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Molecules in intense laser fields: Ultrafast dynamics and high-resolution spectroscopy

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By pump-probe measurements with high temporal resolution using near-IR few-cycle laser pulses, we revealed that the yield of H_3^+ generated from CH_3OH^{2+} exhibits a long-lasting periodic increase reflecting the motion of the vibrational wave packet in methanol cation along the C-O bond stretching coordinate, showing that the time-resolved measurement of the yields of fragment ions is an efficient tool not only for probing ultrafast nuclear dynamics of molecular cations but also for deriving their vibrational frequencies. We further performed pump-probe measurements of methanol and its isotopologues (CH_3OH, CH_3OD and CD_3OH), and obtained the vibrational mode frequencies of methanol and methanol cations by Fourier transform (FT) of the yields of the parent ion and the fragment ions recorded as a function of the pump-probe delay time. In order to examine how high the resolution of a frequency domain spectrum obtained by strong field FT spectroscopy could be, we performed pump-probe measurements of the yields of D_2^+ and D^+ produced after the photoionization of D_2 using intense near-IR few-cycle laser pulses. The yields of D_2^+ and D^+ recorded up to the pump-probe delay time of 527 ps exhibited oscillatory structures reflecting the motion of the created vibrational wave packet of D_2^+, and the FT of the data in time domain revealed that the vibrational level separations can be determined with high precision, showing a potential application of the strong-field pump-probe measurements to high-resolution spectroscopy of molecular ions.