Supplementary Information

Probing sub-diffraction optical confinement via polarized Raman spectroscopy of a single-walled carbon nanotube

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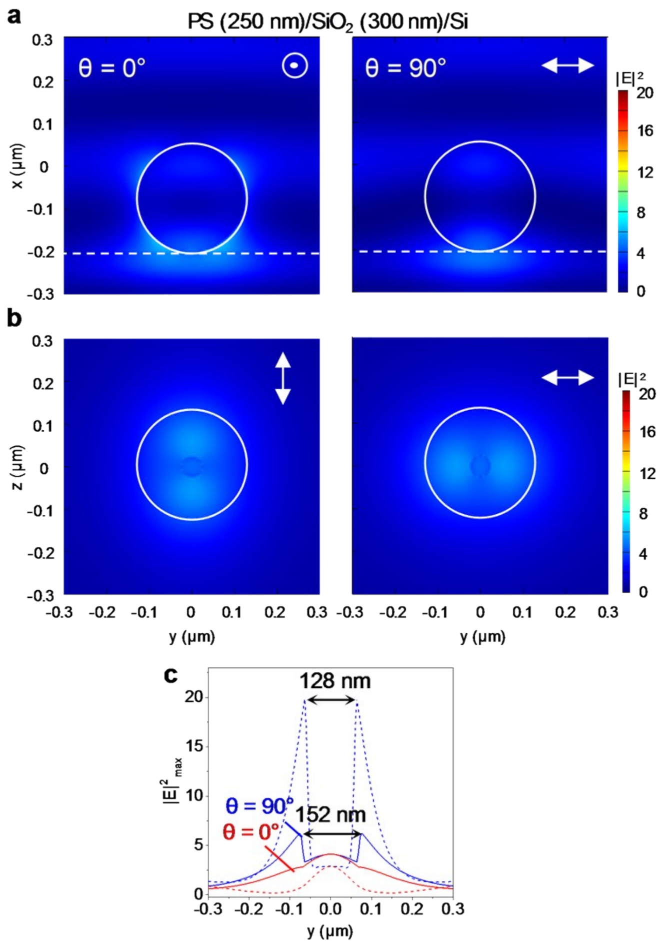

Figure. S1. Marginal enhancement of electric field by PS nanoparticle upon illumination with plane-polarized light. FDTD simulation of a PS nanoparticle (d = 250 nm, n = 1.5983) on SiO_2 substrate illuminated by a plane-polarized wave at 532 nm. The polarization orientation is indicated by white arrows. The electric field distribution on a) xy-plane (z = 0) and b) yz-plane (x at maximum electric field). c) Intensity profile along y-axis at z = 0.

