A VOYAGE OF DISCOVERY: FROM MODEST BEGINNINGS TO DETECTING THE VIBRATIONS OF SINGLE ATOMS

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My interest in building instrumentation for electron microscopy (EM) begun when I was a post-doc at UC Berkeley, where I built, with key help on software from Peter Rez, a serial electron energy loss spectrometer (EELS) [1,2]. The instrument gave about 2.5 eV energy resolution, as needed for compositional analysis, but it had its problems too. I learned a lot from the experience and found that designing new instruments and using them to solve important scientific problems suited me to a T. What followed was a fun and fruitful voyage that involved designing instruments for EM, either alone or with a small team, and using them for pioneering applications.

The principal instruments and software that we developed include:

- parallel-detection EELS using a single magnetic sector and quadrupoles
- imaging filter using quadrupoles plus multipoles (for aberration correction)
- acquisition box for driving the beam in an SEM or STEM and digitizing signals
- EL/P EELS acquisition and quantification software
- DigitalMicrograph image acquisition and processing software
- aberration corrector for a STEM that set a key spatial resolution record
- whole scanning transmission electron microscope that includes a cold field emission gun, an aberration corrector for all aberrations up to 5th order, and an ultra-stable sample stage
- Python-based data acquisition and processing software called Swift
- ground potential monochromator for ultra-high energy resolution EELS
- ultra-high energy resolution EEL spectrometer
- other developments, such as an SE detector and an ultra-stable liquid He stage

The most remarkable of these developments was the combination of the ground potential monochromator, based on a concept I came up with in the 1990s but put aside until 2008, with the ultra-high energy resolution spectrometer and our ultra-stable STEM. It has led to an EELS energy resolution of 2.6 meV [3] – a nearly 1000x improvement relative to the first spectrometer I built.

The large improvement has opened a whole new experimental field: vibrational spectroscopy with atomic resolution. The results include analyzing the vibrations of single atoms, detecting quasiparticles such as magnons, measuring temperatures down to around 100 K with nm-scale resolution using gain-loss spectroscopy, and several other applications, all of which will be described in the talk.

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Comparing 2D and 3D phase imaging in TEM, STEM, and SEM

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Due to their high spatial resolution and wide range of signals that can be collected, electron microscopes offer a plethora of possibilities to characterize materials at the nanoscale. In this talk the focus will be on holographic techniques in the transmission- and scanning electron microscope (TEM & SEM) for mapping electrostatic potentials in a diverse range of thin specimen (see Fig. 1 for the three holography variants being compared). In addition to comparing their intrinsic strengths and limitations, recent results will be presented, including the visualization of the charge in a 2D electron gas [1] via a hybrid holography variant [2], the retrieval of the phonon dispersion from multi-object ptychography [3], sub-Ångstrom-resolution ptychography in an SEM (despite an effective illumination source size of 3.4 Å and an instrument specification of only 6 Å STEM-resolution), and the application of machine learning to optimize probe positions [4] (see Fig. 2).

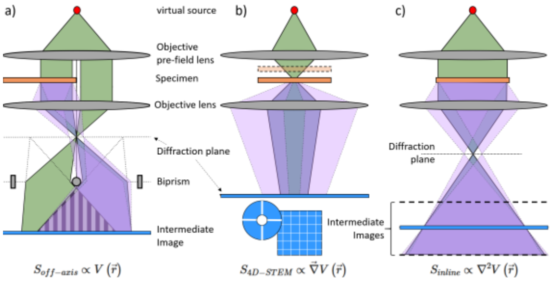

Fig. 1: Comparison of the different experimental setups for a) off-axis electron holography, b) 4D-STEM – with signal recording by a segmented (DPC) detector or a pixelated detector, and c) inline electron holography.

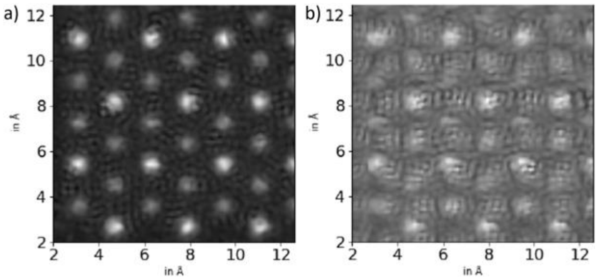

Fig. 2: Ptychographic reconstruction from diffraction patterns simulated for 59 probe positions on MoS_2. a) with optimized probe positions, b) with probe positions on a cartesian grid.

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Atomic Coordination Measurement at the Single-Atom Limit

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Atomic coordination governs material functionality, yet its quantitative analysis has been primarily accessible only through ensemble-averaged spectroscopic techniques such as extended X-ray absorption fine structure (EXAFS) spectroscopy. This averaging obscures local structural heterogeneity that is often critical at interfaces, defects, dopant sites, and catalytically active sites. Here we demonstrate that extended energy-loss fine structure (EXELFS) spectroscopy in the electron microscope, mechanistically analogous to EXAFS but employing an electron beam instead of X-rays, can overcome this limitation to provide element-specific coordination information at atomic resolution and with single-atom sensitivity. Specifically, we resolve atomic-layer-by-atomic-layer coordination and bond-length disorder across the epitaxial graphene/SiC interface, and distinguish the coordination of individual Si impurities in graphene lattice. These results establish an atomic-scale coordination probe in electron spectroscopy, reshaping how coordination and local order can be experimentally accessed in complex materials.

