

Indistinguishable Photons from Independent Semiconductor Nanostructures

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We demonstrate quantum interference between photons generated by the radiative decay processes of excitons that are bound to isolated fluorine donor impurities in ZnSe/ZnMgSe quantum-well nanostructures. The ability to generate single photons from these devices is confirmed by autocorrelation experiments, and the indistinguishability of photons emitted from two independent nanostructures is confirmed via a Hong-Ou-Mandel dip. These results indicate that donor impurities in appropriately engineered semiconductor structures can portray atomlike homogeneity and coherence properties, potentially enabling scalable technologies for future large-scale optical quantum computers and quantum communication networks.

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Many schemes for scalable optical information processing rely on quantum interference between single photons generated by a large number of independent, triggered sources [1–4]. Sufficient homogeneity is certainly available in trapped atoms [5] and ions [6,7], but the scalability of such systems is challenging, in large part due to the requirements of laser cooling and trapping. On the other hand, solid-state quantum computers and quantum communication networks pose a different challenge: finding materials in which many independent atomlike levels efficiently generate identical single photons or scatter single photons identically. For example, quantum computation using only linear optics is possible [2], and all of the optical components may be implemented on a chip [8], but still lacking is a large array of triggered, independent sources of indistinguishable single photons.

Single photons sequentially generated from one semiconductor source, such as a self-assembled InAs quantum dot (QD) in a distributed-Bragg-reflector (DBR) [9] or photonic crystal (PC) [10] microcavity, have been used to demonstrate linear optics quantum computing concepts [11]. Such sources also show another important advantage of a semiconductor system: the possibility of electrical pumping [12]. However, quantum interference between photons emitted by independent QDs has not yet been realized, principally because self-assembled QDs are not identical and consequently show a broad distribution of emission wavelengths.

Here, we experimentally demonstrate indistinguishability of independent semiconductor sources based on the radiative recombination of excitons bound to neutral donors isolated by semiconductor nanostructures. In particular, we use the fluorine donor in ZnSe/ZnMgSe quantum wells (QWs) [13,14].

Isolated impurity-bound excitons are attractive as single-photon emitters since the impurity-bound-exciton-related emission has a well defined emission wavelength

with a small inhomogeneous linewidth. Further, the single electron spin of the neutral donor in its orbital ground state can be used as a long-lived matter qubit. Donors are better suited to such applications than acceptors [15] or isoelectronic impurities [16], since acceptor-bound holes suffer from the rapid, spin-orbit-related relaxation times and isoelectronic impurities do not have the needed metastable ground states. Several proposals [3] and a recent demonstration with trapped ions [7] demonstrate that indistinguishable photon emission into two orthogonal modes, in our case corresponding to emission from the neutral donor-bound exciton state (D^0X) to the two-electron spin states of a neutral donor (D^0), may entangle distant spins heralded by an unbunched, two-photon coincidence count; the degree to which these schemes are robust to interferometric stability and timing jitter is indicated by the width of the Hong-Ou-Mandel (HOM) dip, which we measure here. The electron spin states might also be interfaced with a photonic quantum communication bus through cavity enhancement of the transitions following a variety of proposals [4]. These proposals require high homogeneity in the optical transitions, which is also prerequisite for the HOM dip. Our demonstration therefore suggests that this system may be useful as an optically addressable quantum memory in future quantum repeaters for long-distance quantum communication or in quantum computers with chip-based photonic “wiring”.

The fluorine donor in ZnSe, depicted in Fig. 1(a), is particularly well suited for such applications, since nuclear decoherence of the electron spin may be suppressed by isotopic purification [17,18], unlike in III-V systems. Moreover, the spin-1/2 ^{19}F nucleus with 100% abundance provides even more potential for long-lived quantum memory, since electron entanglement could be transferred to the longer-lived nuclear spin via double resonance techniques while building larger entangled states [19]. At the same time, this system can be engineered using many of the

