Measurement of X-Ray Pulse Widths by Intensity Interferometry

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The pulse width of hard undulator radiation (32 ps width, energy 14 keV) was determined by intensity interferometry. The method, in combination with various x-ray monochromators, enables measurements to be taken over a wide range of time frames, from ns to fs. The applicable target includes measurements of ultrafast x-ray pulse widths from fourth generation synchrotron light sources.

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Ultrafast x-ray pulses [1–4] provide a powerful probe for investigating structural dynamics in biological and material sciences. The upcoming linac-based undulator sources, such as self-amplified spontaneous emission free-electron-laser (SASE FEL) [5,6] or energy recovery linac (ERL) [7], are capable of generating brilliant x-ray pulses of ~100 fs. Determination of such ultrafast profiles is crucial for developing and utilizing these advanced x-ray sources. However, it was quite difficult, or impossible, to measure x-ray pulse widths from ps to fs region. This is because the autocorrelation technique with second harmonic generation (SHG), which is widely used for measuring ultrafast pulses of visible light, cannot be applied to the x-ray region due to negligibly small nonlinear interactions. As an alternative method, the state-of-the-art x-ray streak camera has shown time resolutions of ~1 ps [8]. However, drastic improvement of resolution for the camera appears to be difficult.

In this Letter, we report intensity interferometry, which is a technique initially developed by Hanbury-Brown and Twiss [9] and recently extended to the x-ray region [10–13], is capable of measuring x-ray pulse widths. We demonstrate the validity of the method using SPring-8, a modern synchrotron providing one of the fastest hard undulator radiations (32 ps width, energy 14 keV) currently available. This method can be easily extended to even fs region by using simpler monochromators.

Most hard x-ray sources, which are both currently available and under development including SASE FEL in the linear regime [14,15], are considered to generate chaotic light. In this case, intensity interference is observed as an enhancement of the coincidence rate between the two detectors that receive the spatially and temporally coherent portions of the beam; this enhancement results from larger fluctuations of the mean intensity. In particular, when the method is applied to pulsed beams, the enhanced ratio includes information on the temporal pulse width s_t with respect to the longitudinal coherence time σ_t [10]. Because the coherence time σ_t is directly given by the energy bandwidth ΔE of light, evaluation of the enhanced ratio with a knowledge of the bandwidth enables one to determine the pulse width s_t. It is important for correct measurement of the pulse width to evaluate influence of transverse coherence, because the enhanced ratio also depends on the coherent mode numbers in the transverse directions.

Experiments were performed using beam line 19LXU of SPring-8, which is equipped with a 27-m undulator, the most brilliant x-ray source currently available [16]. High peak brilliance is desirable for reducing the time required to measure the coincidence enhancement with a good signal-to-noise ratio (S/N). The experimental setup is shown in Fig. 1. A monochromator consisting of four-bounced asymmetric reflections [17] (horizontal diffractions of Si 11 5 3, asymmetric angles $\alpha = 78.4°$) was used as an energy filter. A technique was employed to control the bandwidth ΔE (i.e., the coherence time σ_t) of the monochromatic beam. If we choose the incident energy E so as to make the angles $\theta_B - \alpha$ or $\theta_B + \alpha$ small (θ_B is the Bragg angle), a slight shift of E produces a large change in the asymmetric factor $b = \sin(\theta_B - \alpha)/\sin(\theta_B + \alpha)$. This leads to control of the

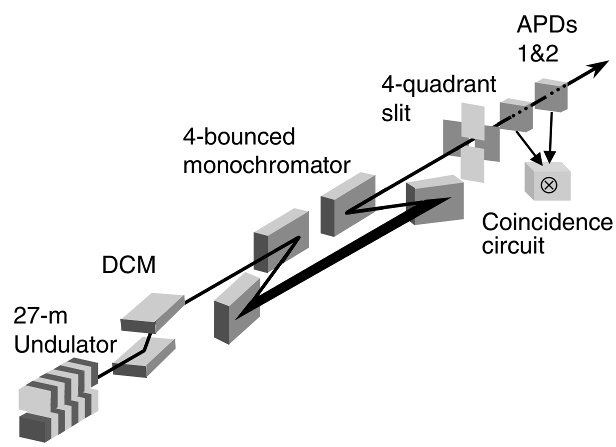

FIG. 1. Schematic view of the experimental setup. Undulator radiation is premonochromatized with a Si 111 double crystal monochromator (DCM). The four-bounced monochromator and the four-quadrant slit were employed to extract the longitudinally and transversally coherent portion of the beam, respectively. Two semitransparent avalanche photodiodes (APDs) were aligned in tandem on the light axis. Outputs of the detectors were connected to the coincidence circuit.

