LIFETIME MEASUREMENTS OF PLASMON-EXCITON POLIARITONIC EMISSION USING SUPERCRITICAL ANGLE FLUORESCENCE MICROSCOPY

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Surface plasmon polaritons (SPPs) are electromagnetic waves that propagate along a metal-dielectric interface. Their excitation relies on the coupling between coherent oscillations of free electrons and photons, which can be drastically enhanced with the grating coupler approach [1]. Scattered light in the periodic arrays of metallic nanosotructures interfere constructively with damping of localized plasmon resonance (LPR) of individual structures, resulting in ultra-narrow plasmonic resonance known as a surface lattice resonance (SLR) [2]. SLR has a wide range of applications in sensing [3], but can also be used to enhance the spontaneous emission rates of organic fluorescent molecules deposited on top of structured metallic surface. In this work we present various energy exchange dynamics, differing in decay rates, by implementing a near-field collection via supercritical angle fluorescence (SAF) microscopy.

We investigate a metallic structure of gold bumps (period around 500 nm) on 50 nm thick gold, with drop-caster and evaporated fluorescent dye of rhodamine R6G, dissolved in water on top of the structure. The fluorescence was excited with a 515 nm femtosecond laser through oil immersion objective, collected in an epi-configuration and filtered at 600 nm. Additionally, a 4F optical system with an adjustable aperture was used for spatial filtering of the back focal plane (BFP) to select particular fluorescence signals (Fig. 1). Near-field and far-field emissions were examined and attributed to SAF and undercritical angle fluorescence (UAF) respectively. UAF mostly contains the uncoupled dye molecules in the bulk dye, thus exhibiting a longer fluorescence lifetime. On the contrary, the SAF decay is fastest and limited by the instrumental response function (IRF) of the detector. This is due to the combination of radiative energy enhancement and non-radiative fluorescence quenching in our sample [2]. The latter effect occurs when an excited-state fluorophore is in close proximity to a metal surface and an instant energy transfer to surface electrons is induced. As radiative and non-radiative decay rates increase, the total lifetime of SAF decreases. In addition, the first-order diffraction rings, formed from multidirectional leakage radiation by SLR, were also observed and had a mix of lifetimes due to UAF influence and filtering limitations.

The lifetime decay measurements were consistent with theoretical predictions, despite being IRF limited. Research on SLR-enhanced coherent emission can lead to development and improvement of quantum nanophotonic devices as well as advanced microscopy and optical biosensing [4].

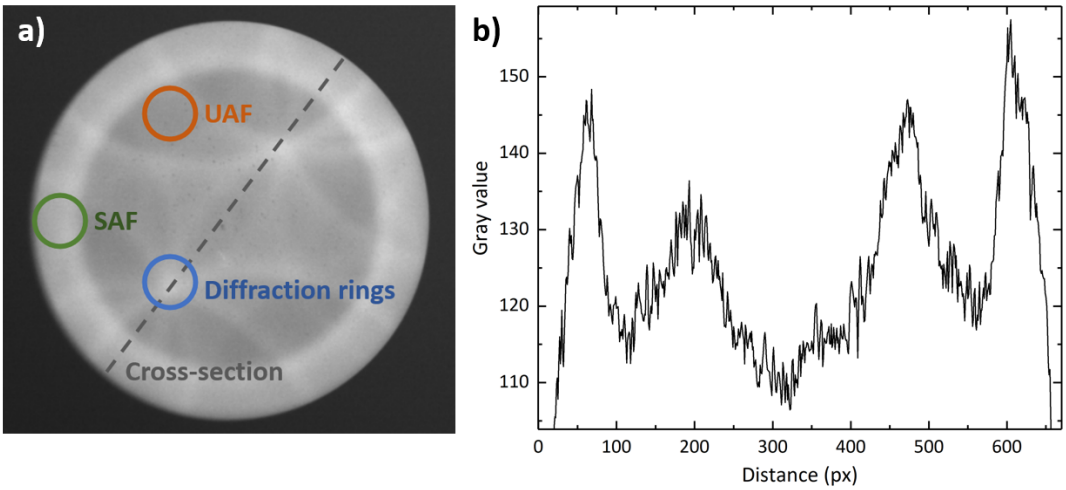

Fig. 1. a) BFP with standard filtering of examined sample, b) Corresponding cross-section taken from a).

[1] E. Stankevičius, K. Vilkevičius, M. Gedvilas et al., "Direct laser writing for the formation of Large-Scale gold microbumps arrays generating hybrid lattice plasmon polaritons in VIS–NIR range," Advanced Optical Materials, vol. 9, no. 12, Apr. 2021, doi: 10.1002/adom.202100027.
[2] P. Jurkšaitis, J. Anulytė, E. Spalinskaitė et al., "Plasmon–exciton polaritonic emission lifetime dynamics under strong coupling," Nanophotonics, vol. 14, no. 14, pp. 2485–2493, Jun. 2025, doi: 10.1515/nanoph-2025-0129
[3] U. F. S. Roggero, C. J. Rojas-Bejarano, C. A. Herreño-Fierro et al., "Surface lattice resonances for high-performance sensing: a study on gold nanoparticle arrays," Optics Express, vol. 33, no. 14, p. 30328, Jun. 2025, doi: 10.1364/oe.564444
[4] W. Deng and E. M. Goldys, "Plasmonic approach to enhanced fluorescence for applications in biotechnology and the life sciences," Langmuir, vol. 28, no. 27, pp. 10152–10163, May 2012, doi: 10.1021/la300332x