Thermal x-ray diffraction and near-field phase contrast imaging

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Abstract – Using higher-order coherence of thermal light sources, the resolution power of standard x-ray imaging techniques can be enhanced. In this work, we applied the higher-order measurement to far-field x-ray diffraction and near-field phase contrast imaging (PCI), in order to achieve superresolution in x-ray diffraction and obtain enhanced intensity contrast in PCI. The cost of implementing such schemes is minimal compared to the methods that achieve similar effects by using entangled x-ray photon pairs.

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Introduction. – The Hanbury-Brown and Twiss (HBT) effect [1] was at the heart of the development of quantum optics, which fundamentally reveals the higher-order coherence of a multimode thermal source. Thermal light ghost imaging based on the HBT effect could be interpreted as a quantum mechanical two-photon interference effect [2–9]. After two decades of intense debate, the disagreement on the quantumness and classicality of thermal light ghost imaging still persists [10,11]. However, recent rigorous investigations revealed evidence that in the low-illumination (i.e., photon counting) regime, it must require a quantum model to describe the ghost image formation [12–14]. It was demonstrated that even the so-called “classical” thermal light must contain nonzero genuine quantum correlations as measured by quantum discord [15,16]. In the large-photon-number regime, the ghost imaging system could transition into the classical regime, nevertheless the quantum-mechanical description evidently remains valid and is quantitatively identical to the classical one, since it is the classical limit of quantum states of thermal light. Leaving aside the quantum vs. classical debate, the ghost imaging actually offers enormous possibilities for x-ray imaging hitherto not fully exploited [9,17,18]. Here we show that using HBT type measurements, we can enhance the resolution power of conventional x-ray imaging techniques, namely x-ray far-field diffraction imaging and x-ray phase contrast imaging (PCI) in the near field. This paper is organized as follows: in the Methods section, we demonstrate how the two-photon interference effect can be utilized to x-ray diffraction imaging to double the resolution, we then investigate two-photon interference with respect to the near-field phase contrast imaging and demonstrate an enhanced image contrast in PCI. We then discuss the experimental feasibility of implementing the proposed schemes in the x-ray regime, and the methods to realize the far- and near-field imaging setups without beam splitter, which would pose technical difficulties in the x-ray regime.

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Methods.  

Enhanced x-ray diffraction.  According to the quantum theory of photo-detection [4,5], an idealized pixel photo-detector measures the probability of observing a photo-detection event at spacetime point \((\vec{r}, t)\)

\[
G^{(1)}(\vec{r}, t) = \text{tr}\left\{\hat{\rho}\hat{E}^+(\vec{r}, t)\hat{E}^-(\vec{r}, t)\right\},
\]

where \(\hat{\rho}\) is the density operator of the quantized photon field, \(\hat{E}^-(\vec{r}, t)\) and \(\hat{E}^+(\vec{r}, t)\) are the negative and positive frequency parts of the electric-field operators. A joint-detection of two independent pixel photo-detectors \(D_A\) and \(D_B\) measures the probability of a coincident event of two photons at spacetime points \((\vec{r}_A, t_A)\) and \((\vec{r}_B, t_B)\)

\[
G^{(2)}(\vec{r}_A, t_A; \vec{r}_B, t_B) = \text{tr}\left\{\hat{\rho}\hat{E}^+(\vec{r}_A, t_A)\hat{E}^-(\vec{r}_B, t_B)\hat{E}^+(\vec{r}_B, t_B)\hat{E}^-(\vec{r}_A, t_A)\right\}.
\]

