Spectral Density Matrix of a Single Photon Measured

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We propose and demonstrate a method for measuring the spectral density matrix of a single photon pulse. The method is based on registering Hong-Ou-Mandel interference between a photon to be measured and a pair of attenuated and suitably delayed laser pulses described by a known spectral amplitude. The density matrix is retrieved from a two-dimensional interferogram of coincidence counts. The method has been implemented for a type-I down-conversion source, pumped by ultrashort laser pulses. The experimental results agree well with a theoretical model which takes into account the temporal as well as spatial effects in the source.

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Development of single photon sources brings the promise of implementing novel quantum-enhanced technologies. In many applications, including quantum computing based on linear optics [1], photon sources are required not only to deliver single light quanta but also to supply them in a well-defined mode. This is a necessary condition for quantum interference between independent sources [2] which is required in the above schemes. Besides, characterizing single photons is also interesting from the fundamental point of view. Historically photons were first described within the framework of quantum field theory, but more recently it was pointed out that a photon wave function can be introduced [3]. This kind of description, generalized by the introduction of the density matrix constructed out of projectors on the states with specific wave functions, seems to be the most elegant and effective theoretical tool for developing quantum-enhanced technologies [4].

Until now measurements of polarization [5] and spatial density matrix [6] of a single photon were reported. The temporal characteristics of single photons were assessed only by verifying whether they interfere with other sources [7,8] or between themselves [9]. Only recently a method for complete temporal characterization of single photons using first-order interference and frequency shifting has been proposed [10]. In this Letter we devise and demonstrate a novel method for complete characterization of the temporal degree of freedom: a measurement of the spectral density matrix of a single photon. We show that the two-dimensional map of coincidence counts recorded as a function of delays between an unknown photon and a pair of weak reference pulses can be used to reconstruct the magnitude and the phase of the density matrix. We present a measurement for a type-I spontaneous down-conversion process in a bulk β-barium borate (BBO) crystal, and compare the results of the reconstruction with theoretical predictions.

When a single photon is launched into a single-mode fiber, the situation is significantly simplified since the spatial mode is well defined. If, moreover, polarization of the photons is fixed, the only remaining degree of freedom is the spectral one. In this case a single photon component of the field can be described by the following density operator:

$$\hat{\rho} = \iint d\omega d\omega' \rho(\omega, \omega') \hat{a}^\dagger(\omega)|0\rangle\langle 0|\hat{a}(\omega'), \qquad (1)$$

where $\hat{a}(\omega)$ is an operator annihilating a photon of frequency ω in the fiber, while $\rho(\omega, \omega')$ is a density matrix of a single photon given in the spectral domain.

Our method for measuring $\rho(\omega, \omega')$ is based on the Hong-Ou-Mandel interference effect between the single photon to be characterized and a local oscillator (LO) pulse of known shape attenuated to a single photon level. The visibility of the two-photon interference dip is proportional to the overlap between the modes of interfering photons. Measuring it for a suitable class of LO pulses suffices to retrieve the spectral density matrix of a single photon. In a broad sense, our experiment is a single photon analog of the homodyne method for measuring quantum correlations within a light pulse [11].

The method is presented in Fig. 1. The first part is a Michelson interferometer which serves as a LO pulse

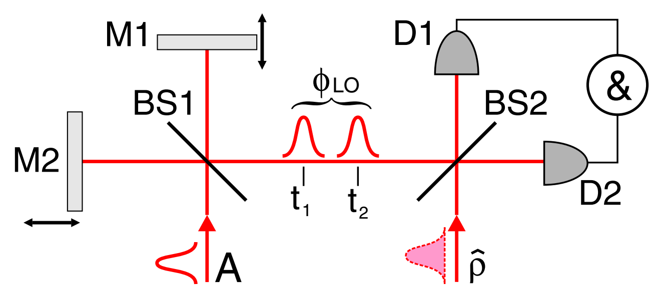

FIG. 1 (color online). Scheme of the experiment. A weak laser pulses described by a spectral amplitude $A(\omega)$ are split in a Michelson interferometer, comprising mirrors M1 and M2 and a 50/50 beam splitter BS1, to form the local oscillator $\phi_{\mathrm{LO}}(\omega)$. Then they interfere with an unknown photon described by the density operator $\hat{\rho}$ on a 50/50 beam splitter BS2 and may give rise to a coincidence click of the detectors D1 and D2.

