Rabi oscillations in a stretching molecule

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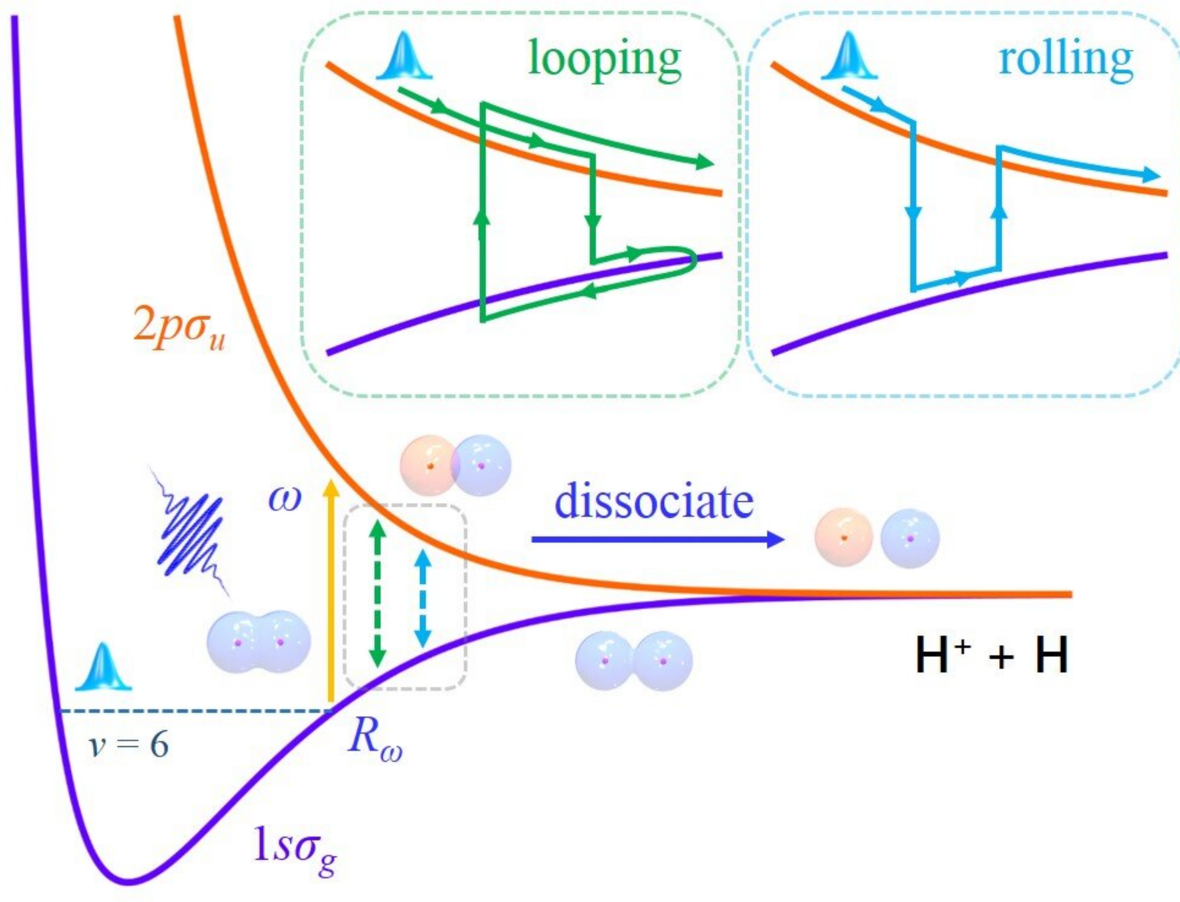

