## In Situ Laser Heating and Excitation in the (Scanning) Transmission Electron Microscope for Real time imaging and excited state spectroscopy

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A new in situ optical delivery tool for the (scanning) transmission electron microscope ((S)TEM) has been developed by Waviks Inc. The applications of this tool include imaging high temperature micro- and nanostructure evolution and excited state materials phenomena at the nano and atomic scale. The tool is mounted on a Zeiss Libra 200 (S)TEM system and contains two optical delivery channels: the current configuration houses two laser diodes (785 nm and 473 nm). The both laser diodes are coupled to a 5 µm mode field diameter single-mode fiber capable of delivering ~200 mW to the sample surface. The lasers can be operated in continuous wave (cw) or repetitive mode with pulse widths as low as a few ns and repetition rates up to 16 MHz can be achieved. Spatial control of the system is achieved via a 3 axis (+/-x,y,z) nanomanipulator; the x and y positions are used to align the laser spot location coincident to the electron beam, while the z position adjusts the focal position. There is an offset between the 785 nm and 473 nm lasers of approximately 500 µm in the z direction (due to the chromatic aberration) and 100 nm in the y direction due to the bundled fibers positions. The optical delivery tool is mounted perpendicular to the electron gun; thus, the sample is tilted (~30-45 degrees) relative to normal to the electron beam. The system contains a lens system to re-image the fiber optics (1x) at a working distance of ~10 mm, which is long enough eliminate charging and minimizes re-deposition of material. The functionality of the tool is demonstrated through photothermal annealing and dewetting of a supersaturated Ag-Ni thin film, and excited state electron energy gain spectroscopy (EEGS) of plasmonic silver nanoparticles. The Ag-Ni work will demonstrate the flexibility and control available using various laser powers, pulse widths, pulse numbers, and laser radii, while the EEGS illustrates the capabilities of a dual laser configuration.



*Figure 1.* Picture of optical delivery system (red circle) mounted on TEM (*top left*). Example of core-loss and low-loss EELS mapping of partially dewet Ag-Ni and a Ag-Ni Janus particle. The core-loss colors in a) and d) correspond to Ag (red) and Ni (green). The low-loss maps illustrate various plasmon resonances in the bimetallic nanoparticle (*top right*). Example of a laser spot in a Ag thin film (*bottom right*).