

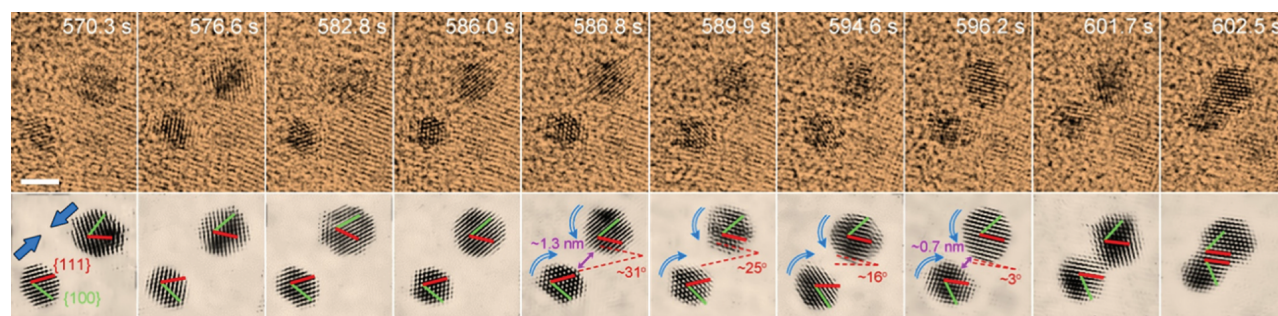
## *In-situ* liquid cell TEM investigation on oriented attachment of gold nanoparticles

With the support by the National Natural Science Foundation of China, the research team directed by Prof. Sun LiTao (孙立涛) from SEU-FEI Nano-Pico Center, Key Laboratory of MEMS of Ministry of Education, Southeast University, recently reported the direct imaging of ligand directed oriented attachment (OA) of Au nanoparticles, which was published in *Nature Communications* (2018, 9: 421).

Inside a liquid solution, the growth of nanocrystals through OA is now widely accepted as an alternative pathway to the conventional growth by Ostwald ripening. The considerable scientific importance of this technique for the synthesis of many materials has been well recognized. However, due to the difficulties to real-time image the dynamic process of OA especially at atomic-scale, the driving force responsible for the occurrence of OA has not yet been clearly identified in any specific case.

Here for the first time, Sun's group have tried to understand the driving force of OA by imaging the OA process of gold nanoparticles in real-time using a home-made liquid cell mounted within a high resolution transmission electron microscope (HRTEM). They show that when the separation distance between a particle pair is larger than twice the thickness of the surface ligand layer (sodium citrate), the particles rotate randomly in solution. In contrast, as they get so close that the surface ligands on the particles come into contact, the particles are no longer independent but behave as a single entity, which triggers off directional rotation, where the particles in a pair rotate in opposite directions until they reach a perfect alignment of their  $\{111\}$  facets. Eventually, the perfectly aligned particle pair makes a sudden contact with the concomitant expulsion of the ligands. Density function theory (DFT) calculations indicate that the lower ligand binding ability of the  $\{111\}$  facets is responsible for the preferential attachment of the two particles at their  $\{111\}$  facets rather than at  $\{100\}$  facets. All these results lead to a conclusion that the OA process is indeed controlled by the ligands adsorbed on particle surfaces

Above findings have provided a universal conceptual framework for understanding the OA process in depth, which is applicable to a variety of other ligand-mediated solution systems. Moreover, these results provide substantial experimental input to develop improved theories on nano-growth, and also enable the design of future multifunctional hierarchical nanomaterials.



**Figure** Image sequences showing the OA process of small gold nanoparticles at  $\{111\}$  facets, evolving into a twin structure.