Fundamentals and applications of electron–light interactions in nanoplasmonics

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Recently, progresses in instrumentation have been enabling extraordinary achievements in electron-beam-based spectroscopies [1], such as atomic-scale spectroscopy (down to a single atom) of vibrational and excitonic modes in two-dimensional materials as well as quantum optical investigations using free-electrons [1]. Firstly, we exploit the capability of electron beams (e-beams) to produce deeply subwavelength near-fields that carry large wave vector components commensurable with those governing the quantum-mechanical response of metals, and demonstrate their ability to interrogate quantum nonlocal effects imprinted on the optical response of metallic structures [2]. In particular, we discuss how electron energy-loss spectroscopy (EELS) and cathodoluminescence (CL) can be used to infer quantum surface response functions (e.g., the Feibelman *d*-parameters) that encapsulate those quantum effects (Fig. a1). We show that quantum nonlocal effects lead to substantial resonance shifts and broadening of the EELS and CL peaks associated with plasmon excitations in several relevant metallic nanostructures (Fig. a2 shows an example for Ag spheres) [2]. Our findings pave the way for new approach for measuring the quantum response of metals using state-of-the-art e-beam spectroscopies. In the second part, we harness the potential of photoemission spectroscopies to unveil quasiparticle dynamics and investigate multi-plasmon effects imprinted in the photoemission spectrum from plasmonics nanostructures. Such effects, driven by electron–plasmon interactions, lead to the emergence of satellite peaks distanced from the main core-level peak by quanta of the plasmon energy (Fig. b1) [3]. Our work extends previous theories which have thus far been limited to semi-infinite planar systems and offers an additional route to study multiple plasmon excitations in nanoplasmonics. We derive universal scaling laws for the plasmon satellite probabilities and investigate the impact of the system's morphology and dimensionality [4]. We further predict the appearance of energy-gain satellites (whereby quanta of the plasmon energy are transferred to the outgoing photoelectron) in optically pumped (preceding the photoemission event) nanostructures (Fig. b2) [4]. Our work opens enticing new directions for investigating ultrafast electron–plasmon interactions in nanophotonics.

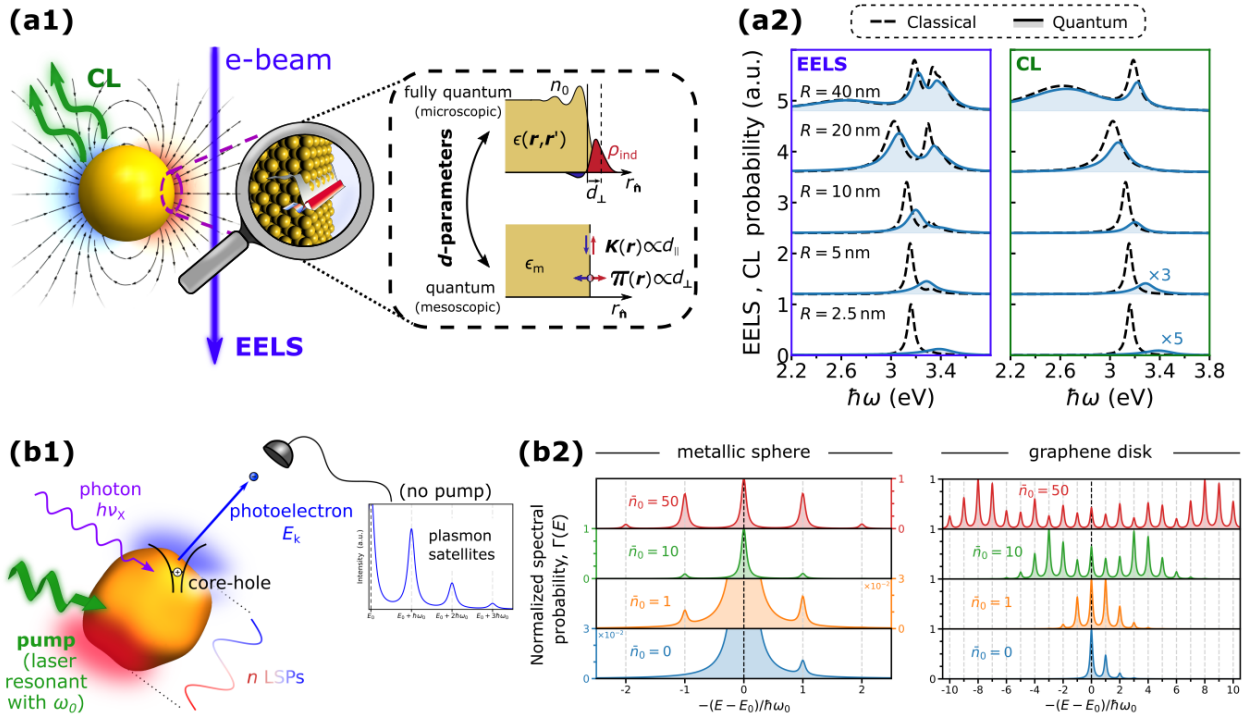

Fig. 1. (a1) Probing the quantum surface response of metals using e-beam spectroscopies. **(a2)** Comparison of classical and quantum treatments of the EELS and CL spectra in Ag spheres of different radii R in a host $\epsilon_d = 2$. **(b1)** Schematic of core-level photoemission and ensuing multi-plasmon excitation. **(b2)** Photoelectron spectra from a $D = 10$ nm silver sphere (left) and from a $D = 20$ nm graphene disk (right) exhibiting multiple plasmon satellite sidebands (when optically pumped prior to photoemission, the initial average number of plasmons is $\bar{n}_0 > 0$).

References

[1] Polman, A., et al. 2019. *Nat. Mater.*, **18**, 1158–1171. García de Abajo, F. J. and Di Giulio, V. 2021. *ACS Photonics*, **8**, 945–974.
[2] Gonçalves, P. A. D. and García de Abajo, F. J. 2023. *arXiv:2301.10040.*
[3] S. Hüfner, *Photoelectron Spectroscopy: Principles and Applications*. 2003, Springer, Berlin.
[4] Gonçalves, P. A. D. and García de Abajo, F. J. 2023. *submitted.*