



Supporting Online Material for

**Probing Quantum Commutation by Addition and Subtraction of
Single Photons to/from a Light Field**

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Direct Probing of Quantum Commutation Rules by Addition and Subtraction of Single Photons to/from a Light Field

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Materials and methods

Creation and annihilation operators and the wave-particle duality

In order to derive Planck's formula for blackbody radiation, it was hypothesized (*S1*) that photons are identical particles and any number of photons can occupy the same quantum state, which leads to the important conclusion that photons take symmetrical states. The proportionality factors \sqrt{n} and $\sqrt{n+1}$ are a direct consequence of this symmetric nature of photon states (*S2*) and indicate that the probability of subtracting a photon from a state is proportional to the number of photons originally in that state, while the probability of adding a photon contains an additional term due to spontaneous emission. This shows that photon (in a general term, boson) annihilation and creation operators have exactly the same mathematical structures as the ladder operators that appear in the quantum harmonic oscillator, which bears a clear wave-like nature. Dirac described this equivalence as "one of the most fundamental results of quantum mechanics, which enables a unification of the wave and corpuscular theories of light to be effected" (*S2*).

The apparent paradox of photon subtraction

It is particularly instructive to see what happens when applying the annihilation operator \hat{a} to an arbitrary quantum light state. As already shown in the context of photodetection

theory (S3,S4), one can easily find that the average number of photons in the “photon-subtracted” state is related to the initial one as:

$$\bar{n}_{sub} = \bar{n} - 1 + F$$

where F is the Fano factor (related to the Mandel q parameter (S5) by $q = F - 1$) of the initial state, and given by

$$F \equiv \frac{\overline{(\Delta n)^2}}{\bar{n}},$$

($\overline{(\Delta n)^2}$ being the photon number variance) which is greater, equal to, or less than unity for Super-Poissonian, Poissonian, or Sub-Poissonian initial states, respectively. So, depending on the character of the initial state, the mean photon number in the final state can be less than, equal to, or even larger than before “photon subtraction” and, unless the initial state is a coherent one, the whole statistics of the field is changed by this process.

The reason for this behavior stems from the \sqrt{n} (or $\sqrt{n+1}$) factor in Eqs. (1) and (2) of the paper which, besides shifting the photon distribution to the left (or right), favors the action of the operators upon the higher excited states, so that it may effectively increase the relative weight of higher-energy components in the final state (this, in fact, plays an important role in quantum-state engineering). Indeed, the successful event of particle subtraction by \hat{a} , just preferentially selects the subset of states having a larger probability of giving a particle away, i.e., those containing a larger number of photons. It should be stressed that this is not a quantum effect, since a similar increase in the mean photon number may also be expected in a classical case if particle subtraction is performed in a probabilistic way.

In certain cases, photon subtraction by \hat{a} may even be more effective than photon addition by \hat{a}^\dagger in increasing the mean number of quanta in a field (S4). After subtracting a photon from a thermal field, the average photon number grows from \bar{n} to $2\bar{n}$. The operation of photon addition increases the mean photon number of the thermal field even further to $2\bar{n} + 1$, while it scales as $\frac{1 + 6\bar{n} + 6\bar{n}^2}{1 + 2\bar{n}}$ and as $\frac{2\bar{n}(2 + 3\bar{n})}{1 + 2\bar{n}}$ for the subtracted-then-added and the added-then-subtracted sequences, respectively. Note that the expected difference in

the mean photon number is exactly 1 both between the added and the subtracted thermal states, and between the states resulting from the two alternated sequences of operations; our experimental findings are in complete agreement with such expectations.

