Supplementary Information

Electronically Controlled Semiconductor Nanoparticle Array for Tunable Plasmonic Metasurfaces

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I. Aluminum Anodization

Fig. 1(a) is a schematic representation of device fabrication steps. Anodized aluminumis considered as perfect template for cost-effective fabrication of nanowires. The self-assembly process offers highly ordered array of densely packed nanopores formed inside a high-k

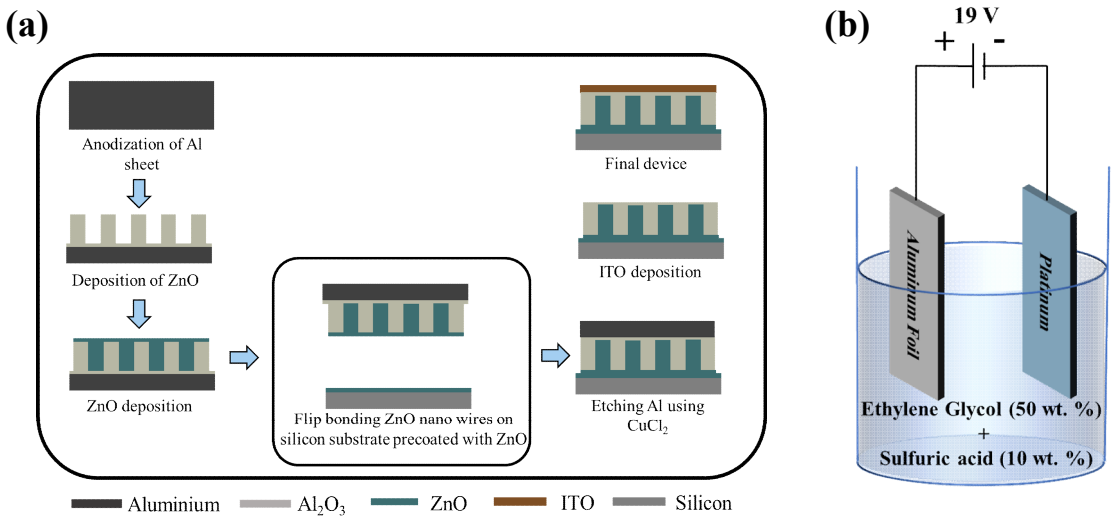

Fig. 1*Devicefabrication steps and aluminum anodization setup.* ***(a)*** *Schematic representation of device fabrication steps.* ***(b)*** *Anodization setup using aqueous solution of sulfuric acid in ethylene glycol with 50 and 10 % by weight respectively at -2 ºC. Aluminum foil and platinum wire net is used as anodes and cathodes respectively for an applied voltage of 19 V.*

dielectric (Aluminum Oxide). Additionally, simple fabrication and effective control over its feature size allows room for much required structural engineering making it an ideal template for growth of reproducible semiconductor nanostructures in the proposed configuration. To fabricate the structure, initially an aluminum foil (from Sigma Aldrich) with a purity of 99.999% is cleaned with (Deionized) DI water, acetone, and ethanol in a sonicator. The film is then annealed at 400º C in Nitrogen environment for 3 hours to yield a defect free surface. To reduce the surface roughness, the Al foil is subsequently electropolished in a solution of ethanol and perchloric acid with 1:3 ratio by volume under a constant voltage of 15 V for 5 mins, after which the foil is thoroughly cleaned with DI water. To produce highly ordered porous film two-step anodized approach is followed where first step anodization is used to create self-organized porous pattern with high degree of regularity which will be utilized as a template for second step anodization. In order to obtain small pore size, the first step of anodization is carried out at 19 V in an aqueous solution of ethylene glycol and sulfuric acid (50 wt. % and 10 wt. % respectively) for 6 hours[1,2]. Fig. 1(b) shows the anodization setup, where aluminum is placed as an anode in anaqueous solution of ethylene glycol and sulfuric acid (50 wt. % and 10 wt. % respectively).A platinum wire net is used as a cathode and a voltage of 19 V is applied across the electrodes. This concentration of ethylene glycol is employed as it yields smallest pore size. After first step anodization the porous oxide layer produced is then etched in an aqueous solution of chromic acid and phosphoric acid (7% and 1.8% by weight) to yield a regular pattern on the Al foil. The second step of anodization with similar parameters is followed to finally achieve the desired porous structure. Fig. 2(a) shows the SEM images of the anodized film where a pore size of around 8 nm in diameter and a period of 25 nm can be clearly seen.

II. Pulsed Laser Deposition

After anodization Gallium doped (10%) ZnO is deposited inside the nanoholes formed in Anodized Aluminum Oxide (AAO) using Pulsed Layer Deposition (PLD) system to form dense array of ZnO nanowires inside AAO. The deposition parameters are described in table 1. Fig. 2(b) displays XRD plot of the resulting structure clearly indicating presence of GZO along with along with highly crystalline aluminum with dominating peaks in the plot 3,4. Fig. 2 (c) shows SEM image of the cross section of AAO template assisted GZO nanowires, clearly depicting

nanowires of diameter around 8 nm. Subsequently, for the next step a layer of ZnO is coated on a separate silicon substrate using sol-gel process. The AAO assisted ZnO nanowire array is then flip bonded on ZnO coated silicon substrate, for which both the layers were first cleaned thoroughly with acetone and Isopropyl Alcohol (IPA), the spin coated ZnO was then dipped in HCl solution for 20 sec then rinsed in deionized (DI) water and dried. The samples are immediately joined together with one sample up-side down and a pressure of 2 MPa was applied. The whole system is then loaded inside a furnace where it is heated to 400 ºC for 2 hours with a ramp of 5 ºC. The sample are found to be bonded after they are removed from the furnace when it is cooled to room temperature. The residual aluminum is etched in a solution of CuCl2 leaving an array of ZnO nanowire inside porous alumina oxide over a glass substrate. Finally, ITO is deposited over the barrier layer with the help of RF sputtering with argon and oxygen gas flow of 10 and 0.08 sscm respectively to obtain the final device as shown in fig. 1(a).