modulator. The second part is the beam splitter on which the Hong-Ou-Mandel interference occurs. Master laser pulse described by a spectral amplitude function $A(\omega)$ is split into two pulselets centered around delays t_1 and t_2 determined by the length of the arms in the interferometer. The normalized spectral amplitude of LO pulses prepared this way reads

$$\phi_{\mathrm{LO}}(\omega) = A(\omega)\frac{\exp(-i\omega t_1) + \exp(-i\omega t_2)}{\sqrt{2S(t_2 - t_1)}}, \quad (2)$$

where $S(t_2 - t_1)$ is the probability that a single photon will pass through the Michelson interferometer. The modulated LO pulses interfere with unknown single photons on a 50/50 beam splitter BS2. If the master laser pulse contains a photon with probability l, while the source produces photon described by spectral density matrix $\rho(\omega, \omega')$ with probability f per pulse, we can calculate the probability of registering coincidence with detectors of quantum efficiency η to be [4]

$$p_c(t_1, t_2) = flS(t_2 - t_1)\frac{\eta^2}{2}[1 - Q(t_1, t_2)], \quad (3)$$

where we have assumed that each of the input states contains at most one photon and $Q(t_1, t_2) = \iint d\omega d\omega' \phi^*_{\mathrm{LO}}(\omega)\rho(\omega, \omega')\phi_{\mathrm{LO}}(\omega')$ is an overlap between the local oscillator $\phi_{\mathrm{LO}}(\omega)$ and the density matrix $\rho(\omega, \omega')$. We can partially evaluate $Q(t_1, t_2)$ using Eq. (2) and insert the result into Eq. (3). Rearranging factors we can obtain

$$Np_c(t_1, t_2) = 2S(t_2 - t_1) - 2\mathrm{Re}\tilde{\rho}(t_1, t_2) - D(t_1) - D(t_2), \quad (4)$$

where Re stands for the real part, N is a normalization factor combining the probabilities of detecting a photon from the master laser l and the source f, as well as detector efficiency η, while

$$\tilde{\rho}(t_1, t_2) = \iint d\omega_1 d\omega_2 e^{i\omega_1 t_1 - i\omega_2 t_2} A^*(\omega_1)\rho(\omega_1, \omega_2)A(\omega_2) \quad (5)$$

is the inverse Fourier transform of a density matrix restricted to the spectral domain of the master laser pulses and $D(t) = \tilde{\rho}(t, t)$ is the function describing the standard Hong-Ou-Mandel interference dip. At this point let us note that the density matrix $\rho(\omega, \omega')$ is nonzero only near $\omega = \omega' = \omega_0$ in case of narrow band photons, where ω_0 is the central frequency of the single photons and the master laser. Therefore $\mathrm{Re}\tilde{\rho}(t_1, t_2)$ oscillates like $\cos[\omega_0(t_2 - t_1)]$. Also $S(t_2 - t_1)$ contains such oscillations. On the other hand, $D(t)$ is a slowly varying function of its argument. Therefore, individual components of the right-hand side of Eq. (4) can be separated in the frequency domain. Let us apply the Fourier transform $\iint dt_1 dt_2 \exp(-i\omega_1 t_1 + i\omega_2 t_2)\ldots$ to both sides of Eq. (4) and rearrange terms:

$$A^*(\omega_1)\rho(\omega_1, \omega_2)A(\omega_2) + A(-\omega_1)\rho^*(-\omega_1, -\omega_2)A^*(-\omega_2) = \delta(\omega_1 - \omega_2)|A(\omega_1 + \omega_2)|^2 - N\tilde{p}_c(\omega_1, \omega_2) + \cdots, \quad (6)$$

where by dots we have denoted Fourier transform of $D(t_1)$ and $D(t_2)$, while $\tilde{p}_c(\omega_1, \omega_2)$ is the Fourier transform of coincidence counts. Additionally we used the fact that $|A(\omega)|^2$ is the Fourier transform of $S(t)$.

