Mueller, J. D., Samantaray, N., & Matthews, J. C. F. (2020). A practical model of twin-beam experiments for sub-shot-noise absorption measurements. *Applied Physics Letters*, *117*, [034001]. https://doi.org/10.1063/5.0015576

Supplementary material: A practical model of twin-beam experiments for sub-shot-noise absorption measurements

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(Dated: 8 July 2020)

Appendix A: Comparison of twin-beam absorption estimators

Estimators are mathematical formulas applied to finite data sets for determining physical parameters of a system. One such parameter used to characterize *e.g.* biological samples is spectral absorption $\alpha(\lambda)$. Typically, for a given wavelength λ, measuring sample absorption involves comparing the intensity of a light source with and without a sample in its path:

$$\alpha_c = 1 - \frac{n_1'}{\mathrm{E}[n_1]}, \tag{S1}$$

where $0 \leq \alpha_c \leq 1$ is the direct classical absorption estimator, n_1 is the probe beam intensity for each measurement trial, and the prime denotes beam intensity after a lossy interaction with the sample. For the remainder of these discussions, α without a subscript refers to the population estimate (unbiased estimate based on an infinitely-large data set), and with refers to sample estimates (realistic finite-sized data sets).

The precision of this absorption measurement is limited by the Poissonian quantum nature of light, the Shot-Noise Limit (SNL), as

$$\mathrm{Var}[\alpha_c] = \frac{(1-\alpha)}{\mathrm{E}[n_1]}. \tag{S2}$$

A first example of a twin-beam absorption estimator for quantum parameter estimation was presented in Ref. 1 and further explored by Ref. 2:

$$\alpha_l = 1 - \gamma\frac{n_1'}{n_2'}, \tag{S3}$$

where n_2 is the reference beam intensity, and $\gamma = \mathrm{E}[n_2]/\mathrm{E}[n_1]$ accounts for unbalanced channel efficiency. Primes in this case denote the measurement stage in general, and the sample is only placed in the path of the probe beam n_1.

In the case of balanced channel efficiency ($\gamma = 1$) and no optical or detector noise, one may write

$$\mathrm{Var}[\alpha_l] = \mathrm{Var}[\alpha_u] + 2\frac{(1-\alpha)^2}{\mathrm{E}[n_1]}\sigma^*, \tag{S4}$$

where $\mathrm{Var}[\alpha_u] = \alpha\mathrm{Var}[\alpha_c]$ is the ultimate quantum limit of an absorption measurement, associated with binomial measurement statistics, attainable with *e.g.* Fock states or when $\sigma = 0$[2,3], and $\sigma^* = 1-\eta$ is the noiseless, balanced-detection Noise-Reduction Factor (NRF). To compare this twin-beam estimator to the classical direct case, we use their relative estimator efficiency

$$\Gamma_i = \frac{\mathrm{MSE}[\alpha_i]}{\mathrm{MSE}[\alpha_c]} \tag{S5a}$$

$$= \frac{\mathrm{Var}[\alpha_i] + (\mathrm{E}[\alpha_i] - \alpha)^2}{\mathrm{Var}[\alpha_c]}, \tag{S5b}$$

for some estimator i, where $\mathrm{MSE}[\alpha_i]$ is the mean squared error, which equals $\mathrm{Var}[\alpha_i]$ in the case of unbiased parameter estimation (as implicitly assumed in Refs. 2 and 4). When $0 \leq \Gamma_i < 1$, the estimator efficiency is sub-SNL. This regime is exclusive to quantum-correlated twin beams, similar to $0 \leq \sigma < 1$.

Comparing Eqs. S2 and S4 yields

$$\Gamma_l = \alpha + 2(1-\alpha)\sigma^*. \tag{S6}$$

One finds $\Gamma_l > 1$ for all $\sigma^* > 0.5$. Thus, even though beams may display sub-Poissonian intensity correlations, one cannot always perform sub-SNL absorption measurements with this estimator. One can gain insight into this counter-intuitive result by considering how α_c is an even less suitable estimator for the twin-beam case, as $\Gamma_c = 1$ for all values of σ.

Ref. 4 presents another twin-beam absorption estimator:

$$\alpha_m = 1 - \frac{n_1' - k\delta n_2' + \delta E}{\mathrm{E}[n_1]}, \tag{S7}$$

where $\delta n_2' = n_2' - \mathrm{E}[n_2']$, k is a weight factor used to maximize the estimator's precision, and $\delta E = \mathrm{E}[k\delta n_2']$ is a correction factor used to ensure that the estimator is unbiased (*i.e.* $\mathrm{E}[\alpha_m] = \alpha$). Contrary to Refs. 2 and 4, α_m is indeed biased in the presence of classical intensity fluctuations, as we demonstrate at the end of this section. We also correct the estimator to be unbiased.

