Broadband Nonlinear Superlens Based on 2-ENZ Nano-Composites

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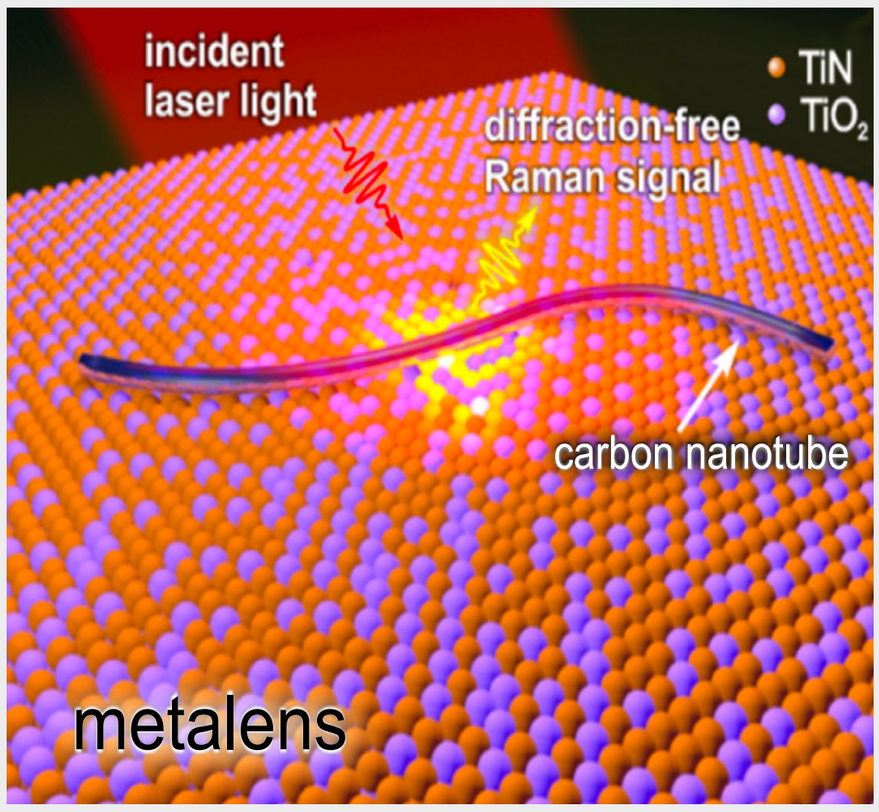
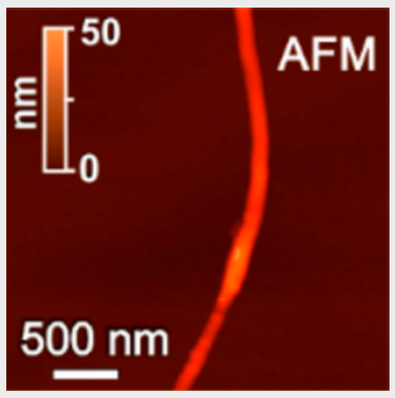
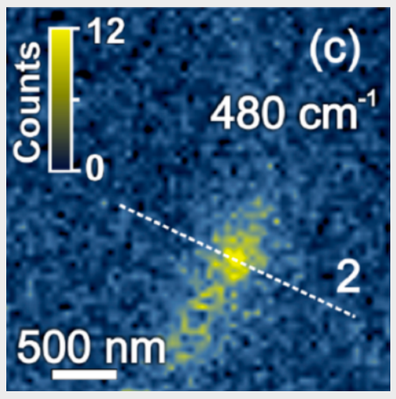
Abstract. In this work, we suggest a concept for far-field broadbandwidth superlens. For this purpose we used titanium oxynitride thin film, that is a disordered metal-dielectric nanocomposite. TiO_xN_y films exhibit a double epsilon-near-zero (2-ENZ) behavior near the percolation threshold. This unususal behavior of permittivity opens up novel opportunities for the excitation of plasmon resonance at several distinct frequencies within visible and NIR range. Thus, enhanced light-mater interactions and superlensing condition can be realized at multiple wavelengths. We experimentally demonstrate a far-field superlensing effect by showing a sub-diffraction resolution λ/6 at different wavelengths using multiwalled carbon nanotubes directly dispersed on the metalens. We have shown that superresolution is achieved due to enhanced Raman nonlinearity and multiplicative nature of four-wave mixing process. We believe that our findings will bring us one step closer to developing robust far-field color imaging system with subdiffraction spatial resolution.

