Chirped Seeded Free-Electron Lasers: Self-Standing Light Sources for Two-Color Pump-Probe Experiments

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We demonstrate the possibility of running a single-pass free electron laser (FEL) in a dynamical regime, which can be exploited to perform two-color pump-probe experiments in the vacuum ultraviolet or x-ray domain, using the free-electron laser emission both as a pump and as a probe. The studied regime is induced by triggering the free-electron laser process with a powerful laser pulse, carrying a significant and adjustable frequency chirp. As a result, the output FEL radiation is split in two pulses, separated in time (as previously observed by different authors), and having different central wavelengths. We show that both the spectral and temporal distances between FEL pulses can be independently controlled. We also provide a theoretical description of this phenomenon, which is found in good agreement with experiments performed on the FERMI@Elettra free-electron laser.

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In recent years, the advent of ultrafast tabletop laser sources has boosted the development of new experimental methodologies, based on pump-probe techniques. In the latter, two light pulses with adjustable time delay and different wavelengths are used to investigate the processes occurring during chemical and physical reactions. The first pulse (pump) initiates the reaction, by breaking a bond or exciting one of the reactants. The second pulse (probe) is then used to interrogate the state of the reaction after a certain time delay from process start. By varying the time delay between pump and probe, and observing sample responses, one is able to make a "movie" of the reaction. Thanks to this technique, very deep scientific and technological insight was gained in different fields, ranging from quantum communications [1,2], to wave-function reconstruction in reacting molecules [3,4].

State-of-the-art tabletop lasers can generate photon pulses with durations of the order of few tens of femtoseconds, thus allowing us (through pump-probe methods; see, e.g., Ref. [5]) to gain access to the characteristic time scale of several basic chemical and physical processes. On the other hand, the photon energies they produce are limited to the IR-visible-UV spectral range. Such a limitation actually prevents the possibility of reaching the frontier of the length scales of the intermolecular distances and the energy scales of the bonds holding electrons in correlated motion with near neighbors.

This constraint is removed by free-electron lasers (FELs), which are able to deliver photon pulses of femtosecond time duration in the vacuum ultraviolet (VUV) or x-ray range [6]. However, FEL-based pump-probe techniques have to cope with practical hurdles, which pose severe limits to their development. The strategies presently considered to set up pump-probe schemes with FELs are basically two. In the simplest case, one can synchronize the FEL with an external laser, which can be used as the pump or as the probe. The first drawback of this scheme is that the possible advantage in using the short-wavelength FEL radiation is limited either to the pump or to the probe. A second issue is related to the jitter between the FEL and the external laser. Being of the order of several tens of femtoseconds, the latter is typically larger than achievable pulse durations. Therefore, the experimental time resolution is jitter limited. A second possible scheme to implement pump-probe FEL experiments is based on splitting an FEL pulse, delaying the two obtained subpulses and recombining them at the sample's location. This approach does not suffer from the limitations of the previous scheme. However, it does not allow us to independently set the wavelengths of the pump and of the probe, since they are generated by the same FEL pulse.

Stimulated by recent results obtained at the FERMI@Elettra FEL facility [7], in this Letter we demonstrate the possibility of operating an FEL in a regime allowing us to perform two-color pump-probe experiments in the VUV or x-ray domain. In the proposed configuration, the FEL is seeded by a powerful laser pulse, carrying a significant frequency chirp. As a result, the output FEL radiation is split in two pulses, separated in time (as previously observed by different authors [8]), and having different central wavelengths. We show that both the spectral and temporal distances between FEL pulses can be independently controlled, providing the possibility of using the FEL at the same time as a pump and as a probe.

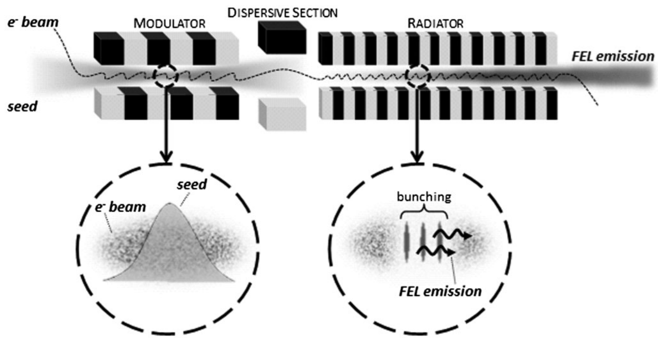

FIG. 1. Schematic layout of a seeded single-pass FEL (harmonic generation scheme).

