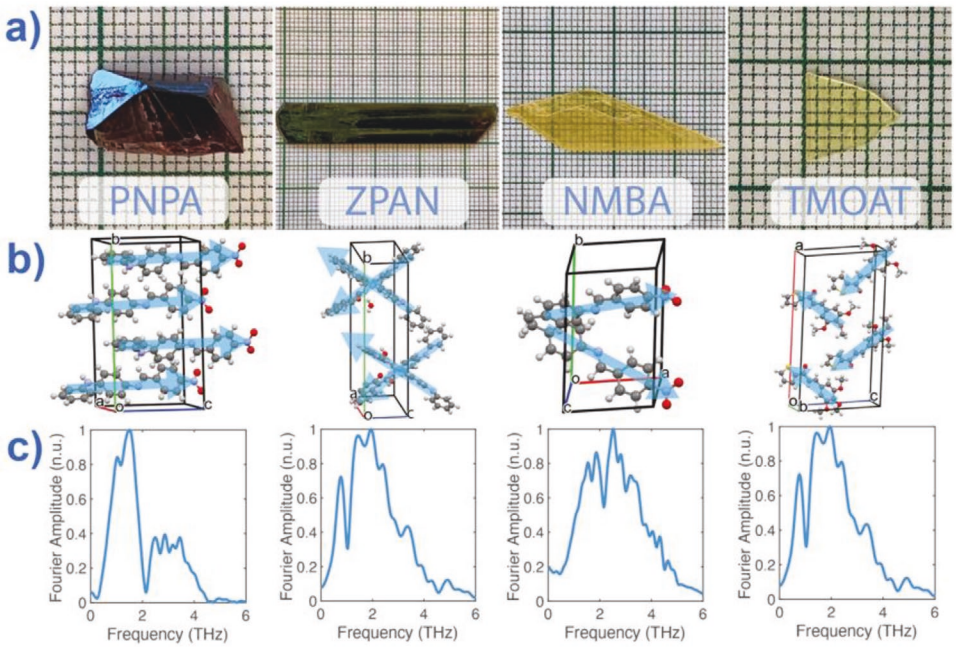

Figure 3. a) Large single crystals grown via slow evaporation protocols (square = 1 mm^2). b) X-ray crystal structure and direction of hyperpolarizability vector for each new crystal. c) Normalized THz spectrum of each NLO crystal.

