addenda and errata

Determination of X-ray pulse duration via intensity correlation measurement of X-ray fluorescence. Erratum

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Corrections to equations and experimental results in the paper by Inoue *et al.* [(2019). *J. Synchrotron Rad.* **26**, 2050–2054] are made.

The correct versions of equations (2) and (3) in the paper by Inoue *et al.* (2019) are as follows:

$$g_f^{(2)}(r_1, r_2) = 1 + \frac{1}{2}\left|j(r_1, r_2)\right|^2 \int \Pi(\tau)\left|\gamma(\tau)\right|^2 d\tau, \quad (2)$$

$$g_f^{(2)}(r_1, r_2) = 1 + \frac{1}{2}g_0^{(2)}\exp\left(-\frac{\Delta x^2}{2l_x^2}\right)\exp\left(-\frac{\Delta y^2}{2l_y^2}\right). \quad (3)$$

The additional factors of 1/2 on the right-hand sides of these equations represent a decrease in intensity correlation of X-ray fluorescence due to the unpolarized nature of the fluorescence (Trost *et al.*, 2020; Goodman, 2007).

Accordingly, the degree of intensity correlation $[g_0^{(2)}]$ and the XFEL duration $[2\sqrt{2\ln 2}\,\sigma_t]$ evaluated by the experiment also need to be corrected; the values of $g_0^{(2)}$ and $2\sqrt{2\ln 2}\,\sigma_t$ shown in Section 4 should be 0.0262 ± 0.008 and 5.1 ± 0.2 fs, respectively. The determined XFEL duration is consistent with previous estimations by other methods (Inubushi *et al.*, 2017; Inoue *et al.*, 2018), in which the XFEL duration was evaluated to be less than 10 fs. Although the determined XFEL duration is shorter than the electron bunch duration measured by a radiofrequency deflector ($\sim$10 fs in FWHM), such discrepancy could be explained by insufficient time resolution of the deflector ($\sim$10 fs) (Ego *et al.*, 2015).

Acknowledgements

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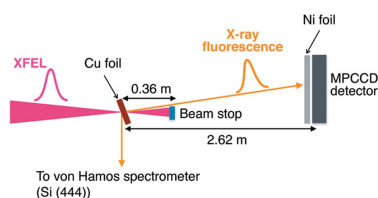
Determination of X-ray pulse duration via intensity correlation measurements of X-ray fluorescence

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A simple method using X-ray fluorescence is proposed to diagnose the duration of an X-ray free-electron laser (XFEL) pulse. This work shows that the degree of intensity correlation of the X-ray fluorescence generated by irradiating an XFEL pulse on metal foil reflects the magnitude relation between the XFEL duration and the coherence time of the fluorescence. Through intensity correlation measurements of copper $K\alpha$ fluorescence, the duration of 12 keV XFEL pulses from SACLA was evaluated to be $\sim$10 fs.

1. Introduction

X-ray free-electron lasers (XFELs) based on the self-amplified spontaneous emission scheme (McNeil & Thompson, 2010; Kondratenko & Saldin, 1980; Bonifacio et al., 1984) are newly developed light sources with femtosecond duration. In combination with their excellent brilliance and high degree of transverse coherence (Gutt et al., 2012; Inoue et al., 2015), the ultrafast XFEL pulses, which are shorter than pulses from the third-generation synchrotron-based X-ray sources by a few orders of magnitude, have opened up novel scientific opportunities, such as capturing transient states of matters (Clark et al., 2013; Kim et al., 2015), damage-free structural analysis (Neutze et al., 2000; Chapman et al., 2011), and measurements and applications of femtosecond X-ray interactions with materials (Yoneda et al., 2015; Inoue et al., 2016). Furthermore, the recent advent of attosecond XFELs (Huang et al., 2017; Marinelli et al., 2017) would further expand the fields of ultrafast sciences that the XFELs can explore.

