Entanglement contains one of the most interesting features of quantum mechanics, often named quantum non-locality\cite{1, 2}. This means entangled states are not separable regardless of the spatial separation of their components. Measurement results on one particle of a two-particle entangled state define the state of the other particle instantaneously with neither particle enjoying its own well-defined state before the measurement.

So far experimental confirmation of entanglement has been restricted to qubits, i.e. two-state quantum systems including recent realization of three- \cite{3, 4} and four-qubit \cite{3, 4} entanglements. Yet, an ever increasing body of theoretical work calls for entanglement in quantum system of higher dimensions\cite{7, 8}. For photons one is restricted to qubits as long as the entanglement is realized using the photons polarization. Here we report the first realization of entanglement exploiting the orbital angular momentum of photons,

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which are states of the electromagnetic field with phase singularities (doughnut modes). This opens up a practical approach to multi-dimensional entanglement where the entangled states do not only consist of two orthogonal states but of many of them. We expect such states to be of importance for the current efforts in the field of quantum computation and quantum communication. For example, quantum cryptography with higher alphabets could enable one to increase the information flux through the communication channels [9, 10, 11].

Multi-dimensional entanglement is another possibility, besides creating multi-particle entanglement, for extending the usual two-dimensional two-particle state. Thus far there also have been suggestions [12, 13] and only a proof-of-principle experiment [14] for realizing higher order entanglement via multiport beam splitters. In the following we present an experiment in which we employed a property of photons namely the spatial modes of the electromagnetic field carrying orbital angular momentum to create multi-dimensional entanglement. The advantage of using these modes to create entanglement is that they can be used to define an infinitely dimensional discrete (because of the quantization of angular momentum) Hilbert space.

The experimental realization proceeded in the following two steps, also reflected in the organization of the present paper. First we confirmed that spontaneous parametric down-conversion conserves the orbital angular momentum of photons. This was done for pump beams carrying orbital angular momenta of $-\hbar$, $0$, and $+\hbar$ per photon respectively. In a further step it was shown that the state of the down-converted photons can not be explained by
assuming classical correlation in the sense that the photon pairs produced are just a mixture of the combinations allowed by angular momentum conservation. We proved that in contrast they are a coherent superposition of these combinations and hence they have to be considered as entangled in their orbital angular momentum.

For paraxial light beams Laguerre-Gaussian (LG) modes define a possible set of basis vectors (Figure 1). As predicted by Allen et al.\cite{15} and observed by He et al.\cite{16} LG modes carry an orbital angular momentum for linearly polarized light which is distinct from the angular momentum of photons associated with their polarizations. This external angular momentum of the photon states is the reason why they are often have been suggested for gearing micro machines and it was shown that they can be used as optical tweezers\cite{17,18,19}.

To demonstrate the conservation of the orbital angular momentum carried by the LG modes in spontaneous parametric down conversion we investigated three different cases for pump photons possessing orbital angular momenta of $-\hbar$, 0, and $+\hbar$ per photon respectively. As a pump beam we used an Argon-ion laser at 351 nm which we could operate either with a simple Gaussian mode profile ($l = 0$) or in the first order LG modes ($l = \pm 1$) after astigmatic mode conversion (for a description of this technique see Ref.\cite{22}). Spontaneous parametric down conversion was done in a 1.5 mm thick BBO crystal cut for type-I phase matching (that is both photons carry the same linear polarization). The crystal cut was chosen such as to produce down-converted photons at 702 nm at an angle of 4° off the pump direction.

The mode detection of the down-converted photons was performed for
Gaussian and LG modes. The Gaussian mode \((l=0)\) was identified using mono-mode fibers (Figure 2) in connection with avalanche detectors. All other modes have a larger spatial extension and therefore cannot be coupled into the mono-mode fiber. The LG modes \((l \neq 0)\) were identified using mode detectors consisting of computer generated holograms and mono-mode optical fibers (Figure 2).

