

Probing Atoms by Periodically Modulated Electron Bunches

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When passing through an undulator in a free electron laser, dense bunches of relativistic electrons split into microbunches, attaining a periodic space-time structure. We show that the field of such periodically modulated bunches is tremendously influenced by coherence effects, resulting in a novel type of beam-atom interaction. Our results indicate that employing such bunches (alone or in combinations with the radiation they emit) offers a multitude of new opportunities for exploring atomic dynamics on a femtosecond timescale.

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Interactions of photons and charged particles with matter belong to the basic processes studied by physics and the results of these studies have numerous important applications in other fields of research, ranging from astrophysics to biology and chemistry.

Until recently beams of charged particles (electrons, ions) had relatively low densities and their collisions with atomic systems were driven by interactions with individual particles of the beam [1–5]. However, today high-density bunches of extreme relativistic electrons are generated by conventional and laser or plasma wakefield accelerators [6–9]. When colliding with an atomic target a significant fraction of bunch electrons may hit the target within its transition time. As a result, these electrons could coherently interact with the target, which strongly affects the target dynamics [10].

In a free electron laser (FEL) a dense bunch of highly relativistic electrons from a linear accelerator passes through an undulator, generating nearly monochromatic intense radiation [11–14]. When passing through an undulator the bunch—due to the interaction with the magnetic field of the undulator and the emitted radiation—splits into microbunches, attaining a periodic space-time structure of a “diffraction grating” that moves with an extreme relativistic velocity. According to our estimates, depending on the bunch and undulator parameters, the diffraction grating may keep its shape over distances from several meters to several tens of meters after leaving the undulator.

In this Letter we show that, due to exceptional properties of such grating projectiles—a periodic structure, a large

number (up to $\sim 10^6$ – 10^7) of electrons in a microbunch, and a very short ($\lesssim 100$ as) microbunch duration—their collisions with atoms are driven by a novel type of beam-atom interaction. Its novelty originates in a multilevel coherence, where a coherent action of the microbunches reshapes the high-frequency part of the bunch field into nearly monochromatic lines, whose intensity is strongly amplified by a coherent action of electrons within each microbunch, and a coherent action of the entire bunch creates an extremely intense maximum at low frequencies. Exploiting this interaction offers new opportunities for exploring atomic dynamics on very short timescales.

The field of a highly relativistic charged particle moving with a *constant* velocity can be thought of as consisting of the so called “equivalent photons” [15–17]. Such photons may closely resemble real photons (even though fundamental differences between them still remain [18]). We will show that the high-frequency part of the bunch-atom interaction is transmitted by coherent equivalent (CE) photons whose energies coincide with energies of the real photons emitted by the bunch in the undulator and whose intensities can be made close to those of the real photons.

Let the target atom, which is at rest in the laboratory frame, be bombarded by a bunch of extreme relativistic electrons, which move in the laboratory frame with a velocity $\mathbf{v}$ along the z axis, $\mathbf{v} = (0, 0, v)$. We shall suppose that the bunch was modulated along this axis that transformed it into a train of equidistantly separated microbunches with a similar shape.

The cross section ($d\sigma_{\text{fi}}/d^2\mathbf{q}_{\perp}$) for atomic transition, differential in the transverse momentum transfer $\hbar\mathbf{q}_{\perp}$ ($\mathbf{q}_{\perp} \cdot \mathbf{v} = 0$) to the atom, is given by (see End Matter)

$$\frac{d\sigma_{\text{fi}}}{d^2\mathbf{q}_{\perp}} = \frac{d\sigma_{\text{fi}}^{(1\text{ el})}}{d^2\mathbf{q}_{\perp}} \times \left\{ |F(\mathbf{q})|^2 + N_t \right\}. \quad (1)$$

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Here, $(d\sigma_{\text{fi}}^{(1\text{ el})}/d^2\mathbf{q}_{\perp})$ is the cross section in the collision with a single electron from the bunch. It reads

$$\frac{d\sigma_{\text{fi}}^{(1\text{ el})}}{d^2\mathbf{q}_{\perp}} = \frac{4e_0^4}{\hbar^2 v^2} \frac{|\langle\psi_f|e^{i\mathbf{q}\cdot\mathbf{r}} - \frac{v}{2m_e c^2} \left(e^{i\mathbf{q}\cdot\mathbf{r}} \hat{p}_z + \hat{p}_z e^{i\mathbf{q}\cdot\mathbf{r}} \right) |\psi_i\rangle|^2}{\left(q^2 - \frac{\omega_{\text{fi}}^2}{c^2} \right)^2}, \quad (2)$$

where e_0 and m_e are the electron charge and mass, respectively, $\mathbf{r} = (x, y, z)$ is the coordinate of the atomic electron with respect to the atomic nucleus, $\hat{p}_z = (\hbar/i)[\partial/\partial z]$, ψ_i and ε_i (ψ_f and ε_f) are the initial (final) atomic state and its energy, respectively, $\omega_{\text{fi}} = (\varepsilon_f - \varepsilon_i)/\hbar$ is the atomic transition frequency, $\hbar\mathbf{q} = (\hbar\mathbf{q}_{\perp}, \hbar q_{\parallel})$ is the momentum transfer to the atom, $\hbar q_{\parallel} = \hbar\omega_{\text{fi}}/v$ is the minimum momentum transfer, and c is the speed of light.

