Crystal engineering of organic molecular solids has been practiced for the last 50 years or so and it is convenient to divide the chronology of its progress into three generations of effort (Figure 1a). First generation crystal engineering was about recognizing that there is a connection between crystal structure and crystal property. The term “Crystal Engineering” was invoked more as a challenge as to how one could go about designing a particular crystal structure. This first stage also saw development of various models that explained observed crystal structures in terms of the packing features and/or intermolecular interactions involved. Second generation crystal engineering dealt with the actual strategy and logic driven methodology of crystal design; the concept of the supramolecular synthon is crucial to the planning of crystal synthesis. But there was no prediction of a property. Here we have tried to take a step in this direction and to utilize predesigned modular structures to modulate properties and exemplified with hand-twisted helical crystals, finding importance in different fields such as negative-index invisible materials, enantiosensitive plasmonic sensors, lithography techniques, chiroptic materials.

A stepwise design strategy from one-dimensional (1D) plastic crystal → 1D elastic crystals → two-dimensional (2D) elastic → 2D plastic crystals is outlined. This type of 2D plasticity represents a new generation of bendable crystals in which plastic behavior is seen with a fair degree of isotropic character in the crystal packing. The presence of two sets of bendable faces, generally orthogonal to each other, allows for the possibility of hand twisting of the crystals to give grossly helical morphologies (Figure 1b). Accordingly, we propose the name hand-twisted helical crystals for these substances. In third generation crystal engineering a solid state property may be obtained through supramolecular synthon based logic driven design rather than merely by optimizing a crystal property that is obtained from an isolated serendipitous result.


**Figure 1**: (a) Different generations of crystal engineering. (b) A hand-twisted helical crystal.

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