Broadband high-resolution X-ray frequency combs

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Optical frequency combs have had a remarkable impact on precision spectroscopy1–3. Enabling this technology in the X-ray domain is expected to result in wide-ranging applications, such as stringent tests of astrophysical models and quantum electrodynamics4, a more sensitive search for the variability of fundamental constants5, and precision studies of nuclear structure6. Ultraprecise X-ray atomic clocks may also be envisaged7. In this work, an X-ray pulse-shaping method is proposed to generate a comb in the absorption spectrum of an ultrashort high-frequency pulse. The method employs an optical-frequency-comb laser, manipulating the system's dipole response to imprint a comb on an excited transition with a high photon energy. The described scheme provides higher comb frequencies and requires lower optical-comb peak intensities than currently explored methods8–10, preserves the overall width of the optical comb, and may be implemented using currently available X-ray technology11.

The spectrum of an optical frequency comb consists of equally spaced, precisely known peaks, centred at an optical frequency 12. Optical-comb generation was initially pursued via intracavity phase modulation12,23 and subsequently with stabilized mode-locked lasers14. Optical frequency combs are employed1, for example, in precision spectroscopy15, all-optical atomic clocks16, astronomical spectrographs calibration17, attosecond science18 and control of atomic coherence19. X-ray frequency combs would enable the aforementioned applications in the X-ray range. Stringent tests of fundamental physics may be pursued, such as accurate measurements of transition energies in highly charged ions, which are predicted to be more sensitive to the variability of fundamental constants20 than currently investigated species.

At present, extreme-ultraviolet (XUV) combs (∼30 eV) are generated by means of intracavity high-order harmonic generation (HHG)8,9. A femtosecond enhancement cavity is utilized to reach the required peak intensities8–10 of up to ∼1 × 1014 W cm−2. However, relativistic effects limit the efficiency of HHG at high harmonic orders20. The investigation of schemes to further increase the carrier frequency of the comb at accessible driving intensities is therefore required.

Short-wavelength light sources with improved brilliance and bandwidth11 enable studies of X-ray quantum optics, for example, in highly charged ions or nuclei14,6,21. Recently, an amplitude-shaping scheme was put forward to imprint a comb onto narrowband X-rays22. Comb generation was also suggested via quantum phase modulation24. However, these schemes are conditioned either by demanding requirements on the X-ray source bandwidth22, or by the spectral width of the emerging comb23. Here, in contrast, the predicted comb requires X-ray pulses as presently provided by free-electron lasers15 (FELs) and, being as wide as the employed optical frequency comb, is suitable to bridge the gap between a reference level and a nearby unknown level12,23.

The key idea behind our method is introduced by considering the three-level system of Fig. 1, and we then prove its viability in a realistic atomic implementation. The excited level of the high-energy transition 1 → 2 is coupled by an optical laser to a third metastable level. This is chosen such that it cannot be excited in a one-photon transition and decays via two-photon emission. Hence, the decay rate Γ 2 of this dark state is orders of magnitude lower than the decay rate Γ 1 of the bright (fast decaying) level 2. The system is described via the density operator P(t) of matrix elements p ij (t), where i,j ∈ {1, 2, 3}. The diagonal element p ii (t) describes the occupation probability of level i, whereas the off-diagonal element p ij (t) describes the coherence between states i and j. Atomic units are used throughout unless otherwise stated.

As shown in Fig. 1, an X-ray pulse (X1) excites the atomic system, whose dipole response is given by the coherence p 12(t). While propagating through the medium, the pulse interferes with the radiation Xout emitted by the driven system, thus creating gain or

Figure 1 | Three-level scheme used to describe the interaction between the model system and the driving fields. a, A low-density ensemble of ions, modelled as a three-level system, is driven by an ultrashort, broadband X-ray pulse (X1, solid blue) exciting the fast decaying level 2, followed by an optical pulse (L1, dashed red) coupling this excited state to the metastable state 3. The system is therefore prepared in an initial state that is a superposition of states 1 and 3. b, An optical frequency comb (L2, solid red) is subsequently used to periodically drive the optical transition 2 → 3. The emitted X-rays (Xout, dashed wavy blue) lead either to gain or attenuation of the incident pulse X1 while it propagates through the medium. This is detected via the absorption spectrum of the transmitted pulse X1. c, Input and output pulses from a and b. The pulses are linearly polarized along the z-direction and are co-propagating.