Further, N_t is the total number of electrons in the bunch and $F(\mathbf{q})$ is the elastic form factor of the bunch,

$$F(\mathbf{q}) = \int d^3\mathbf{R} \rho(\mathbf{R}) e^{-i\mathbf{q}\cdot\mathbf{R}}, \quad (3)$$

where $\rho(\mathbf{R})$ is the bunch density and the integration runs over the bunch volume [19]. In (1) the terms $\sim N_t$ and $\sim |F(\mathbf{q})|^2$ correspond to the independent (incoherent) and coherent, respectively, action of the bunch electrons in the collision. The coherent channel may become very efficient when there is a large number of electrons in the coherence volume $V_c \simeq (\pi/q_{\perp}^2 q_{\parallel})$ [10].

If the bunch is a train of equidistantly separated microbunches with a similar shape, we obtain (see End Matter)

$$F(\mathbf{q}) = \mathcal{F}(\mathbf{q}) \mathcal{G}(\mathbf{q}), \quad (4)$$

where

$$\mathcal{F}(\mathbf{q}) = \int d^3\mathbf{R} \rho_0(\mathbf{R}) e^{-i\mathbf{q}\cdot\mathbf{R}} \quad (5)$$

is the elastic form factor of a single microbunch with the density $\rho_0(\mathbf{R})$ and

$$\mathcal{G}(\mathbf{q}) = \sum_k f_{k,0} e^{-ikq_{\parallel} \mathcal{L}} \quad (6)$$

is the structure factor of the bunch. In (6) the sum runs over the microbunches, $\mathcal{L}$ is the distance between them, and $f_{k,0}$ are coefficients describing the relative intensities of the microbunches. This factor arises due to the interference between the contributions to the atomic transition amplitude from the interaction with different microbunches. Its form closely resembles that of the factor describing a multislit interference of light scattered off a diffraction grating. In our

case the effective grating arises due to a periodic structure of the electron bunch [20].

The factor $|\mathcal{G}|^2$ reaches its maximum at $q_{\parallel} \mathcal{L} = 2\pi n$, where $n = 1, 2, 3, \dots$ which corresponds to resonant frequencies, $\omega_n = (2\pi/T)n$, where $T = \mathcal{L}/v$ is the time interval between the arrival of two consecutive microbunches. When the number of the microbunches is very large and $f_{k,0}$ varies slowly with k , $|\mathcal{G}|^2$ strongly peaks at the resonant frequencies.

In what follows we shall consider electron bunches by relying on parameters of two high-repetition-rate superconducting FEL facilities, FLASH [21] and the European x-ray free electron laser (XFEL) [22]. The former, which is driven by bunches with electron energies 0.5–1.35 GeV, operates in the extreme ultraviolet and soft x-ray regimes and comprises two undulator lines. One of these has been upgraded to a high-repetition-rate seeded FEL line [23] and is expected to deliver fully coherent photon pulses for user operation in the near future. For simplicity, we consider a special regime proposed to improve seeding performance, namely two-bunch seeding [24], in which only the fundamental harmonic of the FEL undulator is imprinted on the electron bunch [25].

Let us consider the ionization of H(1s) atoms by a (FLASH) electron bunch with an electron energy of 1.35 GeV, a total charge of 250 pC (corresponding to $N_t = 1.6 \times 10^9$ electrons) and a bunch length of $L = 30 \mu\text{m}$. The bunch passed through a linearly polarized undulator with the period $\lambda_U = 3.5 \text{ cm}$ and deflection parameter $\mathcal{K} = 3.8$. With an average beta function of 7 m in the undulator and the normalized emittance [6] of 0.5 mm mrad, the bunch radius is $a_0 = \sqrt{2}\sigma_x = 51 \mu\text{m}$. In the undulator the electron bunch splits into microbunches separated along the bunch propagation by the fundamental wavelength of the emitted radiation [11]. As our estimates show, after leaving the undulator the bunch keeps this periodic structure over distances of several meters [26,27].

We shall assume that the shape of the electron microbunches is Gaussian,

$$\rho_k(\mathbf{R}) = \frac{\mathcal{N}_k}{\pi^{3/2} a_0^2 l_0} e^{-Z^2/l_0^2} e^{-R_{\perp}^2/a_0^2}, \quad (7)$$

where $\mathcal{N}_k$ is the number of electrons in the k th microbunch, and l_0 and a_0 are the effective length and radius, respectively, of the microbunches [28]. The coefficients $f_{k,0}$ in (5) are chosen to reflect the Gaussian longitudinal profile of the entire bunch.