In the experiment we can directly measure $p_c(t_1, t_2)$ as a function of t_1 and t_2. In the case of narrow band single photons retrieving of the spectral density matrix runs as follows: First, Fourier transform of $p_c(t_1, t_2)$ is computed. A region in the frequency space where contribution from $S(t_2 - t_1)$ and $\tilde{\rho}(t_1, t_2)$ lies is separated. Next, a large contribution from $S(t_2 - t_1)$ is calculated and subsequently subtracted by measuring $p_c(t_1, t_2)$ for large t_1 and t_2 where $\tilde{\rho}(t_1, t_2)$ is zero. This way we obtain the Fourier transform of $\tilde{\rho}(t_1, t_2)$, which equals $A^*(\omega_1)\rho(\omega_1, \omega_2)A(\omega_2)$. It is divided by spectral amplitude of master laser pulses $A(\omega)$, which is measured separately and finally $\rho(\omega_1, \omega_2)$ is found. Note that the last step is well defined only for single photons of bandwidth narrower than that of the master laser.

To illustrate the above dry formulas a typical coincidence pattern and its Fourier transform are depicted in Fig. 2. The diagonal fringes in Fig. 2(a) come from $S(t_2 - t_1)$, the vertical and horizontal stripes from $D(t_1)$ and $D(t_2)$, while the most interesting term $\tilde{\rho}(t_1, t_2)$ contributes only in the very center of the picture. It is easier identified in Fourier transform plot in Fig. 2(b) where a diagonal cloud corresponds to $A^*(\omega_1)\rho(\omega_1, \omega_2)A(\omega_2)$, $D(t_1)$ and $D(t_2)$ contribute a cross around zero frequency while Fourier transform of $S(t_2 - t_1)$ is seen as a ridge along $\omega_1 = \omega_2$.

In the above derivations we have assumed interference between single photons. In our experiment we interfere a weak coherent state with a multimode thermal state. Whereas the visibility of such interference can be exactly calculated using the semiclassical theory [8], two photon terms of the coherent state and the thermal state contribute only towards a constant background of coincidence counts. However, the shape of the interference pattern does characterize the single photon component of the signal field. Its density matrix can still be retrieved in the way described above.

Our experimental setup is depicted in Fig. 3. The master laser (RegA 9000 from Coherent) produces a train of 165 fs FWHM long pulses at a 300 kHz repetition rate centered at 774 nm. Most of the energy goes to the second harmonic generator, based on a 1 mm thick BBO crystal cut for type-I process. Ultraviolet pulses produced this way have 1.3 nm bandwidth and 30 mW average power. They are filtered out of fundamental using a pair of dichroic mirrors (DM) and a

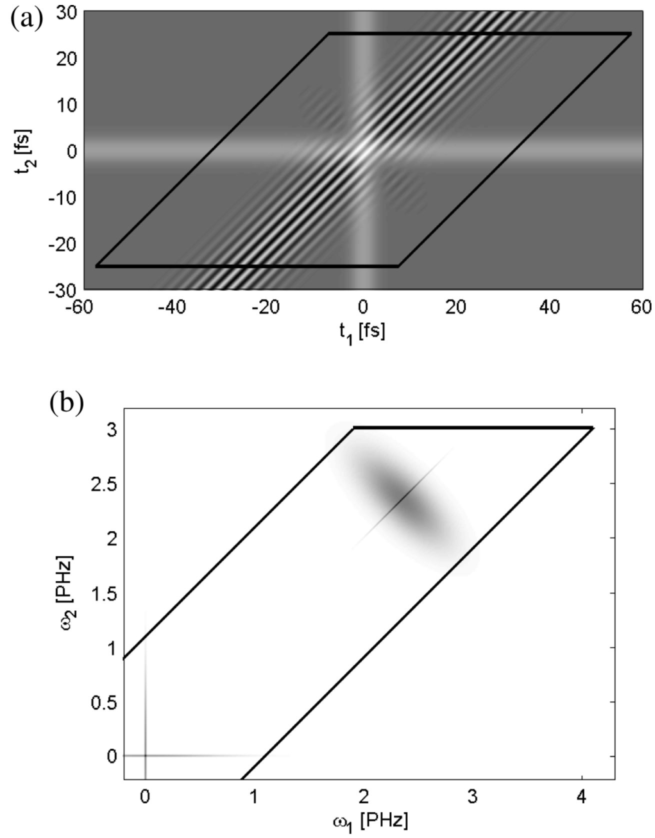

FIG. 2. (a) A typical coincidence interferogram as a function of delays t_1 and t_2 and (b) its Fourier transform. The black parallelogram in (a) defines the scan range, which with suitable sampling density yields the region of interest in the frequency domain, outlined in (b).

