

Hanbury Brown–Twiss Interferometry at a Free-Electron Laser

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We present measurements of second- and higher-order intensity correlation functions (so-called Hanbury Brown–Twiss experiment) performed at the free-electron laser (FEL) FLASH in the non-linear regime of its operation. We demonstrate the high transverse coherence properties of the FEL beam with a degree of transverse coherence of about 80% and degeneracy parameter of the order 10^9 that makes it similar to laser sources. Intensity correlation measurements in spatial and frequency domain gave an estimate of the FEL average pulse duration of 50 fs. Our measurements of the higher-order correlation functions indicate that FEL radiation obeys Gaussian statistics, which is characteristic to chaotic sources.

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Hanbury Brown and Twiss, in their pioneering experiments [1,2], demonstrated that one can get fundamental information on the statistics of light sources by measuring intensity correlations at two separated spatial positions. Originally designed as a robust method to determine the size of stars, these experiments initiated developments in the field of quantum optics [3]. Statistical properties of thermal sources [1,2], lasers [4], semiconductor microcavities [5], and, recently, Bose-Einstein condensates [6,7] have been studied using this technique. The recent advent of x-ray free-electron lasers (FELs) [8–11] with their unprecedented peak brilliance and ultrashort pulse duration has opened the route to a number of spectacular ground-breaking experiments including femtosecond nanocrystallography [12] and single particle coherent imaging [13,14]. Many of these experiments exploit the high degree of coherence of the FELs.

Coherence is the defining feature of a laser source and is described by correlation functions within statistical optics [15,16]. The first-order correlation properties of FEL sources have been extensively investigated recently [17–22]. It was experimentally demonstrated that FELs based on the self-amplified spontaneous emission (SASE) process have a high degree of transverse coherence but poor temporal coherence. To get a more detailed picture of the statistical properties of these sources, higher-order field correlations must be studied. These can be explored, for example, by utilizing intensity correlation measurements in a Hanbury Brown–Twiss (HBT) experiment. In this Letter we present measurements of second- and higher-order intensity correlation functions at the free-electron laser FLASH [8].

The core idea of the HBT experiment [1,2] is to determine the normalized second-order intensity correlation function

$$g^{(2)}(\mathbf{r}_1, \mathbf{r}_2) = \frac{\langle I(\mathbf{r}_1) \cdot I(\mathbf{r}_2) \rangle}{\langle I(\mathbf{r}_1) \rangle \langle I(\mathbf{r}_2) \rangle} \quad (1)$$

by measuring the coincident response of two detectors at separated positions \mathbf{r}_1 and \mathbf{r}_2 (see for review [23]). In Eq. (1), $I(\mathbf{r}_1)$, $I(\mathbf{r}_2)$ are the intensities of the wave field, and the averaging is done over a large ensemble of different realizations of the wave field. It is well established that chaotic light can be described in the frame of Gaussian statistics [15] and is completely determined in spatial domain by the first-order correlation function known as the normalized spectral degree of coherence (SDC) $\mu(\mathbf{r}_1, \mathbf{r}_2)$. It is defined as [15] $\mu(\mathbf{r}_1, \mathbf{r}_2) = W(\mathbf{r}_1, \mathbf{r}_2) / \sqrt{S(\mathbf{r}_1)S(\mathbf{r}_2)}$, where $W(\mathbf{r}_1, \mathbf{r}_2)$ is the cross spectral density function and $S(\mathbf{r})$ is the spectral density.

