Precision calculation of intense-laser-field multiphoton ionization (MPI) rates of H_2^+ at critical internuclear distances

Son, Sang-Kil and Chu, Shih-I
Department of Chemistry, University of Kansas

Abstract

We extend the time-independent non-Hermitian Floquet formalism1) for high precision calculation of the MPI rates of H_2^+ at internuclear distances (R) from 2.0 to 20.0 a.u. in intense laser fields with intensity 1×10^{14} W/cm^2 and wavelength 791 nm. The procedure involves the use of the complex-scaling generalized pseudospectral (CSGPS) method for non-uniform spatial discretization of the Hamiltonian expressed in prolate spheroidal coordinates. We found that the MPI rates strongly depend upon R and are significantly enhanced at several critical distances in good agreement with the recent experimental results2).

Introduction

- Multiphoton ionization of H_2^+ in intense laser fields is enhanced at certain critical internuclear distances.
- Coulomb explosion occurs at critical internuclear distances after the ionization process.
- Recent experiment estimated critical distances of *R*~8, 11, and 15 a.u. from the kinetic energy spectra of fragments.2)
- Fully *ab initio* precision calculations of the real 3D H_2^+ system can provide detailed resonance structure and dynamical behavior in MPI processes.

Computational Method

The Hamiltonian of H_2^+ $$\hat{H}_0 = -\frac{1}{2}\nabla^2 - \frac{1}{|\mathbf{r} - \mathbf{R}_1|} - \frac{1}{|\mathbf{r} - \mathbf{R}_2|}$$

In prolate spheroidal coordinates (ξ, η, φ),

$$\hat{H}_0 = -\frac{1}{2a^2}\frac{1}{(\xi^2 - \eta^2)}\Big[\frac{\partial}{\partial\xi}(\xi^2 - 1)\frac{\partial}{\partial\xi} + \frac{\partial}{\partial\eta}(1 - \eta^2)\frac{\partial}{\partial\eta} + \frac{\xi^2 - \eta^2}{(\xi^2 - 1)(1 - \eta^2)}\frac{\partial^2}{\partial\varphi^2}\Big] - \frac{2\xi}{a(\xi^2 - \eta^2)}$$

$$\begin{cases} x = a\sqrt{(\xi^2 - 1)(1 - \eta^2)}\cos\varphi \\ y = a\sqrt{(\xi^2 - 1)(1 - \eta^2)}\sin\varphi \\ z = a\xi\eta \end{cases}$$

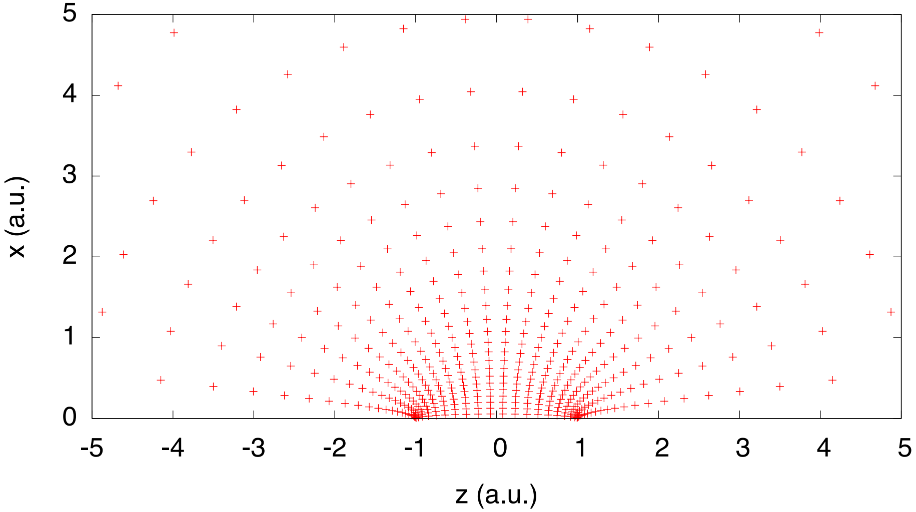
$$1 \leq \xi < \infty$$

$$-1 \leq \eta \leq 1$$

$$0 \leq \varphi \leq 2\pi$$

Non-uniform grid structure

Generalized Pseudospectral (GPS) method provides optimal non-uniform spatial distribution of grid structure.



