individually may choose between single bunch and multibunch SR at each beamline and switch between these modes readily. Since the vertical emittance of SR storage rings is significantly smaller than the horizontal, the spectral monochromatization requires utilizing the vertical as the spectral dispersion plane. In consequence the separation between multi and single bunches should ideally utilize the orthogonal horizontal plane although more challenging. In this work we show how pulse picking by resonant excitation (PPRE) allows for separating photons from a single bunch horizontally, fully orthogonal to the dispersion planes, from the multi-bunch fill pattern. We verify the performance by time-of-flight electron spectroscopy results.

Prior to our work, vertical separation of single- and multi-bunch patterns has been shown at the Advanced Light Source (ALS) by Sun *et al.*[1] through a vertical kick-and-cancel scheme that generates a "pseudo-single-bunch" at variable frequency by a vertical kicker in combination with a specific beamline that is capable of vertically separating photons emitted from a camshaft bunch that travels on a different closed orbit. Using the same method, first attempts at other facilities have been made[2]. However, for realizing the dream of using separated photon pulses at any beamline of an existing light source on demand, we need to preserve the vertical plane for spectral monochromatization. The larger angular/spatial horizontal separation required is prohibitive to the existing kick-and-cancel scheme. We thus devised the Pulse picking by resonant excitation (PPRE) scheme based on a combination of increased horizontal emittance of a single (camshaft or hybrid) bunch by a quasi-resonant incoherent excitation and a static local horizontal deflection of the electron beam that emits unwanted photons already upstream the beamline into a dump and only photons from the excited bunch into the beamline.

Since pulse picking by resonant excitation (PPRE) provides an x-ray source ideally suited for high-resolution time-resolved photoelectron spectroscopy (TRPES) and we show the potential of this approach with angle resolved time of flight (ARTOF) spectroscopy. Here, the Coulomb repulsion between all the emitted electrons on a sample's surface sets a limit to the number of photons per x-ray pulse that can be used in order to reach a certain energy resolution[3-5], and this number decreases with decreasing x-ray spot size and pulse duration. Peak brilliance is therefore not the main figure of merit for a high resolution photoemission set-up. The performance is instead improved by increasing the repetition rate of the source and by optimizing the transmission of the spectrometer in order to accept a fraction of the emitted photoelectrons as large as possible. The angular resolved TOF (ARTOF [6, 7]) provides this feature, combining outstanding transmission with high energy and angular resolution. The instrument also allows operation at a few MHz repetition rates, which is perfectly suited to the presented PPRE scheme. We show that the ARTOF instrument works very well with our new PPRE scheme, even when the storage ring is filled with ultrashort bunches in low-α mode. The ARTOF spectrometer and other time resolved techniques have many attractive characteristics. All of these will

benefit from the possibility to create single bunch radiation on demand at any undulator beamline during regular operation of the facility.

Quasi-Resonant excitation of a single bunch

The employed PPRE scheme is based on a quasi-resonant excitation of incoherent betatron oscillations of electrons in one particular bunch out of the entire temporal fill pattern. The light emitted from this bunch has a larger horizontal divergence than light emitted by bunches which are not excited. As shown in Figure 1, this particular beam property makes it possible to use a knife-edge aperture to pick just the pulse of undulator radiation from the excited bunch only.

Note that the separation is achieved by moving the radiation of the unexcited beam horizontally away from the beam line's axis with a closed orbit bump of the electron beam along the undulator. In this way, by using the on-axis radiation, we preserve the polarization properties of the picked photon beam pulse, even if emitted from a helical insertion device (like in the slicing case[8]). If only one bunch, e.g. a camshaft bunch in the clearing gap is selectively excited, the photon beam arriving at the experiment mimics a single electron bunch's temporal emission pattern with 1.25 MHz repetition rate corresponding to the electron's revolution time of 800 ns in the storage ring. The quasi-resonant excitation, however, has not to be done at 1.25 MHz but close to the horizontal betatron tune and corresponding sidebands of multiples of the revolution frequency. Therefore, and as explained below, a much lower frequency of ~190 kHz can be chosen which leads to stationary but incoherent elongations of electrons at the revolution frequency of 1.25 MHz.

