Soft x-ray femtosecond coherent undulator radiation in a storage ring

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Abstract. We propose to produce femtosecond pulses of soft x-ray coherent undulator radiation in a storage ring for user pump–probe experiments using two energy exchanges between a picosecond relativistic electron bunch and two external ultra-short laser pulses. The coherent emission is generated thanks to the two laser–electron interactions that modulate the longitudinal charge distribution of the electron bunch at a harmonic of the laser wavelength, such as in the echo-enabled harmonic generation in free-electron lasers. Application to the SOLEIL storage ring in the soft x-ray range leads to coherent radiation and improvement of the flux of the photons by several orders in magnitude compared to the conventional slicing scheme. This is also accompanied by a significant enhancement of the signal-to-noise ratio.

With the recent advance of femtosecond (fs) spectroscopy \cite{1}, observing out-of-equilibrium molecular motion, disordered media and distorted crystal lattices in real time became possible. In the usual pump–probe technique, the pump (an ultra-short laser) creates a wave packet, the evolution of which reflects that of the ensemble of excited molecules that is probed by a second ultra-short laser pulse providing a spectroscopic signature that is further converted into structural dynamics. Extending the studies from diatomic molecules to larger and more complex
systems encounters some ambiguity with the conversion of the spectroscopic to structural information, requiring the use of an ultra-fast x-ray probe with techniques such as diffraction for crystals [2–5] or x-ray absorption spectroscopy for dilute and amorphous systems [6–10].

To reveal the structural dynamics in real time on an atomic time scale, intense stable fs x-ray sources are required. One can select from among laboratory sources either high-order harmonic generation from an intense laser focused in a rare gas [11], still limited to the order of 1 keV [12], or laser-generated plasma sources resulting from the emission of an intense laser beam onto a metal or liquid target [13], still with a rather low intensity at a high repetition rate [14].

Storage ring-based synchrotron light sources [15] provide tunable high-brilliance x-ray pulses thanks to large average current and very small electron beam emittance (i.e. small values of the horizontal and vertical root mean square (rms) beam sizes $\sigma_x, \sigma_y$ and divergences $\sigma_x', \sigma_y'$). The typical pulse length is directly linked to the electron bunch length $\sigma_z$ and is approximately tens of picoseconds long. The so-called ‘slicing’ technique [16], which is currently under operation at ALS [17], BESSY [18] and SLS [19] and is under preparation at SOLEIL [20], can produce sub-ps synchrotron radiation pulses. In this scheme, a resonant interaction of an electron beam with an external intense ultra-short laser pulse occurs in a region of periodic magnetic field (an undulator first harmonic being tuned at the laser wavelength). Its induces an energy modulation at the laser wavelength of the electron bunch that is several times as large as the rms beam energy spread $\sigma_E$. Along the bunch transport in the storage ring, where there is some dispersion (i.e. positions of electrons depending on their energy), this energy modulation is transformed into the modulation of an electron transverse coordinate or angle with an amplitude much larger than corresponding beam sizes ($\sigma_x, \sigma_x'$), and finally the radiation can be collected separately, giving synchrotron radiation from vacuum ultraviolet (VUV) to hard x-ray with approximately the same duration as the duration of the laser pulse. As only a fraction of the electrons participate in the sub-ps synchrotron radiation, the peak flux of photons is reduced by approximately a factor of 10. Moreover, the total number of photons in a sub-ps pulse is reduced proportionally to the pulse length, and the repetition rate of the sub-ps pulses is defined by that of the laser, which is significantly smaller than the bunch repetition rate in a typical storage ring. Thus, the price for obtaining sub-ps pulses in a storage ring is a significant loss in the average flux and brightness of the source. Another reason for the small flux is because of the incoherent nature of the electron emission resulting in a condition when the peak power of the radiation is proportional to the peak electron current. A substantial gain in photon flux is achieved with free-electron lasers (FELs) [15, 21–23] thanks to the coherent nature of the emission when the peak power of the radiation is proportional to the square of the peak electron current due to microbunching of the electrons. Compared to the self-amplified spontaneous emission (SASE) FELs that are currently under operation [23–25], seeded FELs show better temporal and spectral properties [26]. Since microbunching in the seeded FEL originates due to the electron beam interaction with an external laser, such a configuration is better adapted for pump–probe experiments due to intrinsic synchronization with the pump laser. The implementation of seeded FELs in storage rings has been demonstrated in the VUV range and below [27–30]. However, both in storage rings and in FELs, limitations arise at shorter wavelengths because of low microbunching efficiency at high harmonic of the laser frequency. The new proposed seeding technique called echo-enabled harmonic generation (EEHG) [31, 32] significantly improves efficiency for an electron microbunching at a high harmonic of the seeding laser.
A proof-of-principle experiment on linac has also been carried out at NLCTA [33] and at SDUV-FEL [34], proving the feasibility of this scheme, but still at long wavelengths.