The intensity correlation function then reduces to [24]

$$g^{(2)}(\mathbf{r}_1, \mathbf{r}_2) = 1 + \zeta_2(D_\omega) |\mu(\mathbf{r}_1, \mathbf{r}_2)|^2, \quad (2)$$

where the contrast function $\zeta_2(D_\omega)$ strongly depends on the bandwidth D_ω of the radiation. It was earlier demonstrated [25] that for stationary chaotic sources the contrast $\zeta_2(D_\omega)$ is determined by the ratio τ_c/T , where $\tau_c = 2\pi/D_\omega$ is the coherence time of the wave field and T is the time resolution of the detectors. Equivalently, the contrast $\zeta_2(D_\omega)$ determines the number of longitudinal modes $M_T = 1/\zeta_2(D_\omega)$ for a chaotic source. By definition the SDC $|\mu(\mathbf{r}_1, \mathbf{r}_2)| \leq 1$ and intensity correlation function $g^{(2)}(\mathbf{r}_1, \mathbf{r}_2) \leq 2$.

For the full statistical description of the wave field the n th order correlation functions can be introduced [3,15]

$$g^{(n)}(\mathbf{r}_1, \dots, \mathbf{r}_n) = \frac{\langle \prod_{i=1}^n I(\mathbf{r}_i) \rangle}{\prod_{i=1}^n \langle I(\mathbf{r}_i) \rangle}. \quad (3)$$

For chaotic light $g^{(n)}(\mathbf{r}_1, \dots, \mathbf{r}_n)$ is completely described by the SDC $\mu(\mathbf{r}_1, \mathbf{r}_2)$ due to the Gaussian moment theorem [15,24]. A comparison between correlation functions of different orders determines whether the field obeys Gaussian statistics or not. In particular, for chaotic light

the n th order correlation function $g^{(n)}(\mathbf{r}, \mathbf{r}, \dots, \mathbf{r})$ is equal to $n!$ for a single longitudinal mode [15].

For pulsed sources the intensity correlation measurements are naturally gated by the pulse duration T [25]. FEL sources, with pulses of few tens of femtoseconds, are ideally suited for intensity correlation measurements. According to FEL theory [26,27] these sources should obey Gaussian statistics in the linear and deep nonlinear regime of operation. In these conditions the correlation function $g^{(2)}(\mathbf{r}_1, \mathbf{r}_2)$ has the form of Eq. (2) and provides access to the transverse coherence properties of an FEL as well as to its pulse duration [25].

The experiment was carried out at FLASH that was operated with six undulator modules and a total undulator length of 30 m. The electron bunch charge was 600 pC, and the electron energy 1.08 GeV resulting in a photon wavelength of $\lambda = 5.5$ nm. The average photon pulse energy was about 110 μ J, which corresponds to about 3×10^{12} photons per pulse at this photon energy. The measurements were performed at the PG2 beam line [28,29] (see Fig. 1). The optical system focuses the beam at a distance of 71.5 m downstream from the undulator exit. A monochromator comprised of a plane grating (PG), collimating (M1) and focusing (M2, M3) mirrors, and an exit slit with variable slit width was utilized to modify the bandwidth. The plane grating has a line density of 200 lines/mm and was tuned to its third order resulting in a dispersion in the exit slit plane of 0.64 eV/mm. The resolution of the monochromator was $\Delta E = 7$ meV, which corresponds to an energy bandwidth

of $\Delta E/E = 3.1 \times 10^{-5}$. Exit slits of 30, 60, 150, 300, and 500 μ m in size were used to select the spectral width. These values correspond to energy bandwidths $\Delta E/E$ of 0.8×10^{-4} , 1.7×10^{-4} , 4.0×10^{-4} , 0.8×10^{-3} , and 1.4×10^{-3} .

In contrast to the original HBT experiment [1,2] with two separated detectors, we utilize a pixel detector for correlation measurements. It allows us to determine second- and higher-order correlation functions at all separation points simultaneously. An in-vacuum CCD (Andor Ikon, 2048×2048 pixels, each $13.5 \mu\text{m} \times 13.5 \mu\text{m}$ in size) was positioned at a distance of 3.3 m behind the focus of the beam line. The detector was operated at a repetition rate of 10 Hz with a region of interest $2047(\text{H}) \times 460(\text{V})$ pixels and binning by five pixels in the vertical direction. A silicon nitride film $14 \text{ mm} \times 14 \text{ mm}$ in size and $1 \mu\text{m}$ thick was positioned approximately 30 cm upstream from the camera to attenuate the beam. The transmission of the film at this energy was 4×10^{-6} . About 2×10^4 intensity profiles were recorded for each monochromator setting [24]. Typical single pulse and averaged intensities are shown in Figs. 1(b)–1(d). Shot-to-shot fluctuations in these intensity profiles, as a consequence of the SASE process are clearly visible. The intensity profiles were averaged along the vertical direction, which is the dispersive direction of the monochromator [see Figs. 1(e)–1(g)]. The intensity correlation analysis was performed in the horizontal direction.