Evaluation of the pulse duration of the XFELs is essential for designing and analyzing XFEL experiments, as well as for controlling the temporal profiles of XFEL pulses by giving effective feedback to machine operation. Several approaches for the temporal diagnostics of XFELs have already been demonstrated. One such approach is single-shot spectroscopy of XFEL pulses (Inubushi et al., 2012, 2017; Zhu et al., 2012; Makita et al., 2015); the XFEL duration can be inferred from the spike width of the spectrum by assuming quantities of electron bunch that generate the XFEL pulse, such as emittance, temporal shape and energy chirp. Temporal diagnostics of electron bunches generating XFEL pulses by radio-frequency deflectors are also explored for estimating XFEL duration (Ding et al., 2011; Behrens et al., 2015). The best temporal resolution of this diagnostic method reported to date is 4 fs root mean square (Behrens et al., 2015), which is not sufficient for evaluating XFEL pulses with durations of less than 10 fs.
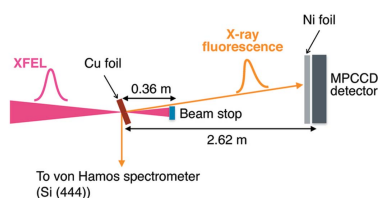
Here we propose a simple and direct method for evaluating the duration of femtosecond and attosecond XFEL pulses using X-ray fluorescence generated by irradiating XFEL pulses on metal foil. We also apply this method to 12 keV XFEL pulses generated from SACLA (Ishikawa *et al.*, 2012; Yabashi *et al.*, 2015).

2. Principles

In the proposed method, the spatial distributions of the X-ray fluorescence are measured shot-by-shot with a two-dimensional detector. Then, two-point intensity correlation of the X-ray fluorescence is evaluated for estimating the duration of fluorescence. For a pulsed chaotic light, it is known that the degree of intensity correlation of light is determined by the magnitude relation between the coherence time of light and the pulse duration (Miyamoto *et al.*, 1993; Yabashi *et al.*, 2002). As the X-ray fluorescence is chaotic light, the pulse duration of the fluorescence could be determined from the measured intensity correlation of the fluorescence. From the pulse duration of the fluorescence, the XFEL duration could be directly determined because the fluorescence lifetime is well known.

We consider the correlation function of X-ray fluorescence intensity at two positions (r_1, r_2) that are located beneath the path of the transmitted XFEL beam given by

$$g_f^{(2)}(r_1, r_2) = \frac{\langle I(r_1) I(r_2) \rangle}{\langle I(r_1) \rangle \langle I(r_2) \rangle}, \quad (1)$$

where $I(r)$ is the intensity of fluorescence at r and the brackets represent the average over different pulses. Owing to the chaotic nature of the fluorescence, the intensity correlation function is related to the intensity envelope function $P(t)$ and the power spectral density $S(\omega)$ of the fluorescence as (Inoue *et al.*, 2018)

$$g_f^{(2)}(r_1, r_2) = 1 + |j(r_1, r_2)|^2 \int \Pi(\tau) |\gamma(\tau)|^2 \, d\tau, \quad (2)$$

where $j(r_1, r_2)$ is the equal-time complex degree of coherence (Wolf, 2007), $\Pi(\tau) = \int P(t) P(t + \tau) \, dt / [\int P(t) \, dt]^2$ is the normalized autocorrelation function of $P(t)$, and $\gamma(\tau) = \int S(\omega) \exp(-i\omega\tau) \, d\omega / \int S(\omega) \, d\omega$ is the normalized Fourier transformation of $S(\omega)$. In the case when $r_1 = r_2 \equiv r$, the right side of equation (2) contains an additional term, $1/\langle I_{ph}(r) \rangle$, where $\langle I_{ph}(r) \rangle$ is the average number of detected photons per pulse at r due to the photon statistics (Singer *et al.*, 2014; Inoue *et al.*, 2018). From the definition, $\Pi(\tau)$ $(|\gamma(\tau)|^2)$ is non-zero only when τ is comparable with or shorter than the pulse duration (the coherence time) of the fluorescence. Therefore, the degree of intensity correlation of the fluorescence reflects the magnitude relation between the coherence time and the pulse duration. Since the coherence time of X-ray fluorescence is of the order of sub-femtosecond to femtosecond, the durations of attosecond and femtosecond X-ray pulses could be readily evaluated using this method.