Let us now consider a thermal light source from which we detect the light fields at \((\vec{r}_A, t_A)\) and \((\vec{r}_B, t_B)\). This can be either a thermal light source subject to a certain propagation distance or a coherent synchrotron source. In the low-photon-number regime, the light field can be modeled by an effective multimode photon wave function \(|\Psi\rangle \approx |0\rangle + \sum_{\vec{k}} f(\vec{k})|\vec{k}\rangle|0\rangle\), where \(f(\vec{k})\) is the probability amplitude for the radiation field to be in the single-photon mode of wave vector \(\vec{k}\) [2]. We assume that the light fields possess a certain coherence time \(\tau_c\) over which second-order coherence is present and a time window for coincidence detection \(\tau = t_2 - t_1 \ll \tau_c\). Following this assumption we can omit the time dependency from the correlation function. Note that the second-order correlation function of a thermal light field measured at the same position is then given by temporal coherence of \(G^{(2)}(\tau \sim 0) \sim 2\), a fact well-known as photon bunching [2]. We use the notation \(\vec{r}_i = (\vec{p}_i, z_i; \vec{k})\) and \(\vec{k} = (\vec{\kappa}, \sqrt{\kappa^2 - \kappa^2})\) and the thermal light can be modeled as a mixed state with density operator [2]

\[
\hat{\rho} \approx |0\rangle\langle 0| + \sum_{\vec{\kappa}} |f(\vec{\kappa})|^2|\vec{\kappa}\rangle\langle \vec{\kappa}|1_\kappa 1_\kappa|1_\kappa 1_\kappa|,
\]

where \(1_\kappa = \hat{a}_\kappa^\dagger|0\rangle\). The transverse part of the photon field on the two detectors \(D_A\) and \(D_B\) can be written as

\[
\hat{E}^+(\vec{p}_j) \simeq \sum_{\vec{\kappa}} g_j(\vec{p}_j, z_j; \vec{\kappa})\hat{a}_{\vec{\kappa}},
\]

where \(g_j(\vec{p}_j, z_j; \vec{\kappa})\) is the Green’s function for a single-mode photon. Then, the spatial part of the second-order coherence function, which is proportional to the joint photon counting rate, reads

\[
G^{(2)}(\vec{p}_A, z_A; \vec{p}_B, z_B) = 
\sum_{\vec{\kappa}, \vec{\kappa}’} |1_\kappa 1_{\kappa’}| \langle \hat{\Psi}^+(\vec{p}_A, z_A)\hat{\Psi}^-(\vec{p}_B, z_B)\hat{\Psi}^+(\vec{p}_B, z_B)\hat{\Psi}^-(\vec{p}_A, z_A)\rangle.
\]

To calculate the measurement signal quantified by eq. (6), we propagate the thermal light field from the source to the detector plane. The required Green’s function reads

\[
G^{(1)}(\vec{p}_A, z_A; \vec{p}_B, z_B) = \sum_{\vec{\kappa}} g_A(\vec{p}_A, z_A; \vec{\kappa})g_B(\vec{p}_B, z_B; \vec{\kappa}),
\]

where \(g_j(\vec{p}_j, z_j; \vec{\kappa})\) is the Thomson scattering amplitude of a photon with the \(i\)-th atom at position \(\vec{r}_i\). Suppose the two-photon interference, which is imprinted in the photon number fluctuation, is from scattering from atoms in two adjacent lattice plane with a distance of \(d\) and reflection angle \(\theta\). Then the interference of the two indistinguishable two-photon quantum paths
acquires a doubled optical path difference compared to conventional Laue diffraction, which translates to a maximally achievable resolution of \( \frac{\lambda}{4} \) (according to Abbe’s criteria for resolution and under the assumption that the measurement is taken over the full solid angle). From the Green’s function \( g_j(\vec{r}_i, \vec{p}_j, z_j; \vec{r}) \) for a single-mode photon that propagates to detector \( D_j \) and scatters from the atom at \( \vec{r}_i \), we have

\[
G^{(1)+}(\vec{p}_A, z_A; \vec{p}_B, z_B)G^{(1)}(\vec{p}_A, z_A; \vec{p}_B, z_B) = \\
\sum_{\vec{r}} \sum_{ii'} g_A(\vec{r}_i, \vec{p}_A, z_A; \vec{r})g_B(\vec{r}_{i'}, \vec{p}_B, z_B; \vec{r}) \\
\times \sum_{\vec{r}'} g_A(\vec{r}_i, \vec{p}_A, z_A; \vec{r}')g_B(\vec{r}_{i'}, \vec{p}_B, z_B; \vec{r}'). \tag{8}
\]