The normalized second-order correlation function $g^{(2)}(x_1, x_2)$ for a narrow bandwidth of 0.8×10^{-4} is shown in Figs. 2(a) and 2(c). Remarkably, it reaches the maximum value of two at small separations [see Fig. 2(c)], which indicates that the contrast $\zeta_2(D_\omega)$ at that monochromator setting is close to one. This contrast is significantly higher than at synchrotron sources [30–32], where it did not exceed 0.3. This 100% contrast indicates that at these conditions there is no degradation of the averaged coherence due to optics imperfection. A Gaussian fit, $1 + \exp(-\Delta x^2/l_c^2)$, to the second-order correlation function $g^{(2)}(\Delta x)$ as a function of the separation $\Delta x = x_2 - x_1$ around the center of the beam [see Fig. 2(c)] provided a transverse coherence length of $l_c = 0.93 \pm 0.04$ mm. This value is substantially larger than the measured beam size in the horizontal direction [0.45 mm (FWHM)], indicating a high coherence of the beam. We quantified it by evaluating the degree of spatial coherence ζ_S defined as [27,33] $\zeta_S = \int |W(x_1, x_2)|^2 dx_1 dx_2 / (\int S(x) dx)^2$ and obtained $\zeta_S = 0.78 \pm 0.01$, which is in a good agreement with the Young's double pinhole measurements at FLASH [21]. Importantly, we can estimate the degeneracy parameter [15], which is the number of photons in a single mode. Our estimates, obtained by integrating the total flux on the detector at the narrow bandwidth, yield a value of 10^9 comparable to Ref. [21], which is significantly higher than at any synchrotron sources.

The second-order correlation function $g^{(2)}(x_1, x_2)$ for a larger bandwidth of 1.4×10^{-3} is shown in Figs. 2(b) and 2(d). This larger bandwidth is equivalent to a shorter

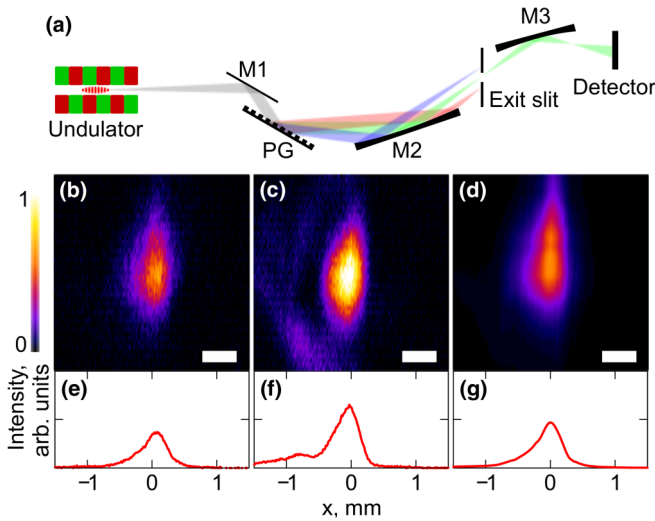


FIG. 1 (color online). (a) Scheme of the experiment. FEL radiation is generated in the undulator and is transmitted through the beam line including one collimating (M1) and two focusing (M2, M3) mirrors, a plane grating (PG), and an exit slit. Intensity profiles of individual femtosecond pulses are measured at the detector. (b),(c) Typical single pulse intensity profiles and (d) an average over 2×10^4 pulses for a bandwidth of $\Delta E/E = 0.8 \times 10^{-4}$. (e)–(g) Projections of the pulse intensities along the vertical direction. The scale bar is 0.5 mm long.

