Heterogeneous Forward and Backward Scattering Modulation by Polymer-Infused Plasmonic Nanohole Arrays

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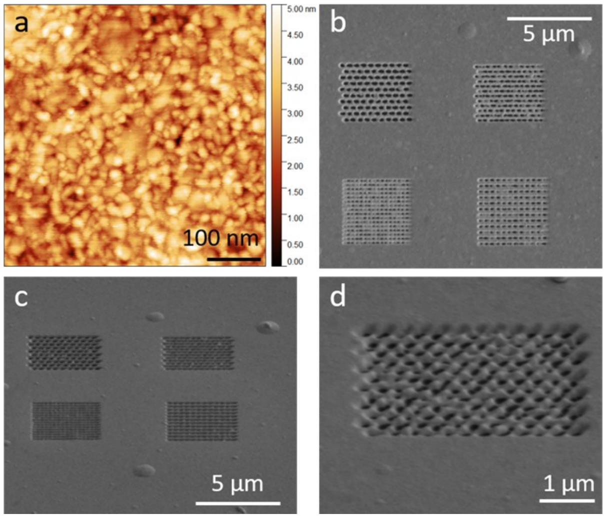

Figure S1. (a) AFM topographical image of E-beam evaporated gold on Si substrate. (b) Top-view SEM image of NHAs. (c) Oblique side-view SEM image of NHAs. (d) Oblique side-view SEM image of p340h

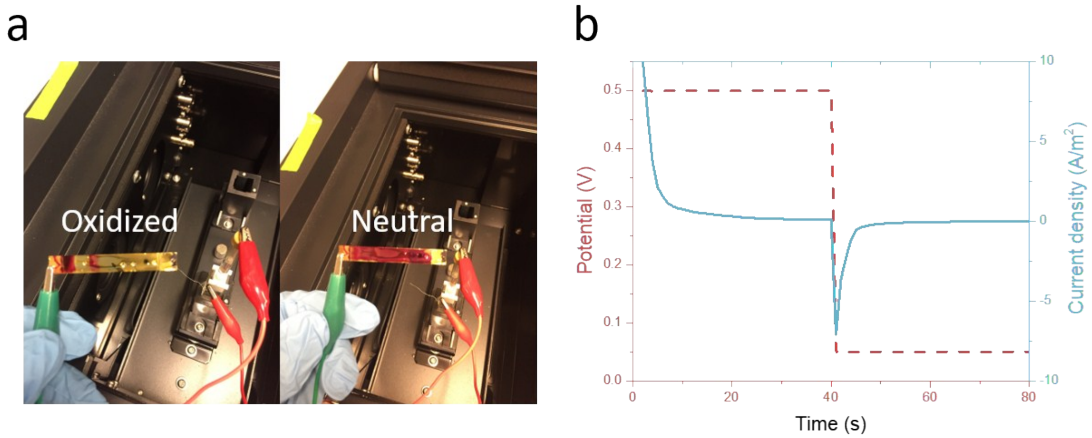

Figure S2. (a) Optical photograph showing the ECP coated nanohole array substrate in the oxidized and neutral state. (b) Current density changes vs time in response to applied potential.

