Experimental Characterization of Nonlinear Harmonic Radiation from a Visible Self-Amplified Spontaneous Emission Free-Electron Laser at Saturation

A. Tremaine,[1] X. J. Wang,[2] M. Babzien,[2] I. Ben-Zvi,[2] M. Cornacchia,[3] H.-D. Nuhn,[3] R. Malone,[2] A. Murokh,[1] C. Pellegrini,[1] S. Reiche,[1] J. Rosenzweig,[1] and V. Yakimenko[2]

[1]*Department of Physics & Astronomy, UCLA, Los Angeles, California 90095*
[2]*Accelerator Test Facility, NSLS, BNL, Upton, New York 11973*
[3]*SSRL, SLAC, Stanford, California 94309*

(Received 20 September 2001; published 3 May 2002)

Nonlinear harmonic radiation was observed using the VISA self-amplified, spontaneous emission (SASE) free-electron laser (FEL) at saturation. The gain lengths, spectra, and energies of the three lowest SASE FEL modes were experimentally characterized. The measured nonlinear harmonic gain lengths and center spectral wavelengths decrease with harmonic number, n, which is consistent with nonlinear harmonic theory. Both the second and third nonlinear harmonics energies are about 1% of the fundamental energy. These experimental results demonstrate for the first time the feasibility of using nonlinear harmonic SASE FEL radiation to produce coherent, femtosecond x rays.

DOI: 10.1103/PhysRevLett.88.204801 PACS numbers: 41.60.Cr, 41.60.Ap, 41.85.Ja

The explosive growth of synchrotron light source applications in the last few decades is due largely to the improved brightness of the x-ray beams they produce [1]. During the same time, high power, femtosecond laser technologies have opened new frontiers in spectroscopy and dynamics system studies [2]. The free electron laser (FEL) is one of the technologies that will bridge the gap between laser light (coherent, femtosecond pulse duration at long wavelengths) and synchrotron light (incoherent, picosecond pulse duration at x-ray wavelengths).

A FEL can operate as either an oscillator or a single-pass amplifier. For coherent, femtosecond x-ray generation, oscillators and laser-seeded FEL amplifiers are not feasible due to the nonexistence of highly reflective mirrors and laser seeds in the x-ray region. All proposed x-ray FEL facilities [3,4] are based on the self-amplified, spontaneous emission (SASE) FEL, which is a high-gain, single-pass FEL amplifier that starts from noise [5,6]. SASE x-ray facilities rely on high-brightness, low energy-spread electron beams and novel undulators to achieve saturation within a reasonable distance.

One of the natural consequences of the SASE FEL is nonlinear harmonic generation (NHG) [7–10], which occurs only in the high gain and saturation regime. Compared to other harmonic radiation, NHG is driven by the fundamental mode and has faster growth rates and higher intensities. Both simulation and theory predict the NHG 3rd harmonic to be about 1% of the fundamental for an x-ray SASE FEL [9,10]. This is about 8 orders of magnitude brighter than current third generation synchrotron light sources. NHG is transversely coherent, and it makes the possibility for SASE FELs to have broad spectral coverage. In addition, NHG is less sensitive to the electron beam parameters, undulator errors, and wakefields compared to other linear harmonic generation schemes [11].

As an electron beam passes through the periodic magnetic field of an undulator, such as in a SASE FEL, spontaneous radiation is emitted due to the electron beam/magnetic field interaction. A ponderomotive potential is formed by the interaction of this radiation field with the undulator magnetic field. The ponderomotive potential longitudinally microbunches the electron beam on the scale of a radiation wavelength, λ_r. As the beam microbunches further, the coherent radiation emitted from the beam grows exponentially through the undulator. At saturation, the radiation field stops growing and an oscillatory exchange of energy between the radiation field and electron beam continues until the end of the undulator.

The wavelength of fundamental radiation emitted from this process is given by

$$\lambda_r = \frac{\lambda_u}{2\gamma^2}\left(1 + \frac{K^2}{2}\right), \tag{1}$$

where λ_u is the undulator period, γ is the electron beam energy, and K is the undulator parameter. Equation (1) is also the period at which the electron beam is longitudinally microbunched, $\lambda_{\mathrm{mb}} = \lambda_r$. High-gain SASE FELs have been demonstrated from IR to UV wavelengths [12–22], and saturation of SASE FELs has been observed recently at the LEUTL [19], VISA [20,21], and TTF [22] facilities. In addition, measurements of the longitudinal microbunching of the electron beam from SASE FELs have been reported [23,24].

