# Rotational Excitation of Asymmetric Top Molecules Induced by An Intense Femtosecond Laser Field

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The rotational wave packet, which is a coherent superposition state of rotational eigenstates, can be created by the anisotropic polarizability interaction, i.e., the impulsive rotational Raman excitation induced by an intense nonresonant femtosecond laser field. The time evolution of the rotational wave packet exhibits transient and periodic molecular alignment and anti-alignment. Research on such rotational dynamics has been mainly focused on linear molecules because of the simplicity and regularity of rotational energy structure [1]. On the other hand, few studies on the rotational dynamics of asymmetric top molecules induced by the intense laser field were reported [2]. In this work, we investigate the rotational motion of a simpler, lighter, and asymmetric top $H_2O$ molecule.

In our experiment, an output from a Ti:Sapphire femtosecond laser system (784 nm, 130 fs, 5 mJ/pulse, 10 Hz) is introduced into a Michelson interferometer to generate pump and probe pulses. A mixture gas of $H_2O$ vapor (2 kPa), $N_2$ (20 kPa) as a reference, and Ne (180 kPa) as a buffer is expanded into a vacuum chamber equipped with a time-of-flight mass spectrometer (TOF-MS) as a supersonic molecular beam. The pump and probe pulses are focused on the molecular beam. Both pump and probe polarizations are set to be parallel to the TOF axis. The yield of $H_2O^+$ is measured as a function of the delay between the pump and probe pulses.

Figure 1 shows the ion yields of $H_2O^+$ and $N_2^+$ as a function of the delay. Clear periodic peaks at multiples of a quarter of the rotational period, $T_{\mathrm{rot}} = 8.4$ ps, are observed as a rotational motion for $N_2^+$. The increase (decrease) of $N_2^+$ yield from the baseline means that the molecular axis is aligned parallel (perpendicular) to the polarization direction because the ionization probability depends on the angle between the molecular axis and the polarization. The angular dependence of the ionization probability reflects the shape of the molecular orbital [3], from which the electron is ejected. The $N_2$ molecule ionizes easier when the molecular axis and the polarization of the probe pulse are parallel than perpendicular because the observed $N_2^+$ is created by the electron ejection from the HOMO, $3\sigma_g$ orbital.

On the other hand, the $H_2O^+$ yield does not show periodic peaks due to the irregularity of the rotational energy structure. By comparing the observed signal with the calculation, we found that the $H_2O^+$ ion is generated by the electron ejection from the HOMO. In addition, we determined the polarizability of $H_2O$ vapor, for which the two different values have been reported [4].

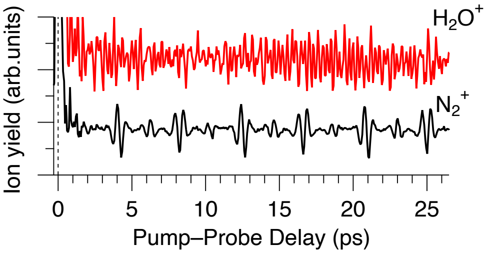

Fig.1 The delay dependence of the $H_2O^+$ and $N_2^+$ yields

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