Breaking Rayleigh’s curse for two unbalanced single-photon emitters using BLESS technique

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According to the Rayleigh criterion, it is impossible to resolve two independent point sources separated by a distance below the width of the point spread function (PSF). Almost twenty years ago it was shown that the distance between two point sources can be statistically estimated with an accuracy better than the PSF width. However, the estimation error increases with decreasing distance. This effect was informally named Rayleigh’s curse. Next, it was demonstrated that PSF shaping allows breaking the curse provided that all other source parameters except for the distance are known a priori. In this work, we propose a novel imaging technique based on the target Beam moduLation and the Examination of Shot Statistics (BLESS). We show that it is capable of breaking Rayleigh’s curse even for unbalanced point sources with unknown centroid and brightness ratio. Moreover, we show that the estimation precision is close to the fundamental limit provided by the quantum Cramér–Rao bound.

1 Introduction

The standard diffraction theory claims that the far-field linear optical imaging resolution is restricted by the Rayleigh limit: two point sources cannot be resolved if the distance between them is smaller than the point spread function (PSF) width, which is proportional to the radiation wavelength [1]. There are many techniques allowing to overcome the Rayleigh limit [2]. Some of them are based on nonlinear light-matter interaction [3], complex systems of excitation and suppression of luminescence [4], and consequently have a very limited field of application.

A different strategy allowing super-resolved imaging is based on the usage of a prior information about the object. Describing the whole object with a few parameters reduces the imaging problem to the problem of statistical estimation. In particular, van den Bos group considered the problem of two point sources localization [5, 6]. Using the model of Gaussian PSF, they concluded that if the distance $d$ between sources is larger than PSF width $\sigma$, its estimation error $\Delta_d$ is proportional to $\sigma \sqrt{K}$, where $K$ is the number of registered events. If $d < \sigma$ then $\Delta_d \propto \sigma \sqrt{Kd/\sigma}$. Hence, the distance between two close point sources cannot be accurately estimated with a limited amount of data. This problem was named Rayleigh’s curse [7].

Later Tsang et al showed the possibility to overcome Rayleigh’s curse [7–9]. They considered the problem of resolving two equal point sources in terms of quantum Fisher information (QFI). They proved that QFI was independent of the distance $d$ value, which means that the parameter can be precisely estimated beyond the Rayleigh limit. Moreover, the QFI limit can almost be saturated by practical measurement protocols: SPAtial-Mode DEmultiplexing (SPADE) [7] and SuperLocalization by Image inVERsion interferometry (SLIVER) [8]. Both protocols make use of the PSF (or detection/target mode) shaping. In particular, the use of odd PSF instead of even Gaussian PSF breaks Rayleigh’s curse. This has been demonstrated in the set of proof-of-principle experiments [10–13].
But it was later shown that the Rayleigh’s curse can be overcome for the two-parameter object model only [14–20]. Two unbalanced point sources with unknown brightness ratio [15, 17] or more than two equal sources [14] cannot be precisely localized beyond the Rayleigh limit: the position estimation error increases polynomially with decreasing distance between sources.

In general, any object can be parameterized by its intensity moments. It was shown that the estimation errors of the first and the second intensity moments are independent of the object size, but the Rayleigh’s curse still holds for higher $k$-order moments $M_k$, resulting in $\Delta M_k \propto d^{1-k/2}$, so they cannot be well estimated beyond the Rayleigh limit [18–20].

To perform precise imaging of complex objects one needs to extract additional information from measurements. Previously, most of the imaging statistical estimation problems were considered in the weak source approximation, where the number of detected photons was not greater than 1, and the spatial distribution of mean photon number was measured [7–20]. However, higher order intensity (or photon number) moments give benefits for solving imaging problems. One of the first demonstrations of this was done by Brown and Twiss in their stellar interferometer experiment [21]. It was later shown that PSF for $N$-order intensity moment measurement was $\sqrt{N}$ times narrower than the first-order one [22]. This was experimentally applied to the single-photon emitters imaging [23–25]. Also, photon statistics analysis was used in STochastic Optical Reconstruction Microscopy (STORM) [26] and PhotoActivated Localization Microscopy (PALM) [27] techniques, where the positions of independently blinking point sources were estimated from a set of frames obtained at different times. Using higher-order correlations for statistical reconstruction was recently reported [28, 29], but the break of the Rayleigh’s curse was not demonstrated.

Below in Section 2 we propose an approach that combines statistical estimation of image parameters, PSF shaping and the examination of photon statistics distribution. In Section 3 we describe the motivation behind using this approach. Using the classical and quantum Cramér–Rao bound (Section 4) we show that this technique allows breaking Rayleigh’s curse for two unbalanced single-photon emitters (Section 5).

## 2 BLESS technique

Consider a 1D imaging problem for two uncorrelated single-photon point sources $S_a$ and $S_b$ located at $x_a$ and $x_b$ respectively (Fig. 1). The sources have the following photon number distributions:

$$P_{a,b}(n) = \delta_0(1-\mu_{a,b}) + \delta_1 n \mu_{a,b}, \quad (1)$$

where $\delta_{ij}$ is the Kronecker delta, $\mu_a$ and $\mu_b$ are mean photon numbers. One can describe this object using the following set of 4 parameters:

- distance $d = x_a - x_b$,
- total mean photon number $\mu = \mu_a + \mu_b$,
- centroid $x_c = (\mu ax_a + \mu bx_b) / \mu$,
- relative brightness $\gamma = (\mu_a - \mu_b) / \mu \in [-1, 1]$.

