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First attosecond pulse control by multilayer mirrors above 100 eV photon energy

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INTRODUCTION. Multilayer XUV mirrors serve as key components in the generation of the attosecond pulses from high harmonic radiation[1]. Those pulses pave the way to investigation of dynamics of electronic motion in atoms, molecules and nanostructures with a never before achieved precision and thus allow to draw conclusions on the basic underlying physics[2, 3]. Each experiment requires its perfectly tailored attosecond XUV pulse, fully optimized to perfectly match the experimental requirements as spectral resolution with sufficient signal to noise, temporal resolution and chirp. Until recently, experiments were limited to photon energies below 100 eV due to the available HH intensities as well as X-ray optics. Here, we have performed for the first time characterization of single isolated attosecond pulses at about 130 eV by attosecond electron streaking measurements and applied these pulse to photoionization experiments in Xe atoms. In order to achieve improved spectral resolution and high time resolution simultaneously, as is required for comparing attosecond electron dynamics of two adjacent electronic states, we have spectrally cleaned the attosecond pulse by suppressing unwanted “out-of band” radiation by improved a-periodic multilayer technology, resulting in an improved signal-to noise ratio for time-resolved electron spectroscopy experiments. This opens the way to study electron dynamics of energetically adjacent electronic states with a temporal resolution which is well below the attosecond pulse length itself[4]..

EXPERIMENTAL METHODS. The attosecond experiments were performed at the AS3 beamline at the Max Planck Institute for quantum optics in Garching, Germany. Numerical algorithms as the "needle optimization" [5] have been used to design the aperiodic ternary coatings of La/Mo/B_4C for normal incidence reflection at about 130 eV and a bandwidth of about 5 eV FWHM... While standard periodic multilayer structures exhibit secondary Kiessig fringes in their

reflectivity spectrum, these can be efficiently suppressed by advanced a-periodic multilayer structures Two different mirrors, with and without Kiessig fringe suppression, have been produced via ion beam deposition and controlled by insitu-ellipsometry. ., The reflectivity of both mirrors has been characterized by reflectivity measurements at the Advanced Light Source (ALS)[6] . The spectral phase of such mirrors, which controls the chirp of the XUV pulse, is much more difficult to access. Here we use attosecond electron streaking in Ne in a XUV pump/IR probe setup[7] and a FROG/CRAB algorithm[8] to fully retrieve the temporal and spectral intensity of the XUV pulse as well as its phase. Finally, the Xe photoelectron time-of flight spectra have been measured and compared for the two different cases of spectrally improved and non-improved XUV pulses.

RESULTS AND DISCUSSION. Figure 1 displays a comparison between the measured photoelectron spectrum of Xe atoms for the periodic mirror designed for maximum reflectivity at 130 eV (left panel) and that of a noise suppressor mirror centred at 122 eV (right panel), together with their corresponding reflectivity measurements. Although the out-of band reflectivity suppression was not perfectly implemented as indicated by a small residual reflection at the low energy side of the Bragg peak, it is obvious, that the photoelectron spectrum is much cleaner in the case of the aperiodic mirror. This spectral cleaning allows to separate the Xe 5p state from the Xe 5s state, the latter being hidden in the electronic background for the non-optimized multilayer.

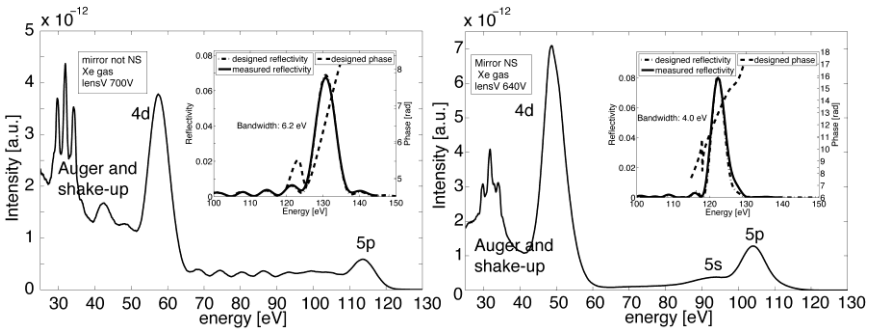

Figure 1 : Measured photoelectron spectra in Xe for attosecond pulses reflected at 130 eV from multilayer mirrors without (left) and with spectral purification. See corresponding reflectivity measurements for comparison.

Figure 2 displays the first attosecond streaking spectrograms measured in Neon gas together with the result from its FROG/CRAB retrieval. The retrieved temporal pulselength of 250 attoseconds is close to the expected Fourier limit.

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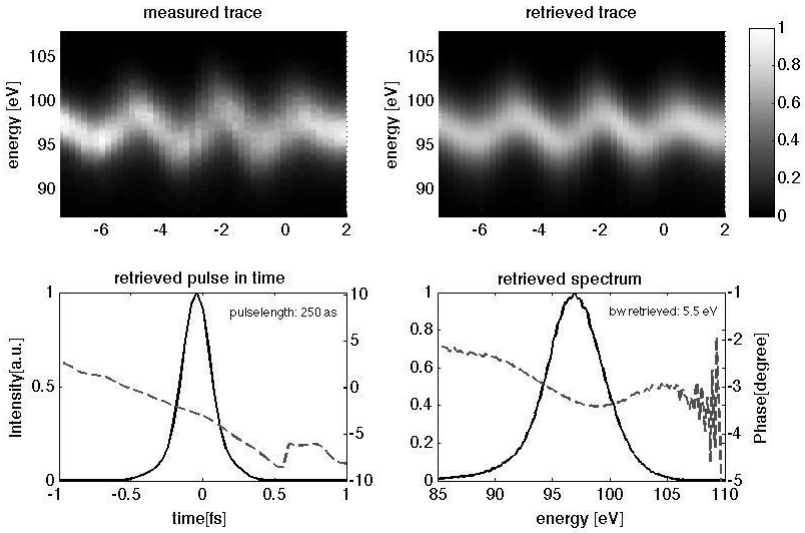

Figure 2 : Attosecond electron streaking measurement in Ne (upper left) and retrieval (upper right) for attosecond pulses at 130 eV. The corresponding temporal (lower left) and spectral (lower right) pulse retrieval displays a pulse duration of about 250 asec and a spectral bandwidth of about 5 eV FWHM..

CONCLUSIONS. First characterization of single isolated attosecond pulses from High Harmonics at ~ 130 eV photon energy has been performed and ~ 250 asec pulses have been filtered by multlilayer mirrors of 5 eV FWHM bandwidth. These pulses have been applied to measure photoelectron spectra in Xe An iproved spectral resolution and signal-to noise ratio in these spectra can be achieved by applying specialized aperiodic multilayer optics, which suppress unwanted radiation outside of the main Bragg maximum. Even for a relative small spectral bandwidth of 5 eV, resulting in ~250 asec pulse duration, spectral filtering results in improved electron spectra and thus allows for future study of electron dynamics even from energetically adjacent electronic states with temporal reolution, which can be well below the temporal pulse length. This concept of adjusting the spectral bandwidth and cleaniness of the attosecond pulse by the multilayer mirror can also be adapted to future investigation of the relative time dynakics of direct photoelectron and shake-up channels in surface experiments with temporal pulse durations of ~ 250 asec and time resolution well below 100 asec.

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