Taken the summation over transverse momentum as integral, and denote \( \delta = d \sin \theta \), we find the following term in eq. (8) that gives the doubled optical path difference from the two-photon interference depicted in fig. 1(a) (see the derivation in the Supplementary Material).

For x-ray diffraction, the doubled optical path difference translates to enhanced resolution, since we obtain a modified Bragg condition for the thermal light two-photon diffraction

\[
4d \sin \theta = n \lambda, \tag{10}
\]

for an integer \( n \). It implies that structures with periods of \( \frac{\lambda}{4} < d < \frac{\lambda}{2} \) are able to form Bragg peaks in the two-photon signal and can thus be resolved by inverse Fourier transformation and phase retrieval techniques. Compared to the two-photon diffraction using entangled photon pairs from x-ray parametric down-conversion (XPDC), which has rather low photon flux, the present scheme using a thermal source can be experimentally favorable. In the case of wavefront distortion or femtosecond serial nanocrystallography that effectively produces powder diffraction patterns, we can replace \( D_A \) and \( D_B \) by two pixel detectors and a sufficient large beam splitter covering the angular spread of the reflected photons, provided each pixel of the detectors are pairwise connected by a coincidence circuit. Or we can record the photon count map and correlate the image in post processing.

The thermal instability of the source, changes in refractive index in the optical paths, and shot-to-shot variation of wavefront of the x-ray beams from x-ray free electron lasers can introduce turbulence into the diffraction and imaging system. However, the two-photon interference can be free of these types of turbulence [12], hence the x-ray ghost diffraction system could be robust and does not require wavefront correction. As depicted in fig. 1(b), the turbulence can be treated as an arbitrary phase perturbation to the Green’s function in eq. (5), and we show in the SM, that the final diffraction pattern determined by \( G^{(2)}(\vec{r}_A, \vec{r}_B) \) is invariant under phase perturbations. It can be similarly shown that the two-photon interference could be free of amplitude perturbation (see appendix).

This is a strong evidence of quantum correlation in ghost image formation, since the classical speckle-to-speckle correlation is sensitive to such turbulence [12].

**Enhanced phase contrast imaging.** As a complementary technique to the far-field x-ray diffraction, x-ray phase contrast imaging (PCI) is a widely applied near-field imaging technique which provides a direct image of the sample [19–21]. PCI is suitable for samples that only weakly absorb photons or only induce phase shift of the photon, the image of the sample is formed on a pixel detector with point-to-point projection. For the assumption that the scatterers in the sample introduce a small phase \( \varphi \), which and the sample no further inhomogeneous phase shift, the image is formed with point-to-point projection.

For the setup shown in fig. 2, the joint intensity can be applied to PCI and enhance the intensity contrast.
written as

\[ (I_A(\vec{\rho}_A)I_B(\vec{\rho}_B)) = G^{(2)}(\vec{\rho}_B, \vec{\rho}_A) = G^{(1)}(\vec{\rho}_B, z_B) G^{(1)}(\vec{\rho}_A, z_A) + \left| G^{(1)}(\vec{\rho}_B, z_B, \vec{\rho}_A, z_A) \right|^2. \]