The light from the source is passed throw a 4f imaging system with 1:1 magnification. Since the imaging lens has a limited numerical aperture (NA), the light from the source $s$, located at $x_a$ has a Gaussian far-field electric field

$$\tilde{\Psi}_a(q) = \frac{1}{(\pi \sigma^2)^{1/4}} \exp \left(-\frac{q^2 \sigma^2}{2} + ix_a\right), \quad (2)$$

which is related to the Gaussian near-field electric field

$$\Psi_a(x) = \frac{1}{(\pi \sigma^2)^{1/4}} \exp \left(-\frac{(x-x_a)^2}{2\sigma^2}\right), \quad (3)$$

Figure 1: The principal imaging scheme. SLM – spatial light modulator, SMF – single-mode fiber, PNRD – photon number resolving detector.
called Point Spread Function (PSF). Its width \( \sigma \sim \lambda/\text{NA} \). For the source, located at \( x_b \) we have the similar equation for \( \Psi_b(x) \).

In the image plane light is coupled to the single-mode fiber (SMF) collimator which forms a Gaussian target (detection) mode at position \( x_D \) with waist \( \sigma_0 \):

\[
\Psi_0(x) = \frac{1}{(\pi\sigma_0^2)^{1/4}} \exp \left[ -\frac{(x-x_D)^2}{2\sigma_0^2} \right].
\]

(4)

Here and below lower index \( D \) corresponds to the detection process. Scanning \( x_D \) one can measure the image profile. Additionally, one can place a Spatial Light Modulator (SLM) between the lenses which transforms the Gaussian HG\(_0\) Target Mode into the first Hermite-Gaussian mode HG\(_1\) with the field distribution

\[
\Psi_1(x) = \frac{\sqrt{2}(x-x_D)}{\sigma_0(\pi\sigma_0^2)^{1/4}} \exp \left[ -\frac{(x-x_D)^2}{2\sigma_0^2} \right].
\]

(5)

Below we will show that this target beam modulation combined with a photon number distribution measurement plays a key role in precise emitters localization.

In order to measure the photon number distribution, the SMF output is connected to a photon number resolving detector (PNRD). For simplicity we assume the detector quantum efficiency to be 100\%. The probability of detecting a single photon emitted by the point source \( S_a \) is then

\[
T_a^{(0,1)} = \left| \int \Psi_{0,1}(x)\Psi_a(x)dx \right|^2.
\]

(6)

Here and below we consider the case \( \sigma_0 = \sigma \) which is a trade-off between the high resolution and high efficiency. Under this condition

\[
T_a^{(0)} = \exp \left[ -\frac{(x_a-x_D)^2}{2\sigma^2} \right]
\]

(7)

for HG\(_0\) Target Mode and

\[
T_a^{(1)} = \frac{(x_a-x_D)^2}{2\sigma^2} \exp \left[ -\frac{(x_a-x_D)^2}{2\sigma^2} \right]
\]

(8)

for HG\(_1\) Target Mode.

Then the total probability of detecting \( k \) photons from the source \( S_a \) is

\[
P_{a,D}(k|\theta, x_D) = \sum_{n=k}^{\infty} \binom{n}{k} P_a(n)T_a^{(1)}(1-T_a)^{n-k}.
\]

(9)

The probability distribution \( P_{b,D} \) for the source \( S_b \) is calculated in the similar way.

Since the sources are uncorrelated, the convolution of \( P_{a,D} \) and \( P_{b,D} \) gives the detected photon number distribution, which depends on the detector position \( x_D \) and image parameters \( \theta = \{d, \gamma, \mu, x_c\} \):

\[
P_D(k|\theta, x_D) = \delta_{0k}(1-M_a)(1-M_b) + \delta_{1k}(M_a + M_b - 2M_aM_b) + \delta_{2k}M_aM_b,
\]

(10)

where \( M_a = \mu_aT_a \), \( M_b = \mu_bT_b \). The mean detected photon number for this distribution is

\[
M_D(x_D) = M_a + M_b,
\]

(11)

the variance is

\[
\sigma_D^2(x_D) = M_a(1-M_a) + M_b(1-M_b),
\]

(12)

and the normalized second-order correlation function is

\[
g_D^{(2)}(x_D) = \frac{2M_aM_b}{(M_a + M_b)^2}.
\]

(13)

The process of the statistical reconstruction of image parameters is performed through the measurement of photocounts across detector positions. The acquisition time is divided into the intervals comparable to the emitters’ lifetimes (shots). This division enables to measure photocount histograms reflecting the detected photon number distribution. The object parameters are then derived by fitting function \( P_D \) with parameters \( \theta \) to histograms. This innovative approach makes use of both the target Beam modulation and the Examination of Shot Statistics (BLESS), effectively enabling us to achieve subdiffraction emitter localization.

Meanwhile, the standard approach exploits the integrated number of registered photons: one estimates the parameters from measuring (11) only.

Experimentally, the use of integrated statistics corresponds to a single measurement with a long exposition time, while the shot statistics analysis assumes many measurements with a short exposition time.

