Supporting Information

Self-Assembled Epitaxial Au-Oxide Vertically Aligned Nanocomposites for Nanoscale Metamaterials

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Using pulsed laser deposition, we fabricated metal-oxide nanocomposite films with vertically aligned Au nanopillars (~ 20 nm in diameter) embedded in various oxide matrices with high epitaxial quality. The supplementary data below summarizes the statistical pillar dimension distribution, nanopillar density tuning, nanopillar growth on different substrates, and large-scale uniformity of the nanopillars.

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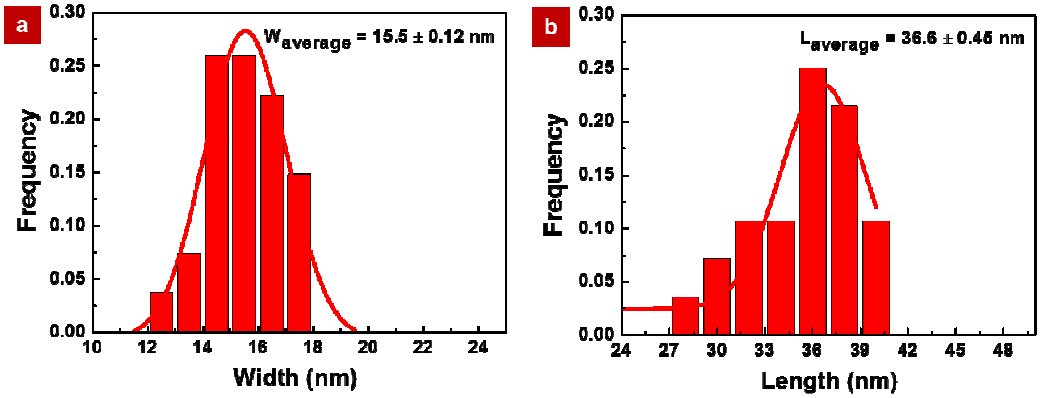

Figure S1. Statistical size distribution of the (a) width and (b) length of the rectangular shaped Au nanopillars in $BaTiO_3$ (BTO) matrix corresponding to the sample in Figure 3 of the manuscript.

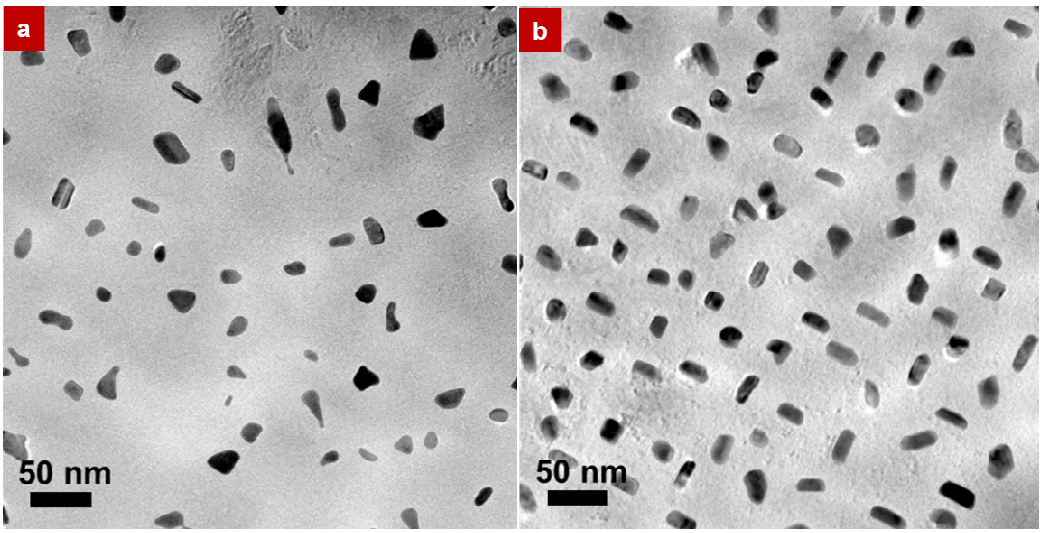

Figure S2. Plan-view transmission electron microscopy (TEM) images of epitaxial Au-BTO thin films grown on $SrTiO_3$ (STO) (001) substrate with tunable Au nanopillar densities. (a) a lower areal density of ~ 3.1×10^{10} cm^{-2} and (b) a higher areal density of ~ 4.9×10^{10} cm^{-2}. The Au nanopillars are uniformly distributed in the BTO matrix and the nanopillar density can be easily tuned by controlling the composition of the composite Au-BTO target.

