Multicolor Operation and Spectral Control in a Gain-Modulated X-Ray Free-Electron Laser

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We show that the spectral properties of a self-amplified spontaneous emission x-ray free-electron laser can be controlled by modulating the gain in magnetic undulators, thus producing one or several spectral lines within a single few femtosecond pulse. By varying the magnetic field along the undulator and the electron beam transport line, the system we demonstrate can tailor the x-ray spectrum to optimally meet numerous experimental requirements for multicolor operation.

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The x-ray free-electron laser (X-FEL) is the brightest source of coherent radiation in the nanometer and subnanometer wavelength regime. Operational X-FEL facilities such as the Linac Coherent Light Source (LCLS, at SLAC) [1], SACLA (RIKEN) [2], and FLASH (DESY) [3] have already shown remarkable scientific capabilities in biology, chemistry, material science, atomic and molecular physics, as well as many other disciplines (see, e.g., Refs. [4–6]). These existing X-FEL facilities operate in the self-amplified spontaneous emission (SASE) mode, and as a result, their spectrum is noisy with a bandwidth of the order of the FEL parameter ρ [7], typically about 0.001. Such pulses are nearly diffraction limited in the transverse dimension, but they are not Fourier limited in the longitudinal (temporal) dimension. Typical temporal profiles are composed of several uncorrelated spikes [8]. Several methods have been proposed to improve the FEL longitudinal coherence using external laser seeding [9–11] or self-seeding [12,13]. More recently it has been shown that near Fourier limited pulses can be obtained by delaying the electron bunch with respect to the radiation pulse in a controlled way, a technique known as improved SASE (or high-brightness SASE) [14–16].

Two-color operation of x-ray FELs is the subject of intense research at x-ray user facilities. In the field of physical chemistry, for instance, there is a desire to extend the traditionally optical techniques of stimulated Raman spectroscopy to the x-ray regime [17–23]. In the condensed phase, the prospect of stimulated resonant inelastic x-ray scattering in solids could bring key time resolution to this already advanced field [24,25]. Furthermore, extending x-ray scattering techniques such as multiwavelength anomalous diffraction [26] to time-resolved interactions could allow such advanced phase retrieval for diffraction studies of femtosecond scale dynamically evolving molecular structures.

First observed with an infrared FEL oscillator [27], two-color operation of a high-gain x-ray free-electron laser has been recently demonstrated in the soft-x-ray regime in Ref. [28] and also for a seeded FEL in Ref. [29]. In contrast to Ref. [28], however, the so-called gain-modulated FEL reported here can achieve two-color pulses with a time delay shorter than the slippage length. In our experiment this is much less than the few femtosecond pulse duration, as required by the classes of experiments mentioned above. We report the generation of single X-FEL pulses with two narrow spectral lines whose relative photon energy separation can be tuned by as much as 2%.

The gain-modulated FEL technique shown in Fig. 1 uses periodic modulation of the undulator field to produce two or more narrow x-ray spectral lines with tunable photon energy separation. In the case of a FEL, the modulation of the undulator magnetic field was first explored theoretically for a simple biharmonic undulator [30]. In this Letter we show that a periodic modulation of the undulator field is equivalent to introducing a sequence of electron delays at two different resonant wavelengths. As the electron bunch propagates, the sequence of resonance-delay sections develops an interference pattern that gives rise to multiple spectral lines. Each line has a narrower bandwidth than pure SASE radiation, but multiple lines can be distributed over a spectral range that is wider than the intrinsic SASE bandwidth. We present a universally scaled one-dimensional theory of this new mode of operation and present its first experimental demonstration at LCLS.

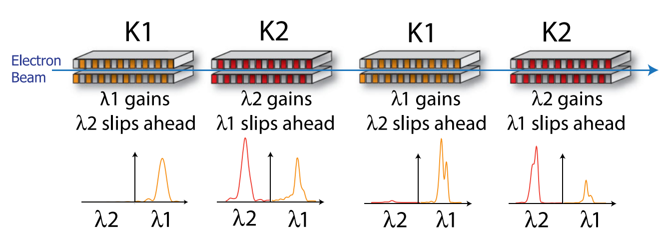

FIG. 1 (color online). Schematics of a gain-modulated FEL. The undulator parameter K varies periodically along the FEL beam line. In each undulator section, the resonant wavelength is amplified exponentially while the nonresonant wavelength slips ahead of the electrons without amplification. The resulting spectra exhibit multiple spectral lines.

