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Experimental Demonstration of Three Mutually Orthogonal Polarization States of Entangled Photons

Tedros Tsegaye,¹ Jonas Söderholm,¹ Mete Atatüre,⁴ Alexei Trifonov,^{1,2} Gunnar Björk,^{1,*}
Alexander V. Sergienko,³ Bahaa E. A. Saleh,³ and Malvin C. Teich^{3,4}

¹*Department of Electronics, Royal Institute of Technology (KTH), Electrum 229, SE-164 40 Kista, Sweden*

²*Ioffe Physical Technical Institute, 26 Polytekhnicheskaya, 194021 St. Petersburg, Russia*

³*Quantum Imaging Laboratory, Department of Electrical and Computer Engineering, Boston University,
8 Saint Mary's Street, Boston, Massachusetts 02215*

⁴*Quantum Imaging Laboratory, Department of Physics, Boston University,
8 Saint Mary's Street, Boston, Massachusetts 02215*

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Linearly polarized classical light can be expressed in a vertical and a horizontal component. Geometrically rotating vertically polarized light by 90° will convert it to the orthogonal horizontal polarization. We have experimentally generated a two-photon state of light which evolves into an orthogonal state upon geometrical rotation by 60° . Rotating this state by an additional 60° will yield a state which is mutually orthogonal to the first two states. Generalizing this procedure, one can generate $N + 1$ mutually orthogonal N -photon states that cyclicly evolve from one to another upon a geometric rotation by $180/(N + 1)$ degrees.

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Polarized light is often described in terms of its Stokes parameters [1], which were introduced to classify the polarization properties of classical light in the middle of the 19th century [2]. With the advent of lasers and photon-counting technology, it was natural to introduce the Stokes operators [3], describing the polarization properties of quantized fields. Some time later it became evident that sensitive measurements of polarization were limited by the inherent polarization noise of the fields [4], and soon thereafter several proposals of polarization squeezed light were put forth [5]. The Stokes operators classify the second order correlations only between orthogonal field modes [6], but for quantized fields higher order correlations can lead to interesting results. Therefore the classification of polarization has been extended to fourth-order correlations [7]. Recently, operational definitions and their experimental implementations of quantum polarization have been discussed [8]. In this paper we report an alternative approach to quantum polarization, where we find the eigenstates of the generator of geometrical rotation and then construct a complementary operator through the operator's eigenstates.

Our starting point is to find states which are invariant under geometrical rotation. This is much in the spirit of Prakash and Chandra [9], and Agarwal [6] who defined *unpolarized* quantum states of light as the states invariant under any geometrical rotation *and* any differential phase shift. (Later, and apparently independently, Lehner *et al.* defined unpolarized states of light in the same manner [10].) We expand the electromagnetic field in two plane waves with horizontal-vertical linear polarization (indicated by the subscript symbol $+$) and expand the field state in the number basis $|m, n\rangle_+ \equiv |m\rangle_+ \otimes |n\rangle_+$. In this expansion the operator generating geometric rotation is given by

$$i(\hat{a}^\dagger \hat{b} - \hat{a} \hat{b}^\dagger) \equiv \hat{S}_y, \quad (1)$$

where \hat{a} and \hat{b} are the annihilation operators for the first and second mode, respectively, and \hat{S}_y is the Stokes operator generating rotation around the direction of the wave vector of the field. The associated unitary operator which rotates a field an angle θ is $\exp(i\theta \hat{S}_y)$. The operator (1) can also be seen as the well-known Jaynes-Cummings interaction Hamiltonian used extensively in atomic physics and

in cavity quantum electrodynamics. It describes a linear coupling between two oscillators, in this case, e.g., a vertically and a horizontally polarized electromagnetic field of the same frequency. We note that \hat{S}_y commutes with the total excitation operator $\hat{a}^\dagger \hat{a} + \hat{b}^\dagger \hat{b}$ (which is the Stokes operator \hat{S}_0) so the total excitation number is an invariant under geometrical rotation. It therefore makes sense to look at the field in each excitation manifold separately. Let us look at a state in excitation manifold N spanned by $N + 1$ basis states $|0, N\rangle_+, |1, N - 1\rangle_+, \dots, |N, 0\rangle_+$. It is well known from the theory of linear coupling of bosonic oscillators with total excitation N that the eigenstates to the Jaynes-Cummings Hamiltonian (1) are *dressed states* $|\circ_n^{(N)}\rangle$ differing in eigenvalue by $\Delta E = 2$. The states are nondegenerate and orthogonal, and therefore provide an alternative basis for expanding any state in excitation manifold N . To exemplify, we explicitly express these states in the $N = 2$ excitation manifold:

