Quantum-Optical Catalysis: Generating Nonclassical States of Light by Means of Linear Optics

A. I. Lvovsky*

Fachbereich Physik, Universität Konstanz, D-78457 Konstanz, Germany

J. Mlynek

Humboldt-Universität zu Berlin, D-10099 Berlin, Germany

We report preparation and characterization of coherent superposition states $t|0\rangle + \alpha|1\rangle$ of the electromagnetic field by conditional measurements on a beam splitter. This state is generated in one of the beam splitter output channels if a coherent state $|\alpha\rangle$ and a single-photon Fock state $|1\rangle$ are present in the two input ports and a single photon is registered in the other beam splitter output. The single photon thus plays a role of a "catalyst:" it is explicitly present in both the input and the output channels of the interaction yet facilitates generation of a nonclassical state of light.

         PACS numbers: 03.65.Wj, 03.67.Lx, 42.50.Dv

A. Introduction.—Beam splitter (BS) is the simplest quantum optical device in which two incident light beams interfere to produce two output beams. Quantum properties of the beam splitter are manifested in its ability to generate an entangled output state from a nonclassical, but unentangled input [1]. This feature has been exploited in a variety of fundamental experiments, such as preparation of the Einstein-Podolsky-Rosen [2] and Greenberger-Horne-Zeilinger [3] states, discrete- [4] and continuous-variable [5] quantum teleportation. It is also an integral part of many yet unimplemented theoretical proposals, such as single-photon nonlocality [6], generating arbitrary quantum states of light [7] and efficient linear quantum computation [8].

In this Letter we investigate a curious consequence of beam splitter's entangling property. A single-photon Fock state $|1\rangle$ and a coherent state $|\alpha\rangle$ present in two BS input channels generate entangled two-mode output (Fig. 1). A quantum measurement performed on one of the output channels causes this entangled state to collapse projecting the second ("signal") output onto a certain local ensemble. Our goal is to characterize this ensemble in the event the measurement in the first output channel reveals a quantum state *identical* to that in one of the BS inputs, namely, the single-photon state. Contrary to classical intuition we find the signal ensemble to differ dramatically from the input ("target") coherent state: it is approximated by a highly nonclassical coherent superposition of the vacuum and the single-photon Fock state. We call this transformation *quantum-optical catalysis* because the single photon, while facilitating the conversion of the target ensemble, itself remains unaffected by this interaction. We note that a related effect of entanglement-facilitated catalysis has recently been described theoretically, although for a different setting [9].

In order to get insight into underlying physics let us assume that both the input coherent excitation α and the BS transmission t^2 are small: $\alpha \sim t \ll 1$. In this case, the coherent state can be approximated as $|\alpha\rangle = |0\rangle + \alpha|1\rangle$.

Suppose a single-photon detector (SPD) registers a photon. Where could this photon have originated from? If it comes from the coherent state, the photon $|1\rangle$ from the Fock state input is likely to have been reflected into the signal channel. If, on the other hand, the photon detected by the SPD originates from the Fock state transmitted through a BS, the quantum state in the signal channel is with a high probability vacuum $|0\rangle$.

The quantum properties of the beam splitter manifest themselves in *fundamental indistinguishability* of these two possibilities. If the two initial states are prepared in matching optical modes, there is no way of telling which input state has contributed a photon to the SPD channel. As a result, the quantum state in the signal channel is *not merely a statistical mixture of the states $|0\rangle$ and $|1\rangle$ but their coherent superposition,*

$$|\psi_s\rangle \approx \frac{1}{\sqrt{t^2 + \alpha^2}} (t|0\rangle + \alpha|1\rangle). \tag{1}$$

Although $|\psi_s\rangle$ has the same components as the initial faint coherent state, their fractions can be controlled by changing the ratio between α and t. For a constant, small t the increase of α implements a gradual transition
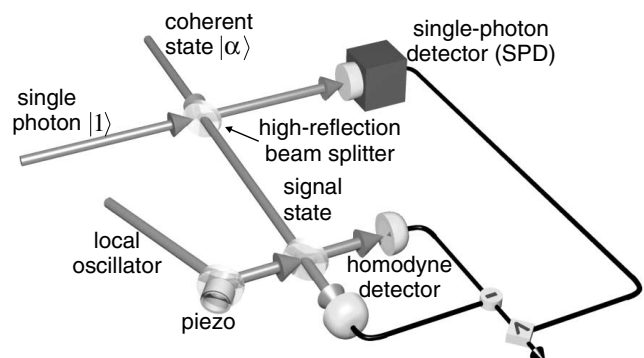

FIG. 1. A conceptual scheme of the experiment. Measurements by the balanced homodyne detector are conditioned on registering a photon by the single-photon detector.

between highly classical ($|0\rangle$) and highly nonclassical ($|1\rangle$) states of light. In this sense the state $|\psi_s\rangle$ can be considered a bridge between the particle and wave aspects of the electromagnetic field.

