Generation of Ultrashort Coherent Vacuum Ultraviolet Pulses Using Electron Storage Rings: A New Bright Light Source for Experiments

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We demonstrate for the first time that seeded harmonic generation on electron storage rings can produce coherent optical pulses in the vacuum ultraviolet spectral range. The experiment is performed at Elettra, where coherent pulses are generated at 132 nm, with a duration of about 100 fs. The light source has a repetition rate of 1 kHz and adjustable polarization; it is very bright, with a peak power several orders of magnitude above that of spontaneous synchrotron radiation. Owing to high stability, the source is used in a test photoemission electron microscopy experiment. We anticipate that seeded harmonic generation on storage rings can lead to unprecedented developments in time-resolved femtosecond spectroscopy and microscopy.

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Over the last few years, a strong demand has emerged for a source of radiation in the vacuum ultraviolet (VUV) spectral range with high brilliance, close-to-full coherence, variable polarization, bandwidth approaching the transform limit, and stable temporal structure in the femtosecond time scale. Much progress in this direction has been achieved through high-harmonic generation using fs lasers [1]. But nowadays, the possibility to realize a source with all the above mentioned characteristics relies principally on single-pass free electron lasers (FELs). A direct impact from FELs is expected on a large number of disciplines [2], including (bio-) materials sciences, nanosciences, plasma physics, and chemistry, where the use of ultrashort VUV and/or x-ray femtosecond pulses can tackle complex transient phenomena with femtosecond time resolution.

In a FEL, the light is generated when a relativistic electron beam passes through the static and periodic magnetic field produced by an undulator. FELs can be operated in several different schemes. The most promising ones are based on the self-amplification of the electron-beam spontaneous emission (SASE), and on the generation of coherent harmonics from an input signal provided, e.g., by a conventional laser (seeding). A SASE source can produce very high brilliance [3], but the pulse temporal structure results from the envelope of a series of micropulses with random intensity and duration. In contrast, seeded FELs can deliver coherent optical pulses with tailored temporal and spectral profiles [4,5].

In the standard seeded scheme, generation of coherent harmonics (CHG) is obtained by coupling the input signal with the electron bunches extracted from a linear accelerator (linac). Proof of principle experiments in this configuration were first performed at Brookhaven laboratory (Upton, USA) [4,5], where coherent emission at the third harmonic (260 nm) of a Ti:sapphire laser was produced. Based on these promising results, several projects are currently under development worldwide (see, e.g., [6]), with the aim of supplying users with new bright and coherent light sources in the VUV and soft x-ray spectral regions. As an alternative to single-pass linac-based devices, electrons can be provided by a storage ring (SR), as shown in Fig. 1. Electron bunches circulating into a SR are generally characterized by lower peak currents and higher relative energy spreads compared to bunches delivered by modern linacs. As a consequence, linac-based FELs can be expected to produce optical pulses whose energy is 3–4 orders of magnitude larger than that of pulses produced by SR CHG. However, using electrons recirculated in a SR presents some important advantages. Indeed, in a SR, the electron-beam properties, notably the mean energy and the

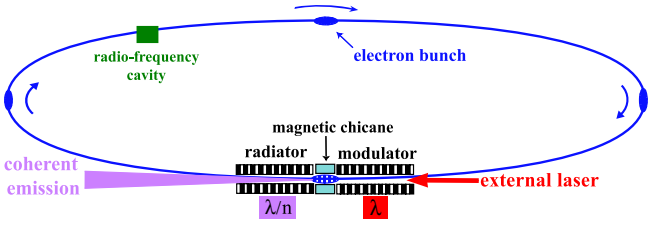

FIG. 1 (color online). Schematic layout of the CHG setup implemented on the Elettra SR. The SR is filled with a relativistic electron bunch, which interacts in the modulator with an external (Ti:sapphire) laser of wavelength λ. In the magnetic chicane, electrons are subdivided in micro-bunches separated by λ. Finally, in the radiator, they emit coherently at λ/n, n being an integer number. The radio-frequency cavity is used to provide electrons with the energy they lose during one turn of the ring.

