A Hybrid Environmental Transmission Electron Microscope for Probing Plasmons and Excitons

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The applications of transmission electron microscope (TEM) related techniques have extended from *ex situ* nanoscale characterization of structure and chemistry of reaction products to dynamic measurements of nanostructures during reaction processes. Commercially-available modified TEM specimen holders and TEM columns (e.g. environmental scanning-transmission electron microscopes or ESTEMs) are being routinely employed to follow the structural and chemical changes at elevated temperatures and/or under controlled atmospheres. The combination of atomic-resolution images and high spatial and energy resolution electron energy loss spectroscopy (EELS) has successfully revealed the functioning of catalyst nanoparticles. Recently, plasmonic nanoparticles, such as Au, Ag and Al, have been reported to initiate gas molecule dissociation at a lower reaction temperatures, even as low as room temperature. However, the optical methods used so far lack the spatial resolution to reveal the reaction sites. Here, we reveal the gas adsorption sites and room-temperature chemical reactions assisted by Au nanoparticles by using an ESTEM (modified column), operated at 80 kV, equipped with monochromated field emission electron source (gun) and image corrector.

We utilize a combination of ESTEM based techniques such as low-loss and core-loss EELS maps, STEM and high-resolution TEM images, and electron diffraction, to identify the gas adsorption sites, morphological changes, structural modifications and reaction products. While the low-loss EELS provides the information about the local surface plasmon resonance and bulk plasmon energies in vacuum and gas environment, core-loss EELS is used to characterize the reaction products.

Moreover, we have designed and built a unique platform that has extended *in situ* spectroscopy measurements to include micro-Raman and cathodoluminescence (CL). We have used this correlative microscopy platform i) to measure the temperature from a 60 m^2 area using Raman shifts, ii) to investigate light/matter interactions in plasmonic particles iii) to perform concurrent optical and electron spectroscopy such as EELS, Raman and/or CL. Details of the design, function, and capabilities of the optical spectrum collection platform will be illustrated using the results obtained for hydrogen dissociation on Au and Al nanoparticles, room temperature CO disproportionation reaction on Au nanoprisms and electron-hole recombination rates in CdTe solar cells.