In this paper, we propose to join EEHG with the storage ring and enable coherent undulator radiation (CUR) in the soft x-ray region providing higher photon flux within sub-ps x-ray pulses and larger signal-to-noise ratio compared to a ‘slicing’ source. The proposed technique is rather generic and can be implemented at most of the existing third-generation soft x-ray light sources.

A typical setup is shown in figure 1. Here the laser provides an initial light pulse with approximately 100 fs pulse width and a few mJ pulse energy. Then this pulse is divided into two pulses and each secondary pulse is fed into the corresponding undulators called modulator 1 and 2 after careful adjustment of the pulse energy and time delay. It is important that the arrival of the laser pulse in the modulator is synchronized with the arrival of the electron bunch. Thus, it is convenient to use one laser, although using two independent lasers even with two different frequencies is also possible. It is plausible that existing undulators can be adjusted to play the role of modulators. One additional piece of the entire scheme is the dispersive section that is used for a fine-tuning of the coherent radiation in the radiator, which is another undulator that often can also be chosen from one of the existing undulators. The evolution of the electron bunch density distribution in this scheme starts from a Gaussian distribution along the transverse directions $x, x', y, y'$ and along $p$, the energy difference with respect to the electron beam energy $E_0$ normalized to the rms energy spread $\sigma_E$. Since the electron bunch is much longer than the fs laser pulse, the electron bunch distribution $f$ can be considered as uniform along the longitudinal coordinate $z$ (taken in meters), i.e. $f(x, x', y, y', p) = \frac{N}{(2\pi)^{\frac{5}{2}}\sigma_x\sigma_{x'}\sigma_y\sigma_{y'}} \cdot \left(\frac{1}{\sigma_x}\frac{1}{\sigma_{x'}}\frac{1}{\sigma_y}\frac{1}{\sigma_{y'}}\right)$, with $N$ being the number of electrons per unit of length of the beam.

Table 1 shows the change in the coordinates taking place along the different steps and figure 3 illustrates the electron bunch distributions in the longitudinal phase space. Here we use beam parameters given in table 2 taken from SOLEIL [20] that are rather typical for all third-generation soft x-ray light sources.

The first laser–electron interaction in the
laser wavelength $\lambda_L$, which again modulates the electron energy at $\lambda_L$ in a length of the order of one $L_1 \times c$, with $L_1$ being the laser rms pulse width and $c$ the light velocity (figures 3(a) and (a')). The laser pulse of waist $w_1$ is transversely centered at the electron bunch center. Then, the electron bunch travels in the storage ring section where it experiences dispersion (step b): as the path taken by electrons depends on their energy, the induced laser energy modulation drifts in the longitudinal direction (figures 3(b) and (b')) and usually in the transverse directions (dispersion strengths being related to the transfer matrix coefficients $R_{ij}$ with $i, j = 1–6$). However, the structure after the transport (figure 3(b')) appears only if the optics is configured such that the transverse dispersion between the two modulators is zero or very weak ($R_{51}^{(1)} \simeq R_{52}^{(1)} \simeq 0$). Thus, from symmetric considerations ($R_{16}^{(1)} \simeq R_{26}^{(1)} \simeq 0$), the fs energy modulated electrons cannot be separated transversely from the ps electron bunch, like in the slicing scheme [16]. The longitudinal dispersion ($R_{56}^{(1)}$) should not be too strong to prevent the detailed structure to be destroyed by the energy fluctuations of amplitude $\Delta E$ introduced by incoherent synchrotron radiation (ISR) in bending magnets ($\Delta E^2 = \frac{55aE_0^2}{48\sqrt{3}} \frac{L}{R} \nu^2$, with $\alpha$ being the fine structure constant, $h$ the Planck constant, $\gamma = E_0/m_0c^2$ the normalized energy, $m_0$ the electron mass and $L$ and $R$ the length and radius of the bending magnet, respectively [35]). In the SOLEIL case, the transverse dispersion is canceled using a Chasman–Green lattice, and the longitudinal dispersion is decreased using an optimized additional chicane system (see figure 2). Then, the electrons are resubmitted to a second laser (step c) of waist $w_2$, which again modulates the electron energy at $\lambda_L$ (figures 3(c) and (c')). It can be seen from figure 3(b') that the phase information of the first laser is smeared out at the second laser stage, so that the process is insensitive to phase errors between the two lasers.