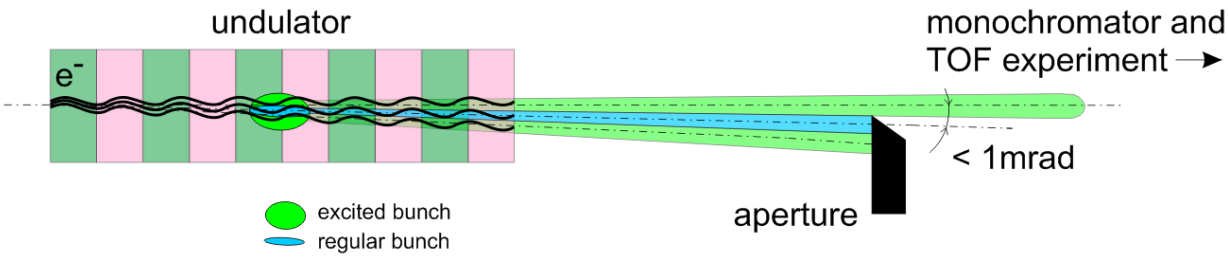

Figure 1 | Schematic illustration (view from above) of pulse picking by resonant excitation (PPRE). Undulator light (blue) emitted from the regular electrons in other buckets (2 ns distance) is dumped into an aperture. In regular operation the bump is turned off and the non-excited bunches (blue ellipse) emit undulator radiation on-axis. With the bump turned on, this radiation hits the knife-edge being dumped there and only radiation from a resonantly excited (green) bunch can enter the beamline.

The original pseudo-single-bunch approach of the ALS[1] requires kicking the bunch on a turn-by-turn basis with always a similar angle, which is technically very challenging, especially in the horizontal plane. So far, only vertical kicks of less than 0.09 mrad with the necessary steep rise and fall times and the required repetition rate (~1 MHz) have been achieved. This approach provides single bunch capabilities at a few beamlines, which furthermore have to be adjusted to utilise the kicked beam at the experiment.

The approach at BESSY is much different. In cases where the full intensity of the bunch is not required, the following approach can be used: A typical bunch consists of a few times 10^{10} electrons which all perform transverse oscillations with frequencies spread around the so-called betatron tune. These oscillations are incoherent because the emission of synchrotron radiation by an electron is random and independent of the other electrons. The resulting excitation and damping process leads to a random distribution of phases and amplitudes of all individual oscillations and finally to a transverse Gaussian particle distribution. Based on our experience with emittance manipulation[9] we use a quasi resonant excitation of the bunch to primarily blow up the beam horizontally. Only comparatively small kicks (< 1 μrad) are required and these small kicks add up more or less coherently and thus create a considerable increase of the horizontal beam size. The bunch is kicked with the help of a stripline kicker. We use a dedicated kicker similar to the one of the transverse bunch-by-bunch feedback system. The kicker is powered by a 100 W-amplifier with a bandwidth ranging from 0.1 to 250 MHz. With the help of a ~5 ns long gate synchronised to the revolution frequency of 1.25 MHz a signal is fed into the amplifier which thereby excites only the selected bunch in the ion-clearing gap. The frequency of the input signal is close to the lowest horizontal betatron resonance of 190 kHz and modulated in frequency over a few hundreds of Hz with a sweep frequency of 100 Hz. This, together with the intrinsic tune spread among the electrons (created by wake fields and the energy and amplitude dependence of the tune) and the tune modulation of the resonance due to small mains ripples of the quadrupole gradients helps to excite the electrons as incoherently as possible. As a result, the centre of gravity oscillation of the bunch is much smaller than the blow-up of the beam size.

Figure 2 shows calculated photon flux curves emitted by a 5 mA single bunch in an undulator at BESSY II for different electron beam emittances. The vertical dashed line marks the position of the knife-edge aperture. With increased horizontal beam emittance an increasing fraction of the light will pass through the aperture. A tenfold increase of the horizontal beam emittance (about a factor of 3 in divergence) is usually achieved with the excitation power levels used here (see supplemental material). The light flash, although at lower intensity, creates a pulse every time the blown-up bunch passes by. The emittance at BESSY II is further discussed in the supplementary material. As shown in Figure 2 it takes horizontal angles larger than of 0.5 mrad in order to effectively separate the light produced by the excited single bunch from light emitted by the rest of the electrons.

