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Attosecond time delays in C_{60} valence photoemissions at the giant plasmon

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Synopsis We perform time-dependent local density functional calculations of the time delay in C_{60} HOMO and HOMO-1 photoionization at giant plasmon energies. A semiclassical model is used to develop further insights.

Oscillations of the correlated electron density in nanosize systems stimulated by external radiation can quantize, forming many-body quasi-particle, the plasmon. For nanoclusters, the direct consequence of the plasmon is the formation of a giant resonance in the response spectra that holds promise in optical applications. For the C_{60} molecule, a giant dipole plasmon can be ionized by XUV photons around 20 eV which therefore can serve as a spectral laboratory to study the effects of the collective motions on the emission time delay.

We compute the phases of HOMO and HOMO-1 photoamplitudes of C_{60} , using the time-dependent local density approximation (TDLDA) [1] that includes many-particle correlation effects. Previous calculations [2] were improved by including a scheme that angle-integrates the dipole amplitudes to calculate the total phase, whose energy-derivatives gave the Wigner-Smith time delays of emission. A semiclassical description is utilized to interpret some aspects of the result.

Numerical results for HOMO and HOMO-1 delay are presented in Figure 1(a). The comparison with single-particle LDA predictions, that omit correlations, clearly reveals the role of the plasmon to affect the delay. A roughly opposite delay pattern between the growing versus waning part of the plasmon resonance (peaked at 20 eV) is noted.

The complex interaction between the 240 valence electrons of C_{60} and the incident photon can be mimicked without solving the full time-dependent many-body problem, by introducing

an effective Hamiltonian. Based on a semiclassical model, we show that the plasmon acts as a screening effect that modifies the phase of the electron wavepacket. It induces a local variation of the angular distribution which is accompanied by a variation of the photoemission delays on the attosecond timescale.

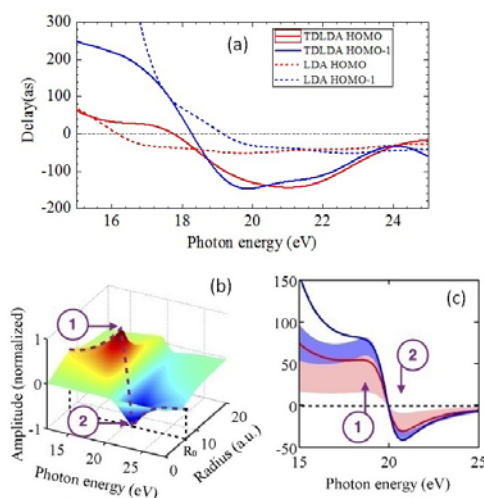


Figure 1. (a) TDLDA and LDA delays for HOMO and HOMO-1 emissions. (b) Real part of the screening potential. (c) Delays from semi-classical model.

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References

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