The development of convenient sources of entangled photons is an important task in quantum information [1]. Entangled photons have been used to realize fundamental quantum information procedures such as quantum key distribution [2, 3, 4], quantum teleportation [5, 6, 7] and entanglement swapping [8]. The latter task is an essential element of quantum repeaters [9], which would allow the distribution of entanglement over very long distances. Recently entangled photons were also used to implement simple quantum logic gates [10].

The standard source of entangled photons at the moment is parametric down-conversion [11], which is based on the conversion of pump photons into pairs of photons inside a non-linear optical crystal. An important drawback of down-conversion sources is the fact that they cannot be made to produce exactly one pair of photons. They always generate a statistical distribution of pairs. If the probability to create a single pair with a given pump pulse is \( p \), then there is a probability of order \( p^2 \) to create two or more pairs with the same pump pulse. This feature of down-conversion sources leads to limitations on their performance for various quantum information procedures, such as teleportation [11], quantum cryptography [12] and entanglement purification [13]. It would thus be very desirable to have a convenient source of individual pairs of entangled photons, where one can be sure that no more than one pair is emitted.

A natural approach towards realizing such sources is to use photonic cascades from atoms or semiconductor quantum dots. Atomic cascades were used to produce polarization-entangled photons in the first tests of Bell inequalities [14]. Quantum dot sources are attractive because they are compact and can be fairly easily integrated into semiconductor microcavity structures to enhance the probability for emission of the photons into a well-defined mode. These features have recently been demonstrated for single-photon sources [15]. There is a recent proposal for a quantum dot source of single pairs of polarization entangled photons [16], based on the biexciton-exciton cascade. However, the generation of polarization entanglement with this source requires the two intermediate exciton states with different spin to be exactly degenerate, which is not the case for currently available quantum dots, due to their lack of exact rotational symmetry around the direction of growth. In consequence, current quantum dots can emit polarization-correlated, but not polarization-entangled, photons [17].

Creating single time-bin entangled photon pairs

When a single emitter is excited by two phase-coherent pulses with a time delay, each of the pulses can lead to the emission of a photon pair, thus creating a “time-bin entangled” state. Double pair emission can be avoided by initially preparing the emitter in a metastable state. We show how photons from separate emissions can be made indistinguishable, permitting their use for multi-photon interference. Possible realizations are discussed. The method might also allow the direct creation of \( n \)-photon entangled states \((n > 2)\).

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Here we propose to create single pairs of time-bin entangled photons from single emitters such as atoms or quantum dots. Time-bin entanglement was first introduced for down-conversion sources in Ref. [18], based on the principle of Ref. [19]. It requires a source that can generate a pair of photons at two different well-defined times. This creation has to happen coherently, such that no information about the time of emission of the photon pair is stored anywhere in the emitting system or in the environment. Under this condition, the generated state is a superposition of both photons having been emitted at the earlier or later time, of the schematic form

\[
|\text{early}\rangle|\text{early}\rangle + |\text{late}\rangle|\text{late}\rangle
\]

which is clearly an entangled state. Methods for detecting and using this form of entanglement were first described in Ref. [18]. In recent years time-bin entanglement has been successfully used to implement various quantum information tasks [4, 7, 20]. It is particularly well suited for long-distance transmission in optical fibers because it is insensitive to polarization fluctuations.

Our proposed source emits at most one time-bin entangled photon pair. This property is achieved by using a single two-photon emitter, which is initially prepared in a metastable state to eliminate the possibility of creating two pairs, as we will now explain. Fig. 1 shows the required level scheme. There are three levels \( e, i \) and \( g \) in a cascade configuration, plus a metastable excited state \( m \). If the system is prepared in the excited state \( e \), it quickly emits two photons, one each from the transitions \( e \rightarrow i \) and \( i \rightarrow g \). For entanglement generation, the system is first promoted to the metastable state \( m \). Then the system is made to interact with two pump pulses that are

FIG. 1: Level scheme for the proposed source of single time-bin entangled photon pairs.
in resonance with the transition from \(m\) to \(e\). The two pulses have a fixed time delay and relative phase. The first pulse excites the system to \(e\) with a probability \(p_1\). If the system is excited, it quickly emits a pair of photons and goes to the state \(g\). In this case, the second pump pulse has no effect, because it is far out of resonance. Starting from the metastable state \(m\) instead of the ground state \(g\) thus ensures that at most one pair of photons is emitted by the source. The second pulse can excite the system from \(m\) to \(e\) with probability \(p_2\). Thus, if the system did not emit a pair of photons after the first pulse, it can emit a pair after the second pulse. Provided that the pumping is on resonance and that there is no coupling to the environment, no information about the time of emission of the photons remains in the system. The process thus creates a pair of photons in a superposition of having been emitted at two different points in time. Schematically, the described procedure creates an entangled two-photon state

\[
\psi = \sqrt{p_1} |\text{early}\rangle |\text{early}\rangle + e^{i\phi_\rho} \sqrt{(1-p_1)p_2} |\text{late}\rangle |\text{late}\rangle, \tag{2}
\]

where \(\phi_\rho\) is the relative phase between the two pump pulses.

