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Evolution from the Plasmon to Exciton State in Atomically Precise Gold Nanoparticles MENG ZHOU, CHENJIE ZENG, YUXIANG CHEN, SHUO ZHAO, Department of Chemistry, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213, USA, MATTHEW SFEIR, Center for Functional Nanomaterials, Brookhaven National Laboratory, Upton, New York 11973, USA, MANZHOU ZHU, Department of Chemistry and Center for Atomic Engineering of Advanced Materials, Anhui University, Hefei, Anhui 230601, P. R. China, RONGCHAO JIN, Department of Chemistry, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213, USA — The evolution from the metallic (or plasmonic) to molecular state in metal nanoparticles constitutes a central question in nanoscience research because of its importance in revealing the origin of metallic bonding and offering fundamental insights into the birth of surface plasmon resonance. Here, we utilize the atomically precise gold nanoparticles protected by thiolate ranging in size from 1 nm to 3.5 nm (including Au₂₅, Au₃₈, Au₁₄₄, Au₃₃₃, Au_{~520}, Au_{~940}) and investigate the grand transition from metallic to molecular state by femtosecond transient absorption spectroscopy, as well as the impact of the transition on catalytic reactions. By directly probing the electron-phonon coupling of the gold nanoparticles, we have mapped out that the transition occurs between 2.3 nm (Au₃₃₃) and 1.7 nm (Au₁₄₄). This study paves the way for future exploitation of the grand transition and its impact on the physicochemical properties of metal nanoparticles, in particular the applications in energy transfer and utilization.

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