ENHANCING COHERENT HARMONIC GENERATION USING TILTED LASER WAVEFRONTS*

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Abstract

Coherent Harmonic Generation (CHG) to produce ultrashort pulses of synchrotron radiation is based on the interaction of relativistic electrons in a storage ring with femtosecond laser pulses in an undulator. The resulting periodic energy modulation is converted into a density modulation by a dispersive chicane, giving rise to coherent emission at harmonics of the laser wavelength in a second undulator. If the first undulator is in a section with non-zero dispersion, the density modulation can be enhanced using tilted laser wavefronts, thus delaying the phase-space distributions of electrons with different energy with respect to each other. The most simple way to produce a wavefront tilt would be a small crossing angle between the electron and laser beam. Details are discussed for the case of the CHG short-pulse facility at DELTA, a 1.5-GeV synchrotron light source at the TU Dortmund University, but HGHG and EEHG seeding of free-electron lasers could also be enhanced this way.

INTRODUCTION

Synchrotron radiation with short wavelength is the standard tool to study the structure of matter on the atomic level. However, synchrotron radiation pulses with a duration of 30 to 100 ps (FWHM) are insufficient to study dynamic processes such as chemical reactions, phase transitions, fast magnetic changes, lattice vibrations etc. which take place on the sub-picosecond scale. The femtosecond regime, on the other hand, has been made available by mode-locked lasers at wavelengths (e.g. 800 nm in the case of titanium-doped sapphire lasers) which are unsuitable to probe inner atomic shells or to provide spatial resolution on the atomic scale.

The need for radiation with short wavelength *and* short pulse duration has prompted new developments in laser physics, such as high-harmonic generation (HHG), as well as in accelerator physics, notably free-electron lasers (FELs) providing extremely brilliant short-wavelength radiation with femtosecond pulse duration. To date, only four linacbased FEL facilities at short wavelengths are in user operation (in chronological order: FLASH, LCLS, SACLA, and FERMI) while more than 50 synchrotron light sources worldwide [1] provide up to 40 beamlines simultaneously with brilliant and tunable radiation. It is therefore worthwhile to study methods which allow to generate conventional synchrotron radiation with shorter pulse duration.



Figure 1: Setup for seeding schemes: a) HGHG or CHG and b) EEHG with respective electron distributions in phase space, i.e. longitudinal coordinate *z*, here normalized to the laser wavelength λ , versus relative energy deviation $\Delta E/E$.

SHORT-PULSE GENERATION

Some methods to generate sub-ps radiation pulses at storage rings are borrowed from FEL seeding schemes using a femtosecond laser pulse to modulate the energy of electrons at the center of a long electron bunch. FEL seeding aims at assisting a positive feedback loop of exponentially growing microbunching and coherent radiation. In a storage ring, on the other hand, the laser pulse is used to define a short "slice" within the electron bunch. A short radiation pulse is emitted by the energy-modulated electrons together with a long pulse from the rest of the bunch. Off-energy electrons can be transversely displaced by dispersion in order to separate the short and long components of incoherent undulator radiation spatially - this scheme is known as "femtoslicing" [2]. Alternatively, a magnetic chicane may convert the energy modulation into a density modulation giving rise to a short pulse of coherent radiation at harmonics of the laser wavelength, which is brighter than the long incoherent pulse. As long as the signal-to-background ratio

$$\frac{P_{\text{short}}}{P_{\text{long}}} = \frac{n_{\text{short}}^2 b_h^2}{n_{\text{long}}} = f^2 n_{\text{long}} b_h^2 \quad \text{with} \quad f \equiv \frac{n_{\text{short}}}{n_{\text{long}}} \quad (1)$$

is tolerable, no geometric separation is required. Here, $f \approx 10^{-3}$ is the ratio between the number of electrons in the slice

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and in the bunch, and b_h is the bunching factor given by the Fourier transform of the longitudinal charge distribution at the *h*th harmonic of the laser wavelength. With $n_{\text{long}} = 10^{10}$, to give an example, $b_h = 0.1$ would yield an excellent signalto background ratio of 10^2 .

The standard scheme of short-pulse generation by laserinduced energy modulation in an undulator (the "modulator"), microbunching in a magnetic chicane, and coherent emission in a second undulator (the "radiatior") is known as coherent harmonic generation (CHG, top part of Fig. 1) [3] and bears a close similarity to an FEL seeding scheme called high-gain harmonic generation (HGHG) [4]. Here, the bunching factor decreases with increasing harmonics as $b_h \sim \exp(-h^2)$. In the same way, echo-enabled harmonic generation (EEHG, bottom part of Fig. 1) [5] involving a two-fold energy modulation in order to reach higher harmonics with $b_h \sim h^{-1/3}$ can be used for FEL seeding as well as for short-pulse generation in a storage ring.