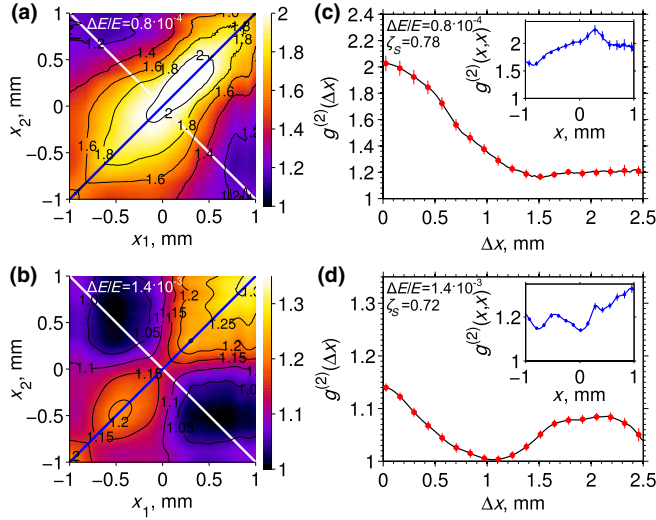


FIG. 2 (color online). Intensity correlation analysis. (a),(b) Intensity correlation function $g^{(2)}(x_1, x_2)$ for a bandwidth of 0.8×10^{-4} (a) and 1.4×10^{-3} (b). (c),(d) Intensity correlation function $g^{(2)}(\Delta x)$ taken along the white line in (a),(b) around the center of the beam. Insets in (c),(d) show intensity fluctuations $g^{(2)}(x, x) = \langle I^2(x) \rangle / \langle I(x) \rangle^2$ taken along the blue line in (a),(b). The error bars in (c),(d) are obtained by statistical analysis of 20 individual subensembles (10^3 shots each) from the whole set of 2×10^4 shots. The number of points displayed in this figure with error bars is reduced for better visibility.

coherence time, and the contrast of $g^{(2)}(x_1, x_2)$ is reduced as expected from Eq. (2). Unexpectedly, at these conditions we observed an oscillatory shape of the correlation function $g^{(2)}(\Delta x)$ [see Fig. 2(d)]. This may originate from the contribution of two independent sources in the lasing conditions of FLASH [34]. It could also be a more general feature of the nonlinear coupling of the electrons and emitted SASE radiation. To disentangle this question, further experimental and simulation studies of the FEL radiation at different lasing conditions will be required. We want to remark that such fine features of FEL lasing would be difficult to observe by other means. For example, our averaged spectral measurements [see Fig. 3(b)] do not show any notable features.

Results of our experiments also indicate that the statistical properties of the FEL beam are not spatially uniform. This is well visualized by the inspection of the beam fluctuations $g^{(2)}(x, x) = \langle I^2(x) \rangle / \langle I(x) \rangle^2$ along the beam profile [see insets in Figs. 2(c) and 2(d)]. They vary significantly both for small and large bandwidths.

We also analyzed the contrast in the center of the beam $\zeta_2(D_\omega) = g^{(2)}(0) - 1$ as a function of the coherence time τ_c [see Fig. 3(a)]. It has a linear dependence as a function of τ_c at a large bandwidth and reaches saturation at a small bandwidth, as described by Eq. (2) [24]. A theoretical fit to the contrast values yields an average pulse duration of 47 ± 21 fs (FWHM).

