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New Advances in Stimulated Electron Energy Gain and Loss Spectroscopy

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Electron Energy Gain Spectroscopy (EEGS) and Stimulated Electron Energy Loss Spectroscopy (sEEGS) are powerful tools capable of yielding information on local excitations of nanostructured systems using a Transmission Electron Microscopes (TEM) equipped with optical illumination [1]. We report the application of an innovative *in situ* optical delivery system for the TEM [2]. The optical probe contains three independent optical channels that can deliver different discrete laser wavelengths or tunable broadband sources. Each optical channel can produce continuous wave (CW) or pulsed irradiance. The focused laser spot (< 10 mm diameter) is positioned with a computer-controlled nanomanipulator and can be configured for high vacuum and column bake. This non-contact laser irradiation system can also be used for simultaneous pinpoint sample heating with minimal thermo-mechanical drift and rapid temperature slew rates.

EELS in the TEM has revealed valuable information about the unique optical properties of surface plasmon resonances in nanostructured materials with nanoscale spatial resolution. This novel optical delivery system has been used to experimentally explore the photon-stimulated sEELS and EEGS responses of individual plasmonic nanoparticles via the simultaneous irradiation of a continuous wave laser and a monochromated electron probe.

Using a 215 mW 785 nm single-mode fiber coupled laser diode producing an ~8 µm diameter Gaussian spot, Liu et al. reported sEELS and EEGS responses of individual plasmonic nanoparticles [3]. By linearly varying the laser irradiance in the range of $0.5–4\times10^8$ W/m^2 , Liu et al. were able to observe equivalent sEELS and EEGS probabilities without excessive heating. The group used theoretical modeling of the rod-like nanostructures to produce an experimental spectra which helped uncover the role of cathodoluminescence in stimulated electron-energy loss events. This study is just the beginning to unlocking key components in quantum physics of excited plasmons [4].

Collette et al. used continuous wave photon sEELS and EEGS to image the near field of optically stimulated LSPR modes in nanorod antennas. The experiment was designed such that the nanorod lengths would resonate with the 785nm laser at modes of 1, 2, and 3. This allowed them to explore retardation effects as well as the effects of different nanorod orientations. They observed that odd modes produce bright EEGS responses and even modes can be tilted to produce an EEGS response. This was confirmed through electron and optical beam simulations. These observations show that this novel optical delivery system is an effective tool for imaging the full set of plasmon modes of either parity [5].

The opportunities for combining optical illumination with electron spectroscopes in the monochromated TEM are now emerging. The application of this new approach to the characterization of cutting-edge material systems is now underway.

