Extreme plasmonics with atomically smooth monocrystalline gold flakes

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In this contribution, I will present an overview of some of our recent and ongoing research programs on extreme plasmonics, where the main player is a chemically synthesized monocrystalline gold flake. First, we removed ambiguity in reported values of the absorption losses for the perfect gold [1]. This was done by directly measuring propagation losses of surface plasmon polaritons (SPP), supported by the gold-air interface, with a scattering-type scanning near-field optical microscopy (SNOM). We designed our experiment to remove all possible excuses and correction factors. It was found that a commonly used reported dielectric function of monocrystalline gold predicts ~1.5 shorter SPP propagation length than the one measured in our experiment. Surprisingly, the best match with our measurements was found for a dielectric function reported for properly fabricated polycrystalline gold.

The second part of my contribution is, as promised by the title, devoted to the extreme plasmonics [2]. Particularly, we decided to probe the influence of the nonlocal electrodynamic surface response of the electron gas, which predicts corrections to the propagation constant of the highly confined plasmonic mode. Particularly, nonlocal effects predict higher propagation losses, therefore we designed our experiment to remove all possible sources of losses (scattering on surface roughness and domain boundaries) by using atomically smooth monocrystalline gold flakes and atomic layer deposited alumina. The influence of nonlocal response becomes non-negligible only for highly confined modes, therefore we selected a gap SPP mode (GSP) as a test platform, where two gold flakes are separated with a nanometer-thin alumina. We use scanning near-field optical microscopy to directly access the near-field of such confined gap plasmon modes and measure their dispersion relation via their complex-valued propagation constants. We compare our experimental findings with the predictions of the generalized nonlocal optical response theory to unveil signatures of nonlocal damping, which becomes appreciable for few-nanometer-sized dielectric gaps.

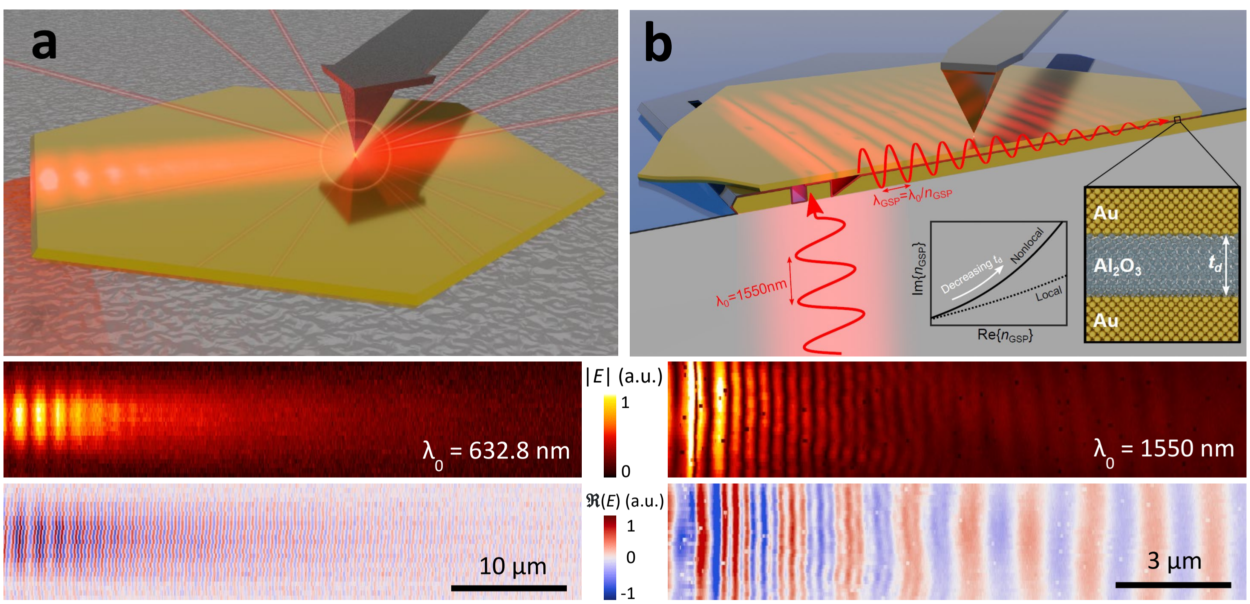

Fig. 1 Direct near-field probing of plasmonic modes, supported by atomically smooth monocrystalline gold flakes. **a** Measurements of the SPP propagation length, directly related to the absorption losses of gold. **b** Measurements of the GSP propagation constant, verifying the generalized nonlocal optical response theory.

References

[1] Lebsir, Y., Boroviks, S., Thomaschewski, M., Bozhevolnyi, S. I., and Zenin, V. A. 2022. *Nano Lett.* **22**, 5759-5764.

[2] Boroviks, S., Lin, Z.-H., Zenin, V. A., Ziegler, M., Dellith, A., Gonçalves, P. A. D., Wolff, C., Bozhevolnyi, S. I., Huang, J.-S., Mortensen, N. A. 2022. *Nat. Commun.* **13**, 3105.