JITTER-FREE TIME RESOLVED RESONANT CDI EXPERIMENTS USING TWO-COLOR FEL PULSES GENERATED BY THE SAME ELECTRON BUNCH

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Abstract

The generation of two-color FEL pulses by the same electron bunch at FERMI-FEL has opened unprecedented opportunity for jitter-free FEL pump-FEL probe time resolved coherent diffraction imaging (CDI) experiments in order to access spatial aspects in dynamic processes. This possibility was first explored in proof-of-principle resonant CDI experiments using specially designed sample consisting of Ti grating. The measurements performed tuning the energies of the FEL pulses to the Ti M-absorption edge clearly demonstrated the time dependence of Ti optical constants while varying the FEL-pump intensity and probe time delay. The next planned CDI experiments in 2013 will explore transient states in multicomponent nanostructures and magnetic systems, using the controlled linear or circular polarization of the two-color FEL pulses with temporal resolution in the fs to ps range.

INTRODUCTION

In the last years the FEL pump-FEL probe experiments have been based either on time-delayed holography [1], or autocorrelator devices [2,3]. These approaches give access only to single color experiments. In 2013, at LCLS [4] it has been generated a pair of temporally and spectrally separate soft X-ray FEL pulses via a double undulator scheme. However, due to the underlying self-amplified spontaneous emission configuration, these pulses have partial longitudinal coherence and limited shot-to-shot repeatability in both energy and central wavelength. In the present paper we report the successful generation of two FEL pulses with precisely controlled time delay, wavelength and intensity ratio, using two-color, two-pulse external laser seeding of the High Gain Harmonic Generation (HGHG)-based FERMI@Elettra FEL. These highly coherent FEL pulse pairs are used in a proof-of-principle XUV pump - XUV probe experiment examining the dynamics of a thin-metal layer structure exposed to high intensity XUV excitation.

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GENERATION OF TWIN FEL PULSES

In a HGHG FEL [5,6] like FERMI@Elettra the output radiation properties (spectrum, duration, arrival time) are very tightly correlated with those of the input seed laser pulse. This scheme has demonstrated outstanding performances in terms of stability10, wavelength and polarisation tenability [7]. As proposed by Freund *et. al.* [8], a straightforward method for the generation of multiple X-ray pulses from an HGHG FEL consists of seeding the electron bunch with multiple laser pulses. In the specific configuration demonstrated here, we use two ultraviolet (UV), 180 fs-long (FWHM) seed pulses at slightly different central wavelengths (independently tunable in the 260-262 nm range) with variable time separation and intensity ratio. As schematically shown in Fig. 1a these pulses are focused in the modulator stage of the HGHG FEL and interact with a 750 fs-long electron bunch, for which attention was devoted in preserving the temporal uniformity of the beam parameters (mainly current and energy) using also the X-band RF cavity to linearize the longitudinal phase space of the electron beam to obtain a nominally flat current. After acceleration to 1.2 GeV, the electron beam is first energy-modulated by the seed pulses (in the modulator) and then density-modulated into two regions in which the phase/amplitude properties of the two seed pulses are encoded into the electrons. In the radiator undulator sections, having the magnetic strengths (K) tuned for fundamental FEL resonance with the N^{th} harmonic ($N = 7$ in this case) of the average seed wavelength, the two regions emit two temporally and spectrally independent XUV pulses having wavelengths scaled by a factor of 7 with respect to the seeding pulses. Tuning the delay between the input

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seed pulses can control the relative delay between the two pulses. Although the seed laser is linearly polarized, the FERMI undulators have variable polarization, permitting choice of either linear or helical output polarization.

A fine scan of the relative seed/electron-beam timing (Fig. 1c) ensures that a sufficiently wide, unused portion of electrons toward the tail can accommodate the second UV pulse (referred to as the "probe", while the first is the "pump"). When both seed pulses are used, double peaked spectra were observed if the temporal delay between the two pulses was set in the proper range. The pump-probe delay for this experiment was between about 300 fs and 700 fs. The limitations to the delay arise from the electron bunch length (upper side) and from the seed pulse duration (lower side). Figure 1b shows a typical spectrometer image of the double FEL pulses generated with a time delay of 500 fs. As it can be seen, the spectrum in this double-pulse seeded regime is extremely clean and stable.