color glass filter (BG) (Shott BG39), and imaged using 20 cm focal length lens (IL) on a down-coversion crystal X, where they form a spot measured to be 155 μm in diameter. The crystal X is a 1 mm thick BBO crystal cut 29.7° to the optic axis, and oriented for maximum source intensity. A portion of down-converted light propagating at an angle of 2.5° to the pump beam passes through a 10 nm interference filter centered at 774.5 nm and is coupled into a single-mode fiber. This defines the spatial mode in which the down-conversion is observed [12]. About 4% of energy of master laser pulses is reflected towards a LO preparation arm. The pulses first go through a half wave plate (HWP) and a polarizer (P) allowing for fine control of the energy. Then they are delayed in a computer-controlled delay line and enter a Michelson interferometer which allows for generation of double pulses with a well-defined temporal separation $\Delta t = t_2 - t_1$. Finally, the LO is attenuated to contain less than 0.1 photon on average and coupled to a single-mode fiber, where its polarization is adjusted using a fiber polarization controller (FPC) to match the polarization of the photon coming from the down-conversion source. Both the down-conversion and local oscillator photons interfere in a 50/50 single-mode fiber coupler and are detected using single photon counting modules (SPCM) (PerkinElmer SPCM-AQR-14-FC) connected to fast coincidence counting electronics (suitably programmed Virtex4 protype board ML403 from Xilinx) detecting events in coincidence with master laser pulses.

For calculating the actual density matrix of unknown single photons, a characterization of master laser pulses was necessary. This was accomplished using the frequency-resolved optical gating (FROG) technique [13]. Retrieved spectral intensity $|A(\omega)|^2$ and phase $\arg A(\omega)$ are plotted in Fig. 4.

The complete measurement consisted in a series of 6 scans of a rectangular grid depicted in Fig. 2(a) spanned by 4000×25 points, where the latter number refers to the direction along the fringes. The corresponding mesh was 0.233 fs $\times$ 66 fs and coincidences were counted for 80 ms at each point. The reconstructed spectral density matrix of a single photon is depicted in Fig. 5. We compare it with

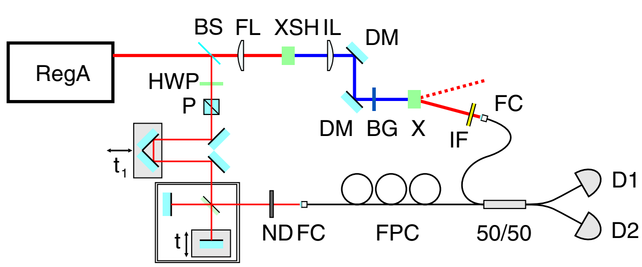

FIG. 3 (color online). The experimental setup. BS: beam splitter; FL: focusing lens; XSH: BBO crystal for generation of the second harmonic; IL: imaging lens; DM: dichroic mirrors; BG: blue glass filter; X: down-conversion crystal; IF: interference filter; FC: fiber coupling stage, HWP: half wave plate; P: polarizer; ND: neutral density filter; FPC: fiber polarization controller; D1 and D2: single photon counting modules.