3 Why BLESS?

To understand how the target mode shaping and photon statistics measurements can improve the
two point resolution, consider the example presented in Fig. 2. The object consists of two point sources $S_a$ and $S_b$ with coordinates $x_a = \sigma/\sqrt{2}$, $x_b = -\sigma/\sqrt{2}$ and mean emitted photon numbers $\mu_a = 0.1$, $\mu_b = 0.2$. Usual imaging scheme allows one to measure the mean photon number $M_D$ versus the detector position $x_D$ (we assume the Gaussian PSF of the detector with the width $\sigma$). This dependence is presented by the blue dashed line on the left plot. This image looks like a single Gaussian function, which center is shifted to the left, since the left source is brighter, and it is really hard to resolve two sources from this picture. If one transforms the Target Mode to the Hermite-Gaussian mode $HG_1$ and again measures the mean photon number, they obtain an image, presented by the green solid line on the right plot. Here we can see the two peaks and a dip between them. Its depth depends both on the distance $d$ between two sources and on their relative brightness $\gamma$, so if one of these parameters is known a priori, it is easy to estimate another one from this plot, but if both parameters are unknown, it is still difficult to estimate them. However, one can measure the photon number distribution at each position $x_D$ and calculate the second-order correlation function (13) which is presented as a green solid line on both plots. Both $g^{(2)}_D$ plots differ from $M_D$ plots and they can carry some additional information about the object parameters. For the $HG_1$ Target Mode case the improvement is really huge and quite visible. If the detector position $x_D$ exactly equals the position $x_a$, it register no light from the source $S_a$ since $HG_1$ function is asymmetrical and gives zero overlap with the Gaussian PSF (8). So, in this point detector register the light from the only one single-photon source $S_b$ and therefore the $g^{(2)}_D$ function exactly equals zero at this point. By the same reason $g^{(2)}_D$ has a minimum at $x_D = x_b$. This means that correlation function has narrow dips at the positions of single-photon sources, which can be used for their precise localization.

This approach can be generalized to a large number of independent single-photon sources $N$ since even in this case $g^{(N)}_D$ correlation function has narrow minima at the sources locations. Therefore, the photon number distribution measurement (which contain information about all the correlation functions) combined with the asymmetric shaping of the target mode, can provide the subdiffraction photon source localization.

4 Limits on parameters estimation

4.1 Cramér–Rao bound (CRB)

Consider a statistically efficient unbiased estimate $\hat{\theta}$ of four image parameters. For each imaging experiment, $\hat{\theta}$ is a random vector having multivariate normal distribution $f(\hat{\theta})$ centered at point $\theta^*$ of the true parameters values [30, 31]. According to CRB, the covariance matrix $\Sigma$ of $f(\hat{\theta})$ is the inverse of the $4 \times 4$ Fisher information matrix $I$ with elements

$$I_{\alpha\beta} = \sum_k \left[ \frac{\partial_{\alpha} P(k)}{P(k)} \right] \left[ \frac{\partial_{\beta} P(k)}{P(k)} \right] \bigg|_{\theta = \theta^*},$$

where $P$ is the detector photon number distribution (10), and $\partial_{\alpha}$ is its partial derivative with respect to the parameter $\theta_\alpha$ [30, 31]. The values $\Delta_\alpha = \sqrt{I^{-1}_{\alpha\alpha}}$ thus describes the statistical limits of the parameters $\theta_\alpha$ estimation error. Since the Fisher information is additive over independent trials, we define the complete information matrix as $I = \sum_x K_{xD}I_{xD}$. Here we take the sum over various target beam positions $x_D$ with the corresponding sample size $K_{xD}$ and Fisher information matrix $I_{xD}$.

Non-efficient estimators give $\Sigma > I^{-1}$ ($\Sigma - I^{-1}$ is a positive-definite matrix). The bound $\Sigma = I^{-1}$ is usually attainable for the maximum-likelihood estimator (see Appendix A), but the maximization routine becomes slower with decreasing distance $d$. Moreover, we expect
the computation complexity to increase tremendously with the increasing amount of light sources and consequently the number of parameters to estimate. In this regard, it is important to discover efficient methods for solving this optimization problem.

For the integrated statistics we analyze the information carried by the mean photon number $M_D$ in each detector position $x_D$. The Fisher information for estimating $M_D$ is just $K_{x_D}/\sigma_D^2$, so the information matrix for object parameters is

$$I_{a|\beta}^M = \frac{K_{x_D}}{\sigma_D^2(x_D)}[\partial_a M_D(x_D)][\partial_\beta M_D(x_D)].$$

As before, summing up over different $x_D$ gives the complete information matrix. Note that considering the integrated statistics is similar to the widely used weak source approximation [7–20]. Below we will demonstrate that it does not provide enough information in order to break Rayleigh’s curse for unbalanced point sources.

### 4.2 Quantum CRB

Fisher information matrix depends on the particular measurements. However, one might be interested in the ultimate limit over all possible measurements. This could be achieved by computing the quantum Fisher information matrix [7, 14, 32]:

$$I^Q_{a|\beta} = 2 \sum_{k,l;\lambda_k+\lambda_l \neq 0} \frac{\langle \psi_k | \partial_a \rho | \psi_l \rangle \langle \psi_l | \partial_\beta \rho | \psi_k \rangle}{\lambda_k + \lambda_l} \bigg|_{\theta=\theta^*}. \quad (16)$$

Here $\rho$ is the image density matrix, $\rho = \sum_j \lambda_j |\psi_j\rangle \langle \psi_j|$ – its spectral decomposition. The quantum CRB is then the inverse of matrix $I^Q$.

Following the proposed BLESS approach one defines the density matrix $\rho$ of the light in the image plane taking into account both photon number and spatial degrees of freedom. For two single-photon sources one obtains

$$\rho = \psi_0 |0\rangle \langle 0| + w_{1a} |1_a\rangle \langle 1_a| + w_{1b} |1_b\rangle \langle 1_b| + w_2 |2\rangle \langle 2|. \quad (17)$$

Here $w_0 = P_a(0)P_b(0)$ is the probability of vacuum state (0 photons), $w_{1a} = P_a(1)P_b(0)$ ($w_{1b} = P_a(0)P_b(1)$) is the probability of the source $A$ ($B$) to emit a single photon, $w_2 = P_a(1)P_b(1)$ is the probability to get 2 photons from both sources.

