Nonlinear plasmonic response of crystalline few-atom-thick silver films

Álvaro Rodríguez Echarri
Joel D. Cox, F. Iyikanat, and F. Javier García de Abajo
ICFO – Institut de ciències fotòniques, The Barcelona Institute of Science and Technology, Castelldefels (08860), Spain
alvaro.rodriguez@icfo.eu

Abstract : Atomically thin crystalline metallic films are attracting attention as suitable platform material for plasmonics owing to their ability to confine light down to few-nanometer scales with tolerable optical losses, well below those of amorphous metal films [1,2] (see Fig1a). Recently, substantial advances have been made to synthetize films with thickness down to a few atomic planes and demonstrate that they support surface plasmons (SP, collective oscillations of conductions electrons at metal-dielectric interfaces) with high quality factor [3]. These modes exhibit thickness-dependent dispersion relations, as illustrated in Fig1b by means of the loss function for silver films with a (111) crystallographic orientation. Such plasmons confine light, thus boosting the magnitude of linear fields by several orders of magnitudes, making them ideal to explore nonlinear phenomena, which in addition are combined with intrinsic features arising from the band structure of thinner films as demonstrated by comparing panels c and d, where we represent the density of states of a thick film consisting of $N = 100$ and $N = 5$ atomic planes, respectively. In our work, we employ a quantum mechanical (QM) method based on a one-dimensional potential [4] that capture the salient electronic-structure features of the Ag (111) surface to compute the optical response within the random-phase approximation (RPA). We extend this method to calculate the nonlinear response of the metallic films, and as shown in Fig1e for second harmonic generation, we define a figure of merit that facilitates the comparison of the nonlinear plasmonic yield as a function of film thickness. We find that thinner films give rise to larger nonlinear response. We also explore third order processes like third harmonic generation and Kerr effect, showing a similar increase with decreasing film thickness.

REFERENCES

[1] D. N. Basov, M. M. Fogler and F. J. García de Abajo, Science 354, aag1992 (2016).
[2] D. Alcaraz Iranzo et al., Science 360, 291 (2018).
[3] Z. M. Abd El-Fattah, V. Mkhitaryan et al., ACS Nano 13, 7771-7779 (2019).
[4] E. V. Chulkov et al, Surf. Sci. 437, 330-352 (1999).

FIGURES

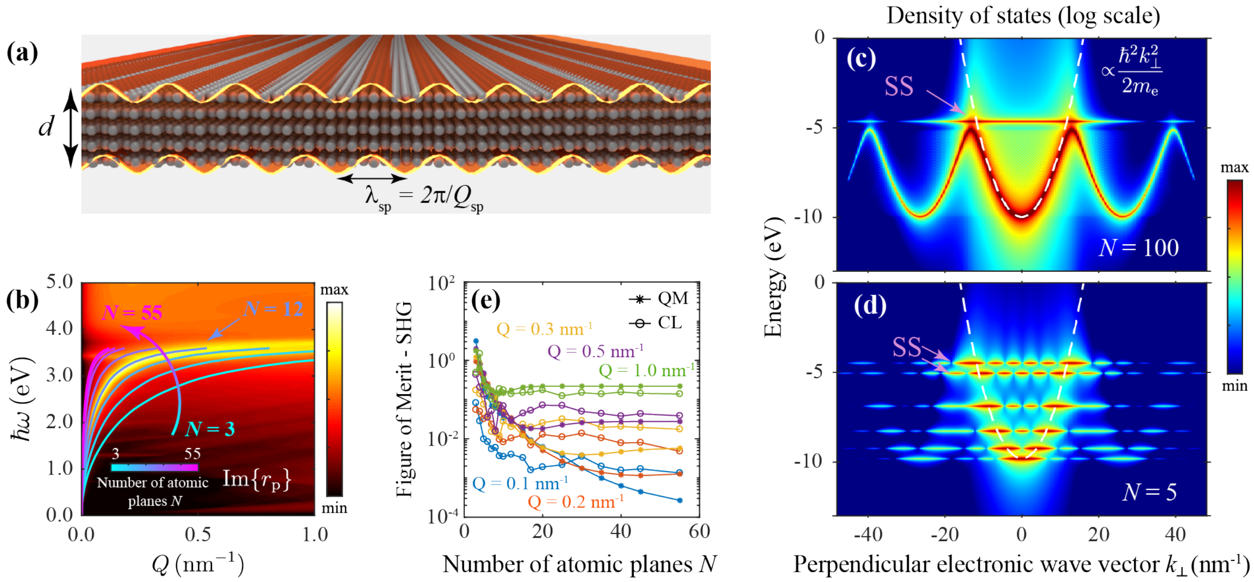

Figure 1: (a) Illustration of surface plasmons (SP) in a few-atom-thick silver (111) film. (b) SP dispersion relation calculated via the loss function (imaginary part of the reflection coefficient) in momentum and energy space. Dispersion curves of SPs for various thicknesses are superimposed on the dispersion map and indicated by the coloured code in the legend. (c,d) Electronic density of states obtained by Fourier-transforming the out-of-plane electronic wave functions for $N = 100$ and $N = 5$ Ag(111) atomic layers, respectively. (e) Figure of merit quantifying the nonlinear plasmonic yield using quantum-mechanical (circles) or classical (stars) models for selected values of the in-plane parallel wave vector Q.