Twisted Epitaxy of Ag Nanoparticles Confined Between Misoriented MoS_2 Bilayers

Pawel Czaja,[a,b] Yi Cui,[a] Prasanna Sarkar,[a] Harold Y. Hwang,[c] Robert Sinclair,[a,*] and Yi Cui[a,*]

[a]*Department of Materials Science and Engineering, Stanford University, Stanford 94305, USA*

[b]*Institute of Metallurgy and Materials Science, Polish Academy of Sciences, 25 Reymonta St., 30-059 Krakow, Poland*

[c]*Department of Applied Physics, Stanford University, Stanford, CA 94305, USA*

Twisted epitaxy is a recently identified phenomenon in metal–2D van der Waals heterostructures, first observed in the Au–MoS_2 system, where epitaxial nanoparticles reorient under the influence of a controlled twist between encapsulating layers. Here, we investigate the extension of this effect to the Ag–MoS_2 system, motivated by the chemical similarity, comparable atomic radius, and crystal structure of silver and gold.

Using high-resolution through-foil transmission electron microscopy (TEM), we examine Ag nanoparticles confined between two mechanically exfoliated MoS_2 layers with a predefined twist angle (0–60°) and subjected to annealing at 500 °C. Initially, the nanoparticles adopt an epitaxial relationship with the bottom MoS_2 (0001) substrate (Ag $\langle 220 \rangle \parallel \langle 1\bar{1}20 \rangle$ MoS_2). Upon annealing, they undergo pronounced morphological and structural evolution. For bilayer twist angles up to ~10°, a gradual reorientation toward an intermediate alignment between the top and bottom MoS_2 lattices is observed. At larger twist angles, the Ag nanoparticles exhibit only minor deviations (~2°) from their initial epitaxial orientation.

Compared to the Au–MoS_2 system, the Ag–MoS_2 heterostructure displays a broader range of twist responses and more complex reorientation behavior. These results demonstrate that twisted epitaxy is not limited to Au-based systems but is likely a more general phenomenon applicable to a wider class of metal–2D material heterostructures.