The corresponding states are

$$|1_a\rangle = q^\dagger_a |0\rangle, \quad |1_b\rangle = q^\dagger_b |0\rangle,$$

$$|2\rangle = \frac{q^\dagger_a q^\dagger_b}{\sqrt{1 + V^2}} |0\rangle,$$

where

$$q^\dagger_{a,b} = \int \Psi_{a,b}(x) a^\dagger(x) dx \quad (19)$$

are the creation operators for Gaussian modes, $a^\dagger(x)$ is the creation operator for coordinate $x$, $\Psi_a(x)$ and $\Psi_b(x)$ are Gaussian functions, centered at $x_a$ and $x_b$ respectively (see eq. (3)). The overlap integral between two modes is

$$V \equiv \int \Psi_a(x) \Psi_b^*(x) dx = \exp \left( -\frac{d^2}{4\sigma^2} \right). \quad (20)$$

The computation of (16) is a bit tricky since operators $q^\dagger_a$ and $q^\dagger_b$ are not orthogonal (do not commute). We first introduce a set of eight non-orthogonal states that supports Rayleigh’s curse for unbalanced point sources.

### 5 Results and Discussion

We have analyzed lower bounds for the estimation accuracy of source parameters depending on the distance $d$ between two unbalanced point sources and their brightness ratio $\gamma$. Fig. 3 shows the estimation errors (standard deviations) for parameters $d$ (a, c, e), $\gamma$ (b, d, f), $\mu$ (g) and $x_c$ (h). Note that we multiply all $\Delta_\gamma$ by $\sqrt{K}$ ($K = \sum_{x_D} K_{x_D}$) to make them sample size independent. The plots for $\Delta_\mu$ and $\Delta x_c$ almost don’t depend on the value of $\gamma$, so we present them for $\gamma = 0.1$ only, but $\Delta_d(d)$ and $\Delta_\gamma(d)$ dependencies are significantly different for distinct $\gamma$. Below we discuss the main observations from the plots.

#### 5.1 Distance $d$ estimation error

The quantum CRB for the distance error $\Delta_d(Q)$ does not depend on $d$ for all the gamma values, which means that the Rayleigh’s curve can be overcome. We have empirically found the following simple relation:

$$\Delta_d(Q) = \frac{\sqrt{2}\sigma}{\sqrt{\mu - \mu^2/2\sqrt{1 - \gamma^2\sqrt{K}}} \cdot (21)}$$
The relative brightness \( \gamma = 0 \) (a, e), \( \gamma = 0.001 \) (b, f), \( \gamma = 0.1 \) (c, d, g, h). Black dotted line corresponds to the quantum Cramér–Rao bound, other lines – to classical Cramér–Rao bounds with different measurement protocols: dashed lines correspond to Gaussian PSF, solid lines – to HG1-mode PSF, red lines correspond to the mean photon number measurements, blue lines – to the shot statistics analysis. For all the protocols detector position \( x_D/\sigma = -2, -1.9, \ldots, 1.9, 2 \) and the centroid position \( x_c/\sigma = 0.001 \). The proposed BLESS technique corresponds to HG1-mode PSF and the shot statistics analysis (blue solid line). Note that the mesh \( x_D \) does not contain the unknown centroid location \( x_c \), and the error for BLESS technique starts to grow when \( d \lesssim 2x_c \) (gray regions on the plots).

This dependency on \( \gamma \) means that \( d \) can be estimated better for the sources with similar brightness than for significantly unbalanced sources. The dependency of \( \mu \) means that for small mean photon number most of the shots have no photons and so do not carry any information about the source.

For classical CRB the errors look different for different measurement protocols. For two equal point sources with \( \gamma = 0 \) (Fig. 3a) the photon number statistics examination give no benefit: both solid lines (blue and red) and both dashed lines are perfectly matched. However, the target mode transformation significantly helps. For HG0 target mode \( \Delta_d \propto d^{-1} \), which corresponds to the Rayleigh’s curse, presented in [5, 6]. At the same time, for HG1 mode Rayleigh’s curse is dispelled: \( \Delta_d \) saturates at \( d/\sigma \sim 0.1 \) and then remains constant for \( d < 0.1\sigma \) as well as it was presented in [7–9].

Here and bellow, \( \Delta_d(d) \) remains constant while there is a node in the detector position mesh in between two sources positions (i.e. \( x_a > x_D > x_h \) for some \( x_D \)). It means that for high resolution one needs to decrease the scanning step or adjust the detector position adaptively. If this condition is not satisfied, the estimation error \( \Delta_d \) starts to grow, which we can see in Fig. 3(a–c) for the blue solid line in the range \( d/\sigma \lesssim 2 \times 10^{-3} = x_c/\sigma \), selected with gray.

For unbalanced point sources with \( \gamma = 0.1 \) (Fig. 3c) and higher, the \( \Delta_d(d) \) dependencies are different. Measuring just a mean value in each image point can not give a precise estimation accuracy of the distance: for both HG0 and HG1 target modes \( \Delta_d \propto d^{-2} \), which matches the results presented in [17]. However, shot statistics examination allows to limit \( \Delta_d \). For HG0 mode \( \Delta_d\sqrt{K} \sim 10^0 \) for \( d/\sigma < 10^{-2} \) and for HG1 mode \( \Delta_d\sqrt{K} \sim 10 \) for \( d/\sigma < 10^{-1} \). So, for this case the target beam modulation can increase the accuracy by two orders of magnitude, but does not qualitatively change the \( \Delta_d(d) \) dependency.

Note, that in [17] quantum CRB leads to
$\Delta_d \propto d^{-1}$ for unbalanced sources, but for our model, considering photon-number distribution, quantum CRB leads to $\Delta_d = \text{const}$, which means, that full model allows better results even from the fundamental point of view.

In the intermediate case with $\gamma = 0.001$ (Fig. 3b), one can see that the combination of the target beam modulation and the shot statistics examination allows limitation of $\Delta_d(d)$ dependence (beating the Rayleigh’s curse), while all the other measurement protocols demonstrate an error growth as $\Delta_d \propto d^{-1}$ and $\Delta_d \propto d^{-2}$.

