Breaking symmetry with light: ultra-fast ferroelectricity and magnetism from non-linear phononics

Andrea Cavalleri (MPI for Structural Dynamics & Oxford University)
Paolo Radaelli (Oxford University)

The use of light to control the structural, electronic and magnetic properties of solids is emerging as one of the most exciting areas of condensed matter physics. One promising field of research, known as femto-magnetism, has developed from the early demonstration that magnetic 'bits' in certain materials can be 'written' at ultra-fast speeds with light in the visible or IR range [1]. More radically, it has been shown that fundamental materials properties such as superconductivity can be 'switched on' transiently under intense illumination [2]. Recently, the possibilities of manipulating materials by light have been greatly expanded by the demonstration of mode-selective optical control, whereby pumping a single infrared-active phonon mode results in a structural/electronic distortion along the coordinates of a second, anharmonically coupled Raman mode – a mechanism that was termed 'nonlinear phononics'. Crucially, the Raman distortion is partially rectified, meaning that it oscillates around a different equilibrium position than in the absence of illumination. Recently it was realised that, under appropriate conditions, the rectified Raman distortion can transiently break the structural and/or magnetic symmetry of the crystal and hypothesised that such symmetry breaking would persist for a time corresponding to the carrier envelope of the pump, which can be less than a picosecond, and can give rise to the ultra-fast emergence of ferroic properties such as ferromagnetism and ferroelectricity [4]. Even more recently, this effect was experimentally demonstrated for the first time in our collaborators' laboratory in Hamburg. Surprisingly, photo-ferroicity persisted for a significantly longer time than the carrier envelope (100s of ps). Although this is not yet fully understood, the most likely explanation is that magnetisation is being transferred to slower electronic/magnonic excitations.

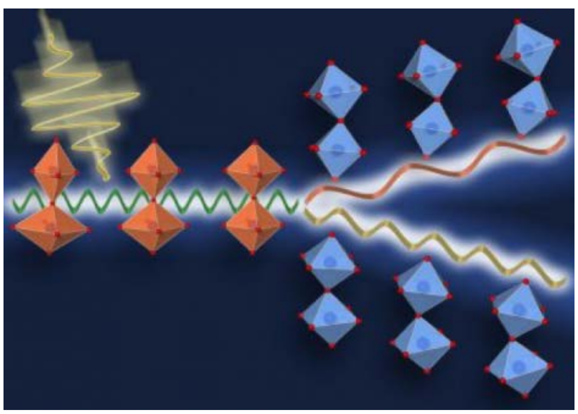

Figure 1: schematic representation of non-linear photo-ferroicity. THz or far-IR 'pump' photons excite an IR-active mode (right), which is coherently coupled with a Raman mode (left). The rectified component of the Raman mode transiently generates ferromagnetism or ferroelectricity, which is probed coherently with a near-IR or visible light beam (top left).

This DPhil project will give the successful candidate the opportunity to pioneer this new field of research. Initial experiments on the 'photo-ferroic' materials that we have already characterised will be performed at the Max Planck Institute for the Structure and Dynamics of Matter in Hamburg, Germany. As a mode-selective pump, we are employing coherent laser radiation in the THz or far-IR range with sub-ps carrier envelopes, while the transient emergence of the ferroic properties will be probed with second-harmonic generation (SHG), Faraday rotation and dichroic absorption of visible/near-infra-red light. Later on in the project, changes in the crystal and magnetic structures of the materials will be probed with X-rays at free electron laser sources such as the European XFEL in Hamburg. Meanwhile, the candidate will develop search strategies for new classes of 'photo-ferroic' materials, based on symmetry and time-dependent density functional theory calculations. He/she will develop the materials specifications in collaborations with crystal growers in Oxford and elsewhere, and will be involved hands on in all aspects of the design and realisation of the experiments and the data analysis.

The experimental part of this project will be predominantly based in Hamburg, so it is essential for the candidate to be willing and able to be based in Germany for extended periods during the DPhil.

[1] "Femtomagnetism: Magnetism in step with light". Uwe Bovensiepen, Nature Physics 5, 461 - 463 (2009) abstract pdf

[2] See for example M. Mitrano,et al., "Possible light-induced superconductivity in K3C60 at high temperature", Nature, 530, 461–464 (2016). [more at this link].

[3] "Nonlinear phononics as an ultrafast route to lattice control", M. Först et. al., Nature Physics, 7, 854–856 (2011).

[4] "Breaking Symmetry with Light: Ultra-Fast Ferroelectricity and Magnetism from Three-Phonon Coupling", P. G. Radaelli, Phys. Rev. B 97, 085145 (2018).