First principles modelling of the excited state and simulated spectroscopy

One of the key challenges in the quantum-mechanical simulation of condensed matter systems lies in a well-documented short-coming of density functional theory. However, it remains a crucial aspect of quantum-chemical simulations that directly comparable data must be available between the theory and experiment, and that this data should be as quantitatively accurate as possible. Over recent years, the calculation of key observables including electrical levels, vibrational quanta and hyperfine interaction tensors have been key to the unambiguous identification of point defect structures, with aspects such as stress coefficients and isotopic shifts being of particular importance.

Perhaps the single largest area for development is the routine quantitative computation of quantitatively accurate spectra for electronic transitions, such as yielded from an optical absorption or photoluminescence experiment. However, such electronic transitions are not strictly within reach of standard density functional calculations, as this theory is based upon the electronic ground state, so that crude estimates for transition energies are often severely underestimated. Additionally, although the energies of vibrational quanta are routinely obtained with fair accuracy, their relative intensities are less accessible.

Recent methodological advances, however, allow for more quantitative accuracy through techniques including the screened exchange functional, and it is the chief aim of the project to connect the advances in computational technique to the determination of quantitatively accurate spectra, both in terms of energy (especially in the context of PL) and intensity (in the context of IR and Raman).

The student will experience the experimental determination of spectroscopic data in the mini-projects, and through these contacts both an appreciation of the data and of the unresolved defect centre structures will be built. Once on the PhD project, it is the aim to maintain the links from the mini-projects, with the establishment of a key range of experimental centres to address during the simulations.

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