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Laser-irradiated inclined metal nanocolumns for selective, scalable, and room-temperature synthesis of plasmonic isotropic nanospheres†

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Plasmonic nanocrystals, which exhibit extraordinary optical properties, are challenging to grow in selective positions with a cost-effective and high-throughput process. We demonstrate that plasmonic isotropic gold nanospheres (AuNSs) can be selectively synthesized on wafer-scale rigid and flexible substrates at room temperature by laser irradiation. First, we prepare gold nanocolumn (AuNC) thin films on sapphire and polydimethylsiloxane substrates with glancing angle deposition (GAD). Then, a KrF excimer laser is exposed at selected positions with a 24 ns pulse duration. Finally, highly isotropic AuNSs as plasmonic nanocrystals are synthesized at the targeted positions. We suggest that the formation of such isotropic AuNSs is caused by reshaping from the top of the AuNCs; this is verified by the temperature distribution in the AuNCs during laser irradiation through finite element method simulations. We further investigate the formation of AuNSs by varying the laser energy density and the kind of substrate. By using a simple mask process, we demonstrate patterning of the letters "KIST" *via* selectively grown AuNSs on a flexible substrate. The simple laser irradiation process on GAD-grown metal NC thin films is expected to be a promising method for scalable synthesis of plasmonic isotropic NSs at targeted positions with a rapid process and at room temperature.

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Introduction

Since its discovery, plasmonics has been extensively studied for next-generation photonic devices owing to the potential for extraordinary optical properties.[1] In particular, nanoplasmonics has garnered a significant amount of research attention due to localized surface plasmon resonance (LSPR), which allows a significant enhancement of the electromagnetic (EM) fields in the vicinity of plasmonic nanocrystals.[2] Generally, the position of LSPR is sensitive to the size, shape, and type of plasmonic nanomaterials.[3,4] Doped semiconductors,[5] metal oxides,[6] chalcogenides,[7] graphene[8] and metallic nanocrystals[9–12] have been utilized for various plasmonic applications such as biosensors,[13,14] solar cells,[15] light-emitting diodes (LEDs),[16] and water splitting.[17] With the significant development of nanotechnology, novel plasmonic nanostructures such as nanorods,[18] nanoprisms,[19] nanocages,[11] nanoshells,[20] nanocolumns (NC),[21] and nanospheres (NS)[22] have been reported. However, low-cost and fast nanofabrication techniques on large areas at selective positions remain a challenge for industrial applications of plasmonic nanostructures. A thermal annealing process is an effective way to provide high-throughput and low-cost post-treatment on large substrates. Interestingly, metallic thin films and nanorods have been converted to NSs by thermal treatment at temperatures much lower than their melting points.[23,24] This phenomenon can be explained by Rayleigh instability[23–25] and thermal reshaping behavior, which act to minimize the surface energy of materials by surface diffusion.[25] Plech *et al.* reported the transformation of gold nanowires to NSs due to surface melting at only 400 K.[26] Furthermore, our group recently developed gold nanotriplets on nanoporous anodic aluminum oxides *via* thermal treatment

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