Supporting Information

Plasmonic Tuning of Aluminum Doped Zinc Oxide Nanostructures by Atomic Layer Deposition

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Figure S1. XRD patterns of 50 nm AZO coatings on planar silicon deposited at varying Al₂O₃:ZnO cycle ratios. The data shows all peaks shifting to larger diffraction angles and an a-axial [increase in (100) peak] preference as Al₂O₃ cycle occurrence is increased.



Figure S2. XRD patterns of Al_2O_3 :ZnO films with a 1:15 cycle ratio on planar silicon deposited at varying temperatures. The data shows all peaks shifting to larger diffraction angles and c-axial preference [increase in (002) peak] as the deposition temperature is increased.



Figure S3. (100) Wurtzite diffraction angle as a function of (left) Al cycle frequency and (right) deposition temperature.



Figure S4. Hall measurements showing carrier concentration and hall mobility of a 50 nm AZO coating on a planar glass substrate for (a) different aluminum cycle frequencies (1:15-1:30 Al_2O_3 :ZnO cycle ratios) and (b) different ALD deposition temperatures at a constant Al cycle frequency of 6.25% (1:15 Al_2O_3 :ZnO cycle ratio).



Figure S5. IR extinction spectra of AZO (250°C, 1:15 Al:Zn) deposited on SiNPs and planar silicon, ZnO coated SiNPs, and uncoated SiNPs.



Figure S6. Real and imaginary dielectric constants determined by ellipsometry for a 1:15 Al₂O₃:ZnO cycle ratio AZO-coated planar substrate deposited at 250°C.

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ω_p	γ_p	f_l	ε_{b}	ωι	γ_l	
1.231	0.281	0.699	2.855	4.392	0.064	

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$$\varepsilon(\omega) = \varepsilon_b - \frac{\omega_p^2}{\omega(\omega + i\gamma_p)} + \frac{f_l \omega_l^2}{\omega_l^2 - \omega^2 - i\omega\gamma_l}$$

The complex permittivity determined experimentally by ellipsometry is fit to the Drude-Lorentz equation where ω_p , γ_p , f_l , ε_b , ω_l , γ_l is the plasma frequency, relaxation rate, oscillator strength, background permittivity, center frequency and damping coefficient, respectively.



Figure S7. IR extinction spectra for AZO-coated SiNP arrays (grown at 250°C with a 1:15 Al₂O₃:ZnO cycle ratio) at different incident angles for (a) TE polarized light and (b) TM polarized light.



Figure S8. (a) IR extinction spectra of a 1:15 Al_2O_3 :ZnO cycle ratio AZO-coated SiNP array with different diameters. The increase in extinction at ~ 2200 nm for the 681 nm diameter pillars is attributed to the photonic modes of the silicon core. (b-d) SEM images of AZO-coated SiNPs with diameters of (b) 303 nm, (c) 490 nm, and (d) 681 nm. (e) Histogram showing the diameter distributions of AZO coated SiNPs with average diameters ~300nm. (f) FDTD simulations of AZO coated SiNPs with varying diameters spanning the size distribution displayed in the previous histogram showing little optical variation of the resonant wavelength.



Figure S9. IR extinction spectra of a 1:15 Al_2O_3 :ZnO cycle ratio AZO-coated SiNP array deposited at 225 °C with varying AZO shell thicknesses. Due to a smaller AZO shell filling fraction, the optical density of the spectra are normalized for better comparison.



Figure S10. (a) IR extinction spectra of a 1:20 Al_2O_3 :ZnO cycle ratio AZO-coated SiNP array deposited at 200°C with different gap sizes. Data points from 2690 nm to 2800 nm are excluded due to hydroxyl impurities in our set-up. SEM images of AZO coated SiNPs with (b) 12 nm (c) 38 nm and (d) 92 nm gap sizes.