$$|\circ_1^{(2)}\rangle = (|0, 2\rangle_+ - i\sqrt{2}|1, 1\rangle_+ - |2, 0\rangle_+)/2, \quad (2)$$

$$|\circ_2^{(2)}\rangle = (|0, 2\rangle_+ + |2, 0\rangle_+)/\sqrt{2}, \quad (3)$$

$$|\circ_3^{(2)}\rangle = (|0, 2\rangle_+ + i\sqrt{2}|1, 1\rangle_+ - |2, 0\rangle_+)/2. \quad (4)$$

These states are eigenstates of \hat{S}_y with eigenvalues $2(n - 1) - N$ and are circularly polarized since they are invariant under any geometrical rotation around the wave vector. If the field represented by the left (right) index in the kets in Eqs. (2)–(4) stands for the oscillation in the horizontal (vertical) direction, then we can identify $|\circ_1^{(2)}\rangle$ and $|\circ_3^{(2)}\rangle$ as left- and right-hand polarized states (assuming a harmonic time dependence $e^{-i(\omega t - \mathbf{k}\cdot\mathbf{r})}$). Expanded in a circularly polarized mode basis (left, right), indicated by the subscript \circ , the states are $|\circ_1^{(2)}\rangle = |2, 0\rangle_\circ$, $|\circ_2^{(2)}\rangle = |1, 1\rangle_\circ$, and $|\circ_3^{(2)}\rangle = |0, 2\rangle_\circ$. The state $|\circ_2^{(2)}\rangle$ is symmetric with respect to the two oscillation modes. This was already noted in [10,11]. We suggest the state be called a neutrally polarized state (which should be distinguished from unpolarized light [6,9,10]). We see that just like classically polarized light, the left-hand polarized state will be transformed into the right-hand polarized state under a relative phase shift of π between the two modes, and vice versa. This can be accomplished with a $\lambda/2$ plate inserted at any angle. The neutrally polarized state is invariant under this transformation. (Similar rotational invariance, but in the context of two polarization-entangled states, has already been demonstrated [12].)

In classical optics the “complementary” polarization to circular is linear. A linearly polarized field has the property that vertical (horizontal) polarization evolves to horizontal (vertical) polarization upon a geometrical rotation of 90° . A set of states with similar properties can be constructed in manifold N as

$$|\zeta_k^{(N)}\rangle = \frac{1}{\sqrt{N+1}} \sum_{n=1}^{N+1} e^{i\pi(k-1)n/(N+1)} |\circ_n^{(N)}\rangle, \quad (5)$$

where $k = 0, 1, \dots, N$. The principle of construction is identical to the construction of Pegg-Barnett phase states and relative phase states [13]. (A somewhat similar construction for excitation manifold 2 is proposed in [14] to achieve a ternary logic based on two-photon states.) All the $N + 1$ states in the set are mutually orthonormal. Furthermore, state $|\zeta_k^{(N)}\rangle$ evolves into state $|\zeta_{k+1}^{(N)}\rangle$, where $k + 1$ should be taken modulus $N + 1$, upon geometrical rotation by an angle $\pi l/(N + 1)$ around the wave vector. In this sense the states are similar to classical linearly polarized fields. However, in contrast to their classical counterparts, the states do not have their electric-field oscillation confined to one spatial direction.

We note that any unitary rotation that leaves the circularly polarized states (2)–(4) invariant will transform the states defined by (5) to states with identical properties with the respect of rotation. Such a unitary transformation must have the form $\hat{U} = \exp(i\mu)|\circ_1^{(2)}\rangle\langle\circ_1^{(2)}| + \exp(i\nu)|\circ_2^{(2)}\rangle\langle\circ_2^{(2)}| + \exp(i\chi)|\circ_3^{(2)}\rangle\langle\circ_3^{(2)}|$. The choice $\mu = 0$, $\nu = \pi/2$, $\chi = 0$ (this particular choice will be motivated below) will generate the set of states

$$|\zeta_1^{(2)}\rangle = \frac{1 - i\sqrt{2}}{\sqrt{12}} |0, 2\rangle_+ - \frac{1}{\sqrt{2}} |1, 1\rangle_+ - \frac{1 + i\sqrt{2}}{\sqrt{12}} |2, 0\rangle_+, \quad (6)$$