To get an independent estimate of the pulse duration, we measured the second-order correlation function

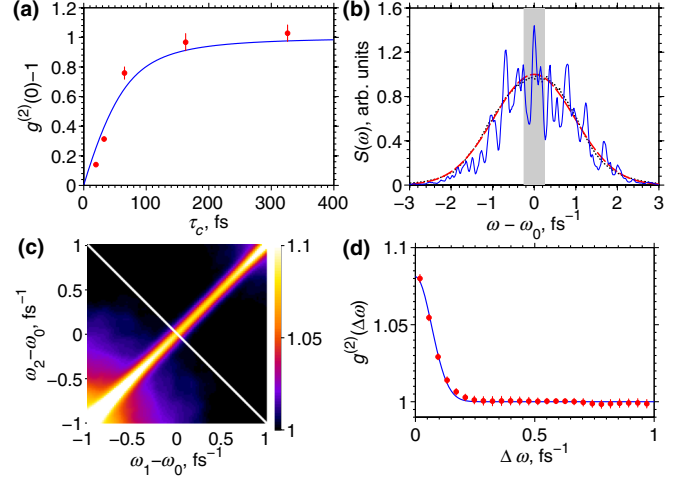


FIG. 3 (color online). Statistical properties of FEL light as a function of the bandwidth. (a) Contrast (points) $\zeta_2(D_\omega) = g^{(2)}(0) - 1$ as a function the coherence time τ_c . The error bars are same as in Figs. 2(c) and 2(d). Theoretical fit (solid line) corresponds to a pulse duration of $T = 47 \pm 21$ fs. (b) Measured single pulse (blue solid line) and average (black dotted line) spectra. Gaussian fit (red dashed line) gives an FEL bandwidth of $\Delta E/E = 6.7 \times 10^{-3}$ FWHM. The width of the largest exit slit used in the experiment and corresponding to the bandwidth $\Delta E/E = 1.4 \times 10^{-3}$ is shown by the gray region. (c) Intensity correlation function $g^{(2)}(\omega_1, \omega_2)$ in the spectral domain. (d) Intensity correlation function $g^{(2)}(\Delta\omega)$ (red circles) taken along the white line in (c). Gaussian fit (blue line) gives an average pulse duration of 27 fs.

$g^{(2)}(\omega_1, \omega_2)$ in the spectral domain as a function of two frequencies [see Fig. 3(c)] [36,37]. About 1.5×10^4 single shot spectra were recorded after the intensity correlation measurements with a detector at the position of the exit slit of the monochromator. This detector is comprised of a scintillating screen (YAG:Ce 0.2%) and an intensified CCD (Andor iStar, DH740), equipped with a lens. The effective pixel size of the detector in the exit slit plane was $19.4 \mu\text{m}$ with the point spread function estimated to be about two pixels (FWHM). The detector was operated at a repetition rate of 10 Hz. Line profiles, obtained from the central part of the beam in the horizontal direction corresponding to about 10% of the beam FWHM were analyzed [24]. A Gaussian fit $1 + \exp(-\Delta\omega^2 T^2)$ was used to determine the pulse duration T from the measurements of the second-order correlation function $g^{(2)}(\Delta\omega)$ [see Fig. 3(d)]. According to this analysis we obtained an averaged pulse duration of 27 fs (FWHM), which lies in the uncertainty range of the previous measurements. In general, our estimates of the pulse duration can be shorter than the intrinsic pulse duration in time domain due to a possible frequency chirp of FLASH pulses [37–39].

To get an insight into the photon statistics of FEL pulses we studied intensity correlation functions of order higher than two. Higher-order correlation functions $g^{(3)}(x_1, x_2, x_3)$ (with $x_3 = 0$) and $g^{(4)}(x_1, x_2, x_3, x_4)$ (with $x_3 = x_1/2$ and