An electron beam in a FEL near saturation is tightly microbunched at the fundamental wavelength. Such a strongly microbunched electron beam drives the microbunching at its harmonics [8], $\lambda_{\mathrm{mb},n} = \lambda_{\mathrm{mb}}/n$, where $n = 1, 2, 3, \ldots$. Since the harmonic microbunching is driven by the fundamental ($n = 1$) SASE process, the harmonic microbunching grows at a rate faster than the fundamental, but it starts later in the undulator. The radiation emitted from such a bunching process is known as nonlinear harmonic radiation [8–10]. The experimental investigation of the nonlinear harmonic radiation at the VISA SASE FEL is presented in this Letter. For the first time, the gain lengths for the three lowest modes of a SASE FEL have been measured and shown to decrease with the harmonic number, n. The measured energies from nonlinear harmonic generation are shown to agree with both nonlinear theory and simulations. In addition, the spectra for these modes have been experimentally characterized.

The VISA FEL is part of the research and development effort for the proposed LCLS x-ray FEL project [3] and is a test bed for both the high-brightness electron beam and SASE FEL physics required for an x-ray FEL. Table I summarizes the key design parameters for the experiment. The VISA experiment was carried out at the Accelerator Test Facility (ATF) of Brookhaven National Laboratory. The ATF high-brightness electron beam is produced by an S band, 1.6-cell photocathode rf gun operating in longitudinal emittance compensation mode [25]. The electron beam is accelerated to 72 MeV by two sections of traveling wave linac. The final match of the electron beam into the VISA undulator is done by a quadrupole magnet triplet. The VISA undulator [26] is a planar magnet array with built-in strong focusing. The strong focusing is introduced by placing dipole magnets with opposite polarity on either side of the electron beam propagation axis [26]. The 4-m undulator has a 1.8-cm period and a 6-mm gap. The combination of strong focusing and bright electron beam demands the straightness of the undulator to be better than 30 μm over one gain length [27].

Both SASE and NHG measurements along the undulator are made using eight diagnostic pop-in ports [28] plus the diagnostic station downstream of the undulator. The diagnostic pop-in screens inside the undulator are spaced at 50-cm intervals, the last pop-in being 25 cm from the exit of the undulator. Nonlinear harmonic growth occurs primarily in the last quarter of the undulator and can be measured at the last three diagnostic pop-ins. The fundamental SASE radiation can be detected at the last five diagnostics ports.

The nonlinear harmonic spectral measurements in Fig. 1(a) were captured in a single shot when high SASE gain was established. The spectrometer (Ocean Optics) has peak sensitivity around 300 nm and attenuates (by about 2 orders of magnitude) at the SASE fundamental. The resolution of the spectrometer is about 1 nm. Figure 1(a) shows the harmonic wavelengths, decreasing according to the following equation:

$$\lambda_{r,n} = \frac{\lambda_f}{n}, \tag{2}$$

where $n = 1, 2, 3, \ldots$ is the harmonic number and λ_f is the fundamental wavelength ($n = 1$). The measured fundamental wavelength is 845 nm while the 2nd and the 3rd harmonics are 423 nm and 281 nm ($n = 2, 3$), respectively. The spectrum obtained in Fig. 1(a) agrees with Eq. (2) within the resolution of the spectrometer.

Figure 1 also shows the spectral bandwidth, $\Delta\lambda_n$ (FWHM), which decreases as the mode number increases. The bandwidth measured for the fundamental at saturation has been reported elsewhere for VISA [20,21] and is $\Delta\lambda_f = 15$ nm. Figure 1(b) provides independent measurements for the 2nd and 3rd harmonics at the VISA saturation. The spectral widths (FWHM) for the 2nd and 3rd harmonics are $\Delta\lambda_2 = 6$ nm and $\Delta\lambda_3 = 3$ nm, respectively. The above data show the nonlinear harmonic bandwidths, $\Delta\lambda_n$, relative to that of the fundamental decrease with an increasing mode number, n. In addition, the relative bandwidths, $\Delta\lambda_n/\lambda_n$, also decrease with mode number, but at a slower rate. Recent studies [29] of the SASE nonlinear harmonic spectra support this observation. A double hump was observed in Fig. 1(a) for the 2nd harmonic. Figure 1(a) was captured when the nonlinear harmonics were just beginning to grow and the double hump is most probably due to the electron beam/harmonic radiation interaction during the SASE NHG start-up process. Figure 1 experimentally confirms that high quality nonlinear harmonic spectra can be

TABLE I. VISA FEL design parameters.

