

A new era in scanning transmission electron microscopy: from single-atom defect engineering and characterisation to vibrational spectroscopy

Q. M. Ramasse¹ D. M. Kepaptsoglou¹ F. S. Hage¹ C. R. Seabourne²
T. Hadcastle² T. Susi² J. Kotakoski³ U. Bangert⁴ A. J. Scott³

¹SuperSTEM Laboratory, SciTech Daresbury Campus, Keckwick Lane, Daresbury WA4 4AD, U.K.

²Institute for Materials Research, SCAPE, University of Leeds, Leeds LS2 9JT, U.K.

³Faculty of Physics, University of Vienna, Boltzmannngasse 5, A-1090 Vienna, Austria

⁴Department of Physics and Energy, University of Limerick, Limerick, Ireland

Modern aberration-corrected scanning transmission electron microscopes (STEMs) have been optimised to provide improved data collection ability and greater flexibility even at low acceleration voltages, spurring what could be arguably described as a new era in nano-scale materials characterisation. A wealth of complementary analytical signals is now available from a single experiment: when combining chemically-sensitive Z-contrast and bright field STEM imaging, 2D chemical mapping using analytical techniques such as electron energy loss spectroscopy (EELS) together with advanced image analysis, it is for instance possible to fully characterise minute chemical variations around nano-scale defects in complex oxide structures. These in turn are related to accurately measured atomic displacements and to the electronic properties of the material determined through theoretical calculations [1]. These developments were also a great benefit to the field of two-dimensional materials such as graphene. By reducing the acceleration voltage to overcome knock-on damage limitations, these structures can be imaged directly at atomic resolution, revealing for instance the successful low-energy ion implantation of single N or B dopants in graphene, a technique widely used by the semiconductor industry and with the potential to revolutionize graphene technology [2]. Furthermore, the sensitivity of EELS is such that it is possible to study how these atoms bond to one another: subtle differences in EELS fine structure can help distinguish unambiguously between tri- and tetravalent bonding configurations of single Si contaminants in graphene [2], while *ab initio* calculations are used to simulate experimental spectra and to rationalize the experiments [3,4]. These otherwise 'gentle' STEM observation conditions can also be precisely tailored to engineer and modify defects in 2-dimensional materials: the electron beam can thus drive the diffusion of substitutional Si dopants through graphene, one atomic jump at a time [5]. There are further exciting times ahead, with the advent of ultra-high energy resolution monochromators, which are boosting the resolution of STEM-EELS into the sub 10meV range and are now opening the

door to vibrational and phonon spectroscopy at unprecedentedly high spatial resolution [6].

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