

excellent generalization performance of the model. Furthermore, the performance of the model was compared with other algorithms, including Support Vector Machine (SVM), K-Nearest Neighbors (KNN), Random Forest (RF), and XGBoost Regression (XGBR). The results indicated that the XGBoost classification model exhibited the highest level of accuracy. The conclusion indicates that ML-assisted theoretical design can accelerate the discovery of 2D doped tellurene and perform effective descriptors to screen 2D doped tellurene with excellent performance in FinFET. Overall, the integration of ML approaches in the design of functional materials shows great promise, not only in the screening of dopants but also in expanding the exploration of new structures with highly desired properties for future applications.

ML is widely recognized as a promising approach to accelerate the discovery of new materials, and researchers in this field strongly believe that it could potentially revolutionize the procedures and inspiration driving the design

of functional materials. Despite being in its early stages, AI is evolving at a remarkable pace. Further exploration is needed to devise and adopt specialized solutions that effectively integrate AI with materials science. In particular, the combination of ML with robotic platforms<sup>9</sup> carries ML into the realm of practical materials preparation. This synergy significantly broadens the scope of ML's applicability. Hence, we believe that ML will play a crucial role in the discovery, synthesis, and characterization of materials, serving as a powerful tool in designing functional materials and offering a plethora of diverse possibilities.

#### DECLARATION OF INTERESTS

The authors declare no competing interests.

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## Synthesis and emergent properties of structurally complex materials with nonrandom disorder

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**This preview discusses the emergent new paradigm of structural complexity engineering, featuring two recent studies on an aperiodic metal-organic framework and nanoassembly of a symmetry-breaking chiral continuum bowtie. These structurally complex materials are not ordered but also not completely random, with controlled lowered symmetry.**

Ordered or crystalline forms of matter have been intensively sought after in diverse areas, such as metallurgy, micro-

electronics, pharmaceuticals, and self-assembly (spanning orders of magnitude in length scales from atoms,

molecules, supramolecules, and nanoparticles, to micron-sized colloids) over the past few decades. Numerous strategies of crystal engineering and theories have been established to build functional materials from units across multiple length scales,<sup>1</sup> such as inorganic

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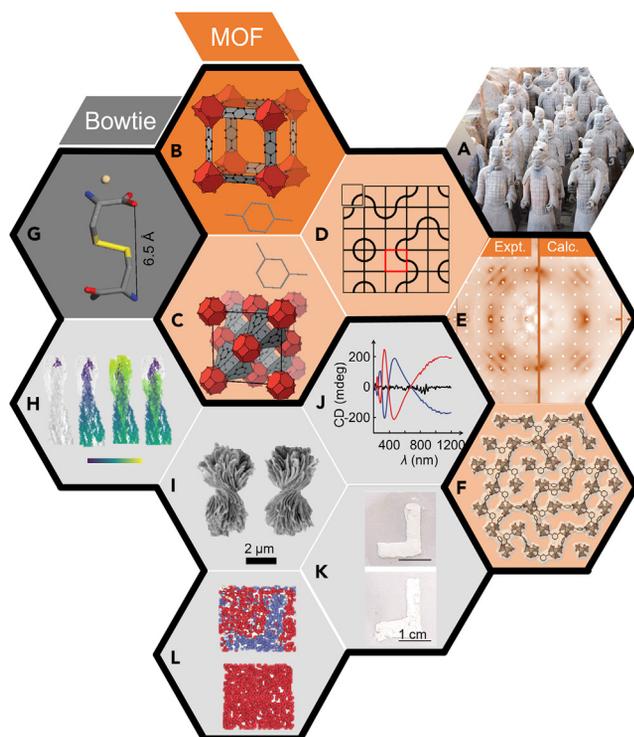
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**Figure 1. Structurally complex structures with nonrandom disorder**

- (A) Terracotta Army where the warriors differ but procession is in order.  
 (B) The 1,4-bdc linker and the resulting crystal structure of MOF-5.  
 (C) The 1,3-bdc linker and the resulting structure of TRUMOF-1.  
 (D) The Truchet tiling.  
 (E) Single-crystal X-ray diffuse scattering and the Bragg reflection positions (white dots) of TRUMOF-1 from experiment and density functional theory (DFT) calculation.  
 (F) The disordered pore network structure of TRUMOF-1.  
 (G) Molecular structure of cystine.  
 (H) Kinetic Monte Carlo simulation showing the growth process of bowties. Violet-to-yellow color bar represents temporal evolution.  
 (I) Scanning electron microscopy images of the bowties with chirality continuum.  
 (J) Property characterization of chiral bowtie using circular dichroism spectroscopy. Peaks are clearly seen for both blue (L-bowties) and red (D-bowties) lines, showing chiroptical activity.  
 (K and L) Application of the bowtie as photonic labels.  
 (C–F) Reprinted with permission from AAAS.<sup>8</sup> (G–L) Reproduced with permission from Springer Nature.<sup>7</sup>

