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Imaging of localized surface plasmonic field at nanoscale by UEM

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The Ultrafast Electron Microscopy (UEM) is one powerful tool to study the ultrafast dynamics of light-matter interaction at a high temporal resolution of sub-picosecond time scale that is 10 orders better than the conventional TEM.[1] The UEM has been widely used in physics, chemistry and materials science. Other than light-matter interaction observed in UEM, the investigation of photon-electron interaction has been enabled by one unique technique called Photon-Induced Near Field Electron Microscopy (PINEM) developed in UEM to capture the evanescent electromagnetic field on its intrinsic time scale and nanometer scale.[2, 3] In free space, the photon-electron interaction is forbidden due to the energy-momentum mismatch. However, the energy-momentum mismatch can be eliminated through the altered dispersion relation of photons using materials. In PINEM, the free electrons gain or loss quantized photon energy when electrons overlap with photons spatiotemporally near the nanostructures or interfaces, where the phase velocity of photons is reduced to the group velocity of free electrons due to the spatial confinement. The images formed by those electrons gaining photon energy are called PINEM image for plasmonic field mapping in UEM.

Surface plasmons, collective oscillating charge wave at the surface of metals or interface of metals/dielectrics under light excitation, are among the most attractive candidates for next-generation information revolution due to their ability to extremely confine electromagnetic field and empower strong coupling of light and matter beyond the diffraction limit. The localized surface plasmon resonances (LSPR) of a nanoscale metal particle have been widely used in biological sensing [4], waveguiding [5], Surface-Enhanced Raman Scattering (SERS) [6], etc. LSPR resonance energies are highly sensitive to the shape, size, and composition of the plasmonic materials and the surrounding dielectric environments.[7] Therefore, investigation of the spatial distributions and temporal evolution of the electromagnetic field at a high spatiotemporal resolution will be very critical for plasmonics, biomedical sensing, quantum information and processing within the scientific community.

There are not enough of such kinds of equipment with high spatiotemporal resolution like UEM that the scientists can get access to in the world. We recently established one state-of-art UEM user facility at the Center for Nanoscale Materials (CNM), Argonne National Laboratory. Besides the capabilities of imaging and diffraction, the UEM at the CNM is equipped with one GIF spectrometer, which enables it to work in energy filtering mode and record the PINEM image. We have demonstrated its success in capturing the optical excited evanescent electromagnetic field of the silver nanoparticle and copper grid edge on our UEM platform. Fig.1a shows the UEM established at the CNM with capabilities of imaging, diffraction and spectrum, which is modified from one JEOL 2100 Plus microscope with a LaB_6 gun. Fig.1b is one bright field image of silver nanoparticle with diameter of 100 nm. Fig. 1c-e show the PINEM images recorded at the delay time of t=0 ps, which represent the captured evanescent plasmonic field distributions within 100 nm range around the nanoparticle under 1030 nm optical excitation different light polarizations, respectively. Fig. 1f and g shows the copper grid edge and its PINEM image at delay time of t=0 ps under 515 nm optical excitation. As seen from the recorded PINEM images, the electromagnetic field around the nanoparticle or copper grid edge and vacuum is enhanced and confined within 100-160 nm range, which is far bellow the optical excitation wavelength. The visualization of the spatial distribution and temporal evolution of the evanescent electromagnetic field near the nanostructures or interface in femtoseconds time scale and at nanometer scale by UEM is critical for the nanophotonics applications.

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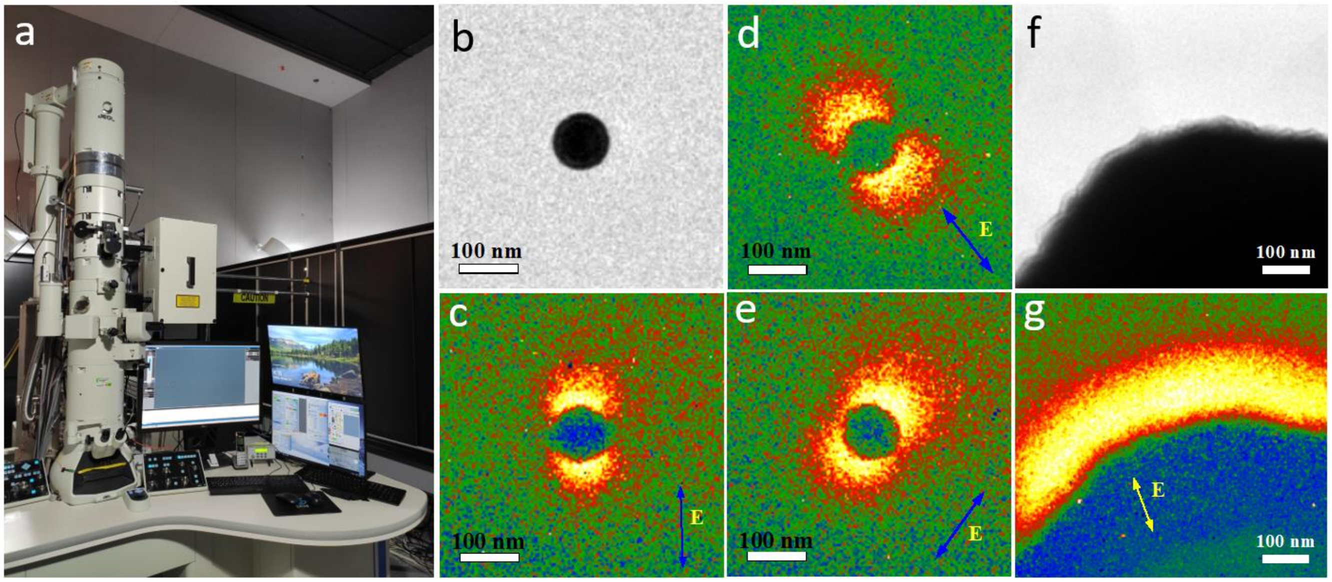

Figure 1. Figure 1. a. UEM at the CNM. b Bright field image of one silver nanoparticle. c-e. PINEM images of the silver nanoparticle obtained at the delay time of 0 ps under 1030 nm optical excitation with different light polarization, respectively. The laser fluence is 0.5 mJ/cm2. f. The bright field of the Cu grid edge. g. PINEM image of the Cu grid edge under 515 nm optical excitation at delay time of 0 ps with the laser fluence of 3.2 mJ/cm2.

References

[1] Zewail A. H. Four-Dimensional Electron Microscopy, *Science*, **328**, 187-913 (2010).

[2] Barwick B., David J. Flannigan & Ahmed H. Zewail. Photon-induced near-field electron microscopy, *Nature*, **462**, 902-904, (2009).

[3] Liu H.H., Baskin, J. S., and Zewail, A. H. Infrared PINEM developed by diffraction in 4D UEM, *PNAS*, **113**, 2041-2046 (2016).

[4] Anker, J. N. et al. Biosensing with plasmonic nanosensors.*Nat. Materials***7**, 442–453 (2008).

[5] Maier, S.A., Atwater, H.A. Plasmonics: Localization and guiding of electromagnetic energy in metal/dielectric structures. *J. App. Phys.***98**, 011101 (2005).

[6] Laura, R. et al.Zeptomol detection through controlled ultrasensitive surface-enhanced Raman scattering.*J. Am. Chem. So*c. **131**(13), 4616–4618 (2009).

[7] Kelly, K. L., Coronado, E., Zhao, L.L., Schatz, G. C. The optical properties of metal nanoparticles: The influence of size, shape, and dielectric environment. *J. Phys. Chem. B.***107**(3), 668-677 (2003).