One may perform a similar analysis as the previous estimator, now with[2]

$$\mathrm{Var}[\alpha_m] = \mathrm{Var}[\alpha_u] + 2\frac{(1-\alpha)^2}{\mathrm{E}[n_1']}\sigma^*(1 - \frac{\sigma^*}{2}), \tag{S8}$$

in the noiseless, balanced-detection case with optimized k[4]:

$$k_m^{\mathrm{opt}} = \frac{\mathrm{Cov}[n_1', n_2']}{\mathrm{Var}[n_2']}. \tag{S9}$$

Comparing this to the classical direct measurement with $\gamma = 1$,

$$\Gamma_m = \alpha + 2(1-\alpha)\sigma^*(1 - \frac{\sigma^*}{2}). \tag{S10}$$

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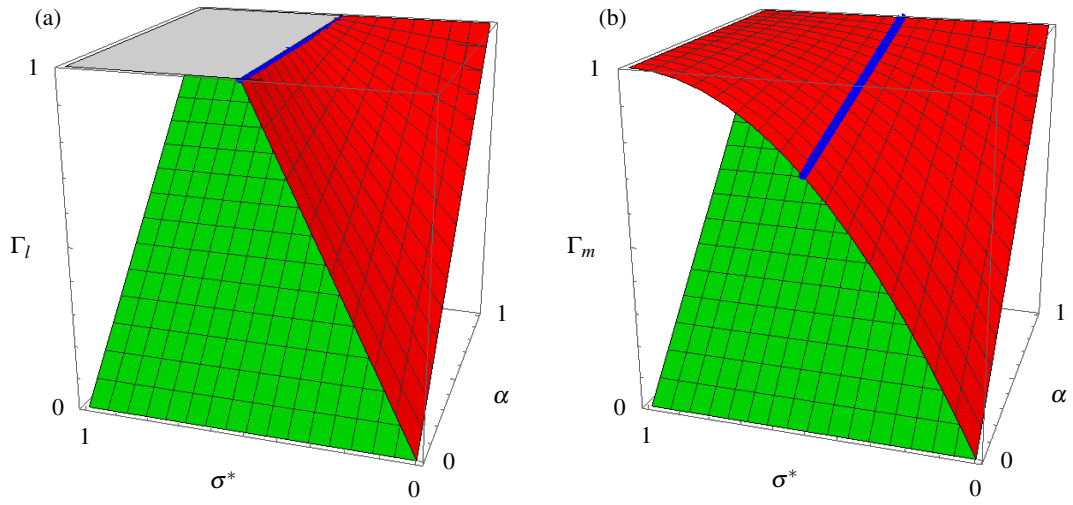

FIG. S1. Comparing relative SNL performance metrics (a) Γ_l and (b) Γ_m in the case of balanced channel efficiency and no optical or detector noise. The green plane $\Gamma_u = \alpha$ is the ultimate quantum limit, and the blue line is the $\sigma^* = 0.5$ contour.

We now find sub-SNL Γ_m for all $\sigma^* < 1$, and $\Gamma_m < \Gamma_l$ for all $\sigma^* > 0$ and $\alpha < 1$. The performance of α_m and α_l is compared graphically in Fig. S1. We see in this figure that α_m is a superior estimator to α_l when appropriately calibrated. In the case discussed here, one achieves sub-SNL measurement statistics for any values of $\eta_{\{1,2\}} > 0$ using α_m, relaxing the requirement that $\eta_{\{1,2\}} > 0.5$ when using α_l, stated in Ref. 1.

Although we do not derive it here, we expect from our discussions of the NRF in the main text that super-Poissonian intensity noise with unbalanced channel efficiency and other uncorrelated noise sources further reduce the efficacy of α_l and α_m for achieving sub-SNL measurement statistics.

We will also show that twin-beam estimators are not only more precise than the direct classical absorption estimator, but also more accurate in general.

For stationary processes (processes whose mean and variance do not change with time), α_c is indeed unbiased, as $\mathrm{E}[n_1'] = (1-\alpha)\mathrm{E}[n_1]$, and $\mathrm{E}[\alpha_c] = \alpha$. For non-stationary processes, however, the probe and reference beam powers are changed by an amount $\varepsilon \geq -1$:

$$\mathrm{E}\left[n_1'\right] = (1-\alpha)(1+\varepsilon)\mathrm{E}[n_1] \tag{S11}$$

$$\mathrm{E}\left[n_2'\right] = (1+\varepsilon)\mathrm{E}[n_2]\,. \tag{S12}$$

This may occur experimentally if the probe beam power is changed between the calibration and measurement phases. Because α_c does not have access to the reference beam, substitution of Eq. S11 into Eq. S1 yields

$$\mathrm{E}[\alpha_c] = 1-(1-\alpha)(1+\varepsilon), \tag{S13}$$

which is biased without knowledge of ε. Simply, the direct classical absorption estimator cannot distinguish probe beam intensity fluctuations from sample absorption.

