Images of Unpaired Electron Density in Molecular Crystals Obtained using Experimentally Constrained Wavefunctions

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Unpaired electrons are responsible for many of the magnetic properties of crystalline systems. Traditionally, unpaired electrons in crystalline systems have been imaged using the technique of polarised neutron diffraction (PND). However, these experiments are difficult, and relatively few data are obtained compared with X-ray diffraction measurements.

In this talk I will present and compare images of unpaired electron density in molecular crystals. These images are obtained from a molecular or cluster Hartree-Fock wavefunctions, which have been constrained to reproduce X-ray diffraction data, to reproduce PND data, and to reproduce both of these data simultaneously.

Two crystalline systems will be considered: a system displaying the photomagnetic LIESST effect, and a simple molecular magnetic system. The difficulties and results will be discussed, including the physical meaning of the orbital energies, and their shapes.

Keywords: quantum crystallography, unpaired electron density, constrained wavefunctions