

Abstract Submitted
for the DAMOP13 Meeting of
The American Physical Society

Controlling generation process of an attosecond electron wave packet in a molecule HIROMICHI NIIKURA, Waseda University — When a molecule is exposed to intense laser fields, tunnel ionization may occur from not only the highest molecular orbital but also the lower electronic states. This process creates electron wave packet motion in a molecule. Using unaligned molecules, we have demonstrated that the attosecond electron wavepacket motion can be mapped onto the 2D high-harmonic spectra measured as a function of the delay between the two-color laser pulses in 2011. Here we extend this approach to control and identify which electronic states are included in the electron wave packet. We combine a molecular alignment technique with the two-color field approach. Using a linearly polarized laser pulse, we create a rotational wave packet that exhibits field-free alignment. Then we probe the electron wave packet by using the orthogonally polarized 800 nm and 400 nm pulses. By aligning molecules, we control the tunnel ionization probability of each electronic state. We observe that the 2D high-harmonic spectra generated by the two-color fields depends on the molecular alignment angle. We show that the portion of the electronic states responsible for the high harmonic emission, thus, generation process of the attosecond electron wave packet can be controlled.

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Date submitted: 25 Jan 2013

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