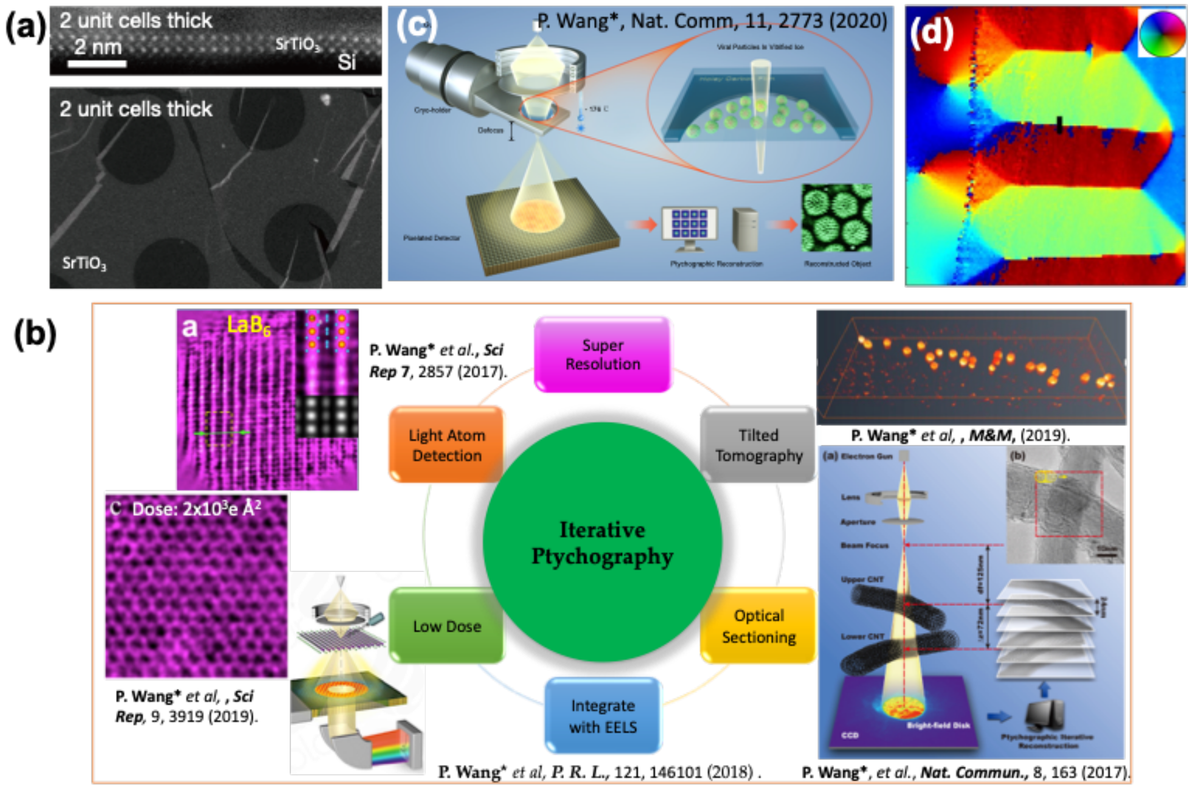
High-Resolution Probing Ferroic-ordering by Cryogenic Electron Ptychography

Direct visualisation of **ferroic orderings (ferromagnetism and ferroelectricity) in materials at the atomically thin limit** [1] as shown in Fig. a) is exciting not only from a fundamental physics perspective, but is also critical for the characteristics of ferroelectric and ferromagnetic materials used in applications that include information storage and logic technologies. Quantitative information about electric and magnetic fields with a high spatial resolution towards an atomic scale is indispensable for a complete understanding of the underlying physics behind the ferroic phenomena. Three-dimensional topological phases, such as skyrmions that hold promise for the applications for future topological spintronics are extremely small, with diameters in the nanometre range; the limitations in imaging technology have so far kept researchers from observing them in more than two dimensions. The imaging methodologies are required to investigate these ferroic-ordering structures in materials and devices at resolutions of a few atoms, both in projection and in three dimensions, and ideally reveal dynamic events in real-time while the specimen is subjected to external stimuli such as elevated or reduced temperature, applied voltage, or light.

