X-RAY FREE-ELECTRON LASERS AND ULTRAFAST SCIENCE AT THE ATOMIC AND MOLECULAR SCALE *

Claudio Pellegrini, UCLA, Los Angeles, CA 90095, USA .

Abstract

Generation of coherent radiation at nanometer and subnanometer wavelength with free-electron lasers (FELs) is a subject of great current interest, as shown by the many FEL being developed or built in Asia, Europe and the USA. This paper reviews the theoretical and experimental status of X-ray and Soft X-ray FELs. After a presentation of the main characteristics of LCLS, the X-ray FEL now under construction at SLAC, we discuss the basic physical properties of short wavelength, single pass FELs. We show that the experimental results obtained from the late 1990's to today are in remarkable agreement with the theory. The final part discusses some of the directions for further developments.

INTRODUCTION

The present interest in X-ray and soft X-rays FELs, at wavelengths from about 0.1 nm to a few nanometers, and a few to 100 nm, is motivated by their characteristics of tunability, coherence, high peak power, short pulse length. X-ray FELs can explore matter at the length and time scale of atomic and molecular phenomena, the Bohr atomic radius, about 1 Å, and the Bohr period of a valence electron, about 1 fs. The peak brightness of soft X-rays and X-ray FELs is about ten orders of magnitudes larger than that of synchrotron radiation sources. The characteristics of FELs are also superior to those of plasma based lasers sources.

The large number of coherent photons/pulse and short pulse duration of short wavelength FELs open the door to:

a. Single shot measurements of the structure of complex molecules, like proteins, and nanoscale systems;
b. Study of non linear phenomena;
c. Study of high energy density systems;
d. Studies of inner levels atomic physics.

Short wavelength FELs will allow the study of the properties matter at the atomic and molecular level with unprecedented space-time resolution.

FELs operating in the visible or infrared spectral region are mainly designed and built as oscillators, amplifying the radiation stored in an optical cavity with a train of electron bunches. Short wavelength FELs are instead single pass devices, because of the difficulties inherent in building a low loss optical cavity in the short wavelength region, and of the problems associated with handling the very large power density at the mirrors.

After many years of research and development, going as far back as the 1980s, the first X-ray FEL operating in the 0.15 to 1.5 nm wavelength range, LCLS [1], proposed in 1992 [2], is now being built and will be completed by 2009.

LCLS main electron beam characteristics are: energy, E ~ 14 GeV, peak current, I_{peak} ~ 3.4 kA, normalized emittance ~ 1.2 mm mrad, pulse duration~100 fs. The LCLS beam will be the brightest ever produced. The undulator length is about 130 m. The peak characteristics of LCLS radiation are given in Table 1.

X-ray FELs in Japan and Korea will follow shortly after LCLS. A European X-ray FEL is being developed with a target date of 2012. Peak characteristics of X-ray pulses for these FELs are similar to those of LCLS. The average brightness and power of X-ray FELs is related to the choice of the electron linac technology. LCLS and the Korean project use a warm temperature S-band linac, the Japanese project a warm C-band linac. The European X-FEL uses a superconducting linac in band L.

More FELs operating from a few to 100 nanometers are being designed and built in Asia, Europe and the US, and are discussed in these Proceedings. A single pass SASE-FEL, Flash, in the wavelength region 100 to 10 nm, built at DESY as a user facility and an X-ray FEL prototype, is already in operation. Some of the results from FLASH will be discussed later in this paper.

Table 1: X-FEL typical characteristics

Wavelength (fundamental)	1.5	0.15	nm
Undulator period/parameter	3/3.5		cm
Undulator length	130		m
Peak saturation power	4	8	GW
Pulse length, FWHM	140	76	mm
Coherent photons per pulse	10.6	1.1	x 10^{12}
Peak brightness	0.28	15	x 10^{32*}

*Brightness measured as number of photons/s/mm^2/mrd^2/0.1%band-width.

The FEL radiation is transversely coherent. While in a typical 3rd generation synchrotron radiation source there is less than one photon in a coherent volume, in LCLS there are more than 10^9. The energy of coherent photons can be pooled together to create multi-photons excitations and carry out non-linear X-ray experiments, a largely unexplored area of science.

In this paper we summarize the physical properties of short wavelength, single pass FELs, and review the main experimental data obtained until now. These results are in

---

*Work supported by the USDOE.

very good agreement with our theoretical understanding down to a wavelength of about 10 nm, and give us confidence that FELs in the sub-nanometer region will operate according to their design.