Therefore, for all the values of $\gamma$ BLESS protocol limits the $\Delta_d(d)$ dependency which is qualitatively close to quantum CRB $\Delta_d = \text{const}$, but for $d \to 0$ the quantum CRB value is $\sim$ 10 times lower than the best considered classical CRB. It means, that our measurement protocol is still not optimal and can be improved. For example, parallel image acquisition in all the image pixels instead of point-by-point scanning can significantly boost the measurements, but it requires much more complicated equipment.

5.2 Brightness ratio $\gamma$ estimation error

As one can see from (Fig. 3c–g), the brightness ratio estimation error $\Delta_x$, almost does not depend on the choice of the target mode (dashed and solid lines stay very close). Independently on the $\gamma$ value integrated statistics measurement leads to $\Delta_x \propto d^{-3}$. Shot statistics examination allows better precision. For balanced sources with $\gamma = 0$ (Fig. 3e) it leads to $\Delta_x \propto d^{-1}$ and for unbalanced sources with $\gamma = 0.1$ (Fig. 3g) $\Delta_x$ saturates to the constant level for $d/\sigma < 0.1$ for shot statistics examination. For the intermediate case with $\gamma = 0.001$ (Fig. 3f) the error dependency $\Delta_x(d)$ also saturates, but at higher level and for smaller values of $d$. For all the plots presented in Fig. 3(e–g) one can note that quantum CRB curve is close to the classical CRB for shot examination protocols, but a bit lower.

5.3 Integral parameters $\mu$ and $x_c$ estimation errors

The integral source parameters $\mu$ and $x_c$ (Fig. 3d, h) can be well-estimated with all the measurement protocols. For $d < 0.1\sigma$ all CRB for all the protocols as well as quantum CRB lead to $\Delta_\mu(d) = \text{const}$ and $\Delta_{x_c}(d) = \text{const}$. However for the centroid $x_c$, BLESS allow higher accuracy than other protocols.

The increase of $x_c$ estimation error for high values of $d$ (which takes place for all the CRB plots) can be explained with two reasons. First, for higher values of $d$ larger part of the image is not covered by the mesh $x_D/\sigma = -2, -1.9, \ldots, 1.9, 2$. Second, it can be shown that for $d \gg \sigma$ the error $\Delta_{x_c}(d) = d$. By definition

$$x_c = \frac{\mu_a x_a + \mu_b x_b}{\mu_a + \mu_b},$$

For well-separated sources all the parameters $x_a$, $x_b$, $\mu_a$, and $\mu_b$ can be independently estimated with the corresponding errors. The error of the $x_c$ value can be calculated as an indirect measurement error:

$$\Delta^2_{x_c} = \frac{d^2(\mu_a^2 \Delta^2_{x_a} + \mu_b^2 \Delta^2_{x_b})}{\mu_a^2 + \mu_b^2},$$

so, indeed for large $d$ values $\Delta^2_{x_c} \propto d^2$.

5.4 Applicability of BLESS approach

Since our technique requires less prior information about the studying object (in comparison with all the previous methods beating the Rayleigh’s curse [7–9]), it can be used for a wider range of real metrological applications. This includes improving both lateral [10–13] and axial [33, 34] resolution in microscopy, as well as elevating temporal [35] and spectral [36] resolution capabilities.

However, BLESS approach is limited by the localization of single-photon emitters, so its main application can be found in Single Molecule Localization (SML) field aimed to localize dye molecules attached to the biological samples. Typical molecule size $\sim 10$ nm is about 10 times smaller than the confocal microscope PSF width $\sigma \sim 200 – 300$ nm. As follows from (21), for $\mu \sim 10^{-2}$ (which fits with an order of total emitter, detector, and imaging system efficiency) one needs to acquire about $K \sim 10^6$ shots to obtain 10 times resolution enhancement ($\Delta_d \sim \sigma/10$).

According to Fig. 3e, the distance error $\Delta_d$ for BLESS can be 10 times bigger than quantum CRB, so real number of shots can reach $K \sim 10^8$. However, the shot duration should be about a dye molecule lifetime $\tau \sim$ ns, therefore the total acquisition time is less than 1 second, which is

7
suitable for many imaging applications. In contrast with other SML techniques like STED [4], STORM/PALM [26, 27], BLESS technique does not require switchable dyes, so it can be used for a wider range of biological samples.

5.5 Research prospects

Our study is the first step in a comprehensive research program. This program includes a study of the scalability of BLESS with an increase in the number of emitters [14, 37] and an assessment of its robustness against experimental imperfections such as background illumination, detector noise, etc [38–40]. Moreover, our approach can be extended to point sources with different photon statistics, including thermal sources and partially coherent sources [35, 41–43]. The use of adaptive measurement strategies for the localization of point sources can also be promising [44].

6 Conclusion

In conclusion, we have proposed a novel imaging technique. This technique is based on the multi-shot photon number measurements in the modulated target beam that scans the object. Image parameters are then estimated by fitting the photon number distribution model to the collected data. The approach has been theoretically studied on the example of two unbalanced single-photon sources. The Cramér–Rao bound (both classical and quantum) analysis has shown that even for infinitely close sources the estimation error of the distance between them is limited. Thus, we have demonstrated that the form of photon number distribution should be used in the statistical estimation of image parameters since it provides an additional information, and in particular allows one to break Rayleigh’s curse. The proposed method can be used for the single molecule localization and other microscopy applications.