The squared norm of the state \(\psi\) describes the overall probability to produce a photon pair. For \(p_1 = 1/2\) and \(p_2 = 1\) the source produces exactly one maximally entangled pair.

For a real system, the schematic wave functions of Eqs. \(^{\text{i}}\)\(^{\text{12}}\) are not completely accurate. In particular, the components of the state referring to an individual cascade, which were denoted by \(|\text{early}\rangle|\text{early}\rangle\) and \(|\text{late}\rangle|\text{late}\rangle\) in the idealized discussion above, are a priori not of the simple product form suggested by the notation, but are themselves entangled states. Below we will explain why this is a potential problem for multi-photon interference experiments such as entanglement swapping, and show how it can be solved. First we describe the nature of the unwanted entanglement. It arises because there is a time ordering of the two transitions in a cascade - the \(e \to i\) transition has to occur before the \(i \to g\) transition. This leads to a temporal correlation between the two photons.

The two-photon wave function referring to a single cascade is \(^{\text{21, 22}}\)

\[
\psi(t_A, t_B) = 2\sqrt{\Gamma_A\Gamma_B} e^{-\Gamma_A t_A} \theta(t_A) e^{-\Gamma_B(t_B-t_A)} \theta(t_B-t_A). \tag{3}
\]

Here the indices \(A, B\) refer to the first and second photon from the cascade, \(t_{A,B}\) are the emission times (relative to the excitation at \(t = 0\)), and \(\Gamma_{A,B}\) the decay rates for the two transitions (cf. fig. 1). This wave function is entangled due to the presence of the factor \(\theta(t_B-t_A)\) that describes the temporal ordering of the emission events in the cascade. (The function \(\theta(x)\) is equal to \(1\) for \(x \geq 0\) and equal to \(0\) for \(x < 0\).)

This entanglement is not problematic for applications where only a single pair is used at any given time, such as quantum key distribution. However, it becomes an issue for entanglement swapping and related applications. Entanglement swapping between a pair of time-bin entangled photons labelled \(A_1\) and \(B_1\) and another time-bin entangled pair labelled \(A_2\) and \(B_2\) proceeds by detecting the photons \(B_1\) and \(B_2\) at an intermediate location in such a way that any information about their origin is erased, see fig. 4 of Ref. \(^{\text{18}}\) and the related discussion. Note that photons \(B_1\) and \(B_2\) have to come from the same stage of their respective cascades, otherwise they could be distinguished by their frequencies. Also, for single-pair sources as the one proposed here, in contrast to down-conversion sources \(^{\text{i1}}\), there is a vanishing probability that both photons detected at the intermediate location were emitted by the same source. The erasure of information about the origin of the two photons \(B_1\) and \(B_2\) can only be perfect if the arrival times of photons \(A_1\) and \(A_2\) contain no information about which of the two photons detected at the intermediate location was \(B_1\) and which was \(B_2\). This requires that there is no temporal correlation between the photons in each pair \((A_1B_1\) and \(A_2B_2\) apart from the time-bin entanglement, which means that the wave function \(\psi\) has to have product form.

The problem can be analyzed in terms of wave function overlap. For the entanglement swapping to work perfectly, the two photons \(B_1\) and \(B_2\) have to have perfect overlap. If photon \(B_1\) is entangled with \(A_1\) and photon \(B_2\) with \(A_2\) (in addition to the time-bin entanglement), then their quantum states given by tracing over the \(A\) photon in Eq. \(^{\text{4}}\) are not pure, but mixed: \(\rho_B = \sum_i p_i |\chi_i\rangle \langle \chi_i|\) (with \(\chi_i\) the eigenstates of \(\rho_B\)) is equal to \(\sum_i p_i^2\), since the overlap is one if the systems are both in the same eigenstate \(\chi_i\) (which happens with probability \(p_i^2\)), and zero otherwise. The latter expression is equal to \(Tr\rho_B^2\), whose departure from unity therefore gives the order of magnitude of the expected error in the entanglement swapping. A straightforward calculation using Eq. \(^{\text{4}}\) gives

\[
1 - Tr\rho_B^2 = \frac{\Gamma_B}{\Gamma_A + \Gamma_B}. \tag{4}
\]

