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Hydration Layer-mediated Pairwise Interaction of Nanoparticles resolved by in situ TEM

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Hydration forces play an important role in various physical, chemical and biological processes. They are responsible for colloidal stability, lubrication, crystallization, and interactions between biological molecules, etc [1]. From earlier studies conducted at a macro and micro scale, it is known that these short-range repulsive forces may stall the approach of interacting surfaces [2]. However, the role of repulsive hydration forces on the interaction dynamics between nanoparticles remains unresolved. Here, we use in situ TEM imaging [3-5] and new image processing algorithms we visualize and quantify the pairwise interactions between gold nanoparticles in water and show that: *1*) hydration monolayer around each gold nanoparticle is largely responsible for exponentially increasing hydration repulsion, and *2*) the balance between van der Waals (vdW) force and hydration force gives rise to a transient nanoparticle pair [6].

We track the separation between multiple interacting gold nanoparticle pairs in water and find that when the separation between two interacting gold nanoparticle surfaces is two water molecules thick, they form a transient stable pair. When thermal fluctuations drain the water between nanoparticle surfaces, hydration forces vanish and as a result of attractive vdW these nanoparticles jump to contact. The implications of our results for nanoparticle self-assembly will also be discussed [7].

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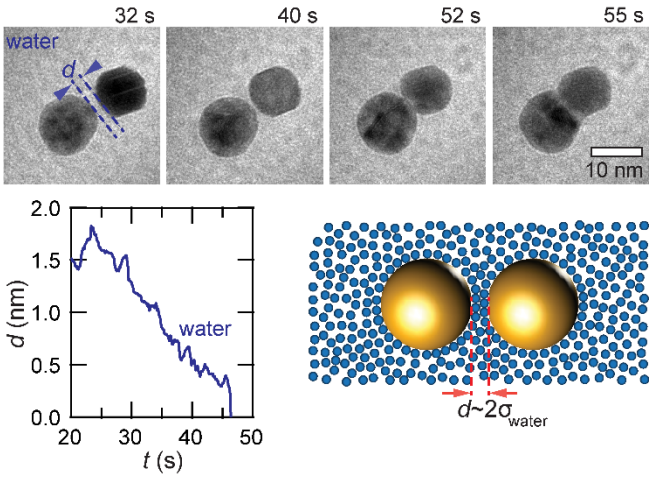

Figure 1. Pairwise interaction of gold nanoparticles. Top: Time-series TEM images of two gold nanoparticles undergoing coalescence in water. **Bottom-left:** Separation *d* between the two interacting gold nanoparticles; jump-to-contact occurs at a separation of ~5Å. **Bottom-right:** The corresponding schematic of these nanoparticle interactions in water (blue spheres-water molecules, orange sphere-gold nanoparticle) that arise when gold nanoparticles come into contact with each other with their one-water-molecule-thick hydration layer.

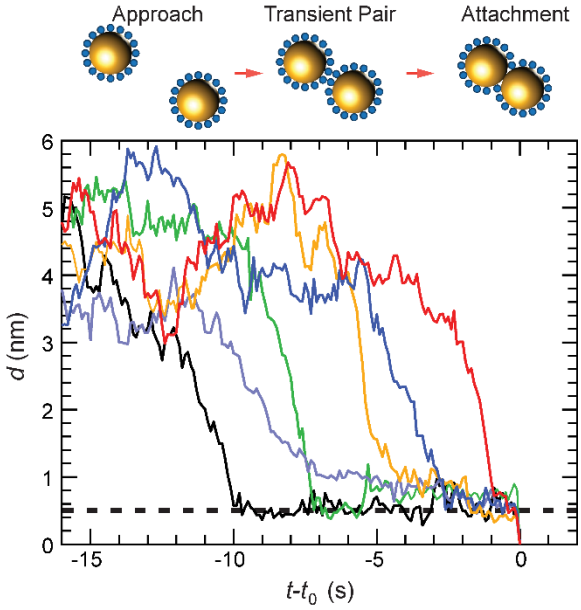

Figure 2. Pairwise separation between interacting gold nanoparticles in water. The plots of the pairwise separations between nanoparticles show the approach down to a pairwise separation of ~ 0.5 nm separation set by the combined thickness of the hydration layer of each nanoparticle, at which point the these nanoparticles form a sterically stabilized transient pair. Once the surface-bound water molecules between nanoparticles are drained, the nanoparticle surfaces come into contact at $t - t_0 = 0$ s, and coalesce, as schematically illustrated in the top panel. The dashed line shows the spacing which corresponds to two diameters of water molecules.

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