Supporting Information

Emergent, Collective Oscillations of Self-Mobile Particles and Patterned Surfaces under Redox Conditions

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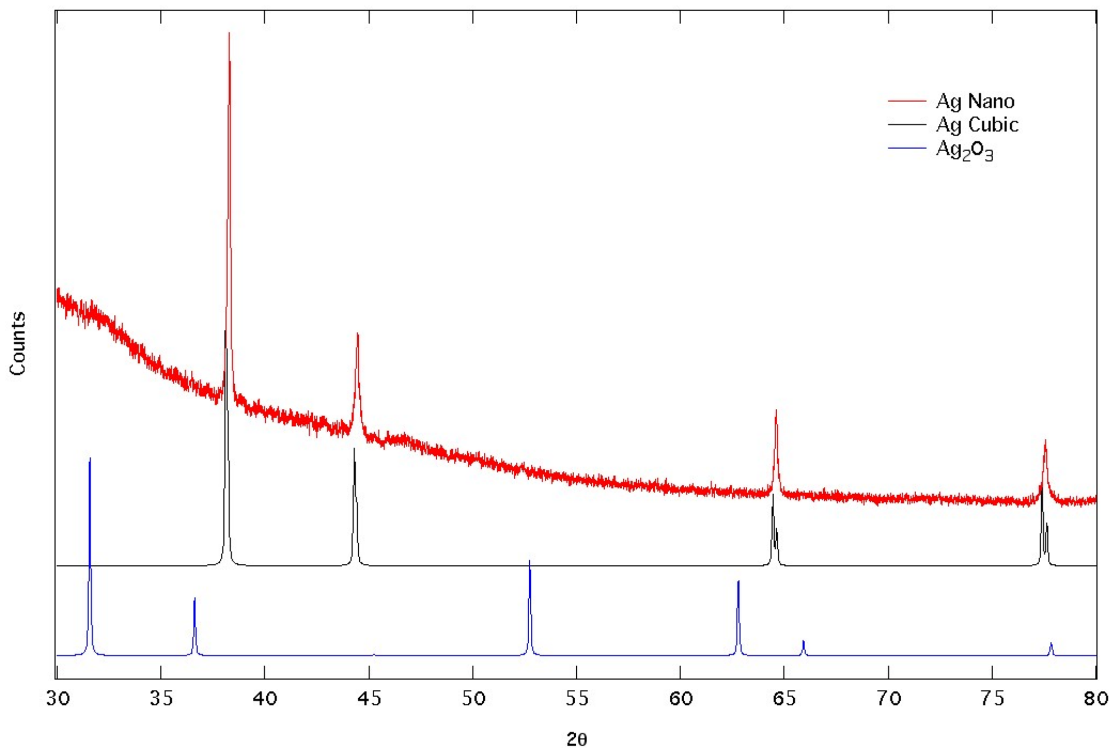
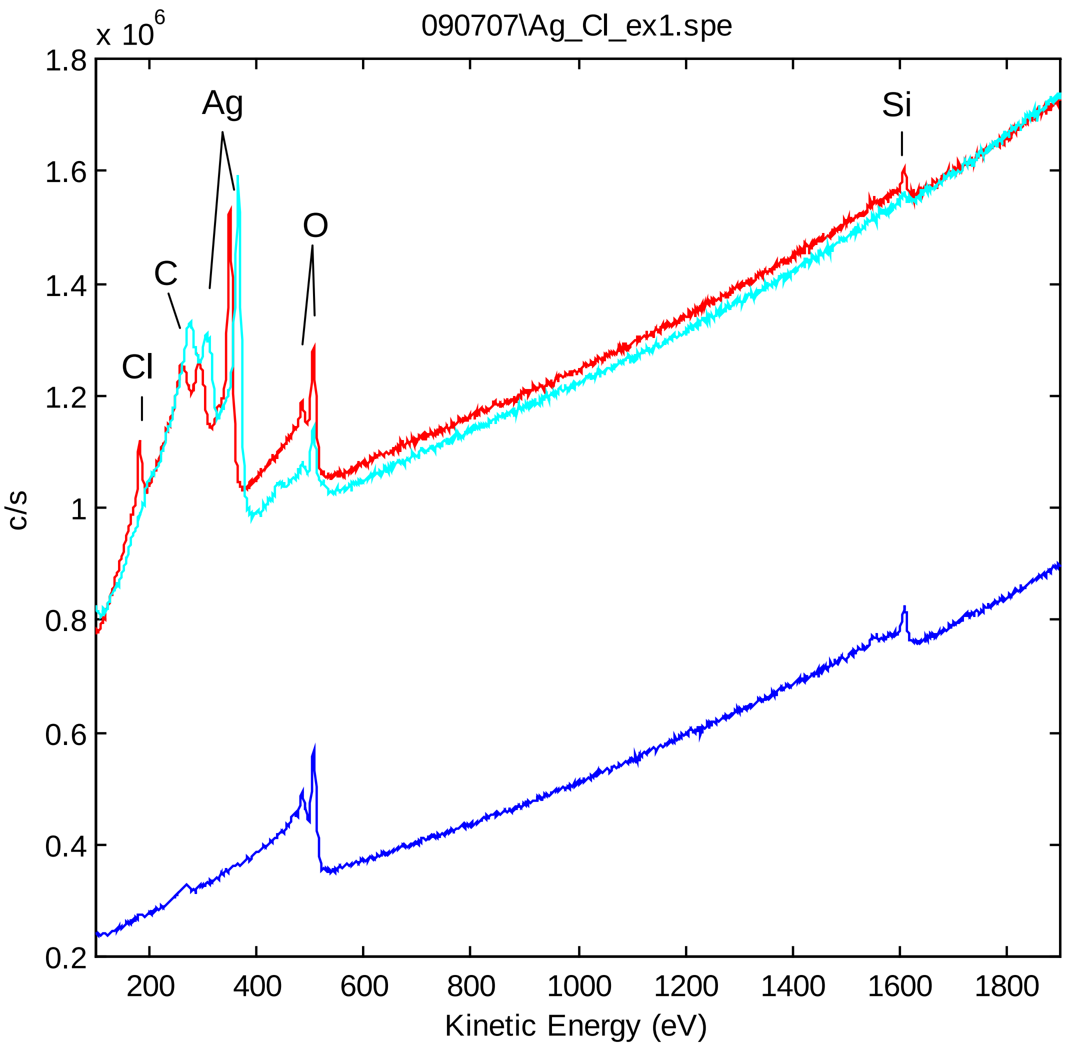
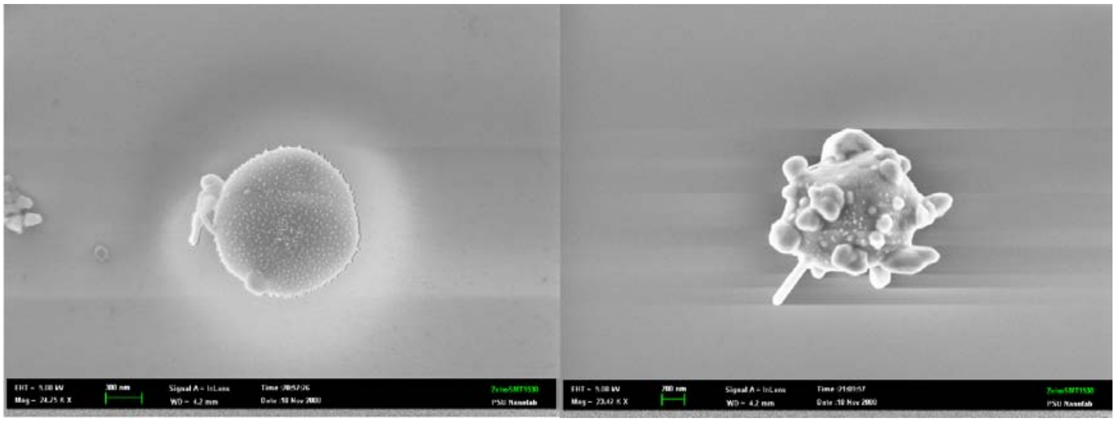