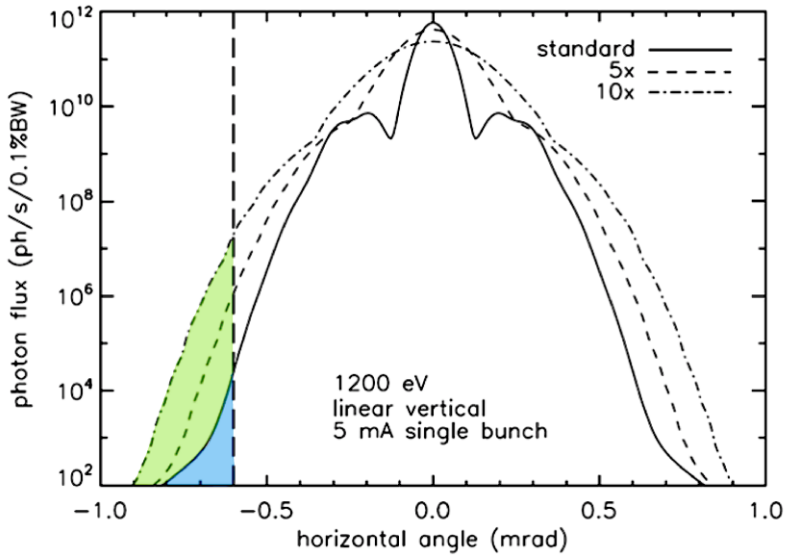

Figure 2 | Calculated angular distribution of x-rays emitted from a single bunch at 5 mA that passes the UE56 (Apple II) undulator at BESSY II. The solid line represents emission from standard operation. The dashed curves represent emission from resonantly excited bunches of 5 fold and 10 fold increased effective emittance, respectively. It can be seen that accepting photons beyond a cut at about -0.6 mrad gives a high purity of up to 10^4 for the PPRE radiation (green) that passes the aperture compared to the (blue) pedestal from the unexcited bunch.

Operating in the top-up-mode with such a blown-up bunch is possible if the excitation is turned off shortly before an injection takes place. Otherwise electrons of this bunch can get lost at the injection septum magnet. The blank-out pulse indicating an injection to the users roughly 20 ms in advance is used to suppress the excitation for 50 ms. This approach is compatible with normal user operation of BESSY II.

Experimental results

(i) Regular user mode

The PPRE method in the regular operation mode of the storage ring is demonstrated in Figure 3. Currently, the regular fill pattern of BESSY II in 2012/2013 contains 4 camshaft bunches in a 200 ns clearing gap as indicated by signals from an Avalanche Photodiode (APD, negative record) behind the monochromator under the condition of the scheme drawn in Figure 1, but yet at rather weak excitation of the camshaft bunch that was in the center of the gap. Here, the aperture was set such to still detect some background signal from the regular multi-bunch at an angular bump setting of -0.6 mrad. With PPRE switched on, under these conditions (Figure 3b), the signal from the excited bunch considerably grows indicating that more photons pass the closed aperture.

