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Electron Orbital Angular Momentum Transfer to Nanoparticle Plasmon Modes

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We observed the decay of an electron vortex beam from a state with orbital angular momentum $l = 1\hbar$ to $l = 0$ by interaction with a gold nanoparticle. Here, we present evidence which suggests that angular momentum is transferred to surface plasmon modes on the nanoparticle. Several optical studies have induced plasmon vortices using optical vortices and circularly polarized light and suggested their use in nanophotonic and plasmonic devices [1,2]. Direct observation of angular momentum transfer from electron vortices allows for unique identification of the orbital angular momentum associated with localized plasmon excitations down to the nanometer scale.

The electron vortex was produced in an FEI Titan transmission electron microscope (TEM) at 300 kV with large spot size and strong gun lens to maximize the coherence of the beam. A forked diffraction grating (Fig. 1) is located in the condenser lens aperture of the microscope. The first diffracted order of the grating is focused to a nanoscale probe in the plane of the sample to be analyzed; this beam carries $1\hbar$ orbital angular momentum [3]. To fully account for the interaction between the electron vortex and the 54 nm-diameter gold particle, we recorded a transmission electron micrograph of both the vortex beam on the electron-transparent substrate far from the nanoparticle (Fig. 2) and the vortex beam concentric on the nanoparticle (Fig. 3) at the same magnification and beam intensity. We also imaged the nanoparticle under normal illumination without orbital angular momentum (Fig. 4).

The peak intensity of the vortex beam forms a ring (Fig. 2). Inside this ring, the intensity of the beam drops nearly to zero, as the azimuthal phase term of a vortex beam, $e^{il\phi}$, is singular at the center of the beam. A gaussian beam with no angular momentum, however, has no phase singularity and the peak intensity is at the center of the beam. Thus, if an electron vortex beam with one unit of angular momentum transfers that angular momentum to a plasmon mode, the scattered component of the beam will be distinguishable as an increase in the intensity of the center of the beam.

This increase in the intensity of the center of the probe beam is exactly what we observed when the probe beam was concentric with a nanoparticle (Fig. 3). A comparison of the integrated radial intensity of the beam off the nanoparticle and on the nanoparticle (Fig. 6) clearly shows an absolute increase in intensity in the center of the beam. This central intensity increase is visible even though a significant fraction of the incoming beam intensity is lost to high-angle scattering in interaction with the particle. To isolate the intensity due to electron vortex-nanoparticle interaction, we subtracted the intensities of the vortex beam (Fig. 2) and nanoparticle (Fig. 4) from the micrograph of the vortex-nanoparticle interaction (Fig. 3). The resulting image (Fig. 5) of scattered intensity shows a clear peak in the center of the vortex beam. The calculated peak scattered intensity is 16% of the original unscattered vortex beam peak intensity (Fig. 6)

We have thus observed transfer of orbital angular momentum from an electron vortex to a gold nanoparticle. Preliminary electron energy loss spectroscopy (EELS) data from the vortex on the nanoparticle indeed shows increased loss peaked at 2 eV. Further work with energy-filtered TEM can better identify the energy losses corresponding to transfer of orbital angular momentum. As forked diffraction gratings which generate beams with up to $100\hbar$ angular momentum have been demonstrated [3], the technique we demonstrate here can be repeated for electron vorticies with any integer number of angular momentum. Thus, this technique makes possible the identification of localized plasmon modes of specified angular momentum on nanoparticles and nanostructures with high spatial resolution.