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References

[1] F. R. S. Rayleigh. “Xxxi. investigations in optics, with special reference to the spectroscope”. The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science 8, 261–274 (1879).
[2] Philip R. Hemmer and Todd Zapata. “The universal scaling laws that determine the achievable resolution in different schemes for super-resolution imaging”. Journal of Optics 14, 083002 (2012).
[3] Jeff Squier and Michiel Muller. “High resolution nonlinear microscopy: A review of sources and methods for achieving optimal imaging”. Rev. Sci. Instrum. 72, 2855 (2001).
[4] Tobias Müller, Christian Schumann, and Annette Kraegeloh. “Sted microscopy and its applications: New insights into cellular processes on the nanoscale”. ChemPhysChem 13, 1986–2000 (2012).
[5] S. Van Aert, A.J. den Dekker, D. Van Dyck, and A. van den Bos. “High-resolution electron microscopy and electron tomography: resolution versus precision”. Journal of Structural Biology 138, 21–33 (2002).
[6] E. Bettens, D. Van Dyck, A.J. den Dekker, J. Sijbers, and A. van den Bos. “Model-based two-object resolution from observations having counting statistics”. Ultramicroscopy 77, 37–48 (1999).
[7] Mankei Tsang, Ranjith Nair, and Xiao-Ming Lu. “Quantum theory of superresolution for two incoherent optical point sources”. Physical Review X 6, 031033 (2016).
[8] Ranjith Nair and Mankei Tsang. “Interferometric superlocalization of two incoherent optical point sources”. Optics Express 24, 3684 (2016).
[9] Ranjith Nair and Mankei Tsang. “Far-field superresolution of thermal electromagnetic sources at the quantum limit”. Physical Review Letters 117, 190801 (2016).
[10] Pauline Boucher, Claude Fabre, Guillaume Labroille, and Nicolas Trep. “Spatial optical mode demultiplexing as a practical tool
for optimal transverse distance estimation”. Optica 7, 1621 (2020).

[11] Martin Paúr, Bohumil Stoklasa, Jai Grover, Andrej Krzic, Luis L. Sánchez-Soto, Zdeněk Hradil, and Jaroslav Reháček. “Tempering rayleigh’s curse with psf shaping”. Optica 5, 1177 (2018).

[12] Martin Paúr, Bohumil Stoklasa, Zdenek Hradil, Luis L. Sánchez-Soto, and Jaroslav Rehacek. “Achieving the ultimate optical resolution”. Optica 3, 1144 (2016).

[13] Weng-Kian Tham, Hugo Ferretti, and Aephraim M. Steinberg. “Beating rayleigh’s curse by imaging using phase information”. Physical Review Letters 118, 070801 (2017).

[14] Evangelia Bisketzi, Dominic Branford, and Animesh Datta. “Quantum limits of localisation microscopy”. New Journal of Physics 21, 123032 (2019).

[15] Kent A G Bonsma-Fisher, Weng-Kian Tham, Hugo Ferretti, and Aephraim M. Steinberg. “Realistic sub-rayleigh imaging with phase-sensitive measurements”. New Journal of Physics 21, 093010 (2019).

[16] Lijun Peng and Xiao Ming Lu. “Generalization of rayleigh’s criterion on parameter estimation with incoherent sources”. Physical Review A 103, 1–10 (2021).

[17] J. Řehaček, Z. Hradil, B. Stoklasa, M. Paúr, J. Grover, A. Krzic, and L. L. Sánchez-Soto. “Multiparameter quantum metrology of incoherent point sources: Towards realistic superresolution”. Physical Review A 96, 062107 (2017).

[18] Mankei Tsang. “Subdiffraction incoherent optical imaging via spatial-mode demultiplexing: Semiclassical treatment”. Physical Review A 97, 023830 (2018).

[19] Mankei Tsang. “Quantum limit to subdiffraction incoherent optical imaging”. Physical Review A 99, 1–12 (2019).

[20] Sisi Zhou and Liang Jiang. “Modern description of rayleigh’s criterion”. Physical Review A 99, 1–20 (2019).

[21] R. Hanbury Brown and R. Q. Twiss. “A test of a new type of stellar interferometer on sirius”. Nature 178, 1046–1048 (1956).

[22] T Dertinger, R Colyer, G Iyer, S Weiss, and J Enderlein. “Fast, background-free, 3d super-resolution optical fluctuation imaging (sofi)”. Proceedings of the National Academy of Sciences 106, 22287–22292 (2009).

[23] D. Gatto Monticone, K. Katamadze, P. Traina, E. Moreva, J. Forneris, I. Ruoberchera, P. Olivero, I. P. Degiovanni, G. Brida, and M. Genovese. “Beating the abbe diffraction limit in confocal microscopy via nonclassical photon statistics”. Physical Review Letters 113, 143602 (2014).

[24] O Schwartz and D Oron. “Improved resolution in fluorescence microscopy using quantum correlations”. Phys. Rev. A 85, 33812 (2012).

[25] Osip Schwartz, Jonathan M Levitt, Ron Tenne, Stella Itzhakov, Zvicka Deutsch, and Dan Oron. “Superresolution microscopy with quantum emitters.”. Nano Lett. 13, 5832–5836 (2013).

[26] Michael J. Rust, Mark Bates, and Xiaowei Zhuang. “Sub-diffraction-limit imaging by stochastic optical reconstruction microscopy (storm)”. Nature Methods 3, 793–796 (2006).

[27] Eric Betzig, George H. Patterson, Rachid Sougrat, O. Wolf Lindwasser, Scott Olenych, Juan S. Bonifacino, Michael W. Davidson, Jennifer Lippincott-Schwartz, and Harald F. Hess. “Imaging intracellular fluorescent proteins at nanometer resolution”. Science 313, 1642–1645 (2006).