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Figure 2 | Time evolution and X-ray frequency comb comb absorption spectrum driven by a resonant optical frequency comb. The three-level system of Fig. 1 is used to describe Be$_2^{+}$ ions driven by a train of optical pulses tuned to the 2 → 3 transition. The driving comb is characterized by present-day parameters\textsuperscript{10}, that is, a single-pulse FWHM duration of 100 fs, a spectral width of 0.07 eV and a repetition frequency of 100 MHz. The peak intensity of 2.57 × 10\textsuperscript{10} W cm\textsuperscript{-2} corresponds to a 2π pulse area. a, Time evolution of $|\rho_2(t)|$ during the first 50 pulses of the optical frequency comb. Inset: $|\rho_2(t)|$ (blue, solid line) and $|\rho_2(t)|$ (red, dashed line) in the presence of the first pulse. b, Peak amplitudes of the frequency comb featured by the absorption spectrum, normalized to the maximum of the single-peak Lorentzian spectrum in the absence of optical control. The comb is centred at the X-ray transition energy $\omega_{21} = 123.7$ eV. c,d. Comb structure (c) and shape of a single comb tooth (d). For an X-ray pulse\textsuperscript{11} tuned to the transition energy $\omega_{21}$, with a duration of 100 fs, beam radius of 50 μm, peak intensity of 2.5 × 10\textsuperscript{12} W cm\textsuperscript{-2} and bandwidth of 1 eV, driving a sample of ions with density\textsuperscript{4} of 1 × 10\textsuperscript{4} cm\textsuperscript{-3} over a length of 2 cm, this corresponds to a power per comb line of ~30 pW (Supplementary Section IV), on the same order of magnitude as XUV combs generated via HHG\textsuperscript{10}.

Attenuation in the absorption spectrum

$$\sigma(\omega) \propto -\omega \text{Im}(\hat{\rho}_{12}(\omega))$$

of the transmitted pulse X1, with the frequency-dependent dipole response\textsuperscript{25} $\hat{\rho}_{12}(\omega) = \int_{-\infty}^{\infty} \rho_{12}(t) e^{-i\omega t} dt$. Here, $\sigma(\omega) > 0$ corresponds to absorption and $\sigma(\omega) < 0$ to gain.

If only the pulse X1 is used, spontaneous decay of the bright level follows its X-ray excitation. The resulting absorption spectrum, centred at the transition energy $\omega_{21}$ between states 2 and 1, exhibits a Lorentzian profile of width $\Gamma_2$. Based on previous experiments on time-domain control\textsuperscript{12,18}, we augment this X-ray-only scheme with two optical fields (Fig. 1a,b). The first optical pulse (L1 in Fig. 1a) depletes the fast decaying state 2 by transferring coherences and population into the metastable level 3. By preparing the system in a long-lived initial state that is a superposition of states 1 and 3, the fast decay of the system due to the large decay rate of level 2 is circumvented (Supplementary Section II). An optical frequency comb (L2 in Fig. 1b) is subsequently used to periodically modulate the emission out of the bright state, leading to a pulse train at the X-ray transition energy $\omega_{21}$.