In the present paper we generate $|\psi_s\rangle$ and investigate it by means of quantum homodyne tomography [10]. This work was inspired in part by a recent experiment by Rarity *et al.* who overlapped a single photon with a coherent state on a beam splitter and studied the nonclassical photon statistics in the output [11]. Means of preparing the single photon in a well-defined spatiotemporal mode and pulsed time-domain homodyne detection have been elaborated in our group's earlier work [12–15]. Applications of conditional measurements on a beam splitter to generate a wide variety of quantum states of light were studied by Dakna *et al.* [7].

Our experiment can be seen as an implementation of the first of two stages of the nonlinear sign shift quantum gate, the basic element of the recent linear-optical quantum computation proposal [8]. Such a gate is another example of quantum optical catalysis, because the required modification of the target state is achieved with the two ancilla channels involved remaining in their original quantum states.

B. Theory.—We start by assuming that the input state $|1\rangle$ is prepared with a 100% efficiency in an optical mode that perfectly matches that of the coherent state $|\alpha\rangle = e^{-|\alpha|^2/2} \sum_n \alpha^n/\sqrt{n!}\,|n\rangle$ at the BS input. The entangled state in the beam splitter output can be found by applying the BS transformation operator

$$|\Psi_{\text{out}}\rangle = \hat{B}|1, \alpha\rangle, \tag{2}$$

where

$$\hat{B}|m, n\rangle = \sum_{j,k=0}^{m,n} \sqrt{\frac{(j+k)!\,(m+n-j-k)!}{m!\,n!}} \binom{m}{j}\binom{n}{k}$$
$$\times (-1)^k t^{n+j-k} r^{m-j+k}$$
$$\times |j+k, m+n-j-k\rangle, \tag{3}$$

$r^2 = 1 - t^2$ is the BS reflectivity and $\binom{a}{b}$ are binomial coefficients [16].

A commercial single-photon detector generates a "click" when $n \geq 1$ photons are incident upon its sensitive area. It is described by the following positive operator-valued measure (POVM):

$$\hat{\Pi}_{\text{no-click}} = \sum_{n=0}^{\infty} (1 - \eta_{\text{SPD}})^n |n\rangle\langle n| \,;$$
$$\hat{\Pi}_{\text{click}} = \hat{1} - \hat{\Pi}_{\text{no-click}}, \tag{4}$$

where η_{SPD} is its quantum efficiency. A measurement via the SPD leads to a collapse of the state $|\Psi_{\text{out}}\rangle$ projecting it, in the event of a "click," upon the following ensemble in the signal channel:

$$\hat{\rho}_s = \text{Tr}_1(|\Psi_{\text{out}}\rangle\langle\Psi_{\text{out}}|\hat{\Pi}_{\text{click}}), \tag{5}$$

which is readily calculated.

In our experiment the initial single photon is prepared by means of conditional measurements on a biphoton generated via parametric down conversion. As discussed in our previous publications [12–14], imperfections associated with this technique result in the photon being prepared with a substantial admixture of the vacuum state: $\hat{\rho}_{|1\rangle} = \eta_{|1\rangle}|1\rangle\langle 1| + (1 - \eta_{|1\rangle})|0\rangle\langle 0|$, where $\eta_{|1\rangle}$ is the preparation efficiency. The vacuum fraction of $\hat{\rho}_{|1\rangle}$, interacting on the beam splitter with the coherent state, does not entangle itself with it [1] but produces *coherent states* with amplitudes αt and αr in the two BS output channels. Taking this circumstance into account, we write the output ensemble as

$$\hat{\rho}'_s = \eta'_{|1\rangle}\hat{\rho}_s + (1 - \eta'_{|1\rangle})|\alpha t\rangle\langle\alpha t|. \tag{6}$$

Note that the quantity $\eta'_{|1\rangle}$ in the above equation is always higher than $\eta_{|1\rangle}$ because a photon present in the input increases the likelihood for the SPD to trigger. For $\alpha \ll t$, $\eta'_{|1\rangle}$ tends to 1, for $\alpha \gg t$ it approaches $\eta_{|1\rangle}$.