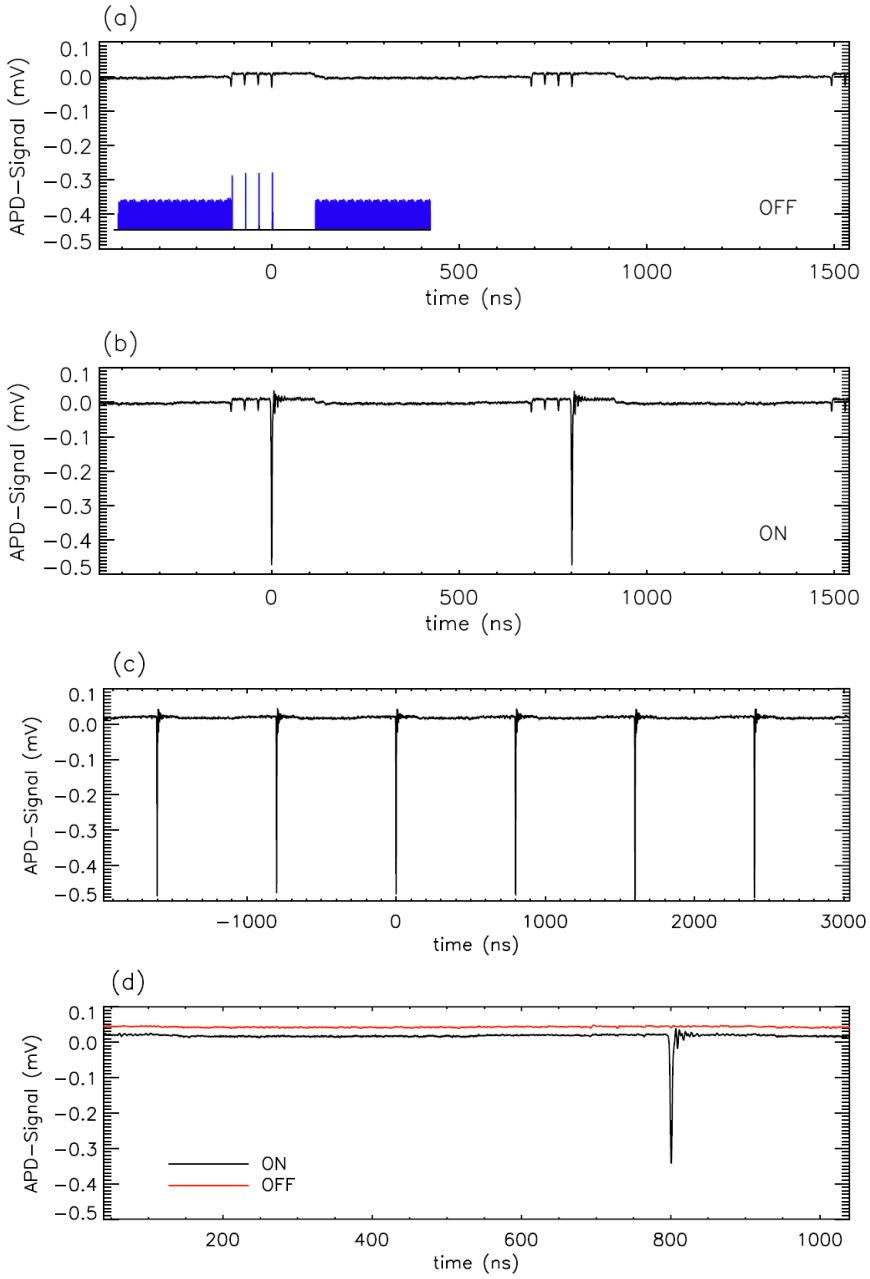

Figure 3 | Raw oscilloscope traces of the avalanche photodiode (APD) signal behind the exit slit at a photon energy of 1200 eV for three different situations: (a) Normal hybrid mode with four camshaft bunches of mod (36 ns) distance in an ion clearing gap of 200 ns. The inset shows the electron bunch structure (fill pattern). (b) Weak excitation of one camshaft bunch and in (c) and (d) a stronger excitation. The red curve (shifted for clarity) in (d) shows the result for the excitation switched OFF. The signal contribution from the multi-bunches is not detectable even with a signal-to-noise limit > 10^3.

Closing the aperture more (accepting less background signal) and increasing the amplitude of the quasi-resonant excitation, the core beam can be completely suppressed (Figure 3 c) and a single bunch signal of 10^7 -10^9 ph/s at a purity of up to 10^2-10^4 is detected. Such a situation is achieved in the results displayed in Figure 3d, where the red curve (slightly set off for clarity) indicates a complete disappearance of the radiation if the excitation is switched off. It can be noted that a signal of this magnitude is even

approaching the maximum acceptable level for very high resolution PES on a micron-sized sample[4]. The same method was also successfully tested with similar results at a different undulator beamline (UE52-beamline and SGM – monochromator[10]. Even at very low photon energies down to 50 eV and up to 1200 eV a sufficiently clear separation of the single bunch pulses was possible. Most promising was a first test at the L-absorption edge of Iron (at ~700 eV) in circular mode of the helical undulator indicating a feasibility of time-resolved magnetic experiments based on XMCD, also using photoelectron- or Auger electron detection.

(ii) Low-α mode

Bunch lengths of ~2-3 ps (rms) are available using BESSY II's low-α mode[11, 12] or even shorter at lowest bunch currents[13]. Therefore, a special fill pattern with a bunch current of < 100µA of the camshaft bunch was created depicted by the green curve in Figure 4. Albeit at somewhat less purity, the PPRE separation method applies here as well, as confirmed by the suppression of the multi-bunch APD-signal when blocking the regular beam more and more by the aperture (Figure 4a and b). As will be seen below, this provides a roadmap for an excellent source of ps pulses at MHz repetition rates for short pulse electron spectroscopy.