A free-electron laser operating in the SASE mode amplifies the microbunching that is produced by beam shot noise. This generates coherent radiation at a central wavelength,

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

where λ_u is the undulator period, γ the beam energy normalized to the rest energy mc^2, and the undulator parameter is given by $K = eB_u\lambda_u/2\pi mc$, where B_u is the peak magnetic field of the undulator and e is the electron charge. The gain bandwidth is of the order of the FEL parameter $\rho = ((K/4\sqrt{2})(JJ\lambda_u\omega_p/2c\pi))^{2/3}$ [7], where c is the speed of light and $\omega_p = \sqrt{4\pi e^2 n_0/m\gamma^3}$ is the relativistic beam plasma frequency, with n_0 being the beam volume density and m the electron mass. Finally, $JJ = J_0(K^2/(4 + 2K^2)) - J_1(K^2/(4 + 2K^2))$, where J_0 and J_1 are the zeroth and first-order Bessel functions of the first kind. The FEL parameter also defines the energy transfer at saturation from the electron beam to the photon pulse. In the SASE mode, the spectrum is composed of several uncorrelated spikes distributed within the amplification bandwidth and the temporal profile is given by several spikes with a characteristic length of λ_r/ρ. In a typical SASE FEL, the undulator parameter K is constant during the amplification process, except for a small linear taper (on the order of 1% over the entire undulator beam line) that is used to compensate beam energy losses due to spontaneous radiation and wakefields.

The gain-modulated FEL, shown in Fig. 1, is analogous to gain modulation in solid-state lasers, e.g., quantum cascade lasers [31] where a periodic modulation of the active medium gives rise to discrete energy bands that are determined entirely by the geometry of the medium. In the context of FELs, gain modulation is achieved by alternating the undulator magnetic field with a periodicity $L_u \gg \lambda_u$. This allows for nearly simultaneous generation of two colors in a single pulse. By producing two distinct values of the undulator parameter $K_{1,2}$, radiation at the two resonant wavelengths $\lambda_{1,2} = \lambda_u(1 + K_{1,2}^2/2)/2\gamma^2$ is alternately amplified as the beam travels along the undulator, resulting in a time separation smaller than the slippage in a single modulation period $\lambda_r L_u/\lambda_u$. This type of configuration can be easily implemented in all existing X-FELs, since typical X-FEL undulators are divided into many modules with tunable strength parameter K.

The evolution of the system in an undulator section is described with a simple one-dimensional model. We use the formalism introduced in [8], in which the FEL dynamics is described in terms of three universally scaled collective variables. The radiation electric field $E_{\rm rad}$ is normalized to the saturation power as $A = E_{\rm rad}/(2\pi\rho P_b)^{1/2}$, where $P_b = \gamma n_0 mc^2$ is the electron beam energy density. The electron beam bunching factor is given by $B = (1/N)\sum_j \exp[-i(k_u + k)z_j + ikct]$, where $k_u = 2\pi/\lambda_u$ is the undulator frequency, $k = 2\pi/\lambda$ is the radiation frequency, z_j is the longitudinal position of the jth electron, and the sum is performed over all the N electrons in the bunch. The energy modulation, normalized to ρ is given by $P = (1/N)\sum_j(\eta_j/\rho)\exp[-i(k_u + k)z_j + ikct]$, where η_j is the relative energy deviation of the jth particle. In the frequency domain, and in the linear growth regime, the FEL evolves according to the following [8]:

$$\frac{\partial A}{\partial \tau} + i\delta A = B, \qquad \frac{\partial B}{\partial \tau} = -iP, \qquad \frac{\partial P}{\partial \tau} = -A, \tag{2}$$

where the interaction time is normalized to the FEL gain length as $\tau = ct/L_g$ with $L_g = \lambda_u/4\pi\rho$, $\delta = (\lambda_r - \lambda)/2\lambda_r\rho$ is the normalized detuning, $\lambda = 2\pi/k$ is the wavelength of interest, and λ_r is the resonant wavelength in each module. The solution of the linear system (2) is expressed in compact form with a transfer matrix:

$$\begin{pmatrix} B \\ P \\ A \end{pmatrix} = M_\tau(\delta)\begin{pmatrix} B_0 \\ P_0 \\ A_0 \end{pmatrix}, \tag{3}$$

with

$$M_\tau(\delta) = I + \sum_{j=1}^{3} \frac{i[\exp(i\mu_j\tau) - 1]}{3\mu_j^2 + 2\delta\mu_j}\begin{pmatrix} \frac{i}{\mu_j} & -\frac{i}{\mu_j^2} & -1 \\ -i & \frac{i}{\mu_j} & \mu_j \\ -\mu_j & +1 & -i\mu_j^2 \end{pmatrix}, \tag{4}$$