Next we need to account for the SPD dark count events. In such an event, the quantum state in the signal channel is not conditioned on that in the SPD channel. The BS acts upon the incident single-photon state simply as a lossy reflector, reducing its efficiency by a factor of r^2. In addition, the coherent field causes the phase-space displacement of this state, producing a statistical mixture of a *displaced Fock state* [17,18] and a coherent state:

$$\hat{\rho}_{\text{DC}} = \eta_{|1\rangle} r^2 \hat{D}(\alpha t)|1\rangle\langle 1|\hat{D}^\dagger(\alpha t)$$
$$+ (1 - \eta_{|1\rangle} r^2)|\alpha t\rangle\langle\alpha t|, \tag{7}$$

where $\hat{D}(\alpha t)$ is the displacement operator. The above ensemble admixes to $\hat{\rho}'_s$ in the proportion determined by the SPD dark count rate.

Finally, in order to compare our prediction with the experimental results we need to account for the nonunitary quantum efficiency η_{HD} of the balanced homodyne detector. This is done by means of the generalized Bernoulli transformation well described in the literature [16,19].

C. Experimental apparatus.—The core of our apparatus consisted of the setup for generating the single-photon Fock state, which was the same as in our previous experiments [12–14]. A 82-MHz repetition rate train of 1.6-ps pulses generated by a Spectra-Physics Ti:sapphire laser at 790 nm was frequency doubled and directed into a beta-barium borate crystal for down conversion. The down conversion occurred in a type-one frequency-degenerate, but spatially nondegenerate configuration. A single-photon detector, placed into one of the emission channels, detected photon-pair creation events. This detector firing ensured that a photon has as well been emitted into the other down-conversion channel, thus preparing single-photon Fock states by conditional measurements. All further measurements were conditioned upon a biphoton production event. We have obtained between 300 and 400 such events in a second.

Pulses containing conditionally prepared photons entered the optical arrangement shown in Fig. 1, which had to be maintained interferometrically stable throughout the experimental run. We used a Perkin-Elmer SPCM-AQ-131 single-photon counting module with $\eta_{SPD} \approx 0.5$ and a beam splitter with a reflectivity of $r^2 = 0.925$. The choice of the BS was dictated, on one hand, by the requirement that t be small compared to 1; on the other hand, the SPD count rate, proportional to $t^2 + \alpha^2$ for small α, should have been sufficiently high in order to prevent domination of the dark counts and to allow acquisition of a large amount of data sufficient for quantum tomography.

The local oscillator for balanced homodyne detection [15], as well as the target coherent state, have been provided by the master Ti:sapphire laser. The spatiotemporal modes of these pulses had to match each other, as well as that of the single-photon pulse emerging from the down converter. To this end, we first mode matched the single-photon pulse with the local oscillator via the technique described in [12,14]. The coherent state was then mode matched with the local oscillator by optimizing the visibility of the interference pattern observed between these two classical fields.

A set of preliminary measurements was carried out in order to test the system and evaluate its main characteristics. First, we calibrated the homodyne detector (HD) by observing the quantum noise of the vacuum state with the beam paths of both the coherent state and the Fock state blocked [Fig. 2(a)]. Second, we unblocked the coherent state input so the quantum state entering the HD was also a coherent state with amplitude αt [Fig. 2(b)]. Performing homodyne tomography on this state allowed us to determine α to within 0.15. Third, we blocked the coherent state but unblocked the Fock state. In this case, the quantum noise measured by the HD was associated with the statistical mixture of the states $|1\rangle$ and $|0\rangle$ with an efficiency $\eta_{tot} = r^2 \eta_{|1\rangle} \eta_{HD}$ [Fig. 2(c)]. By analyzing the statistics of this noise, we found $\eta_{tot} = 0.58 \pm 0.02$. Since it was known from previous measurements that $\eta_{HD} = 0.91$ [15], we estimated $\eta_{|1\rangle} = 0.69$.