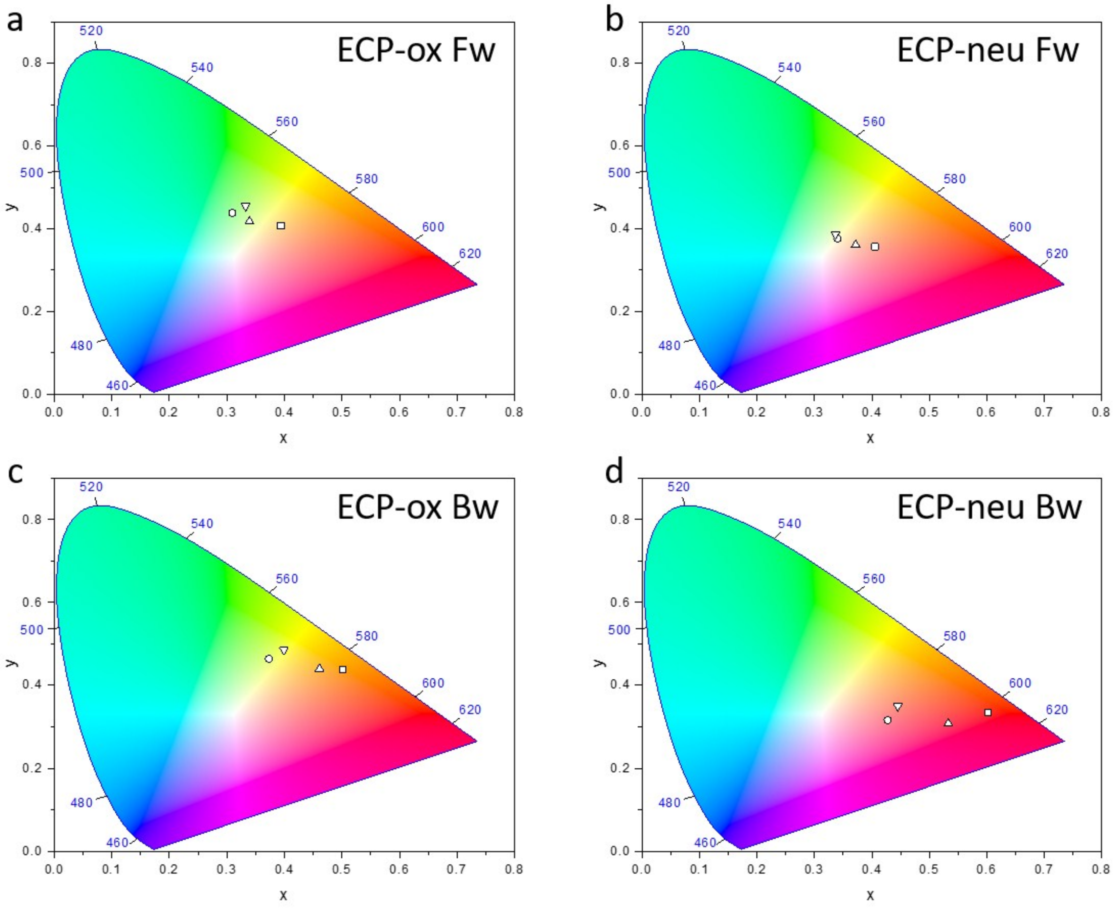

Figure S3. CIE 1931 coordinates (standard illuminant E) calculated from normalized spectrum of NHAs under different conditions □: p250, ○: p350, △: p340h, ▽: p420h (a) ECP-ox Fw, (b) ECP-neu Fw, (c) ECP-ox Bw, (d) ECP-neu Bw

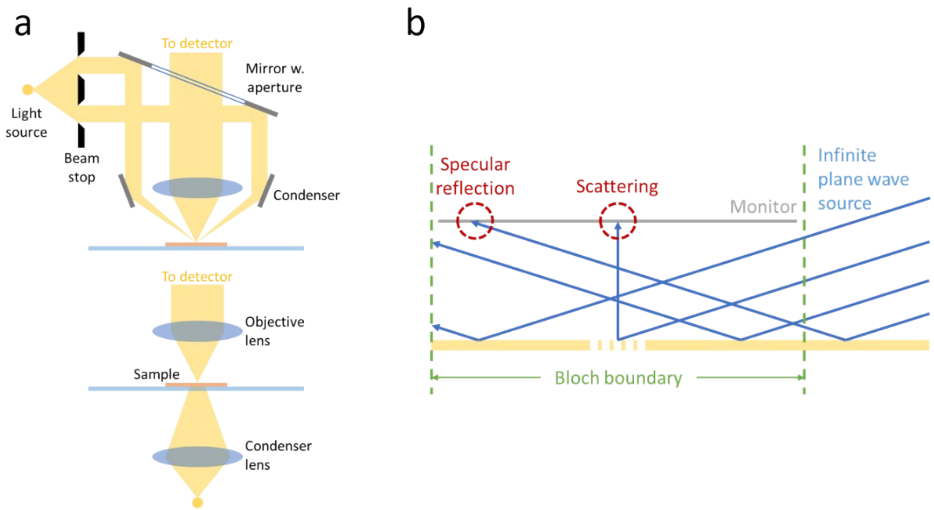

Figure S4. (a) Schematics showing backward scattering (top) and forward scattering (bottom) illumination setup. (b) Schematics showing the FDTD simulation setup in back scattering illumination condition.

Compared to forward scattering, backward scattering is much more challenging to simulate and is rarely performed. This is because when a plane wave source is injected at an incidence angle other than 0°, different frequency components of the source pulse will have different angles of incidence due to phase differences. Since the angle of incidence of the center frequency is already very high (73°), incidence angle for some of the frequency components can easily approach 90°, coupled with periodic boundaries, these components can propagate indefinitely in the XY plane, easily leading to divergence. Thus, to perform such simulation, extreme narrowband sources need to be used, leading to long pulse length (50 fs), and individual simulation must be run at each frequency point to produce the spectrum in a specific range, quickly draining computational resources and increasing computational time.