$$|\zeta_2^{(2)}\rangle = \frac{\sqrt{2} + i}{\sqrt{6}} |0, 2\rangle_+ - \frac{\sqrt{2} - i}{\sqrt{6}} |2, 0\rangle_+, \quad (7)$$

$$|\zeta_3^{(2)}\rangle = \frac{1 - i\sqrt{2}}{\sqrt{12}} |0, 2\rangle_+ + \frac{1}{\sqrt{2}} |1, 1\rangle_+ - \frac{1 + i\sqrt{2}}{\sqrt{12}} |2, 0\rangle_+, \quad (8)$$

in the second excitation manifold. It is straightforward to show that geometrically rotating state $|\zeta_2^{(2)}\rangle$ by $\pi/3$ ($-\pi/3$) will make it evolve into the orthogonal state $|\zeta_3^{(2)}\rangle$ ($|\zeta_1^{(2)}\rangle$). The reason we made the particular choice $\mu = 0$, $\nu = \pi/2$, $\chi = 0$ is that $|\zeta_2^{(2)}\rangle$ can be generated from the state $|1, 1\rangle_+$ by the means of a phase plate.

The $|1, 1\rangle_+$ state can experimentally be generated by spontaneous parametric down conversion (SPDC). In this process a pump photon is converted into a pair of photons with lower energy. SPDC is possible in nonlinear, birefringent materials via the conservation of energy $\omega_p = \omega_s + \omega_i$ and momentum $\mathbf{k}_p = \mathbf{k}_s + \mathbf{k}_i$, where the subscripts refer to the pump (p) and the down-converted photons, usually denoted signal (s) and idler (i). There are two types of SPDC; in type-I SPDC the signal and idler have the same polarization, whereas in type-II SPDC they are orthogonally polarized.

Our experimental setup is outlined in Fig. 1. Similar setups have been used for other measurements of fringe visibility [12]. We use type-II SPDC so the state after

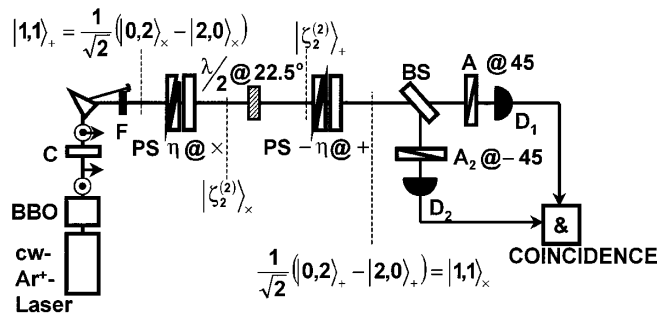


FIG. 1. A photon pair produced in BBO via type-II spontaneous parametric down-conversion impinges on a compensator plate C (used to make the two photons overlap in time) and then on a filter F (used to narrow the spectral width of the pair). A relative phase shift of $\eta = -\arccos(1/3)$ is applied in a basis rotated by 45° from the vertical, so the outgoing state is the $|\zeta_2^{(2)}\rangle_x$ state. A $\lambda/2$ plate inserted at 22.5° gives the same state in the horizontal and vertical basis. A second relative phase shift of $-\eta$ is then applied between the horizontal and vertical basis. A symmetric beam splitter is followed by two analyzers aligned at 45° and 135° . The two detectors are connected to a coincidence circuit that projects out the state $|1,1\rangle_x$.

the nonlinear crystal is $|1,1\rangle_+$. In a linear basis rotated by 45° around the wave vector (indicated by the subscript \times), this state can be expressed $(|0,2\rangle_x - |2,0\rangle_x)2^{-1/2}$. Inserting a variable birefringence component (in our case a birefringent prism pair) with its principal axes at 45° and 135° , a state of the form $[|0,2\rangle_x - \exp(i\eta)|2,0\rangle_x]2^{-1/2}$ will be generated. Specifically the state $|\zeta_2^{(2)}\rangle_x$ is generated if the relative phase shift (indicated with the letters PS in the figure) is chosen to be $\eta = -\arccos(1/3) \approx -70.5^\circ$.