Complex-scaling generalized pseudospectral method

$$\xi(x) = 1 + L\frac{1+x}{1-x}e^{i\alpha}, \qquad -1 \le x \le 1$$

$$\eta(y) = y, \qquad -1 \le y \le 1$$

ξ is complex-rotated by α. ξ and η are discretized by GPS method with Gauss-Legendre abscissas $\{x_i\}$ and $\{y_j\}$.

Time-independent non-Hermitian Floquet formalism[1)]

With lineraly polarized laser field of frequency ω and field strength F along the internuclear axis $\hat{z}$

$$\hat{H} = \hat{H}_0 + Fz\cos\omega t.$$

From the Floquet theorem, $\Psi(\mathbf{r},t) = e^{-i\varepsilon t}\sum_{m=-\infty}^{\infty}\Phi_m(\mathbf{r})e^{im\omega t}$

where ε: quasienergy, Φ_m: quasienergy-state Fourier component.

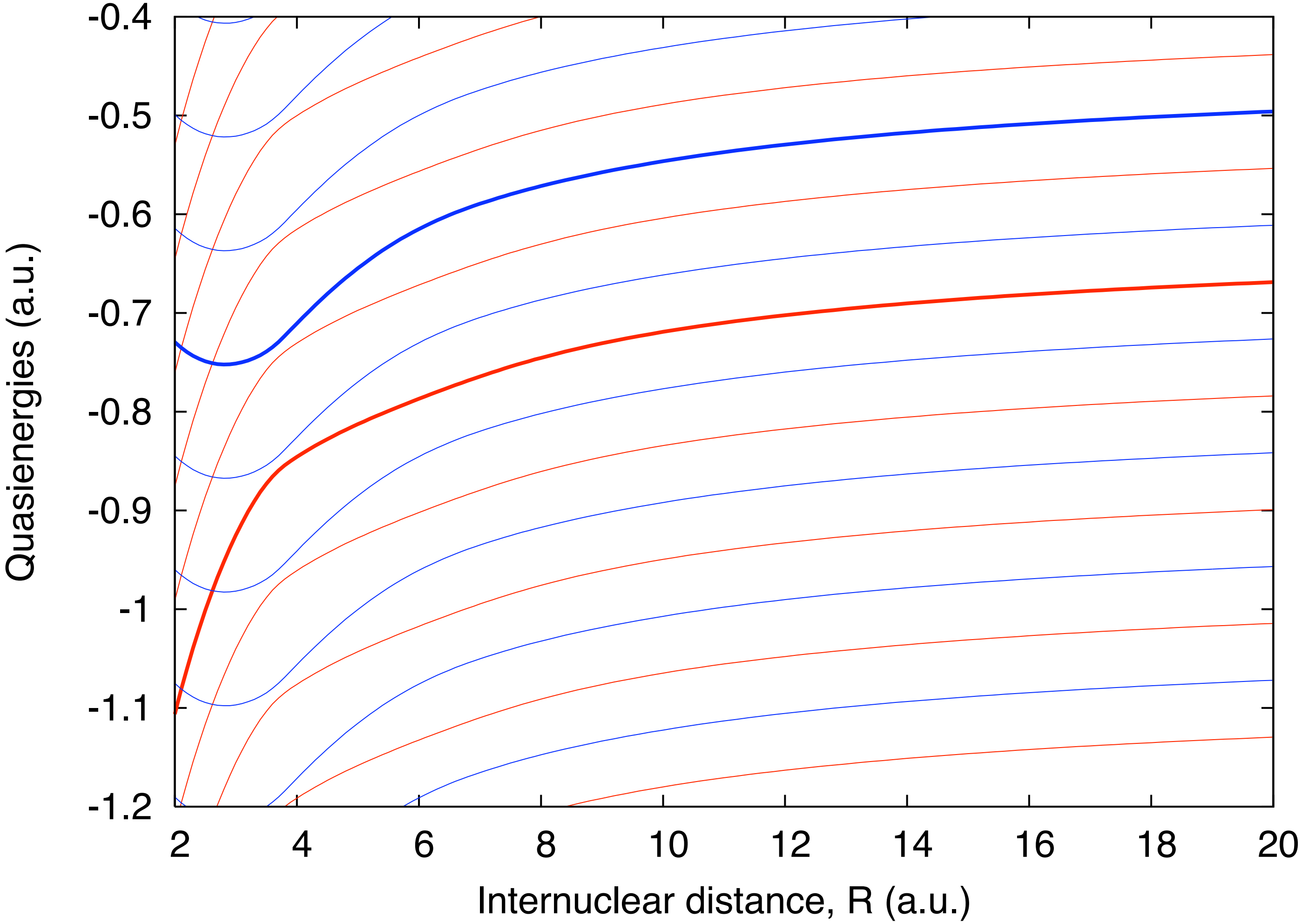
The equivalent time-independent Floquet Hamiltonian is given by