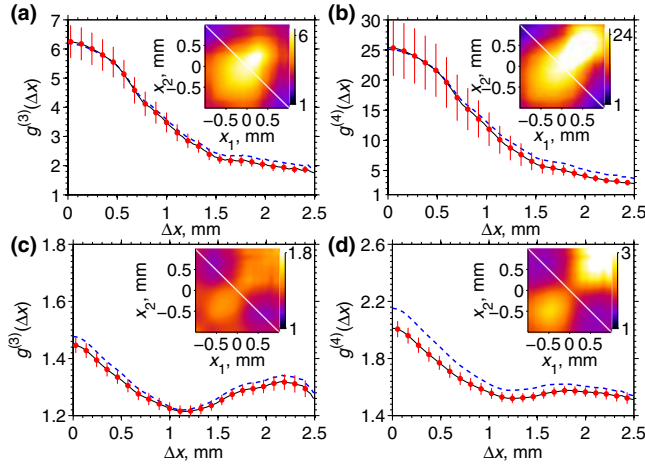


FIG. 4 (color online). Higher order correlation functions for a bandwidth of 0.8×10^{-4} (a),(b) and 1.4×10^{-3} (c),(d). (a),(c) Third-order correlation function $g^{(3)}(x_1, x_2)$ (shown in the insets) and $g^{(3)}(\Delta x)$ (black solid line) taken along the white line in the insets. (b),(d) Fourth-order correlation function $g^{(4)}(x_1, x_2)$ (shown in the insets) and $g^{(4)}(\Delta x)$ (black solid line) taken along the white line in the insets. The same functions obtained under assumption of the Gaussian statistics are shown as blue dashed lines. The number of points displayed in this figure with error bars is reduced for better visibility. Error bars are evaluated in the same way as in Fig. 2.

$x_4 = x_2/2$) are presented in the insets of Fig. 4 for two different bandwidths as functions of x_1 and x_2 . In Fig. 4 the same quantities are presented as a function of Δx around the center of the beam with $x_1 = -\Delta x/2$, $x_2 = \Delta x/2$. The higher-order correlation functions were also calculated under the assumption of Gaussian statistics [24] from the measured second-order correlation function $g^{(2)}(x_1, x_2)$ (see Fig. 4). The excellent agreement between these two curves confirms the chaotic nature of the FLASH source at these operation conditions. In particular, at a small bandwidth we observed a factorial behavior of higher order correlation functions at zero point separation $g^{(n)}(\mathbf{r}, \mathbf{r}, \dots, \mathbf{r}) \approx n!$ [see Figs. 4(a) and 4(b)], which is typical for Gaussian statistics. At a wider bandwidth the maximum value is substantially reduced compared to a narrow bandwidth case [see Figs. 4(c) and 4(d)].

In summary, we have presented high-order intensity correlation measurements at FLASH which provide a simple, robust, and versatile tool for monitoring basic beam properties of FELs including the degree of spatial coherence, average pulse duration, and details of the photon statistics. We obtained an averaged pulse duration less than 50 fs, degree of transverse coherence of about 80%, and degeneracy parameter of more than 10^9 . Such values are similar to laser sources and were never observed at these wavelengths at conventional synchrotron radiation sources. However, our measurements of the higher-order correlation functions indicate that present FEL sources based on the SASE principle are essentially chaotic sources obeying Gaussian statistics.

The measurements presented in this work were performed at XUV photon energies that can be conveniently reached at FLASH. We anticipate that there are no principal limitations to extend our approach to every existing or planned FEL source, including hard x-ray FELs. This is in contrast to Young's double pinhole experiments, which are limited to the soft x-ray energy region due to high penetration depth of hard x rays and difficulties of manufacturing thick opaque apertures.

An interesting further application of the methods developed in this work would be the study of seeded FEL sources [40–42]. An intriguing question is whether seeded FELs are fully coherent sources in all orders according to Glauber [3] and, in this way, are equivalent to conventional single mode lasers, or if they obey Gaussian statistics like SASE FELs. We could also foresee that intensity correlation analysis might be applied to study the dynamics of ultrafast processes at FELs. For example, the intensity correlation analysis of the Coulomb explosion in single molecule imaging experiments [43] could provide detailed information about the disintegration of these molecules on a femtosecond time scale. The exploding molecule can be considered as a secondary source and its size at different instants of time can be studied using the intensity correlation method.

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