Phase-coherent, time-delayed harmonic pulses for high resolution spectroscopy in the XUV

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Pairs of time-delayed, phase-locked laser pulses have proved successful to perform Time-Delay Spectroscopy (TDS) and reach spectral resolutions normally unachievable to ultrashort-pulse sources [1, 2, 3]. The possibility to apply TDS in connection with high-order harmonic radiation is very attractive for the study of high-lying and autoionizing states [4] with one-photon transitions; such states have been mainly studied by synchrotron radiation up to now. Because of the intrinsic difficulty of splitting harmonic pulses in the XUV, we propose to generate phase-locked harmonic pulses by focusing two delayed identical laser pulses in the same position of the gas jet.

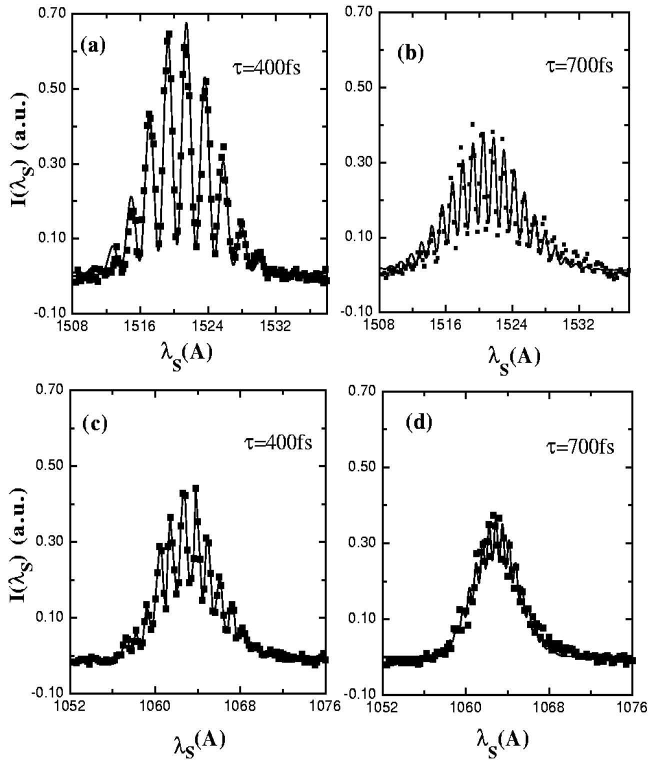

Figure 1: Spectra of the 5^{th} (a and b) and 7^{th} (c and d) harmonics for different time delays τ of the generating laser pulses. The solid lines are fits to the experimental data.

The 400 μJ, 100 fs pulses emitted by an amplified Ti:Sapphire laser (λ=800 nm) pass through a Michelson interferometer used to split them into two identical time-delayed pulses with a well controllable relative delay. The two-pulse sequence is focused by a 20 cm lens into the region of interaction with a supersonic jet of xenon which is injected into the vacuum chamber by a pulsed electromagnetic valve. The 5^{th}, 7^{th}, and 9^{th} harmonics produced in the interaction are observed with a phosphor screen and a phototube after dispersion by a spherical diffraction grating. By closing the output slit of the monochromator to 10 μm we achieve a spectral resolution of about 0.04 nm.
In Figure 1 we show the two-pulse spectra obtained for different delays of the pump pulses: the observed contrast of the fringes that modulate the single-pulse spectrum is well consistent with the predictions of a simple model that takes into account the instrumental resolution as the only source of contrast degradation.
These results demonstrate that phase-coherent harmonic pulses can be generated regardless of the significant ionization of the medium and that the passage of the first pulse through the gas jet does not perturb the generation of the second harmonic pulse.
Considering the possibility to apply this source to TDS, we have also performed some measurements varying the relative delay of the two pulses while selecting just a narrow spectral component of the emitted radiation: by doing this we effectively stretch in time the two harmonic pulses and allow them to overlap even for relatively long delays, when the two pump pulses no longer interfere. In principle this is similar to the real situation in a TDS experiment, where the atom, with its narrow resonances, plays the role of the spectral filter (the slit of the monochromator in our case). We find that clear fringes can be observed from the 5^{th} to the 11^{th} harmonic when the time delay is varied continuously up to about 1 ps. Also in this case the agreement with the results of our simple model is good enough to affirm that it is indeed possible to produce copropagating, phase-locked harmonic pulses that can be successfully applied to Time-Delay Spectroscopy in the XUV.

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