$$\left(\hat{H}_0 - m\omega\right)\Phi_m + \tfrac{1}{2}Fz\left[\Phi_{m-1} + \Phi_{m+1}\right] = \varepsilon\Phi_m.$$

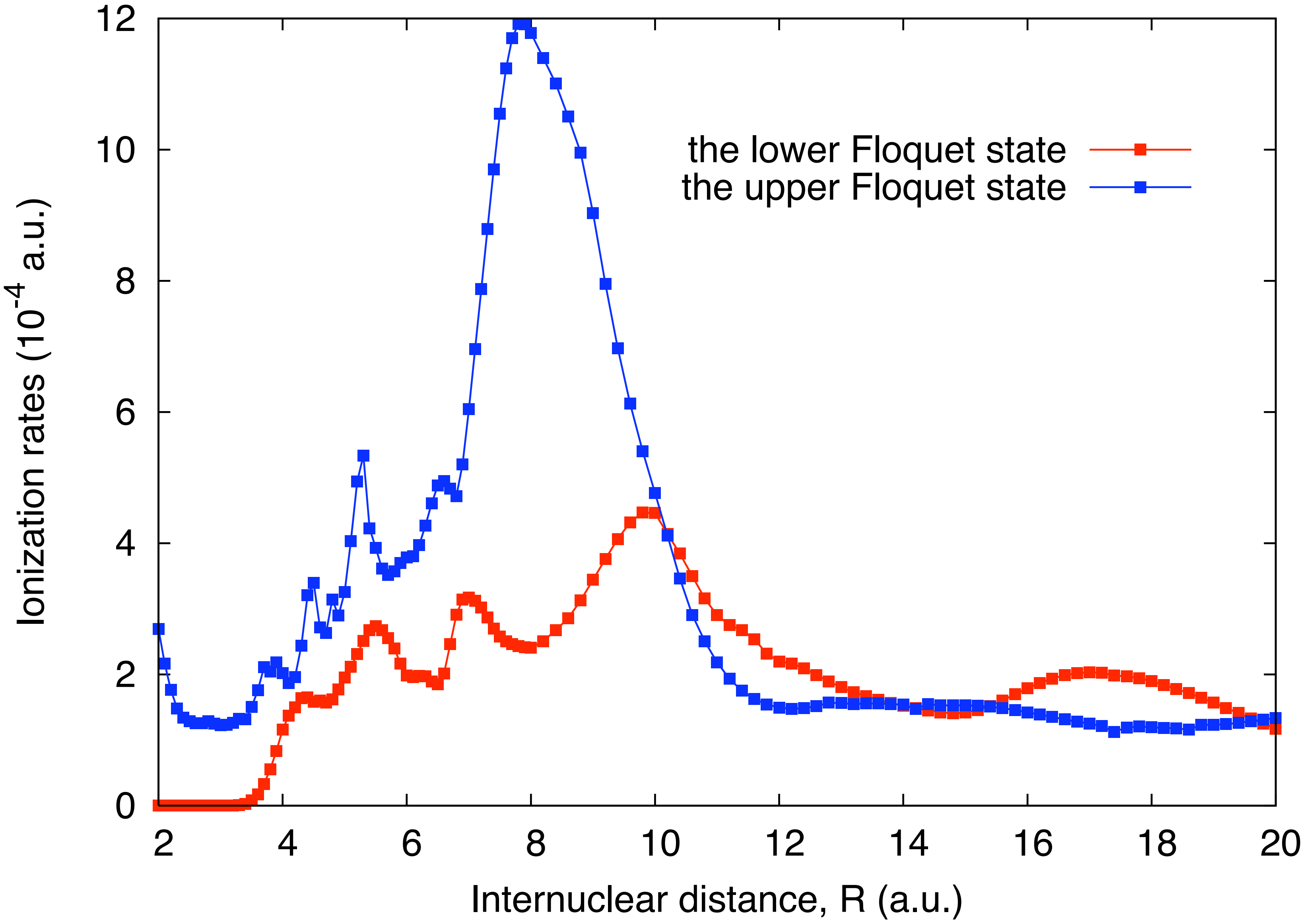
CSGPS applies to the time-independent non-Hermitian Floquet matrix to determine complex quasienergies.