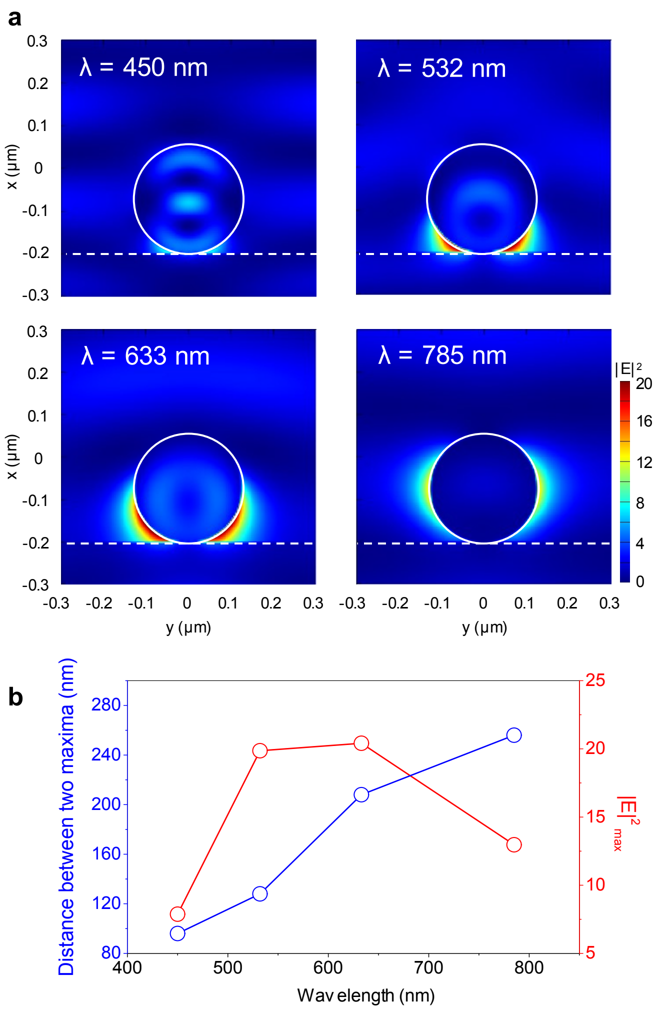

Figure S2. Laser wavelength dependence of nanoscale light confinement. a) FDTD simulation of dielectric TiO_2 nanoparticles (d = 250 nm) on SiO_2/Si substrate illuminated by polarized (θ = 90°) plane waves at 450, 532, 633, and 785 nm. b) Dependence of peak-to-peak distance (blue) and maximum intensity of electromagnetic field (red) on the laser wavelength.

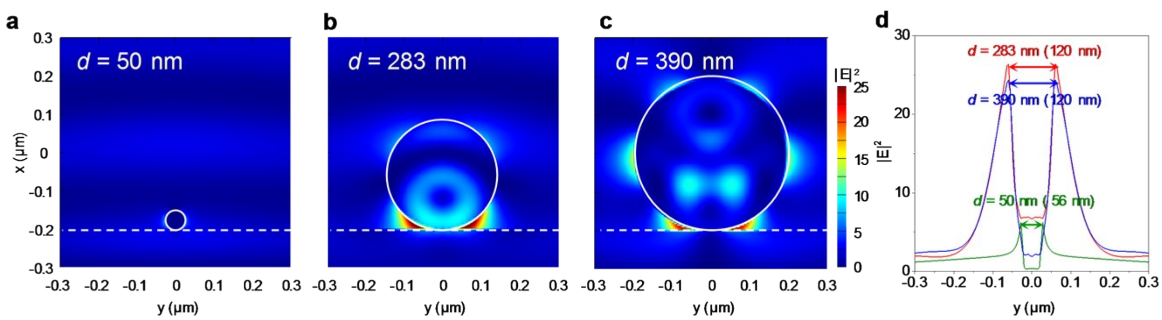

Figure S3. Dependence of peak-to-peak distance on the diameter of TiO_2 nanoparticle. a-c) FDTD simulations of dielectric TiO_2 nanoparticles of different sizes (d = 50, 283 and 390 nm) on SiO_2/Si substrate under polarized incident light (θ = 90°) at 532 nm. d) Maximum intensity profile at the TiO_2-substrate contact indicating a different peak-to-peak distance and maximum intensity. The TiO_2 nanoparticle with d = 50 nm shows the lowest values in both the intensity and the peak-to-peak distance.