The generated state can subsequently be geometrically rotated by a $\lambda/2$ -plate. If the plate's fast axis is set at an angle $\alpha/2$ from the 45° direction, the state is geometrically rotated by α degrees, e.g., if the fast axis is set at 22.5° from the vertical, the state $|\zeta_2^{(2)}\rangle$ is rotated so that the bases refer to the horizontal and vertical directions. (It should be noted that a $\lambda/2$ plate does not strictly correspond to a geometrical rotation. For a general state expressed in some linear basis it corresponds to a pure geometrical rotation of the basis, followed by a relative phase shift of π in the rotated basis. However, for the state $|\zeta_2^{(2)}\rangle$ which only has components with an even number of photons, 0 or 2, this particular phase shift is inconsequential as pointed out in the paragraph following Eq. (4). In other cases it can be canceled with the help of a second $\lambda/2$ -plate, succeeding the first, oriented with its principal axes in the rotated basis directions.)

To detect the state we transform it to a $|1,1\rangle$ state which subsequently can be detected using coincidence counting. To this end a second relative phase shift of $-\eta$ is applied in the horizontal-vertical basis. If the state in this basis prior to the second relative phase shift is $|\zeta_2^{(2)}\rangle_+$, the state after the phase shift is $(|0,2\rangle_+ - |2,0\rangle_+)2^{-1/2}$. Following the

second relative phase shift is a nonpolarizing beam splitter (BS) and two polarization analyzers (A_1 and A_2), one for each arm. The analyzer in the transmitted arm is oriented at 45° and the analyzer in the reflected arm is oriented at 135° . When the $\lambda/2$ plate is inserted at 22.5° we see a maximum in the coincidences since at this angle we project the state $|\zeta_2^{(2)}\rangle$ onto itself. By rotating the $\lambda/2$ plate to $22.5 + \beta/2$ the coincidence rate is proportional to

$$|\langle \zeta_2^{(2)} | \exp(i\beta \hat{S}_y) | \zeta_2^{(2)} \rangle|^2 = \frac{\sin^2(3\beta)}{9 \sin^2(\beta)}. \quad (9)$$

At $\beta = 60$ ($\beta = -60$) degrees this projection probability is zero since the state $|\zeta_2^{(2)}\rangle$ evolves into the orthogonal state $|\zeta_3^{(2)}\rangle$ ($|\zeta_1^{(2)}\rangle$).

In the experiment our light source is a single-mode cw argon-ion laser with a wavelength of 351.1 nm. The SPDC crystal is β -BaB₂O₄ (BBO) with a length 0.5 mm. The crystal is aligned so that collinear orthogonally polarized photon pairs with equal energy are produced. The pump is separated from the photon pairs by a dispersion prism. Pinholes and one interference filter (F), with a bandwidth of 10 nm and centered at 702.2 nm, further select photon pairs that have the same energy and that travel in the same spatiotemporal mode. Because of the linear dispersion of the crystal the horizontally and vertically polarized photons are separated in time. A birefringent crystalline quartz plate (C) is used to compensate for the linear dispersion. The detectors (D_1 and D_2) are actively quenched single-photon counting modules (EG&G SPCM).

The experimental data presented in Fig. 2 is just as collected (raw data) without any background subtraction. The achieved visibility is 90%. The minima are achieved for rotations of approximately $45^\circ \pm 60^\circ$ from the vertical as predicted (corresponding to a rotation of the $\lambda/2$ plate by $22.5^\circ \pm 30^\circ$ from the vertical). The nonperfect visibility is explained by the fact that the measurement requires careful mode matching and alignment of 7 birefringent and 4 nonbirefringent optical components. Furthermore, the

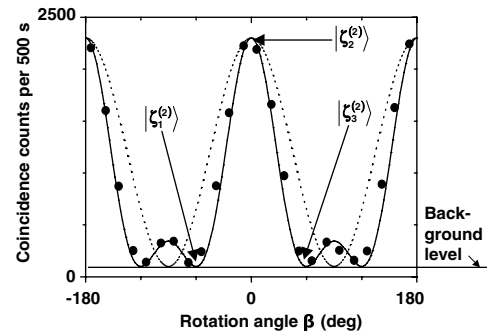


FIG. 2. Experimentally measured values of the coincidences as a function of twice the rotated angle of the $\lambda/2$ plate from its original position at 22.5° . At $\beta = \pm 60^\circ$, the coincidence rate is reduced to the background level. The dotted line indicates the behavior of a classical state under the same geometrical rotation.

compensator plate gives imperfect dispersion cancellation between the vertically and horizontally polarized photon. The state is then in a mixture between the desired state and the states $|0, 1\rangle_+$ and $|1, 0\rangle_+$. These latter states slightly deteriorate the quality of the measurement.

