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Meeting-report

Measuring Phase and Symmetries in STEM-EELS

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Under certain conditions, STEM/EELS can be used to probe the phase and symmetry of nanoscale excitations. While demonstrations of this are now decades old [1], recent technologies provide a new way to measure polarization and symmetry of optical transitions at small lengthscales. For instance, electron beams with engineered phase can be used to induce optical transitions in samples with polarization sensitivity, such as using electrons with helical phase to probe chiral plasmonic excitations in nanoparticle clusters [2].

More recently, we developed an interferometric STEM EELS technique in which an electron coherently divided into two separate probes can induce a dipole excitation in a sample yet remain in a superposition, retaining phase coherence between the two paths [3]. This work built on our previous elastic interferometry [4–7]. In this experiment, the probing electron induces an optical dipole transition to an excited state in a sample at a superposition of two locations. In the case of the first experiment [3], both electron paths passed near an aluminum nanoparticle. When filtering for that part of the wavefunction that lost energy corresponding to an optical dipole plasmon transition in the nanoparticle, it was found that the two electron paths still interfered. That is, if the interferometer was initially tuned to have complete destructive interference at the output going to the EELS detector, introduction of the nanoparticle and inducement of a plasmon would imprint a relative π phase shift between the two paths, resulting in a maximum of counts at the detector. Conversely, if the interferometer output were initially maximized due to constructive interference, the phase shift induced by the plasmon excitation would result in destructive interference at the detector. This behavior can be seen in Figure 1c-d, which shows the interferometer output for both elastically and inelastically scattered electrons for the two complementary Mach-Zehnder output ports, as the dual probes are raster scanned across a single nanoparticle. The electrostatic potential surrounding the nanoparticle introduces a varying phase between the probes that can be modeled.

If one of the probe paths is incident upon the sample, electrons can get scattered away from the detector yet this process can still be measured using the STEM interferometer (Fig. 1e). Scattering of one probe path eliminates the destructive interference that occurs at the interferometer output, allowing a non-zero probability current to be incident on the detector. We demonstrated this type of so-called "interaction-free" measurement in an interferometric STEM arrangement [8] and have improved it using a more efficient direct electron detector and weak measurement probes (Fig. 1e). Possibilities of using this type of counter-factual measurement for STEM/EELS will be discussed [9].

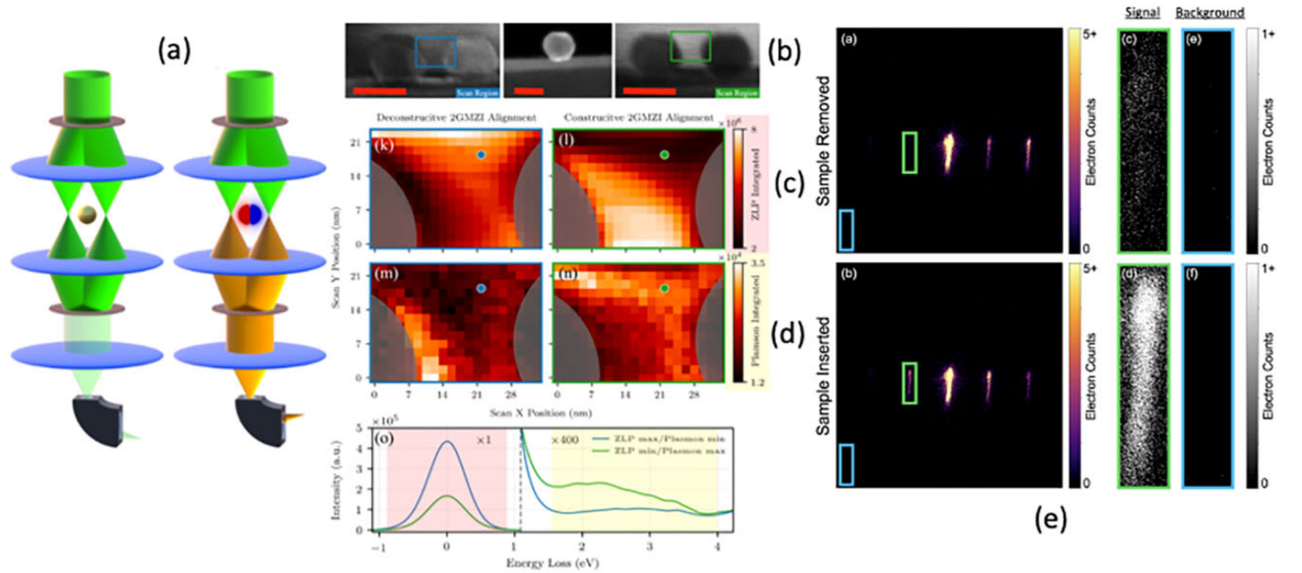

Fig. 1. *(a-d)* Inelastic interferometry of scattered electrons (*borrowed from* [3]). Schematics show the setup for interferometric STEM-EELS to probe an optical dipole excitation (plasmon) on an aluminum nanoparticle. The probes are coherently divided and the recombined by holographic beamsplitter gratings place before and after the specimen. (b) Dark field STEM image of a 60 nm gold nanoparticle (NP) isolated on the edge of a carbon substrate. The nanoparticle appears in double when a superposition of two beams is used as the imaging probe. Comparison of images of the space surrounding the particle using zero-loss (c) and plasmon-loss (d) energy peaks at the output of the interferometer show opposite contrast, indicative of a π phase shift introduced between scattered paths. (e) When one of the two probes is incident on the particle, electron counts in one of the STEM interferometer brightfield outputs *increases*, serving as a demonstration of "interaction-free" measurements [8].

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