peak current, are ''thermalized'' by a long-lasting periodic dynamics. This in principle results in a very good reproducibility of the seeding process and, as a consequence, in an excellent shot-to-shot stability of the optical pulse produced at the end of the device. On the contrary, successive electron bunches delivered by a linac are generally characterized by significant fluctuations of the mean electron-beam energy and current. According to theoretical calculations [7], this may result in a conspicuous shot-to-shot instability of the emitted harmonic power. Moreover, SR CHG allows the production of harmonic pulses at relatively high repetition rates (order of 1 kHz, or even higher), to the benefit of average harmonic power. Only FELs based on superconducting linacs can perform the same, while normal-conducting devices are limited to repetition rates of the order of 100 Hz. Finally, the coherent harmonic light generated by a SR is naturally synchronized to the synchrotron radiation emitted by undulators and bending magnets, and this makes easier the design and the realization of pump-probe experiments. Proof of principle of SR CHG was first performed at LURE (Orsay, France) [8] using a Nd:YAG laser (fundamental wavelength at 1.06 μm) to generate (third) harmonic coherent radiation at 355 nm. Recently, coherent emission at 260 nm, the third harmonic of a Ti:sapphire laser, was observed at UVSOR (Okazaki, Japan) [9].

In this Letter we demonstrate the possibility of using SR CHG to produce powerful and stable VUV radiation. Taking advantage of the FEL undulator system installed on the Elettra SR [10], we have generated ultrashort (close to 100 fs FWHM) coherent UV pulses at 132 nm (sixth harmonic of a Ti:sapphire laser), with fully adjustable polarization, relatively high repetition rate (1 kHz) and bandwidth close to the transform limit. The pulse train is very stable and the peak power is orders of magnitude above that of standard synchrotron radiation. The radiation was used for a test experiment in which a topographic image of a patterned SiO_2 sample was recorded using a SPELEEM microscope [11]. In the following, we describe the experimental setup, provide a complete characterization of the source, and briefly report about the test experiment.

The source layout is shown in Fig. 1. The region of seed-electron interaction is composed of two Apple-II identical independent undulators (10-cm period, 2-m long), separated by a magnetic chicane. Apple-II undulators allow us to freely determine the polarization of the emitted light. The seed laser is a Ti:sapphire, having a fundamental wavelength of 794 nm, a maximum repetition rate of 1 kHz and a maximum energy per pulse of 2.5 mJ. A nonlinear optical crystal was used to generate second harmonic pulses with energy of up to 800 μJ and duration of 120 fs FWHM, so that CHG was seeded at 397 nm. The process leading to CHG can be described as follows [12]. The laser is focused into the first undulator (called the modulator, and tuned at the seed wavelength), and synchronized with the incoming electron bunch. The laser-electron interaction in the modulator leads to a modulation of the electron energy. When the beam crosses the magnetic chicane, the energy modulation is converted into a spatial microbunching of electrons at the period of the seed wavelength. As this microbunching is nonsinusoidal, when analyzed in the frequency domain it contains significant harmonics of the fundamental. Finally, these microbunched electrons radiate coherently in the second undulator (called the radiator), which is tuned at a harmonic of the first. The extracted power is proportional to the square of number of seeded electrons. Figure 2 shows the intensity of the monochromatized radiation at 132 nm (i.e., the third harmonic of the seed) vs the acquisition time. The radiator is tuned either for circular [Fig. 2(a)] or for linear [Fig. 2(b)] polarizations. The enhanced peaks correspond to the coherent signal generated by the seeded electron bunch, while the small side peaks represent the spontaneous (i.e., unseeded) incoherent radiation. When the radiator field is set for circular polarization, the field-electron coupling is strengthened and, as a consequence, the optical gain is enhanced. This explains why the signal in circular polarization is about a factor 2 stronger than that generated when the radiator is in planar polarization. The electron bunch revolution frequency is about 1.16 MHz, corresponding to an interbunch period of 864 ns. This means that the bunch is seeded (i.e., generates coherent emission) only once every about 1160 turns. Hence, the repetition rate of the spontaneous (incoherent) emission generated by the beam at every pass through the undulators is much higher than that of coherent emission. Spontaneous emission produces background noise, which may be disturbing when the light is used for experiments. As we shall see, the problem can be solved by using a gated detector. As shown