From Eq. \(^{\text{4}}\) one sees that the error can be made small by making the decay rate for the first transition in the cascade, \(\Gamma_A\), much bigger than that for the second transition, \(\Gamma_B\). (It is easy to see intuitively that the correlation between the photons is very strong in the opposite case of \(\Gamma_B \gg \Gamma_A\) because then the uncertainty of their time difference is much smaller than the uncertainty of the emission of each photon individually.) If \(\Gamma_A\) is not much bigger than \(\Gamma_B\) in a given system, the decay rates can be influenced through external cavities, by having a more significant Purcell effect for the first transition.

The problem of unwanted temporal entanglement also exists for down-conversion sources, although the entanglement has a slightly different character. For these sources, which are very broad in energy since the emission process is extremely fast (on a fs timescale), the wave function \(\psi(t_A, t_B)\) can be made to have essentially product form by frequency filtering \(^{\text{5, 7}}\). This approach does not work for single emitters as discussed here because they are already very narrow in frequency. However, the fact that the emission can be comparatively slow for single emitters (on a ns timescale) may
allow an approach based on time-resolved detection, which could be used in some situations where tuning the decay times is not feasible, or to achieve even better indistinguishability. This second approach is based on noticing that the wave function $|\psi(t_A - t_B)|$ is of product form apart from the factor $\delta(t_A - t_B)$. Therefore, if one detects photons of type $A$ only in an interval $[0, T_1]$, and photons of type $B$ only in an interval $[T_2, T_2 + \Delta T]$, where $T_2 > T_1$, then the wave function is projected onto a perfect product state in the space defined by the two intervals. The timing of one photon then carries no information whatsoever about its partner. This remarkable property of the wave function $|\psi\rangle$ is a consequence of the exponential behavior of the photon emission.

We will now discuss the perspectives for an experimental realization of our proposal. As mentioned in the introduction, atomic cascades were used in the first Bell experiments [14]. Recently a single-photon source that generates photons in a well-defined radiation mode was realized with atoms in a high-finesse cavity [23]. Metastable states as required in the level scheme of fig. 1 certainly exist in atoms, for example the $D_{3/2}$ and $D_{5/2}$ states in the $^{40}$Ca$^+$ ions used in recent quantum information processing experiments [24]. These ion traps have also been integrated with high-finesse cavities [24]. Compared to quantum dots, the decay times in atoms are typically longer. For decay times of the order of ns and longer, the time-resolved detection described above becomes realistic. The most commonly used avalanche photodiodes have a time resolution of the order of hundreds of ps. However, new single-photon detectors based on superconductors can achieve a time resolution as fine as 18 ps [26]. We believe that realizing the proposed protocol with atoms is an interesting possibility that deserves further investigation.

Here we will focus on implementing the proposal with quantum dots. As mentioned in the introduction, a single-mode single-photon source was recently realized based on a quantum dot embedded in a micro-pillar shaped semiconductor micro-cavity [15]. The requirement that no information about the emission process may leak into the environment (which is essential for creating the time-bin entanglement, cf. above), is similar to the requirements for the emission of individual photons into a single temporal mode, which was demonstrated using the above-mentioned type of source in Ref. [27]. We have also mentioned before the recent proposal to create polarization entangled photon pairs via exciting the bi-exciton state in a quantum dot [12]. The resulting two-photon cascade via the single-exciton states is a natural candidate for our proposal as well. For the present source, one should select definite polarizations for the two photons, thus reducing the cascade to a single intermediate state. The correlation in polarization between the two emitted photons was demonstrated in Ref. [17].

The metastable state $m$ can be realized by using a dark exciton state, which is connected to the quantum dot ground state by an optically forbidden transition ($\Delta J_z = \pm 2$). The lifetime of such dark excitons are orders of magnitude larger than those of bright (optically allowed) excitons [28]. Preparing the system in the metastable state can simply be done by excitation to the conduction band with subsequent relaxation. The system will relax to a dark exciton state with a probability of order $1/2$. Exciting the system from the dark exciton state to the bi-exciton state requires driving a transition that is optically forbidden. However, it should be possible with realistic light intensities, since in real quantum dots there is always some coupling of dark and bright excitons due to valence band mixing. For example, radiation from the dark exciton was recently observed in II-VI quantum dots, showing that the transition is not strictly forbidden [29]. In the same experiment, the ratio of the lifetimes of the dark and bright exciton states was determined to be of order $100$ [30]. Similar results are expected for III-V quantum dots, where discrimination of the radiation from the dark exciton is more difficult because of a smaller splitting between the dark and bright exciton levels.

