THz Streaking of the Autoionization Dynamics of O₂ at the Free-Electron-Laser FLASH

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Synopsis With a XUV-pump/THz-probe scheme the decay of excited O₂ cations that were produced by irradiation with XUV photons has been investigated using a Reaction Microscope (ReMi) at the free-electron-laser FLASH in Hamburg. The temporal profile of electrons emitted due to autoionization of Rydberg states $C^4 \Sigma_u^-(n l \sigma_g)$ has been traced using THz streaking. This way the relaxation dynamics was followed. The relative shift in phase between photoelectrons and autoionization electrons was analyzed with respect to emission time delays or autoionization lifetimes.

A proper understanding of the dynamical response of atomic and molecular systems to intense pulses of XUV radiation is of vital importance for the description of the relaxation dynamics and multiple ionization process in the X-ray regime. As a consequence, the interaction of XUV radiation with small molecules leading to the population of highly excited states that evolve on ultrafast time-scales has attracted considerable theoretical and experimental interest. Technologically, attosecond streaking has become a well-established technique to obtain time-resolved information about the electronic processes as well as for the characterization of XUV pulses [1, 2]. Here, we use the FEL pulses (60 eV photons, intensity about 10¹³ W/cm²) together with synchronized THz pulses (wavelength 152 μ m, oscillation period 507 fs).

Within a kinematically complete experiment the emitted electrons are streaked by the simultaneously acting THz-field and they are detected in coincidence with the emerging ion(s). The highly excited states in the molecular ion are created by absorption of one (or more) XUV photons. Both, the photoelectron as well as the secondary Auger or autoionization electron are exposed to the streaking field. This leads to a momentum or energy shift that allows determining the moment of ionization. Thus, the phase shift between the photoelectron and the secondary electron carries information about the time difference between both emission steps or the autoionization lifetime. For autoionization of singly charged O₂ molecules we expect lifetimes between a few 10 and up to 300 fs [3, 4]. Using an XUV-pump/THzprobe scheme, we performed a direct measurement of the autoionization process of singly charged excited oxygen ions. As shown in Fig.1, the autoionizing Rydberg states $C^4 \Sigma_u^- (n l \sigma_g)$ have lifetime of 96 ± 21 fs.



Figure 1. The temporal profiles of the momenta of the photoelectrons and autoionization electrons.

Theoretical work on the autoionization mechanisms of Rydberg series of O_2 cations is needed for further understanding of the relaxation dynamics of the class of excited states.

References

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