TILTED LASER WAVEFRONTS

A primary goal in using CHG or EEHG is to reach high harmonics, i.e. short wavelengths, to extend the range of scientific applications of the short-pulse source. One of the limitations is the finite length of the microbunches given by the sinusoidal nature of the energy modulation and by the electron energy spread σ_E . It may be possible to linearize the energy modulation in a sawtooth-like fashion by seeding with several laser harmonics simultaneously [6, 7], but a given laser pulse energy is usually better invested in obtaining a high modulation amplitude $\Delta E_{\text{max}}/\sigma_E$ rather than in additional harmonic generation. The effect of the energy spread can be reduced if there is a correlation between the energy offset ΔE of each electron and another parameter. The obvious candidate for such a parameter is the transverse coordinate x in the presence of dispersion D(s). In a storage ring, dispersion is introduced in the horizontal plane by the dipole magnets and its value at a particular position, e.g. at the CHG or EEHG modulator, depends on the quadrupole settings. The ΔE -x correlation is to good approximation linear, i.e. $x = D \cdot \Delta E/E$, but is smeared out by the emittance-dependence of the horizontal beam size

$$\sigma_x(s) = \sqrt{\varepsilon_x \beta_x(s) + D^2(s) \sigma_E^2 / E^2},$$
 (2)

where ε_x is the horizontal beam emittance and $\beta_x(s)$ is the beta function. Reference [8] suggests to use an undulator with transverse gradient as modulator in which the phase advance with respect to the laser field depends on the transverse position. Another and even more simple possibility to make use of the ΔE -x correlation is to seed with tilted laser wavefronts as recently proposed by [9] for FEL seeding. This way, a phase shift of the energy modulation is introduced which depends on x and thus also on ΔE .

An example is shown in Fig. 2 with D = 0.5 m, $\sigma_E/E =$ $8 \cdot 10^{-4}$, $\varepsilon_x = 15$ nm rad, and $\beta_x = 2$ m. In this case, the optimum wavefront tilt is 0.1 mrad and the bunching factor is roughly doubled.



Figure 2: Energy and density modulation without (left) and with (right) wavefront tilt. The top figures show the phase space distribution, the bottom figures the longitudinal electron density. Electrons with an original energy deviation of $\Delta E > \sigma_E$ (and $\Delta E < -\sigma_E$) are shown in red (and blue).

Table 1: Parameters of the Storage Ring DELTA

beam energy	1.5 GeV
circumference	115.2 m
beam current (multibunch)	130 mA
beam current (single bunch)	20 mA
horizontal emittance	15 nm rad
min. horizontal beta function @CHG	2 m
min. vertical beta function @CHG	20 m
horizontal dispersion @CHG	-0.1 m
relative energy spread	0.0008
bunch length (rms)	13 mm

CHG AT DELTA

The 1.5-GeV electron storage ring DELTA with parameters as shown in Table 1 is operated as a synchrotron light source by the TU Dortmund University. In 2011, a shortpulse facility based on CHG was constructed in order to provide ultrashort coherent synchrotron radiation pulses in the VUV and THz regimes for users [10, 11], and an extension based on EEHG including a femtoslicing source is planned [12]. The bunch length corresponds to a pulse duration of 100 ps (FWHM) while the CHG pulse length is below 100 fs. For a typical single-bunch current of 10 mA, the number of electrons is $N_{\text{long}} = 2.4 \cdot 10^{10}$ and the ratio f (cf. Eq. 1) is below 10^{-3} . Seeding is presently performed using Ti:sapphire laser pulses at 800 nm or frequency-doubled pulses thereof, and a signal-to-background exceeding 10^2 is routinely observed for low harmonics [10]. The beam size is 20 $\sigma_x \approx 170 \,\mu\text{m}$ according to the first term of Eq. 2 while the second term with D = -0.1 m in the CHG section amounts only to 80 µm.

and



Figure 3: Bunching factors calculated for CHG at the 5th harmonic with the beam parameters of DELTA and different values of dispersion D at the modulator. The laser field is assumed to consist of plane waves arriving under a tilt angle α with respect to the electron beam.