Computer generated holograms often have been exploited in the past for creating LG modes of various orders.\[^{23}\] Our holograms were phase gratings 5 x 5 mm\(^2\) in size with 20 lines per mm which we first recorded on holographic films and bleached afterwards to increase the transmission efficiency (Figure 2). We made holograms which had one or two dislocations in the center and designed them to have their maximum intensity in the first diffraction order, so we could distinguish between LG modes \(l = -2, -1, 0, 1, 2\) using all holograms in the first diffraction order only for which order they have been blazed. For analyzing a LG mode with a negative index the holograms were just rotated by 180° around the axis perpendicular to the grating lines. The total transmission efficiency of all our holograms was about 80% and they diffracted 18% of the incoming beam into the desired first order. These characteristics were measured at 632 nm as a laser source at 702 nm was not available to us.

The diffraction efficiency is not the only loss that occurs. Also, we have to account for Fresnel losses at all optical surfaces (95% transmission), imperfect coupling into the optical fibers (70% for a Gaussian beam), non-ideal interference filters (75% center transmission), and the efficiency of the detectors (30%). A conservative estimate of all the losses yields an over-
all collection efficiency of 2 to 3 percent. Comparing the unnormalized 
\( (l_{\text{pump}} = l_1 = l_2 = 0) \) coincidence rates of about 2000 s\(^{-1}\) to the sin-
gles count rates of about 100,000 s\(^{-1}\) we deduce an efficiency of 2%, well in agreement with the above estimation.

The mode analysis was performed in coincidence for all cases where mode 
filter 1 was prepared for analyzing LG modes \( l_1 = 0, 1, 2 \) and mode filter 2 
for those with \( l_2 = -2, -1, 0, 1, 2 \). For analyzing a LG mode with mode 
index \( l = 0 \), i.e. a Gaussian mode, the dislocation of the hologram was 
shifted out of the beam path. The beam was sent through the border of 
the hologram where it acts as a customary grating without changing the 
photons angular momentum. The results are shown in Figure 3 for different 
values of orbital angular momenta of the pump beam. Within experimental 
accuracy coincidences were only observed in those cases where the sum of 
the orbital angular momenta of the down converted photons was equal to 
the pump beams orbital angular momentum. However the absolute count 
rates of these cases are not equal. This fact is most likely due to unequal emission probabilities of the photons into the different modes in the down 
conversion process.

These results confirm conservation of the orbital angular momentum in 
parametric down-conversion. The achieved signal to noise ratios were as 
high as \( V = 0.976 \pm 0.038 \) and \( V = 0.916 \pm 0.009 \) for pump beams with 
and without pump orbital angular momentum respectively. \( V \) is defined as 
\( V := \frac{I_{\text{out}} - I_{\text{in}}}{I_{\text{out}} + I_{\text{in}}} \), where \( I_{\text{in}} \) and \( I_{\text{out}} \) denote the maximum and the minimum of 
the coincidences with the dislocation of the hologram in and out of the beam 
path respectively.
It is important to mention that only by using a coincidence measurement we could show that the conservation of the orbital angular momentum holds for each single photon pair. In contrast, cumulative detection methods using many photons result in an incoherent pattern \cite{24} since each beam from parametric down-conversion by itself is an incoherent mixture. Therefore Arlt et al. \cite{24} using these classical detection methods which are in principle unsuitable at the single photon level were led to believe that the orbital angular momentum is not conserved in spontaneous parametric down-conversion.

Given this experimental verification of the orbital angular momentum conservation one may expect to find entanglement between the two photons produced in the conversion process. But for explaining the conservation of the orbital angular momentum the photons do not necessarily have to be entangled. It would be sufficient to assume classical correlation. However further experimental results showed that the two-photon state goes beyond classical correlation and indeed we were able to prove the entanglement for photon states with phase singularities.

