**2D Freestanding Janus Gold *Plasmene* nanosheets**

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Recently, a new concept in 2D plasmonics nanoassemblies– ‘*Plasmene*’ was established, which is defined in analogy to graphene as free-standing, one-particle-thick, ordered nanoassemblies of nanoparticles (“meta-atoms”) from the “plasmonic periodic table”. *Plasmene* shows great potential in the design of SERS substrates due to its unique structure-dependent physicochemical functionalities. Previous *Plasmene* nanosheets are typically constructed from symmetrical constituent building blocks, which have identical structural and functional properties on both sides. Here, a general ligand symmetry-breaking strategy is reported to grow 2D Janus gold nanocrystal *Plasmene* nanosheets with nanocube morphology on one side yet with nanostar on the opposite side. Such asymmetric metallic structures lead to distinct wetting and optical properties as well as surface-enhanced Raman scattering (SERS) effects. In particular, the SERS enhancement of the nanocube side is about 20-fold of that of the nanostar side, likely due to the combined “hot spot lightening-rod” effects. This is nearly 700-fold of SERS enhancement as compared with the symmetric nanocube *Plasmene* without Janus structures. This work therefore opens up the possibility of a new 2D nanoassembly design rule through breaking the morphology symmetry and provides an effective approach to strengthen the material properties by the integration of different properties into one structure.