where I is the unit matrix and the eigenvalues μ_j are the solutions of the cubic dispersion equation:

$$\mu^3 + \delta\mu^2 + 1 = 0. \tag{5}$$

In a gain-modulated FEL there are two resonant wavelengths $\lambda_{1,2}$. We define the average resonant wavelength as $\bar{\lambda}_r = (\lambda_1 + \lambda_2)/2$. For a given wavelength we define the average detuning parameter $\bar{\delta} = (\bar{\lambda}_r - \lambda)/2\bar{\lambda}_r\rho$ and the normalized frequency separation $\Delta = (\lambda_2 - \lambda_1)/4\bar{\lambda}_r\rho$. The FEL dynamics is then described by the following evolution matrix:

$$\begin{pmatrix} B \\ P \\ A \end{pmatrix} = \prod_{n=1}^{N_s} M_{\tau_u}(\bar{\delta} + (-1)^n\Delta)\begin{pmatrix} B_0 \\ P_0 \\ A_0 \end{pmatrix}, \tag{6}$$

where $\tau_u = L_u/2L_g$ is the normalized length of an undulator section (i.e., the length of half a modulation period), N_s is the total number of sections, and B_0, P_0, A_0 are the initial values of the collective variables and the normalized radiation field. For small values of the effective detuning $\delta = \bar{\delta} + (-1)^n\Delta$, the undulator is resonant and the FEL is in the exponentially growing regime. For large values of the detuning parameter, instead, the single undulator transfer matrix can be approximated as

$$M \simeq \begin{pmatrix} 1 & -i\tau_u & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \exp(-i\delta\tau_u) \end{pmatrix}. \tag{7}$$

Equation (7) describes the evolution of the system in a nonresonant interaction regime. The microbunching evolves linearly as a function of time, $B = B_0 - iP_0\tau_u$, since the finite undulator dispersion transforms the electron beam energy modulation P_0 into density modulation. The electron bunch slips behind the radiation field to produce the term $\exp(-i\delta\tau_u)$, which corresponds to a forward temporal translation of the radiation field with respect to the electrons. The nonresonant radiation pulse also slips ahead of the resonant pulse, since the group velocity of the exponentially growing FEL pulse is $v_g = c(1 - 2\lambda_r/3\lambda_u)$ [32] while the nonresonant pulse travels at the speed of light. The induced slippage in the detuned undulators increases the effective cooperation length [14,33]. This very same physical principle is employed in an improved SASE FEL [14] whereby the delay is induced by magnetic chicanes placed between undulator modules such that the bandwidth can be reduced to the Fourier limit. Therefore, a gain-modulated FEL can be described as a two-color improved SASE scheme where each resonant wavelength $\lambda_{1,2}$ alternates between an exponential growing section and a temporal translation section. We define the gain function G_f as the amplitude of the field variable divided by the initial bunching factor $G_f = |A|/|B_0|$ starting from the following condition: $P_0 = 0$, $A_0 = 0$. The gain function squared is the average spectral power gain in a gain-modulated FEL starting from shot noise. Figure 2 shows the gain curve (defined as $\log|G_f|^2$) as a function of the detuning parameter $\bar{\delta}$ and the normalized wavelength separation Δ for $\tau_u = 1/\sqrt{3}$ [Fig. 2(a)] and $\tau_u = 4/\sqrt{3}$ [Fig. 2(b)], corresponding to an undulator periodicity of two and eight power gain lengths, respectively. The spectral power gain is also shown for three different values of Δ [Figs. 2(c) and 2(d)]. The spectrum exhibits several peaks at the frequencies $\bar{\delta}\tau_u = \ell\pi$, where ℓ is an integer number, corresponding to a constructive interference between the radiation emitted in two consecutive resonant undulators. In physical units, the relative wavelength separation of these peaks corresponds to the periodicity of the undulator modulation divided by the undulator frequency; i.e., $d\lambda/\lambda = \lambda_u/L_u$. Note that, for an undulator periodicity of several gain lengths [such as the case shown in Figs. 2(b) and 2(d)], the spectral peaks tend to merge into two dominant modes at the corresponding resonance frequencies $\bar{\delta} = \pm\Delta$, and the gain-modulated FEL generates two colors. For a shorter periodicity, instead, the system generates three or four colors depending on the specific value of Δ.

