

Noble Metal Metaplasmonics

Zarko Sakotic^{1*}, Amogh Raju,¹ Noah Mansfeld,¹ Daniel Krueger¹, Alexander Ware¹, Divya Hungund¹, and Daniel Wasserman¹

¹ Electrical and Computer Engineering, University of Texas at Austin, Austin, TX 78758, United States

*zarko.sakotic@austin.utexas.edu

Abstract: We introduce metaplasmonics, a novel plasmonic modality with a host of appealing properties. Using ultrathin perforated nanoribbons we demonstrate large confinement, high quality factors, and large near-field enhancements across a broad range of infrared wavelengths. © 2024 The Author(s)

Plasmonic response in metals is typically limited to a narrow frequency range below the metals' plasma frequency. This limits the use of noble metals for plasmonics to the visible range. Efforts to extend plasmonic response to the infrared have been focused on materials with lower plasma frequencies such as doped semiconductors or metal-oxides, or atomically thin conductive materials such as graphene. The former still suffer from significant scattering loss, limiting the propagation length and confinement of propagating and localized plasmonic modes, while the latter are better suited for longer mid-IR wavelengths and the THz. Accounting for the difficulty of scaling of 2D-materials, this leaves a rather significant technological gap, extending from the near-infrared through to the long wave infrared ($1 - 14\mu\text{m}$), for scalable plasmonic materials with large figures of merit i.e. strong confinement and high quality factors. The recent emergence of atomically thin metals offers one promising path to fill this gap [1, 2], extending the plasmonic materials palette into the NIR. However, the challenging growth required to achieve planar atomically-thick metals remain an impediment to their wide applicability, and perhaps more fundamentally, these films suffer from strongly increased losses due to the intrinsic electron confinement effect (i.e. Landau damping) that emerges at these length scales [3, 4].

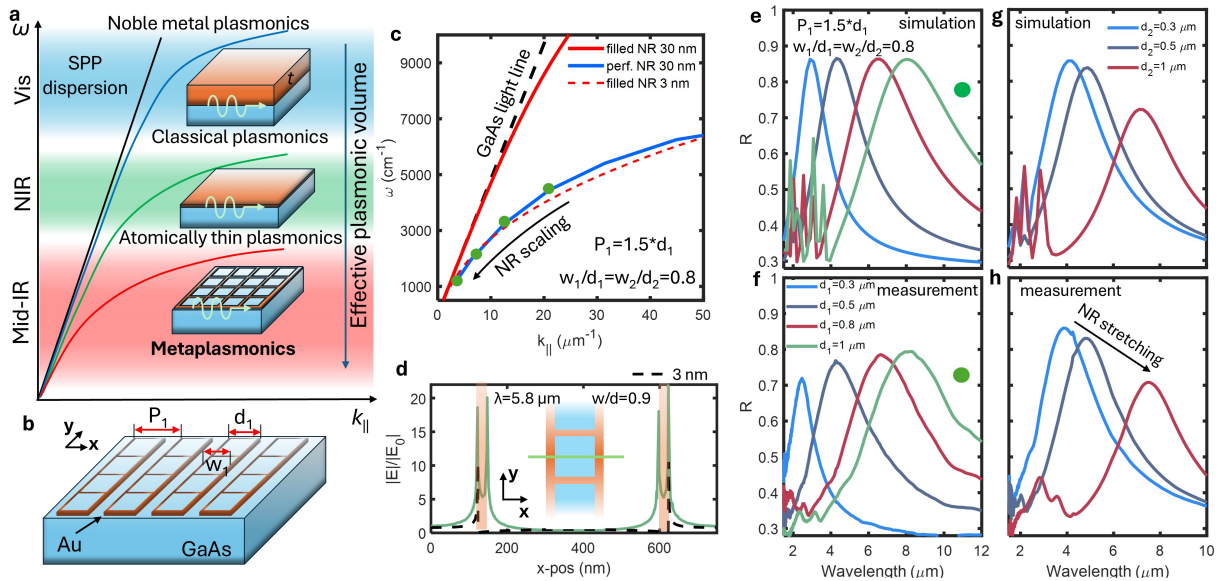


Fig. 1. (a) Schematic of classical, atomically thin, and the metaplasmonics concepts with associated SPP dispersion. (b) Sketch and geometrical parameters of the fabricated nanoribbons (c) numerically obtained dispersion of the modes localized on the NR, along with the dispersion of the filled NRs with thicknesses of 30nm and 3nm for comparison. (d) Simulated field enhancement along the middle of the NR unit cell (for comparison, same is shown for the 3nm planar NR of the same size/resonant wavelength). (e,f) simulated and measured IR reflection response of the fabricated NRs with different scale, tracing the dispersion from (c). (g,h) simulated and measured IR reflection response of the NRs that are stretched in the y-direction, showing increasing confinement factor.