The chosen bunch parameters result in the value of $\lambda_1 \approx 2 \times 10^{-6} \text{ cm}$ for the fundamental wavelength, given by $\lambda_1 = [\lambda_U/2\gamma^2](1 + 0.5\mathcal{K}^2)$ [11] with $\gamma = 1/\sqrt{1 - v^2/c^2}$, for the electromagnetic radiation emitted by the bunch along its propagation direction. The fundamental frequency $\omega_1 = (2\pi v/\lambda_1)$ corresponds to the energy $\hbar\omega_1 \approx 60 \text{ eV}$.

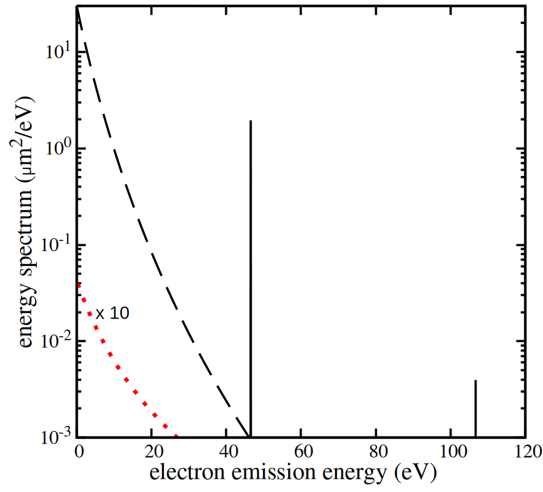


FIG. 1. Energy spectrum of electrons emitted in ionization of H(1s) atoms by a bunch of 1.35 GeV electrons passed through an undulator. $N_t = 1.6 \times 10^9$, $L = 30 \mu\text{m}$, $a_0 = 51 \mu\text{m}$, $\lambda_U = 3.5 \text{ cm}$, $\mathcal{K} = 3.8$, and $l_0 = 0.005 \mu\text{m}$. Dotted curve: all bunch electrons act incoherently (multiplied by 10). Dashed curve: coherence within a microbunch is taken into account but the microbunches act incoherently. Solid curve: coherences within a single microbunch and of different microbunches are taken into account. The cross sections are given *per bunch*.

We further obtain that the effective number of the microbunches $\simeq L/\lambda_1 = 2 \times 10^3$ and the number $\mathcal{N}_k$ of electrons in the most intense microbunches is $\simeq 7 \times 10^5$. The length l_0 of a microbunch is proportional to λ_1 : $l_0 = \eta\lambda_1$. We take $\eta = 0.25$, which yields $l_0 \simeq 5 \times 10^{-7} \text{ cm}$ (the corresponding microbunch duration is $\approx 17 \text{ as}$).

In Fig. 1 we show results of three different calculations for the energy spectrum of the emitted electrons (represented by the ionization cross-section differential in the emission energy): (i) the dotted curve was obtained by assuming that all 1.6×10^9 bunch electrons interact with the hydrogen atoms individually (in order to make this spectrum more visible, in the figure it was multiplied by 10); (ii) the dashed curve accounts for the coherent action of the electrons within each microbunch but assumes that the microbunches act incoherently; (iii) the solid curve was obtained by taking into account the coherent action not only within each microbunch but also across all microbunches.

Figure 1 demonstrates that the coherence within a microbunch strongly enhances the ionization process whereas the coherent action of the microbunches drastically changes the shape of the emission spectrum by compressing it into sharp peaks separated by the energy $2\pi\hbar(v/\lambda_1)$. This points to the presence of strong (quasi) monochromatic lines $\omega_n = (2\pi v/\lambda_1)n$ ($n = 1, 2, 3, \dots$) in the energy spectrum of the CE photons carried by the electron bunch passed through the undulator.

In Fig. 2 we show the energy spectrum of equivalent photons for two electron bunches. One of them is the bunch considered above. The other can be operated at the

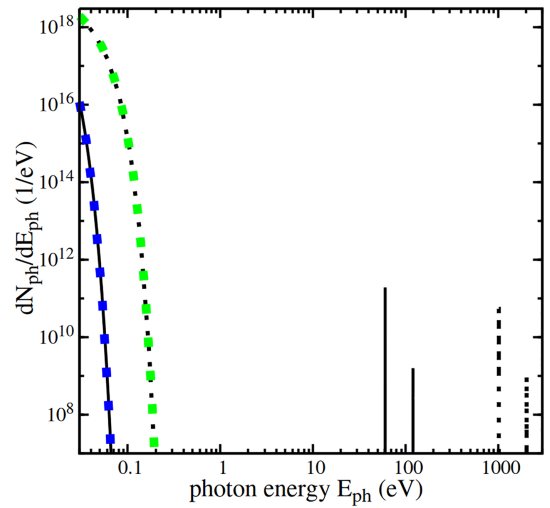


FIG. 2. The energy spectrum of equivalent photons carried by 1.35 GeV (solid curve) and 14 GeV (dashed curve) electron bunches (for each bunch only the first two high-frequency lines are shown). The spectra of equivalent photons, carried by the corresponding unmodulated bunches, are shown by square markers. For more explanations see text.