Another problem with simulating backward scattering is that the periodic boundary condition will necessarily cause specular reflection to be collected. As shown in Figure S4, when both the planewave source and the structure is infinite in the XY plane, 2D monitors will unavoidably collect radiations from specular reflection. Such effect is demonstrated in Figure 6c, in which we simulated the “back scattering spectrum” of a solid perfectly smooth 150-nm thick Au film (Bulk Au). Because such film is incapable of producing back scattered light, as all the light will either be absorbed or specularly reflected, the intensity should be zero across the board. However, we do see spectrum features, meaning specular reflection is collected.

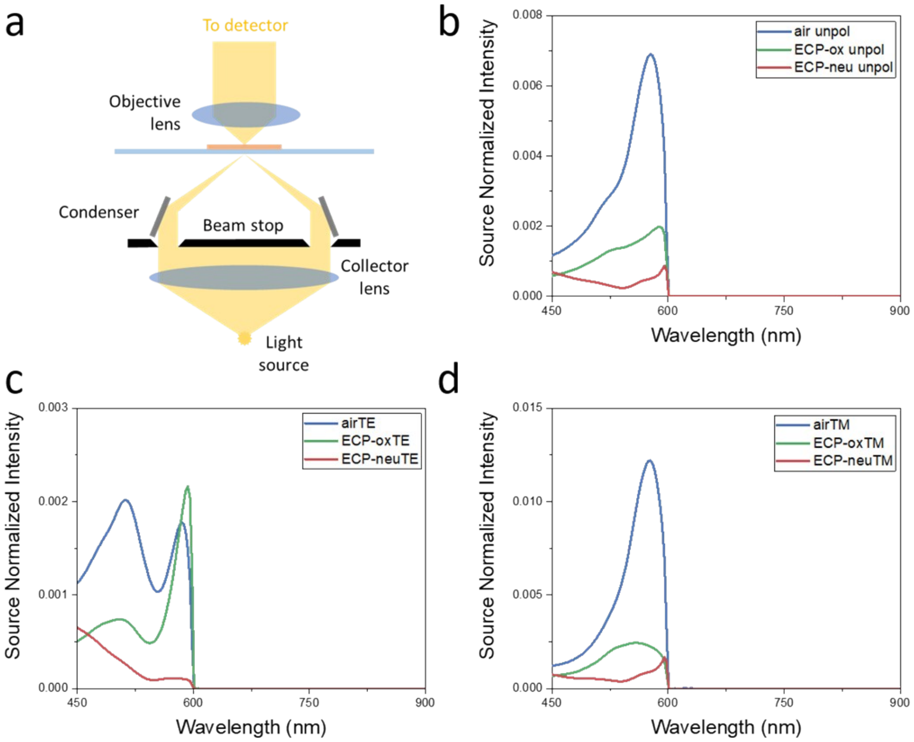

Figure S5. (a) Schematics showing the transmission darkfield illumination condition. (b) Unpolarized scattering spectra of p250s array in air (blue), ECP-ox (green), and ECP-neu (red) from FDTD simulation. Decomposition of FDTD simulated spectra into (c) TE and (d) TM components.

We also simulated the spectra for the darkfield transmission setup (Figure S5a), in which light is coming from beneath the substrate at a highly oblique angle and the spectrum is collected by objectives lens above the substrate. Figure S5b shows the transmission spectrum for unpolarized illumination. In general, the transmission intensities are very low, less than 1% of the source intensity is transmitted across the spectrum with pristine NHAs. When ECP is added, the transmission dropped even lower to below 0.2%. This is intuitive, as grazing angle incidence can be shown to enhance reflectivity by simple Fresnel equation analysis using complex refractive indices. There also appears to be a cut-off frequency below which transmission intensity is negligible. Figure S5c and S5d shows the decomposition of the simulated spectra into TE and TM components. By comparing with the unpolarized spectra, we can see the general spectra trend is dominated by TM components where TE components only have a small contribution. However, we want to accentuate here that these simulations are not experimentally verified, sharp transitions such as the one shown in ECP-neu TM mode might be due to simulation instabilities and may not be physically accurate. Nevertheless, these preliminary results are fascinating, especially the appearance of a cut-off frequency and the markedly different TM/TE response and would be of great interest for future research.