Hybrid Metrology for the Development of Self-Assembled Metamaterials

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With the revision of the International System of Units (SI), as approved by the General Conference on Weights and Measures (Conférence générale des poids et mesures [CGPM]) in 2019, units are defined based on fixed, with an exact value, fundamental physical constants. This opened new possibilities for the direct realization of the SI units for any experiment that correlates a measurable quantity to a fundamental constant of nature. In this framework, the recent advances in the generation and manipulation of individual photons have become of great interest for their employment as a primary traceable route to the SI. This is particularly true for emerging quantum technologies, where sources that generate single photons at a known rate can be exploited on the *mise en pratique* for the practical realization of candela (cd) in the SI, as reported by the Consultative Committee for Photometry and Radiometry (CCPR). In this context, nanoplasmonic structures such as hyperbolic metamaterials (HMMs), play a key role on the development of single-photon sources that operate at room temperature, guaranteeing efficient directional and broadband single-photon spontaneous emission enhancement. To date, the intrinsic constrains imposed by conventional lithographic and material deposition/growth techniques, limit the fabrication of HMMs in terms of configurations and compositions, with consequent restrictions on the resulting optical properties. In this work, the development of a new lithographic technique, based on self-assembly (SA) of block copolymers (BCPs), is proposed as a suitable fabrication route for the realization of HMMs, with in-plane optical axis, that operate in Vis frequencies. The novelty of this research activity relies on the employment of a dewetting process of BCP/homopolymer blend thin films over topographically defined patterns. The combination of bottom-up BCP nanopatterning and top-down optical lithography leads to ordered periodic lamellar nanostructures, in a hierarchical configuration, representing ideal platforms for the subsequent pattern transfer into hybrid Au/air HMMs. The hyperbolic behavior of the proposed material is supported by a strong reduction in the fluorescence lifetime dynamics of defects in nanodiamonds placed on top of the HMM, along with a computed Purcell factor as high as 32 at 580 nm. However, due to the great sensibility of the optical properties of HMMs to their dimensional and compositional parameters, even slight variations in terms of nanofeature's height or refractive index determine a significant modulation of the photon emission enhancement. With the aim to optimize the optical performances, it is therefore crucial to establish a relationship of materials' functionality over their geometric and compositional parameters. This can be achieved by implementing, in a hybrid metrology approach, different analytical techniques to gain a comprehensive characterization of such nanostructures. This measurement strategy requires therefore the fabrication of materials that could be employed as model systems for a reliable interpretation and

correlation of materials' functionality to their underpinning physical and chemical properties. Novel synthetic methods, based on the selective infiltration and growth of metal oxides, such as the sequential infiltration synthesis (SIS), are developed in order to provide the degree of control on the material fabrication, needed to meet the best conditions for enhanced optical performances. Then, an analytical strategy based on a hybrid metrology approach is applied to reveal dimensional and analytical information on such nanomaterials. The implementation of synchrotron-based traceable X-ray techniques with scanning probe and electron microscopies, along with a good knowledge of the relationship of each targeted measurand to the specific physical or chemical quantities of interest, allows for mutual validation and more complete, coherent description of complex samples.

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