Considering now the twin-beam estimator α_l, we may substitute Eqs. S11 and S12, yielding

$$\mathrm{E}[\alpha_l] = 1-\gamma\mathrm{E}\left[\frac{n_1'}{n_2'}\right] \tag{S14a}$$

$$\approx 1-\frac{\mathrm{E}[n_2]}{\mathrm{E}[n_1]}\frac{\mathrm{E}[n_1']}{\mathrm{E}[n_2']} \tag{S14b}$$

$$= \alpha, \tag{S14c}$$

where the approximation in line two is valid for large n[1,5]. This estimator is therefore unbiased in the large-photon-flux limit, which is the regime where intensity-correlated measurements are most practical.

Finally, we consider the absorption estimator α_m, which we previously showed to obtain the greatest measurement precision of the three discussed estimators. The form of this estimator, as originally presented in Ref. 4 and discussed further in Ref. 2, is biased, obtaining the same functional form for $\mathrm{E}[\alpha_m]$ as Eq. S13:

$$\mathrm{E}[\alpha_m] \approx 1-\frac{\mathrm{E}[n_1']-\mathrm{E}[k\delta n_2']+\delta E}{\mathrm{E}[n_1]} \tag{S15a}$$

$$= 1-\frac{\mathrm{E}[n_1']}{\mathrm{E}[n_1]} \tag{S15b}$$

$$= 1-(1-\alpha)(1+\varepsilon). \tag{S15c}$$

This is because α_m is derived from α_c, which implicitly requires a stationary twin-beam intensity to be unbiased. We present here a new, unbiased form of α_m, denoted α_{lm}, using α_l as the starting point:

$$\alpha_{lm} = 1-\gamma\frac{n_1'-k\delta n_2'+\delta E}{n_2'}. \tag{S16}$$

This estimator is unbiased for optimized k, as $\mathrm{E}[\alpha_{lm}] = \mathrm{E}[\alpha_l] = \alpha$.

The k which maximizes the precision of α_{lm} is found by minimizing $\mathrm{Var}[(n_1' - k\delta n_2')/n_2']$. This variance may be approximated according to Ref. 5, yielding

$$k_{lm}^{\mathrm{opt}} \approx k_m^{\mathrm{opt}} - \frac{\mathrm{E}[n_1']}{\mathrm{E}[n_2']}. \tag{S17}$$

Appendix B: Noise-reduction factor with uncorrelated noise on both detection channels

We derived the NRF Eqs. 10a–10d for the case of optical noise on only one detection channel and balanced detector noise on both channels, for simplicity. These equations may be generalized to include uncorrelated optical and detection noise on each channel following the same procedure outlined in the main text, with the following result:

$$\sigma_p = 1 - \frac{2\eta_1\eta_2}{(1+\rho_1)\eta_1 + (1+\rho_2)\eta_2 + d_1 + d_2} \tag{S18a}$$

$$\sigma_{sp} = \frac{(\eta_1 - \eta_2)^2(F-1)}{(1+\rho_1)\eta_1 + (1+\rho_2)\eta_2 + d_1 + d_2} \tag{S18b}$$

$$\sigma_\rho = \frac{\eta_1^2\rho_1(F_{\rho_1}-1) + \eta_2^2\rho_2(F_{\rho_2}-1)}{(1+\rho_1)\eta_1 + (1+\rho_2)\eta_2 + d_1 + d_2} \tag{S18c}$$

$$\sigma_d = \frac{d_1(F_{d_1}-1) + d_2(F_{d_2}-1)}{(1+\rho_1)\eta_1 + (1+\rho_2)\eta_2 + d_1 + d_2}, \tag{S18d}$$

where $N_{\{1,2\}} \to N_{\{1,2\}} + N_{\rho_{\{1,2\}}} + N_{d_{\{1,2\}}}$. Setting $\rho_1 = 0$, $d_1 = d_2$, and $F_{d_1} = F_{d_2}$ yields the derived Eqs. 10a–10d.

Appendix C: Details of noise-reduction factor simulation for experimental model

The simulations shown in Fig. 2 were performed according to the following procedure.

We first define the mean and variance the distributions N, N_ρ, and N_d from which the signal counts and optical and detector noise counts are sampled. These distributions are Gaussian for large mean values, where the degree to which they are super-Poissonian can be set by the relative values of their means and variances. We also define the number of trials t for the data to be averaged over, as well as channel detection efficiency η_1.

For each count source (twin beams, optical noise, and detector noise), an integer list of length t is generated, with each element sampled from its corresponding distribution. This represents the number of pre-loss photons or detector dark counts, for each measurement trial.

A loop is performed over η_2 from 0 to 1. Within this loop, a loop over t is performed, where for each trial and each count source, a list of pseudo-random numbers between 0 to 1, inclusive, is generated whose length is given according to the the specified element from the previous step. To determine if the photon is detected as a count, these pseudo-random numbers are compared to the correspondingly defined channel efficiency, and replaced with a one if the pseudo-random number is less than $\eta_{\{1,2\}}$, zero otherwise (detector noise counts, independent of detector efficiency, do not undergo this comparison). The list is then summed and stored as the number of detected counts for that trial. In this way, we can simulate the random loss associated with the photon-count sources.

Finally, the signal and noise counts are summed for each channel, the NRF is calculated for the specified η_2, and η_2 is incremented.

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