# Correlation drives a strong attractive force on plasmonic photoelectrons

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**M Magrakvelidze[1], M E Madjet[2 *] and H S Chakraborty[3 †]**

[1]Department of Physics, University of Mary Washington, Fredericksburg, Virginia 22401, USA
[2]Qatar Environment and Energy Research Institute, Hamad Bin Khalifa University, Qatar Foundation, P.O. Box 5825, Doha, Qatar
[3]Department of Natural Sciences, D L Hubbard Center for Innovation, Northwest Missouri State University, Maryville, Missouri 64468, USA

**Synopsis** We show that the dynamical electron correlation suppresses the dipole field of incoming photon while inducing a strong attractive force at the spectral region of the giant plasmon resonance of $C_{60}$. A signature of this force is seen in the form of a binding well in the imaginary part of the induced potential. This attraction can significantly influence the emerging photoelectron's temporal delay behavior.

Fullerene molecules are a highly stable form of nanoscopic carbon allotrope that can exist at room temperature. Thus, they are routinely attractive for spectroscopic studies, including current interests in probing their photoresponse in real time [1]. Of particular interest is the spectral zone around 20 eV where a collective excitation occurs that decays *via* the electron continuum producing a giant plasmon resonance in the photoelectron intensity [2]. Theoretical studies of the electrons' collective motions at this resonance can thus be invaluable in developing deeper insights, particularly in understanding attosecond photoemission measurements in either RABITT or streaking spectrometry.

In the present study, a jellium potential representing 60 $C^{4+}$ is constructed by smearing the total charge over a spherical shell of proper radius and thickness. The Kohn-Sham equations for a system of 240 electrons, made up of four valence electrons from each C atom, are solved to obtain the ground state structure in local density approximation [3]. Considering the dipole response of the system to the incoming photon, the energy dependent induced electron-density is then computed from the many body susceptibility in a linear response frame. This subsequently yields the complex radial induced potential $V_{\rm ind}(R,E)$ which, at plasmon energies, probes the collective response of the molecule.

At energies below/above the resonance peak Re[$V_{\rm ind}$] screens/anti-screens the external potential over the shell region of maximum wavefunction overlaps. The switch between these modes occures roughly at the plasmon peak where Re[$V_{\rm ind}$] is closely zero. In contrast, Im[$V_{\rm ind}$] in Figure 1 exhibits a deep minimum at the resonance peak, while falling off moving either side from the peak. This shape of Im[$V_{\rm ind}$] indicates a transient attractive force on electrons which may cause their time-delayed emergence.

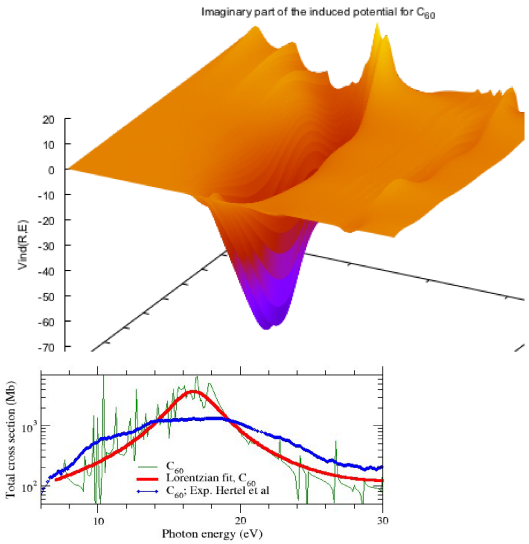


**Figure 1.** (Top) Im[$V_{\rm ind}$] as a function of $C_{60}$ radius and photon energy. (Bottom) Calculated [3], Lorentzian fit, and measured [2] (red-shifted) ionization cross sections.

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## References

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* E-mail: mmadjet@hbku.edu.qa
† E-mail: himadri@nwmissouri.edu