The example shown in Fig. 2 was based on the DELTA parameters except for an increased dispersion value of |D| = 0.5 m, which can in principle be achieved by modifying the quadrupole settings of the storage ring, and work to this end is in progress in order to perform an experimental investigation of the tilted-wavefront effect. Calculated bunching factors for different dispersion values are shown in Fig. 3 as function of the tilt angle. As expected, the improvement with |D| = 0.1 m is marginal while an experiment with |D| = 0.3 m might already yield an observable effect.

A wavefront tilt can be obtained by simply introducing a small crossing angle between the electron and laser beam axes. A tilt produced by other means, e.g. by passing through a prism, will result in a wavefront rotation at the laser waist [13] which is not desired here. A crossing angle of the order of 0.1 mrad is well controllable by the motorized mirrors upstream of the modulator (an electromagnetic undulator with 7 periods of 25 cm length) and would result in an offset of 90 μ m at either end of the modulator, which is small compared to the laser beam size. A variation of the incident laser beam angle presently exhibits a broad maximum of the CHG yield. With larger dispersion, the tilted-wavefront effect is expected to result in a more pronounced optimum angle.

In CHG operation, coherent THz radiation is generated over several turns due to the fact that the energy-modulated electrons leave a gap in the longitudinal charge distribution [14]. This gap gives rise to coherent radiation at wavelengths above $\sim 50 \,\mu\text{m}$ and should be insensitive to the sub- μm effects of titled wavefronts. Since the absolute crossing angle is not easily determined, an offset between the optimum laser angle for CHG and for THz may be the most obvious signature for the tilted-wavefront effect for a fixed dispersion value. If *D* can be tuned on the fly, an improvement of the CHG signal with increasing dispersion should be observed.



Figure 4: Phase shift of energy modulation for a transverse displacement of $\Delta x = 0.1$ mm and a wavefront tilt angle of $\alpha = 0.1$ mrad as function of longitudinal coordinate *z*, normalized to the Rayleigh length z_R , for the case of a plane wave (blue) and a Gaussian laser waist at z = 0 (red).

MORE REALISTIC WAVEFRONTS

Distortions of the laser wavefronts have not been studied so far but are likely to occur due to the passage of the laser pulses through air and glass (lenses and vacuum windows). Furthermore, even idealized wavefronts of a Gaussian laser beam are curved. Up to now, plane waves were tacitly assumed leading to a constant phase shift of the energy modulation given by

$$\Delta \varphi = \frac{2\pi \, \alpha}{\lambda} \Delta x \tag{3}$$

for a transverse displacement Δx , a tilt angle α , and a laser wavelength λ . For a Gaussian beam, the wavefront is flat at the center of the laser waist, such that Eq. 3 holds, but nearly spherical in the far field, where $\Delta \varphi$ reduces to zero. In between, the radius of curvature at a distance z from the waist is

$$R(z) = z + \frac{z_R^2}{z}$$
 with $z_R \equiv \frac{\pi w_0^2}{\lambda}$ (4)

in paraxial approximation [15], where z_R is the Rayleigh length and w_0 is the $1/e^2$ waist radius. As shown in Fig. 4, the resulting phase shift decreases with increasing distance z. At $z = \pm z_R$, for example, it is only half of the maximum value. Thus, making use of the tilted-wavefront effect may require to increase the Rayleigh length which corresponds to reducing the electric field acting on the electrons. According to theory [16], the Rayleigh length for maximum energy modulation on the beam axis is about 1/4 of the modulator length, but this condition will usually be relaxed due to the finite electron beam size. For the case of DELTA, an optimized value of z_R is 1/2 of the modulator length or even larger [12].

The reduced phase shift due to curved laser wavefronts can be compensated by an increased crossing angle. Since the phase shift is now a function of the position z along the modulator, the improvement of the bunching factor depends on several parameters, such as the undulator length, the Rayleigh length, the tilt angle etc., and its prediction requires an integration of the laser-electron energy exchange over z. Furthermore, the electric field of the laser pulse and thus the amount of energy modulation is a function of all coordinates. If the chicane is set for optimum bunching at the center of the laser pulse, electrons at any other longitudinal or transverse position will contribute less to the coherent emission of radiation.