Results

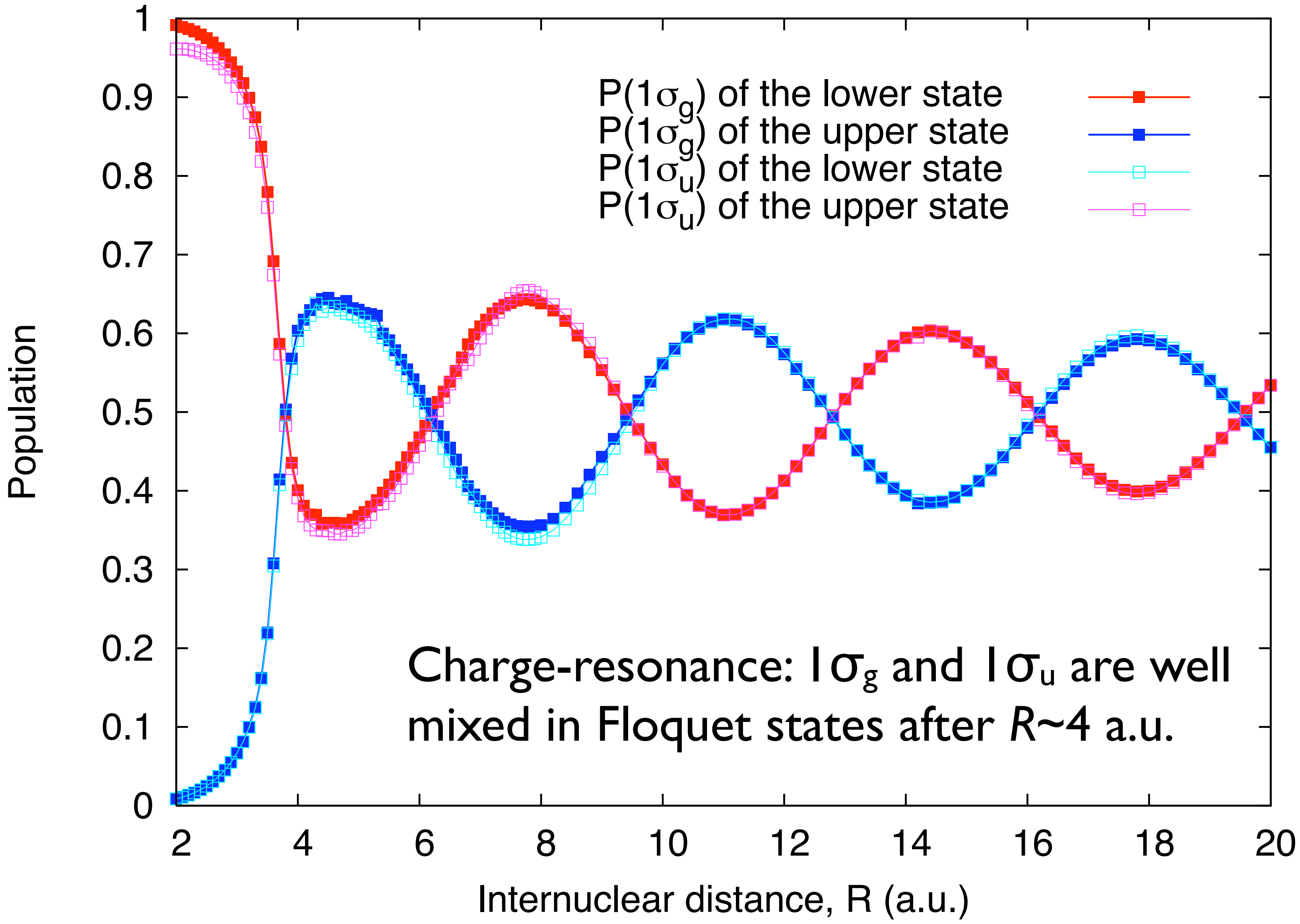
Real parts of quasienergies



R-dependent multiphoton ionization rates

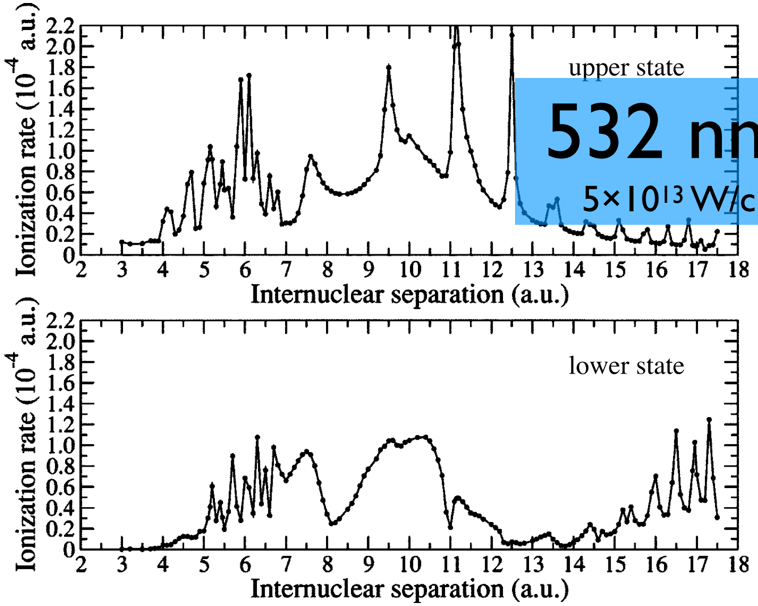