FEL PHYSICS

FEL gain and saturation

Consider an electron beam moving along the axis of an undulator magnet and executing an oscillation transverse to the direction of propagation. An electromagnetic wave co-propagates with the beam and modulates its energy. Each electron in the beam radiates a field, which is added to the initial field. The total field acts on other electrons, establishing a collective interaction. The interaction produces a transition of the beam to a novel state, consisting of micro-bunches separated by the radiation wavelength, and emitting coherent radiation with large intensity [3].

Let λ_w, B_w, N_w be a helical undulator period length, magnetic field and number of periods. Consider a single electron. When going through the undulator it emits a wave train with N_w waves, at a wavelength

$$\lambda = \lambda_W (1 + K^2 + \gamma^2\theta^2) / 2\gamma^2 \qquad , \qquad (1)$$

where γ is the beam energy in rest mass units, θ is the angle between the undulator axis and the direction of observation of the radiation, and $K = eB_W \lambda_W / 2\pi mc^2$ is the undulator parameter. The definition of K and some of the other formulae that follow must be slightly modified in the case of a planar undulator [3]. The line width is $\Delta\lambda / \lambda \approx 1 / N_W$.

Because of the angular dependence of the wavelength, the "coherent angle", corresponding to a line width $1/N_W$, is $\theta_C = \sqrt{\lambda / \lambda_W N_W}$, and the effective, diffraction limited, source radius is $a_C = \sqrt{\lambda\lambda_W N_W}$, so that $a_C\theta_C = \lambda / 4\pi$. For an X-ray FEL we have typically $a_C \approx 10\mu m,\ \theta_C \approx 1\mu rad$.

The average number of coherent photons/electron in the coherent solid angle and line-width is

$$N_{Ph} = \pi\alpha K^2 / (1 + K^2) \sim 0.01 \qquad (2)$$

where α is the fine structure constant. The production of coherent photons by one electron is a very inefficient process.

Consider now an electron beam. In the initial state, when entering the undulator, the electrons have a random longitudinal position. The wave trains from each electron superimpose with random phases, and the intensity is proportional to the number of electrons, the physical situation described as spontaneous radiation.

However, if the undulator is long enough, the radiation emitted by the electrons can act on other electrons in the beam, modulating their energy on a scale length equal to the radiation wavelength. This energy modulation is then transformed into a longitudinal density modulation in the undulator magnetic field. Because of the density modulation more electrons emit in phase, leading to a larger intensity at the wavelength (1). The larger intensity produces a larger energy modulation, establishing a feedback loop. As a result the intensity and the density modulation grow exponentially until a saturation level is reached.

The process is characterized by the FEL parameter [4]

$$\rho = [(K / 4\gamma)\Omega_P \lambda_W / 2\pi c]^{2/3} \quad , \qquad (3)$$

where Ω_P is the beam plasma frequency. Typical values of the FEL parameter for Soft X-ray or X-ray FELs are in the range 10^{-3}-10^{-4}. The exponential growth rate, saturation power and undulator saturation length are given, in this simple 1-D model by:

$$L_G = \lambda_w / 4\pi\rho \qquad (4)$$

$$P_{Sat} = \rho I_{Peak} E \qquad (5)$$

$$L_{Sat} \simeq 10 L_G \qquad (6)$$

where I_{Peak}, E are the beam peak current and energy.

The number of coherent photons per electron of energy $E_{Photon} = hc / \lambda$ at saturation is $N_{Ph} = \rho E / E_{Photon}$. For E_{Photon}=10keV, E=15 GeV, $\rho=10^{-3}$, the number of coherent photons is about 10^3, a gain of 5 orders of magnitude respect to the spontaneous radiation case. It is important to notice that there is room for further increase of the number of photons, to reach the limit of intensity proportional to the square of the number of electrons, going beyond saturation by a factor of about 10^4.

The exponential growth occurs if some conditions on the beam emittance, ε, and energy spread, σ_E, are satisfied:

$\sigma_E < \rho$, cold beam condition,

$\varepsilon \approx \lambda/4\pi$, phase space matching.

An additional condition is that the losses of coherent radiation due to diffraction must be less than the gain, to obtain gain guiding. are in band S for, and band C for the These conditions require very high brightness, high peak current beams. X-ray FELs are pushing the science of beam generation, acceleration and control to a new level of sophistication.