Regarding transmission electron microscopy (TEM), as the electron wave passes through a sample, an electron scatters, producing an exit wavefront rich in information. Using the **wave nature of electrons**, a wide range of solid properties, from electric and magnetic field strengths to specimen thickness, strain maps and mean inner potentials, can be extrapolated from its phase and mapped at the nanoscale or even at an atomic level Therefore, accurate sensitive imaging of the electron phase signal at high resolution has been a long-term goal pursued in the field of electron microscopy.

With ultrafast direct detectors, we have been developing a high-resolution diffractive **phase imaging technique**, so-called **ptychography**. Ptychography is a phase recovery method based on scanning in-line holography, as originally proposed by Hoppe. This approach uses a radiation source to illuminate a sample and records a series of diffraction patterns as a function of the illumination position to recover the sample exit plane wavefunction using iterative algorithms. Ptychography has attracted considerable interest from both X-ray and electron communities for its potential applications in super-resolution imaging, reaching a resolution of 0.39 ångströms, which set a world record by tripling the resolution of a state-of-the-art electron microscope. Electron ptychography enables high-contrast light-element detection [2], high dose-efficiency [3], 3D optical sectioning [4] and coupling to spectroscopic data acquisition [5], as performed by us and as shown in Fig. b). Recently, at the cryogenic temperature we for the first time demonstrated electron ptychography can provide high contrast and high sensitivity of phase information of samples [6], as shown Fig. c). The potential of phase imaging is not only recognized to allow light elements to be detected at the super-resolution and low dose, but also to provide unique information about local variations in magnetic and electrostatic fields. Fig. d) shows our preliminary result of a colour-coded vector map of the projected magnetization recovered using electron ptychography. Therefore, electron ptychography can potentially enable high-sensitivity observations of the ferroic-orderings within functional materials (such as 2D ferroic thin films) at unprecedentedly high resolution and sensitivity at low temperature.

In this proposed research project, we will **develop a novel ordering field imaging using electron ptychography at a cryogenic temperature** together with "big data" processing methods and exploit opportunities by accelerating its application in functional materials. The quantitative imaging of the magnetization configurations will be extracted by comparison with detailed image simulations. Combined with tomographic methods [7], the electromagnetic field mapping will be extended into **three-dimensional vector fields** inside nanostructured materials, rather than simply in projection. Furthermore, we will look at **dynamic behaviours of materials *in situ*** and study how they respond to a changing external stimulus (such as electric, magnetic field, temperature and light [8]) at timescales required. This time-resolved phase imaging will be capable of opening the doors to many high-resolution *in situ* experiments such as real-time observation of phase transitions in relation to external stimuli (electric bias) or access to intermediate phases which exist only in narrow temperature windows. It can also lead to exciting opportunities in next-generation integrated flexiable electronics and nanoscale devices.



References:

[1] Ji, D.,…., **Wang*, P.**, Nie*, Y. & *et al*. Freestanding crystalline oxide perovskites down to the monolayer limit. ***Nature*** **570**, 87-90, (2019).

[2] **Wang* P.**, *et al.* Electron Ptychographic Diffractive Imaging of Boron Atoms in LaB_6 Crystals. ***Scientific Reports***, **7**, 2857 (2017).

[3] Song J.,…, **Wang* P.**, *et al.* Atomic resolution defocused electron ptychography at low dose with a fast, direct electron detector. ***Scientific Reports***, **9**, 3919 (2019).

[4] Gao S.,…, **Wang* P.**, *et al.* Electron ptychographic microscopy for three-dimensional imaging. ***Nature Communications,*** **8**, 163 (2017).

[5] Song B.,…,**Wang* P.** Hollow electron ptychographic diffractive imaging. ***Physical Review Letters***, **121**, 146101 (2018).

[6] Zhou, L., …. & **Wang*, P.** Low-dose phase retrieval of biological specimens using cryo-electron ptychography. ***Nature Communications,*** 11, 2773, (2020).

[7] Ding, Z., …. & **Wang*, P.** Three-dimensional electron ptychographic phase imaging for organic-inorganic hybrid nanostructures, ***Nature Communications,*** in Revision (2021).

[8] Cai, S., …& **Wang*, P.** Development of in situ optical–electrical MEMS platform for semiconductor characterization. ***Ultramicroscopy*** **194**, 57-63, (2018).