Attosecond dynamics measured in the C state of molecular nitrogen

Content

Due to the large number of accessible overlapping states, the ultrafast photoionization dynamics in molecules is rich and complex. In order to access the ultrafast evolution of the system experimentally, it is advantageous to detect all the charged products in coincidence. For such experiments an attosecond beamline driven by a 100 kHz noncollinear optical parametric chirped pulse amplification (NOPCPA) system was developed [1]. The beamline incorporates a reaction microscope (REMI) for performing electron-ion coincidence detection measurements. The NOPCPA delivers few-cycle laser pulses with approximately 7 fs duration at 800 nm central wavelength. Using these pulses, relatively short attosecond pulse trains (APTs) are obtained through high-order harmonic generation (HHG) [2]. The APTs are combined with synchronized near infrared (NIR) pulses in a pump-probe configuration.

Here we report on a series of RABBITT (Reconstruction of Attosecond Beating By Interference of Two photon Transitions) experiments performed in gas mixtures of N_2 with argon or helium. In these experiments the photoionization dynamics associated with the photoinduced transitions to the predissociating $C\,^2\Sigma_u^+$ state of N_2^+ was investigated. Using the electron-ion coincidence capability of the REMI, the photoelectrons accompanying the formation of different vibrational states ($v =$ 3-7) of the C state were identified. Photoionization delays corresponding to the formation of the $C\,^2\Sigma_u^+$ state were extracted from the acquired RABBITT spectrograms and compared with a noble gas reference. Theoretical calculations required for a better understanding of the observed non-trivial photoionization delay energy dependence have been performed and show a very encouraging agreement with the experiment.

[1] Federico J. Furch et al., "CEP-stable few-cycle pulses with more than 190 μJ of energy at 100 kHz from a noncollinear optical parametric amplifier" Opt. Lett. 42, 2495-2498 (2017)

[2] M. Osolodkov et al., "Generation and characterisation of few-pulse attosecond pulse trains at 100 kHz repetition rate" J. Phys. B: At. Mol. Opt. Phys. 53, 194003 (2020)

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