In the case when the profile of the XFEL beam is described by a two-dimensional Gaussian function, $g_f^{(2)}(r_1, r_2)$

is related to the XFEL beam size in a simple form as shown below. According to the van Cittert–Zernike theorem, $|j(r_1, r_2)|$ is given by $|j(r_1, r_2)| = \exp[-(k^2 \sigma_x^2 / 2R^2) \Delta x^2] \exp[-(k^2 \sigma_y^2 / 2R^2) \Delta y^2]$, where k is the wavenumber of the fluorescence, σ_x (σ_y) is the horizontal (vertical) XFEL beam size in standard deviation, Δx (Δy) is the horizontal (vertical) distance between r_1 and r_2, and R is the distance between the foil and the detector. By substituting this equation into equation (2), we obtain

$$g_f^{(2)}(r_1, r_2) = 1 + g_0^{(2)} \exp\left(-\frac{\Delta x^2}{2l_x^2}\right) \exp\left(-\frac{\Delta y^2}{2l_y^2}\right), \quad (3)$$

where $l_x = R/(\sqrt{2} k \sigma_x)$, $l_y = R/(\sqrt{2} k \sigma_y)$ and $g_0^{(2)} = \int \Pi(\tau) |\gamma(\tau)|^2 \, d\tau$.

3. Experimental

Based on the above principle, we evaluated the duration of XFEL pulses with a photon energy of 12 keV at SACLA BL3 (Tono *et al.*, 2013). A total of 1047 pulses with pulse energies of 460 μJ ± 20 μJ were used in the following analysis. Fig. 1(*a*) represents a schematic of the experimental setup. The XFEL beam was focused by using a Kirkpatrick–Baez mirror (Yumoto *et al.*, 2013). The focused beam profile measured by a knife-edge scan method was well described by a two-dimensional Gaussian function with $\sigma_x = 0.4$ μm and $\sigma_y = 0.5$ μm. A copper foil with a thickness of 20 μm was tilted by 30° from the normal incidence and placed on the focus as a target. The $K\alpha$ fluorescence was measured shot-by-shot with a multi-port charge-coupled device (MPCCD) detector (Kameshima *et al.*, 2014) located 2.62 m downstream of the foil. Here, a 20 μm-thick nickel foil was placed in front of the detector to reduce the number of $K\beta$ fluorescence photon impinges on the detector to a negligibly small level compared with that of $K\alpha$. Furthermore, a beam stop made of carbon with a diameter of 3 mm was placed to avoid irradiation of the transmitted 12 keV X-ray beam on the detector. We distinguished $K\alpha$ fluorescence photons and 12 keV photons originating from the incident XFEL beam through the photon-energy sensitivity of the MPCCD detector, and only $K\alpha$ fluorescence photons were used for the following analysis. Simultaneously, the spectrum of the $K\alpha$ fluorescence emitted from the Cu foil was measured using a spectrometer in the von Hamos geometry consisting of a silicon (444) curved crystal and a MPCCD detector [Fig. 1(*b*)], and thereby $|\gamma(\tau)|$ was experimentally determined [Fig. 1(*c*)].