The experiment described above is an experimental demonstration of Heisenberg-limited polarimetry, although not the first [12]. The rotation sensitivity of the states defined by Eq. (5) scales as $\pi/(N + 1)$, clearly exceeding the standard quantum limit $\propto 1/\sqrt{N}$. A problem with the scheme is that the states are highly entangled. Therefore, in higher excitation manifolds, highly nonlinear optical components will be needed to generate the states from, e.g., two-mode number states. This is a difficulty this scheme shares with many other applications where entanglement is used to optimize the device performance, such as quantum computing.

Let us now briefly discuss the difference between our formalism and the traditional Stokes operator formalism. Our starting point was finding the eigenstates of the generator of geometrical rotation \hat{S}_y . From these, we constructed the eigenstates of a complementary operator through Eq. (5). It is clear that the set of states is associated with a Hermitian operator since in each excitation manifold the set is complete. The complementary nature of the operator follows from the fact that $|\langle \sigma_k^{(N)} | \zeta_l^{(N)} \rangle|^2 = 1/(N + 1) \forall k, l = 1, 2, \dots, N + 1$. Complete knowledge of the circular polarization state of a field precludes any knowledge of its complementary polarization state (all outcomes are equally probable), and vice versa. It is also clear that the latter is not true for the eigenstates of the Stokes operators, except in manifold $N = 1$. The state $|0, 2\rangle_+$ is a linearly polarized state and is an eigenstate of $\hat{S}_z = \hat{a}^\dagger \hat{a} - \hat{b}^\dagger \hat{b}$. However, the fact that $|\langle \sigma_2^{(2)} | 0, 2\rangle_+|^2 = 1/2 \neq 1/3$ shows that \hat{S}_y and \hat{S}_z are not complementary in spite of the fact that they do not commute. Similarly, $|\langle \zeta_2^{(2)} | 0, 2\rangle_+|^2 = 1/2$, showing that \hat{S}_z is not complementary to the polarization operator that can be defined through Eqs. (6)–(8) either.

There are three noncommuting Stokes operators \hat{S}_x , \hat{S}_y , and \hat{S}_z , and one Stokes operator \hat{S}_0 which commutes with all the other three, defining a SU(2) algebra. So far, in our theory of polarization rotation, we have talked about only three Hermitian operators, \hat{S}_y , \hat{S}_0 , and the operator defined by its set of eigenstates (5). However, as shown in [15], in every manifold there exists (at least) one more operator, defined through the set of states (11), (17), and (36) in [15], which commutes with \hat{S}_0 . In excitation manifold $N = 1$ the three noncommuting operators exhaust the set of mutually complementary operators, they define a SU(2) algebra and therefore a choice can be made so that they coincide with the Stokes operators. However, in a manifold $N > 1$, defining a Hilbert space of dimension $N + 1$, there exist at least three, and sometimes (when $N + 1$ is a prime or a power of a prime [15]) as many as $N + 2$, mutually com-

plementary operators, all commuting with \hat{S}_0 . Therefore this algebra is richer than the SU(2) group. However, the operators' detailed properties, e.g., their commutation relations, will have to be left out of this paper due to space limitations. What is important to retain is that, except for one photon state, linearly polarized states and circularly polarized states are not eigenstates of complementary operators. The set of complementary polarization operators may include either \hat{S}_y or \hat{S}_z , but not both. In this paper we made the choice to start from \hat{S}_y , but we could just as well have started from \hat{S}_z . With the latter choice, the states corresponding to Eqs. (6)–(8) would have been the two-mode relative phase states [13].

Let us now briefly touch upon the difference between the classical and a quantum mechanical definition of polarized light. In classical physics, polarization is a manifestation of correlations between the *two* transverse field *modes* defined in an Euclidian space. Therefore only second order correlations are needed to fully classify the state of polarization. A quantized transverse field with a fixed number of photons N , on the other hand, is defined in a Hilbert space of dimension $N + 1$. In this case, the polarization is a manifestation of correlations between $N + 1$ *quantum states*. Therefore, if $N > 1$, the quantum description offers more degrees of freedom than the classical description. Just as it is possible to use quantized fields to improve the resolution of interferometers, it is possible to use quantized fields to increase the resolution of polarimetry.

Finally, it may be noteworthy to point out that the quantized polarization states derived in this paper defy the correspondence principle. They will be increasingly nonclassical as the excitation N increases. They share this feature with many modern applications of quantum mechanics such as quantum computers. The reason is that the states are maximally entangled irrespectively of their excitation, and therefore lack classical counterparts.

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*Electronic address: gunnarb@ele.kth.se

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