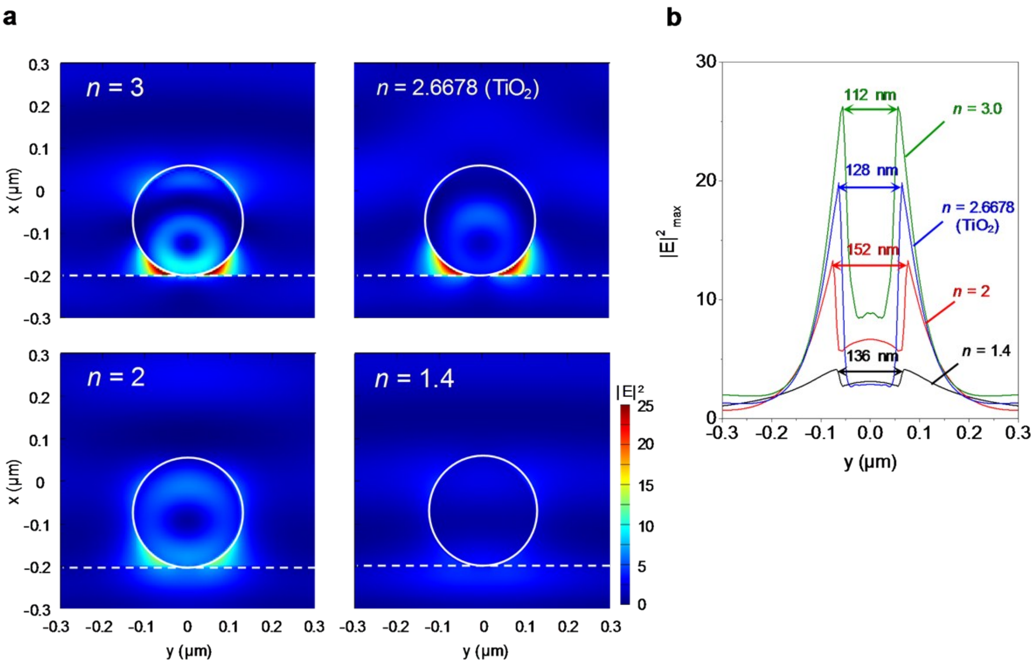

Figure S4. Dependence of nanoscale light confinement on the refractive index of the TiO_2 nanoparticle. a) FDTD simulation of electromagnetic field of 250 nm dielectric nanoparticles having different refractive indices (n = 3, 2.8, 2, and 1.4), under polarized incident light (θ = 90°) at 532 nm. b) Maximum intensity profile at the TiO_2-substrate contact showing difference in the peak-to-peak distance and the maximum intensity. Strong nanoscale light confinement starts to appear at refractive indices above ~2, and the intensity increases with the refractive index of the particle.

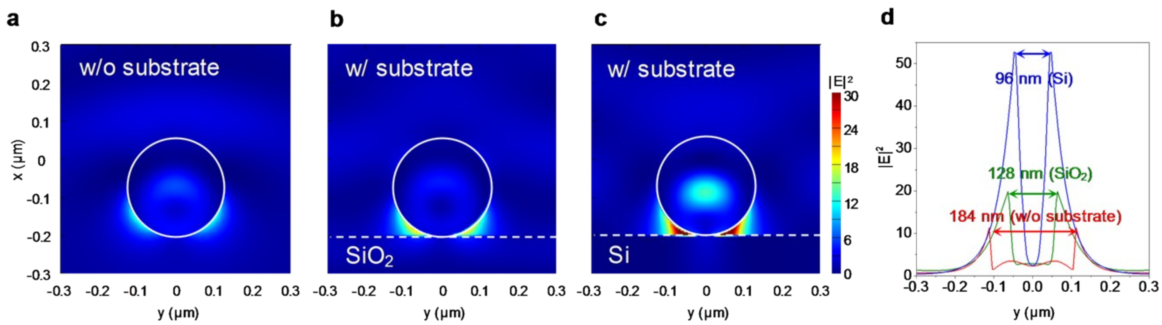

Figure S5. The effect of substrate on the dual-band confinement. FDTD simulation of electromagnetic field around a TiO_2 nanoparticle (d = 250 nm) illuminated by a polarized light at 532 nm (a) in air without a substrate, (b) on a SiO_2 substrate (n = 1.4607, k = 0), and (c) on a bare Si (n = 4.1520, k = 0.051787). (d) Maximum intensity profile at the TiO_2-substrate contact indicates that existence of substrate and high refractive index of the substrate enhances nanoscale light confinement.

