

## Imaging Chiral Materials with Photon-Induced Near-Field Electron Microscopy

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Chirality is central to a number of open scientific questions and technologically relevant materials, including broken symmetry in the biochemistry of life, CP violation, magnetic skyrmions and metamaterials. The sub-nanometer resolution routinely achievable in transmission electron microscopy could improve our understanding of chirality. Three-dimensional chirality can only be characterized by tomography or with a probe that has controllable helicity. The latter approach allows for investigation of chiral quantum states and transitions that have no direct link to three-dimensional structure. Some progress has been made toward such a technique with electron vortices [1-3]. Another approach is to use electrons to probe the interaction between circularly polarized light and chiral materials.

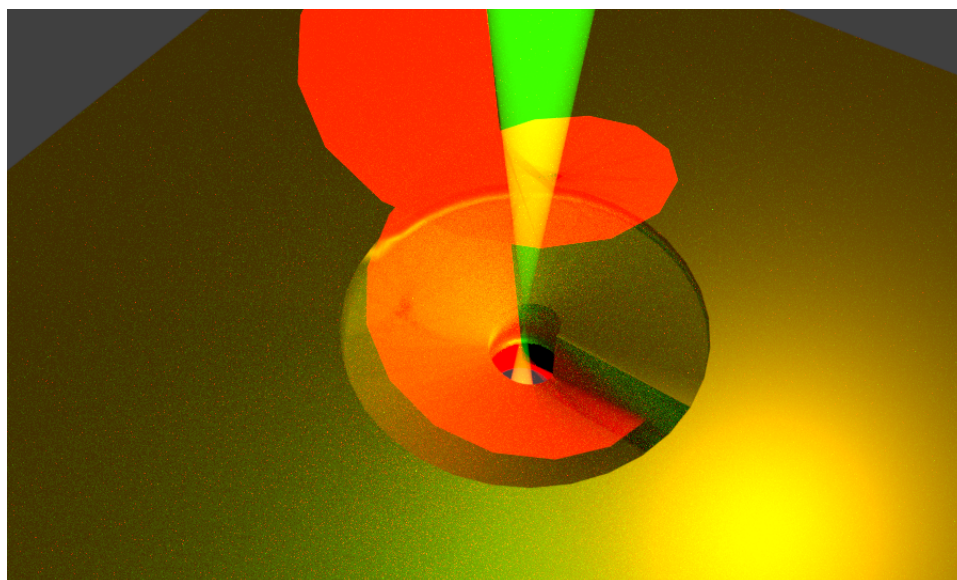
Although absorption or emission of a single photon by an electron is forbidden in free space by energy-momentum conservation, this interaction is possible in the presence of a surface. A surface breaks translation symmetry and allows for coupling between electron momentum and the electromagnetic field amplitude [4]. The strength of this coupling,  $g$ , depends on the shape and optical properties of the surface, as well as the incident optical power [5]. Because electron beams can be focused to sub-nanometer spots in electron microscopes, it is also possible to map inelastic electron-light scattering on the same lengthscale. This technique, called photon-induced near-field electron microscopy (PINEM), can be employed to image plasmonic modes and optical properties with nanometer spatial resolution.

We demonstrate the ability to probe chirality with PINEM. By illuminating a sample with left- and right-circularly polarized light (see Figure 1) and measuring the difference in coupling strength with electrons, we probe chiral optical near fields with nanometer spatial resolution.

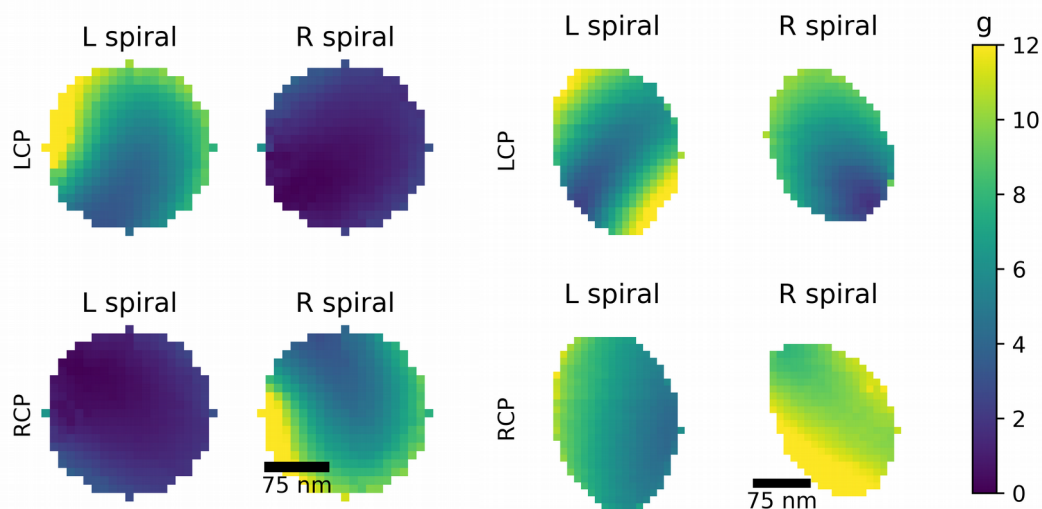
We use a FIB-milled spiral gold structure as a prototypical chiral sample. We find, as predicted by finite-element simulations, that there is a difference in the average coupling strength for left and right-circularly polarized illumination on a structure of one handedness, as well as spatial variations linked to the particular shape of the chiral structure (Figure 2). Dichroism (Figure 3) is opposite for the structure of opposite handedness. This technique may enable the investigation of chiral optical and electronic states in plasmonic nanostructures, molecules and atoms with sub-nanometer spatial resolution.

### References:

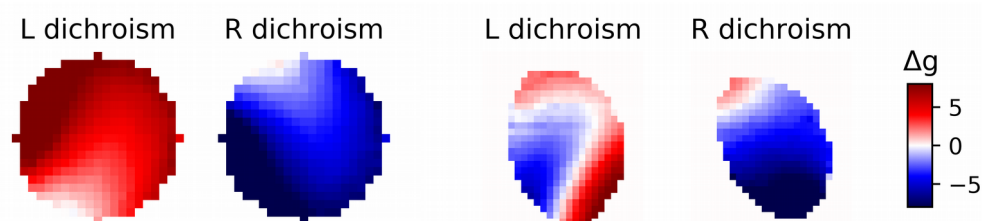
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**Figure 1.** Illustration depicting circular dichroism experiment with PINEM. A circularly polarized optical beam (red) illuminates a chiral structure (gold), while the electron beam (green) probes it.



**Figure 2.** (left) Simulated coupling strength  $g$  for left- and right-circularly polarized (LCP, RCP) illumination on left- and right-handed spirals. (right) Experimentally measured  $g$  (color bar shared).



**Figure 3.** (left) Simulated dichroism  $\Delta g$  calculated by subtracting RCP-illumination  $g$  from LCP-illumination  $g$ . (right) Experimental dichroism (color bar shared).