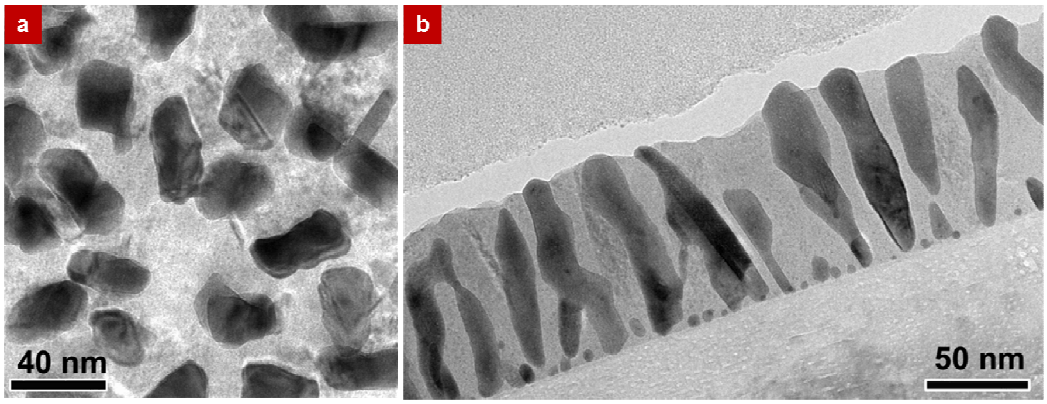

Figure S3. Plan-view (a) and cross-sectional (b) TEM images of Au-BTO thin film grown on *c*-cut α-Al_2O_3 substrate. The TEM images indicate that epitaxial Au nanopillars embedded in BTO matrix can also be fabricated on *c*-cut α-Al_2O_3 substrate.

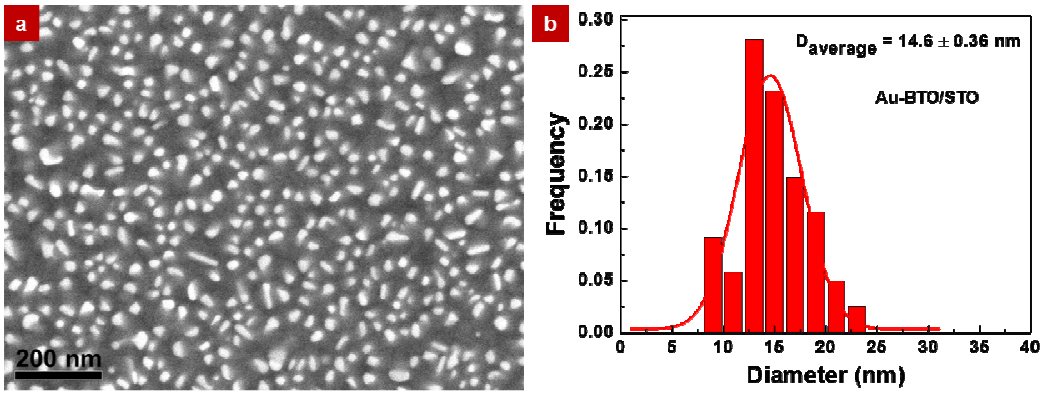

Figure S4. (a) A typical low magnification scanning electron microscopy (SEM) image of Au-BTO thin film grown on STO (001) substrate and (b) the statistical distribution of the nanopillar diameters from the corresponding SEM image. The low magnification SEM image from a large area indicates the uniformly distributed epitaxial Au nanopillars in BTO matrix. The statistical size distribution shows that the average diameter of Au nanopillars is at nanometer scale of around 15 nm. This clearly demonstrates the advantages of the pulsed laser deposition method for creating epitaxial Au nanopillars by one-step self-assembly direct growth.