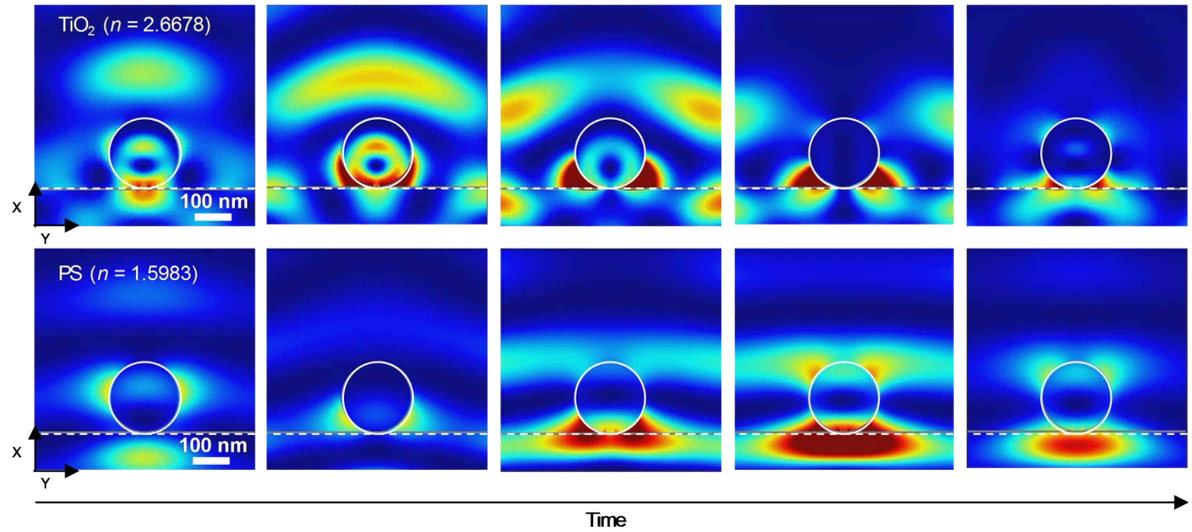

Figure S6. Propagation of the electrical field dependent on the refractive index of the dielectric nanoparticle. Dielectric nanoparticles (d = 190 nm) with different refractive indices (TiO_2 = 2.6678, PS = 1.5983) on a SiO_2/Si substrate are illuminated by a plane polarized wave at 532 nm. Time series of electric field distribution shows that strong enhancement occurs at the TiO_2-substrate contact (top), whereas such enhancement does not occur at the PS-substrate contact (bottom).