Geometric Constraints on Quantum Measurement Revealed by Spatially Resolved EELS

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For most of the 20th century, the large size of electron probes restricted our view of nanoscale physical science to electron scattering measurements of energy and momentum. Thus, clusters of nanoparticles showed strong surface plasmons, but very little bulk behavior, suggesting that small structures are dominated by surface/bulk ratio considerations.[1] When nm probes became available, spatially resolved EELS produced local, probe-specimen scattering, rather than momentum resolved averages over many particle volumes. [2] These new results contributed to an exciting period of EELS equipment development aimed at high spatial and energy resolution that continues today.[3]

Theoretical work began with plane waves, to predict energy loss spectra as a function of transferred momentum. Energy transfer was assumed to be *stochastic*, carrying random phase information after scattering. In 1976, Rose proposed a Mixed Dynamical Form Factor to describe simultaneous scattering of waves having well known phases to explore complex shapes of surface plasmons.[4] Nearly 20 years later, in 1993, I used electron probe channeling to cleanly separate plasmon from inter-band scattering in diamond, based on the even or odd lateral parity of the scattered electron wavefunction.[5]

After the success of sub-Ångstrom imaging using aberration correction at the end of the 20th century, observation of individual particle motion under a nanoscale electron beam became obvious, but still difficult to understand.[6] Generally an attractive dielectric response charge would dominate the force between a passing keV electron and a metal particle. Yet for very close approaches, nanoparticles were forced away from the electron beam. Using *time-dependent* calculations based on electromagnetic forces, we found that repulsive magnetic forces overcame dielectric forces at small distances.[7]

These findings of significant dependence on space and time, as well as their *Duals*, momentum and energy, suggest that EELS presents unique capabilities to measure quantum systems that simultaneously display *Dual Behavior*, allowing simultaneous measurements of Dual variables, even near Uncertainty Principle limits.[8] When Dual behavior exists, the geometric space-time algebra of Hestenes can be used to simplify calculations by describing electromagnetic response within a single wavefunction, based on a four dimensional, space-time energy density, and subject to operators constructed to respect well known relationships among electric and magnetic fields.[9] In addition, this kind of treatment couples with ongoing work seeking to unify quantum gravity and electromagnetic behavior, through the AdS-CFT correspondence, a recent conjecture that relates electromagnetic Conformal Field Theory to quantum gravity using String Theory in the Anti-de Sitter geometry that describes space-time near a black hole.[10]

There are experimental results that support this discussion. To illustrate this, I will show a 1973 momentum-energy resolved EELS experiment from Vincent and Silcox [11] that can be Fourier analyzed and transferred to a space-time Penrose Diagram, showing a strong resemblance to entropy information exchange through an event horizon of a Black Hole, described recently by Penington and co-workers.[8]

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UNVEILING PHONON-ASSISTED PROCESSES USING ATOM-WIDE ELECTRON BEAMS

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Abstract

The development of atom-sized monochromatic electron beams in scanning transmission electron microscopes has made possible the realization of phonon spectroscopy studies on nanomaterials with (sub)-nanometer spatial sensitivity. Specifically, spectroscopy and imaging of phonons in both real and reciprocal space can be performed using spatially and momentum-resolved monochromated EELS. These capabilities are enabling us to reveal phonon-assisted processes in the realms of infrared nanophotonics and nanoscale heat transfer.

In this talk, we will describe progress in these two areas by highlighting two main components: (i) Imaging and spectroscopy of hyperbolic phonon polaritons in twisted low-symmetry crystals. We fabricated suspended twisted α-MoO_3 structures and probe their phonon polariton response down to the far IR range (35 meV). The focus is on the understanding the excitation and propagation of hyperbolic phonon polaritons using fast electrons. Spatially confined canalization patterns that depends on the twist angle emerge from phonon coupling, offering opportunities to tune the IR response of engineered twisted materials. (ii) Studies of the phonon response across a chemically diffuse and strained AlN/GaN interface. We identified a ~ 4 nm layer of $Al_xGa_{1-x}N$ with non-uniform stoichiometry and under strain gradient (1 - 5%) across the AlN/GaN interface. The phonon behaviour was mapped and studied along high-symmetry crystallographic directions (i.e., Γ-A, Γ-M). A strong phonon hardening effect on optical phonon modes is identified.

These results from the first component advance our understanding of hyperbolic polaritons in twisted optics by extending knowledge of phonon coupling in twisted layers. Meanwhile, the second component of the work provides physical insights into factors such as strain and stoichiometry that can be used to tune the phonon behavior at interfaces.