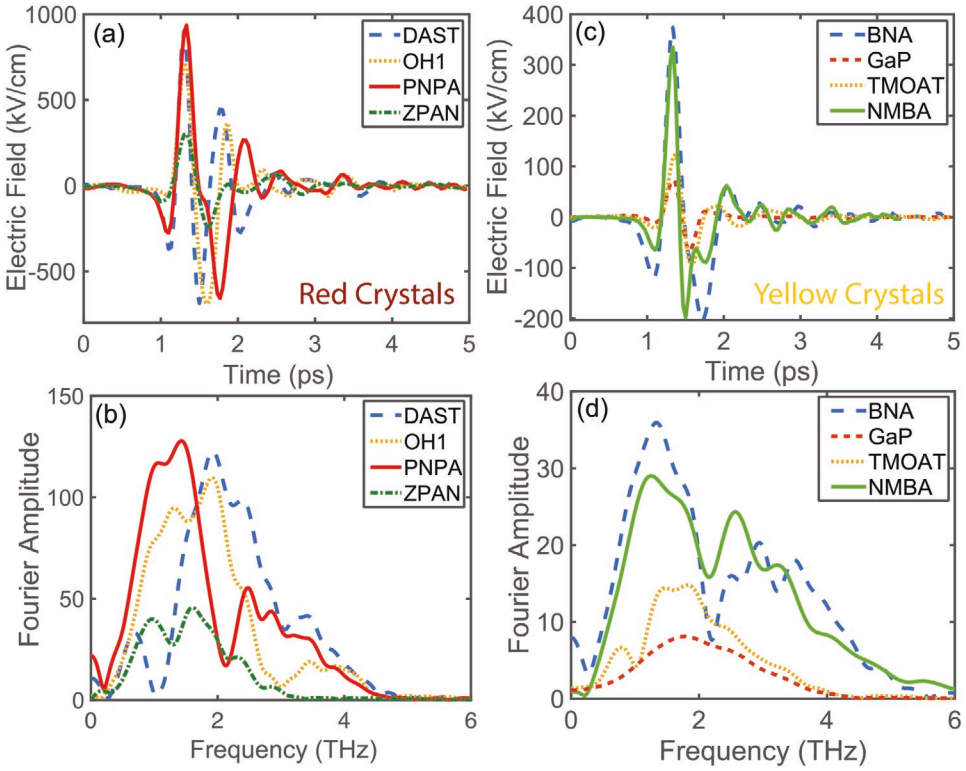

Figure 4. a) Generation comparison of red organic crystals at a pump wavelength of 1450 nm. b) Spectra of red crystals. c) Generation comparison of yellow organic crystals pumped at 1250 nm (and inorganic GaP). d) Spectra of yellow crystals and GaP.

of PNPA extracted from the time trace exceeds that of both DAST and OH-1. The THz spectrum and efficiency of PNPA rivals and even exceeds both DAST and OH-1, with a spectral peak at 1 THz and frequency components extending to 5 THz. Despite the mild absorption at 2 THz, PNPA provides the benefits of large amplitudes at <1.5 THz compared to DAST, as well as more intense THz at >4 THz compared to OH-1. ZPAN, although notably less efficient at THz generation, has a broad spectrum with only small absorptions below 3 THz. Having an order parameter of 0.55, ZPAN does not exhibit the head-to-tail chromophore configuration, but rather the X-configuration that has been garnering recent attention due to potentially large off-diagonal components of the nonlinear susceptibility.[25,26,38]

Yellow organic crystals often exhibit the ability to be pumped efficiently at shorter wavelengths than their red counterparts just described. NMBA and TMOAT have nearly clear, but still slightly yellow colors. We compared THz generation using our new NMBA (370 µm thick) and TMOAT (540 µm thick) crystals to well-known and characterized yellow crystal BNA (400 µm thick) and the red inorganic crystal GaP (300 µm thick). Figure 4c shows the electric field traces of the three yellow organic crystals compared to GaP, all pumped with exactly the same conditions, including the 1250-nm pump wavelength. We see that NMBA and BNA have very similar peak-to-peak electric field strengths. Figure 4d shows the Fourier transforms of these four crystals, showing that THz generation using NMBA is very broadband, and rivals that of BNA (with larger amplitude at higher frequencies). TMOAT does not generate THz as efficiently as BNA or NMBA but has a broad spectrum with only one main absorption at 1 THz. The intensity of THz generated with TMOAT is consistent with its lower calculated hyperpolarizability and order parameter compared to NMBA.

The combined data mining and computational approach predicted that these new crystals should perform relatively in this order: PNPA the strongest, followed by NMBA, ZPAN, and TMOAT the weakest. From THz generation results shown in Figure 4, we see a slight difference in the ordering, giving PNPA, ZPAN, NMBA, and TMOAT. We note that the next step to improve our predictive power should involve calculating phase-matching conditions for each material to better understand spectral shapes and relative THz intensities. Overall, these results confirm that our combined data mining and computational approach can identify new materials capable of intense THz generation with high levels of success and provide useful information about relative THz generation efficiencies between materials.

In conclusion, our data mining approach enables the rapid identification of new organic materials suitable for intense THz generation. These results demonstrate that data mining of structural databases combined with DFT calculations provide a productive and powerful method for identifying new organic materials for any material science application. Through this approach, unique molecular structures (chromophores) and materials can be identified that were originally reported for different materials applications, but that can be applied to a new field and enable new materials discovery. We rapidly identified and crystalized four new organic NLO

crystals with unique molecular structures in the amount of time that would previously have been required to discover a single new NLO material. In particular, the discovery of PNPA as an efficient THz generator that outperforms several benchmark organic nonlinear optical crystals will be beneficial to the growing research area of high-field THz science. These results coincide with the goals of the materials genome initiative, which are to expedite the development of advanced materials. Here, we show for the first time that it is possible to identify and produce organic solid materials with optimal nonlinear optical properties that make them excellent for THz generation.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Note: Figure 1 was reset on April 21, 2022, after initial publication online, to correct a spelling mistake in the figure.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