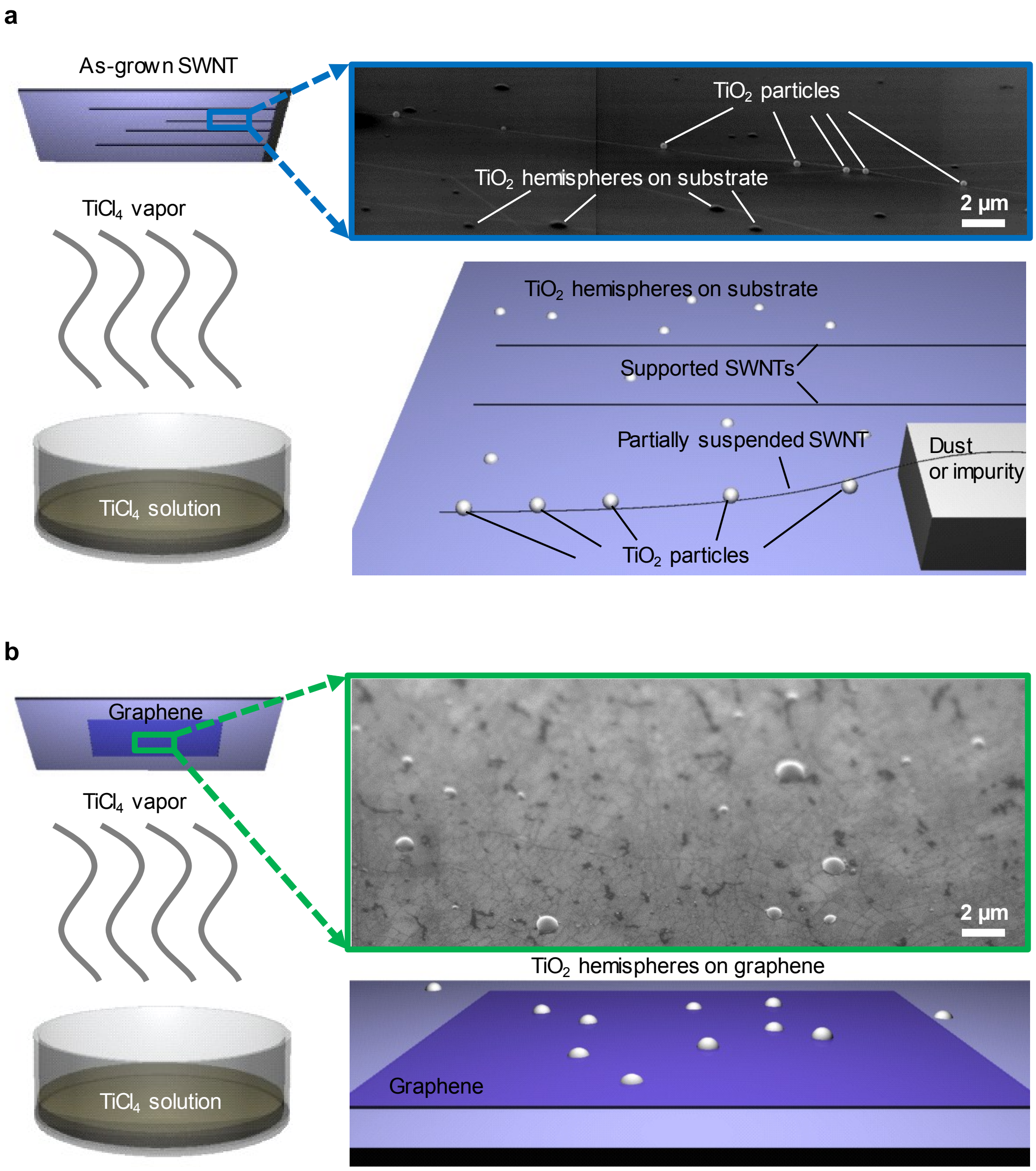

Figure S7. a) Schematic illustrating the process of decorating a partially suspended nanotube with TiO_2 nanoparticles. The TiO_2 nanoparticles were formed selectively on the nanotube when nanotubes were partially suspended due to impurities on the substrate, whereas TiO_2 hemispheres were formed randomly on the substrate and supported nanotube. The SEM image shows distinct morphology of TiO_2 particles and hemispheres. b) Schematic illustrating the formation of TiO_2 hemispheres on graphene.

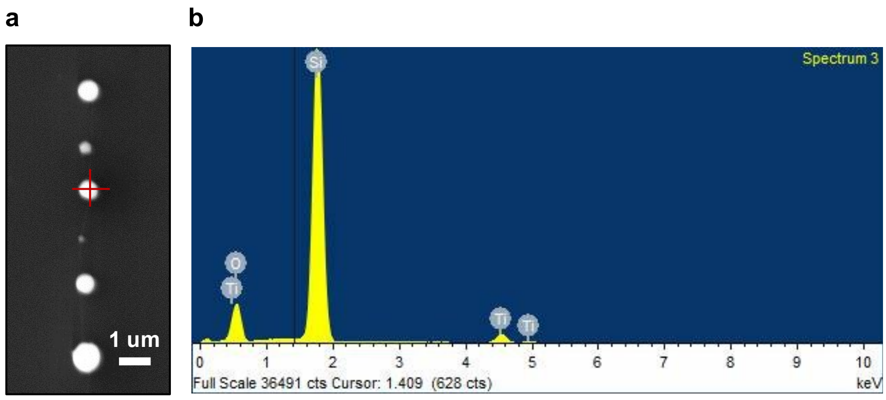

Figure S8. a) SEM image of TiO_2 nanoparticles formed along the nanotube. b) Corresponding energy-dispersive X-ray spectroscopy result confirms that the nanoparticles are TiO_2.