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TABLE I. Experimental conditions for coincidence measurements.

E (keV)	θ_B (deg)	b	ΔE_T (meV)[a]	ΔE (meV)[a]
14.267	84.92	2.53	0.755	0.763 ± 0.002
14.299	83.64	3.38	0.495	0.508 ± 0.004
14.333	82.52	4.56	0.315	0.334 ± 0.005
14.365	81.60	6.13	0.203	0.231 ± 0.008
14.412	80.41	10.4	0.097	0.145 ± 0.012

[a]These are values in FWHM.

bandwidth ΔE, as it is proportional to $b^{1.5}$ [17]. Numerical calculation shows that changing energy from 14.267 to 14.412 keV varies the bandwidth over a factor of seven. The energy shift is so small that other experimental conditions (transverse coherence lengths, for example) were only negligibly changed. Five energies were chosen from the above range for coincidence measurements, results of which are summarized in Table I. We note that the energy bandwidths were calibrated using the nuclear forward scattering (NFS) of ^{57}Fe, because minor imperfections in crystals possibly broaden the actual bandwidths ΔE from the theoretical values ΔE_T [18].

The true coincidence rate C_S and the accidental one C_N were measured with coincidence circuits and an electric delay of 4.79 μs that corresponds to the revolution frequency of the storage ring [11]. Here, the enhancement $R = C_S/C_N - 1$ of the coincidence rate is given by the inverse of the mode number $M = M_x M_y M_t$, where M_x and M_y are the transverse mode numbers on the detectors and M_t is the longitudinal mode number included in the single pulse [10,13,19]. Because the incident light is partially coherent in the transverse directions, we used a four-quadrant slit in order to extract the coherent portion of the beam (Fig. 1). Figure 2 shows values of R on varying horizontal slit widths w_x, at a constant vertical width of $w_y = 30$ μm. For each energy, R increases as w_x decreases. The extrapolated value of R for $w_x \to 0$ ($M_x \to 1$) is regarded as $M_y^{-1} M_t^{-1}$. Assuming that the horizontal coherence profiles are Gaussians, they were well fitted using only two parameters: the coherence length σ_x, and the scale factor $M_y^{-1} M_t^{-1}$. The lengths σ_x ($\sim$8 μm) were close to those calculated from the angular acceptances of the monochromator, i.e., the angular size of the virtual source, 6 μm. Similar measurements taken while scanning the vertical widths w_y assured that values of M_y at $w_y = 30$ μm were almost unity [13]. Thus the longitudinal mode number M_t was determined at each condition.

Figure 3 shows the mode number M_t plotted as a function of the bandwidth ΔE. Assuming that the pulse envelope and the longitudinal coherence profiles are both Gaussian distributions, M_t is given from theory by

$$M_t = \sqrt{1 + \frac{s_t^2}{\sigma_t^2}}, \tag{1}$$

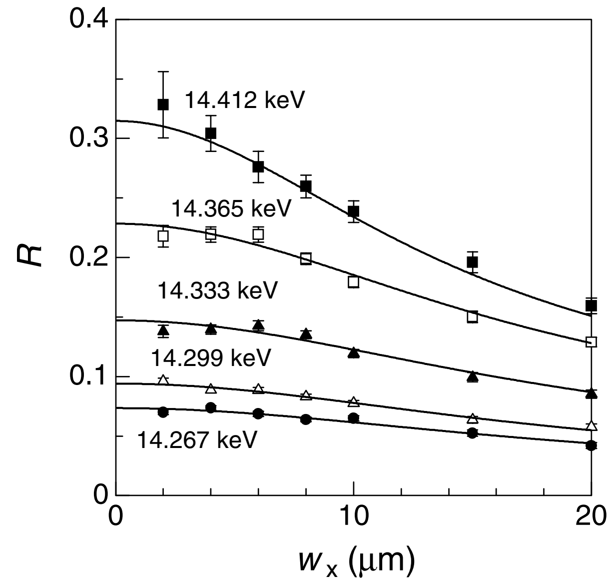

FIG. 2. The enhancement ratio of the coincidence rate R as a function of the horizontal slit width w_x, measured at different energies. The lines represent the fit results.

where s_t is the pulse width in full width at half maximum (FWHM). σ_t is the longitudinal coherence time given by

$$\sigma_t = \frac{4\hbar \ln 2}{\Delta E}, \tag{2}$$

where ΔE is the bandwidth in FWHM. Using these equations, the data were fitted with one fitting parameter, the pulse width s_t. The width s_t was determined to be 32.7 ± 1.6 ps in FWHM. This value was compared to that measured with a streak camera, 32 ps [20,21]. This level of agreement was excellent. We confirmed that intensity interferometry is applicable to measuring x-ray fast pulse widths with high accuracy. However, if the coherence time