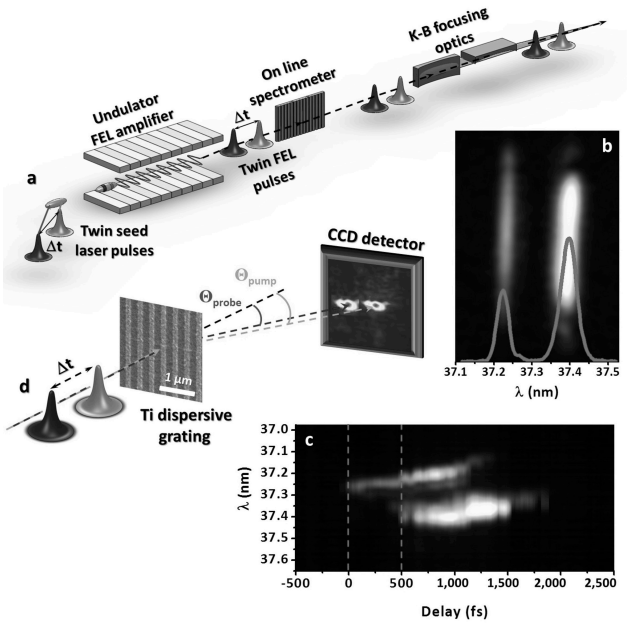

Figure 1: a) Scheme of the generation of the double pulse; b) double pulse as measured by the energy spectrometer; c) sequence of FEL spectra during a temporal scan of the electron bunch-seed laser delay; d) scheme of the experiment.

The switching between single and double FEL emission is obtained by blocking one of the seed laser arms, without changing neither the electron beam characteristics nor the undulator strength parameter *K*. A fit of the two-color FEL spectra as a sum of two independent Gaussians shows a negligible difference between the single and double FEL emission, confirming the robustness of the pulses generation scheme. The analysis of the spectra (Fig. 2a) shows that the bandwidth RMS is about 25×10^{-3} nm, corresponding to about 0.05% of the central wavelength, together with a shot-to-shot peak position RMS jitter of about 3×10^{-3} nm (~ 0.005% of the central wavelength, Fig. 2c). Figure 2b shows that the absolute single wavelength intensities of double FEL emission, obtained from the corresponding peak area of the spectrum, is comparable to those of single FEL emission, with a RMS shot-to-shot stability of about 15% (Fig. 2d). In the present scheme the ratio between the intensities of the pump and the probe pulses can be tuned easily by changing the relative intensity of either seed laser pulses.

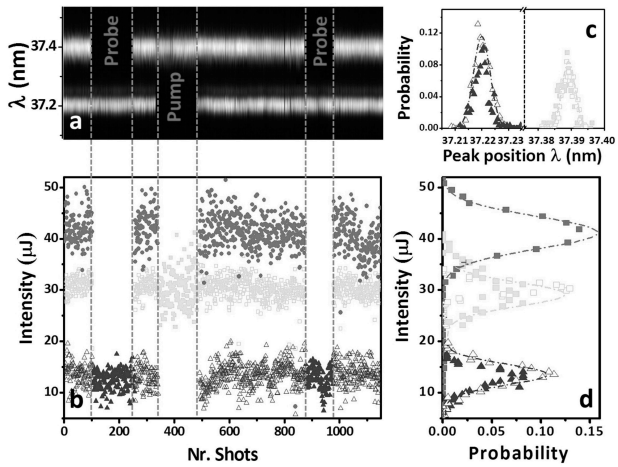

Figure 2: a) Sequence of spectra of both pump and probe as measured by the energy spectrometer; b) intensity of the pulses as measured by both the I0 monitor and the energy spectrometer used as an intensity detector; c) stability of the peak positions; d) stability of the peak intensities.

PROOF OF PRINCIPLE EXPERIMENT

As the wavelengths of the double-pulses are close together, a Ti grating (Fig. 1d) was used, acting as a spectral analyzer, and separating the diffraction peaks of the pump and probe pulses in angle. The wavelength and intensity of each pulse were measured by the I_0 gas monitor and the energy spectrometer [9] (Fig. 1b). The maximum pulse intensity of the pump pulse (30 μJ) corresponds to a fluence *F* of ~18.5 J/cm^2 on the sample, and it was attenuated using a gas cell and/or Al filters. The diffraction measurements were carried out at the DiProI endstation [10], where the FEL pulses were focused to a 195±30 μm^2 spot on the sample by a K-B active optical system [11]. As shown in Figure 1d, the pulses hit the sample at normal incidence and diffract along directions defined by angles depending on the FEL wavelengths, the diffraction order, and the grating pitch (see [12]).

Tuning the two-color pulses to wavelengths in the slope region above the Ti $M_{2/3}$ edge (λ~38 nm) the measurement is more sensible in terms of photon deflection angle and absorption, since the wavelength dependence of the complex refractive index undergoes sharp changes through the atomic resonances [13]. Consequently, the intensities and positions of the diffraction peaks are highly sensitive to the instantaneous Ti ionization state.

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Figures 3 a-c present the results in the low-*F* regime (F < 150 mJ/cm^2, the latter being the radiation damage threshold) when the radiation induces minor changes in Ti electronic structure. In this regime the diffraction patterns have the expected peak position and the two-color (pump-probe) pattern is a sum of the pump (Fig 3a) and the probe (Fig 3b) peaks (see [12] for further details). Figure 3d presents the single-shot, pump-probe pattern for the high-*F* regime: the diffraction pattern shows the contribution of the probe peak strongly attenuated. As the duration of the experiment (estimated FEL pulse length ~ 90 fs [14]; delay ~ 500 fs) is shorter than the time scales of hydrodynamic expansion 1 - 10 ps [1,15], the two pulses probe the same grating geometry (the estimated line expansion of the grating is smaller than 2.5 nm).