Population analysis on $1\sigma_g$ and $1\sigma_u$



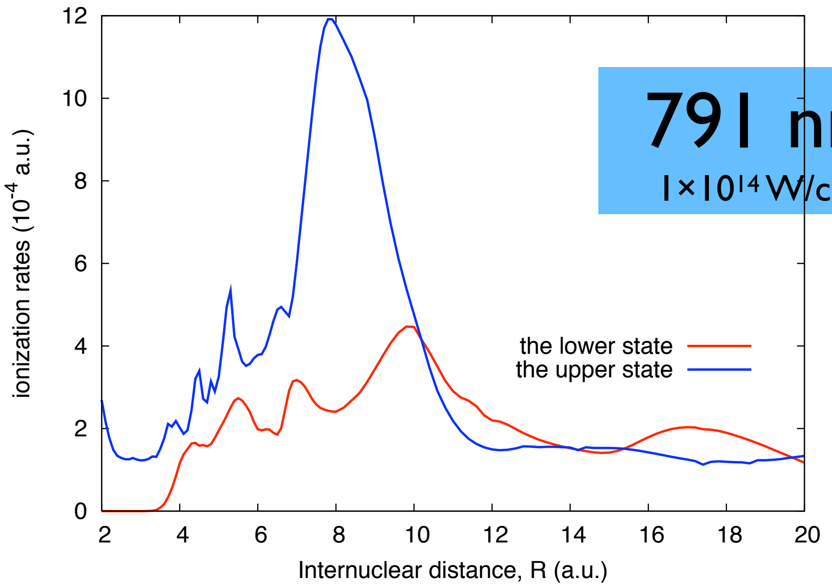
Charge-resonance: $1\sigma_g$ and $1\sigma_u$ are well mixed in Floquet states after R~4 a.u.