Slippage, Cooperation Length, Time Structure

The radiation propagates faster than the electron (it "slips" by λ per undulator period); thus electrons communicate only with the ones in front of them; the total slippage $S=N_w\lambda$ is also the wave train length.

The cooperation length, [5], is the slippage in one gain length, $L_C = \lambda / 4\pi\rho$, and defines the radiation longitudinal coherence.

When the FEL starts from spontaneous radiation, a noisy signal, the radiation is "spiky", and the number of "spikes" is the bunch length/$2\pi L_c$. This is called a SASE-FEL. Since in this case the process starts from noise the intensity in each spike fluctuates, according to a negative exponential, as for a thermal source. When many spikes are present in the final radiation pulse the intensity distribution becomes a Gamma function distribution [6], and the rms fluctuation width decreases as the square root of the number of spikes.

Figure 1 shows the calculated evolution of the temporal structure of the LCLS intensity within the fundamental line, from the initial spontaneous radiation noise, to the development of the spikes. The figure shows a part of the bunch and a few spikes, but is representative of the whole bunch, with about 200 spikes.

For the LCLS case we have: L_c=0.04 mm; spike length ~ 0.3 µm (1 fs); $\Delta\lambda/\lambda$ ~ 3x10^{-4}; spike number ~ 200. The expected intensity fluctuation is about 7%, if the fluctuations due to beam shot to shot changes in energy, charge and peak current can be reduced below this value.

Reduction of line width: seeding, self seeding and harmonics

Instead of starting from noise, as in SASE, the FEL can be used as an amplifier, seeded by an external laser field, at the wavelength (1). If the laser seed produces a beam energy modulation larger than that due to the spontaneous radiation, and the laser pulse is longer than the electron pulse and has a transform limited line width, then one can expect the FEL pulse to be also transform limited, with a line width smaller than in the SASE case. In the LCLS case this would give, for the same bunch length, a line-width smaller, respect to SASE, by a factor equal to the number of spikes, or $\Delta\lambda/\lambda \sim 4\ 10^{-6}$.

An option available when there is no external laser seed is self-seeding, using a first undulator followed by a monochromator to produce the radiation field needed to seed the FEL in a second undulator [7]. The resulting line width is defined by the monochromator resolution. The self-seeding scheme will be tested in the near future at FLASH. It is expected to increase the peak brightness by about one to two orders of magnitude.

Another option is the generation and amplification of harmonics. The FEL can generate and amplify harmonics of (1) [8]. Bonifacio and his group [9] considered the possibility of using harmonics to operate an FEL at short wavelengths, using a cascade of undulators tuned at the fundamental and harmonics. Yu and collaborators proposed the High Gain Harmonic Generation (HGHG) FEL[10], using both harmonics and high gain, an approach which is being considered now by many groups for soft X-ray FELs.

In HGHG the beam is modulated in energy at a wavelength lambda, and the energy modulation is transformed into a longitudinal density modulation in a dispersive section. The density modulation is rich in harmonics, and if the beam is sent through an undulator tuned at the wavelength lambda/n it will produce and amplify coherent radiation at the harmonic. The process can be repeated in a cascade scheme. The radiation has at each step a large amplification. For the processes to be effective the initial energy modulation must be small enough to prevent gain losses in the second or higher stages. The beam peak current and emittance needed for large gain at the final wavelength is comparable to what is needed to amplify directly with high gain at the final wavelength.