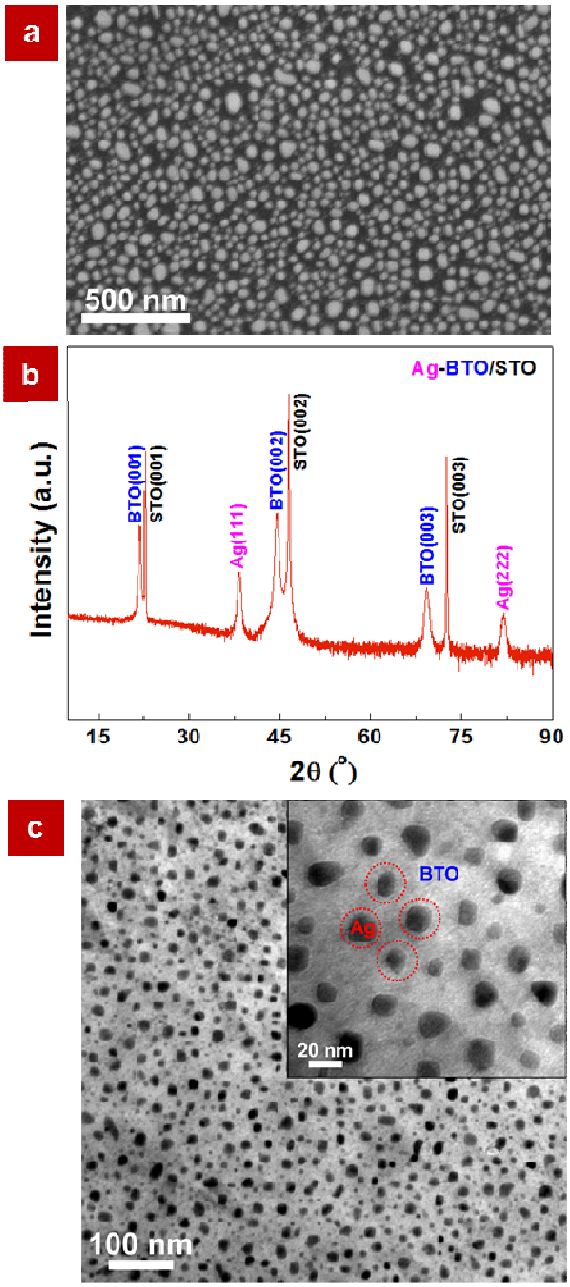

Figure S5. (a) SEM image of Au-ZnO nanocomposite VAN structure grown on *c*-cut α-Al_2O_3, XRD pattern (b) and plan-view TEM image (c) of Ag-BTO nanocomposite demonstrating the potential and versatility of the metal-oxide nanocomposites for nanoscale metamaterial systems.

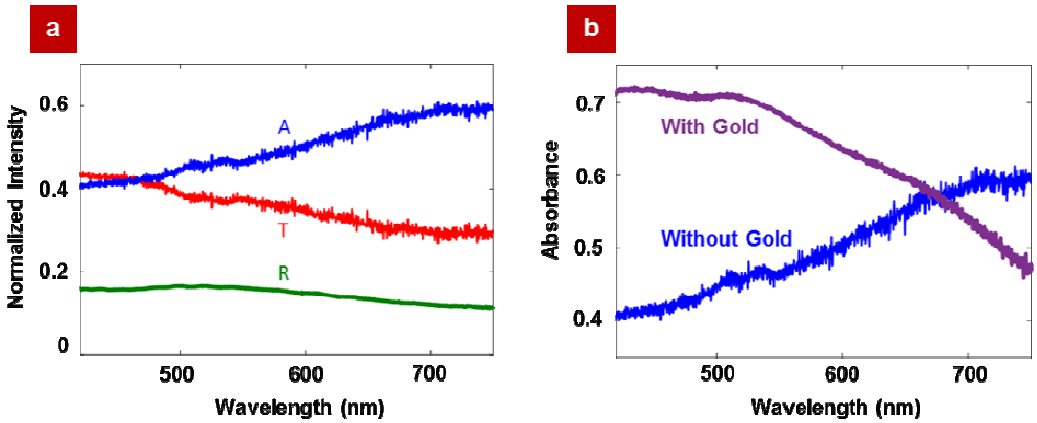

Figure S6. Experimental absorbance (blue), reflectance (green) and transmittance (red) spectra of a control 100 nm thick BTO film on a STO substrate (a) are compared with the absorbance (purple) of the equivalent VAN structure with Au pillars (b), as shown in Figure 4.