Wavelength (fundamental)	λ_f	800 nm
E-beam energy	E	72 MeV
Peak current	I_p	200 A
Emittance	ε	2 mm · mrad
Energy spread	$\Delta\gamma/\gamma$	0.17%
Power gain length	L_g	17.5 cm
Spot size	σ	60 μm

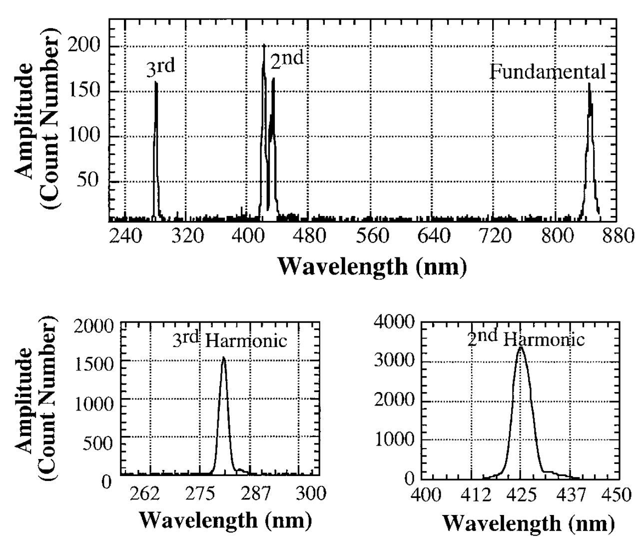

FIG. 1. (a) Single shot harmonic spectra of the fundamental, 2nd and 3rd harmonics. (b) Harmonic spectra independently measured at peak VISA gain.

expected for future short wavelength SASE FELs driven by high-brightness electron beams.

The nonlinear harmonics as a function of distance for the three lowest SASE FEL modes were measured using factory-calibrated Molectron joulemeters. Two bandpass filters were placed before the joulemeter for each harmonic measurement. Each filter attenuates the fundamental signal by at least a factor of 1000; thus combining two filters leads to 6 orders of magnitude attenuation outside of the passband. Figure 2 shows a log-linear plot for fundamental, 2nd and 3rd harmonics vs distance along the second half of the 4-m VISA undulator. The deviation of the last two points from exponential growth for the fundamental mode demonstrates SASE saturation. The fundamental saturation length is about 3.8 m.

A fundamental gain length of $L_g = 19$ cm is obtained from Fig. 2. To obtain the harmonic gain lengths, $L_{g,2}$ and $L_{g,3}$, the data in the linear harmonic region are excluded. The 2nd harmonic gain length is measured to be $L_{g,2} = 9.8$ cm. Because of the limited number of diagnostic pop-ins, only two points for the nonlinear 3rd harmonic regime could be used to estimate the gain length. Multiple measurements resulted in a 6-cm average growth rate, which is consistent with the gain length relation quoted in Eq. (3) below. Figure 2 shows the nonlinear harmonic radiation grows faster compared to the fundamental. Our measurements confirm nonlinear harmonic generation theory [8–10], in which the gain length decreases with the harmonic number, n.

$$L_{g,n} = \frac{L_g}{n}. \tag{3}$$

Finally, the energy for each mode was measured downstream of the undulator. Table II shows the nonlinear harmonic energies measured at the end of the undulator and

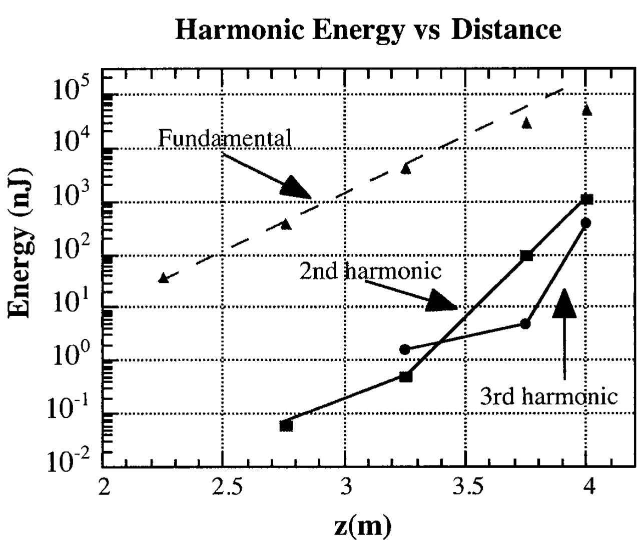

FIG. 2. Energy vs distance for the fundamental, 2nd and 3rd harmonics. The gain lengths for these are 19, 9.8, and 6.0 cm, respectively.