SUMMARY

A crossing angle between laser and electron beam can improve the bunching factor for FEL seeding or for the generation of ultrashort pulses in a storage ring via CHG or EEHG, provided the modulator is placed in a section with significant dispersion. The smaller the beam emittance, the larger is the effect of tilted wavefronts for a given dispersion value. It may therefore be advantageous to introduce vertical dispersion. In the case of DELTA, however, vertical dispersion would require additional magnets while horizontal dispersion can be created by changing the quadrupole setting. For this and other reasons (larger vertical beta function, larger horizontal aperture), first experimental investigations of the titled-wavefront effect will be under horizontal crossing angles only. General statements on the result can only be given in the crude approximation of plane waves and an infinite laser beam size. More realistic predictions require a thorough simulation and are only valid for a particular set of parameters.

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REFERENCES

- For a list of synchrotron radiation sources see e.g. http://www.lightsources.org
- [2] A.A. Zholents and M.S. Zolotorev, "Femtosecond X-Ray Pulses of Synchrotron Radiation", *Phys. Rev. Lett.* 76, p. 912, 1996.
- [3] R. Coisson and F.D. Martini, "Free-electron relativistic scatterer for UV-generation", in *Physics of Quantum Electronics IX*, edited by S. F. Jacobs et al., Addison-Wesley, 1982.

- [4] L.H. Yu, "Generation of Intense UV Radiation by Subharmonically Seeded Single-Pass Free-Electron Lasers", *Phys. Rev.* A 44, p. 5178, 1991.
- [5] G. Stupakov, "Using the Beam-Echo Effect for Generation of Short-Wavelength Radiation", *Phys. Rev. Lett.* 102, p. 074801, 2009.
- [6] D. Ratner, A. Chao, "Seeded Radiation Sources with Sawtooth Waveforms", *Proc. 33th Free-Electron Laser Conf.*, Shanghai/China, 2011, pp. 53-56.
- [7] G. Stupakov, M.S. Zolotorev, "Using Laser Harmonics to Increase Bunching Factor in EEHG", *Proc. 33th Free-Electron Laser Conf.*, Shanghai/China, 2011, pp. 45-48.
- [8] H. Deng, C. Feng, "Using Off-Resonance Laser Modulation for Beam-Energy-Spread Cooling in Generation of Short-Wavelength Radiation", *Phys. Rev. Lett.* 111, p. 084801, 2013.
- [9] C. Feng, T. Zhang, H. Deng, Zh. Zhao, "Three-dimensional manipulation of electron beam phase space of seeding soft x-ray free-electron lasers", *Phys. Rev. ST - Accel. Beams* 17, p. 070701, 2014.
- [10] S. Khan, F. Bahnsen, S. Cramm, S. Döring, J. Greve, M. Höner, H. Huck, M. Huck, R. Molo, P. Ungelenk, L. Plucinski, A. Schick, C. M. Schneider, "Generation of Ultrashort and Coherent Synchrotron Radiation Pulses at DELTA", *Sync. Rad. News* 26(3), p. 25, 2013.
- [11] A. Meyer auf der Heide, S. Hilbrich, M. Höner, H. Huck, M. Huck, S. Khan, C. Mai, R. Molo, H. Rast, A. Schick, P. Ungelenk, "Coherent Harmonic Generation at the DELTA Storage Ring: Towards User Operation", in these Proceedings: *Proc. 36th Free-Electron Laser Conf.*, Basel/Switzerland, 2014, TUP082.
- [12] S. Hilbrich, M. Höner, H. Huck, M. Huck, S. Khan, C. Mai, A. Meyer auf der Heide, R. Molo, H. Rast, A. Schick, P. Ungelenk, "Upgrade Plans for the Short-Pulse Facility at DELTA", in these Proceedings: *Proc. 36th Free-Electron Laser Conf.*, Basel/Switzerland, 2014, MOP087.
- [13] S. Akturk, X. Gu, P. Bowlan, R. Trebino, "Spatio-Temporal Couplings in Ultrashort Laser Pulses", J. Opt. 12, p. 093001, 2010.
- [14] P. Ungelenk, M. Bakr, M. Höner, H. Huck, S. Khan, R. Molo, A. Nowaczyk, A. Schick, M. Zeinalzadeh, "Recent Developments at the DELTA THz Beamline", *Proc. Int. Part. Accel. Conf.*, New Orleans/LA/USA, 2012, pp. 768-770.
- [15] A.E. Siegman, "Lasers", University Science Books 1986.
- [16] A.A. Amir, Y. Greenzweig, "Three-dimensional Theory of the Free-Electron Laser", *Phys. Rev.* A 34, p. 4809, 1986.