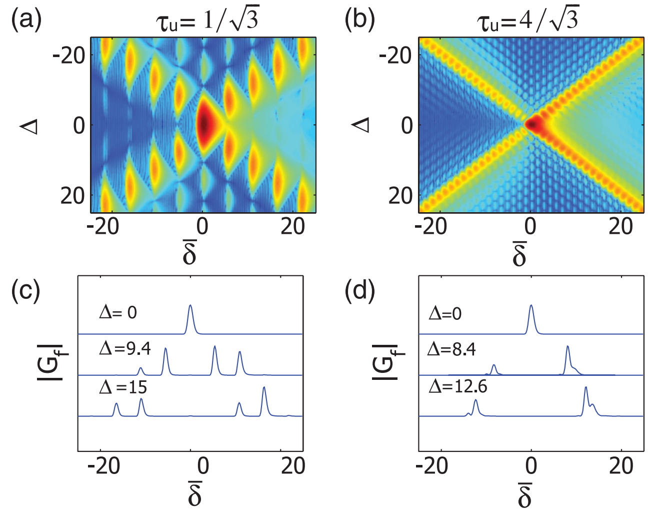

FIG. 2 (color). Gain curve (logarithm of the gain function G_f) as a function of the average detuning $\bar{\delta}$ and the undulator detuning Δ (upper images) for an undulator periodicity of two power gain lengths (a) and eight power gain lengths (b). (c),(d) Linear plots of the gain function for three different values of Δ.

The generation of multicolor pulses with a gain-modulated FEL has been demonstrated experimentally at LCLS. The experimental beam parameters are energy $E_b \simeq 4.35$ GeV, peak current $I_p = 2$ kA, and normalized transverse emittance $\epsilon_{x,y} \simeq 0.34$ μm. The average photon energy is tuned to $E_{\rm ph} \simeq 830$ eV, corresponding to a wavelength of $\bar{\lambda}_r \simeq 1.5$ nm. The pulse duration is limited to 20 fsec FWHM by use of an emittance spoiler [34]. The spectrum is measured with a soft-x-ray grating spectrometer [35]. The LCLS undulator is composed of modules of 110 periods each, which limits the undulator modulation periodicity to multiples of $220\lambda_u$. Figure 3 shows the average spectrum as a function of beam energy for a configuration with undulator periodicity of two undulator modules (i.e., alternating the K value at every undulator module), corresponding to a sideband separation of $\delta\lambda/\lambda \simeq 0.44\%$. The alternating undulators have $K_1 = 3.49 \times (1 + 4.375 \times 10^{-3})$ and $K_2 = 3.49 \times (1 - 4.375 \times 10^{-3})$. The data have been binned in energy to deconvolve the effect of shot-to-shot beam energy fluctuations. The spectrum clearly shows the appearance of three lines, spaced by multiples of the normalized undulator periodicity 0.44%. The position and relative amplitude of the spectral peaks are in good agreement with the 1D theoretical model. The measured pulse energy at saturation $E_{\rm pl} \simeq 15$ μJ is roughly one-tenth of the single-color SASE pulse saturation energy for the same beam conditions and the same average photon energy [36]. Saturation was reached after 18 undulators, corresponding to 9 gain-modulation periods. By increasing the periodicity of the undulator modulation, the spectral separation of the

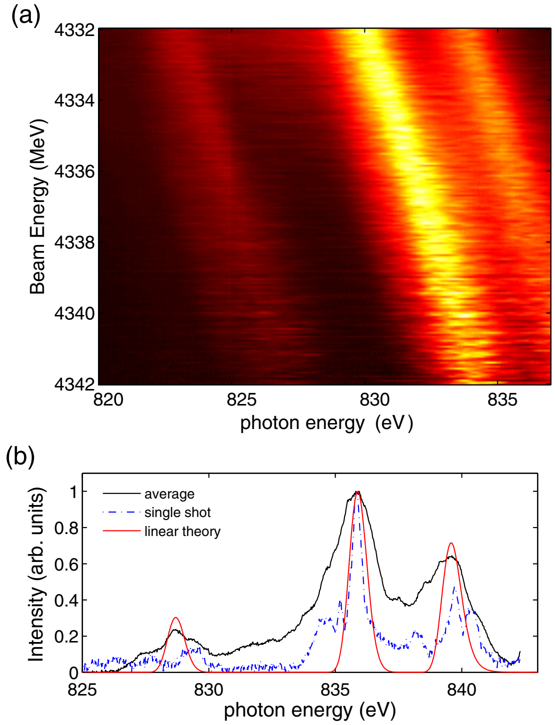

FIG. 3 (color online). (a) Average intensity as a function of beam energy and photon energy for an undulator periodicity of 2 undulator modules. (b) Average spectrum (black line) and single-shot spectrum (dashed blue line) for a beam energy of 4334 MeV. For comparison, the spectrum predicted from the linear theory is plotted as a red line.