Supporting Figure 1: (Red) XPS spectra of the smooth, uncapped, single crystal silver nanoparticles synthesized for this publication. (Black) Reference spectra of cubic phase Ag. (Blue) Reference spectra of Ag_2O_3. Peaks for the particles synthesized for this publication match the spectra for cubic Ag.



Supporting Figure 2: Auger spectra of (Teal) the uncapped single crystal silver nanoparticles used in this publication, (Red) the nanoparticles after they had been exposed to 15% H_2O_2, UV light, and 0.5 M KCl for 7min, and then washed with deionized water several times, and (Blue) the signal from the SiO2 wafer.



Supporting Figure 3: (Left) AgCl colloid before UV illumination. Very small white dots on the colloid are FESEM charging artifacts. (Right) AgCl colloid after 2 hours of UV illumination. Large silver islands are seen obscuring the colloid's surface. Images reproduced from Reference 24.

Supporting Video 1:

UV-illuminated silver chloride (AgCl) particles undergoing "stick-slip" motion in solution over a microscope slide. The solution contains 0.33% H_2O_2 in water. The video is 194 μm in width. Movie plays in real-time.

Supporting Video 2:

A solution containing AgCl particles (darker objects), silica tracer spheres (lighter objects), and 1% H_2O_2 in water. The particles are illumined with UV light over a microscope slide. The AgCl particles are seen to move through solution, and alternately bind and release the silica tracer particles. The video is 98 μm in width. Movie plays in real-time.

Supporting Video 3

A solution containing single crystal silver particles (darker objects), silica tracer spheres (lighter objects), 0.25 mM KCl, and 0.63% H_2O_2 in water. The particles are illuminated with UV light over a microscope slide. The silver particles are seen to move through solution, and alternately bind and release the silica tracer particles (qualitatively similar to Video 2). The video is 98 μm in width. Movie plays in real-time.

Supporting Video 4

A solution containing AgCl particles and 1% H_2O_2 in water under UV illumination. The AgCl particles alternately attract then repel one another. Traveling waves of particle motion can be seen traversing the illuminated area. Synchronization of particle attractions/repulsions is also observed over a variety of length scales. The video is 194 μm in width. Movie plays in real-time.

Prior to this movie being recorded and prior to the particles' exposure to H_2O_2, the particles were placed in pure deionized water and illuminated with UV light for 25 min. H_2O_2 was then added to the system to reach a final concentration of 1%. This movie was recorded approximately 5 minutes after the addition of H_2O_2.

Supporting Video 5

A UV-illuminated aqueous solution containing 2.3 μm silica tracer particles (lighter objects), silver chloride colloids (darker objects), and 1% (v/V) H_2O_2. A reaction wave moves in from the upper left and is joined by a second wave from the upper right. After the front passes, the tracer particles are clumped. The field of view is 485 μm wide. Movie plays in real time.

Supporting Video 6

A UV-illuminated aqueous solution containing 2.3 μm silica tracer particles, 0.33 mM KCl, and 0.17% H_2O_2 is imaged above a 25 μm diameter silver disk that has been patterned onto a SiO_2 surface. The silica tracer particles are alternately attracted to and repelled from the silver surface. The silver disk is discolored by deposits of AgCl which have formed in situ. One such deposit resembles a long line that traverses the entire disk. Such spatially defined deposits are common in this system. Movie plays in real-time

Supporting Video 7

A UV-illuminated aqueous solution containing 2.3 μm silica tracer particles, 0.33 mM HCl, and 0.17% H_2O_2 is imaged above an array of 9 μm diameter silver disks with 11 μm spacings. As a traveling wave of tracer particle motion passes the array, the disks appear to flash on and off as their color alternates between reflective silver and darkened AgCl.

The movie has two distinct segments. The first segment is a real-time recording of the phenomenon taken at 30 frames per second. The second segment shows the movie slowed down to 1/10th speed (3 fps). Because of the relatively slow frame rate of the camera used verses the speed at which this remarkably fast phenomenon occurs, it is possible that some of the color variations of the surface features were not captured on film because they occurred between the frames.

Supporting Video 8

A UV-illuminated aqueous solution containing 2.3 μm silica tracer particles, 0.17 mM HCl, and 0.17% H_2O_2 is imaged above a 90 μm diameter silver disk that has been patterned onto a SiO_2 surface. Connected to the silver disk is a silver wire that leads out along the surface to a silver contact pad located outside the liquid. The silver wire is coated with an insulating Su-8 polymer to isolate it from solution. Movie plays in real time

The silver disk is discolored by deposits of AgCl which have formed in situ. The silica tracer particles are alternately attracted to and repelled from the silver surface. As this motion occurs, the current is measured between the silver disk working electrode and a gold wire counter electrode placed in the above solution. This current is plotted below the video. Negative currents are oxidative with respect to the silver disk. Attractive motions of the tracer particles accompany oxidative current spikes.