Probing the chiral behavior of nano-optical near-fields through angular momentum resolved PINEM

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Background incl. aims

Usually performed in a ultrafast transmission electron microscope (UTEM), photon induced near-field electron microscopy (PINEM) consists in mapping the magnitude of an optical near-field pumped by a laser with a fast electron beam [1]. Benefiting from the sub-nm spatial resolution of TEMs as well as the high fluence and energy resolution of ultrashort lasers, this technique can probe deep-subwavelength structural details of the optical near-field in resonant and non-resonant structures.

Methods

In addition, and contrary to electron energy-loss spectroscopy (EELS), PINEM displays a high degree of selectivity thanks to the laser illumination. For example, by properly tailoring e.g. the wavelength and polarization of the laser, one can selectively excite individual optical modes [2]. This idea can be extended to probe the chirality of nano-optical near-fields. Indeed, as first demonstrated by Harvey and collaborators in 2020 [3], one can perform a so-called “chiral PINEM” experiment – where a right- and left-handed polarized laser beam is sequentially used to excite the structure and measure the differential PINEM signal. In that configuration, spin angular momentum (SAM) of the incoming beam is tuned so that this technique is denoted as SAM-dichroic PINEM (see figure 1).

Nevertheless, by shaping its phase profile, a laser beam can also carry orbital angular momentum (OAM), and a long-standing question remains on the difference between OAM- and SAM-based dichroic experiment [4]. A natural candidate to tackle this issue is thus a combined SAM-dichroic and OAM-dichroic PINEM experiment – an experimental scheme that we will refer to as angular momentum resolved PINEM (AM-PINEM).

Results

In this conference, we will present our latest work on angular momentum resolved PINEM, showing its application to the probing of the coupling between chiral light and optical near-fields. In a first part we will show that the theoretical framework based on the local density of states (LDOS) used to explain EELS and CL experiments [5] can be extended to PINEM, combining in an elegant way these three main TEM spectroscopies. In particular, we will demonstrate that AM-PINEM can be connected to the concept of chiral radiative LDOS.

In a second part we will focus on the numerical simulation of AM-PINEM experiments using pyGDM - an electrodynamics simulation toolbox based on the green dyadic method that we enhanced to simulate polarized electron spectroscopies. The large library of different illuminations allows for simulation of both SAM- and OAM-resolved PINEM experiments. We will focus on practically realizable structures such as the gold trimer (see figure 1) in which OAM- and SAM-dichroism present clear differences.

Conclusions

In a last part, we will show our first experimental results on SAM-dichroic PINEM realized with a new cold field emission UTEM on simple achiral structures, revealing dichroic behaviors at the deep sub-wavelength scale. Finally, we will present our preliminary work towards the realization of the OAM-dichroic PINEM experiments.

Graphic:

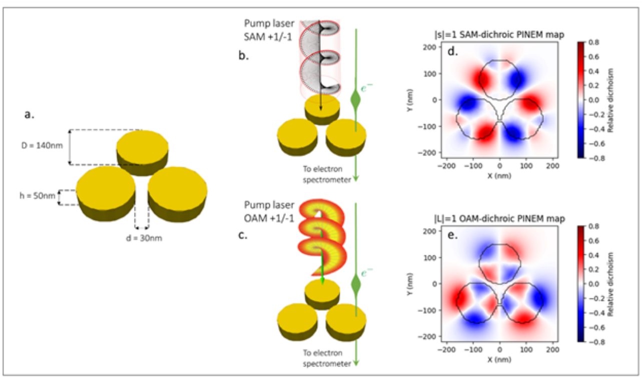

Figure 1 - (a) Gold trimere in vacuum with dimensions, **(b,c)** representation of the dichroic PINEM experiment where the sample is pumped with **(b)** SAM +1/-1 beam, **(c)** OAM +1/-1 beam at 650nm wavelength. **(d,e)** Corresponding resulting dichroic maps. Dichroic maps are made by computing the difference of two maps with the same norm of angular momentum but opposed helicity for the illumination beam.

Keywords:

PINEM, Angular momentum, Numerical, nano-optics

Reference:

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