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Non-Born-Oppenheimer electronic wavepacket in molecular nitrogen at 14 eV probed by time-resolved photoelectron spectroscopy C. MARCEAU, Joint Attosecond Science Laboratory, University of Ottawa and National Research Council, 100 Sussex Drive, Ottawa, ON K1A 0R6 Canada, V. MAKHIJA, University of Ottawa, P. PENG, M. HERVE, P. B. CORKUM, A. YU. NAUMOV, Joint Attosecond Science Laboratory, U. Ottawa and NRC, A. STOLOW, U.Ottawa and NRC, D.M. VILLENEUVE, Joint Attosecond Science Laboratory, U. Ottawa and NRC — Attosecond pulses provide a means to coherently excite a set of electronic states in atoms and molecules. Here we use a train of attosecond pulses to excite a pair of electronic states in molecular nitrogen. The excitation frequency is centered at 14 eV, and populates two coupled electronic states – the valence state $b'\ ^1\Sigma_u^+\ v\ =\ 13$ and the Rydberg state $c_4'\ ^1\Sigma_u^+\ v\ =\ 4$. The resulting electronic wavepacket is probed by a time-delayed 800 nm pulse which, through two-photon absorption, ionizes the molecule. The pump-probe final state-resolved photoelectron spectrum exhibits a quantum beat with a period of about 240 fs, corresponding to a frequency of 4.1 ± 0.2 THz. The phase of the quantum beat depends on the vibrational quantum number of the final state in the cation. The zeroth order Born-Oppenheimer representations of the electronic states strongly perturb each other since they have the same symmetry. Optical excitation of the intermediate levels causes major zeroth order population oscillations at the observed frequency that are then mapped onto the photoelectron spectrum, qualitatively reproducing the experimental observations.

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