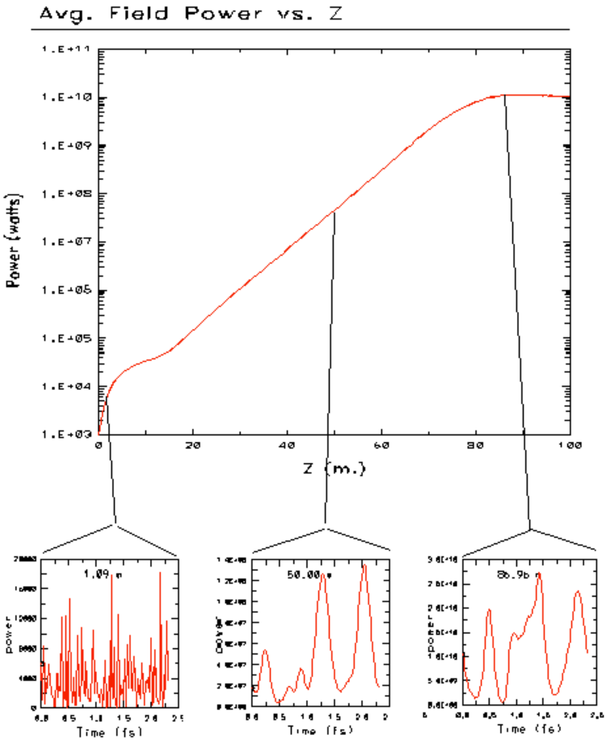

Figure 1 Evolution of the radiation intensity along the bunch, the temporal structure, from the initial noise to the spike structure, evaluated for LCLS [1] .

The self-seeding and HGHG systems are sensitive to beam energy jitter, which can lead to large intensity fluctuations. The effect of energy variation along the electron bunch, including quadratic and cubic terms, on the minimum line width has been studied recently for the Fermi FEL [11]. The results show that an energy variation along the bunch limits the minimum line width to a value larger than the transform limit. A new mode of operation of the Fermi photoinjector gun-linac system to minimize these effects is also suggested in [11].

The line width reduction, and the consequent increase in brightness, obtainable with self-seeding and HGHG is comparable.

Short pulse production

Many ideas have been proposed to reduce the X-ray pulse length: reducing the electron bunch length; chirping the electron beam energy and the radiation pulse wavelength and then selecting part of the bunch with a monochromator in a two undulator system; or producing a local increase of the electron bunch emittance and/or energy spread so that only part of the bunch large gain.

The emittance spoiler method has been proposed and investigated in reference [12], and can produce few fs long pulses. It uses a foil with a slit, located in the middle of a chicane, where a correlated energy chirp is transformed into a transverse electron distribution. Only the part of the bunch going through the slit maintains a small emittance and has large gain. The part of the bunch going through the slit corresponds to a small part of the bunch in the longitudinal distribution, and thus has a short duration.

The two-undulator chirped-pulse system is described in reference [13], and can reach an X-ray pulse length around 10 fs. This scheme is similar to the self-seeding system for line width reduction. In this case one uses an energy chirped electron beam, with a chirping larger than the gain band width. A monochromator is used to select a part of the radiation pulse. The selected part is then amplified again in the second undulator.

Enhanced SASE, proposed by Zholents [14], is another interesting option, The energy of the electron bunch is modulated by an external laser interacting with it in an undulator. This modulation is transformed in microbunching at the external laser wavelength prior to entering the undulator. The microbunching increases the local peak current and the gain. The X-ray pulse duration is controlled by changing the duration of the external laser pulse, down to the femtosecond level.

Wakefields in the linac and/or the undulator can be used to produce a sharp peak in a part of the electron bunch longitudinal distribution [15]. Lasing will occur only near the peak. FLASH can operate in this mode and has produced soft X-ray pulses as short as 20 fs [16].

Another concept [17] for production of high power coherent attosecond X-ray pulses is based on generation of harmonics in a multistage HGHG FEL starting from shot noise. Single-spike phenomena occurs when the electron bunch goes through a sequence of undulators. The statistical properties of the SASE FEL high-harmonic radiation are used to select radiation pulses with a single spike.

EXPERIMENTAL RESULTS FOR SINGLE PASS FELS

When the LCLS was first proposed in 1992 the high gain, spikes and other parts of SASE theory and harmonic generation had not been verified experimentally. The first results showing high gain were obtained in the microwave region [18] [19] [20], the only wavelength region that could be studied with the beam characteristics available at that time. The situation changed in the late 90s. Beams produced by photoinjector guns allowed FELs to operate in the infrared region, measuring high gain SASE [21], including intensity fluctuations due to spikes [22]. The experiment reported in [22] was the first to achieve large gain, $3x10^5$, demonstrating the real possibility to use SASE for short wavelength, high peak power FELs. The next step was to reach saturation and even shorter wavelengths. Both objectives were reached in SASE FELs at Argonne [23], Desy [24] and by VISA [25], a UCLA-SLAC-BNL collaboration, at wavelengths from 0.8 to about 0.1 μm. More recently the FLASH group reached saturation at 32 nm, and lasing at a wavelength as short as 13 nm [16].