Typical lifetimes for the bi-exciton to exciton and the exciton to ground state transitions in III-V quantum dots (such as InAs) are of order $0.6$ ns and $1.4$ ns respectively [31]. The decay rate $\Gamma_A$ is thus larger than $\Gamma_B$ already without a cavity. In order to further reduce the unwanted temporal entanglement, in order to further reduce the unwanted temporal entanglement, this ratio could be significantly enhanced by embedding the quantum dot in a micro-cavity as in Ref. [15]. For example, for the above values of the decay rates, a Purcell factor of 20 for the first transition in combination with a Purcell factor of 2 for the second transition would already reduce the error due to the temporal correlations given by Eq. 3 to the level of 5 percent. A Purcell factor of order 6 was reported in Ref. [15]. It should be possible to achieve such a combination of Purcell factors with a single micro-cavity, since the frequency difference between the two transitions (which is due to the exchange interaction between the excitons in the bi-exciton state) is of the same order of magnitude as a typical micro-cavity linewidth. For example, the splitting between the exciton and bi-exciton lines in Ref. [31] is of order $3$ meV (or $2$ nm), whereas the cavity linewidth in Ref. [32] is of order $2$ meV. For an appropriately chosen quantum dot and micro-cavity, one should thus be able to bring the bi-exciton to exciton line into exact resonance with the cavity (by temperature tuning) to maximize the Purcell effect, while still achieving a smaller Purcell effect for the exciton to ground state transition.

Demonstration experiments with a quantum dot source would proceed in analogy to the experiments and setups described in Ref. [18], which were inspired by the proposal of Ref. [15]. For example, the setup for verifying the presence of time-bin entanglement contains three interferometers with identical path length difference and adjustable phases between the two paths. The first one is placed in the pump beam to create the two pump pulses from the output pulse of a mode-locked laser. There is also an interferometer at each observer station, $A$ and $B$. After their emission, the photons can be split by a suitable wavelength-sensitive element, and photon $A(B)$ is directed to observer $A(B)$. Interference occurs between the possibility that the photons were created by the first pump pulse, and then both took the longer path in the interferometers at $A$ and $B$, and the possibility that they were created
by the second pump pulse and then both took the shorter path in the interferometers at $A$ and $B$. As a consequence the coincidence probabilities between interferometer outputs at $A$ and $B$ vary sinusoidally with the combined phase $\phi_P - \phi_A - \phi_B$, where $\phi_P$ is the phase between the paths of the pump interferometer and $\phi_A(B)$ is the phase of the interferometer in $A(B)$. Ref. [18] describes how time-bin entanglement can be used for quantum key distribution, entanglement swapping and other multi-photon interference experiments. Several of these proposals were realized for down-conversion sources in Refs. [4, 7, 20]. The basic detection methods described in these papers are equally applicable to our proposed source.

Current experiments on light emission from quantum dots at the single photon level typically use III-V (InAs) quantum dots with exciton wavelengths around 900 nm [15, 17, 27, 31, 32]. However, InAs quantum dots have been shown to be capable of emitting around 1.3 $\mu$m [33], and recently even close to 1.5 $\mu$m [34]. There thus seems to be a real possibility of realizing a source of single time-bin entangled photon pairs at telecommunication wavelengths, which would be very valuable for long-distance quantum communication.

Another very interesting perspective is the possibility to create time-bin entanglement not only of photon pairs, but of larger numbers of photons directly from a single emitter, e.g. a quantum dot. This could be done by having the pump pulses in our scheme be in resonance with the tri-exciton or even higher excitonic states instead of the bi-exciton. This would create states of the form

$$|\text{early}\rangle_A|\text{early}\rangle_B|\text{early}\rangle_C|\ldots + |\text{late}\rangle_A|\text{late}\rangle_B|\text{late}\rangle_C|\ldots,$$

(5)

where again the ideal product form of the two terms could be achieved by tuning of the decay rates or by time-resolved detection. A three-photon cascade was demonstrated in Ref. [35] based on two-photon coincidence measurements. The demonstration and use of time-bin entanglement requires the collection and detection of all the photons from the cascade. Considering the external quantum efficiency of close to 40% reported for the single-photon source of Ref. [15], the coincident detection of three or more photons does not seem at all unrealistic with current single-photon detectors. A source that produces entangled $n$-tuplets of photons directly and efficiently would be of major interest for quantum information processing and quantum communication.

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