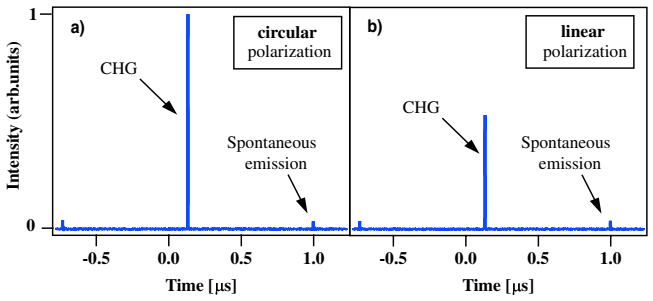

FIG. 2 (color online). Intensity of the UV pulses vs acquisition time. The signal was acquired using a photomultiplier (PMT) placed downstream a monochromator. Note that the PMT does not allow to resolve the sub-ps temporal scale on which the coherent pulse evolves. This, in turn, does not permit direct detection and therefore appreciation of the effective amplitude difference between the seeded and the spontaneous signals, their true ratio being a factor about 10^4 (see text). In (a) the radiator is tuned for circular polarization; in (b) the radiator is tuned for linear polarization.

in Fig. 2, the seeded signal in circular polarization is a factor about 25 above spontaneous emission, which becomes a factor about 10^4 if one takes into account the difference between spontaneous and coherent pulse durations, i.e., about 35 ps and 100 fs (FWHM), respectively. We assume here that the duration of the harmonic coherent pulse, Δt, is slightly shorter than the seed pulse duration. Such an assumption is supported by numerical simulations performed using the numerical code GENESIS [13]. For the reported experiment, the Elettra SR was operated in single-bunch mode, the electron energy was tuned at 1.1 GeV and the current per bunch was about 0.6 mA (corresponding to a peak current of 14 A). In these conditions, the number of measured photons per coherent pulse was of the order of 10^9.

The quadratic dependence of the harmonic signal on beam current, which is the signature of coherence, is shown in Fig. 3. Data were obtained from a scan of the relative position of the seed pulse within the electron bunch. The CHG signal is then plotted versus the effective electron-beam current at the location of the seed pulse. The current is normalized to the bunch peak current.

The spectrum of the coherent signal at 132 nm (radiator for linear polarization) is shown in Fig. 4(a), after subtraction of the radiator spontaneous emission. A direct measurement of the spectral width gives a bandwidth, $\Delta\lambda$, of about 0.33 nm FWHM. Assuming Gaussian temporal and spectral profiles, at the transform limit one would get $\Delta t = 0.44\lambda^2/(c\Delta\lambda) \simeq 77$ fs FWHM, which is close to the value predicted by GENESIS. Data shown in Fig. 4(b) demonstrate the good spectral stability of the source. In this case, the radiator is tuned at $\lambda_0 = 203$ nm, i.e., is slightly mismatched with respect to $\lambda_2 = 198.5$ nm, which is the second harmonic of the seed laser. As already explained, at the undulator entrance, the electron beam is "prepared" to emit coherently at one of the harmonics of the seed. Coherent emission at λ_2 still occurs provided the relative mismatch $(\lambda_0 - \lambda_2)/\lambda_0$ does not exceed the FEL gain width $\simeq 1/(N)$, N being the number of radiator periods. The effect of the mismatch on the radiator emission is shown in Fig. 4(b), where the measured spectrum is reported. As it can be seen, while the part of the spectrum generated by spontaneous emission is centered around the "detuned" wavelength (i.e., 203 nm), the position of the coherent signal is "locked" to the second harmonic of the seed laser. For the considered case, $(\lambda_0 - \lambda_2)/\lambda_2 \simeq 2.3\%$, which is well within the FEL gain width $1/N = 5\%$.