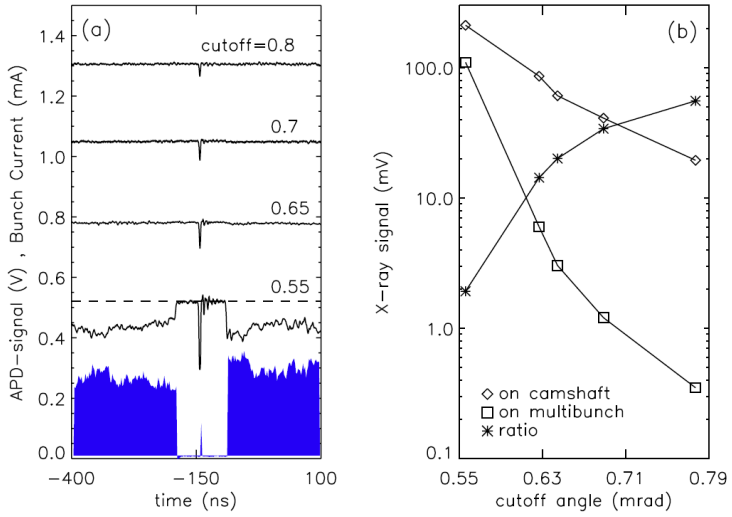

Figure 4 | X-ray signals in low-α mode measured at the UE56/1 with PPRE for different cut-OFF angles. The envelope of the temporal current distribution in the storage ring as measured in the accelerator (blue curve in (a)) indicates the single bunch at <100 µA bunch current in the center of a gap of 100 ns. The bunch is surrounded by a (non-uniform) pattern from the multi-bunch fill. The oscilloscope traces of the X-ray signal plotted in black in (a) reflect different aperture settings (cut-OFF angles). The analysis of the measurements in (b) shows the peak signals for different aperture settings: squares- background signal from multi-bunch; diamonds - peak signal from the single bunch; black dots - purity (signal-over-background).

(iii) Photoelectron spectroscopy results

We have successfully recorded photoelectron spectra using the PPRE scheme, using the newly developed ARTOF spectrometer. The ARTOF technique is based on the combination of an advanced electron lens with two-dimensional time sensitive detector. The angular resolving mode of such a lens is based on the fact that the kinetic energy of the photoelectron and its take-off angles lead to a unique position at the focal plane at the end of the lens. However, the angular resolved electrons are not projected on an entrance slit but hit the surface of a delay line detector. This detector measures the arrival time of the pulses and their x and y coordinates. Thus, we record in parallel the whole 2D angular pattern instead of only one angular coordinate. The kinetic energy of electrons can be recovered with a very high precision from the measured flight time in conjunction with the position on the detector[7]. A great advantage of the ARTOF type of electron analyzers is the much higher acceptance, the resulting increased information rate in comparison to classical hemispherical analyzers and the much better resolution compared to classical time-of-flight analyzers[6]. The increase in acceptance (about a factor of 10^3) in comparison to a hemispherical electron analyzer more than compensates for the intensity loss due to our PPRE scheme.