4. Results and discussion

Fig. 1(*d*) shows the average number of detected $K\alpha$ fluorescent photons by the downstream MPCCD detector. Although the number of detected photons at the bottom of the detector is much smaller than that at other detector regions due to the presence of the beam stop, the number of detected photons in the remaining area is almost the same [0.08 photons/(pulse × pixel)], reflecting isotropic emission of
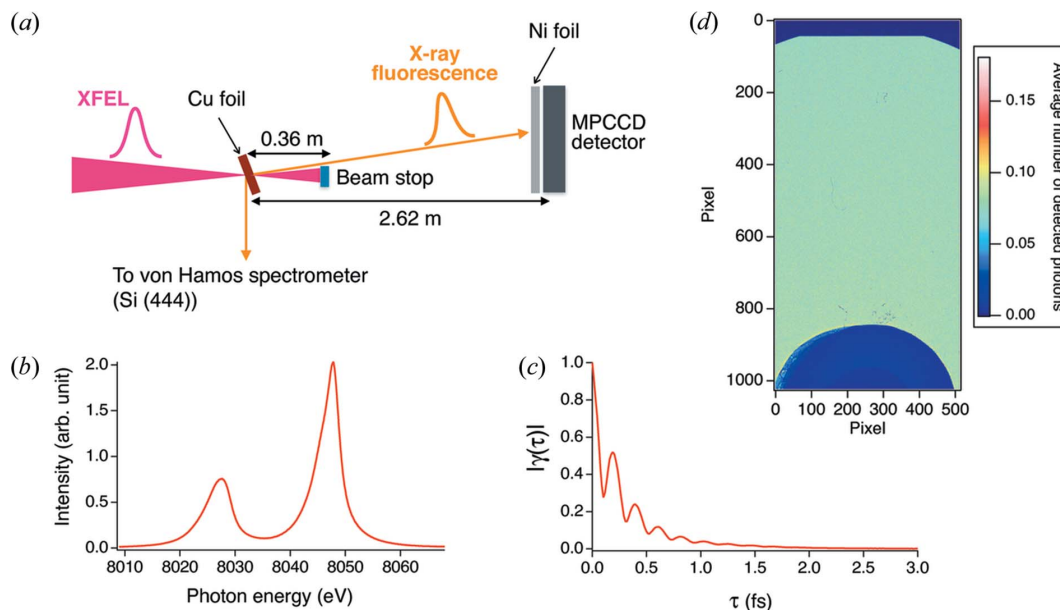
Figure 1
(*a*) Schematic of the experimental setup. (*b*) Spectrum of Cu *Kα* fluorescence measured with a von Hamos spectrometer. (*c*) $|\gamma(\tau)|$ calculated from the spectrum of Cu *Kα* fluorescence. (*d*) Average number of detected Cu *Kα* fluorescent photons per pulse at each detector pixel.

the *Kα* fluorescence. The flat intensity region is used for evaluating the intensity correlation of the fluorescence. Fig. 2 shows the intensity correlation function of the *Kα* fluorescence evaluated by

$$g_f^{(2)}(\Delta x, \Delta y) = \langle n_0(x + \Delta x, y + \Delta y)\, n_0(x, y) \rangle, \qquad (4)$$

where the brackets represent the average over pulses and pixels, and $n_0(x, y)$ is the number of the detected *Kα* fluorescent photons at the pixel (x, y) normalized to the average value over all pulses. $g_f^{(2)}(\Delta x, \Delta y)$ for small Δx and Δy exceeds unity, indicating that intensities of the fluorescence at different positions are significantly correlated.

The measured $g_f^{(2)}(\Delta x, \Delta y)$ are well fitted by equation (3), and the degree of intensity correlation $g_0^{(2)}$, the transverse
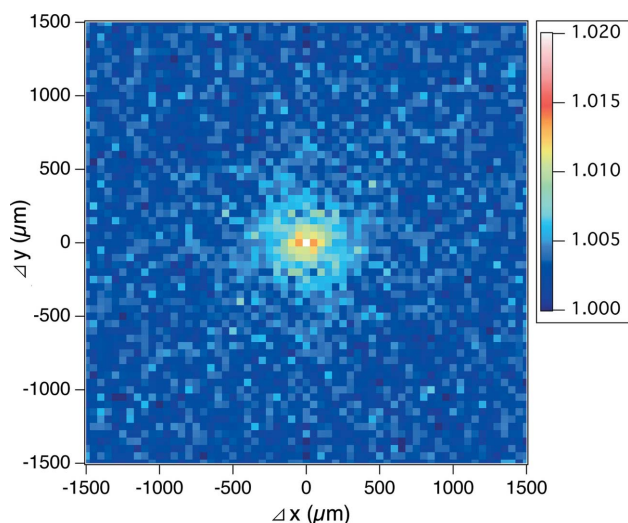
Figure 2
Intensity correlation function of Cu *Kα* fluorescence.