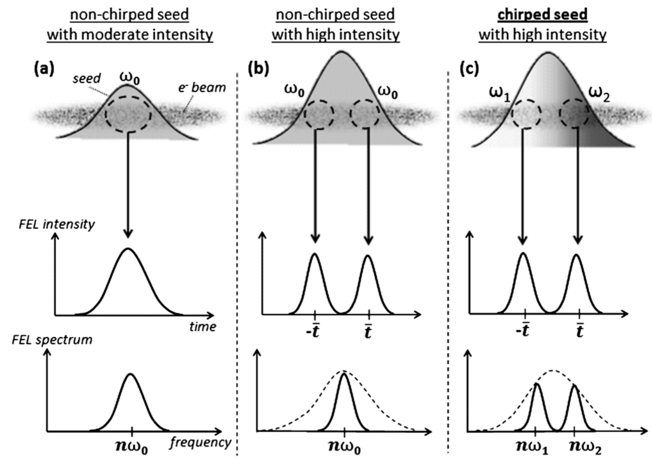

FIG. 2. Seed-electron interaction and resulting FEL (temporal and spectral) outputs for different seed configurations: no chirp and moderate seed intensity (left panel), no chirp and high seed intensity (central panel), chirped seed with high intensity (right panel). The meaning of the symbols is explained in the text.

The method we propose overcomes all the issues related to the previously mentioned schemes.

Seeded FELs can rely on different configurations [9–14]. In this work, we focus on the so-called harmonic generation scheme, in which a relativistic electron beam propagating through the static and periodic magnetic field generated by an undulator (called a modulator) interacts with a collinear externally injected optical pulse (seed) having wavelength λ_0; see Fig. 1. The interaction modulates the electron-beam energy. Energy modulation is transformed into spatial bunching, when the electron beam propagates through a magnetic chicane (dispersive section). The bunching (as the energy modulation) has a periodicity equal to the seed wavelength. However, it also presents significant components at the harmonics of the latter, i.e., at $n\omega_0$ (where $\omega_0 = 2\pi c/\lambda_0$, c being the speed of light and n an integer number). Finally, the bunched electron beam is injected into a long undulator chain (called a radiator), where it emits coherently at one of the seed harmonics. In the radiator, the electromagnetic intensity generated from bunched electrons is amplified, until, due to bunching deterioration, electrons are no longer able to supply energy to the wave and the process reaches saturation.

Consider now an FEL, in which a homogeneous electron beam is seeded by a Gaussian monochromatic laser pulse, e.g., the one generated by a Ti:Sa laser. In standard operation mode, the seed peak intensity, the strength of the dispersive section, and, as a consequence, the bunching at the radiator entrance are tuned, so as to maximize the emission from the part of the electron beam seeded by the center of the Gaussian pulse [see Fig. 2(a)]. The parts of the beam seeded by the tail of the Gaussian pulse will instead arrive at the radiator entrance with a local bunching smaller than optimum. As a result, the output FEL pulse will approximately reproduce the Gaussian shape of the seed, both in time and in spectrum. Suppose now we maintain constant the strength of the dispersive section and steadily increase the seed intensity [(see Fig. 2(b)]. For high enough intensities, the part of the electron beam that, in the modulator, interacted with the center of the Gaussian seed will have, at the radiator entrance, a bunching larger than optimum (overbunching). Because of the large laser-induced energy spread, the FEL emission from this part of the bunch will be significantly attenuated. The larger the seed peak intensity, the larger the overbunched zone will be. The beam portions having optimum bunching (seeded by the lateral parts of the Gaussian seed) will be located both at the right and at the left of that zone. As a consequence, in these conditions the FEL pulse will be characterized, in the time domain, by two lateral peaks [(see Fig. 2(b)] or, if the seed is sufficiently intense to suppress the emission from the central part, by two separated subpulses [8]. Because the seed is assumed to be monochromatic, the two subpulses have the same wavelength.

The situation changes if the seed carries a significant frequency chirp. Indeed, in this case, the two subpulses will be also characterized by different wavelengths; see Fig. 2(c). This opens up the possibility of using the FEL as a self-standing source to carry out two-color pump-probe experiments.

Let's consider a seed pulse carrying a controllable linear chirp [15]. The pulse electric field reads [16]:

$$E(t) \sim \exp(-\Gamma_R t^2 - i\Gamma_I t^2), \qquad (1)$$

where Γ_R and Γ_I are constant parameters controlling, respectively, the pulse duration and bandwidth. According to the previous equation, one can define the time-dependent phase of the pulse as $\phi(t) = \Gamma_I t^2$. Switching to intensity ($\simeq |E(t)|^2$), one finds that the rms pulse temporal duration, σ_l, is given by $\sigma_l = 1/(2\sqrt{\Gamma_R})$.