Refernces:

[1] Kharintsev, S.S., Kharitonov A.V., et al. // ACS Appl. Mater. Interfaces, 2020, **12**, 3862-3872
[2] Kharitonov A.V. et al. // Opt. Mater. Express, 2020, **10**, 513-531
[3] Braic L. et al. // ACS Appl. Mater. Interfaces, 2017, **17**, 29857-29862
[4] Kharintsev, S.S., Kharitonov A.V., et al. // Nanoscale, 2019, **11**, 7710-7719
[5] Kharintsev, S.S., Kharitonov A.V., et al. // Nano Lett., 2017, **17**, 5533-5539

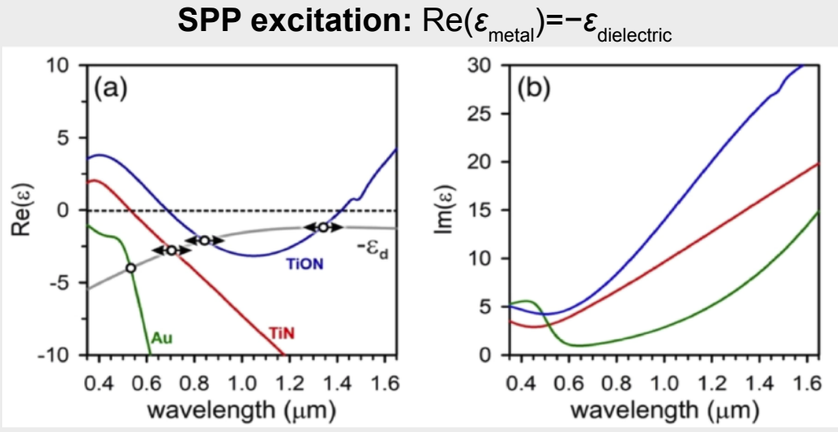




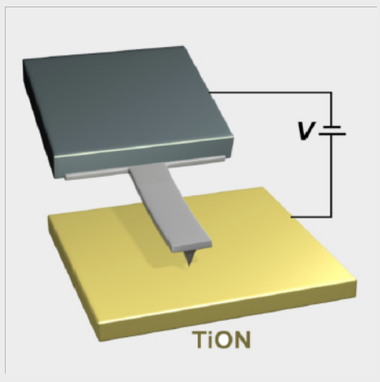


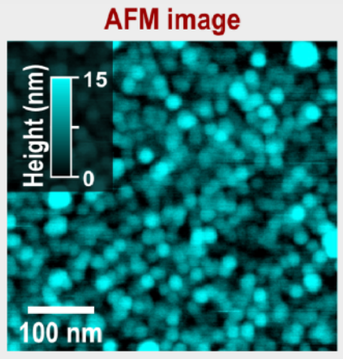
2ENZ plasmonic materials. In the case of conventional plasmonic materials (Au, Ag, ...), the SPP excitation condition is fulfilled at a *single fixed* wavelength. For this reason, vast majority of plasmonic devices have only one working wavelength. Alternative plasmonic media, such as TiN, enable the SPP excitation at a *single tunable* wavelength. 2ENZ materials constitute a novel class of plasmonic media, which allows for excitation of *tunable multi-band* plasmon resonance.