Experimental realization of a thermal light source

The photon number distribution for a thermal light state exhibits a more or less rapid exponential decay when going to higher photon numbers depending on the temperature of the light source. The higher the temperature, the broader the distribution and the larger the mean number of photons in the field. Since the generation of a true thermal source with a non-negligible number of photons per mode in the spectral region around 800 nm is a complicated task due to the impractically-high temperatures involved, we have used a pseudo-thermal light source obtained by inserting a rotating ground glass disk (RD) in the path of the laser beam. By coupling a fraction (much smaller than the typical speckle size) of the randomly scattered light into a single-mode fiber, the radiation at the fiber output is in a clean spatial mode with random amplitude and phase, yielding the photon distribution typical of a thermal source (S6).

Experimental implementation of photon addition/subtraction and of their sequences

Experimentally, a single photon subtraction is performed by detecting a photon after splitting the field by a beam splitter of high transmittivity. The field of density operator $\hat{\rho}^{(0)}$ in mode ‘a’ and a vacuum $|0\rangle\langle 0|$ in mode ‘b’ are two inputs to the beam splitter. After the output field mode ‘b’ is measured by a conventional on-off photodetector, described by the operator $M = \mathbf{1} - |0\rangle\langle 0|$, which discriminates there being photons but is not able to discern how many photons there are, output mode ‘a’ collapses into

$$\hat{\rho}_a^{(s)} = N \text{Tr}_b \left(M_b \hat{B} \hat{\rho}_a^{(0)} |0\rangle_b \langle 0| \hat{B}^\dagger \right) \quad ; \quad \hat{B} = \exp(\theta(\hat{a}^\dagger \hat{b} - \hat{a} \hat{b}^\dagger)), \quad (\text{S1})$$

where \hat{B} is the beam splitter operator with transmittivity $T = \cos^2(\theta)$. Throughout this Section, N denotes a normalization constant for the respective state.

On the other hand, a single photon addition is done using a parametric down-converter, which generates twin photons into two modes ‘a’ and ‘b’. By detecting a photon in one

mode (say, ‘**b**’), it is certain that the twin photon is in the other mode ‘**a**’. This process converts field $\hat{\rho}^{(0)}$ in mode ‘**a**’ into

$$\hat{\rho}_a^{(a)} = \text{NTr}_b \left(M_b \hat{U} \hat{\rho}_a^{(0)} |0\rangle_b \langle 0| \hat{U}^\dagger \right) \quad ; \quad \hat{U} = \exp(\lambda(\hat{a}\hat{b} - \hat{a}^\dagger\hat{b}^\dagger)), \quad (\text{S2})$$

where \hat{U} is the parametric down-conversion operator whose gain factor is λ . Again, a vacuum has been assumed in input mode ‘**b**’. We can easily extend these analyses to get the density operator $\hat{\rho}^{(sa)}$ corresponding to the application of the sequence of experimental apparatuses for photon addition-then-subtraction, and $\hat{\rho}^{(as)}$ corresponding to the other sequence.

Ideally, a sequence of a single photon addition followed by a single photon subtraction, should turn the initial thermal state, whose density operator is diagonal as

$$\hat{\rho}_{th} = \sum_{n=0}^{\infty} P(n) |n\rangle \langle n| \quad ; \quad P(n) = \frac{\bar{n}^n}{(1+\bar{n})^{1+n}}, \quad (\text{S3})$$

into the state of density operator, $\hat{\sigma}^{(sa)} = \text{N} \hat{a} \hat{a}^\dagger \hat{\rho}_{th} \hat{a} \hat{a}^\dagger$. On the other hand, with the reverse sequence of a single photon addition after subtracting a photon, the initial thermal state becomes the state of density operator, $\hat{\sigma}^{(as)} = \text{N} \hat{a}^\dagger \hat{a} \hat{\rho}_{th} \hat{a}^\dagger \hat{a}$.