The instantaneous frequency within the seed pulse, $\omega_{\rm inst}(t)$, is defined by the following relation:

$$\omega_{\rm inst}(t) - \omega_0 = \frac{{\rm d}\phi(t)}{{\rm d}t} = 2\Gamma_I t. \quad (2)$$

In order to illustrate our findings, we consider the paradigmatic example of the FERMI@Elettra FEL [17], whose relevant parameters are listed in the caption of Fig. 3. The left panel of Fig. 3 shows the measured spectral evolution of the FERMI@Elettra FEL pulse, as a function of the seed power.

As expected, above a given power threshold the FEL pulse splits, both in time and in spectrum [18]. Larger seed powers correspond to larger extensions of the overbunched zone around the pulse center and, therefore, to larger temporal and spectral separations.

The central panel of Fig. 3 reports the spectral evolution of the FEL pulse, simulated using the numerical code PERSEO [19]. As can be seen, the agreement with experiments (left panel) is very satisfactory. This confirms the correctness of our interpretation of the pulse splitting mechanism. The left panel of Fig. 3 shows the correspondent (simulated) temporal pulse splitting. We remark that the maximum obtainable temporal split is limited by the electron-beam duration and by the possibility of generating long-enough (chirped) seed pulses characterized by significant local power at their tails.

In the following, we concentrate on the analysis of the so-called low-gain regime. In this low-gain regime, the relative FEL gain bandwidth B (approximately equal to the relative maximum spectral separation) is proportional to the inverse of the number of radiator periods (N_u): $B \simeq 1/N_u$.

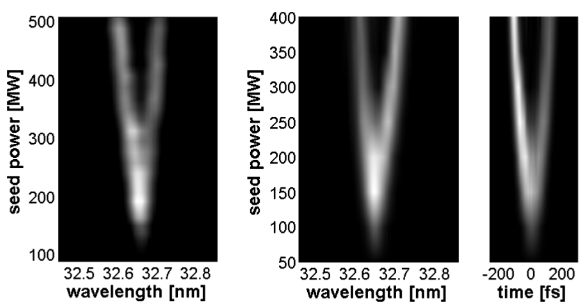

FIG. 3. Projected spectral and temporal FEL intensities for different seed powers. Left panel: experimental spectral splitting, measured at FERMI@Elettra [7]; central and right panels: simulated spectral and temporal splitting (using the code PERSEO). The set of parameters, valid both for the experiment and for the simulation, is the following: normalized energy $(\gamma_0) = 2.544 \times 10^3$, energy spread $(\sigma_\gamma) = 2.544 \times 10^{-1}$, peak current $(I_{\rm peak}) = 200$ A, bunch duration = 1 ps (rms); modulator period = 0.1 m, modulator length $(L_u) = 3$ m, modulator parameter $(K) = 5.7144$, radiator period $(\lambda_w) = 55 \times 10^{-3}$ m, number of radiator periods $(N_u) = 264$, central modulator wavelength $(\simeq 2\pi c/\omega_0) = 261$ nm, central radiator wavelength $(\lambda_{\rm rad}) = 32.6$ nm, harmonic number $(n) = 8$; seed-laser power $(P_0) = 50$–500 MW, strength of the dispersive section $(R_{56}) = 20\ \mu$m, laser spot size in the modulator $(\sigma_r) \simeq 300\ \mu$m, $(\Gamma_R, \Gamma_I)_3 = (5 \times 10^{-5}\ {\rm fs}^{-2}, 3.2 \times 10^{-5}\ {\rm fs}^{-2})$.

Exploiting a chirped seeded FEL for carrying out pump-probe experiments relies on the possibility of controlling independently the spectral and temporal distance between subpulses. Let us show how this can be achieved.

Suppose we want to generate, at the radiator exit, two subpulses having constant spectral distance and variable temporal separation. First, let's see how, for a given chirp, one can decide the subpulses' spectral distance. We indicate the latter with $\hat{\Delta}\omega = \hat{\omega}_1 - \hat{\omega}_2$, where $\hat{\omega}_1 = n\omega_1$ and $\hat{\omega}_2 = n\omega_2$ are the central frequencies of the two subpulses, ω_1 and ω_2 being the corresponding instantaneous frequencies carried by the seed [determined according to Eq. (2)], and n the selected harmonic number; see Fig. 2. In general, $\hat{\Delta}\omega$ will be given by $\alpha B \omega_{\rm rad}$, where α is a factor (smaller or slightly larger than one) fixed by the user and $\omega_{\rm rad} = n\omega_0$ is the central frequency at which the radiator is tuned.