Experiments have verified other aspects of the theory, besides gain and saturation. Microbunching of the electrons at the radiation wavelength was observed through the detection of coherent transition radiation [26], [27]. Harmonic generation was observed at VISA, [28] and LEUTL [23]. Transverse coherence of the SASE radiation was measured on FLASH at about 100 nm [29] and more recently at 32 nm [25].

The FLASH results also demonstrate the possibility of producing very short, about 20 fs, pulses, using wakefields and collective effects to shape the electron bunch, and obtain large peak current in a limited part of the bunch.

HGHG operation has been demonstrated at Brookhaven [30], using an 800 nm seeding pulse -longer than the electron bunch- to induce an energy modulation slightly larger than the intrinsic energy spread. The dispersive section converts it into a current modulation, and its third harmonic, at 266 nm, provides the seeding signal, which is then amplified. Saturation has been reached in the 3rd harmonic. Chirped pulse amplification in HGHG has been demonstrated more recently [31].

CONCLUSION

The great progress in the physics and technology of high brightness electron beams, and the exploitation of the FEL collective interaction, has made possible to design and build powerful X-ray FELs in the 1Å spectral region, opening the way to new opportunities to explore the properties of matter at the atomic length and time scale.

R&D work should be continued in many areas -like high brightness electron sources, beam stability, diagnostics, undulators, X-ray optics, synchronization of the X-ray probe pulse with a pump pulse, short fs pulses, higher peak power- to increase even more the future capability of single pass FELs.

REFERENCES

[1] "LCLS Conceptual Design Report", Stanford Linear Accelerator Center, SLAC-R-593, 2002.

[2] C. Pellegrini, "A 4 to 0.1 nm FEL based on the SLAC linac," in Proc. Workshop IV Generation Light Sources, M. Cornacchia and H. Winick eds., 1992, p. 364.

[3] For a comprehensive review of single pass FELs see C. Pellegrini and S. Reiche, "The development of X-ray Free-electron Lasers", IEEE J. of Selected Topics in Quantum Electronics, vol. 10, pp. 1393-1404, 2004; for a general FEL review see C. Pellegrini and S. Reiche, "Lasers, Free-electron", in Optics Encyclopedia, p. 1111-1134, Wiley-VCH Verlag, Berlin, 2003.

[4] R. Bonifacio, C. Pellegrini, and L. Narducci, “Collective instabilities and high-gain regime in a free electron laser,” Opt. Comm., vol. 50, pp. 373–378, 1984.

[5] R. Bonifacio et al., “Spectrum, temporal structure, and fluctuations in a high gain FEL starting from noise”, Phys. Rev. Lett. 73, pp. 70-73 (1994).

[6] E. L. Saldin, E.A, Schneidmiller and M. V. Yurkov, “Statistical Properties of the radiation from SASE FELs operating in the linear regime”, Nucl. Instr. And Meth. A, vol. 407, pp. 291-295, 1998.

[7] J. Feldhaus et al., “Possible application of X-ray optical elements for reducing the spectral bandwidth of an X-ray SASE FEL “, Nuclear Instr. and Meth. A, vol. 393, pp. 162-166, 1997.

[8] I.Boscolo and V. Stagno, Nucl. Instrum. Methods A, vol. 198, p. 483, 1982; J. B. Murphy, C. Pellegrini, and R. Bonifacio, “Collective instability of a free electron laser including space charge and harmonics,” Opt. Commun., vol. 53, pp. 197–202, 1985; Z. Huang and K.-J. Kim, “Three-dimensional analysis of harmonic generation in high-gain free-electron lasers,” Phys. Rev. E, vol. 62, p. 7295, 2000.

[9] R. Bonifacio, L. De Salvo Souza, P. Pierini, and E. T. Scharlemann, “Generation of XUV light by resonant frequency tripling in a two-wiggler FEL FEL amplifier,” Nucl. Instrum. Methods A, vol. 296, pp. 787–790, 1990.

[10]L. H. Yu, “Generation of intense UV radiation by subharmonically seeded single-pass free-electron lasers,” Phys. Rev. A, vol. 44, pp. 5178–5193, 1991; I. Ben-Zvi, L. F. Di Mauro, S. Krinsky, M. G. White, and L. H. Yu, “Proposed UV FEL user facility at BNL,” Nucl. Instrum. Methods A, vol. 304, pp. 181–186, 1991; “Theory of high gain harmonic generation: an analytical estimate “, Nuclear Instr. and Methods A, vol. 483, pp. 493-498, 2002

[11] M. Cornacchia et al., “Formation of electron bunches for harmonic cascade FELs”, in Proc. of this conference.