**Table 1.** Coordinate changes at each step of the proposed scheme.

| Step   | Expression                                                                 |
|--------|---------------------------------------------------------------------------|
| a      | $p = p + A_1 \cdot \exp\left(-\frac{z^2}{2\sigma_{E_1}^2}\right) \cos\left(\frac{2\pi z}{L_1}\right) \cdot \exp\left(-\frac{z^2}{2w_1^2}\right)$ |
| b      | $z \simeq z + p \cdot R_{56}^{(1)} \frac{\sigma_{E_1}}{E_0}$              |
| c      | $p = p + A_2 \cdot \exp\left(-\frac{z^2}{2\sigma_{E_2}^2}\right) \cos\left(\frac{2\pi z}{L_2}\right) \cdot \exp\left(-\frac{z^2}{2w_2^2}\right)$ |
| d      | $z \simeq z + p \cdot R_{56}^{(2)} \frac{\sigma_{E_2}}{E_0}$              |

**Table 2.** SOLEIL parameters used in this study.

| Parameter                        | Value   |
|----------------------------------|---------|
| Nominal energy $E_0$ (GeV)       | 2.75, 2.79 |
| Energy spread $\sigma_E$ (MeV)   |         |
| Bunch dimensions $\sigma_x$ (mm) | 10.5, 147, 33 |
| $\sigma_y$ (µm)                 | 10.0, 4.8 |
| Peak current $I_{\text{peak}}$ (A) | 134 |
| Storage ring damping time (ms)   | 3.3     |
| Radius $R$ (m) and length $L$ (m) | 5.39, 1 |
| Chasman length (m) and field (T) | 0.65, 0.7 |
| Modulator 1 and 2 period length (mm)/number | 150, 13 |
| Radiator period length $\lambda_u$ (mm)/number $N_u$ | 80, 19 |
| Laser wavelength $\lambda_L$ (nm)/energy (mJ) | 800, 5 |
| rms laser pulse length $\sigma_{L_1}$ (fs), $\sigma_{L_2}$ (fs) | 43, 118 |

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Figure 2. $R_{56}^{(1)} (\cdot)$ and $R_{51}^{(1)} (\cdot \cdot)$ values between the two modulators (modulator 1 output at $z = 0$ and modulator 2 input at $z \simeq 20$ m). Final $R_{56}^{(1)}$ of 4 mm. D: dipole; FQ and DQ: focusing and defocusing quadrupole. Dashed: additional chicanes.

Figure 3. Calculated electron bunch density in the longitudinal phase space $(z, p)$, using a linear six-dimensional (6D) macroparticule code including noise from ISR (a, b, c, d: at the scale of the laser pulse lengths; and $a', b', c', d'$: at the scale of the laser wavelength), after the first laser interaction (a, a'), after the first dispersive section (b, b'), after the second laser interaction (c, c') and after the second dispersive section (d, d'). Parameters: $A_1 = -5$, $A_2 = -2.95$, $R_{56}^{(1)} = -1.5$ mm, $R_{56}^{(2)} = -48 \mu$m, $\sigma_{L1} = 21$ fs, $\sigma_{L2} = 118$ fs, $w_1 = w_2 = 600 \mu$m ($A_2$ and $R_{56}^{(2)}$ chosen to optimize the bunching factor of the thirtieth harmonic). For the figure clarity, some parameter values are different from those in table 2.

Just after the second modulator (step d), the electron bunch passes through an adaptive dispersive section with a longitudinal dispersive strength of $R_{56}^{(2)}$ (figures 3(d) and (d')). With a proper set of parameters, the longitudinal charge distribution $\rho(z)$ of the electron distribution $f$ (with $\rho(z) = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} f(x, x', y, y', z, p) dx dx' dy dy' dp$) is modulated at a harmonic $k$ of the laser wavelength along the overlap of the two laser pulses [31, 32]. The second laser pulse length $\sigma_{L2}$ is chosen quasi-uniform at the $\sigma_{L1}$ scale to provide a good bunching all along $\sigma_{L1}$. Thus, these so-called ‘bunched electrons’ can emit in phase a train of independent sub-fs pulses, at a repetition rate of the laser frequency, in the tuned radiator and produce CUR at a harmonic number of the laser wavelength ($\lambda_r = \lambda_L/k$), with a duration near the first laser.
pulse duration, which is typically 100 fs full-width at half-maximum (FWHM). Jitter between the two lasers should be smaller than rms laser width, i.e. about 50 fs. The very weak transverse dispersion should permit us to get shorter pulse duration compared to the slicing scheme. After the energy spread enhancement due to the laser–electron interactions, the electron bunch naturally returns to its equilibrium state thanks to the synchrotron radiation damping, whose time scale (3.3 ms at SOLEIL) is of the same order as a standard laser repetition period (kHz). An alternative scheme for increasing the relaxing time is to change the electron bunch target at each laser pulse.

