

***In situ* TEM Investigation on Rotation and Coalescence Behaviors of Au Nanoparticles on h-BN Substrate**

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Two dimensional (2D) materials have opened wide possibility for novel heterostructures. For instance, 2D nanosheet decorated with metal nanoparticles can be of great advantages in catalyst, sensor, and energy storage device applications. However, a major question related to the stability of nanoparticles on 2D substrates still remains unclear.

Among the 2D family, hexagonal boron nitride (h-BN) is attracting increased attentions since it's unique 2D form combined with wide band gap insulating intrinsic. Gold nanoparticles decorated h-BN composites have been shown of great activity for oxygen reduction reaction (ORR) [1], hydrogen peroxide detection, and surface-enhanced Raman scattering (SERS) [2]. These applications require the Au particles to be stable enough with minimum sintering. However, little is known of how and when the sintering of Au behaves on h-BN substrate.

In situ TEM provides a powerful way to study such dynamic material behaviors. In this study we utilized electron beam to trigger the diffusion and coalescence processes of Au nanoparticles on h-BN substrate. The dynamic processes were then captured in real time. It is found that Au has strong trend to rotate at h-BN edge, and the coalescence behavior of nearby Au particles on h-BN were analyzed.

h-BN was first exfoliated into monolayer by liquid phase exfoliation method. Au nanoparticles were deposited onto h-BN from metal precursor. The synthesized Au nanoparticles are in size range of 2-10 nm. Au h-BN heterostructure sample was drop casted onto TEM grid and irradiated by electron beam with 300 kv acceleration voltage. The intensity and dose rate of electron beam were kept consistent during TEM experiments. Atomic resolution movies were acquired to capture the sintering behavior of Au particles in real time. Figure 1a shows the dynamics of an individual Au particle rotational process at h-BN edge. The corresponding FFT of each frame is shown in Figure 1b. It was found that the Au particle is rotating continuously with its (002) surface changing direction in a clockwise manner. The rotation is in addition accompanied by translational movement of the center of mass and surface reconstruction (Figure 1a), indicating the rolling behavior of Au particle at h-BN edge. Figure 2a shows the coalescence process of two adjacent Au particles on h-BN surface, as well as the corresponding FFTs focusing on whole area and individual particle area. The two Au particles were initially in different orientations, however with gradual rotation the two particles aligned with each other in terms of crystal orientation, and finally coalesced into a whole particle after surface reconstruction. Figure 2b shows an ABF image of several Au particles in the intermediate stage of coalescence. Although the orientation of particles is yet aligned, the formation of necking area between each particle is clear to be observed (Figure 2c). The current study can provide insights into how sintering of Au particles happens on h-BN substrate, and help gain deeper understanding of controlling the nanoparticle morphology on 2D material based heterostructures [3].

References:

[1] G Elumalai et al. *Electrochemistry Communications* **66** (2016), p. 53.

[2] Q Cai et al., *ACS Applied Materials & Interfaces* **8(24)** (2016), p. 15630.

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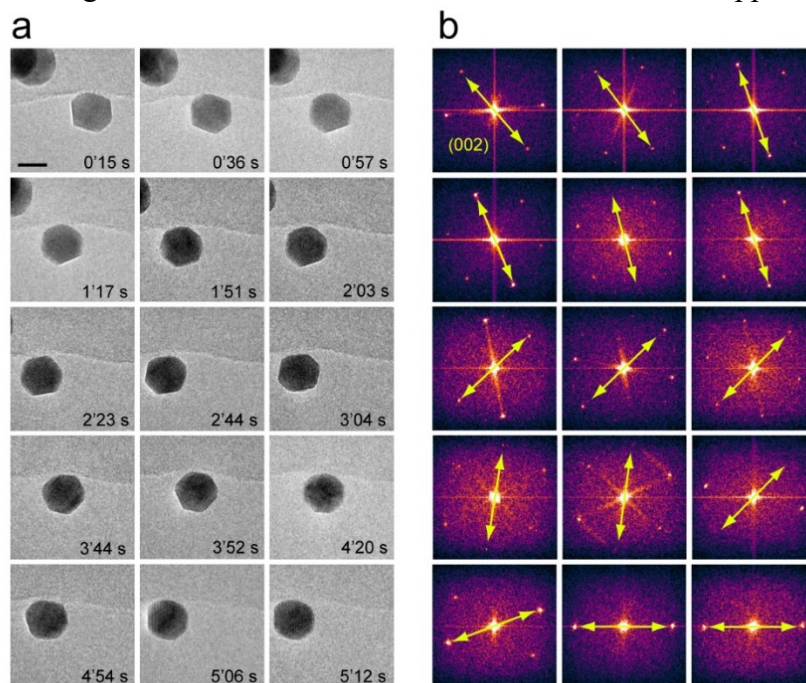


Figure 1. (a) Snapshots of Au particle rotation at h-BN edge. Scale bar: 5 nm. (b) FFT patterns from a. The arrows indicate the orientation of Au (002) surfaces.

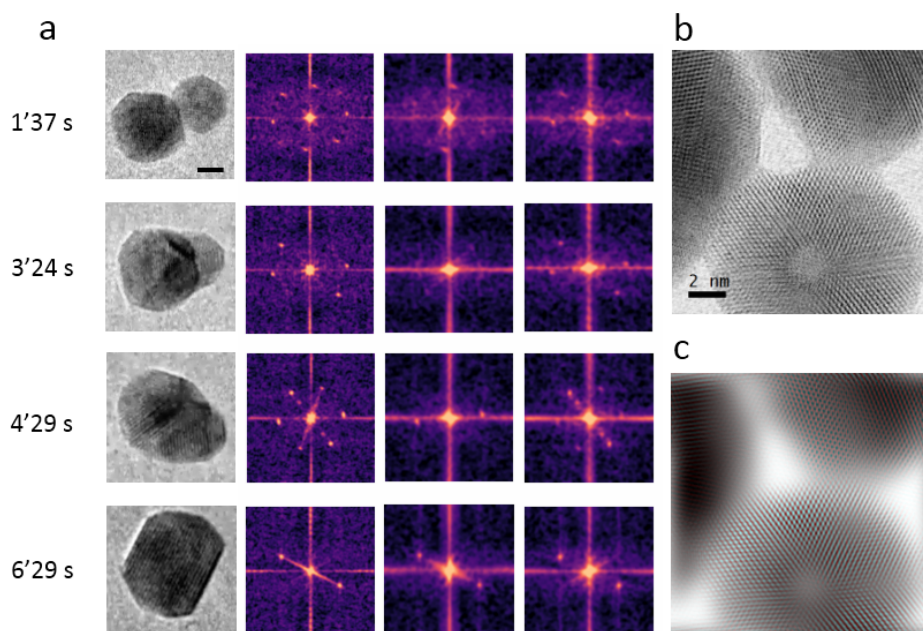


Figure 2. (a) Snapshots of Au particles coalescence on h-BN substrate. FFTs following each image corresponding to the whole image, smaller particle, and larger particle, respectively. Scale bar: 2 nm. (b) ABF image of Au particles in coalescence progress. (c) FFT filtered image corresponding to b.