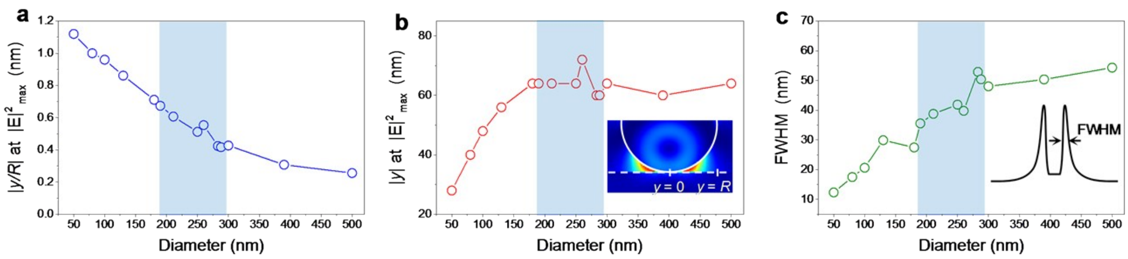

Figure S9. a) y/R at maximum electric field intensity versus diameter of TiO_2 nanoparticle. b) Distance between the particle-substrate contact (y = 0) and the location at maximum electric field intensity (y at $|E|^2$max), plotted versus diameter of TiO_2 nanoparticle. c) FWHM of each band versus diameter of the TiO_2 nanoparticle. Areas shaded in blue indicates the diameter range tested experimentally in Figure 4.

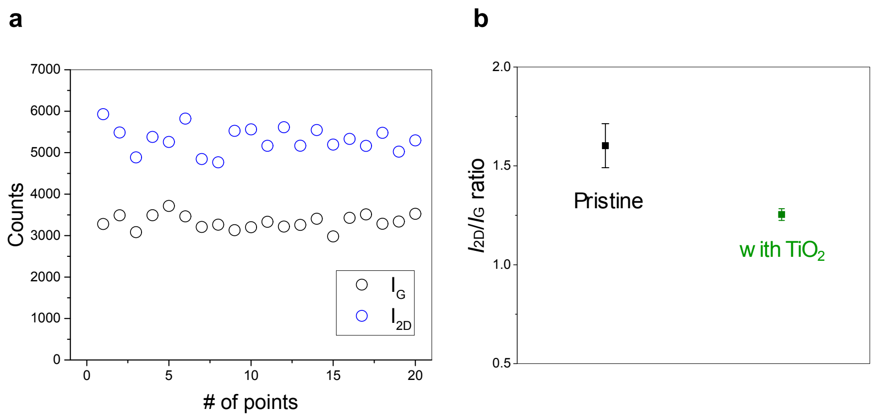

Figure S10. a) Intensity of G and 2D modes of pristine graphene. b) I_{2D}/I_G obtained from pristine graphene or graphene in the presence of TiO_2 nanoparticles.

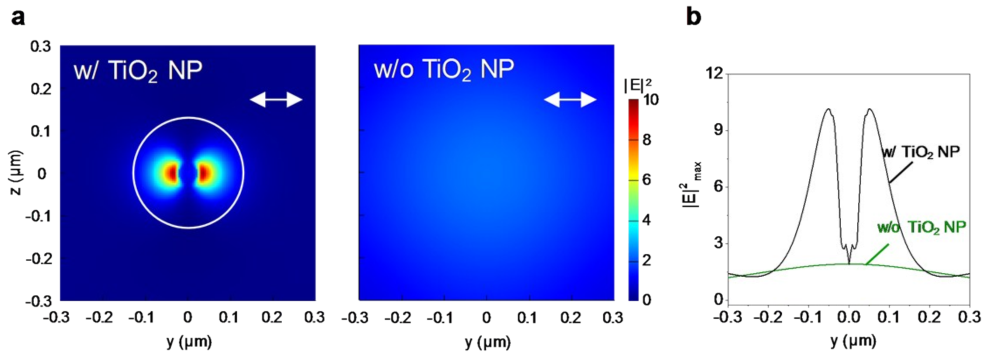

Figure S11. a) FDTD simulation of SiO_2/Si substrate with (left) or without (right) TiO_2 nanoparticle (d = 250 nm) illuminated by polarized plane wave (θ = 90°) at 532 nm. b) Corresponding intensity profile on the substrate.

