Dynamics of molecular rotational wave packets induced by intense ultrashort pulsed laser fields

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When molecules are exposed to an intense ultrashort laser pulse, a superposition of the rotational eigenstates called a rotational wave packet (RWP) is created by the impulsive rotational Raman transition. Dynamics of the RWP of linear molecules has been extensively investigated [1], and it has been known that the molecular axis distribution evolves in time periodically and the alignment of the molecular axis with respect to the laser polarization direction is achieved under the field-free condition at the rotational revival times.

In the present study, we examine the RWP dynamics of H_2O. H_2O is one of the simplest non-linear molecules whose moments of inertia along the principal axes are different to each other, and this type of molecules is classified as an asymmetric top. Based on the dependence of the tunnel ionization probability of H_2O on the angle between the molecular axis and the laser polarization axis, the RWP dynamics is investigated by the pump-probe measurement of the yield of the molecular ions, H_2O^+, in which the RWP is created by the pump laser pulse and the ionization is induced by the probe laser pulse.

A linearly polarized femtosecond laser pulse from the Ti:Sapphire laser amplification system was split into two by the Michelson interferometer and the split pulses were focused on the supersonic gas jet of H_2O vapor diluted with Ne in a vacuum chamber. The yield of the produced H_2O^+ was recorded as a function of the delay between the pump and probe pulses in the delay range of -0.3-26.7 ps. We found that the recorded H_2O^+ yield varies irregularly depending on the delay. This result is ascribed to the irregular rotational energy level structure of the asymmetric top. The Fourier transform spectrum of the recorded delay dependent H_2O^+ yield exhibits the strong peaks at 70 cm^{-1}, 111 cm^{-1} and 113 cm^{-1}, which are assigned as the rotational energy differences of J_{KaKc} = 2_{02}-0_{00}, 2_{21}-1_{01}, and 3_{03}-1_{01}, respectively. These results mean that the rotational Raman transitions between these energy levels are induced by the laser field. From the recorded delay dependent H_2O^+ yield, we also found that the tunnel ionization probability of H_2O is enhanced when the molecular c axis, which is perpendicular to the molecular plane of H_2O, is parallel to the laser polarization direction. This result indicates that the molecular orbital from which an electron is emitted is the highest occupied orbital of H_2O, whose lobes are located along the c axis. In this talk, we also briefly report the ionization dynamics revealed from the similar pump-probe experiments on the RWPs of NO [2] and O_2.

On the other hand, in some molecular species, the rotational energy levels are split into sublevels by the coupling between molecular rotational angular momentum and another angular momentum. In the case of NO, the rotational energy level is split into two with the small energy splitting by the Λ-type doubling, which originates from the interaction between the rotational and electronic orbital angular momenta. Recently, we reported theoretically [3] that the wave packet composed of the Λ-type doublets can be created by an intense sub-nanosecond laser pulse and evolves in time with the period of 2.8 ns. In order to observe wave packet dynamics with the ns time scale, we generated an intense sub-nanosecond laser pulse from the frequency-doubled output of the injection-seeded nanosecond Nd:YAG laser system by the stimulated Brillouin scattering in water. The generated sub-nanosecond laser pulses can be used for the examination of not only the wave packet composed of the Λ-type doublets but also other types of wave packets which evolve in time with the period of the ns time scale, such as RWPs of large molecules or wave packet composed of the energy levels associated with the hyperfine splitting.

References

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