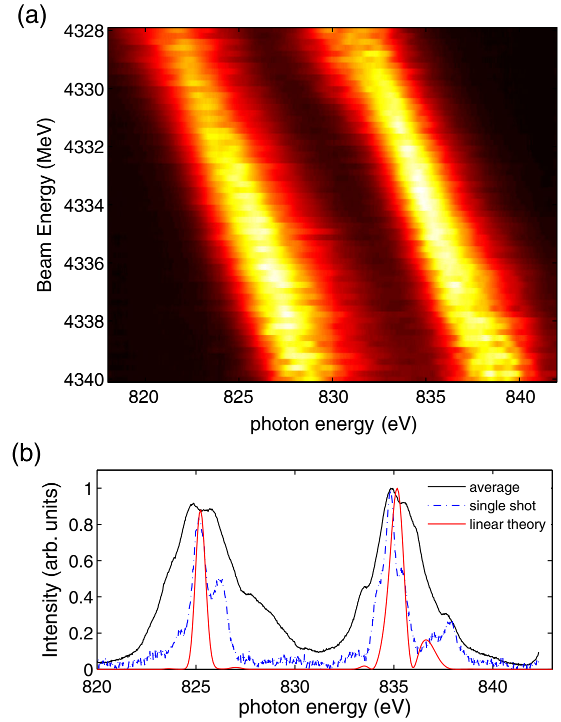

FIG. 4 (color online). (a) Average intensity as a function of beam energy and photon energy for an undulator periodicity of 6 undulator modules. (b) Average spectrum (black line) and single-shot spectrum (dashed blue line) for a beam energy of 4334 MeV. For comparison, the spectrum predicted from the linear theory is plotted as a red line.

sidebands decreases. Figure 4 shows the spectral intensity as a function of photon energy and beam energy for a configuration with a modulation periodicity of six undulator modules with $K_1 = 3.49 \times (1 + 3.5 \times 10^{-3})$ and $K_2 = 3.49 \times (1 - 3.5 \times 10^{-3})$. In this case, the sidebands merge into one dominant peak and the two emitted colors correspond to the resonant wavelengths in the alternating undulators. Note that in a gain-modulated FEL, for long modulation periods, one color typically contains a larger number of photons. This is due to two effects: first, there is a well-known asymmetry in the FEL dispersion relation [7], which makes the gain at one color larger for the same number of undulator modules, and second, the saturation length of the FEL is normally not a multiple of the modulation period. For certain scientific applications, however, it is important that the two colors have the same intensity. This problem can be easily overcome by adding a few extra undulators to boost the emitted photons in one of the two colors. Balancing of the two colors is achieved in this case by adding five undulators tuned to the low-energy photon pulse, close to the saturation point. The resulting spectrum in Fig. 4 shows two colors with approximately the same spectral intensity. The measured pulse energy at saturation in this case is roughly $E_p \simeq 18\ \mu\text{J}$, with a pulse duration of 10 fsec determined by the emittance spoiler aperture. The total number of undulators in this case was 23, corresponding to 6 gain-modulation periods followed by the 5 extra undulators to boost the first color. The position of the spectral peaks is in good agreement with the theoretical prediction. The theory, however, predicts that the two colors are balanced by adding 3.5 undulators tuned to the first color (lower photon energy). This is easily explained by noting that in the experiment we were operating at saturation, while the theory is based on the linear FEL equations.

The 1D model predicts a smaller bandwidth than the measured value (for both the single-shot spectra and the averaged spectrum), this effect is routinely observed in regular SASE operation at the LCLS, where the bandwidth is measured to be roughly 2 times the theoretical 1D value. This is due to a residual variation of the energy along the electron bunch length estimated to be on the order of 0.1%, which is not included in the 1D theory.

In conclusion, in this Letter we introduce the concept of gain-modulated FELs. In this new mode of operation, the magnetic field of the undulator is modulated by alternating the undulator parameter K with a certain periodicity. The spectrum of a gain-modulated FEL is composed of several lines. Each line has a bandwidth smaller than that of SASE, but their separation can be larger than the SASE bandwidth itself. We discuss a simple universally scaled model of this process and present the first experimental demonstration at the LCLS. The results presented in this Letter extend the capabilities of x-ray free-electron lasers by allowing the generation of a single x-ray pulse with multiple spectral lines with tunable photon energy separation.

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