In order to confirm entanglement one has to demonstrate that the two-photon state is not just a mixture but a coherent superposition of product states of the various Gaussian and LG modes which obey angular momentum conservation. For simplicity we restricted ourselves to superpositions of two basis states only. An important distinction between coherent superposition and incoherent mixture of Gaussian and LG modes is that the latter posses no phase singularity. This is because adding the spatial intensity distributions of these two modes will yield a finite intensity everywhere in the resulting pattern. In contrast, in a coherent superposition the amplitudes
are added and therefore the phase singularity must remain and is displaced to an eccentric location (Figure 4). It will appear at that location where the amplitudes of the two modes are equal with opposite phase. Therefore the radial distance of the singularity from the beam center is a measure of the amplitude ratio of the Gaussian to the LG components whereas the angular position of the singularity is determined by their relative phase. Intuitively speaking the position of the dislocation with respect to the beam is equivalent to the orientation of a polarizer.

As discussed in Figure 2 such superpositions of LG and Gaussian modes can experimentally be realized by shifting the dislocation of the hologram out of the center of the beam by a certain small amount. Hence in order to detect a photon having an orbital angular momentum which is a superposition of the Gaussian and the LG mode the hologram was placed in a position such that the dislocation was slightly displaced from the beam center. In the intensity pattern these modes possess an eccentric singularity (Fig. 4). For demonstrating the entanglement we therefore shifted one of the holograms and scanned the Gaussian mode filter on the other side while recording the coincidences.

The results shown in Fig. 4 clearly verify the correlation in superposition bases of the LG (l=±2) and Gaussian (l=0) modes. A closer analysis shows that there are two conditions necessary to obtain the measured curves. First the shifted hologram has to work as described above and second the source must emit an angular momentum entangled state. Assume that the source only emits classically correlated but not entangled singularities. Then on the side with the shifted hologram the various terms of the classical mixture
would be projected onto a state with displaced singularity leaving the total state again in a mixture. Respecting the conservation of angular momentum we would then have to sum the probabilities of the various components on the other side resulting in a coincidence pattern not containing any intensity zeroes. Such a coincidence pattern would also be observed if a shifted hologram together with a mono-mode detector would not be able to analyze for superposition states.

An entangled state represents both correctly the correlation of the eigen-modes and the correlations of their superpositions. Having experimentally confirmed the quantum superposition for \( l=0 \) and \( l=\pm 2 \), it is reasonable to expect that quantum superposition will also occur for the other states. Nevertheless, ultimate confirmation of entanglement will be a Bell inequality experiment generalized to more states \([25]\). Such an experiment will be a major experimental challenge and it is in preparation in our laboratory.

For a pump beam with zero angular momentum the emitted state must then be represented by

\[
\psi = C_{0,0}|0\rangle|0\rangle + C_{1,-1}|1\rangle|-1\rangle + C_{-1,1}|-1\rangle|1\rangle + C_{2,-2}|2\rangle|-2\rangle + C_{-2,2}|-2\rangle|2\rangle + \ldots
\]

(1)

since the LG modes form a infinite dimensional basis. Here the numbers in the brackets represent the indices \( l \) of the LG modes and the \( C_{i,j} \) denote the corresponding probability amplitude for measuring \( |i\rangle|j\rangle \). The state (1) is a multi-dimensional entangled state for two photons, which in general will also contain terms with \( p \neq 0 \). It means neither photon in state (1) possesses a well-defined orbital angular momentum after parametric down conversion. The measurement of one photon defines its orbital angular momentum state.
and projects the second one into the corresponding orbital angular momentum state.

It is conceivable to extend these states to multi-dimensional multi-particle entanglement in the future. A steadily increasing body of theoretical work calls for entanglement of quantum systems of higher dimensions [7, 8]. These states have applications in quantum cryptography with higher alphabets and in quantum teleportation. Since such states increase the flux of information it is conceivable that they will be of importance for many other applications in quantum communication and quantum information too. Also the possibility to use these photon states for driving micro machines and their application as optical tweezers make them versatile and auspicious for future technologies [17, 18, 19].