crystals (e.g., metals, ceramics, biominerals) of rich electronic, photonic, and mechanical properties built from atoms and ions, metal-organic frameworks (MOFs) from coordination of metal ions and organic linkers for gas storage and release, and superlattices assembled from nanoparticles (NPs) as metamaterials.<sup>2</sup> In inorganic crystals, order arises from the precise and periodic arrangements of the constituent atoms or ions, which entails a series of kinetic control over the nucleation and crystal growth habits.<sup>3</sup> For organic-inorganic

crystals (such as MOFs), the primary unit becomes a hybrid motif, where the position, orientation, and spatial configuration of metal ions and linkers need to be commensurate with the symmetries of a large-scale repeatable lattice; for example, a linear linker such as 1,4-benzodicyclohexadiene (1,4-bdc) can serve as the basis of a diversity of MOFs, ranging from MOF-5 of simple cubic pores to MIL-101 of a giant unit cell built from corner-sharing tetrahedrons. In NP superlattices, order emerges from NPs that are highly monodisperse in size

and shape with inter-NP interactions that are annealable.<sup>4</sup> These rules of thumb and the resulting structures of long-range order (in terms of lattice constants and symmetry groups) produce diffraction or scattering patterns<sup>5</sup> under electrons or X-rays for structural characterization and thus allow controlled synthesis.<sup>6,7</sup> However, they significantly limit the varieties of the candidate building blocks and the structural diversity.

Meanwhile, systems that are “messier”—not ordered with spatial heterogeneity, such as intrinsically disordered proteins, gels, polymer membranes, and composites—are emerging to find importance in cell signaling, targeted delivery, molecular separation, and energy storage utilizing the structural flexibility where stringent requirements of order formation are not present. Recently, two studies innovatively demonstrated the formation of structures of greatly enriched complexity with reduced ordering.<sup>8,9</sup> The MOF work<sup>8</sup> intentionally employed a curved organic linker of reduced symmetry from a linear one. The coordination numbers in the formed motifs thus spatially change due to geometric frustration, leading to Truchet tiling analog structures, exhibiting both diffuse scattering patterns from local variations and Bragg reflection characteristics of a statistically averaged lattice structure. The nanoassembly work,<sup>9</sup> on the other hand, used non-uniform building blocks that differ in chirality, which assemble into twisted, symmetry-breaking bowties. Meanwhile, long-range electrostatic repulsion limits the size of the bowties, enabling high synthetic reproducibility and size monodispersity of their geometries. Despite the length scale difference, both studies produced materials of nonrandom disorder—somewhat similar to the macroscopic Terracotta Army in China, Figure 1A: the look and expression of each warrior differ, but their collective procession signifies self-similarity.

Writing in *Science*, Meekel and colleagues<sup>8</sup> show the preparation of nonrandom disordered TRUMOF-1 structure by replacing the linear 1,4-bdc linker producing MOF-5 with a bent 1,3-bdc linker (Figures 1B and 1C). While X-ray diffraction of the MOF structure suggests a stoichiometry of  $[\text{OZn}_4][1,3\text{-bdc}]_3$ , the same to that of MOF-5, the reduced symmetry of the 1,3-bdc linker drastically altered the original simple cubic unit cell of MOF-5 into a complex cubic unit cell.  $[\text{OZn}_4]$  clusters are connected at the corners and face centers by partly occupied trigonally symmetric linker molecules. In addition, while the  $[\text{OZn}_4]$  clusters have 12 nearest neighbors in an ideal lattice, only half of them are connected by the bent linker, contrary to MOF-5 where all nearest neighbors are connected by 1,4-bdc linkers. This coordination number variation creates another complexity in terms of reticular connectivity on top of a complex lattice structure (Figure 1C).

The structural complexity in TRUMOF-1 prompted efforts to map the three-dimensional (3D) nonrandom disorder and to model and understand the structure. The authors traced the connection paths of the  $[\text{OZn}_4]$  clusters and found an analogy to the two-dimensional Truchet tiles (Figure 1D), each of which has a single arc at the corner (corresponding to the curved 1,3-BDC linker) connecting the centers of the neighboring edges (corresponding to the  $[\text{OZn}_4]$  clusters), forming into the complex aperiodic lattice of TRUMOF-1. The Truchet tile captures the structural complexity based on which atomic-scale models were constructed for DFT calculations, reproducing the weak diffuse scattering observed in experiments (Figure 1E). Note that diffuse scattering is related to the disordered connectivity. The aperiodic pore network structure can invite new applications to frustrate elastic instabilities and dampen long-wavelength acoustic phonons, which are not accessible in ordered lattices (Figure 1F).

