Supporting Information:

Nonlinear Photoluminescence Spectrum of Single Gold Nanostructures

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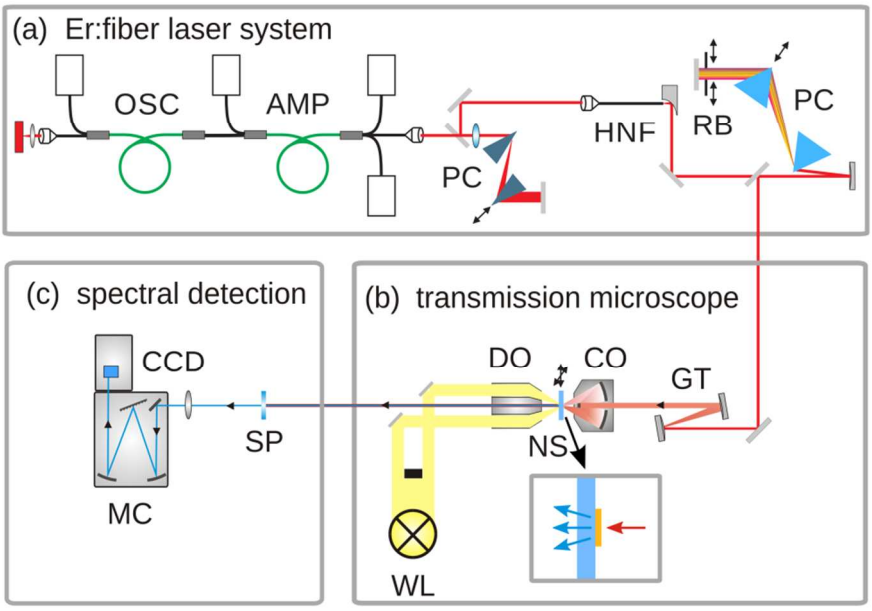

Figure I. Experimental setup for nonlinear spectroscopy of single nanostructures. (a) Diode-pumped Erbium:fiber laser system with supercontinuum generation and pulse compression. OSC, oscillator; AMP, amplifier; PC, prism compressor; HNF, highly nonlinear fiber; RB, razor blade for spectral selection. (b) Transmission microscope with piezo-actuated stage and nanostructures. GT, Galileian telescope; CO, Cassegrain objective; NS, nanostructures; DO, dark field objective; WL, white light source. The dispersionless Cassegrain objective enables few-

femtosecond pulse durations in the sample plane. (c) Spectral detection unit. SP, short pass filter; MC, monochromator; CCD, charge coupled device.

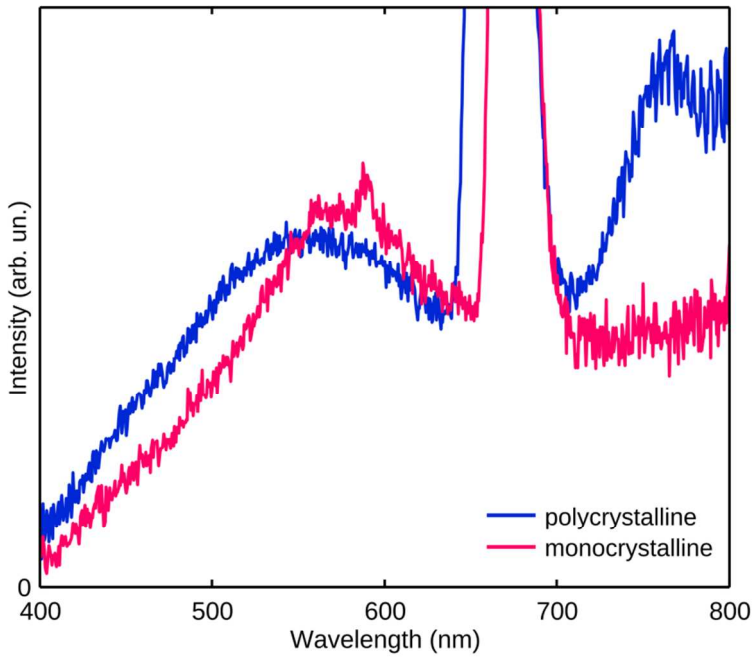

Figure II. Typical PL spectra obtained from a polycrystalline (blue graph) and a monocrystalline (red) nanorod under excitation at a wavelength of 2000 nm. The pulse energy was set to 50 pJ and 43 pJ, respectively. Rod width was 50 nm in both cases. Rod length was significantly longer than for excitation at a fundamental wavelength of 1560 nm: 470 nm for the polycrystalline rod and 350 nm for the monocrystalline specimen, emphasizing again the resonance shift between the two crystallinity types. For comparison, the nanoantennas used for the spectra in Figure 1 resonant to 1560 nm have a length of 370 and 310 nm, respectively. Consequently, the red photoluminescence peak in the polycrystalline sample is red-shifted according to the position of the second-order resonance in the longer nanoantenna. The small peak at a wavelength of 590

nm in the spectrum from the monocrystalline rod originates from parasitic laser radiation that was not fully filtered out.

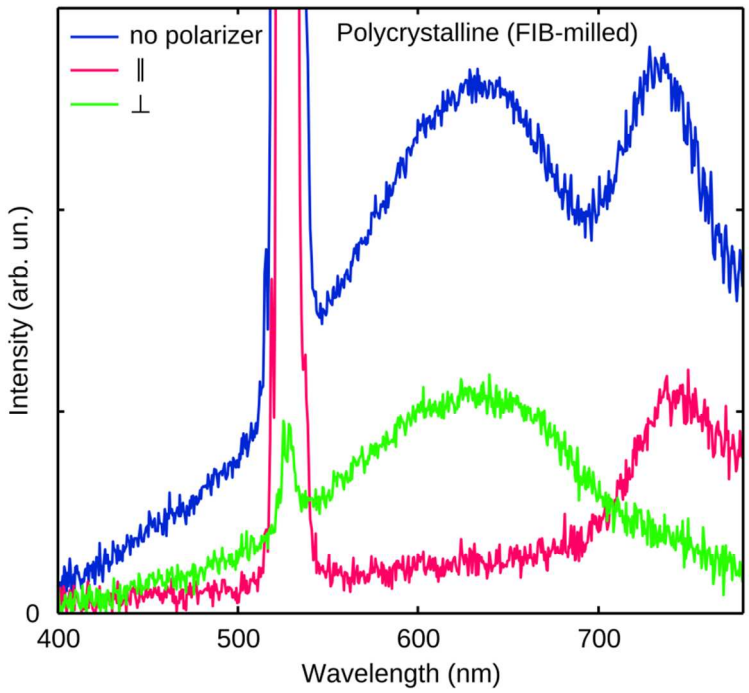

Figure III. Polarization-resolved photoluminescence spectra from a polycrystalline nanorod fabricated with FIB milling into a gold film produced via thermal evaporation: data taken without polarizer (blue curve) and with polarizer set parallel (red graph) and perpendicular (green) to the long rod axis, respectively. The excitation wavelength was set to 1575 nm at a pulse energy of 88 pJ. The green PL peak is red-shifted since this nanorod was fabricated on a quartz substrate with higher index of refraction than fused silica. The spectra show that polycrystalline FIB-milled rods behave like polycrystalline rods fabricated with electron beam lithography, proving that the nanostructuring method does not influence the PL. Furthermore, X-

ray emission spectroscopy proves that our FIB-milled samples do not suffer from implantation of Gallium ions.