Dependence on wavelengths



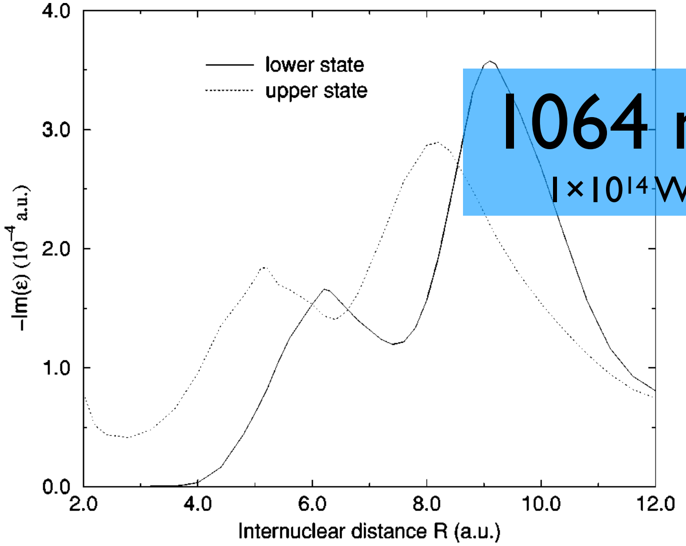
532 nm[3)]

5×10^{13} W/cm^2



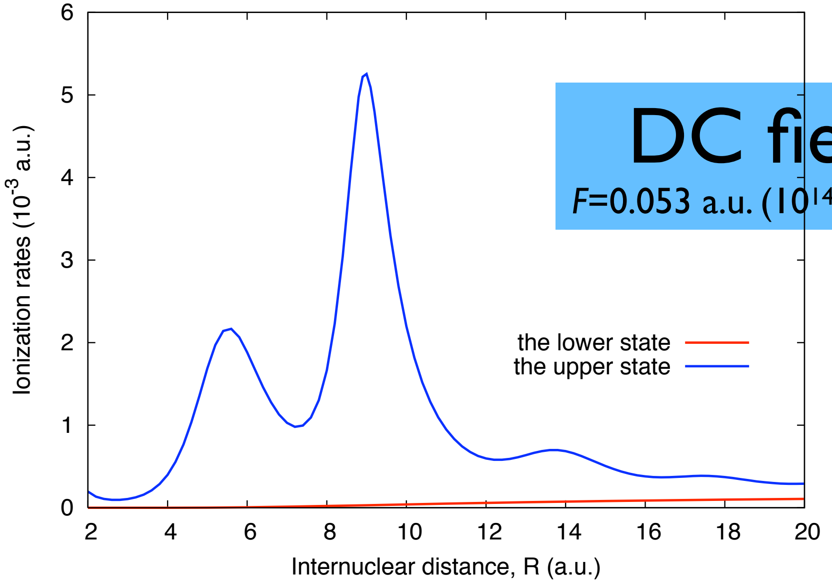
791 nm

1×10^{14} W/cm^2



1064 nm[4)]

1×10^{14} W/cm^2



DC field

F=0.053 a.u. (10^{14} W/cm^2)

Conclusion

- MPI rates of H_2^+ as a function of internuclear distance R are computed by time-independent non-Hermitian Floquet matrix discretized by CSGPS method.
- MPI rates strongly depend on R and are strongly enhanced at some critical distances.
- Rich resonance structures in MPI rates are found for short internuclear distances. Dependence of MPI rate peaks on wavelengths can be examined.
- To analyze the relation of vibrational levels and ionization processes, nuclear motions need to be included.

References

1. S. I. Chu and D. A. Telnov, *Phys. Rep.* **390**, 1 (2004).
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3. D. A. Telnov and S. I. Chu, *Phys. Rev. A* **71**, 013408 (2005).
4. X. Chu and S. I. Chu, *Phys. Rev. A* **63**, 013414 (2001).