[28] A. B. Mikhalychev, B. Bessire, I. L. Karuseichyk, A. A. Sakovich, M. Unternährer, D. A. Lyakhov, D. L. Michels, A. Stefanov, and D. Mogilevtsev. “Efficiently reconstructing compound objects by quantum imaging with higher-order correlation functions”. Communications Physics 2, 134 (2019).

[29] Yunkai Wang, Yujie Zhang, and Virginia O. Lorenz. “Superresolution in interferometric imaging of strong thermal sources”. Physical Review A 104, 022613 (2021).

[30] Harald Cramér. “Mathematical methods of statistics”. Princeton University Press. (1999).

[31] M. G. Kendall and A. Stuart. “The advanced theory of statistics, vol. 2: Inference and relationship”. Charles Griffin and Company Ltd. (1961).

[32] Jing Liu, Haidong Yuan, Xiao-Ming Lu, and Xiaoguang Wang. “Quantum fisher information matrix and multiparameter estima-
[33] D. Koutný, Z. Hradil, J. Řeháček, and L. L. Sánchez-Soto. “Axial superlocalization with vortex beams”. Quantum Science and Technology 6 (2021).

[34] Yiyu Zhou, Jing Yang, Jeremy D. Hasseit, Seyed Mohammad Hashemi Rafsanjani, Mohammad Mirhosseini, A. Nick Vamvakas, Andrew N. Jordan, Zhimin Shi, and Robert W. Boyd. “Quantum-limited estimation of the axial separation of two incoherent point sources”. Optica 6, 534 (2019).

[35] Syamsundar De, Jano Gil-Lopez, Benjamin Brecht, Christine Silberhorn, Luis L. Sánchez-Soto, Zdeněk Hradil, and Jaroslav Řeháček. “Effects of coherence on temporal resolution”. Physical Review Research 3, 033082 (2021).

[36] M. Paúr, B. Stoklasa, D. Koutný, J. Řeháček, Z. Hradil, J. Grover, A. Krzic, and L. L. Sánchez-Soto. “Reading out fisher information from the zeros of the point spread function”. Optics Letters 44, 3114 (2019).

[37] Cosmo Lupo, Zixin Huang, and Pieter Kok. “Quantum limits to incoherent imaging are achieved by linear interferometry”. Physical Review Letters 124, 80503 (2020).

[38] Cosmo Lupo. “Subwavelength quantum imaging with noisy detectors”. Physical Review A 101, 22323 (2020).

[39] Giacomo Sorelli, Manuel Gessner, Mattia Walschaers, and Nicolas Treps. “Optimal observables and estimators for practical super-resolution imaging”. Physical Review Letters 127, 123604 (2021).

[40] Giacomo Sorelli, Manuel Gessner, Mattia Walschaers, and Nicolas Treps. “Moment-based superresolution: Formalism and applications”. Physical Review A 104, 033515 (2021).

[41] Stanislaw Kurdzialek. “Back to sources – the role of coherence in super-resolution imaging revisited” (2021). url: http://arxiv.org/abs/2103.12096.

[42] Mankei Tsang and Ranjith Nair. “Resurgence of rayleigh’s curse in the presence of partial coherence: comment”. Optica 6, 400 (2019).

[43] S.A.Wadood, Kevin Liang, Yiyu Zhou, Jing Yang, M. A. Alonso, X.-F. Qian, T. Malhotra, S. M. Hashemi Rafsanjani, Andrew N. Jordan, Robert W. Boyd, and et al. “Experimental demonstration of superresolution of partially coherent light sources using parity sorting”. Optics Express 29, 22034 (2021).

[44] Erik F. Matlin and Lucas J. Zipp. “Adaptive imaging of arbitrary thermal source distributions with near quantum-limited resolution” (2021). url: http://arxiv.org/abs/2106.13332.

[45] A. Yu. Chernyavskiy. “Calculation of quantum discord and entanglement measures using the random mutations optimization algorithm” (2013). arXiv:1304.3703.

[46] A Yu Chernyavskiy. “Random mutations global optimization solver for python”. https://github.com/a-chernyavskiy/RandomMutations (2023).
where the creation operators for HG source location:

Using this definition one can compute the derivatives of creation operators

Figure 4: Distributions of parameters estimation errors for two single-photon emitters. Simulation parameters: HG target mode; \( x_D = \{-2\sigma, -1.9\sigma, \ldots, 1.9\sigma, 2\sigma\} \); total sample size \( K = 10^3 \); object parameters \( d = 0.1\sigma, \gamma = 0.1, \mu = 0.1, x_c = 0.001\sigma \). Histograms are the estimates from 100 independent numerical experiments. Solid curves show marginal distributions of \( f(\hat{\theta}) \) computed using CRB.

A Attainability of estimation limits

Let us demonstrate that the limits imposed by Cramér–Rao bound (CRB) are attainable by practical methods. We have considered the maximum likelihood estimator (MLE). It is asymptotically efficient, which means the convergence to multivariate normal distribution \( f(\hat{\theta}) \) with \( K \to \infty \). Fig. 4 shows the results of 100 numerical experiments and the corresponding theoretical marginal distributions of \( f(\hat{\theta}) \) calculated via CRB. The image source consisted of two single-photon emitters with parameters \( d = 0.1\sigma, \gamma = 0.1, \mu = 0.1, x_c = 0.001\sigma \). The measurement simulation is performed according to the BLESS protocol (target mode positions are \( x_D = \{-2\sigma, -1.9\sigma, \ldots, 1.9\sigma, 2\sigma\} \)). Since the likelihood function is non-convex we have used the Python implementation of global optimization algorithm based on random mutations [45, 46]. We have used the default algorithm parameters.

Similar plots for other model parameters have also shown a close correspondence between MLE estimates and theoretical distributions.