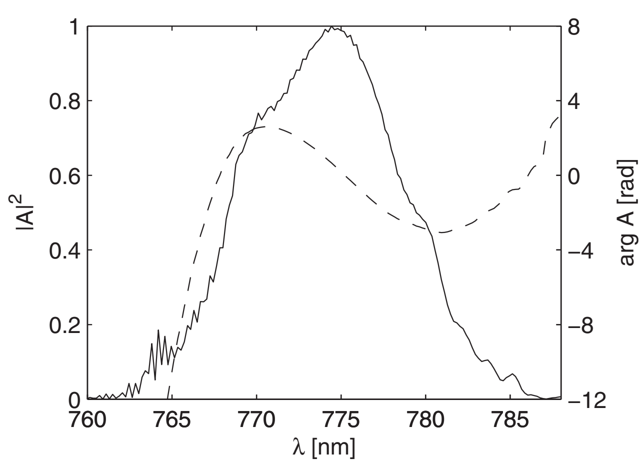

FIG. 4. Spectral intensity $|A(\omega)|^2$ (solid line) and phase $\arg A(\omega)$ (dashed line) of the master laser pulse retrieved using FROG.

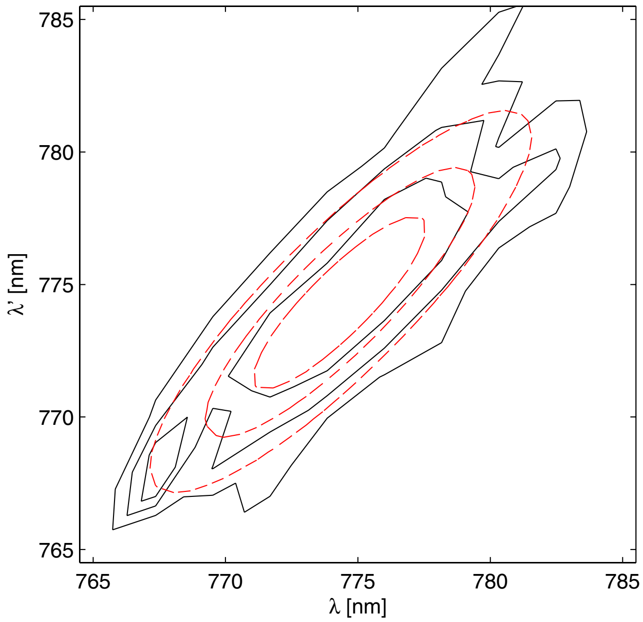

FIG. 5 (color online). Contour plot of the absolute value of the measured spectral density matrix $|\rho(\omega, \omega')|$ as a function of wavelengths $\lambda = 2\pi c/\omega$ (solid lines) and a theoretical prediction for this quantity (dashed red lines). The contours were drawn at 0.75, 0.5, and 0.25 of the maximum values, the outermost encircles 7×5 experimental data points.

theoretical calculations plotted with dashed lines in the same figure. The theoretical model used in these calculations assumed the exact phase matching function of the nonlinear crystal and the ultraviolet pump pulse shape computed from the measured $A(\omega)$ using perturbative approach. The transverse components of the wave vectors for the pump and down-converted beams were treated in the paraxial approximation. The spectral density matrix was calculated for coherent superpositions of plane-wave components of the down-conversion light that add up to localized spatial modes defined by the collecting optics and single-mode fiber. The other photon from the source, which remains undetected, was traced out assuming that it can propagate at any direction and have any frequency that is consistent with the conservation of energy and perpendicular momentum in the down-conversion crystal. As seen in Fig. 5 the theoretical calculations predict more pronounced correlations, i.e., smaller width along the antidiagonal $\rho(\omega_0 + \delta, \omega_0 - \delta)$ than was actually measured. We attribute this discrepancy to a difference between the actual ultraviolet pump pulse shape and the one calculated from $A(\omega)$. Also the tips of the density matrix are measured with reduced accuracy, since in that region the raw experimental result is divided by a relatively small master laser spectral intensity $A(\omega)$ which amplifies errors. The theoretical model predicts the phase of the density matrix to be smaller than $\pi/30$ in the region bounded by the contour at 0.25 maximum. The measured phase in this region is smaller than $\pi/10$ and varies randomly from point to point.

In summary, we proposed and demonstrated a method for measuring the spectral density matrix of a single photon component of the electromagnetic field in a single-mode fiber. The method is based on two photon interference and is thus limited to the spectral range where known reference pulses are available; however, it allows for retrieving both amplitude as well as phase of the density matrix. We have applied this method to a down-conversion-based source of single photons and found that measured density matrix agrees with theoretical predictions.

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