Probing Plasmons Using Structured Electrons

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Electrons with non-trivial phase profiles provide a new way to probe the chirality and spatial coherence of nanoscale plasmonics. New developments in electron optics provide the ability to engineer the spatial phase of electron wavefunctions. One example of this are electron vortices featuring a spiral phase singularity and a corresponding quantized orbital angular momentum (OAM). Other examples, include electrons prepared in superpositions of two linear momentum states or two locations.

We previously reported early results demonstrating symmetry-breaking inelastic interactions between electron vortex beams and chiral nanoparticle clusters [1,2]. We observed that nanoparticle clusters with 3D structural chirality showed slight differences in their EEL spectra that depended on the sign of the OAM in the illuminating electron beam. We ascribed this to a situation in which OAM from the illuminating electron beam was transferred to a chiral surface plasmon mode, yet an independent model was needed to support this interpretation.

To that end, we adapted an analytical theory based on electromagnetic Green's functions [3] and applied it to understand how electron vortex modes might transfer OAM to our target specimens. Modelling a chiral cluster of Al nanoparticles as point dipoles, we use Fermi's golden rule to calculate the energy-dependent transition probability of transferring $1\hbar$ of OAM from an electron vortex beam to the nanoparticle cluster via the Coulomb interaction Hamiltonian. We show that when geometry and nanoparticle sizes in experimental systems are input to this model, the predicted energy-dependent transition probability shows a good qualitative agreement to the experimental spectra. As shown in Figure 2, the experimental dichroism data (gray) is well fit by a model (red line) that assumes a transfer of OAM from the electron vortex probe beam to the nanoparticle cluster.

Electrons coherently divided into two or more positionable probes, used recently for elastic interferometry [4–6], could also be used to probe plasmon modes using inelastic interferometry. This improved theoretical model can serve as a guide for experimental demonstrations of novel plasmonic interactions using structured electron beams. Improved nanoscale electron optics such as grating holograms that can produce isolated electron beams with better currents and phase fidelity could enable a new field of structured electron microscopy, spectroscopy, and interferometry.

References:

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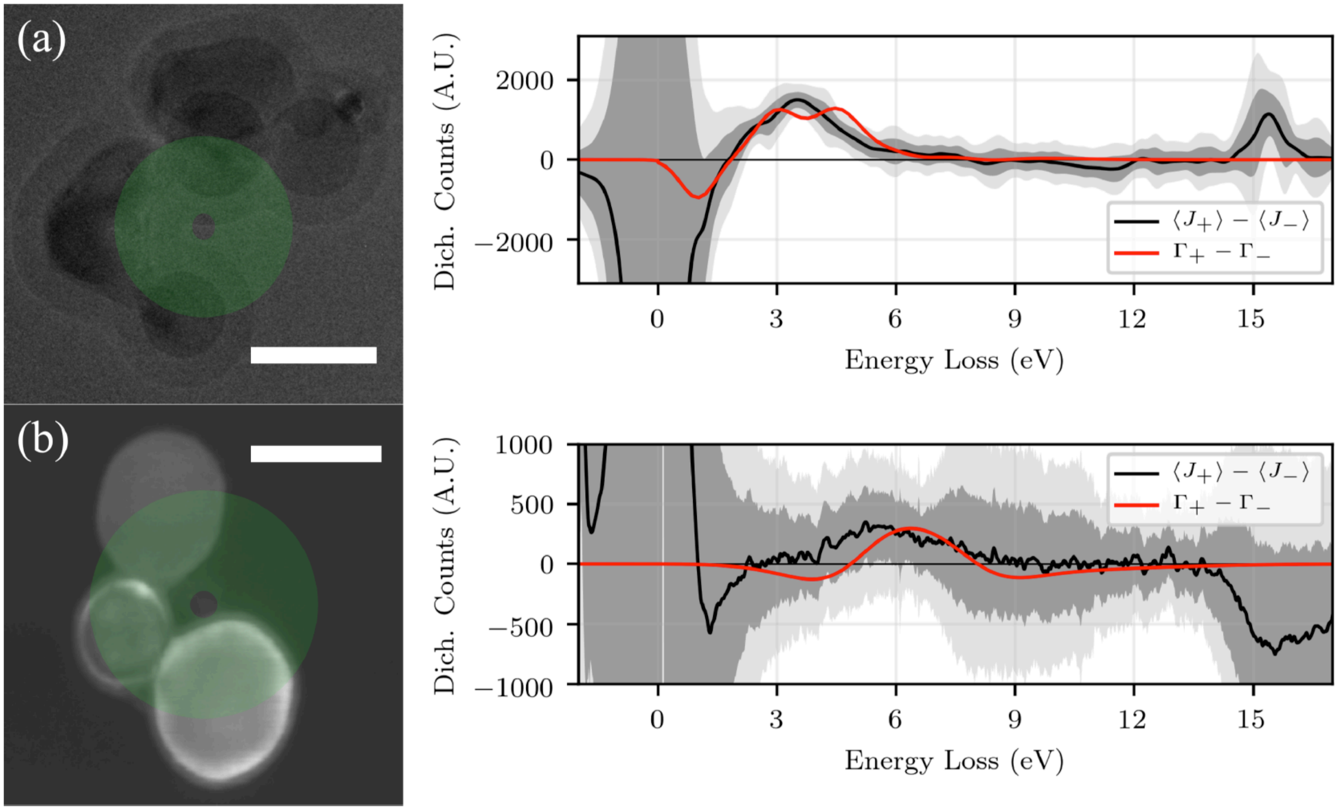

Figure 1. (a) TEM micrograph of an Al nanoparticle clusters (scale bar is 100 nm). The green annulus overlay denotes the position and size of an incident electron vortex. (Right) Experimental dichroic EELS spectrum shows the difference between measured counts $J_{\pm}(E)$ for $\langle L_z \rangle = \pm\hbar$ vortex beam probes. The dark gray region is within one standard deviation, light gray is within two. Red curve is the dichroic spectrum predicted by the theory. (b) Different experimental cluster of aluminum nanoparticles exhibiting extrinsic dichroism (scale bar is 50 nm).