Assume the refractive index of the sample at a point \( \vec{x} \) is \( n_s(\vec{x}) = 1 - \delta_s(\vec{x}) \) with \( \delta_s(\vec{x}) \ll 1 \). The electric field on the detector plane at spacetime \((\vec{r}_i, t)\) can be written under first Born approximation as

\[ E(\vec{r}_i, \omega) = E_{in}(\vec{r}_i, \omega) - \frac{k^2}{2\pi} \int d^3 \vec{x}' e^{ik|\vec{r}_i - \vec{x}'|/|\vec{r}_i - \vec{x}'|} \delta_s(\vec{x}') E_{in}(\vec{x}', \omega), \quad (11) \]

where \( i = A, B \). Defining the coordinates on the pixel detectors as \( \vec{r}_i = (\vec{\rho}_i, z_i) \), the coherence function can be expressed as

\[ G^{(1)}(\vec{\rho}_B, z_B, \vec{\rho}_A, z_A) = \frac{1}{2\pi} \int d\omega_B d\omega_A \delta_1 \left( \hat{E}_{in}^*(\vec{\rho}_B, \omega_B) \hat{E}_{in}(\vec{\rho}_A, \omega_A) e^{i\Delta \omega_{AB}t} \right) - \frac{1}{2\pi} \int d\omega_B d\omega_A \int d^3 \vec{x}' e^{ik|\vec{\rho}_B - \vec{x}'|/|\vec{\rho}_B - \vec{x}'|} \delta_s(\vec{x}') \left( \hat{E}_{in}^*(\vec{\rho}_B, \omega_B) \hat{E}_{in}(\vec{\rho}_A, \omega_A) e^{-i\omega_{AB}t} \right) \]

\[ - \frac{1}{2\pi} \int d\omega_B d\omega_A \int d^3 \vec{x}' e^{ik|\vec{\rho}_A - \vec{x}'|/|\vec{\rho}_A - \vec{x}'|} \delta_s(\vec{x}') \left( \hat{E}_{in}^*(\vec{\rho}_B, \omega_B) \hat{E}_{in}(\vec{\rho}_A, \omega_A) e^{-i\omega_{AB}t} \right), \quad (12) \]

where \( \Delta \omega_{AB} = \omega_B - \omega_A \). Under the condition \( z_A = z_B \) and \( \vec{\rho}_A = \vec{\rho}_B = \vec{\rho} \), we obtain the multipath coherence function as (see the derivation in the SM)

\[ G^{(1)}(\vec{\rho}_B, z_B, \vec{\rho}_A, z_A) = I \left( t - \frac{z}{c} \right) \]

\[ \times \left( 1 - \frac{k}{4\pi z} \right) \int d^2 \vec{\rho} e^{-\frac{z^2}{4\pi^2}} \frac{\nabla^2 \varphi(\vec{\rho} - \vec{\rho}')}{|\vec{\rho} - \vec{\rho}'|^2}. \]

The point-to-plane-formed image on detector plane at \( \vec{\rho} \) is a convolution of the phase contrast \( \nabla^2 \varphi \) centered at \( \vec{\rho} \) with radius \( |\vec{\rho} - \vec{\rho}'| \) and a Gaussian weight. Thus the resolution requirement leads to the condition for the coherence length

\[ \frac{z\lambda}{\pi p} \leq l_c, \quad (13) \]

as well as the near-field condition \( \frac{z}{\pi p} \ll 1 \). Provided these conditions are satisfied, we finally obtain

\[ G^{(1)}(\vec{\rho}_B, z_B, \vec{\rho}_A, z_A) = I \left( t - \frac{z}{c} \right) \left( 1 - \frac{z}{k} \nabla^2 \varphi(\vec{\rho}) \right), \quad (14) \]

which has the same form as \( G^{(1)}(\vec{\rho}, z) \), and does not smear out the image contrast in the baseline signal. In the conventional PCI, the image intensity is determined by [21]

\[ I(\vec{\rho}, z) = G^{(1)}(\vec{\rho}, z) = I \left( 1 - \frac{z}{k} \nabla^2 \varphi(\vec{\rho}) \right), \quad (15) \]

