Nanofluidic Flow-Assisted Assembly (NFAA) of Well-Dispersed Plasmonic Nanostructures into Nanoslit Sensors

Hiroto Izuoka^{1, 2}, Hongsuk Nam¹, Jeong Seop Yoon¹, Wenjie Wan³, and Xiaogan Liang^{1, *}

¹Mechanical Engineering Department, University of Michigan, Ann Arbor, MI 48109

² Mechanical Science and Engineering, Nagoya University, Nagoya, Aichi 464-8603, Japan

³University of Michigan-Shanghai Jiao Tong University Joint Institute and Department of Physics and Astronomy, Shanghai Jiao Tong University, Shanghai 200240, China

The sensing techniques utilizing plasmonic properties of metallic nanostructures (*e.g.*, nanoparticles (NPs) and nanorods (NRs)) have been a subject of considerable interest lately because of their high detection sensitivity and low limit-of-detection (LOD).^{1,2} Examples of such plasmonic device principles include Localized Surface Plasmon Resonance (LSPR) and Surface-enhanced Raman Scattering (SERS).¹⁻³ The key step to fabricate such plasmonic sensors is the deposition of high-density, well-dispersed metal nanostructures on solid surfaces with integration of well-sealed nano/microfluidic structures. However, the requirements of a high coating density and a good dispersion of metal nanostructure aggregation and inter-particle plasmonic coupling, which result in serious degradation of detection sensitivity and LOD. Because of this reason, the regular coating methods usually deposit nanostructures with average areal densities less than 1 nanostructure/ μ m², ³ which limits the sensitivities of resultant plasmonic sensors. Furthermore, the undesirable aggregation of high-density plasmonic nanostructures results in a too poor fabrication yield to enable scale-up biosensor arrays for practical multiplexing sensing applications. New generic nanoplasmonics/nanofluidics-compatible nanofabrication methods are needed to tackle this challenge.

Here, we present a method for assembling plasmonic nanostructures into nanofluidic sensor structures. This method is termed as Nanofluidic-Flow-Assisted-Assembly (NFAA). NFAA utilizes nanofluidic flows with large shear rates to selectively coat high-density, well-dispersed plasmonic nanostructures into sensor areas. Using NFAA, we have demonstrated the coating of Au NPs and NRs with various areal densities into nanolit-based sensors.

Fig. 1a schematically illustrates the NFAA setup. Before a NFAA process, the nano/microfluidic structures for manipulating analyte solutions are firstly fabricated into a quartz substrate and subsequently sealed. Here, a (or a set of) nanoslit channel(s) serves as the critical structure for assembling plasmonic nanostructures as well as the sensing area. During the NFAA process, a colloidal solution of plasmonic nanostructures is filled into the whole fluidic structure, and the solution flow is constantly driven by a pressure drop. Within the nanoslit area, the large shear rate of the nanofluidic flow is important to enable the plasmonic nanostructures uniformly deposited on the top/bottom surfaces and prevents aggregation. The final coating density can be controlled by adjusting the total flow time. Fig. 1b shows the optical micrograph of an as-fabricated nano/microfluidic device for testing NFAA. Fig. 2 displays the SEM images of exemplary plasmonic nanostructures coated using NFAA, including (a) 40 nm \times 80 nm NRs with average areal density of ~ 20 NR/µm²,³ and (b) 5 nm Au NPs with average areal density of ~ 200NP/µm². We will present additional study on the dependence of the resultant plasmonic nanostructure densities/dispersions on the processing parameters such as flow times, nanoslit dimensions, and dimensions of plasmonic nanostructures.

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This work advanced the critical nanofabrication technology for integrating plasmonic nanostructures with nanofluidic structures and enabling new sensing device functionality.



Figure 1: (a) illustration of the NFAA setup; (b) optical micrograph of an as-fabricated nano/microfluidic device for testing the NFAA method.



Figure 2: SEM images of (a) 40 nm \times 80 nm AuNRs with average areal density of ~20 NR/ μ m², and (b) 5 nm spherical AuNPs with average areal density of ~200 NP/ μ m², where were deposited into nanoslit sensor areas using the presented NFAA method.