the harmonic energy as a percentage of the fundamental. It can be seen from Table II that not only is there significant UV (281 nm, 3rd harmonic) energy, but there is also comparable energy in the green (423 nm, 2nd harmonic). The relatively large amount of 2nd harmonic energy from a planar undulator is due to the low electron beam energy used in this experiment. The 2nd and 3rd nonlinear harmonic energies are related by [30]

$$E_2 \approx E_3\left(\frac{K}{\gamma k_u \sigma_x}\right)^2\left(\frac{K_2}{K_3}\right)^2\left(\frac{b_2}{b_3}\right)^2, \tag{4}$$

where $K = 1.28$ is the undulator parameter, γ is the beam energy, k_u is the undulator wave number, σ_x is the rms beam size, and b_n are the harmonic bunching parameters. K_n are the coupling coefficients [30]:

$$\begin{aligned} K_n &= K(-1)^{(n-1)/2}[J_{(n-1)/2}(n\xi) - J_{(n+1)/2}(n\xi)], \\ n &= 1,3,5,\ldots, \\ K_n &= K(-1)^{(n-2)/2}[J^{©}_{n/2}(n\xi)], \qquad n = 2,4,6,\ldots, \end{aligned} \tag{5}$$

where $\xi = K^2/(4 + 2K^2)$ and $J(n\xi)$ is the Bessel function.

Using the parameters in Table I, GENESIS [31] simulations produce harmonic bunching parameters of $b_2 = 0.47$ and $b_3 = 0.28$ near saturation. Inserting the GENESIS results into Eqs. (4) and (5), NHG theory predicts that the 3rd and 2nd harmonic energies should be 1% and 1.6% of the fundamental, respectively. The measured 3rd and 2nd harmonics are 0.8% and 1.8% (Table II) of the fundamental, with the ratio $E_2/E_3 = 2.3$.

The slight discrepancy between the measured and calculated energy ratio could arise from two sources: First, for each harmonic measurement, the signal passes through two narrow band filters. The errors in calibrating the transmission coefficient for such filters is one possible source, especially for the 3rd harmonic where each filter attenuates the desired signal by 80%. Second, the VISA beam profile monitors have a resolution no better than 15 μm. An error in spot size measurements of this order could account for the remainder of the discrepancy described above since the energy ratio given in Eq. (4) strongly depends on the ratio of the wiggle amplitude to the transverse spot size, $K/\gamma k_u{:}\sigma_x$.

The on-axis radiation is dominated by the odd harmonics for a planar undulator. If the off-axis radiation is collected, the even harmonic is observed to be quite significant and comparable to the odd harmonic for the VISA FEL.

TABLE II. Measured harmonic energy for fundamental, 2nd and 3rd harmonics.

Mode	n	1	2	3
Wavelength (nm)	λ_f	845	423	281
Energy (μJ)	E_n	52	0.93	0.40
% of fundamental energy	$\frac{E_n}{E_1} \times 100$		1.8	0.77

Coupling to the 2nd harmonic strengthens as the electron beam energy decreases. The reason for this can be seen as follows: If the electron beam energy decreases, then the wiggle amplitude will increase. The even harmonics for a planar undulator are emitted off axis [32], and as the transverse wiggle amplitude increases relative to σ_x, so will the radiation emitted for the 2nd harmonic. The strong coupling to even the NHG occurs only for low energy FELs, such as VISA, thus providing a unique window to further test the NHG theory.

Though the observation of NHG in the far infrared is claimed [33], we have presented the first experimental characterization of the nonlinear harmonic gain lengths and single shot spectra for the three lowest modes of a SASE FEL. The basic properties of NHG theory have been experimentally confirmed; gain length and harmonic wavelength decrease linearly with harmonic number, n, and harmonic spectral width narrows with harmonic number, n. Furthermore, large energies of the 2nd and 3rd nonlinear harmonics were observed and confirmed the predictions of NHG theory. These results are important not only as a comprehensive verification of nonlinear harmonic SASE FEL theory, but also as a demonstration of an alternative source of high-energy, high-quality spectra, short wavelength radiation.

The authors thank Dr. Z. Huang of the APS for stimulating conversations regarding the nonlinear harmonic evolution process. We also express our gratitude to Mr. R. Harrington and Mr. D. Davis for their support. This work is supported by U.S. Department of Energy Contracts No. DE-AC02-98CH10886, No. DE-FG03-92ER40793, and No. DE-AC03-76F00515.