We now investigate down to which harmonic number of the laser wavelength the CUR is produced in the SOLEIL case [20]. The radiator parameters are those of the TEMPO beamline [20], covering 27.6–0.8 nm, corresponding to a harmonic number \( k \) of the 800 nm Ti:Sa laser wavelength between 29 and 967. The CUR on the \( k \)th harmonic results in the bunching factor \( b(k) \) defined as

\[
b(k) = \frac{1}{N} |\langle \rho(z) e^{ikz/\lambda L} \rangle|,
\]

with \( \lambda = R_{16}^{(i)} \frac{2\pi}{\lambda L} \sigma_\rho / E \) (\( i = 1, 2 \)) and \( J_k \) the Bessel function of order \( k \). Further optimization of equation (1) leads to the simpler expression \( b(k) \approx 0.39 \times k^{-1/3} \) [32], which provides an upper value of the bunching factor versus harmonic number, as shown in figure 4. This scaling is obtained with a fixed value of \( A_1 \) [32]. One of three other parameters \((A_2, R_{16}^{(1)}, R_{16}^{(2)})\) can also be fixed and the other two are tuned for each \( k \). \( b(k) \) is also calculated (with a 6D macroparticule

**Figure 4.** (a) The bunching factor \( b(k) \) just before the radiator versus the harmonic number \( k \) (calculated with a linear (−) and a nonlinear (−−) 6D macroparticule code over one laser wavelength) with \( R_{16}^{(1)} = -4 \) mm and \( A_1 = -5 \), and (⋯⋯) \( b(k) = 0.39 \times k^{-1/3} \). (b) Associated \( A_2 \) (−) and \( R_{16}^{(2)} \) (−−) for linear transport.
code, using equations (a) and (c) in table 1 to model the laser–electron interactions) considering the transverse dynamics, the noise induced by ISR [32], the finite laser pulse dimensions $\sigma_{L1}$, $\sigma_{L2}$, $w_1$, $w_2$ and a limitation imposed on the amplitude of the second energy modulation $A_2$, i.e. of 5$\sigma_b$ in the SOLEIL case (with a laser pulse energy of 5 mJ, $w_2 = 600 \mu m$ and $\sigma_{L2} = 118$ fs [36]). $b(k)$ computed with the linear terms in the transport decreases smoothly with $k$ towards a cut-off corresponding to $|A_2|$ reaching the limit value of 5 (figure 4(b)). The position of the cut-off strongly depends on the $R_{s6}^{(1)}$ value and is of $k \simeq 150 \left(\lambda_e \simeq 5.3 \text{ nm}\right)$ for $R_{s6}^{(1)} = -4$ mm. It can be pushed towards higher harmonics for higher $R_{s6}^{(1)}$ value but the noise induced by ISR plays a larger role as the phase-space structure becomes finer. Associated values of $A_2$ and $R_{s6}^{(2)}$ (figure 4(b)) have been chosen from equation (1), and the absolute value of $A_1$ is taken as 5 since a saturation of the bunching factor arises at about this value [32], $b(k)$ computed with the nonlinear terms (of drift sections, dipoles, quadrupoles and sextupoles) is slightly reduced and the cut-off appears for higher harmonics, in part because $A_2$ and $R_{s6}^{(2)}$ have been further optimized around the value given by the linear study.