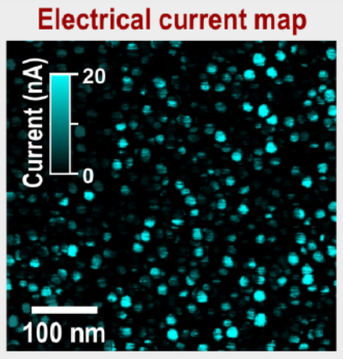
SPP excitation: $\mathrm{Re}(\varepsilon_{metal})=-\varepsilon_{dielectric}$

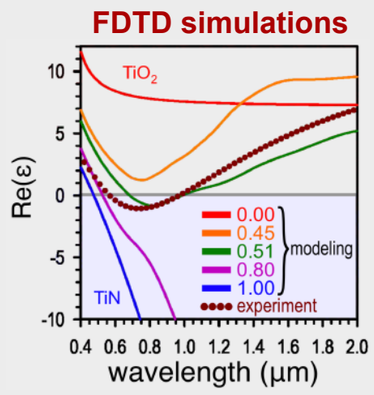


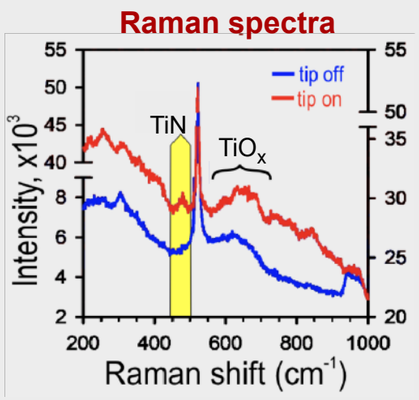
Physical origin of 2-ENZ behavior. Using a wide range of experimental techiques and performing FDTD simulations we have confirmed, that 2ENZ properties of TiON is due to its percolating nanocomposite structure: metallic TiN nanoparticle chains within the multiphase TiO_x/TiO_yN_z host.