After a straightforward calculation, we can find that the density operators for both the ideal and the experimentally realizable states are diagonal. In order to measure how close the experimentally realizable state $\hat{\rho}$ is to the ideal state $\hat{\sigma}$, we calculate the fidelity defined as

$$F = |f|^2 \quad \text{with} \quad f = \text{Tr} \sqrt{\sqrt{\sigma} \rho \sqrt{\sigma}}.$$

For two diagonal matrices (S7), $\hat{\rho} = \sum_i \rho_i |i\rangle \langle i|$ and $\hat{\sigma} = \sum_i \sigma_i |i\rangle \langle i|$, $F = \sum_i \sigma_i \rho_i$. Now, for the experimental values of $T = 0.99$, $\lambda = 0.01$ and the average photon number, $\bar{n} \approx 0.6$ and 1, the fidelities are simply calculated and give above 98% for both $\hat{\rho}^{(as)}$ vs. $\hat{\sigma}^{(as)}$ and $\hat{\rho}^{(sa)}$ vs. $\hat{\sigma}^{(sa)}$.

As the average photon number grows, the experimental realization begins to differ from the ideal state. This can be analytically seen by the Taylor expansion of \hat{B} and \hat{U} in $\hat{\rho}^{(s)}$ and

$\hat{\rho}^{(a)}$ of Eqs.(S1) and (S2). Under the condition $\lambda, \theta \ll 1$, the Taylor expansions are truncated to $O(\theta^5)$ and $O(\lambda^5)$ to give

$$\hat{\rho}_a^{(s)} = \mathbf{N} \left(\left(\frac{\theta^2}{4} - \frac{\theta^4}{48} \right) \hat{a} \hat{\rho}_a^{(0)} \hat{a}^\dagger - \frac{\theta^4}{32} \left(\hat{a} \hat{\rho}_a^{(0)} \hat{a}^{\dagger 2} \hat{a} + \hat{a}^\dagger \hat{a}^2 \hat{\rho}_a^{(0)} \hat{a}^\dagger - \hat{a}^2 \hat{\rho}_a^{(0)} \hat{a}^{\dagger 2} \right) \right) \quad (\text{S4})$$

and

$$\hat{\rho}_a^{(a)} = \mathbf{N} \left(\left(\lambda^2 + \frac{\lambda^4}{3} \right) \hat{a}^\dagger \hat{\rho}_a^{(0)} \hat{a} - \frac{\lambda^4}{2} \left(\hat{a}^\dagger \hat{\rho}_a^{(0)} \hat{a}^2 \hat{a}^\dagger + \hat{a} \hat{a}^{\dagger 2} \hat{\rho}_a^{(0)} \hat{a} - \hat{a}^{\dagger 2} \hat{\rho}_a^{(0)} \hat{a}^2 \right) \right). \quad (\text{S5})$$

When the probability of there being a large number of photons is negligible, only the first term dominates in Eqs. (S4) and (S5), which is the desired result of single photon subtraction and addition, respectively. However, when there is a very large number of photons, the other terms acquire weight so that the experimental state begins to digress from the desired one. For the average photon number of about 0.6 and 1, the experiment well simulates the single photon addition and subtraction.

Experimental details

Single Photon Subtraction – While the acquisition rate (about 160 kHz) for the unperturbed thermal states is just limited by the dead time of the digital oscilloscope used for homodyne measurements, the production rate of photon subtracted states scales with the reflectivity of the beam splitter (set to about 1%) and the quantum efficiency of the on-off photodetectors, and is of the order of 100 kHz. A very good mode matching between the thermal field and the subtraction modes is achieved by using single mode fibers. In particular, the first subtraction mode is the same as that of the fiber used to define the thermal field, while the second subtraction mode is matched to that of homodyne detection. Although the two subtraction modes are different, we have verified that they lead to the same result on the final detected state in this case.