Given a coherence length \( l_c \) and incident photon wavelength, the propagation length \( z \) behind the exit plane cannot be arbitrarily extended, thus in the conventional PCI, the intensity contrast \( \frac{\Delta I}{I} \) cannot be simply enhanced. However, as we show in eq. (14), the photon number fluctuation \( \Delta I = 1 \) should not smear out the signal. Instead it has the same intensity contrast as the baseline signal \( I_A(\vec{\rho})I_B(\vec{\rho}) \), and thus we have the image intensity of joint photo-detection as

\[ \langle I_A(\vec{\rho})I_B(\vec{\rho}) \rangle \simeq I^2 \left( 1 - \frac{2z}{k} \nabla^2 \varphi(\vec{\rho}) \right), \quad (16) \]

which gives a doubled intensity contrast. It can be expected that higher-order coherence measurement of thermal light [22] can further enhance the intensity contrast.

**Discussion.** As shown in the above derivations, the measurement of intensity-fluctuation correlations holds potential to substantially increase the performance of x-ray diffraction imaging and near-field PCI. For the practical implementation of the proposed x-ray imaging techniques based on two-photon interference, we have to consider that the use of any optical element poses a challenge toward the experimental realization. As such we discuss detection schemes here that do not require the beam splitter of figs. 3 and 2. The beam splitter usually enables the use of a pair of single-photon-counting detectors, such as avalanche photo-diodes, for intensity correlation measurements. The advantages are that one can measure in CW mode, avoid dead time effects and have a high temporal
defined as coincidence detection. In (b), the detection of \( D_n \) counting detector, and the single-pixel photon-counting event resolution. However the average count rate on a single detector should be much less than unity and only a single position \( \rho_A = \tilde{\rho}_B \) or \( \rho_A \neq \tilde{\rho}_B \) can be measured at once. Hence the detector pair needs to be raster scanned across the detection plane.

If a pulsed scheme is considered the natural time-gating capability of ultra-short pulses removes the need for high temporal resolution in the detectors, such that a large pixelated detector can be utilized, which additionally covers the entire detection plane at once. Conducting a measurement with two different pixels \( \rho_A \neq \tilde{\rho}_B \) the second-order correlation function is simply given by \( G^{(2)}(\rho_A, \tilde{\rho}_B) \sim \langle \hat{a}^\dagger_A \hat{a}^\dagger_B \hat{a}_A \hat{a}_B \rangle = \langle \hat{a}^\dagger_B \hat{a}_A \rangle \). Here \( \hat{a}^\dagger \) and \( \hat{a} \) denote the creation and annihilation operator of a photon in the (spatial) mode \( \rho_A \) or \( \tilde{\rho}_B \), respectively, and \( \hat{n} \) is the corresponding number operator. Since the two pixels are independent, one can simply multiply the photon counts of different pixels and average over a series of pulses to obtain the second-order correlation function.

However, we are mostly interested in the case of \( \rho_A = \tilde{\rho}_B \). In case of the PCI scheme with a continuous pattern, it is possible to correlate adjacent pixels that sample nearly the same mode created by the sample at the position of the detector (see fig. 4(b)). Then the previous calculation method applies. In case of crystallography with sharp Bragg peaks (see fig. 4(a)) that are much smaller than the pixel size, the adjacent pixel will not sample the same mode such that one would be required to use the same pixel with photon-number-resolving capability.

Then \( G^{(2)}(\rho_A, \rho_A) \sim \langle \hat{a}^\dagger_A \hat{a}^\dagger_A \hat{a}_A \hat{a}_A \rangle = \langle \hat{a}^\dagger_A \hat{a}_A \rangle = \langle \hat{n}^2_A - \hat{n}_A \rangle \), which can equally be evaluated.

**Conclusion.** – In the present work, we have demonstrated that using the higher-order coherence properties of thermal light, we can achieve an enhanced resolution for conventional x-ray diffraction imaging techniques in the far-field and enhanced intensity contrast in the near-field regime.

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