III. Electro-optic Characterization:

The device is characterized for its passive as well as dynamic electro-optic response. To measure

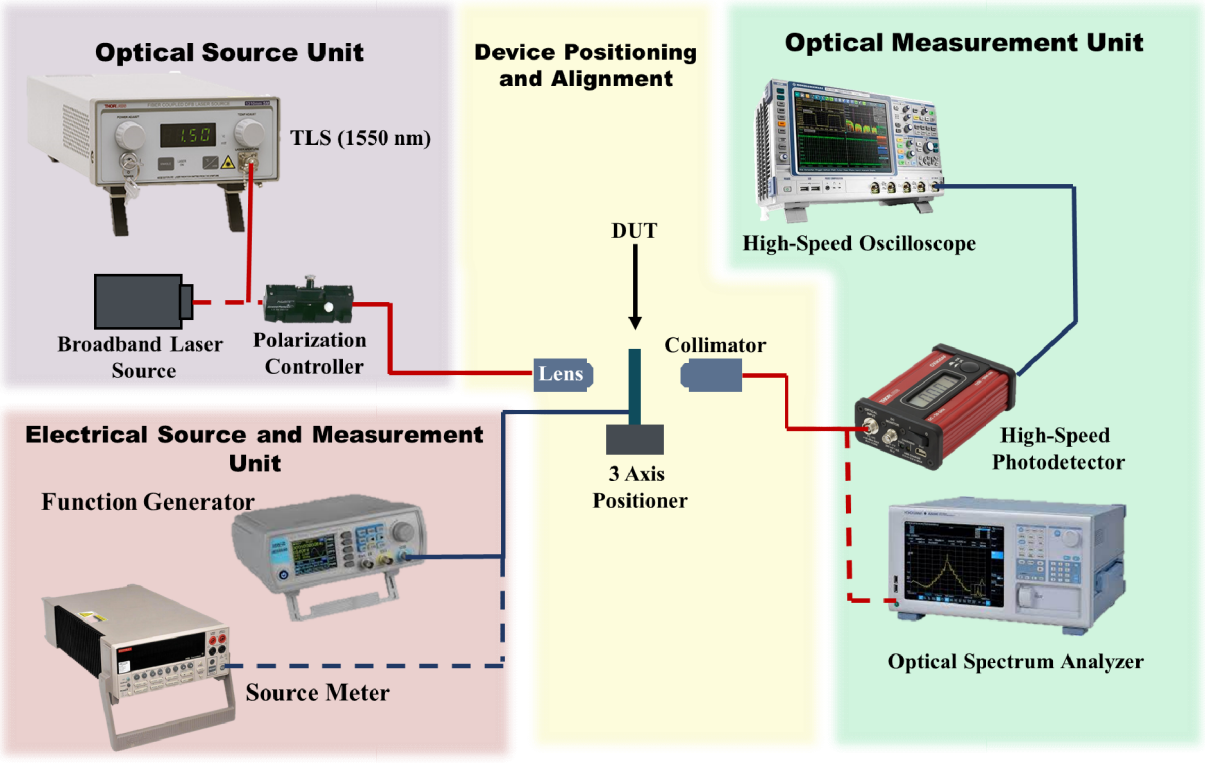

Fig. 3 *Passive and dynamic electro-optic characterization setup. Dash lines represents setup for passive characterization while solid lines show required setup for dynamic characterization.*

its passive response a broadband optical source is used to excite the sample and the response is measured with the help of Optical Spectrum Analyzer (OSA) while voltage bias to the device is provided using a source meter. The connections are shown in fig. 3, where dashed lines represent setup for passive characterization while solid lines show required setup for dynamic characterization. Optical power from broadband source is transmitted through the Device Under Test (DUT) via a polarization controller with the help of optical fiber mounted on a 5-axis positioner. The transmitted optical power is then collected by OSA via a collimator placed on a 5-axis positioner. To ensure proper collection of the optical power the coupling between OSA and the optical source is optimized by aligning the fiber and the collimator through micro-positioner stages. Consequently, the DUT, which is mounted on vacuum stage, is sandwiched in between, and electrical input is provided by source meter unit as shown in fig. 3. The optical absorption is calculated for different applied voltages with the help of the measured optical transmission spectrum.

Measurement of dynamic response of the device is similar to passive response measurement, where broadband source is replaced with a continuous laser source at a fixed wavelength of 1550 nm. Also, a high-speed function generator is used instead of fixed voltage dc source meter to deliver high frequency voltage input to the DUT. Finally, the optical power is fed into a high-speed photodetector and the dynamic response of the device is recorded by an oscilloscope which is connected to the photodetector.

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