[12] P. Emma, et al., “Femtosecond and subfemtosecond X-ray pulses from a self-amplified spontaneous-emission-based free-electron laser,” Phys. Rev. Lett., vol. 92, pp. 074 801 1–074 801 4, 2004.

[13] C. B. Schroeder, C. Pellegrini, S. Reiche, J. Arthur, and P. Emma,“ Chirped-beam two-stage free-electron laser for high-power femtosecond x-ray pulse generation,” J. Opt. Soc. Amer. B, vol. 19, pp.1782–1789, 2002.

[14] A. A. Zholents, “Method of enhanced self-amplified spontaneous emission for x-ray free- electron lasers,” Phys. Rev. ST Accel. Beams **8**, 040701, 2005.

[15] S. Reiche, P. Emma and C. Pellegrini, “ Pulse length control in an X-ray FEL by using wakefields”, Nuclear Instr. and Meth. A, vol. 507, pp. 426-430, 2003.

[16] J. Rossbach, “Results from the VUV-FEL”, in Proc. of this conference.

[17] E. L. Saldin, E. A. Schneidmiller and M. V. Yurkov “Scheme for attophysics experiments at a X-ray SASE FEL “, Nuclear Instr. and Meth. A, vol. 507, pp. 439-444 , 2003.

[18] T. Orzechowski *et al.*, “Microwave radiation from a high-gain free-electron laser amplifier,” Phys. Rev. Lett., vol. 54, pp. 889–892, 1985.

[19] D. A. Kirkpatrick et el., “A high power, 600 m wavelength free electron laser,” Nuclear Instrum. Methods A, vol. 285, pp. 43–46, 1989.

[20] T. Lefevre, et al., “Self-amplified spontaneous emission and bunching at 3 GHz in a microwave free-electron laser,” Phys. Rev. Lett., vol. 82, pp. 323–326, 1999.

[21] M. Hogan *et al.*, “Measurements of high gain and intensity fluctuations in a SASE free-electron laser,” Phys. Rev. Lett., vol. 80, pp. 289–292, 1998.

[22] M. Hogan, et a., “Measurements of gain larger than 10^5 in a 12 mm SASE-FEL,” Phys. Rev. Lett., vol. 81, pp. 4867–4870, 1998.

[23] S. G. Biedron et al., “Measurements of nonlinear harmonic generation at the Advanced Photon Source's SASE FEL “, Nuclear Instr. and Met. A, vol. 483, Pages 94-100, 2002.

[24] V. Ayvazyan *et al.*, “Generation of GW radiation pulses from a VUV free-electron laser operating in the femtosecond regime,” Phys. Rev. Lett., vol. 88, p. 104 802-1-4, 2002.

[25] A. Murokh *et al.*, “Properties of the ultrashort gain length, self-amplified spontaneous emission free-electron laser in the linear regime and saturation,” Phys. Rev. E, vol. 67, p. 066 501, 2003.

[26] A. Tremaine, et al., “Observation of self-amplified spontaneous emission induced electron-beam microbunching using coherent transition radiation,” Phys. Rev. Lett., vol. 81, pp. 5816–5819, 1998.

[27] A. H. Lumpkin et al., “First observations of COTR due to a microbunched beam in the VUV at 157 nm “, Nuclear Instr. and Meth. A, Vol. 528, pp. 194-198, 2004

[28] A. Tremaine et al., “Experimental characterization of nonlinear harmonic radiation from a visible self-amplified spontaneous emission free-electron laser at saturation, pp. 204 801-1–204 801-4, 2002.

[29] R. Ischebeck et al., “Study of the transverse coherence at the TTF free electron laser “, Nucl. Instr. and Meth. A, Vol. 507, pp. 175-180, 2003.

[30] L. DiMauro *et al.*, “First SASE and seeded FEL lasing of the NSLS DUV FEL at 266 and 400 nm,” Nucl. Instrum. Meth. A, vol. 507, pp.15–18, 2003.

[31] A. Doyuran et al., “Chirped pulse amplification of HGHG-FEL at DUV-FEL facility at BNL “, Nucl. Instr. and Meth. A, vol. 528, pp. 467-470, 2004.