Realtime tracking of plasmon dephasing in a sub-nm system

Content

Research on nanoplasmonics has enabled multidisciplinary applications in the fields of solar energy harvesting, ultrafine sensor technology, controlled photocatalysis, etc. [1-3]. While the plasmonic response of particles with diameters exceeding 10 nm is generally well characterized by classical Mie-type models, the collective dynamics below this size limit acquires a predominantly quantum character that is only poorly understood. In particular, the exact origin of plasmon dephasing in this regime is still unknown.

Here, we report on the time-resolved study of the plasmonic dynamics for the sub-nm fullerene C60 [4]. In this system, the coherent dynamics among π and σ band states resulting from photoexcitation give rise to an ultrabroad giant plasmon resonance (GPR) in the continuum, with excitation energy around 20 eV (ionisation potential IP = 7.6 eV) and a bandwidth exceeding 10 eV5. By employing attosecond streaking spectroscopy, we measured the photoelectron emission delay around the GPR, while ab-initio linear-response time-dependent density functional theory (LR-TDDFT) and classical trajectory Monte-Carlo (CTMC) calculations were used to simulate the plasmonic response of C60.

By comparing experimental data and computations, we demonstrate that the GPR dephasing time maps directly to the energy-dependent intrinsic time delay of the electron photoemission. In particular, we identified the contribution originating exclusively from the large-scale correlation-induced collective excitation of the GPR to be the fastest route of dephasing, with a characteristic time between 50 and 200 as (See Fig.1). Our study sets a benchmark for a deeper understanding of the dynamics in nano- or sub-nanomaterials. Given the importance of plasmonics in many fields of science, the study can inspire investigations of rapid decoherence and control of plasmon phenomena on their natural time scale. This knowledge can be particularly important for the development of novel technology involving quantum plasmonics.

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