Finally, we unblocked both the coherent state and the Fock state BS inputs, and performed homodyne measurements conditioned upon the SPD firing. The local oscillator phase was scanned linearly by means of a piezoelectric transducer. The acquired data were then scaled in accordance with the vacuum state noise acquired earlier and each data sample was associated with the appropriate local oscillator phase value. The Wigner function was reconstructed for each data set by means of the inverse Radon transformation implemented via the standard filtered back-projection algorithm [16] with a cutoff frequency of 6.4. This algorithm was applied directly to the quadrature data without calculating marginal distributions associated with each phase value [18].
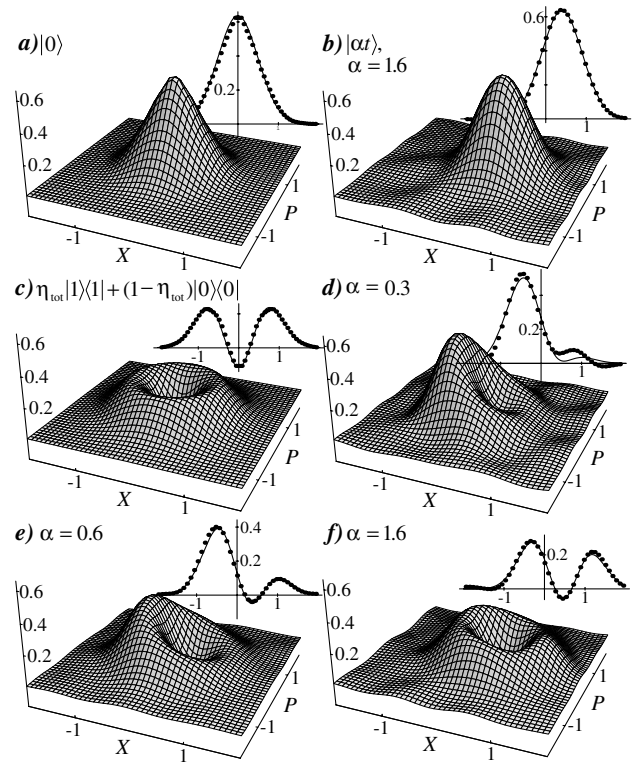

FIG. 2. Wigner functions of the quantum states observed in the experiment. (a)–(c): States obtained during the alignment procedure: (a) vacuum state; (b) coherent state; (c) statistical mixture of states $|1\rangle$ and $|0\rangle$, efficiency $\eta_{tot} = 0.59$. (d)–(f): Quantum catalysis ensembles obtained for various values of α. For low α, the Wigner function resembles that of the vacuum state; for high α—the displaced Fock state. Insets show Wigner functions' cross sections along with theoretical fits.

D. Results and discussion.— Figures 2(d)–2(f) show the Wigner functions reconstructed for various coherent state amplitudes. The three Wigner functions shown allow one to trace the trend the catalysis ensembles follow with increasing α. For $\alpha = 0$, a photon registered in the SPD channel means there is nothing in the signal channel (unless it is a dark count); the signal state must thus be vacuum. We have not reproduced this case experimentally due to an extremely low count rate associated with it. With α increasing to the order of t, the quantum state approximates the coherent superposition (1) with a significant admixture of the vacuum due to a nonunitary $\eta_{|1\rangle}$ [Fig. 2(d)].

For high α's the SPD will fire with almost every laser pulse detecting a large fraction of the reflecting coherent state's photons. Because of the significant role of multiphoton terms in the coherent state decomposition, this regime cannot be viewed as "catalytical" and is no longer described by Eq. (1). It is rather analogous to the dark count case discussed earlier: the measurement of the signal state is effectively no longer conditioned on the SPD ensemble, so a statistical mixture (7) of a displaced Fock state and a coherent state obtains. The ensemble shown in
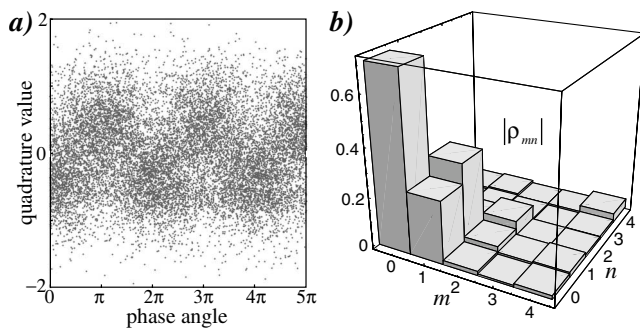
FIG. 3. 14153 raw homodyne detector data (a) and the absolute values of the density matrix elements in the Fock representation (b) for the catalysis ensemble state with $\alpha = 0.3$. The ensemble purity factor $\mathrm{Tr}(\hat{\rho}^2) = 0.77$.

Fig. 2(f) approaches this situation. An experimental study of displaced Fock states will be reported separately [18].

The fits shown in Figs. 2(d)–2(f) were calculated using the experimental parameters determined during the preliminary measurements and varied within their tolerances in order to obtain the best fit. An exception was made for η_{HD} which was set to 0.83 in order to account for the imperfect mode matching between the coherent state and the local oscillator. Precise treatment of this imperfection is rather complicated; reducing the value of η_{HD} is a simplified solution which, however, allows the generation an excellent fit shown in Figs. 2(e) and 2(f). A deviation from the perfect fit seen in Fig. 2(d) is a technical artifact caused by a very low data acquisition rate.

