Comment on "Correlation Quantum Dynamics between an Electron and D_2^+ Molecule with Attosecond Resolution"

In a recent Letter, Hu *et al.* [1] used the time-dependent–wave-packet method to calculate the kinetic energy distribution of the D^+ ions of D_2 molecules by intense femtosecond laser pulses. Their theoretical results are in surprisingly good agreement with the experimental data reported by Niikura *et al.* [2]. Their conclusion, that the D^+ ions came from the recollision induced dissociation between the parent ion D_2^+ and the rescattering electrons in their *first* return, is in agreement with the model of Niikura *et al.*. This conclusion, however, has been previously questioned by Tong *et al.* [3]. The latter concluded that the D^+ ions of the same experiment came from Coulomb explosion of two D^+ ions, due to impact excitation by the rescattering electrons in their *third* return, followed by further laser induced ionization. The disagreement in the interpretation is significant since it amounts to distinct reading of the molecular clocks of a few femtoseconds, even though the clocks can be read with attosecond resolution [2,3].

When a D_2 molecule is placed in a laser field, it is first ionized into the ground state of D_2^+ in the early part of the pulse. The issue under discussion is the relative importance of dissociation vs ionization induced by the rescattering electrons in the subsequent production of D^+ ions. To avoid D^+ ions from other mechanisms, Niikura *et al.* measured D^+ ions perpendicular to the laser polarization and by focusing only on high-energy D^+ ions. Experimentally, ionization can be distinguished from dissociation by detecting two D^+ ions in coincidence, which have been carried out by Alnaser *et al.* [4,5] for a range of laser intensities and pulse lengths. These experiments, which have been shown in good agreement with the modeling of Tong *et al.* [3,6], were not mentioned in Hu *et al.*. When the same theoretical model of Tong *et al.* was used to simulate the noncoincidence experiments of Niikura *et al.*, it was found that D^+ ions were generated mostly from rescattering occurring at the third return, in disagreement with the model of Niikura *et al.* and the new theoretical results from Hu *et al.*.

Since rescattering involves electron impact excitation of the molecular ion, proper theoretical treatment of this process is essential. In Tong *et al.*, electron impact excitation cross sections were carefully modeled, including the magnetic-substate (or alignment) dependence. For laser polarization perpendicular to the molecular axis, it was found that excitation by the rescattering electrons to the π_u state of D_2^+ is much more important than for excitation to the σ_u state. In their wave-packet propagation method, Hu *et al.* made two critical assumptions: (1) the laser polarization direction is irrelevant; (2) the effect of electron-electron interaction can be approximated by a drag term. Under approximation (1), their electronic wave function retains cylindrical symmetry. This in effect implies that the laser field was chosen to be along the molecular axis. Although they projected out the electron wave packet perpendicular to the molecular axis, that contribution is a high order effect due to Coulomb focusing. For their approximation (2), it has not yet been proved that such a drag term can describe electron impact excitation processes.

Since all theoretical calculations involve some form of approximations, to resolve the apparent discrepancy in the reading of the molecular clock from the experiment of Niikura *et al.*, it is desirable that coincidence measurements be carried out for the parameters in that paper. Meanwhile, it is desirable that Hu *et al.* address the coincidence experiments of Alnaser *et al.* [4,5]. Until these have been done, there is no proof that the wave-packet propagation calculation of Hu *et al.* supports the original interpretation of the molecular clock of Niikura *et al.*, which had previously been questioned in Tong *et al.* [3].

X. M. Tong,[1] Z. X. Zhao,[2] and C. D. Lin[1]
[1]J. R. Macdonald Laboratory
Physics Department
Kansas State University
Manhattan, Kansas 66506-2604, USA
[2]Center for Photonics Research
University of Ottawa
Ottawa, K1N 6N5, Canada

Received 5 October 2005; published 26 July 2006
DOI: 10.1103/PhysRevLett.97.049301
PACS numbers: 31.15.Qg, 33.15.Vb, 33.80.Wz, 34.80.Qb

[1] J. Hu, K.-L. Han, and G.-Z. He, Phys. Rev. Lett. **95**, 123001 (2005).
[2] H. Niikura *et al.*, Nature (London) **421**, 826 (2003).
[3] X. M. Tong, Z. X. Zhao, and C. D. Lin, Phys. Rev. Lett. **91**, 233203 (2003).
[4] A. S. Alnaser *et al.*, Phys. Rev. Lett. **91**, 163002 (2003).
[5] A. S. Alnaser *et al.*, Phys. Rev. Lett. **93**, 183202 (2004).
[6] X. M. Tong, Z. X. Zhao, and C. D. Lin, Phys. Rev. A **68**, 043412 (2003).