data mining, organic photonic materials, terahertz generation

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[1] C. R. Groom, I. J. Bruno, M. P. Lightfoot, S. C. Ward, *Acta Crystallogr. B* **2016**, *72*, 171.
[2] S. K. Kauwe, J. Graser, R. Murdock, T. D. Sparks, *Comput. Mater. Sci.* **2020**, *174*, 109498.
[3] B. Meredig, *Chem. Mater.* **2019**, *31*, 9579.
[4] R. J. Murdock, S. K. Kauwe, A. Y.-T. Wang, T. D. Sparks, *Integrating Materials and Manufacturing Innovation* **2020**, *9*, 221.
[5] K. T. Schütt, H. E. Sauceda, P. J. Kindermans, A. Tkatchenko, K. R. Müller, *J. Chem. Phys.* **2018**, *148*, 241722.
[6] A. Y.-T. Wang, S. K. Kauwe, R. J. Murdock, T. D. Sparks, *npj Computational Materials* **2021**, *7*, 77.
[7] T. Xie, J. C. Grossman, *Phys. Rev. Lett.* **2018**, *120*, 145301.
[8] S. Curtarolo, G. L. W. Hart, M. B. Nardelli, N. Mingo, S. Sanvito, O. Levy, *Nat. Mater.* **2013**, *12*, 191.
[9] C. C. Fischer, K. J. Tibbetts, D. Morgan, G. Ceder, *Nat. Mater.* **2006**, *5*, 641.
[10] H. Koinuma, I. Takeuchi, *Nat. Mater.* **2004**, *3*, 429.
[11] N. Marzari, A. Ferretti, C. Wolverton, *Nat. Mater.* **2021**, *20*, 736.
[12] W. Sun, C. J. Bartel, E. Arca, S. R. Bauers, B. Matthews, B. Orvañanos, B.-R. Chen, M. F. Toney, L. T. Schelhas, W. Tumas, J. Tate, A. Zakutayev, S. Lany, A. M. Holder, G. Ceder, *Nat. Mater.* **2019**, *18*, 732.
[13] R. M. Geilhufe, S. S. Borysov, A. Bouhon, A. V. Balatsky, *Sci. Rep.* **2017**, *7*, 7298.
[14] R. M. Geilhufe, S. S. Borysov, D. Kalpakchi, A. V. Balatsky, *Physical Review Materials* **2018**, *2*, 024802.
[15] R. M. Geilhufe, B. Olsthoorn, A. V. Balatsky, *Nat. Phys.* **2021**, *17*, 152.
[16] W. L. Chan, J. Deibel, D. M. Mittleman, *Rep. Prog. Phys.* **2007**, *70*, 1325.
[17] J. B. Baxter, G. W. Guglietta, *Anal. Chem.* **2011**, *83*, 4342.
[18] A. A. Gowen, C. O'Sullivan, C. P. O'Donnell, *Trends Food Sci. Technol.* **2012**, *25*, 40.
[19] W. Zouaghi, M. D. Thomson, K. Rabia, R. Hahn, V. Blank, H. G. Roskos, *European Journal of Physics* **2013**, *34*, S179.
[20] I. F. Akyildiz, J. M. Jornet, C. Han, *Physical Communication* **2014**, *12*, 16.
[21] F. D. J. Brunner, O. P. Kwon, S.-J. Kwon, M. Jazbinšek, A. Schneider, P. Günter, *Opt. Express* **2008**, *16*, 16496.
[22] T. Kampfrath, K. Tanaka, K. A. Nelson, *Nat Photon* **2013**, *7*, 680.
[23] C. H. Matthias, F. J. András, *J. Phys. D: Appl. Phys.* **2011**, *44*, 083001.
[24] C. Vicario, M. Jazbinsek, A. V. Ovchinnikov, O. V. Chefonov, S. I. Ashitkov, M. B. Agranat, C. P. Hauri, *Opt. Express* **2015**, *23*, 4573.
[25] G. A. Valdivia-Berroeta, L. K. Heki, E. W. Jackson, I. C. Tangen, C. B. Bahr, S. J. Smith, D. J. Michaelis, J. A. Johnson, *Opt. Lett.* **2019**, *44*, 4279.
[26] G. A. Valdivia-Berroeta, L. K. Heki, E. A. McMurray, L. A. Foote, S. H. Nazari, L. Y. Serafin, S. J. Smith, D. J. Michaelis, J. A. Johnson, *Adv. Opt. Mater.* **2018**, *0*, 1800383.
[27] G. A. Valdivia-Berroeta, E. W. Jackson, K. C. Kenney, A. X. Wayment, I. C. Tangen, C. B. Bahr, S. J. Smith, D. J. Michaelis, J. A. Johnson, *Adv. Funct. Mater.* **2019**, **n/a**, 1904786.
[28] G. A. Valdivia-Berroeta, K. C. Kenney, E. W. Jackson, J. C. Bloxham, A. X. Wayment, D. J. Brock, S. J. Smith, J. A. Johnson, D. J. Michaelis, *J. Mater. Chem. C* **2020**, 11079.
[29] I. C. Tangen, G. A. Valdivida-Berroeta, L. K. Heki, Z. B. Zaccardi, E. W. Jackson, C. B. Bahr, D. J. Michaelis, J. A. Johnson, ArXiv e-prints, 2020, 2005.05545.
[30] G. A. Valdivia-Berroeta, I. C. Tangen, C. B. Bahr, K. C. Kenney, E. W. Jackson, J. DeLagange, D. J. Michaelis, J. A. Johnson, *J. Phys. Chem. C* **2021**, *125*, 16097.
[31] 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, J. A. Johnson, ArXiv e-prints, 2010.02380, 2020.
[32] M. Shalaby, C. Vicario, K. Thirupugalmani, S. Brahadeeswaran, C. P. Hauri, *Opt. Lett.* **2016**, *41*, 1777.
[33] H. Zhao, Y. Tan, T. Wu, G. Steinfeld, Y. Zhang, C. Zhang, L. Zhang, M. Shalaby, *Appl. Phys. Lett.* **2019**, *114*, 241101.
[34] C. B. Bahr, N. K. Green, L. K. Heki, E. McMurray, I. C. Tangen, G. A. Valdivia-Berroeta, E. W. Jackson, D. J. Michaelis, J. A. Johnson, *Opt. Lett.* **2020**, *45*, 2054.
[35] M. Jazbinsek, U. Puc, A. Abina, A. Zidansek, *Appl. Sci.* **2019**, 9.
[36] J.-H. Jeong, B.-J. Kang, J.-S. Kim, M. Jazbinsek, S.-H. Lee, S.-C. Lee, I.-H. Baek, H. Yun, J. Kim, Y. S. Lee, J.-H. Lee, J.-H. Kim, F. Rotermund, O. P. Kwon, *Sci. Rep.* **2013**, *3*, 3200.
[37] O. P. Kwon, B. Ruiz, A. Choubey, L. Mutter, A. Schneider, M. Jazbinsek, V. Gramlich, P. Günter, *Chem. Mater.* **2006**, *18*, 4049.