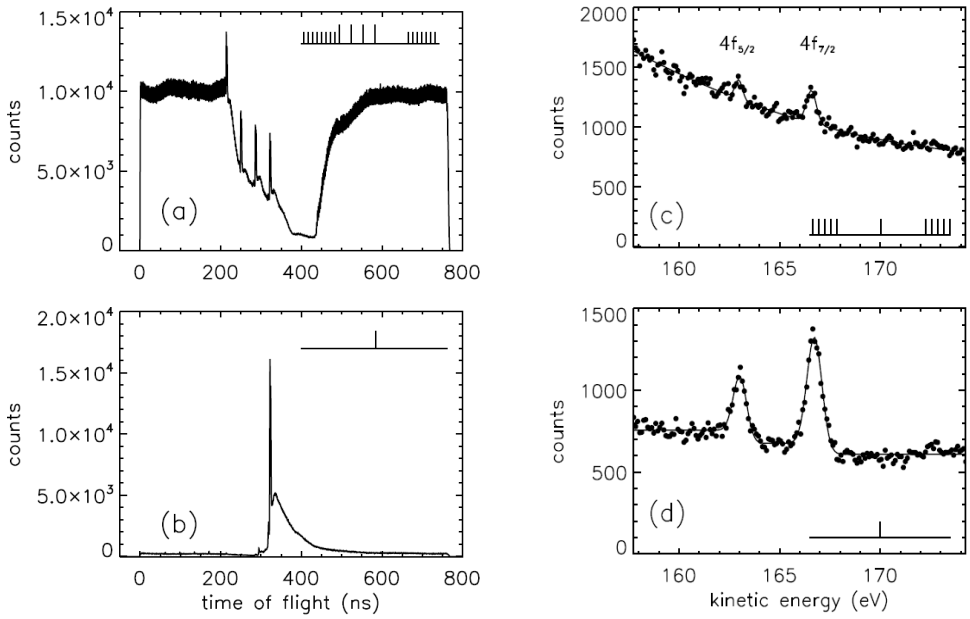

Figure 5 | Time-of-Flight spectra recorded with the ARTOF in regular user mode with PPRE switched OFF (a) and ON (b), respectively and kinetic energy electron spectra from a Gold target taken with the ARTOF in low-a mode with PPRE switched OFF (c) and ON (d). Time-of-Flight data recorded with the ARTOF clearly show a strong contamination from other bunches if pulse separation is switched OFF (a) and (c). Clean spectra (b) and (d) are obtained if only light from the exited bunch arrives at the sample. The bunch fill patterns are seen in the insets in (a) and (c). In (b) and (d) the PPRE single bunch structure is inserted, obtained from the fill patterns in (a) and (c), respectively. The Au 4f spectra at 160 meV analyzer resolution were taken with a photon energy of 250 eV using the 1200 l/mm grating and 100 µm slit (E/ΔE~5000) at the UE56/1-PGM beam line.

For the first proof of principle measurements with an ARTOF and our PPRE scheme we used a Gold sample, see Figure 5a. There we can clearly see the 4f peaks exited by light from each of the four camshaft bunches. In addition, the multibunch structure gives a high undesired background. When the pulse picking is turned on in Figure 5b a very clean Au 4f spectrum is seen. In Figure 5c the ring is operated in hybrid mode with only one camshaft pulse, but under low-α condition. Already in this case, the Au 4f peaks from the hybrid bunch are visible although on a very high background. The fact that they are detectable at all is explained by the clean clearing gap for the low-α mode (no 4 camshaft bunches). When PPRE is turned on, a useful electron kinetic energy spectrum is obtained in only a few minutes, see Figure 5d. The photon energy was 250 eV and the resolution of the spectrometer was set to 160 meV. In case of the low-α mode, the overall flux is very small making it unfeasible to perform any kind of photoelectron spectroscopy with hemispherical analyzers. Pulse picking further reduces the flux, but due to the large acceptance angle and the high transmission of the ARTOF, it is no problem at all to acquire useful spectra.

Conclusions

In summary, we have introduced a resonant pulse picking scheme which allows to separate x-ray pulses at undulator beamlines on demand from single bunches in a storage ring filled with a multi-bunch fill pattern. The method is based on the quasi-resonant excitation of the horizontal betatron oscillations of electrons in one particular bunch. The excitation leads to an increased emittance of this bunch and causes a corresponding horizontal angular blurring of its undulator radiation cone. By deflecting the regular, very narrow unexcited part of the undulator radiation away from the beamline's axis, only light from the excited bunch will reach the experiment with a rate given by the revolution frequency. The setting of the local orbit bump together with slit positions in the beamline's front end that separate the useful photons can be utilized to adapt different widths of the undulator radiation cone at different gap and shift settings[14]. Owing to this kind of separation, there is a loss of flux involved that, however, is acceptable and even desired for photoemission experiments using modern highly transmitting end stations like the ARTOF. We have confirmed this by proof-of-principle experiments with the ARTOF instrument in different modes of BESSY II, at different undulators (UE56, UE52, U125) and at different beamlines: plane grating monochromators (PGM), zone plate monochromators (ZPM) and spherical grating monochromators (SGM). In cases of elliptical insertion devoices, the separation was possible even in circular mode of the helical undulator. Selecting light form ultrashort bunches and applications have also been performed in the low-α mode of BESSY II, even at very low bunch currents of < 100 µA. At last, the user can select to use either the full static flux or alternatively to switch over within minutes to the "single bunch on demand" situation.

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Author contributions

K.H., R.O., P.K. conceived and carried out the experiments supported by R. M., A. S., M. S., M. G., T. L., S. S., N. M. and A.F. Results were analyzed mainly by K.H., R.O. and K. H. wrote 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 K.H.

Competing financial interest

The authors declare no competing financial interests.