Violet and orange curves denote the potential energy curves of the electronic ground (1sσg) and excited (2pσu) states, respectively. The yellow vertical arrow between the two curves indicates the resonant one-photon transition at Rω. The two insets sketch the looping and rolling processes during the molecular dissociation, marked by the gray frame. The dashed horizontal line marks the eigenenergy of the ν = 6 vibrational state of the 1sσg state. Credit: Shengzhe Pan, Chenxi Hu, Wenbin Zhang, Zhaohan Zhang, Lianrong Zhou, Chenxu Lu,

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Over eighty years ago, Rabi oscillations were proposed to describe the strong coupling and population transfer in a two-level quantum system exposed to an oscillatory driving field. As compared to atoms, molecules have an extra degree of vibration, which adds an additional knob to the Rabi oscillations in light-molecule interactions. However, how such a laser-driven Rabi oscillation during the stretching of molecular bonds determines the kinetic energy release (KER) spectrum of dissociative fragments is still an open question.

In a new article published in *Light: Science & Applications*, a joint team of scientists, led by Professor Feng He from Shanghai Jiao Tong University and Professor Jian Wu from East China Normal University has investigated Rabi oscillations in a stretching molecule and discovered the strong-field-induced dissociation dynamics beyond the well-accepted resonant one-photon dissociation scenario. During the dissociation of the simplest molecular ion of H_2^+, coupled with the laser field, the electron hops between the $1s\sigma_g$ and $2p\sigma_u$ states, forming the Rabi oscillations.

The ionization-created nuclear wave packet (NWP) may propagate alternatively along the two potential energy curves towards a larger internuclear distance monotonically, termed as the rolling process, or may propagate outwards along the $2p\sigma_u$ curve followed by the inward propagation in the $1s\sigma_g$ curve and then be relaunched to $2p\sigma_u$ state again followed by subsequent dissociation, termed as the looping process. The rolling and looping dissociation pathways lead to different KERs of the ejected dissociative fragments, which have been verified by comparing experimental measurements with quantum simulation results.

In many fields where Rabi oscillations have been fully investigated, such as quantum optics and quantum dots, the external laser field is weak. While in the ultrafast and strong-field community, a strong femtosecond laser pulse is often utilized to induce the Rabi oscillations and even to observe the ultrafast dynamics of atoms and molecules in femtosecond time scales. These scientists summarize the Rabi oscillations in a stretching molecule:

"Our explanation based on the coupling of nuclear movement and electron Rabi oscillations is fundamentally different from the conventional one-photon bond-softening scenario. Using the conventional Floquet formalism, different dissociation pathways have been well recognized, and it is suggested that lower vibrational states can spill out the potential energy curves when a driving laser pulse becomes stronger. The Floquet formalism works well when the driving laser is a continuum wave, which unambiguously identifies the dissociation pathways from the energy point of view by paying for the loss of time information."

"However, we propose a fundamentally different scenario upon Rabi oscillations to explore more fruitful dynamical processes beyond the previous studies. In the Rabi oscillation scenario, H_2^+ may first absorb one photon and then transit from the $1s\sigma_g$ state to the $2p\sigma_u$ state. Once the accumulated population in the $2p\sigma_u$ state is larger than that in the $1s\sigma_g$ state, H_2^+ in the $2p\sigma_u$ state may emit one photon and dump to the $1s\sigma_g$ state, forming a complete Rabi oscillation. During the dissociation, if the NWP undergoes half-integer multiples of Rabi oscillations, the proton will end with the net-one-photon absorption, which is similar to the bond-softening scenario only from the energy point of view," the researchers add.

"The time-resolved scenario fulfills an essential step of the well-known Rabi oscillations from atoms to molecules, giving birth to a complete understanding of the laser-driven molecular dissociation, particularly the ejection of slow nuclear fragments. The electron hopping mechanism presented here is general for strong-field dynamics of a stretching molecule, which also has implications for complex molecular processes, including the nuclear-electron correlations," the scientists say.

More information: Shengzhe Pan et al, Rabi oscillations in a stretching molecule, *Light: Science & Applications* (2023). DOI: 10.1038/s41377-023-01075-9

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