Comment on “Correlation Quantum Dynamics between an Electron and D2 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\textsuperscript{+} ions of D\textsubscript{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\textsuperscript{+} ions came from the rescollison induced dissociation between the parent ion D\textsubscript{2}\textsuperscript{+} 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\textsuperscript{+} ions of the same experiment came from Coulomb explosion of two D\textsuperscript{+} 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\textsubscript{2} molecule is placed in a laser field, it is first ionized into the ground state of D\textsubscript{2}\textsuperscript{+} 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\textsuperscript{+} ions. To avoid D\textsuperscript{+} ions from other mechanisms, Niikura et al. measured D\textsuperscript{+} ions perpendicular to the laser polarization and by focusing only on high-energy D\textsuperscript{+} ions. Experimentally, ionization can be distinguished from dissociation by detecting two D\textsuperscript{+} 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].

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 \( \pi_u \) state of D\textsubscript{2}\textsuperscript{+} is much more important than for excitation to the \( \sigma_u \) 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].

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