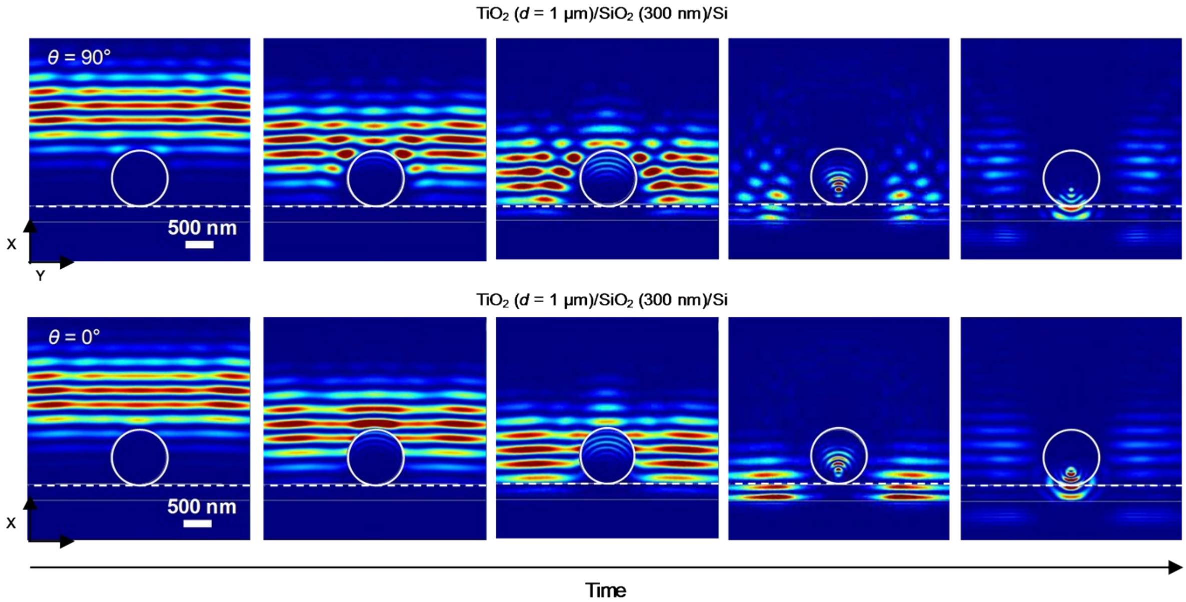

Figure S12. Propagation of electric field along dielectric microsphere. Time series of electric field distribution showing that the electric field at the particle–substrate contact is not significantly enhanced at the TiO_2 microsphere under both incident polarizations at 532 nm.

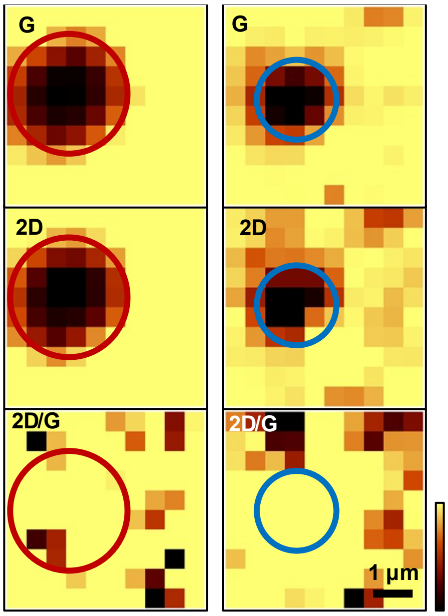

Figure S13. Raman scattering of 2D graphene through a dielectric microsphere. Raman G, 2D, and 2D/G maps of graphene under incident light at 532 nm. The diameters of the TiO_2 microspheres on graphene are 3 μm (red circles) and 2 μm (blue circles).