B Computing QFI

The derivation of (16) is complicated since the infinite number of spatial modes, each containing up to 2 photons. However, one can reduce the task by finding the minimal orthogonal basis that supports \( \rho \) and its derivatives (e.g. see Appendix C in [7]). In this section we describe the numerical procedure for the case of two single photon sources. We use the following definition of HG functions:

\[
\Psi_0(x) = \sqrt{\frac{1}{\sigma \sqrt{\pi}}} \exp\left(-\frac{x^2}{2\sigma^2}\right), \quad \Psi_1(x) = \sqrt{\frac{2}{\sigma \sqrt{\pi}}} \left(\frac{x}{\sigma}\right) \exp\left(-\frac{x^2}{2\sigma^2}\right).
\]  

(24)

Using this definition one can compute the derivatives of creation operators (19) with respect to the source location:

\[
\frac{\partial q_a^\dagger}{\partial x_a} = \frac{f_a^\dagger}{\sqrt{2\sigma}}, \quad \frac{\partial q_b^\dagger}{\partial x_b} = \frac{f_b^\dagger}{\sqrt{2\sigma}}, \quad \frac{\partial q_a^\dagger}{\partial x_b} = \frac{\partial q_b^\dagger}{\partial x_a} = 0,
\]  

(25)

where the creation operators for HG modes are introduced:

\[
f_a^\dagger = \int \Psi_1(x-x_a) a^\dagger(x) dx, \quad f_b^\dagger = \int \Psi_1(x-x_b) a^\dagger(x) dx.
\]  

(26)

Using the chain rule one computes the derivatives of the states (18):

\[
\frac{\partial_a}{\partial_a} |1_a\rangle = \frac{\partial_a x_a}{\sqrt{2\sigma}} f_a^\dagger |0\rangle, \quad \frac{\partial_a}{\partial_a} |1_b\rangle = \frac{\partial_a x_b}{\sqrt{2\sigma}} f_b^\dagger |0\rangle,
\]

\[
\frac{\partial_a}{\partial_a} |2\rangle = \frac{1}{\sqrt{2\sigma}} \left( \frac{(\partial_a x_a) f_a^\dagger q_b^\dagger + (\partial_a x_b) q_a^\dagger f_b^\dagger}{\sqrt{1+V^2}} |0\rangle - \frac{V^2}{1+V^2} \frac{d}{\sqrt{2\sigma}} (\partial_a d) |2\rangle \right),
\]  

(27)
where $\partial_\alpha$ is the partial derivative with respect to the parameter $\theta_\alpha$. The derivative of the density matrix (17) is

$$
\partial_\alpha \rho = (\partial_\alpha w_0) |0\rangle\langle 0| + (\partial_\alpha w_1) (|1_a\rangle\langle 1_a| + w_{1a}[\langle \partial_\alpha |1_a\rangle |1_a\rangle + h.c.]) + (\partial_\alpha w_2) |2\rangle\langle 2| + w_2[(\partial_\alpha |2\rangle \langle 2| + h.c.)].
$$

(28)

One can observe that the initial density matrix (17) and its derivatives are supported by the following set of eight states:

$$
|\varphi_0\rangle = |0\rangle, \\
|\varphi_1\rangle = q_a^\dagger |0\rangle, \\
|\varphi_2\rangle = q_b^\dagger |0\rangle, \\
|\varphi_3\rangle = f_a^\dagger |0\rangle, \\
|\varphi_4\rangle = f_b^\dagger |0\rangle, \\
|\varphi_5\rangle = q_a^\dagger q_b^\dagger |0\rangle, \\
|\varphi_6\rangle = q_a^\dagger f_b^\dagger |0\rangle, \\
|\varphi_7\rangle = f_a^\dagger q_b^\dagger |0\rangle.
$$

(29)

Let us now introduce an orthonormal basis \{|$z_j\rangle$\} such that

$$
|\varphi_n\rangle = \sum_{j=0}^7 c_{jn} |z_j\rangle.
$$

(30)

To get coefficients $c_{kn}$ of the matrix $c$ one has to compute all pair-wise scalar products $K_{mn} = \langle \varphi_m | \varphi_n \rangle$. In our case the matrix $K$ has the following form:

$$
K = \begin{pmatrix}
1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 1 & V_{00} & 0 & V_{01} & 0 & 0 & 0 \\
0 & V_{00} & 1 & V_{10} & 0 & 0 & 0 & 0 \\
0 & 0 & V_{10} & 1 & V_{11} & 0 & 0 & 0 \\
0 & V_{01} & 0 & V_{11} & 1 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 + V_{00}^2 & V_{00}V_{01} & V_{00}V_{10} & 0 \\
0 & 0 & 0 & 0 & V_{00}V_{01} & 1 + V_{01}^2 & V_{00}V_{11} & 0 \\
0 & 0 & 0 & 0 & V_{00}V_{10} & V_{00}V_{11} & 1 + V_{10}^2 & 0
\end{pmatrix},
$$

(31)

where

$$
V_{nm} = \int \Psi_n(x - x_a)\Psi_m(x - x_b) dx.
$$

(32)

In particular,

$$
V_{00} = V, \quad V_{01} = -V_{10} = \frac{d}{\sqrt{2}\sigma} V, \quad V_{11} = \left(1 - \frac{d^2}{2\sigma^2}\right) V.
$$

(33)

The orthonormality of the basis \{|$z_j\rangle$\} gives the following equation:

$$
K_{mn} = \langle \varphi_m | \varphi_n \rangle = \sum_{jk} c_{km}^* c_{jn} \langle z_k | z_j \rangle = [c^\dagger c]_{mn}.
$$

(34)

Matrix $c$ is then found by the Cholesky decomposition of matrix $K$. After that, one can express $\rho$ and $\partial_\alpha \rho$ for any $\alpha$ via \{|$z_j\rangle$\} and compute QFI numerically.