

The importance of Rydberg orbitals in dissociative ionization of small hydrocarbon molecules in intense few-cycle laser pulses

This content has been downloaded from IOPscience. Please scroll down to see the full text.

2015 J. Phys.: Conf. Ser. 635 112043

(<http://iopscience.iop.org/1742-6596/635/11/112043>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 129.130.37.147

This content was downloaded on 24/03/2016 at 14:58

Please note that [terms and conditions apply](#).

The importance of Rydberg orbitals in dissociative ionization of small hydrocarbon molecules in intense few-cycle laser pulses

Bethany Jochim^{*1}, R. Siemering[†], M. Zohrabi^{*}, A. Voznyuk^{**}, J. B. Mahowald^{**}, D. G. Schmitz^{**}, K. J. Betsch^{*}, Ben Berry^{*}, T. Severt^{*}, Nora G. Kling^{*‡}, T. G. Burwitz^{**}, U. Ablikim^{*}, K. D. Carnes^{*}, M. F. Kling^{*‡}, I. Ben-Itzhak^{*}, E. Wells^{**2}, R. de Vivie-Riedle[†]

^{*}J.R. Macdonald Laboratory, Department of Physics, Kansas State University, Manhattan, Kansas 66506 USA

[†]Department für Chemie, Ludwig-Maximilians-Universität München, D-81377 München, Germany

^{**}Department of Physics, Augustana College, Sioux Falls, South Dakota 57197 USA

[‡]Department of Physics, Ludwig-Maximilians-Universität München, D-85748 Garching, Germany

Synopsis We demonstrate the importance of ionization from Rydberg orbitals via experimental and theoretical work focusing on the strong-field dissociative single ionization of small hydrocarbons. Our findings suggest that Rydberg states should be routinely considered when studying polyatomic molecules in intense laser fields.

Much of our intuition about strong-field processes is built upon studies of diatomic molecules, which have electronic states that are typically well separated in energy. In polyatomic molecules, however, the electronic states are closer together, leading to the involvement of more states. We studied a specific process initiated by intense few-cycle laser pulses in C_2D_2 , C_2D_4 , and C_2D_6 , namely single ionization of the parent molecule followed by hydrogen elimination. We measure the momentum of the remaining $C_2D_{n-1}^+$ fragment in each case using velocity map imaging (VMI). Our measurements, along with angle-dependent ionization probability calculations [1], demonstrate that Rydberg orbitals are readily populated for certain orientations of the molecules relative to the laser polarization. Moreover, these commonly-neglected Rydberg orbitals make a key contribution to strong-field ionization.

As an example, the comparison between measured and calculated $C_2D_3^+$ angular distributions from C_2D_4 , shown in Fig. 1, clearly illustrates the need to include contributions from Rydberg states. The general four-lobed structure of $C_2D_3^+$ from dissociative single ionization is independent of pulse intensity and duration (at least up to 45 fs [2]). Calculations that do not include ionization from Rydberg orbitals result in an approximately isotropic angular distribution, as illustrated by Fig. 1(c), while including contributions from Rydberg orbitals yields the four-lobed structure shown in Fig. 1(d), which qualitatively matches the experimental results. This improved understanding of the links between photofragment images and the electronic dynamics in the molecule can improve endeavors towards image-

based strong-field coherent control.

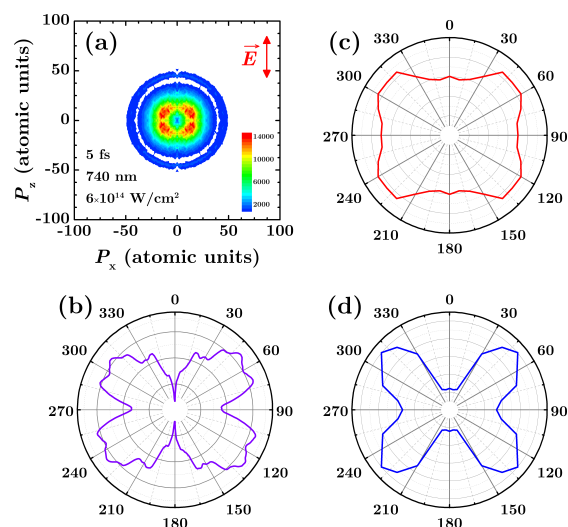


Figure 1. (a) Momentum distribution of $C_2D_3^+$. The inner lobes come from dissociative single ionization of C_2D_4 , and the weaker outer ring is due to dissociative double ionization. (b) Measured yield of the inner lobes as a function of the angle between the $C_2D_3^+$ and the laser polarization. Calculated angular distribution for $C_2D_3^+$ (c) without including Rydberg orbitals and (d) with ionization from Rydberg orbitals included.

JRML is supported by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. Augustana College is supported by NSF, EPSCoR, and the South Dakota Space Grant. BJ is also supported by DOE-SCGF. MFK, R dV-R, and RS are supported by DFG via the Cluster of Excellence: Munich Center for Advanced Photonics.

References

- [1] P. von den Hoff *et al* 2010 *Appl. Phys. B* **98** 659
- [2] E. Wells *et al* 2013 *Nat. Comm.* **4** 2895

¹E-mail: bjochim@phys.ksu.edu

²E-mail: eric.wells@augie.edu