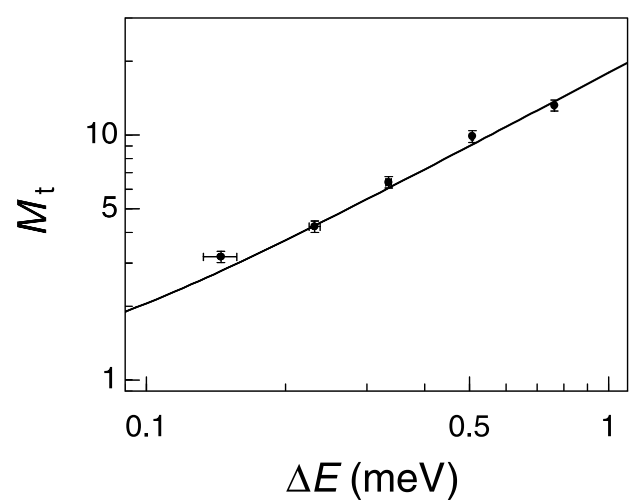

FIG. 3. The longitudinal mode number M_t vs the energy bandwidth ΔE. The line shows the fit result with a pulse width s_t of 32.7 ps.

σ_t were to exceed the original pulse width, the monochromatic pulse would be a Fourier-limited pulse and its width s_t would no longer be equal to the input pulse width. This is actually observed in nuclear resonant decay with synchrotron radiation [22]. Since the coherence times in our measurement were sufficiently smaller than the input pulse width for every condition, no significant differences between the incident and the monochromatic pulse widths (for infinity small w_x) are expected. This assumption was consistent with experimental results. More rigorous treatment of the output pulse response can be calculated using the time-space-dependent dynamical diffraction theory [23].

Next, we discuss the applicable range of the method. In order to observe the considerable enhancement of C_S, it is preferable that σ_t be larger than $\sim 10^{-2} s_t$. However, when the incident beam is too monochromatized (i.e., $\sigma_t \sim s_t$), the output pulse is forced to have a delayed response, as previously discussed. One must select a proper bandwidth corresponding to the pulse width to be measured. Today, various x-ray monochromators having a well-defined bandwidth ΔE can be constructed using perfect crystals. A wide range of ΔE from 10^{-4} to 10 eV can be covered at an energy of $\sim$10 keV. These bandwidths correspond to values of σ_t between 10 ps and 0.1 fs. Therefore, intensity interferometry combined with such monochromators is capable of determining x-ray chaotic pulse widths in the time scale from ns down to fs. Another essential requirement is the brilliance of the light source. The signal-to-noise ratio (S/N) of the coincidence enhancement is given by [10]

$$S/N \propto \lambda^2 \xi B_p \sqrt{f_B \Delta T}\,, \tag{3}$$

where λ is the wavelength, ξ is a product of the detector efficiency and the monochromator throughput, B_p is the peak brilliance, f_B is the pulse repetition rate, and ΔT is the measurement time. In our measurement where 175 over 2436 rf-buckets in the storage ring were occupied with electron bunches (i.e., $f_B = 37$ MHz), B_p was calculated to be $\sim 3 \times 10^{23}$ photons/s/mm^2/mrad^2 in 0.1% bandwidth at 100 mA [24], and the efficiencies ξ to be a few percent. Under these conditions, a measurement time ΔT of $\sim$1 h was necessary with a small slit size in order to obtain a S/N greater than 10. Let us consider to measure ultrafast pulses from the upcoming linac-based, coherent x-ray sources. With the design parameters [5,6], one finds that even spontaneous emission [$B_p \sim 10^{28}(10^{29})$ photons/s/mm^2/mrad^2 in 0.1% bandwidth and $f_B \sim 120(6 \times 10^4)$ Hz for LCLS (TESLA)] is sufficient for our method. The method can be further applied to SASE FEL operation with proper attenuation of intensity or by replacing the coincidence technique with the photon-counting technique [25].

Our method, which is based on the fundamental property of chaotic light, can be easily extended to faster pulse regions, because the optics required are much simpler than those used in the present work. Fast time resolution is unaffected by the timing jitter of the incident pulses and of the trigger signal. At present, the method provides a unique technique for characterizing $\sim$100 fs pulse profiles generated with the forthcoming linac-based, coherent x-ray sources. The method can be applied to characterize much faster x-ray pulses produced by proposed slicing technique of chirped pulses [26] and improved fast Bragg switches [27–30], as well as the secondary pulses from materials.

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