In order to analyze the enhancement of the emitted power with respect to the slicing scheme, the CUR peak power $P_{\text{peak CUR}}$ is estimated with an analytical formula taken from [37] and further modified to take into account $\sigma_x$, $\sigma_y$, $\sigma_{\gamma'}$ and the transverse incoherent part of the radiation: $P_{\text{peak CUR}} = \pi a \hbar \omega_{\text{TrK}}^{3/2} [JJ]^2 \frac{b_{\text{peak}}}{c \epsilon} n_e b^2 \sqrt{\mathcal{J}_2}$, with $n_e = \frac{b_{\text{peak}} \lambda_b N_u}{c e}$ being the number of electrons within the slippage length $\lambda_b N_u$, $N_u$ the radiator period number, $f_2 = \left(\sigma_x \sigma_{\gamma'}\right) / \left(\sqrt{\sigma_x^2 + \sigma_y^2} \sqrt{\sigma_y^2 + \sigma_{\gamma'}^2} \sqrt{\sigma_x^2 + \sigma_{\gamma'}^2} \right)$, $\sigma_x = \sqrt{2 \lambda_b \lambda_u N_u / 4 \pi}$ and $\sigma_{\gamma'} = \lambda_u / 2 \lambda_u N_u$, respectively, the rms size and divergence of the undulator fundamental mode, $\omega = 2 \pi c / \lambda_x$, $e$ the electron charge, $K = \sqrt{4 \lambda_x \gamma^2 / \lambda_u - 2}$, the associated radiator magnetic field $B = K / (93.4 \lambda_u)$, $[JJ] = [J_0(x) - J_1(x)]$ and $x = \frac{k}{4 \pi c \epsilon x}$. At $k = 30 \left(\lambda_x = 26.7 \text{ nm}\right)$ with $b = 0.04$ (figure 4(a)), $P_{\text{peak CUR}} \simeq 120 \text{ kW}$. Assuming a CUR Gaussian emission, the energy per pulse is $P_{\text{peak CUR}} \times \sqrt{2 \pi \sigma_{L1}} \simeq 10 \mu J$.

Figure 5 shows the CUR peak power $P'_{\text{peak CUR}}$ calculated with GENESIS [38] along one laser wavelength with the previously optimized parameters. The second laser peak power and the $R_{s6}^{(2)}$ values are further adjusted to obtain a bunching factor around 4%. The output power $P'_{\text{peak CUR}}$ is about 91 kW, a value in good agreement with that found with the

**Figure 5.** Emitted power along the radiator at 26.7 nm (the 30th harmonic of the laser wavelength). Output power: $P'_{\text{peak CUR}} \simeq 91 \text{ kW}$. Parameters: the first (resp. second) laser peak power $= 8 \text{ GW} \left(0.9 \text{ GW}\right)$, waist of the two lasers $= 0.6 \text{ mm}$, $R_{s6}^{(1)} = 4 \text{ mm}$, $R_{s6}^{(2)} = 119.5 \mu \text{m}$.
analytical expression. In comparison, the power in the usual slicing case $P_{\text{peak ISR}}$, given by $P_{\text{peak ISR}} = \dot{N}_{\text{phot}} \hbar \omega \times \eta$, is 0.135 W at $\lambda_c = 26.7$ nm and for the planned relative bandwidth $\Delta \omega / \omega$ value of 0.05% (with $\eta$ being the percentage of electrons involved in the fs light pulse, typically $\eta = 0.1$ and $\dot{N}_{\text{phot}}(\omega) = \pi \alpha N_{\text{u}} \Delta \omega / \omega$). The power is increased by about six orders of magnitude with the two laser–electron interactions. The signal-to-noise ratio $S/N$, i.e. the fs CUR energy emitted by the bunched electrons compared to the ps ISR energy emitted by all the electrons in the bunch, is $S/N = \frac{P_{\text{peak CUR}} \times \sigma_{L1} \eta c}{P_{\text{peak ISR}} \times \sigma_z}$ $\approx 10^7$ (or $S'/N = \frac{P_{\text{peak CUR}} \times \sigma_{L1} \eta c}{P_{\text{peak ISR}} \times \sigma_z}$ $\approx 83$). Finally, we note that with $\Delta \omega / \omega = 0.05\%$, experimentally adjusted using a monochromator, all the CUR can be collected assuming a Fourier-transform-limited pulse, whereas a large part of the ISR is suppressed.

We have shown that the echo scheme can be applied to storage rings to achieve fs coherent undulator radiation until the soft x-ray region, of great interest for user applications. For the SOLEIL case presented here, for which practical design issues were addressed, the CSR extends down to 5 nm and provides about 120 kW at 27 nm, which is nearly six orders of magnitude higher than the power obtained under the same conditions with the slicing scheme, although slicing can be used with much higher photon energies. The extension of CSR toward shorter wavelengths could be achieved with lower beam energy and shorter laser wavelengths. Indeed at lower energy, noise effects from ISR are reduced and lower laser pulse energy is required since energy spread is reduced.

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