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Evaporative self-assembly of gold nanorods into macroscopic 3D plasmonic superlattice arrays for SERS detection

Penghui Li¹, Xue-Feng Yu¹, Paul K. Chu²

¹ Center for Biomedical Materials and Interfaces, Shenzhen Institutes of Advanced Technology, Chinese Academy of Sciences, No.1068 Xueyuan Avenue, Nanshan District, Shenzhen, 518055, China;

² Department of Physics & Materials Science, City University of Hong Kong, Tat Chee Avenue, Kowloon, Hong Kong, China

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Introduction: The buildup of 3D superlattice with collective electro-optical properties by ordered self-assembly of nanoparticles has become the cutting-edge technology in the research field of nanomaterials [1]. Droplet evaporative self-assembly is a simple and effective technique to build the superlattice structure. However, the well-known coffee-ring effect seriously hinders the ordered packing of the nanoparticles. A strategy to totally suppress or even reverse the coffee-ring effect is highly desirable in order to accomplish homogenous self-assembly of nanoparticles into a macroscopic structure [3].

Materials and Methods: The surface chemistry of gold nanorods and substrate were regulated to achieve the reversing of coffee-ring effect during the evaporative self-assembly [3]. The evaporative self-assembly processes with different parameters were recorded, and the deposition behaviors of gold nanorods during evaporation were analyzed. The morphology and structure of the obtained 3D superlattice were observed with confocal laser scattering microscopy and scanning electron microscopy (SEM). The plasmonic properties of the 3D superlattice array were determined in surface-enhanced Raman scattering (SERS) with malachite green (MG) as the model analyte.

Results and Discussion: The results demonstrated that the coffee-ring effect has been completely reversed and consequently, homogenous deposition is achieved based on the combined effects of Marangoni flow and contact line receding. All the gold nanorods are dense, regular, and nearly vertically aligned with respect to the surface of the substrate, and form the homogeneous macroscopic 3D superlattice array (Figure 1). The SERS detection of MG with this superlattice array exhibit sensitive and reproductive results, with the detection limit down to 1.0×10^{-10} M and reproducibility lower than 10% (Figure 2).

Conclusion: Millimeter-scale 3D superlattice arrays composed of dense, regular, and vertically aligned gold nanorods have been fabricated and demonstrated excellent SERS sensitivity and reproducibility. Such 3D organization of gold nanorods into macroscopic superlattices results in excellent plasmonic substrates suitable for macroscopic sensing applications. This reproducibility and operability of the droplet evaporation strategy can be readily extended to the self-assembly of various plasmonic nanoparticles into macroscopic 3D superlattice arrays and bridge the gap between nanoscale materials and macroscopic applications [4].

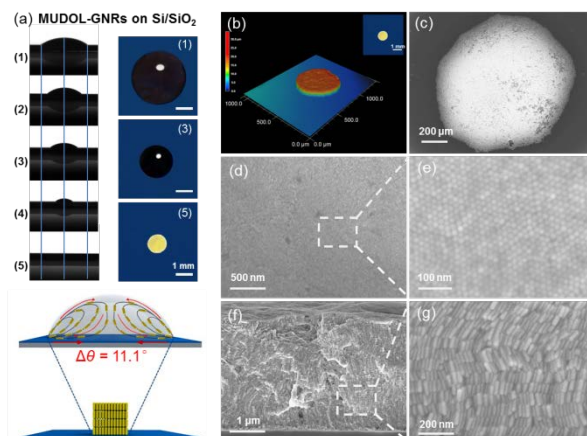


Figure 1 (a) Evaporation process and deposition behavior of gold nanorods during evaporative self-assembly; (b-g) Morphology and structure of the obtained gold nanorods 3D plasmonic superlattice array [3].

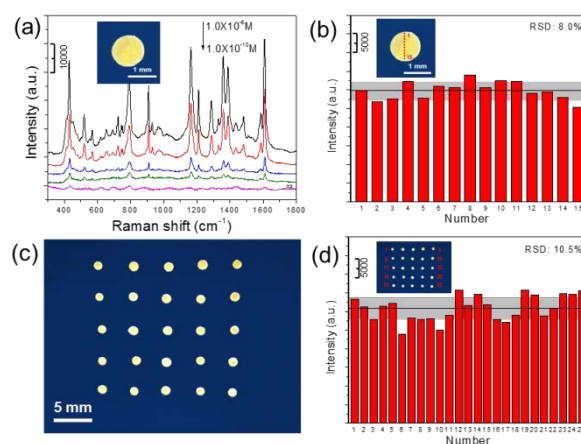


Figure 2 SERS detection performance of MG molecules with the gold nanorods 3D plasmonic superlattice array [3].

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Corresponding:

Email: xf.yu@siat.ac.cn

Tel: 86-755-86392212

Fax: 86-755-86585222