PhD PROJECT PROPOSAL

## **PhD Project Title**

## Strain Tuning Phonon Topology in Flexible Organic Crystals

## **PhD Supervisory Team**

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## **Project Abstract**

A class of organic crystals have been recently discovered that are bendable. This opens an exciting opportunity to design new functional materials but requires a detailed understanding of how bending affects material properties. Of particular importance is to identify how bending affects the vibrational behaviour of the material, because these vibrations play a fundamental role in nearly all functional properties, including thermal and electronic conductivity. By studying how the uniaxial strain of bending affects material vibrations, using both simulation and experiment, this project will develop strategies to identify, predict, and design new flexible crystals suitable for next-generation technologies.

## **Detailed Project Description**

Background: Single crystals are essential components of many functional materials. Unfortunately, single crystals are typically brittle, making technologies fragile and limiting their potential for wearable or mouldable

devices, or in sensing applications. The paradigm of brittle single crystals has been recently challenged with the discovery of mechanically flexible single crystals, which bend without compromising the crystallinity. These fascinating solids have the potential to radically transform the material design landscape, with exciting promise in next-generation nano-electronic and nano-optical devices, for innovations in sensing and display technologies, and in energy harvesting and conversion.1

Mechanical bending exerts marked uniaxial forces on the internal structure of the crystal,2 Fig 1, with potential to impact their functional properties by changing the underlying phonon spectrum.3 In the organic solid state i.e. for most mechanically flexible crystals functional properties are determined by both localised molecular-based phenomena and delocalised, cooperative phenomena. For systems where these phenomena mix through phonon interactions, strain (i.e. during bending) exerts its biggest influence on material properties.4 The phenomenon of phonon interactions in organic solids is often discussed as , which describes crossing of molecular vibrational bands with lattice vibrational bands, though this phenomenon remains largely unexplored. The development of topology provides an excited opportunity to elucidate this amalgamation and hence provide a new dimension for understanding and engineering the organic solid state. Some of the key functional properties envisioned for mechanically flexible crystals are intimately dependent on the nature of phonon amalgamation, including thermal and optical transport, for example to design shapable optical band pass materials. Additionally, with the vision of making flexible nano-electronic devices, charge transport in the organic solid state relies on having poor amalgamation, as strongly amalgamated phonons expedite charge recombination.5 Designing flexible materials for technological applications therefore requires that we understand the topological features responsible for phonon coupling, the structure types that give rise to these topological features, and our ability to make/break these features with uniaxial force.

Project: This project will establish a new dimension of structure-property correlations in the organic solid state with the aim to design next-generation, mechanically flexible functional materials. By computationally characterising, and benchmarking with inelastic neutron scattering spectroscopy, the phonon topology of experimentally reported mechanically flexible crystals, we will establish a library of topologies for a broad range of organic crystals. This represents the first investigation into phonon topology in organic crystals and promises an exciting new direction to understand the organic solid state. Emphasis will be on identifying topological features associated with the interaction between the crystal lattice and the constituent molecules, with the goal to identify which crystallographic and molecular features favour these topologies, for example aided by crystal structure descriptor algorithms. By combining simulation with synchrotron X-ray diffraction, synchrotron X-ray absorption spectroscopy, and vibrational spectroscopy, we will explore how key phonon topologies can be selectively manipulated through uniaxial mechanical strain using unique, custom-built set-ups. Hence, the project will establish a link between atomic structure, bulk mechanical response, and their cooperative role in selectively designing tuneable phononic behaviour to facilitate the design of a new generation of advanced functional material.

Fig.1

Fig 1| Mechanically flexible crystal (left), and compression of crystal axes in bent crystal (right)

Objectives: To establish a strategyto design controllable phonon topologies in mechanically flexible organic crystals. To achieve this the project aims:

i)Toidentify the topological nature of phonons in model organic crystals

ii)To determinewhich topological features are responsible for mixing ofthelocalised and cooperative

behaviourin the organic solid state

iii)To identify the crystallographic and molecular features that give rise tothese topological structures

iv)Toidentify how topological features can be created / annihilated through uniaxial mechanical strain, and hence identify how mechanical bending in organic crystals can give rise to design strategies for mechanically functional materials. Training: The CDT student will receivea highly interdisciplinary portfolio ofin-depth training across

techniques in computational materials science(including phonon simulations within the tight binding and density functional theory paradigms), aspects of materials informatics (high dimensional structural descriptors),

crystal growth,advanced synchrotron-based structural characterisation (standard single crystal and powder X-

ray diffraction and X-ray spectroscopy, and advanced applications under conditions of hydrostatic and uniaxial compression), and inelastic neutron scattering (INS)spectroscopy. To accompany scientific skill development, the CDT studentwill also developskills in coding (e.g.,Python), high-performance computing, and technique/instrument development (uniaxial compression devices and synchrotron beamline construction). The student will also develop transferrable skills, including proposal writing experience (e.g.,beamtime and high-

performance computing time applications), scientific writing (preparing academic journalarticles),

presentation skills (presenting in group meetings and at scientific conferences).

The supervisory team: This project is highly interdisciplinary, integrating computational materials science,

experimental solid-statechemistry, and materials engineering. The project builds on expertise of Dr Michalchuk inphonon dynamicsoforganic solids, including under hydrostatic high-pressureconditions, and his developments in atomistic studies of mechanicallyflexible crystals.6 This expertise combines with that of Dr Sunin method development and study of single crystals under uniaxial strain.Through the strategic Birmingham BAM collaboration, the project usesthe expertise of Dr Emmerling (BAM) in X-ray diffraction and access via BAM to facilities at the Berlin synchrotron (BESSY-II).The project focuses on the UoB strategy

for central facilities research, and will makeextensive use of the Birmingham-Diamond (and Birmingham-

DIAD) collaborations to access facilities at the Diamond Light Source. The CDT student should have a background in chemistry or physics (or related discipline), with a keen interest to combine simulation and experiment tostudy material behaviour.

References: (1)Ahmed, et al. Angew. Chem. Int. Ed. 2018, 57 (29), 8837 8846..

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(4)Michalchuk, A. A. L.,et al. J. Mater. Chem. A 2019, 7 (33), 19539 19553.

(5)Schweicher, G.,et al. Adv. Mater. 2019, 31 (43), 1902407.

(6)Liu, X., et al. Nat. Commun. 2021, 12, 3871.