Single Photon Addition – The production rate of the single-photon-added states drops dramatically with respect to the single photon subtraction. Due to the low parametric gain $\lambda \approx 0.01$ and to the strong spatial and spectral filtering introduced in the trigger channel of

the parametric amplifier, the conditional generation takes place at a rate of about 1 kHz. The generation probability depends on the mean photon number of the injected field and is given by $p = |\lambda|^2 (1 + \bar{n})$. Using this relation it is possible to absolutely calibrate the mean photon number in the detected thermal field by comparing the trigger count rate when the injection channel is open and when it is blocked (S8,S9). When a click of the photodetector along the trigger channel is observed, the single-photon-added state is prepared in the signal mode (approximately coincident with the mode of the classical UV pump beam). The purity of the prepared state depends on the ratio between the pump-beam spatial and spectral widths and that of the filters along the trigger channel. Including also the effect of dark counts in the trigger detector (measured to be of the order of 1%), the preparation efficiency of the photon-added state is calculated to be $\xi = 0.92$.

Balanced Homodyne Detection – Imperfections in the homodyne detection of the states are due to: the limited quantum efficiency of the two photodiodes ($\eta_{ph} = 0.89$), the electronic noise of the high bandwidth amplifier (about 12 dB below the shot noise level when operating at a local oscillator power of about 8 mW, resulting in $\eta_{el} = 0.90$), optical losses ($\eta_{op} = 0.90$), and imperfect mode matching ($\eta_{mm} = 0.91$) between the local oscillator and the conditionally prepared state (the amplified mode of the parametric amplifier). The overall detection efficiency is thus estimated to be $\eta = 0.66$.

Theoretical modeling

In order to keep into account all the experimental imperfections, we consider the following model for analyzing the experimental marginal quadrature distributions:

$$P_{out}(x) = (1 - \xi)P_c(x; \eta) + \xi P(x; \eta) \quad (S6)$$

where $P(x; \eta)$ is the marginal distribution of the expected state as measured with a detection efficiency η , contaminated with a portion $(1 - \xi)$ of the distribution $P_c(x; \eta)$ (also measured with a detection efficiency η). First, we have measured the overall detection efficiency η (including detector losses and mode-matching efficiency) by analyzing a

simple thermal state (i.e. without conditioning) whose mean photon number was obtained by the absolute calibration procedure described above (Fig. 3A). η is obtained from the fit of Eq. (S6) by setting $\xi = 1$ and using the expected marginal of a thermal state for $P(x; \eta)$. The value we obtain ($\eta = 0.64$) is in very good agreement with the expected one and is kept fixed in the following. Photon-subtracted states (see Fig. 3B) are found to fit well their expected marginal distributions without any contamination (i.e., with $\xi = 1$). On the other hand, photon-added thermal states (Fig. 3C) and both sequences of addition and subtraction (Figs. 3D and 3E) are found to fit Eq. (S6) with the expected small contamination ($\xi = 0.92$) related to the imperfect process of photon addition. The contaminant contribution comes from the injected thermal state in the case of the simple photon addition, while it derives from residual photon-subtracted thermal state for the two sequences of addition and subtraction.

The same model, with the above parameters, is used to calculate the expected photon number distributions and the Wigner functions of the different states. The slight discrepancies between the experimental and theoretical results in the sections of the Wigner functions of Fig.4D is indeed due to the fact that we choose to use fixed, independently determined, values for the detection and preparation efficiencies ($\eta = 0.64$ and $\xi = 0.92$) in the calculations. Small fluctuations and slow drifts in these values are likely to have occurred during and between the long experimental acquisitions and may fully account for the observed discrepancies.

Quantum Tomography

We have performed the reconstruction of the diagonal density matrix elements using the maximum likelihood estimation (S10). This method gives the density matrix that most likely represents the measured homodyne data. Firstly, we build the likelihood function contracted for a density matrix truncated to 15 diagonal elements (with the constraints of Hermiticity, positivity, and normalization), then the function is maximized by an iterative procedure (S11, S12) and the errors on the reconstructed density matrix elements are evaluated using the Fisher information (S12). In the reconstructions we correct for the

detection losses by using the value of $\eta = 0.64$ as obtained from the measurement of the thermal state discussed above.

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