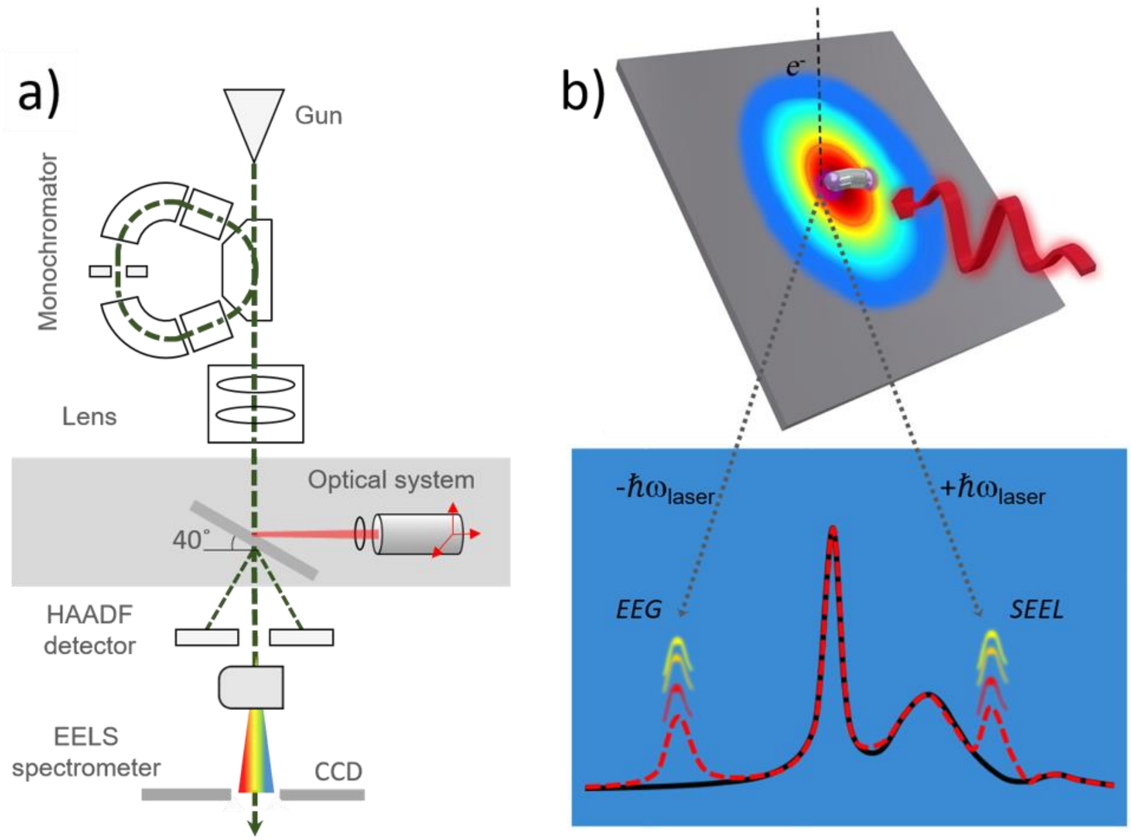

Figure 1. a) Schematic of the monochromated (S)TEM/EELS Instrument with the optical delivery system mounted orthogonal to the electron beam. b) Illustration of the coincident and continuous focused laser light and 200 keV electron beam; the laser spot has a 3.7mm radius Gaussian profile and interacts with the samples to produce signature SEEL and EEG peaks whose intensities vary with laser irradiance.

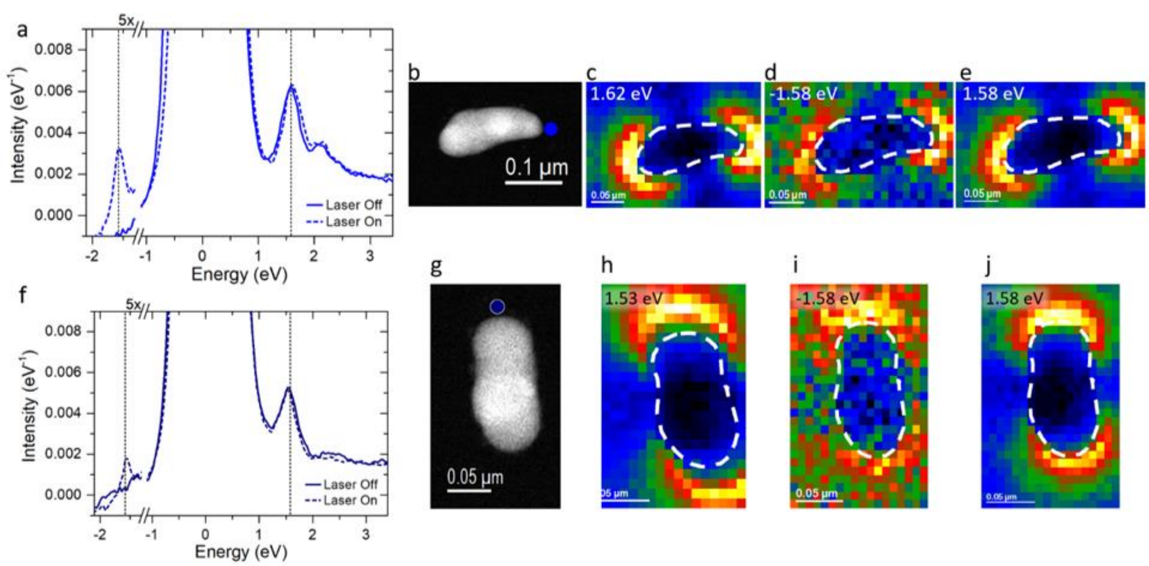

Figure 2. (a) Horizontal nanorod aloof 6-pixel map spectra average with laser off and on. Dashed lines in a and f correspond to the laser energy at ±1.58 eV (b) HAADF image of horizontal nanorod with aloof position indicated by blue circle. (c-e) Horizontal nanorod maps of EEL, sEEG, and sEEL peak intensities, respectively. (f) Vertical nanorod aloof 6-pixel map spectra average with laser off and on. (g) HAADF

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image of vertical nanorod with aloof position indicated by blue circle. (h-j) Vertical nanorod maps of EEL, sEEG, and sEEL peak intensities, respectively.

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