European XFEL, which produces coherent x-ray radiation at three undulator lines driven by a high-energy electron bunch with energies up to 16.3 GeV. The line SASE3 operates in the soft-x-ray regime, enabled by its large undulator period of $\lambda_U = 6.8 \text{ cm}$ and high deflection parameter. We consider an electron energy of 14 GeV and $\mathcal{K} = 7.2$, resulting in a photon energy of $\approx 1 \text{ keV}$. The electron bunch parameters are the same as in the FLASH example, except for a shorter length of $10 \mu\text{m}$ and a smaller radius $a_0 = 24 \mu\text{m}$ (assuming an average beta function of 16 m in the undulator). The considered undulator line operates in the self-amplified spontaneous emission (SASE) regime, which provides only partial longitudinal coherence [14]. Full longitudinal coherence can be achieved using a self-seeding scheme [29,30]. Here, for illustration, we consider the fully coherent case.

Two main features of the photon spectrum are seen in Fig. 2: (i) narrow high-frequency lines of CE photons (for each bunch only the first two are shown), whose positions practically coincide with those of the real photons emitted in the undulator (not shown), and (ii) an extremely intense maximum at very low energies, $\hbar\omega_{\text{ph}} \lesssim \hbar v/L$.

The analysis shows that the integral intensity of each high-frequency line is inversely proportional to the length L of the electron bunch [31] and that the dependence on the bunch radius a_0 is more complicated, varying from $\sim a_0^{-4}$ at $a_0 \gg v\gamma/\omega_n$ to $\sim \ln(1/a_0)$ at $a_0 \ll v\gamma/\omega_n$ [32]. The a_0 dependence is illustrated in Fig. 3.

The maximum at very low frequencies contains equivalent photons with wavelengths comparable to or exceeding the bunch length L and, therefore, its shape [33] is not sensitive to microbunching (see Fig. 2). This maximum is

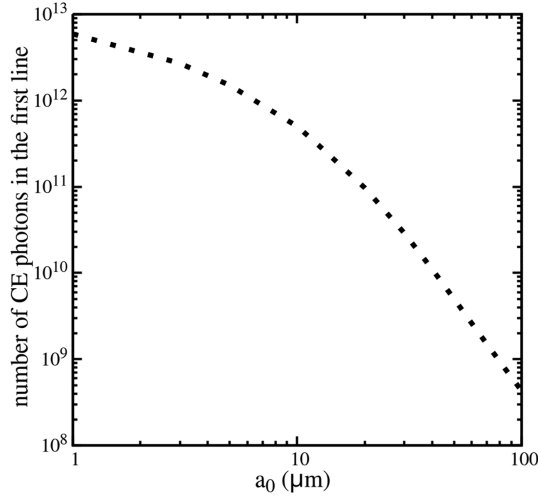


FIG. 3. The number of CE photons in the line centered at $\hbar\omega_1 \approx 60$ eV as a function of the bunch radius a_0 . The other parameters are $N_t = 1.6 \times 10^9$, electron energy of 1.35 GeV, $L = 30$ μm , $\lambda_U = 3.5$ cm, $\mathcal{K} = 3.8$, and $l_0 = 0.005$ μm .

so extremely intense because the entire bunch coherently contributes to it, making its intensity proportional to N_t^2 .

Our calculations for the photon spectrum differential in the distance b from the bunch axis show that even very large b ($b > 100a_0$, [34]) noticeably contribute to the low-frequency maximum. Therefore, its enormous intensity does not necessarily imply strong fields because the low-frequency photons are spread over a large volume.

Single-pulse (unmodulated) electron bunches, whose duration greatly exceeds typical atomic times ($\tau_A \approx 24$ as), can ionize atoms via collisions with individual electrons and tunneling. Tunneling is a coherent mechanism but differs qualitatively from coherent impact ionization [10], which is efficient for short ($\lesssim 10\tau_A$) single-pulse bunches, proceeding via absorption of equivalent photons whose energy exceeds the atomic binding energy.

As our calculations show [35,36], for the bunch parameters in Fig. 1 the tunneling ionization of H(1s) is negligible. However, since the tunneling ionization rate, $\sim (F_a/F) \exp(-2F_a/3F)$ [37] (F is the low-frequency bunch field and $F_a \sim 10^9$ V/cm is the characteristic atomic field), strongly depends on F , this mechanism is extremely sensitive to the bunch parameters [10]. We also note that long before the bunch field reaches values where the tunneling becomes possible, it can already efficiently mix closely lying bound atomic states.