According to Eq. (2),

$$|\hat{\Delta}\omega| = n|\omega_2 - \omega_1| = 4n\Gamma_I \bar{t} \simeq \alpha n B \omega_0, \quad (3)$$

where $\bar{t}$ is the temporal position of ω_1 and ω_2 (symmetric with respect to ω_0; see Fig. 2), determining the (identical) seed powers experienced by the portions of the electron beam emitting in the radiator at the two frequencies $\hat{\omega}_1$ and $\hat{\omega}_2$. Such a power is given by

$$P = P_0 \exp\left(\frac{-\bar{t}^2}{2\sigma_l^2}\right), \quad (4)$$

P_0 being the maximum seed power. Knowing P, one can find the electron-beam energy modulation, $\Delta\gamma$, induced by the laser-electron interaction inside the modulator [20]:

$$\Delta\gamma(r) = \sqrt{\frac{P}{\bar{P}}} \frac{KL_u}{\gamma_0 \sigma_r} J_{0,1}\left(\frac{K^2}{4 + 2K^2}\right) \exp\left(-\frac{r^2}{4\sigma_r^2}\right). \quad (5)$$

Here $\bar{P} \simeq 8.7$ GW, K is the modulator parameter, L_u the modulator length, γ_0 the nominal (normalized) beam energy, $J_{0,1}$ is the difference between the J_0 and J_1 Bessel functions, r is the radial position inside the electron beam, and σ_r is the seed-laser spot size in the modulator. The energy modulation is related to the bunching, b, created in the dispersive section by the following relation [9]:

$$b = \exp\left(-\frac{1}{2} n^2 \sigma_\gamma^2 d\right) J_n(n\Delta\gamma d), \quad (6)$$

where σ_γ is the (normalized) electron-beam incoherent energy spread, d is the strength of the dispersive section and J_n the Bessel function. The parameter d is usually expressed in terms of the parameter R_{56}, as $d = 2\pi R_{56}/(\lambda_0 \gamma_0)$. Our aim is to find the optimum value of the dispersive section which, for a fixed seed power, allows maximizing the FEL emission at the two wavelengths we are interested in. Such a value cannot be found by simply maximizing the bunching parameter defined by Eq. (6) with respect to d, because in this way the emission at $\hat{\omega}_1$ and $\hat{\omega}_2$ would be oversaturated (and, therefore, suppressed) slightly after the entrance of the electron beam inside the radiator. Instead, one should set R_{56} so as to reach maximum bunching in the last part of the radiator.

When the FEL is operated in a low-gain regime, the additional contribution to R_{56}, created within the radiator at a longitudinal distance z, can be written as $2N(z)\lambda_{\mathrm{rad}}$, where $N(z)$ is the number of radiator periods after the longitudinal distance z and $\lambda_{\mathrm{rad}} = 2\pi c/\omega_{\mathrm{rad}}$ is the radiator resonant wavelength.

The value of R_{56} optimizing the emission at $\hat{\omega}_1$ and $\hat{\omega}_2$, called $(R_{56})_{\mathrm{opt}}$, is given by

$$(R_{56})_{\mathrm{opt}} \simeq (R_{56})_{\max} - 2\left(\frac{1}{2}N_u\right)\lambda_{\mathrm{rad}}, \tag{7}$$

where $(R_{56})_{\max}$ is the value of R_{56} obtained by maximizing the bunching defined by Eq. (6). In the previous relation we have set $N(z) = 1/2N_u$. This in practice means that, according to Eq. (7), we impose that the two portions of electron beam emitting at $\hat{\omega}_1 = n\omega_1$ and $\hat{\omega}_2 = n\omega_2$ reach maximum bunching after propagating through half of the radiator [21]. The combination of the previous equations provides a simple recipe on how to use the strength of the dispersive section as a control parameter to generate, for a given chirp, two subpulses with fixed (decided) spectral distance. Let's now focus on the temporal domain. For constant $|\hat{\Delta}\omega|$, the temporal distance between the two peaks depends on $\bar{t}$, which is determined by the chirp parameter Γ_I according to Eq. (3).

In conclusion, the generation of two subpulses with constant spectral distance and variable (adjustable) temporal separation can be be obtained by varying Γ_I and adjusting the strength of the dispersive section as prescribed by Eqs. (3)–(7).

