Abstract Submitted for the DAMOP06 Meeting of The American Physical Society

Measuring Attosecond Electron Motion in Molecules¹ ANDRE D. BANDRAUK, GENNADY YUDIN, Universite de Sherbrooke, PAUL B. CORKUM, National Research Council, ATTOSECOND SCIENCE TEAM — We study analytically the possibility of monitoring electron motion in a molecule using two ultrashort pulses [1]. The first pump pulse prepares a coherent superposition of two electronic molecular states whereas the second (attosecond) probe photoionizes the molecule. We show that direct information about electron dynamics on an attosecond time scale can be obtained from the photoelectron spectra as a function of time delay between the two pulses. In particular asymmetries in photoelectron angukar distributions provide a simple signature of the electron motion within the initial time-dependent coherently coupled two electron molecular states. It is further shown that "chirped" pulses can measure attosecond time scale electron dynamics just as effectively as "transform-limited" attosecond pulses of the same bandwith [2]. Chirped pulses with frequency dependent phases create interferences in the photionization spectrum which are absent from transform-limited pulses. These interferences contain important information about the chirp rate and the attosecond electron dynamics. [1] G Yudin, S Chelkowski, A D Bandrauk, P B Corkum, Phys Rev A 72,051401(2005) [2] G Yudin, A D Bandrauk, P B Corkum, Phys Rev Lett (2006)

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Date submitted: 25 Jan 2006

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