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Attosecond electron dynamics in N$_2^+$ dissociation

Photo-ionization of molecular nitrogen induced by extreme ultraviolet (XUV) radiation is of relevant importance in the photochemistry of the Earth’s upper atmosphere [1]. In this context, XUV attosecond sources provide a unique tool to study, with extremely high temporal resolution, the dynamics of the highly-excited electronic states activated by photoionization [2-3], representing a crucial step in understating the atmospheric radiative-transfer processes. In the seminar I will describe the investigation of ultrafast dissociative dynamics of N2+. The momentum distribution of the produced N+ fragments was recorded using a Velocity Map Imaging spectrometer and the kinetic energy spectrum of N+ ions was studied as a function of the delay between a 300-as pump pulse and a 4-fs VIS/NIR probe pulse.

In the pump-probe measurement we observed a strong depletion of the signal at 1 eV (F-band [4]) 8 fs after zero time-delay. In addition, a clear sub-cycle modulation of the ion yield is visible, showing an evident time vs energy dependence (tilt in the fringe pattern). In order to simulate the pump-probe experiment the Time Dependent Schrödinger Equation was solved in a set of 616 coupled electronic states of N2+ taking into account the VIS/NIR probe pulse. The numerical results are in excellent agreement with the experimental data and provide the physical interpretation of the processes under study. The strong depletion of the F-band is due to two resonant single photon transitions that transfer population to higher excited states of N2+. The sub-cycle modulation, on the other hand, is the interference between two different dissociation paths. The tilt in the interference pattern is caused by the dispersion of the wavepacket during propagation, thus carrying important information on the shape of the N2+ potential energy curves involved in the ultrafast dissociative process. This result demonstrates the possibility to follow and control the ultrafast relaxation dynamics of the excited electronic states activated by the XUV light, which is clearly of high interest for the complete understanding of photo-stability of molecules exposed to ultraviolet radiation.