Rydberg-state excitation suppression of diatomic molecules in strong near-infrared laser fields

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Synopsis: In this article, we perform a comparative study between Rydberg state excitation of diatomic molecules (N_2 and O_2) and their companion atoms (Ar and Xe) in strong 800-nm laser fields. We show both theoretically and experimentally that neutral molecules can also survive strong 800-nm laser fields in high-n Rydberg states, while their behavior is remarkably different in comparison with their companion atoms. The effect of the structures of molecular orbitals is discussed.

It was surprising to find, both theoretically and experimentally [1], that neutral atoms can survive strong laser fields in Rydberg states. While neutral Rydberg state excitation (RSE) of molecules in strong laser fields has yet to be observed. In this study, by employing pulse-field ionization method combing with Time-of-flight mass spectroscopy, we have observed, for the first time, the RSE of diatomic molecules (N_2 and O_2) in strong 800-nm laser fields. The RSE yields of molecules have compared with their companion atoms with similar IPs (N_2 vs Ar and O_2 vs Xe), showing the molecular structure effect on the RSE process [2].

As shown in Figure 1, both the single ionization and Rydberg excitation yields of N_2 are similar to those of Ar. While comparing to Xe, the single ionization of O_2 is suppressed, which is in agreement with previous studies. The RSE suppression of O_2 is more significant than ionization. Our TDSE calculations qualitatively reproduced the experimental observations.

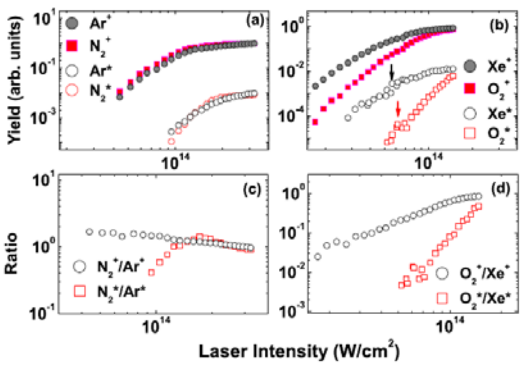

Figure 1. (a) and (b) Experimentally measured single ionization and Rydberg excitation yields. (c) and (d) Ratios of single ionization and Rydberg excitation yields.

Further investigations show that the Rydberg populations and spatial distributions of N_2, Ar, and Xe are very similar, however, the Rydberg electrons of O_2 have lower energy and the spatial distribution apparently deviates from the laser field direction. It is indicated that the difference between the angular distributions of N_2 (similar to atoms) and O_2 is attributed to their HOMO structures.

Moreover, the the angular distribution of O_2 is noticeably wider than those of other species. A wider angular distribution leads to a stronger diffusion of the wave packet of the electron ionized from O_2 when it evolves in the laser field. Therefore, it will be harder for the electron to occupy high Rydberg states which locate relatively farther from the ionic core. This explains the more pronounced suppression of the RSE compared with the ionization for O_2.

In conclusion, we have observed the suppressed RSE probability in O_2 compared to Xe, and the suppression is stronger than that in single ionization. Our TDSE calculations well reproduce the experimental measurements. Analysis indicates that the structure of molecular orbitals is the reason of the suppression of molecular RSE.

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