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Visualizing Au-Au bond formation in solution with femtosecond X-ray scattering

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Bond formation is an essential process in chemical reactions, but it is challenging to keep track of detailed atomic movements associated with bond formation because of its bimolecular nature. Bond formation in solution phase has been especially elusive because it is difficult to initiate and follow such diffusion-limited bimolecular processes with ultrafast time resolution. In this regard, a Au oligomer complex, $[Au(CN)_2^-]_n$, offers a good model system in which to study the dynamics of bond formation in solution [,2].Using femtosecond time-resolved X-ray scattering, we successfully visualized in real time the birth of a gold trimer complex, $[Au(CN)_2^-]_3$, that occurs via photoinduced formation of Au-Au covalent bonds [3]. The ground state of the trimer has Au atoms that are weakly bound to each other by aurophilic interaction and aligned in a bent geometry. Upon photoexcitation, the ground state rapidly converts into the first excited state where Au-Au covalent bonds are formed among Au atoms aligned in a linear geometry. Subsequently, the state transforms to a triplet state in 1.6 ps while accompanying further contraction of Au-Au bonds by 0.1 Å. Later, the triplet state of the trimer converts to a tetramer on nanosecond time scale. This work showcases the possibility of tracking detailed structural changes in solution with sub-ps temporal and sub-angstrom spatial resolutions, thanks to the advent of X-ray free electron lasers and the advance of data analysis of time-resolved solution scattering data.

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