To address these issues, and extend the true plasmonic response of noble metals to the infrared, we propose the metaplasmonic approach, schematically shown in Fig. 1(a) and compared to the classical and atomically-thin

approaches to achieving plasmonic response. The dispersion of the surface-plasmon-polariton (SPP) modes for all three approaches is illustrated, showing that characteristic SPP dispersion bending can be extended well into the mid-infrared using the metaplasmonic approach.

Achieving infrared plasmonic response in atomically thin metals is a result of these films drastically reduced volume; however, since the thickness of these films (usually only a few nanometers) is at least several times smaller than the electron mean free path d_{eff} (10 – 20nm for gold), Landau damping dominates their optical response and the scattering rate (γ , i.e. loss) associated with the film is increased significantly compared to the bulk γ , imposing a strict trade-off between confinement and associated quality factors. Alternatively, plasmonic volume reduction can be achieved with 'dilution' - by starting with a metal film whose thickness is on the scale of, or slightly thicker than d_{eff} , and subsequently perforating the film so as to minimize metal filling factor without introducing features smaller than d_{eff} (in either lateral or vertical dimensions). By doing so, we can circumvent the effects of Landau damping and preserve the bulk optical quality of the metal. The massive reduction in the metal volume enables large plasmonic confinement (large in-plane wavevector $k_{||}$) otherwise unattainable with noble metals. We have recently demonstrated that diluted films can mimic absorbing properties of atomically thin planar metals [5], which gives initial credence to this idea although surface modes in such structures remain unexplored.

To access the highly confined propagating modes of the metaplasmonic film, we fabricate 30nm thick gold metaplasmonic nanoribbons (NRs) using electron-beam lithography (EBL), metallization, and lift-off, on semi-insulating GaAs substrates, Fig 1(b); this enables coupling to SPP modes that are localized on the NRs. In Fig. 1(c) we show the simulated dispersion of 'filled' planar NRs with 30 nm and 3 nm thicknesses (solid and dashed red lines), along with the metaplasmonic NRs (blue line). For the 30 nm thick planar NR, the SPP mode is close to the light line i.e. it is weakly confined. When the 30nm NR is perforated with square holes (dimensions in the figure inset), we achieve strong dispersion bending indicating much stronger confinement across the mid-IR and NIR regions, and crucially with preserved bulk optical quality. Qualitatively similar dispersion bending is achieved by reducing the filled NR thickness to 3nm, however this necessarily results in a scattering rate at least several times larger than the bulk [1,4]. Furthermore, metaplasmonic NRs also leads to very large near-field enhancements, Fig. 1(d), where strong enhancements are present on both sides of the metal edges.

To experimentally verify the plasmonic behavior of the metaplasmonic NRs, we employ infrared reflection spectroscopy under normal incidence, as shown in Fig. 1(e-h). First, we scale the size of the NR and the perforation, Fig. 1(e,f), which traces the dispersion from Fig. 1(c). The measured spectra closely follows the simulations in strength, position, and width, indicating the preserved bulk-quality. The design space also allows for extending the perforation size in the y-direction i.e. we can 'stretch' the NR without changing the lateral size in x-direction (which determines the plasmon wavelength λ_{spp}), and further effectively dilutes the constituent film i.e. increases the level of confinement λ_0/λ_{spp} . Fig. 1(g,h) show the simulated and measured reflection spectra for varying y-period with width of the NR fixed at 500nm. The resonant wavelength is shifted from 4 μ m to 8 μ m, shrinking the plasmon wavelength by 8 times ($\lambda_0/\lambda_{spp} = k_{spp}/k_0 = 8$) with excellent matching between measurement and simulations. Our simulations (not shown) indicate that by using this method the confinement factor λ_0/λ_{spp} can be increased by an order of magnitude, although with less pronounced peaks. This indicates the potential for achieving extreme confinement regime only realized by graphene or Van der Waals layers so far, but with noble metals and standard lithography techniques.

These results challenge the traditionally held notion that noble metals have poor confinement properties in the infrared due to their prohibitively large permittivity, and provides a recipe for turning a material with large carrier concentration into a massively diluted system with plasmonic properties akin to those of true 2D-materials. To conclude, we propose and demonstrate the concept of metaplasmonics, which offers a promising path to fill the near- and mid-infrared technological gap for high quality plasmonic materials, and provides a new material system to study the effects of extreme plasmonic confinement, including nonlinear and quantum plasmonics.

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