[38] J.-A. Lee, W. T. Kim, M. Jazbinsek, D. Kim, S.-H. Lee, I. C. Yu, W. Yoon, H. Yun, F. Rotermund, O. P. Kwon, *Adv. Opt. Mater.* **2020**, *8*, 1901921.
[39] S.-H. Lee, M. Jazbinsek, C. P. Hauri, O. P. Kwon, *CrystEngComm* **2016**, *18*, 7180.
[40] S.-H. Lee, B.-J. Kang, J.-S. Kim, B.-W. Yoo, J.-H. Jeong, K.-H. Lee, M. Jazbinsek, J. W. Kim, H. Yun, J. Kim, Y. S. Lee, F. Rotermund, O. P. Kwon, *Adv. Opt. Mater.* **2015**, *3*, 756.
[41] L. Mutter, F. D. Brunner, Z. Yang, M. Jazbinšek, P. Günter, *J. Opt. Soc. Am. B* **2007**, *24*, 2556.
[42] Z. Yang, L. Mutter, M. Stillhart, B. Ruiz, S. Aravazhi, M. Jazbinsek, A. Schneider, V. Gramlich, P. Günter, *Adv. Funct. Mater.* **2007**, *17*, 2018.
[43] Z. Yang, M. Wörle, L. Mutter, M. Jazbinsek, P. Günter, *Cryst. Growth Des.* **2007**, *7*, 83.
[44] J. Yin, L. Li, Z. Yang, M. Jazbinsek, X. Tao, P. Günter, H. Yang, *Dyes Pigm.* **2012**, *94*, 120.
[45] S. S. Borysov, R. M. Geilhufe, A. V. Balatsky, *PLoS One* **2017**, *12*, e0171501.