The contributions of tunneling, absorption of high-frequency CE photons, and collisions with individual electrons to the ionization of H(1s) by the 14 GeV bunch are shown in Fig. 4. There we see an enormous increase in the tunneling ionization with decreasing a_0 [38] and inefficiency of ionization via absorption of the CE photons because their energies ($\gtrsim 1$ keV) greatly exceed the

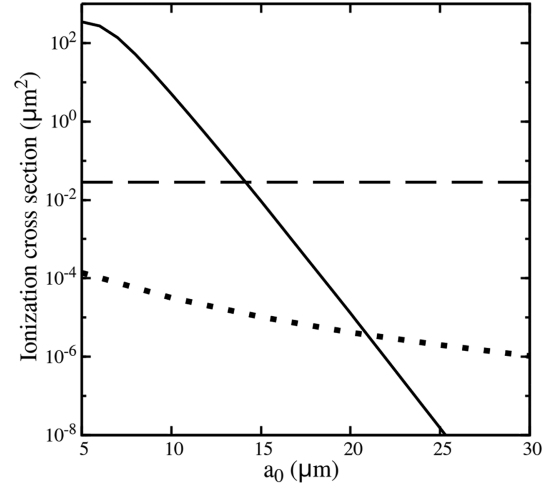


FIG. 4. Ionization of H(1s) by 14 GeV electron bunch as a function of the bunch radius. $N_t = 1.6 \times 10^9$, $L = 10$ μm , $\lambda_U = 6.8$ cm, $\mathcal{K} = 7.2$. Solid curve, tunneling ionization; dashed curve, ionization by N_t individual electrons; dotted curve, ionization via absorption of a CE photon from the first high-frequency line.

hydrogen binding energy (≈ 13.6 eV), making the absorption unlikely [39,40].

High-frequency CE photons are appropriate for exploring multiply charged ions [40] or many-electron atoms. For example, the first CE line, generated by the 14 GeV electron bunch (see Fig. 2) colliding with Ne atoms, would create a vacancy in their K shell with the cross section ≈ 0.053 μm^2 . The emission of a K -shell electron triggers Auger and shakeoff processes, leading to the production of Ne^{2+} and Ne^{3+} ions. Assuming the target density of $\approx 10^{15}$ cm^{-3} , its longitudinal length of ≈ 1 cm and the bunch repetition rate of $\approx 10^3$ Hz, we obtain $\approx 5 \times 10^8$ for the number of such events per second [41].

Thus, an electron bunch, passed through an undulator, carries an electromagnetic field, where nearly monochromatic high-frequency CE photons “coexist” with a field that slowly varies in time [42]. Such a—perfectly synchronized—field combination can, for instance, be used for exploring many-electron atoms where inner and outer electrons are accessed via the high-frequency photons and the low-frequency field, respectively, or studying nondipole transitions in hydrogenlike ions via resonant absorption of a high-frequency CE photon and coupling of nearly degenerate excited states by the low-frequency field. The low-frequency field can also noticeably influence the spectra of electrons emitted from atoms by absorption of high-frequency CE photons [43], which provides additional information about the emission process.

Intensities of high-frequency CE photons can be further strongly enhanced by reducing the radius of the electron bunch. This can, for instance, be done by installing at the undulator exit a quadrupole triplet that focuses the electron

beam by a factor of $\simeq 4$, corresponding to a beta function at the waist of 0.5 m. The total length of the system would only be a few meters.

Our estimates show that for both electron bunches under consideration the radius can be reduced by 4 to 5 times without spoiling their periodic structure. This increases the intensity of high-frequency CE photon lines by 2 orders of magnitude (for an illustration see Fig. 3).

The high-frequency CE photons closely resemble the field of the emitted radiation. However, important differences still hold, including a fundamental one: unlike for a real photon field, the integral $\int_{-\infty}^{+\infty} \mathbf{E}_{\perp} dt$ (where $\mathbf{E}_{\perp}$ is the transverse part of the electric field) does not vanish for the CE photon field. Also, polarization properties can be quite different [44]. This, in general, implies additional differences in the response of atomic systems when they are probed by real and equivalent photons produced by the same electron bunch.

By separating the electron bunch from the emitted radiation at the undulator exit (while preserving its density modulation), the bunch can be used for probing atomic systems on an fs timescale. Several approaches for implementing such a separation have been considered [45], including an achromatic bend or a compact chicane combined with an absorber or deflector for the radiation [46].

A key advantage of the electron beams over real photon beams generated at XFEL facilities is the ability to combine multiple spectral components within a single pulse. Beyond the combination of high- and low-frequency components, additional intermediate frequency bands can be produced through laser-based manipulation in XFEL injectors [47,48], two-color seeding schemes [49] with frequency-mixing techniques [50], or controlled shaping of the electron density distribution in bunch compressors [21].

A pump-probe scheme involving a beam of real photons and a short ($\simeq 250$ as, the corresponding bandwidth $\simeq 2.6$ eV) single-pulse electron bunch was proposed in [51]. Compared to it, the present concept possesses a number of fundamental differences [52], including the fact that here all spectral components can be realized using equivalent photons alone. Moreover, employing an electron bunch with a periodic space-time structure, distinct spectral components can be generated in different temporal slices of the same electron bunch with a controllable delay, enabling pump-probe measurements within a single shot [53]. This unified approach simplifies the experimental layout and facilitates rapid reconfiguration between different measurement modes.