A fundamental feature of a light source to be exploited for user experiments is the shot-to-shot stability of the emitted power. The reproducibility of our source is shown in Fig. 5, where the acquisition of 200 consecutive pulses is reported as a function of time. A very good stability is found, with fluctuations of the order of few percent. As already mentioned, this is mainly due to the very good electron-beam stability, which is a common characteristic of all modern storage rings. An important role is also played by the low seed-electron timing jitter, which is about 2 ps in our case. Such a value is small enough to guarantee that, during consecutive shots, the laser "seeds" beam portions containing very similar numbers of electrons. Concerning longer term reproducibility, for relatively low current values ($\leq$1 mA), i.e., for a relatively long beam lifetime, we observed a very stable behavior over several hours.

The CHG source was employed in a test experiment using a SPELEEM microscope. This instrument combines energy-filtered photoemission electron microscopy (PEEM) with microspectroscopy (micro-XPS) [14]. Data acquisition was synchronous with the photoemission signal generated by seeded or nonseeded pulses, by gating (over

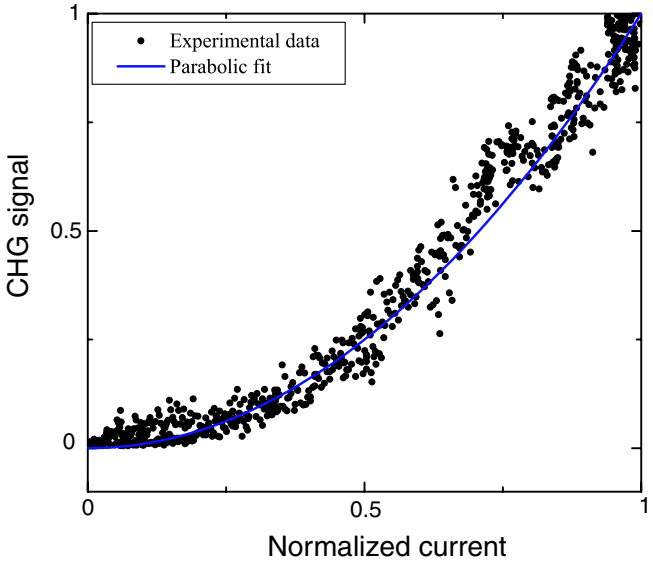

FIG. 3 (color online). Quadratic dependence of the coherent harmonic detected using a PMT vs (normalized) bunch current. Dots represent experimental data; the curve is a fit obtained using a quadratic function.

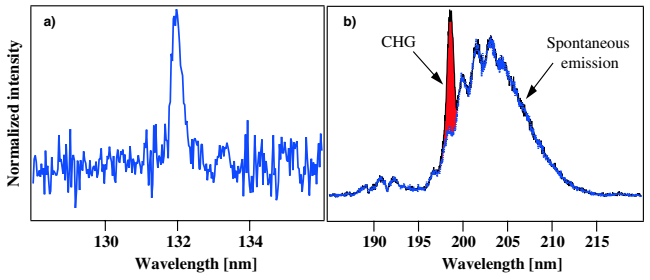

FIG. 4 (color online). (a) Spectrum of the coherent emission at 132 nm (linear polarization). The integration time is 1 ms; the spectrum is obtained after subtraction of the background due to spontaneous emission. (b) Spectrum of spontaneous and coherent emission for the case in which the radiator is tuned at 203 nm, i.e., slightly mismatched with respect to the second harmonic of the seed laser (198.5 nm).

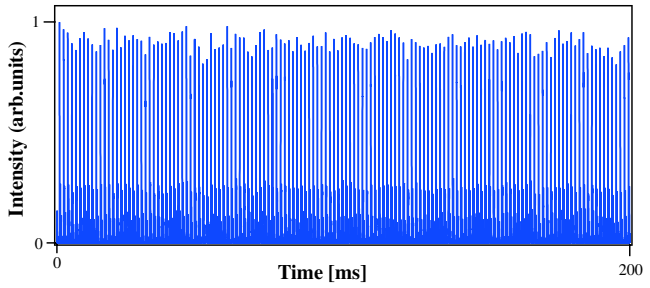

FIG. 5 (color online). Acquisition of the coherent signal during 200 ms (i.e., 200 turns pulses).

about 200 ns) the multichannel plate imaging detector of the microscope at the CHG repetition frequency (1 kHz). Each PEEM image was obtained by accumulating the signal from pulses at fixed time delay from the reference bunch marker and suppressing that from all other pulses. The background generated by the radiator spontaneous emission was subtracted from all CHG spectra and images. In this way, we could eliminate the unwanted contribution of synchrotron radiation, which overlaps the CHG pulses.