After completion of the experimental work presented here related theoretical work was brought to our attention [20, 21].

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Captions:
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Figure 1: The wave front (top) and the intensity pattern (bottom) of the simplest Laguerre Gauss \( (LG_p^l) \) or doughnut mode. The index \( l \) is referred to as the winding number and \( (p + 1) \) is the number of radial nodes. Here we only consider cases of \( p = 0 \). The customary Gaussian mode can be viewed as LG mode with \( l = 0 \). The handedness of the helical wave fronts of the LG modes is linked to the sign of the index \( l \) and can be chosen by convention. The azimuthal phase term \( e^{il\phi} \) of the LG modes results in helical wave fronts. The phase variation along a closed path around the beam center is \( 2\pi l \). Therefore in order to fulfill the wave equation the intensity has to vanish in the center of the beam.

Figure 2: Experimental setup for single-photon mode detection. After parametric down conversion each of the photons enters a mode detector consisting of a computer generated hologram and a mono-mode optical fiber. By diffraction at the hologram the incoming mode undergoes a mode transformation in a way that a LG mode can be transformed into a Gaussian mode. Since it has a smaller spatial extension than all LG modes, only the Gaussian mode can be coupled into the mono-mode fiber. Thus observation of a click projects the mode incident on the fiber coupler into the Gaussian mode. The hologram is a phase grating with \( \Delta m \) dislocations in the center blazed for first order diffraction. An incoming Gaussian laser beam passing through the dislocation of the hologram is diffracted by the grating and the \( n \)-th diffraction order becomes a LG mode with an index \( l = n\Delta m \) and vice versa. Intuitively speaking the phase dislocation exerts a “torque” onto the diffracted beam because of the difference of the local grating vectors in the upper and lower parts of the grating. This “torque” depends on the diffrac-
tion order \( n \) and on \( \Delta m \). Consequently the right and left diffraction orders gain different handedness. Reversing this process a photon with angular momentum \( \Delta m \hbar \) before the grating can be detected by the mono-mode fiber detector placed in the first diffraction order. A photon with zero angular momentum (Gaussian mode) is detected by diffracting the beam at the border of the hologram faraway from the dislocation. All our measurements were performed in coincidence detection between the two down-converted photons.

Figure 3: Conservation of the orbital angular momentum.

Coincidence mode detections for photon 1 and photon 2 in 15 possible combinations of orthogonal states were performed. This was done for a pump beam having an orbital angular momentum of \(-\hbar\), 0, and \(+\hbar\) per photon respectively. Coincidences was observed in all cases where the sum of the orbital angular momenta of the down converted photons were equal to the pump beams orbital angular momentum. The coincidence counts for each fixed value of the orbital angular momentum of photon 1 was normalized by the total number of coincidences varying the orbital angular momentum of photon 2.

Figure 4:

Experimental evidence of entanglement of photon states with phase singularities: The dislocation of the hologram in the beam of photon 1 is shifted out of the beam center step by step (top, middle, bottom). In these positions this hologram together with the mono-mode fiber detector projects the state of photon 1 into a coherent superposition of LG and Gaussian modes. The mode filter for photon 2 with the hologram taken out makes a scan of the second photons mode in order to identify the location of its singularity with
respect to the beam center. The coincidences show that the second photon is also detected in a superposition of the LG and the Gaussian mode. Classical correlation would yield a coincidence picture which is just a mixture of Gaussian and LG modes. In that case the intensity minimum would remain in the beam center but would become washed out. In the experiment a hologram with two dislocations in the first diffraction order was used. This results in a superposition of the l=0 and l=2 modes.
Type-1 Hologram

Mono-Mode Optical Fibres

Detectors and Coincidence Circuit

Normalized Coincidences