The Role of Super-Atom Molecular Orbitals in Doped Fullerenes in a Femtosecond Intense Laser Field

N. Berrah [#1], H. Xiong [#], B. Mignolet[^], L. Fang[&], T. Osipov[*], T. J. A. Wolf [&], E. Sistrunk[&], M. Gühr [&%], and F. Remacle[^]

[#]University of Connecticut, Physics Department, Storrs, CT 06269, USA
[^]Departement de Chimie, B6c, Université de Liege, B4000 Liege, Belgium
[&]Center for High Energy Density Science, University of Texas, Austin, TX 78712, USA
[*]LCLS, National Accelerator Laboratory, Menlo Park, CA 94025, USA
[&]Stanford PULSE Institute, SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA.
[%]Institut für Physik und Astronomie, Universität Potsdam, 14476 Potsdam, Germany

Synopsis The photoionization of complex targets, such as endohedral fullerenes, in strong laser fields has been studied experimentally and theoretically. We used the prototype $Ho_3N@C_{80}$ interacting with intense ($0.1\text{-}5\times10^{14}$ W/cm^2), short (30 fs), 800 nm laser pulses giving rise to multiply charged parent ions. The power law dependence of $Ho_3N@C_{80}^{q+}$, q=1-2, was found to be different from that of C_{60}. Time-dependent density functional theory computations suggest that the fast ionization of the SAMO states in $Ho_3N@C_{80}$ is responsible for the different power law behavior.

The investigations of the behavior of matter in intense laser fields were extended to larger molecules, such as C_{60} [1]. We increased further the complexity by investigating the photoionization of complex targets, such as endohedral fullerenes in strong laser fields. We used the prototype, $Ho_3N@C_{80}$, interacting with intense ($0.1\text{-}5\times10^{14}$ W/cm^2), short (30 fs), 800 nm laser pulses giving rise to multiply charged parent ions as well as fragment ions.

We will report on the measurement and theoretical explanation of the power law for singly ionized $Ho_3N@C_{80}$ yields, and we will compare our findings to C_{60} results carried out under similar conditions [1]. In our work, we distinguish two regions in the ion yield spectra with respect to field strength corresponding to different ionization mechanisms and power laws. For low field strengths, multiphoton ionization dominates, while for higher field strengths, tunneling and ionization over the barrier are the main ionization processes. The power law dependence of $Ho_3N@C_{80}^{q+}$, q=1-2, was found to be different from that of C_{60}. Our time-dependent density functional theory computations revealed different light-induced ionization mechanisms. Unlike in C_{60}, in doped fullerenes, the breaking of the cage spherical symmetry makes super atomic molecular orbital (SAMO) states optically active. Theoretical calculations suggest that the fast ionization of the SAMO states in $Ho_3N@C_{80}$ is responsible for the measured n=3 power law for $Ho_3N@C_{80}^+$ at intensities lower than 1.2×10^{14} W/cm^2. The measured C_{60}^+ power law was found to be n=5 [1]. For C_{60}, both super atomic molecular orbitals (SAMO) and Rydberg states played important roles when exposed to a strong laser field [2]. Both kinds of states can be indirectly populated by vibronic coupling, but because the density of Rydberg states is higher than that of SAMO states, Rydberg state ionization in C_{60} dominates the ionization at laser intensities $>10^{13}$ W/cm^2 [2]. Our theory demonstrated that the SAMO states play a more important role in doped fullerenes than in C_{60}. Endohedral fullerenes respond differently to intense near-infrared femtosecond laser fields compared to C_{60}, and this is reflected by the measured and calculated different slope of the power law for low field strengths.

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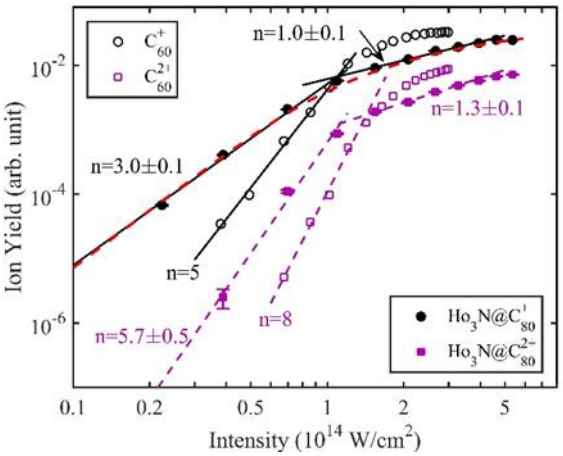

Figure 1. Comparison of the ion yield dependence on peak intensity for the ionization of $Ho_3N@C_{80}^{q+}$ (q=1, 2) (filled symbols) and C_{60}^{q+} (q=1, 2) (open symbols) [1].

References

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[1]E-mail: nora.berrah@uconn.edu