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Microscopy and Modelling to Understand Defects and Phonon Dispersions

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Electron energy loss spectroscopy (EELS) carried out inside a scanning transmission electron microscope (STEM) provides a probe of elemental composition and bonding with atomic resolution. It allows us to map the local chemistry of a material. STEM EELS can be used to tackle a wide variety of materials problems and has been used to identify single atoms [1], determine crystal phases [2], map dopants at grain boundaries [3] and visualise plasmon modes [4]. The flexibility of EELS comes in part because there are different energy-loss mechanisms which provide us with a variety of probes of structure and bonding.

Advances in instrumentation mean that signals unavailable a decade ago are now accessible in the electron microscope. Spectra from single atoms and defects have been obtained using core-loss spectroscopy. These signals are weak simply due to the small number of atoms involved. Obtaining these spectra require stability in both the microscope and the sample. Significant advances in hardware over the last few years have reduced the energy spread of the electron probe making phonon modes now accessible [5]. The low energies of the phonon modes mean that they are present as a week signal on the tail of the zero-loss peak.

Combining experimental EELS with modelling can play an invaluable role in both the interpretation of experimental data and the design of experiments. Crucially, it can allow us to solve problems which experiment or modelling alone cannot. Along with the advances in microscopy, there have also been advances in first principles EELS simulations in the past decade. These include advances in first-principles structure predication [6], density functional theory functionals [7] and theory to simulate phonon EELS spectra which had been previously unnecessary [8,9].

This presentation will focus on several examples where modelling has been necessary to interpret the spectra from weak signals. It will, most importantly, put it in a materials science context to highlight the advances in our knowledge that have resulted from combining modelling and microscopy. I will give examples including counting nitrogen-vacancies in nanodiamond [10] and using a combination of first principles structure prediction, EELS and diffraction to solve the structure of an unknown crystal phase at the interface of a metal and an oxide [2]. I will also talk about the interpretation of momentum resolved vibrational mode measurements which can now be carried out with nanometre spatial resolution [9].

References

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