To better understand this, we analyse the dynamics of the system interacting with the optical pulse train L2. The electric field $\vec{E}_{12}(t) = \vec{E}_{12}(t) \cos(\omega_{12} t) \hat{e}_z$ has envelope $\vec{E}_{12}(t)$ and carrier frequency $\omega_{12}$. The linear polarization $\hat{e}_z$, parallel to the dipole-moment matrix element $d_{23}$ between states 2 and 3, leads to the instantaneous Rabi frequency\textsuperscript{27} $\Omega_{R}(t) = d_{23} \vec{E}_{12}(t) \hat{e}_z$. The envelope $\vec{E}_{12}(t)$ consists of identical pulses, located at $t_j = t_0 + j T_p$, $j \in \mathbb{N}_0$, with repetition period $T_p$ and single-pulse FWHM duration $\tau \ll T_p$. The peak intensity $I$ corresponds to a maximum field
A train of off-resonant 1.5 eV pulses is used, with a detuning of \( \Delta = -0.52 \text{ eV} \). Single-pulse FWHM duration and repetition period are set as in Fig. 2. A, Spectra associated with different values of the driving peak intensity: \( 4.54 \times 10^{11} \text{ W cm}^{-2} \) (red), \( 9.14 \times 10^{11} \text{ W cm}^{-2} \) (blue), \( 1.38 \times 10^{12} \text{ W cm}^{-2} \) (black) and \( 1.85 \times 10^{12} \text{ W cm}^{-2} \) (green). The spectra are normalized to the peak of the Lorentzian spectrum in the absence of optical control. B, C. For the green curve, amplitude of the peaks in the comb (b) and shape of a single comb tooth (c).

In Fig. 2 we use an optical frequency comb tuned to the optical transition. The peak intensity of \( 2.57 \times 10^{10} \text{ W cm}^{-2} \), corresponding to 2\( \pi \) pulses, is several orders of magnitude lower than the intensity required for HHG-based schemes\(^8\)-\(^{10}\). As shown in Fig. 2a, the coherence, stored in \( \rho_{13}(t) \) in the long time interval between two pulses, is transferred to \( \rho_{12}(t) \) only in the presence of an optical pulse, with corresponding decay of the bright state. This results in the X-ray comb of Fig. 2b–d, centred and locked to the natural transition frequency at 123.7 eV, spanning as wide as the driving optical frequency comb and consisting of Lorentzian peaks separated by the optical-comb repetition frequency of 100 MHz (4.1 \( \times \) 10\(^{12}\) eV). The width of these peaks, given by the inverse of the long, effective decay time of the system, is orders of magnitude smaller than the natural width of the bright state. This leads to a correspondingly high spectral resolution and attainable precision of the comb.

In Fig. 3, we use the scheme to analyse off-resonant optical driving. We use pulses with 1.5 eV photon energy from mode-locked Ti:sapphire lasers. The chosen peak intensities, near \( 1 \times 10^{12} \text{ W cm}^{-2} \), are lower than those presently used for XUV-comb generation with HHG\(^8\)-\(^{10}\). The position and line shape of the peaks in the absorption spectrum depend controllably on the peak intensity (Supplementary Section III). Despite its asymmetric shape, the X-ray absorption spectrum is as wide as the optical frequency comb.

The scheme is, in principle, applicable to atomic systems at higher X-ray energies, provided an X-ray source\(^1\) is available at that photon energy and an optical transition can be identified between the bright state and a metastable level. Furthermore, the duration of the optical pulses and the time delay between X1 and L1 have to be smaller than the decay time of the excited state, to enable the preparation of the system and its further manipulation (Fig. 1; see Supplementary Sections II and III).

In conclusion, we have developed a scheme to imprint a frequency comb onto the absorption spectrum of an X-ray FEL pulse\(^1\). The spectrum consists of equally spaced lines whose exact positions, referenced to an atomic transition, may be used to precisely bridge a reference level and an unknown X-ray frequency.
Furthermore, by using transitions with a higher energy, for example, 1s² → 1s np in helium-like ions of higher nuclear charges, or nuclear transitions, the presented scheme may enable comb generation up to gamma-ray frequencies.

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Author contributions

S.M.C. developed the mathematical model, performed the analytical calculations and wrote the manuscript. All authors contributed to the development of ideas, discussion of the technical aspects and results, and preparation of the manuscript.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to S.M.C.

Competing financial interests

The authors declare no competing financial interests.