One can also probe atomic systems by various coherent combinations of CE photons with the emitted radiation. Here, various scenarios are possible. For instance, one can shift azimuthal symmetry axes of beams of equivalent and real photons with respect to each other, producing a high-frequency field with nontrivial polarization. Beams of equivalent and real photons can also be combined at

different angles, enabling multiwave mixing [54], controlled interference effects, and the generation of complex polarization structures.

We note that our proposal is also relevant to the more common SASE regime. In this case, the spectral width increases proportionally to the number of longitudinal modes in the SASE pulse and the pattern changes randomly shot to shot [14], while the integrated spectral intensity remains comparable to that in the seeded case.

In conclusion, in FELs, bunches of extreme relativistic electrons pass through an undulator, attaining a periodic space-time structure that may survive over considerable distances after leaving the undulator. We have shown that the interaction of such bunches with atoms is tremendously modified by a multilevel coherence, where a coherent action of the microbunches transforms the high-frequency part of the bunch field into nearly monochromatic lines, a coherent action of electrons within each microbunch strongly amplifies intensities of these lines and a confluence of these two coherences occurs at low frequencies, resulting in an extremely intense maximum, created by a coherent action of the entire bunch.

In FELs, electron bunches are employed for generating electromagnetic radiation. Our results indicate that such bunches can further be exploited (alone or in coherent combinations with the radiation they emit), offering a multitude of new opportunities for exploring atomic dynamics on a femtosecond timescale.

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Data availability—The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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- [32] The variation of the dependence on the beam radius from $\sim a_0^{-4}$ at $a_0 \gg v\gamma/\omega$ to $\sim \ln(1/a_0)$ at $a_0 \ll v\gamma/\omega$, where ω is the frequency of the equivalent photon, is similar both for periodically modulated electron beams and for beams consisting of just a single bunch. For the discussion of the dependence on the beam radius a_0 in the latter case, see Ref. [10].
- [33] The shape is mainly determined by the factor $\exp(-\omega_{ph}^2 L^2/v^2)$.
- [34] In our consideration we neglect electrodynamic environment but note that $b \sim 100a_0$ would already approach the radius of vacuum chamber that prevents from considering larger values and strongly modifies the low-frequency part of the spectrum.
- [35] Tunneling ionization is a highly nonperturbative process and, in order to calculate its cross section, we used the approach described in detail in [10], which employs tunneling rate expressions from [36] valid for ionization by a constant or slowly varying electric field.
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- [38] In the FLASH case the tunneling is negligible because the electron bunch is longer and broader than that in the XFEL case: this reduces the field strength, having a dramatic effect on the tunneling rates due to their extreme high sensitivity to the field strength. Since the tunneling rate is also extremely

sensitive to the typical interatomic field, an increase in the binding energy of atomic electrons also has a dramatic effect on the tunneling ionization: for instance, replacing hydrogen by helium reduces the tunneling cross section, shown in Fig. 4, by several orders of magnitude. However, even though one can expect that the low-frequency field will normally have a very modest (direct) influence on atomic ionization, effects of a concerted action of the high- and low-frequency fields may still remain, since even a relatively weak low-frequency field can be very efficient in mixing closely lying atomic states with opposite parities.