As a practical example, let's consider again the case of the FERMI@Elettra FEL. Let's fix, for instance, $\hat{\Delta}\lambda (= 2\pi c/\hat{\Delta}\omega) = 0.15$ nm, corresponding to $\alpha B = \hat{\Delta}\lambda/\lambda_{\mathrm{rad}} = 4.6 \times 10^{-3}$. Considering the three chirp configurations reported in the caption of Fig. 4, making use of Eqs. (3)–(7) and of the parameters reported in the caption of Fig. 3, with $P_0 = 600$ MW, one finds [22] $(R_{56})_1 = 29.3\ \mu$m, $(R_{56})_2 = 24.1\ \mu$m, and $(R_{56})_3 = 21.8\ \mu$m. Carrying out simulations with these values using the numerical code GENESIS [23], one gets the spectral and temporal pulses reported in Fig. 4. As can be seen, the agreement between simulations and theory is quite satisfactory. Indeed, as shown in the left panel, the spectral distance between subpulses is close to 0.15 nm for all chirp values. As expected (see the right panel), the subpulses in the time domain are separated by different (adjustable) distances, depending on the chirp parameter Γ_I. The temporal duration depends on the local profile of the seed around the selected time $\bar{t}$: relatively flat local profiles (corresponding to $\bar{t}$ values located at seed tails) correspond to relatively long subpulses, while steeper local profiles (corresponding to $\bar{t}$ values closer to seed center) will turn into shorter subpulses. In the absence of chirp, longer pulses should correspond to narrow bandwidths, and vice versa. However, this trend is counteracted by the presence of the chirp on the seed. The latter is stronger in the case of longer subpulses and weaker when subpulses are shorter. As a net effect [see left panel of Fig. 4(a)], the spectral bandwidth of subpulses remains practically constant for all chirps.

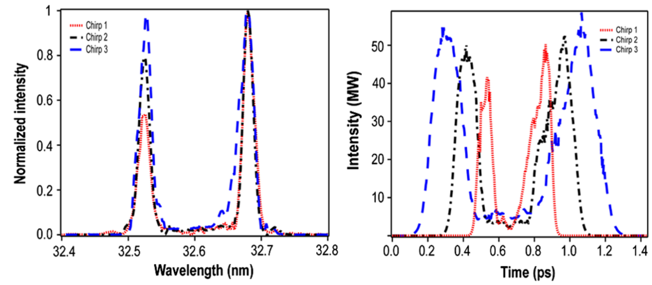

FIG. 4 (color online). Spectral and temporal FEL output for different chirp configurations of the seed pulse: $(\Gamma_R, \Gamma_I)_1 = (3.18 \times 10^{-5}\ \mathrm{fs}^{-2}, -4.33 \times 10^{-5}\ \mathrm{fs}^{-2})$, $(\Gamma_R, \Gamma_I)_2 = (1.08 \times 10^{-5}\ \mathrm{fs}^{-2}, -2.94 \times 10^{-5}\ \mathrm{fs}^{-2})$ and $(\Gamma_R, \Gamma_I)_3 = (5.13 \times 10^{-6}\ \mathrm{fs}^{-2}, -2.1 \times 10^{-5}\ \mathrm{fs}^{-2})$. For the simulation (carried out with the code GENESIS), we have used a set of parameters similar to the one listed in the caption of Fig. 3. Here $P_0 = 600$ MW.

According to simulations, the relative intensity of the two subpulses can be easily controlled by tuning the strength of the radiator (data not shown).

Finally, we remark that we have also performed simulations including possible realistic distortions of the electron-beam distribution (e.g., a current variation along the bunch, or residual linear and quadratic chirps in the energy profile). Obtained results (not reported here) clearly show that the FEL spectrotemporal splitting induced by the chirped seed is particularly robust. This holds in turn for the possibility of exploiting the observed splitting to carry out pump-probe experiments.

In this Letter, we studied an FEL regime in which electrons are seeded by a powerful chirped laser pulse. As a result of this interaction, the FEL output is split in two separated pulses, both in time and in spectrum. By exploiting the interplay between seed chirp and FEL parameters, we proposed a method for controlling the temporal distance between subpulses, while maintaining fixed spectral distance. For the considered parameters, the maximum temporal distance between subpulses is of the order of several hundreds of femtoseconds, their maximum relative spectral separation is of the order of few percent. The analytical prediction has been successfully benchmarked with numerical simulations carried out for the case of the FERMI@Elettra FEL. The latter are in very good agreement with experiments. The proposed method opens up the possibility of performing two-color pump-probe experiments in the VUV or x-ray domain using the FEL light, both as a pump and as a probe.

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