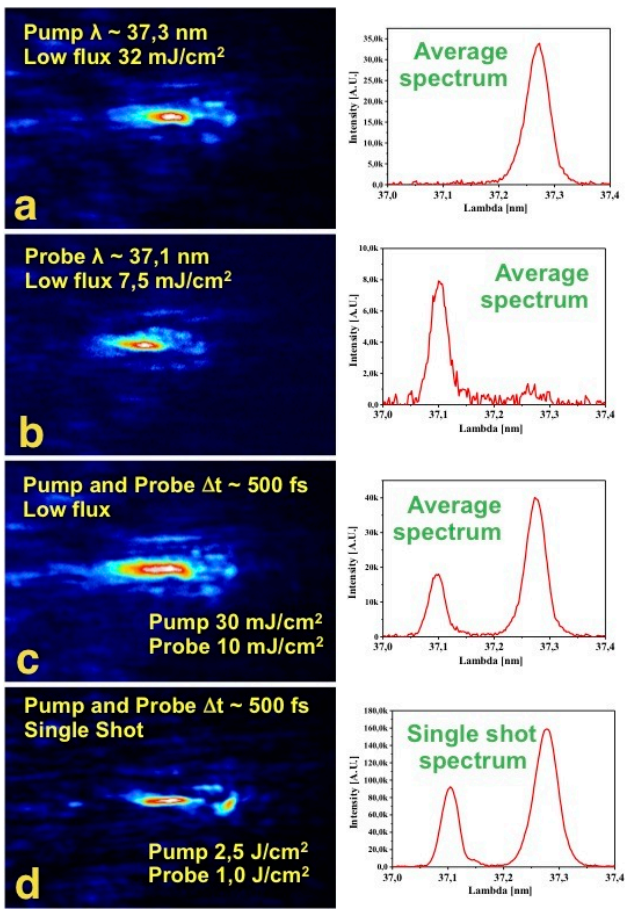

Figure 3: Diffraction images and energy spectra as acquired in different experimental conditions.

Consequently, the difference in the Fig. 3c and Fig. 3d patterns indicates dramatic changes in the Ti electronic structure due to the intense pump pulse. In particular, the high degree of ionization created by the pump shifts the absorption resonances to shorter wavelengths leading to abrupt changes in the complex index of refraction, resulting in a decrease of the diffracted peak intensity and width. In the low-*F* regime, less than 1% of Ti atoms are ionized by the pump and the optical properties of the material are negligibly affected by the material ionization state. On the contrary, at high-*F* almost all Ti atoms are ionized within a few 10's of fs. The generated primary photoelectrons then thermalize creating high ionization states through secondary electron emission and Auger decay. These events occur at time scales shorter than 100 fs [16,17] and, as a result, the absorption edge is shifted towards shorter wavelengths [18,19]

This behavior is similar to the observed photo-induced transparency of Al [20] obtained by high-fluence FEL pulses and to the quenching of the X-ray resonant magnetic scattering signal on Co/Pt multilayer systems using very intense FEL pulses [19]. However, the approach reported here has the remarkable potential advantage of providing time resolution in the sub-ps domain to follow the FEL-induced changes in material electronic structure and non-thermal ion motions.

CONCLUSIONS

The possibility to generate and use two separate XUV FEL pulses with precisely controllable wavelength and relative time delay by using a double-pulse, external laser seeding of the HGHG-based FEL source FERMI@Elettra was experimentally demonstrated. The FEL double pulses used had a relative time delay between 300 and 700 fs and central wavelengths of 37.2 and 37.4 nm, tuned to the Ti M2,3 absorption edge. This newly developed tool was exploited to perform the first all-FEL-based, two-color pump-probe proof experiment with specially designed Ti grating. For fluences greater than 2 J/cm^2 the measured pump-probe diffraction patterns indicate the occurrence of a photo-induced transparency evidencing the high degree of Ti ionization created by the intense pump pulse, which results in a shift of the Ti absorption edge to wavelengths shorter than those of the FEL twin pulses.

This technique can be extended to generate pump-probe pulses with similar wavelength separation in the whole currently available FERMI FEL range (20-65 nm), while the minimum time delay can be decreased to ~ 150 fs. The current upper limit of the delay between the two pulses can also be increased to above 1 ps by optimizing the FERMI photoinjector and linac and producing longer electron bunches with temporally flat output energy distributions.

From a wider perspective, the concepts reported here for generating multiple and multi-color coherent photon pulses with fully controllable parameters will open up the way to extend the most advanced, table-top ultrafast methods into the XUV/X-ray domain, thus potentially adding nanometer spatial resolution and atomic-selectivity to these powerful experimental tools. This will advance the knowledge to the fundamental domains of materials science, paving the road to future nonlinear X-ray technologies that cannot even be foreseen today.

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