- [39] The cross section for photoionization of H(1s) by a ≈ 1 keV photon is by 6 orders of magnitude smaller than by photons whose energies are just slightly above the ionization threshold (13.6 eV) of H(1s); see, e.g., Fig. 66.2 on page 969 of *Springer Handbook of Atomic, Molecular, and Optical Physics*, 2nd ed., by G. W. F. Drake (Springer, Cham, 2023).
- [40] For an illustration of how the ratio between the ionization by the absorption of the first high-energy CE line and by the individual action of the bunch electrons varies with the change in the binding energy of the target electron we present here the corresponding cross sections (in μm^2) in collisions with the bunch, whose parameters are given in Fig. 4 and assuming that $a_0 = 10 \mu\text{m}$: $\sigma_{\text{CE}} = 2.5 \times 10^{-4}$ and $\sigma_{\text{ind}} = 4.5 \times 10^{-2}$ for He(1 s²); $\sigma_{\text{CE}} = 7.5 \times 10^{-4}$ and $\sigma_{\text{ind}} = 6.8 \times 10^{-3}$ for He⁺(1s); $\sigma_{\text{CE}} = 4.4 \times 10^{-3}$ and $\sigma_{\text{ind}} = 2.9 \times 10^{-3}$ for Li²⁺(1s); $\sigma_{\text{CE}} = 1.43 \times 10^{-2}$ and $\sigma_{\text{ind}} = 1.63 \times 10^{-3}$ for Be³⁺(1s); $\sigma_{\text{CE}} = 3.45 \times 10^{-2}$ and $\sigma_{\text{ind}} = 1.0 \times 10^{-3}$ for B⁴⁺(1s); $\sigma_{\text{CE}} = 6.9 \times 10^{-2}$ and $\sigma_{\text{ind}} = 7.0 \times 10^{-4}$ for C⁵⁺(1s); $\sigma_{\text{CE}} = 0.12$ and $\sigma_{\text{ind}} = 5.1 \times 10^{-4}$ for N⁶⁺(1s); $\sigma_{\text{CE}} = 0.19$ and $\sigma_{\text{ind}} = 3.9 \times 10^{-4}$ for O⁷⁺(1s). The number of the CE photons in the first line ($\hbar\omega_1 \approx 1$ keV) is $\approx 3 \times 10^{10}$ and the line energy is $\approx 3 \times 10^{13}$ eV. We also note that for all the above targets the tunnel ionization is negligible.
- [41] In this case the tunneling ionization is very weak, but the atomic electrons can also be removed by collisions with individual electrons. Our rough estimates for the cross section for the removal of a 2p (2s) electron yield $\approx 0.1 \mu\text{m}^2$ ($\approx 0.015 \mu\text{m}^2$). For the cross section for the removal of an electron from the K shell we obtain $\approx 7.8 \times 10^{-4} \mu\text{m}^2$, which is by 2 orders of magnitude smaller than the cross section for the removal of such an electron by the absorption of the CE photon from the first line (≈ 1 keV). We also note that the removal of a 2s electron from Ne does not trigger Auger process (it is not allowed energetically) and, therefore, is unlikely to lead to the production of Ne²⁺ ions.
- [42] Compared to the high-frequency CE lines, the field, which is generated by individual electrons, can also be regarded as an effectively low-frequency field. For a simple discussion of properties of such a field, expressed in terms of equivalent photons, see, e.g., Ref. [17].
- [43] Our results for the emission spectra, shown in Fig. 1, are obtained within the first-order perturbation theory in the interaction with the bunch field. This approximation does not take into account that the emitted electrons can be influenced by the field of the bunch (predominantly by its low-frequency part).
- [44] For instance, electron bunches with azimuthal symmetry generate equivalent photons that are polarized along the bunch radius.
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- [46] The impact of the chicane on longitudinal dynamics can be optimized by operating the FEL at the onset of saturation and avoiding smearing effects associated with the uncorrelated energy spread.
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- [52] As was stressed in the introduction, the field of an electron beam with a periodic space-time structure is tremendously influenced by a multilevel coherence. As a result, such beams affect atoms via a kind of “3-in-1” action: the synchronized interactions with (i) high-frequency CE photons and (ii) the low-frequency field of the beam, which both are coherently created by the beam electrons, are superimposed on (iii) the “background” interaction with the individual electrons. The first two very closely resemble the interactions with intense laser fields, whereas the third one proceeds in the standard regime of collisions with charged particles. By playing with the relative intensities of these channels one can, in particular, bridge the gap between the “standard” collisions and the interaction with intense laser fields.
- [53] See, for example, A. A. Lutman *et al.*, *Nat. Photonics* **10**, 745 (2016).
- [54] F. Bencivenga, R. Cucini, F. Capotondi, A. Battistoni, R. Mincigrucci, E. Giangrisostomi, A. Gessini, M. Manfredda, I. P. Nikolov, E. Pedersoli, E. Principi, C. Svetina, P. Parisse, F. Casolari, M. B. Danailov, M. Kiskinova, and C. Masciovecchio, *Nature (London)* **520**, 205 (2015).
- [55] The results for the cross sections do not depend on the choice of the origin: it can be an arbitrary point that is at rest in the laboratory frame.
- [56] L. I. Schiff, *Rev. Sci. Instrum.* **17**, 6 (1946).

End Matter

Here, we briefly sketch the derivation of the cross section (1). Let an atom be at rest in the laboratory frame and let the bunch electrons move in this frame with a velocity $\mathbf{v} = (0, 0, v)$. It was shown [10] that within the first-order perturbation theory in the bunch-atom interaction the cross section for the atomic transition $i \rightarrow f$ reads

$$\frac{d\sigma_{\text{fi}}}{d^2\mathbf{q}_{\perp}} = \frac{d\sigma_{\text{fi}}^{(1\text{ el})}}{d^2\mathbf{q}_{\perp}} \times \left| \sum_{j=1}^{N_t} e^{i\phi_j} \right|^2. \quad (\text{A1})$$

