Crystal Structure Prediction Applied to Photovoltaic Materials

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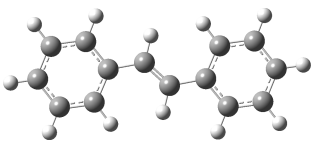
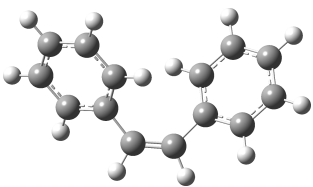


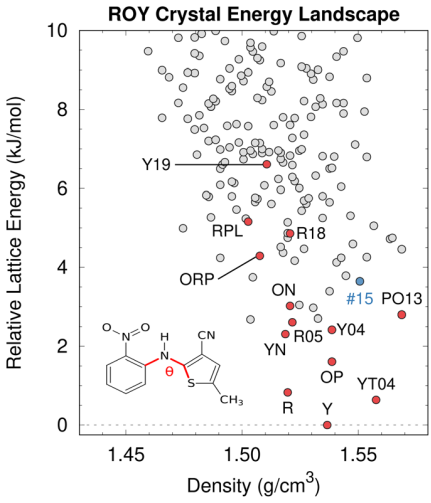


What is Crystal Structure Prediction (CSP)?

Definition: Predicting the crystal structure starting from its molecular diagram.

Why is it important?

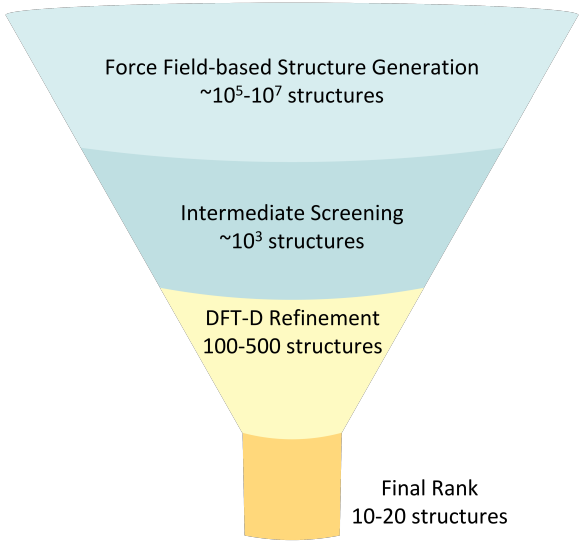
- Physical properties depend critically on crystal packing.
- Determines electronic properties in **organic semiconductors**.
- Fundamental for the design of tailored materials prior to synthesis.



Source: Beran et al., *Chem. Sci.* **14**, 13290

Search strategy: the computational "funnel"

1. **Extensive sampling:** Stochastic generation.
2. **Experimental screening:** Filtering by XRPD similarity.
3. **Accurate ranking:** Refinement via solid state DFT-D.

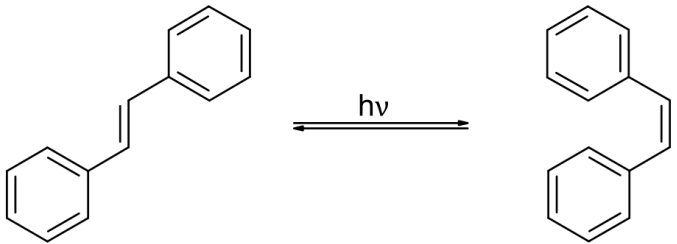


Crystal Structure Prediction of Stilbene

Stilbene is a **molecular photoswitch** that changes its structure upon irradiation:

- ***trans*-stilbene:** Stable isomer, solid at room temperature.
- ***cis*-stilbene:** Metastable isomer, liquid at room temperature.

Technological interest as a **Phase Change Material (PCM)** and for **Molecular Solar Thermal (MOST)** energy storage [1].

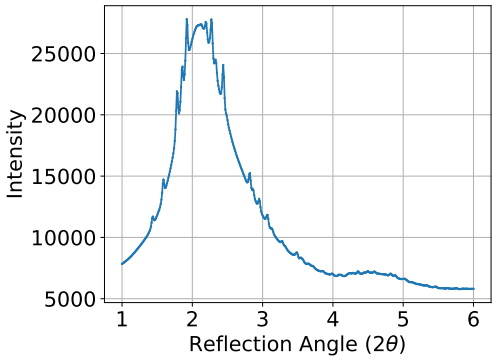


[1] Börjesson et al., *ACS Sustainable Chem. Eng.* **1**, 585 (2013).

Experimental evidence and objective

The experimental problem:

- Sample of *cis*-stilbene measured using Differential Scanning Calorimetry (DSC).
- Under slow cooling, the sample **partially crystallizes**.
- Synchrotron diffraction (Diamond Light Source, Oxford. $\lambda = 0.1617$Å) reveals a powder pattern of this phase.



Objective

Can we use CSP to identify the structure generating this experimental pattern and understand its stability?

Crystal structure prediction for Singlet Fission materials

Singlet Fission (SF) is a photophysical process that multiplies charge carriers:

- **Mechanism:** A single absorbed photon generates a singlet excited state (S_1) that splits into **two triplet states** ($2 \times T_1$)
- **Efficiency:** Allows bypassing the theoretical **Shockley-Queisser limit** ($\sim$30%) in single-junction solar cells.
- **Thermodynamic Criteria:** $E(S_1) \geq 2E(T_1)$ and avoid triplet-triplet annihilation (TTA)$E(T_2) \geq 2E(T_1)$

Evaluation of SF Candidates

- **1. Crystal Structure Prediction:** exploration of boron-doped acenes to map their crystal energy landscapes.
- **2. Dimer Extraction:** Selection of the global minimum lattice energy structure and extraction of the closest interacting molecular dimer.
- **3. Evaluation evaluation of the electronic states** (S_0, S_1, T_1, T_2) using multireference methods.