On the larger length scale of NP building blocks, symmetry-lowered molecular twists, such as chiral ligands, facilitate the formation of solid-state microstructures that are otherwise challenging to obtain. Reporting in *Nature*, Kumar and colleagues<sup>9</sup> recently demonstrated progressively tunable bowtie structures with a chirality continuum, rather than binary left or right chiral phases and shapes, using chiral molecule-facilitated self-assembly (Figures 1G–1I). In an aqueous solution, different ratios of L- or D-cystine molecules are mixed with  $\text{Cd}^{2+}$  cores, where the four cystine molecules are spontaneously anchored and coordinated to a  $\text{Cd}^{2+}$  core. These heterogeneous coordination clusters further pack into twisted nanoribbons 200–1,200 nm in length and 45 nm in thickness, which subsequently stack to form micron-sized bowties. The chirality originates from the helical nature of the nanoclusters, which also allows chemical stability through partial swapping of L-cystine for D-cystine without drastic disruption of the short-range coordination bonds. The assembly is self-limiting and stops when the electrostatic repulsion wins over the addition of new coordinated clusters (Figure 1H).

The correlation between the molecularly derived microstructural chirality and the ensemble functionality was studied in the chirality continuum. By changing the ratio of L- or D-cystine in the aqueous solution, the final bowtie can exhibit continuous handedness from “left” to “right” with tunable twist angle, pitch, width, thickness, and length, exhibiting multispectral photonic activity in terahertz, mid-infrared, and UV-visible-near infrared ranges, a representative spectrum shown in Figure 1J. From a thorough analysis and comparison among descriptors, Osipov-Pickup-Dunmur (OPD) index, which offers continuous and sign-changing chirality measure, was chosen for the chiral bowties. Surprisingly, the OPD measure indeed correlates (expo-

nentially) with the major peaks in the circular dichroism spectrum. The correlation between the chirality measure and the optical signatures allows pattern recognition (Figures 1K and 1L), which can be useful for autonomous driving with scalable manufacturing of printable metasurfaces.

These two studies from the fields covering different length scales show a convergent interest in structural complexity that lowers ordering and symmetry but introduces numerous packing possibilities for widely opened property space. To this end, new synthesis and assembly routes, new characterization techniques and simulation tools, new quantitative description and understanding of the structural complexity,<sup>10</sup> and new bridges that connect structural descriptors to functionalities are needed. We anticipate a new paradigm to emerge in novel material systems that take advantage of the structural complexities of disorder and lowered symmetry.

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## DECLARATION OF INTERESTS

The authors declare no competing interests.

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## Flexible crystals meet dynamic phosphorescence

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**Utilizing a crystal engineering approach to design multifunctional flexible materials is highly desirable but still a challenging goal. By linking crystal structures and luminescent properties, Ma and co-workers demonstrated a series of single-component molecular crystals simultaneously with high elasticity, room temperature phosphorescence, and multi-level stimuli responsiveness. This study thus expands the scope of potential applications of flexible optical materials.**

Flexible materials have made great advances for next-generation electronic and optoelectronic devices with elastic mechanical responses.<sup>1</sup> In this field, molecular crystals have recently attracted much attention, ascribed to the uniformly distributed weak interactions acting as a structural buffer for their flexible and elastic properties.<sup>2</sup> The luminescent properties of elastic organic crystals (EOCs) are important for the extension of flexible device applications. Although a lot of EOCs with short-lived fluorescence have been explored,<sup>3</sup> achieving versatile EOCs with tunable and ultralong room temperature phosphorescence (RTP) is still highly desirable, and the examples are rather limited.

During the last decade, RTP materials with long-lived triplet excited states

have drawn much academic and engineering interest in the fields of anti-counterfeiting, optical imaging, information encryption, and so on.<sup>4</sup> To realize persistent RTP, the crystallization-induced structural rigidification is recognized as an effective way to reduce the non-radiative decay.<sup>5</sup> For molecular crystalline materials with tunable excited triplet states, changes in molecular structure, conformation, or packing would have important effects in their corresponding stimuli-responsive RTP performances.<sup>6</sup> Considering the above factors, it is possible to endow organic crystals with RTP, stimuli responsiveness, and elastic softness simultaneously, which may supply new opportunities for their potential use as dynamic materials and smart devices. However, the development of EOCs with stimuli-responsive RTP is still

in its infancy stage, mainly due to the conflicting requirements of molecular solid-state structures: (1) to realize efficient RTP emission, a rigid microenvironment with strong intermolecular forces is necessary; (2) the formation of slip planes with regionally weak intermolecular interactions is vital to achieve elastic deformation; and (3) to obtain effective stimuli responsiveness, the transformation of the molecular conformation and/or packing mode, which is maintained by relatively weak intermolecular interactions, should be ensured.<sup>7,8</sup>

In a recent issue of *Matter*, Ma and Tian's group from East China University of Science and Technology made progress in EOCs with dynamic RTP. They have devoted great passion into the research of single-component organics beyond typical cocrystals,<sup>2,3</sup> named dialkyl 4,4'-oxalyldibenzoate (n-BZ; n represents the number of carbon atoms in the n-BZ alkyl chain), with combined long-lived RTP and multilevel stimuli-responsive properties (Figure 1).<sup>9</sup> Herein, the benzil unit acted as the chromophore to achieve RTP

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