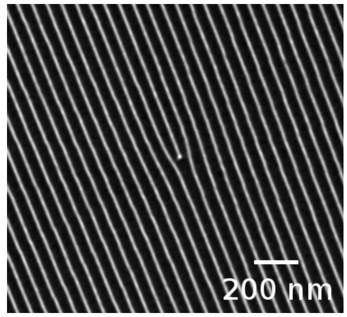

Figure 1: Transmission Electron Micrograph of the single-forked diffraction grating used to produce electron vortices with $n\hbar$ *orbital angular momentum in the* n*th diffraction order.*

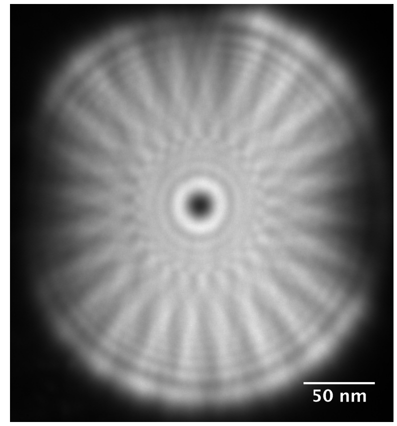

Figure 2: Micrograph of Electron Vortex away from Nanoparticle; taken under identical microscope conditions used to record the vortex-nanoparticle interaction.

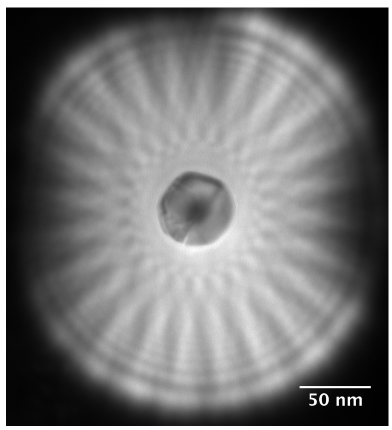

Figure 3: Micrograph of Electron Vortex on Nanoparticle. A slight intensity increase in the center of the vortex suggests transfer of orbital angular momentum.

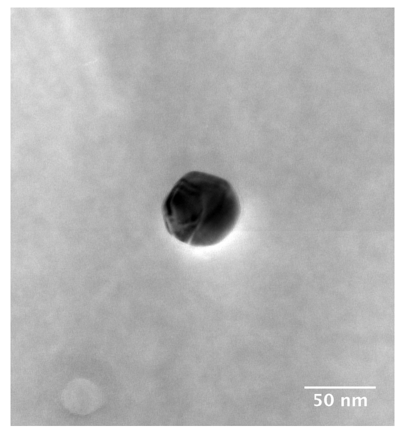

Figure 4: Micrograph of Nanoparticle under normal illumination used as a reference to subtract the unscattered beam in Figure 5.

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[3] B. J. McMorran *et al.*, Science **331** (2011) p. 192.

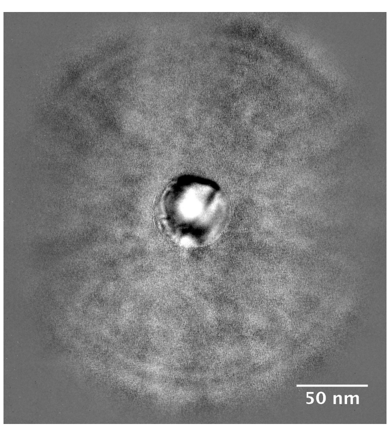

Figure 5: Intensity attributed to electron vortex-nanoparticle interaction. This isolation of vortex-specific scattering was constructed by the subtraction of Figures 2 and 4 from Figure 3.

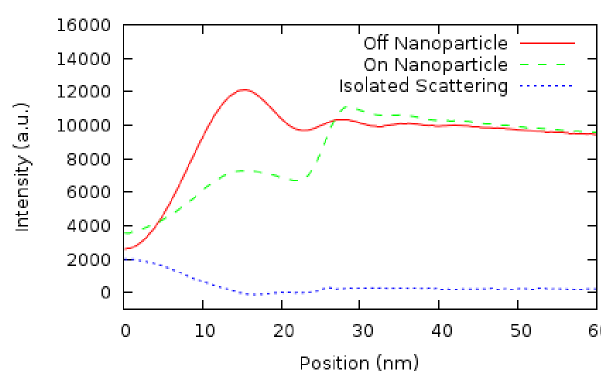

Figure 6: Comparison of the radial integrated intensity of the electron vortex off the nanoparticle (Fig. 2), on the nanoparticle (Fig. 3), and the isolated scattering due to the electron vortex-nanoparticle interaction (Fig. 5).

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