Here, $(d\sigma_{\text{fi}}^{(1\text{ el})}/d^2\mathbf{q}_{\perp})$ is the cross section in the collision with a single electron from the bunch,

$$\frac{d\sigma_{\text{fi}}^{(1\text{ el})}}{d^2\mathbf{q}_{\perp}} = \frac{4e_0^4}{\hbar^2 v^2} \frac{|\langle \psi_f | e^{i\mathbf{q}\cdot\mathbf{r}} - \frac{v}{2m_e c^2} (e^{i\mathbf{q}\cdot\mathbf{r}} \hat{p}_z + \hat{p}_z e^{i\mathbf{q}\cdot\mathbf{r}}) | \psi_i \rangle|^2}{\left(\mathbf{q}^2 - \frac{\omega_{\text{fi}}^2}{c^2} \right)^2}, \quad (\text{A2})$$

where e_0 and m_e are the electron charge and mass, respectively, $\mathbf{r} = (x, y, z)$ is the coordinate of the atomic electron with respect to the atomic nucleus, $\hat{p}_z = (\hbar/i)[\partial/\partial z]$, ψ_i and ε_i (ψ_f and ε_f) are the initial (final) atomic state and its energy, respectively, $\omega_{\text{fi}} = (\varepsilon_f - \varepsilon_i)/\hbar$ is the atomic transition frequency, $\hbar\mathbf{q} = (\hbar\mathbf{q}_{\perp}, \hbar q_{\parallel})$ is the momentum transfer to the atom, $\hbar q_{\parallel} = \hbar\omega_{\text{fi}}/v$ is the minimum momentum transfer, and c is the speed of light.

In (A1) $\phi_j = q_{\parallel} v t_j - \mathbf{q}_{\perp} \cdot \mathbf{R}_{\perp j}$, where $\mathbf{R}_{\perp j}$ is the transverse ($\mathbf{R}_{\perp j} \perp \mathbf{v}$) coordinate of the j th bunch electron and t_j is the time of its closest approach to the origin [55] and the sum runs over the bunch electrons. Following [56] for the term $|\sum_{j=1}^{N_t} e^{i\phi_j}|^2$, ($N_t \leq |\sum_{j=1}^{N_t} e^{i\phi_j}|^2 \leq N_t^2$), we obtain

$$\begin{aligned} \left| \sum_{j=1}^{N_t} e^{i\phi_j} \right|^2 &= N_t + (1 - 1/N_t) |F(\mathbf{q})|^2 \\ &\approx N_t + |F(\mathbf{q})|^2, \end{aligned} \quad (\text{A3})$$

where

$$F(\mathbf{q}) = \int d^3\mathbf{R} \rho(\mathbf{R}) e^{-i\mathbf{q}\cdot\mathbf{R}} \quad (\text{A4})$$

is the elastic form factor of the bunch, $\rho(\mathbf{R})$ is the bunch density, and the integration runs over the bunch volume (both the density and volume are taken at an arbitrarily fixed moment of time).

Let us suppose that the electron bunch is a train of microbunches whose density is given by

$$\rho(\mathbf{R}) = \sum_k \rho_k(\mathbf{R}_{\perp}, Z - \mathcal{Z}_k), \quad (\text{A5})$$

where ρ_k is the density of the k th microbunch and the sum runs over all microbunches. Then for the integral in (A4) we obtain

$$\int d^3\mathbf{R} \rho(\mathbf{R}) e^{-i\mathbf{q}\cdot\mathbf{R}} = \sum_k e^{-iq_{\parallel} \mathcal{Z}_k} \int d^3\mathbf{R} \rho_k(\mathbf{R}) e^{-i\mathbf{q}\cdot\mathbf{R}}. \quad (\text{A6})$$

If the distances $\mathcal{L}_{k+1,k} = \mathcal{Z}_{k+1} - \mathcal{Z}_k$ between two consecutive microbunches are random, then

$$\left| \int d^3\mathbf{R} \rho(\mathbf{R}) e^{-i\mathbf{q}\cdot\mathbf{R}} \right|^2 = \sum_k \left| \int d^3\mathbf{R} \rho_k(\mathbf{R}) e^{-i\mathbf{q}\cdot\mathbf{R}} \right|^2, \quad (\text{A7})$$

and the microbunches act in the collision individually.

If $\mathcal{L}_{k+1,k} = \mathcal{L}$ is constant (or varies with k very little) and the microbunches have a similar shape, $\rho_k = f_{k,0} \rho_0$ ($f_{k,0}$ are coefficients describing the relative intensities of the microbunches), then

$$\begin{aligned} |F(\mathbf{q})|^2 &= \left| \int d^3\mathbf{R} \rho(\mathbf{R}) e^{-i\mathbf{q}\cdot\mathbf{R}} \right|^2 \\ &= |\mathcal{F}(\mathbf{q})|^2 \times |\mathcal{G}(\mathbf{q})|^2, \end{aligned} \quad (\text{A8})$$

where

$$\mathcal{F}(\mathbf{q}) = \int d^3\mathbf{R} \rho_0(\mathbf{R}) e^{-i\mathbf{q}\cdot\mathbf{R}} \quad (\text{A9})$$

is the form factor of a single microbunch with the density ρ_0 and

$$\mathcal{G}(\mathbf{q}) = \sum_k f_{k,0} e^{-ikq_{\parallel} \mathcal{L}} \quad (\text{A10})$$

is the structure factor of the bunch.