Density matrices of the catalysis ensembles have been determined by applying the quantum state sampling method [16,20] directly to the homodyne data. The ensemble shown in Fig. 3 approximates a statistical mixture of the state (1) and the vacuum. Presence of nondiagonal elements ρ_{01} and ρ_{10} distinguishes the ensemble from a statistical mixture of the states $|1\rangle$ and $|0\rangle$ investigated in our earlier papers [12,13]. Unlike the coherent state, on the other hand, our ensemble has negligible matrix elements associated with the multiphoton Fock terms.

In conclusion, we have synthesized and characterized a new quantum state of light, a coherent superposition of the single-photon and vacuum states. This state is generated by means of a "catalytical" process in which the single-photon state enables the interaction without being affected by it. By changing the amplitude of the target coherent state, a gradual transition between a highly classical and a highly nonclassical state of light occurs. This experiment was implemented by means of linear optics, single-photon sources, and detectors and can be viewed as a step towards experimental realization of linear optical quantum computation.

Note added. — A related work in which coherent superpositions of states $|0\rangle$ and $|1\rangle$ were generated by means of seeding the trigger mode of a down-converting crystal has recently been reported [21].

*Email address: Alex.Lvovsky@uni-konstanz.de

[1] M. S. Kim *et al.* Phys. Rev. A **65**, 032323 (2002).
[2] Z. Y. Ou *et al.,* Phys. Rev. Lett. **68**, 3663 (1992).
[3] D. Bouwmeester *et al.,* Phys. Rev. Lett. **82**, 1345 (1999).
[4] D. Bouwmeester *et al.,* Nature (London) **390**, 575 (1997).
[5] A. Furusawa *et al.,* Science **282**, 706 (1998).
[6] S. M. Tan, D. F. Walls, and M. J. Collett, Phys. Rev. Lett. **66**, 252 (1991).
[7] M. Dakna *et al.* Phys. Rev. A **59**, 1658 (1999); M. Dakna *et al.* Eur. Phys. J. D **3**, 295 (1998), and references therein.
[8] E. Knill, R. Laflamme, and G. Milburn, Nature (London) **409**, 46 (2001); T. C. Ralph *et al.* Phys. Rev. A **65**, 012314 (2002) presents a simplified scheme of the nonlinear phase shift gate. It consists of two sequential conditional measurements in which the target state is overlapped with two ancilla modes in states $|1\rangle$ and $|0\rangle$ on beam splitters.
[9] D. Jonathan and M. B. Plenio, Phys. Rev. Lett. **83**, 3566 (1999); G. Vidal and J. I. Cirac, Phys. Rev. Lett. **88**, 167903 (2002).
[10] D. T. Smithey *et al.,* Phys. Rev. Lett. **70**, 1244 (1993); G. Breitenbach, S. Schiller, and J. Mlynek, Nature (London) **387**, 471 (1997); M. Vasilyev *et al.,* Phys. Rev. Lett. **84**, 2354 (2000).
[11] J. G. Rarity, P. R. Tatster, and R. Loudon, quant-ph/9702032.
[12] A. I. Lvovsky *et al.,* Phys. Rev. Lett. **87**, 050402 (2001).
[13] A. I. Lvovsky and J. H. Shapiro, Phys. Rev. A **65**, 033830 (2002).
[14] T. Aichele, A. I. Lvovsky, and S. Schiller, Eur. Phys. J. D **18**, 237 (2002).
[15] H. Hansen *et al.,* Opt. Lett. **26**, 1714 (2001).
[16] U. Leonhardt, *Measuring the Quantum State of Light* (Cambridge University Press, Cambridge, United Kingdom, 1997).
[17] F. A. M. de Oliveira *et al.,* Phys. Rev. A **41**, 2645 (1990).
[18] A. I. Lvovsky and S. A. Babichev, quant-ph/0202163.
[19] T. Kiss, U. Herzog, and U. Leonhardt, Phys. Rev. A **52**, 2433 (1995).
[20] G. M. D'Ariano, U. Leonhardt, and H. Paul, Phys. Rev. A **52** R1801 (1995); U. Leonhardt and M. G. Raymer, Phys. Rev. Lett. **76**, 1985 (1996).
[21] K. J. Resch, J. S. Lundeen, and A. M. Steinberg, Phys. Rev. Lett. **88**, 113 601 (2002).