[1] J.-L. Laclare, in *Proceedings of the 1998 European Particle Accelerator Conference* (Editions Frontières, Gif-sur-Yvette, France, 1998), p. 79.

[2] A. H. Zewail, in *Femtochemistry and Femtobiology,* edited by V. Sundström (World Scientific, River Edge, NJ, 1997).

[3] M. Cornacchia *et al.,* Linac Coherent Light Source (LCLS) Design Study Report, Ó Report No. SLAC-R-521 (Stanford Linear Accelerator Center, Stanford, CA, revised 1998).

[4] *Conceptual Design of a 500 GeV e^+e^- Linear Collider with Integrated X-Ray Laser Facility,* edited by R. Brinkmann, G. Materlik, J. Rossbach, and A. Wagner (Deutsches Elektronen-Synchrotron, Hamburg, 1997).

[5] A. M. Kondratenko and E. L. Saldin, Sov. Phys. Dokl. **24**, No. 12, 986 (1979).

[6] R. Bonifacio, C. Pellegrini, and L. M. Narducci, Opt. Commun. **50**, 373 (1984).

[7] J. B. Murphy, C. Pellegrini, and R. Bonifacio, Opt. Commun. **53**, 197 (1985).

[8] R. Bonifacio *et al.,* Nucl. Instrum. Methods Phys. Res., Sect. A **293**, 627 (1990).

[9] H. P. Freund *et al.,* IEEE J. Quantum Electron. **36**, 275 (2000).

[10] Z. Huang and K. J. Kim, Phys. Rev. E **62**, 7295 (2000).

[11] K. J. Kim and Z. Huang, in *Physics of, and Science with, the X-Ray Free-Electron Laser,* edited by S. Chattopadhyay *et al.,* AIP Conf. Proc. No. 581 (AIP, Melville, NY, 2001).

[12] S. V. Milton *et al.,* Phys. Rev. Lett. **85**, 988 (2000).

[13] R. Prazeres *et al.,* Phys. Rev. Lett. **78**, 2124 (1987).

[14] M. J. Hogan *et al.,* Phys. Rev. Lett. **80**, 289 (1998).

[15] M. J. Hogan *et al.,* Phys. Rev. Lett. **81**, 4867 (1998).

[16] D. C. Nguyen *et al.,* Phys. Rev. Lett. **81**, 810 (1998).

[17] M. Babzien *et al.,* Phys. Rev. E **57**, 6093 (1998).

[18] J. Andruszkow *et al.,* Phys. Rev. Lett. **85**, 3825 (2000).

[19] S. Milton *et al.,* Science **292**, 2037–2041 (2001).

[20] A. Tremaine *et al.,* in *Proceedings of the 2001 Particle Accelerator Conference, Chicago, 2001* (IEEE, Piscataway, NJ, 2001).

[21] A. Murokh *et al.,* in *Proceedings of the 2001 Particle Accelerator Conference, Chicago, 2001* (Ref. [20]).

[22] V. Ayvazyan *et al.* (to be published).

[23] A. Tremaine *et al.,* Phys. Rev. Lett. **81**, 5816 (1998).

[24] A. H. Lumpkin *et al.,* Phys. Rev. Lett. **86**, 79 (2001).

[25] X. J. Wang, X. Qiu, and I. Ben-Zvi, Phys. Rev. E **54**, R3121 (1996).

[26] R. Carr *et al.,* Phys. Rev. ST Accel. Beams **4**, 122402 (2001).

[27] P. Emma *et al.,* in *Proceedings of the 20th International Free Electron Laser Conference (FEL98), Williamsburg, VA, 1998* (Elsevier, Amsterdam, New York, 1999), p. 1198.

[28] A. Murokh *et al.,* in *Proceedings of the 22nd International Free Electron Laser Conference (FEL2000), Durham, NC, 2000* (North-Holland, Amsterdam, 2001).

[29] Z. Huang (private communication).

[30] Z. Huang and K. J. Kim, in *Proceedings of the 22nd International Free Electron Laser Conference, Durham, NC, 2000* (Ref. [28]).

[31] S. Reiche, Nucl. Instrum. Methods Phys. Res., Sect. A **429**, 243 (1999).

[32] H. P. Freund and T. M. Antonsen, Jr., *Principle of Free-Electron Lasers* (Chapman & Hall, London, 1996), 2nd ed.

[33] R. Kato *et al.,* in *Proceedings of the 23rd International Free Electron Laser Conference (FEL2001), Darmstadt, Germany, 2001* (North-Holland, Amsterdam, 2001).