correlation lengths of the fluorescence in the horizontal and vertical directions l_x and l_y are evaluated to be 0.0131 ± 0.004, 161 µm ± 14 µm and 133 µm ± 9 µm, respectively. Here, the data point at $\Delta x = \Delta y = 0$ µm, where $g_f^{(2)}$ is dominated by the photon statistics, was omitted from the fitting to avoid the effect of photon statistics on $g_f^{(2)}$. These measured correlation lengths roughly agree with the theoretical predictions [$l_x = R/(\sqrt{2}k\sigma_x) = 120$ µm and $l_y = R/(\sqrt{2}k\sigma_y) = 90$ µm]. We would like to emphasize that the observed spatial correlation of the fluorescent intensity purely originates from the statistical properties of the fluorescence and is not due to artefacts of the MPCCD detector, because the point-spread function of the detector (less than 10 µm) (Kameshima *et al.*, 2014) is much smaller than the transverse correlation length of the fluorescence (l_x and l_y).

To evaluate the XFEL duration from $g_0^{(2)}$, we assumed that the intensity envelope function of the XFEL pulse $P_0(t)$ is described by the Gaussian function $P_0(t) = C_0 \exp[-t^2/(2\sigma_t^2)]$ with a proportional constant C_0. Then, the intensity envelope function of the fluorescence $P(t)$ may be expressed as

$$P(t) = C \exp\left(-\frac{t}{\tau} + \frac{\sigma_t^2}{2\tau^2}\right) \mathrm{erfc}\left(\frac{\sigma_t}{\sqrt{2}\tau} - \frac{t}{\sqrt{2}\sigma_t}\right), \qquad (5)$$

where C is a proportional constant, the lifetime of the *Kα* fluorescence $\tau = 0.3$ fs (Krause & Oliver, 1979) and complementary error function $\mathrm{erfc}(t) = (2/\sqrt{\pi}) \int_t^\infty \exp(-x^2)\, dx$. By substituting equation (5) and experimentally determined parameters [$g_0^{(2)}$ and $\gamma(\tau)$] into $g_0^{(2)} = \int \Pi(\tau)|\gamma(\tau)|^2\, d\tau$, we obtained $2\sqrt{2\ln 2}\, \sigma_t = 10.3 \pm 0.5$ fs, which corresponds to the XFEL duration in full width at half-maximum. The error in the determined XFEL duration originates from the uncertainty of $g_f^{(2)}$ and can be reduced by increasing the number of XFEL pulses used for the analysis. We note that the duration

of the electron bunch used for generating 12 keV XFEL pulses was estimated to be ~10 fs by a radiofrequency deflector (Otake *et al.*, 2013) which is consistent with the current evaluation.

5. Summary

In this paper, a simple and direct method using X-ray fluorescence is proposed to diagnose the duration of XFEL pulses. Clear intensity correlation of the fluorescence generated by irradiating XFEL pulses on a copper foil was observed, and the duration of 12 keV XFEL pulses at SACLA BL3 was evaluated to be ~10 fs.

This method is applicable to much shorter XFEL pulses and would provide a pathway to temporal diagnostics of attosecond XFELs. Also, our finding that the intensity correlation of the X-ray fluorescence is readily measurable with current XFEL sources would stimulate advanced methodologies using higher-order coherence properties of X-ray fluorescence, such as structure determination based on intensity correlation of the fluorescence (Classen *et al.*, 2017; Schneider *et al.*, 2018).

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