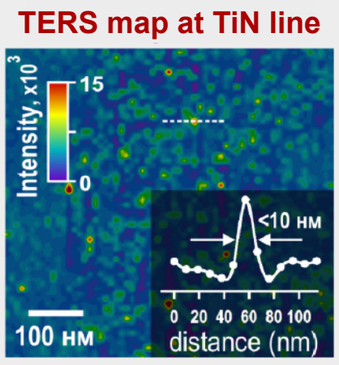


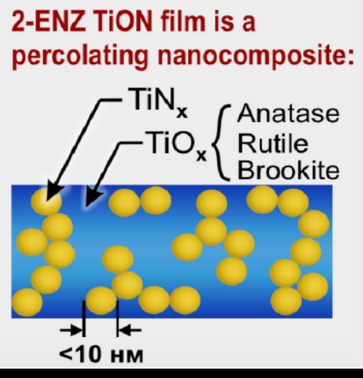




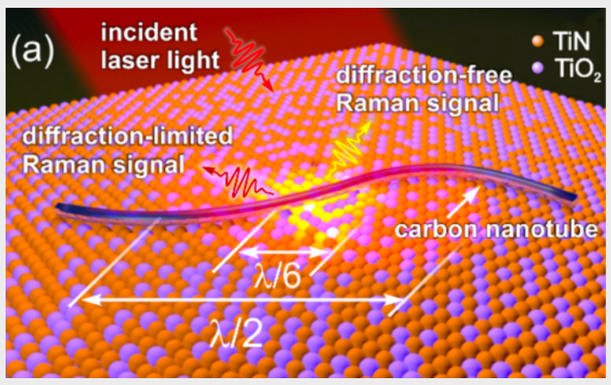


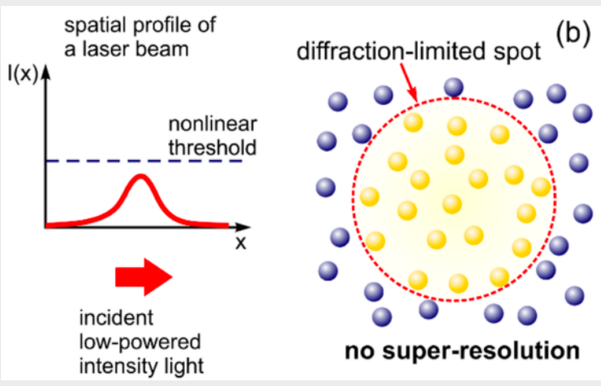


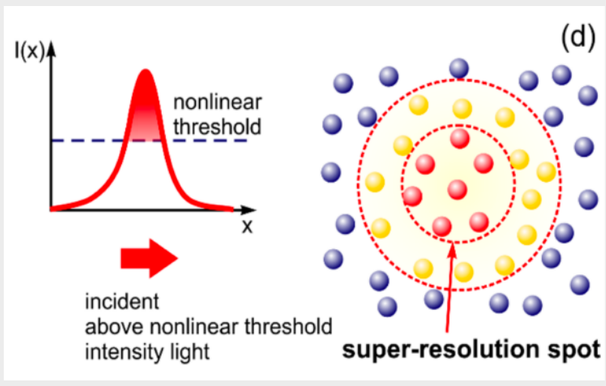


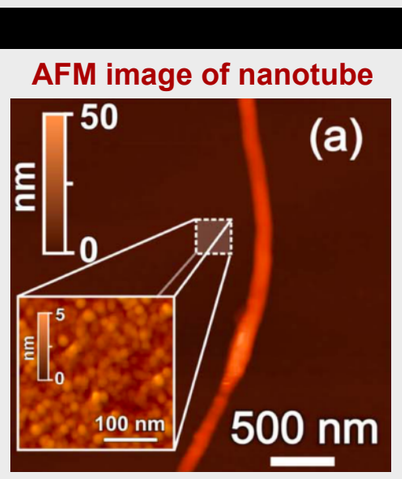


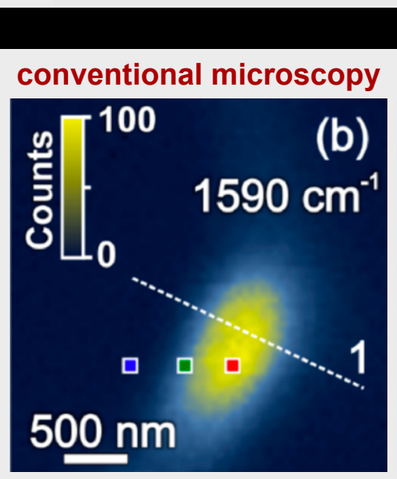
Far-field superlensing. Plasmon multiple scattering within a disordered metal-dielectric media allows one to enhance a cubic susceptibility drastically, and thus stimulated Raman scattering (SRS) can occur at the nanoscale using low-powered pump (see Refs. 4, 5). Hence, an excitation volume is squized due to the nonlinear threshold.

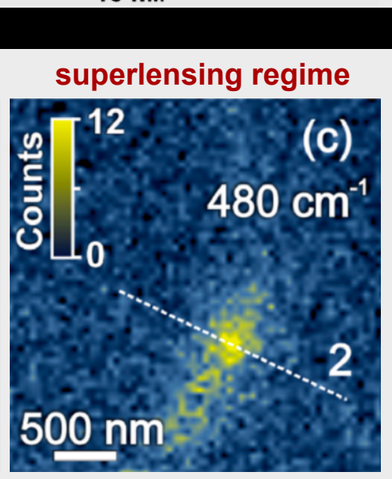




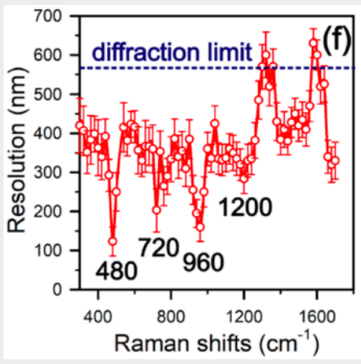


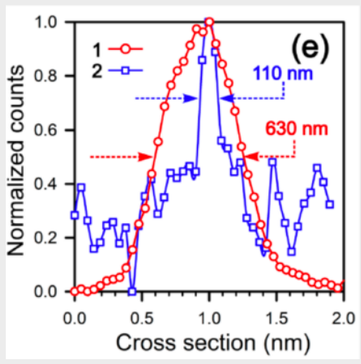






Broadbandwidth superlensing. Super-resolution spot acts as the localized source of coherent Raman light. Scanning over the sample surface allowed us to obtain a superresolution image of nanotube at different wavelengths – overtones of stimulated Raman scattering





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