Using CHG we recorded topographic images of a patterned SiO_2 sample. Results demonstrate feasibility of CHG-PEEM, at least at intermediate lateral resolution, and prove that the source can be fruitfully employed in a realistic photoemission electron microscopy experiment.

In conclusion, we implemented a presently unique light source, which is able to produce ultrashort coherent photon pulses in the VUV spectral region. Our results show that seeded coherent harmonic generation on electron storage rings can significantly extend the capabilities of presently available light sources. In particular, the most evident advantages of coherent harmonic generation with respect to standard synchrotron radiation rely on the time structure and the coherence of the generated optical pulses.

It is worth stressing that there is a quite wide margin for improving the currently achieved performance. Indeed, seeding with the third harmonic of the Ti:sapphire (i.e., 265 nm) will provide the possibility of extending the source wavelength range well below 100 nm (e.g., tuning the radiator at the third harmonic of the seed, 88.3 nm). Moreover, the maximum repetition rate at which the harmonic signal can be produced (1 kHz) is presently limited only by the seed laser repetition rate. Given the rapid progress in the development of high repetition rate Ti:sapphire amplifiers (see, e.g., [15]), we anticipate a significant increase of the average harmonic power in the next few years. Potential applications of the source we have developed range from atomic and molecular physics to the study of electronic excitations on surfaces, interfaces, and nanostructures.

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[1] T. Pfeifer *et al.*, Rep. Prog. Phys. **69**, 443 (2006); J. Seres *et al.*, Nature Phys. **3**, 878 (2007).
[2] R. Neutze *et al.*, Nature (London) **406**, 752 (2000); W. A. Barletta and H. Winck, Nucl. Instrum. Methods Phys. Res., Sect. A **500**, 1 (2003); N. Gedik *et al.*, Science **316**, 425 (2007); A. Cavalieri, Nature (London) **448**, 651 (2007).
[3] A. Kondratenko and E. Saldin, Part. Accel. **10**, 207 (1980); Y. Derbenev, A. Kondratenko, and E. Saldin, Nucl. Instrum. Methods Phys. Res., Sect. A **193**, 415 (1982); R. Bonifacio, C. Pellegrini, and L. Narducci, Opt. Commun. **50**, 373 (1984); M. Babzien *et al.*, Phys. Rev. E **57**, 6093 (1998); S. V. Milton, Phys. Rev. Lett. **85**, 988 (2000); J. Andruszkow, Phys. Rev. Lett. **85**, 3825 (2000); R. Brinkmann, Proceedings FEL Conference 2006.
[4] A. Doyuran *et al.*, Phys. Rev. Lett. **86**, 5902 (2001).
[5] L. H. Yu *et al.*, Phys. Rev. Lett. **91**, 074801 (2003).
[6] G. Bocchetta, A. Mesnek, M. E. Couprie, and L. Poletto *et al.*, in Proceedings FEL Conference 2006.
[7] G. De Ninno *et al.*, Proceedings FEL Conference 2006.
[8] B. Girard *et al.*, Phys. Rev. Lett. **53**, 2405 (1984).
[9] M. Labat *et al.*, Eur. Phys. J. D **44**, 187 (2007).
[10] G. De Ninno *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. A **507**, 274 (2003).
[11] T. Schmidt *et al.*, Surf. Rev. Lett. **5**, 1287 (1998).
[12] L. H. Yu, Phys. Rev. A **44**, 5178 (1991).
[13] S. Reiche, Nucl. Instrum. Methods Phys. Res., Sect. A **429**, 243 (1999).
[14] A. Locatelli *et al.*, Surf. Interface Anal. **38**, 1554 (2006).
[15] D. M. Gaudiosi *et al.*, Opt. Express **14**, 9277 (2006).