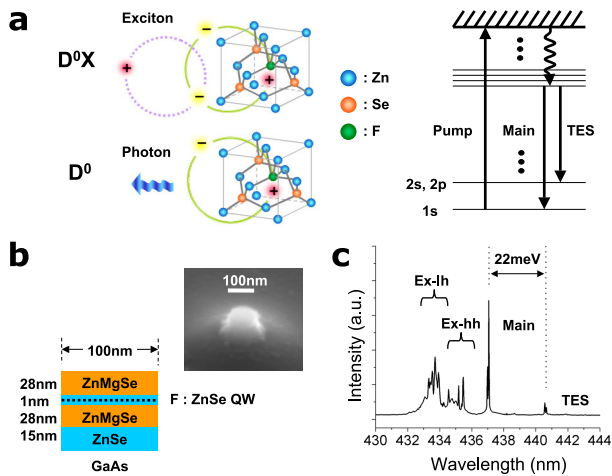


FIG. 1 (color online). (a) Schematic of a neutral fluorine donor in ZnSe (D^0) and the associated donor-bound exciton (D^0X) state, and corresponding energy diagram showing the above-band pump, the D^0X -to- D^0 transition (Main), and the two-electron satellite (TES). (b) The material composition of the single-photon device grown by molecular beam epitaxy, and a scanning electron micrograph of one device structure. (c) Emission spectra of a typical device, showing photoluminescence from the Main and TES transitions indicated by the energy diagram, as well as other related transitions (Ex-hh, Ex-lh).

sophisticated techniques of modern nanotechnology employed with III-V semiconductor alloy systems. Heterostructures and quantum wells (QWs) based on wide band-gap II-VI semiconductor alloys (ZnSe, MgSe, ZnS, MgSe) may be grown nearly defect-free on GaAs substrates [20]. Microdisk cavities and waveguide structures, for example, can be realized by combinations of dry-etching processes of ZnSe with selective wet-chemical etching of the GaAs substrate [21].

A detailed study of the properties of the fluorine donor in ZnSe/ZnMgSe QWs was performed in Ref. [13] and recently, low-threshold microcavity lasers have been realized with similar δ -doped QW structures in Ref. [14]. For the present study, the total multilayer structure shown in Fig. 1(b) was grown on a (100)-GaAs substrate and a 15 nm buffer layer of ZnSe, to guarantee optimal interface properties. The ZnSe QW has a thickness of 1 nm and is sandwiched between two 28-nm-thick ZnMgSe barrier layers with a magnesium concentration of about 13%. The fluorine δ doping in the central region of the ZnSe QW was performed with a sheet donor concentration of approximately $3 \times 10^{10} \text{ cm}^{-2}$. The nanofabrication of posts with 100 nm diameter was done with electron-beam writing and wet-chemical etching [21]; a scanning electron micrograph of one such post is also shown in Fig. 1(b). In the 100 nm-diameter post-nanostructures, there are only 2.4 fluorine donor atoms on average, which may be separately observed due to a small strain-induced inhomogeneous broadening.

Figure 1(c) shows the spectrum of a single donor-bound exciton emitted by a typical device. The excited levels of the D^0X complex correspond to different rotational states of the bound hole, but these states are nearly degenerate in zero magnetic field. Although the D^0X state predominantly relaxes to the $1s$ D^0 state, there is a finite probability that the state relaxes to any of the excited ($2s$, $2p$, etc.) D^0 states, predicted by a simple hydrogenlike model. These two-electron satellite (TES) transitions can be observed about 22 meV below the main line, and serve to verify that these transitions are due to neutral-donor-bound excitons.

From many devices of varying properties, we have chosen two for this study, denoted A and B, with spectra shown in Fig. 2. The devices are excited above the band gap using a 3 ps pulse from a frequency-doubled Ti:sapphire mode-locked laser. The spectra from these particular devices are shifted to shorter wavelengths than those in Fig. 1(c) due to slightly different magnesium content and strain status in the chip. Both devices emit at the same wavelength of 429.06 nm within the 0.01 nm resolution limit of the spectrometer. By fitting the spectra to Lorentzian line shapes, the two center wavelengths are found to be identical within an accuracy of 1.5 GHz. Although not all devices we observe are as homogeneous as this pair, the probability of finding such a pair vastly exceeds that for QDs. This probability could be further enhanced with small amounts of local tuning, which may be possible via local heating or Stark shifting [22]. The right insets of Fig. 2 show the normalized number of coincident photon counts versus delay τ between two photodetectors following a 50/50 beam splitter [$g^{(2)}(\tau)$].

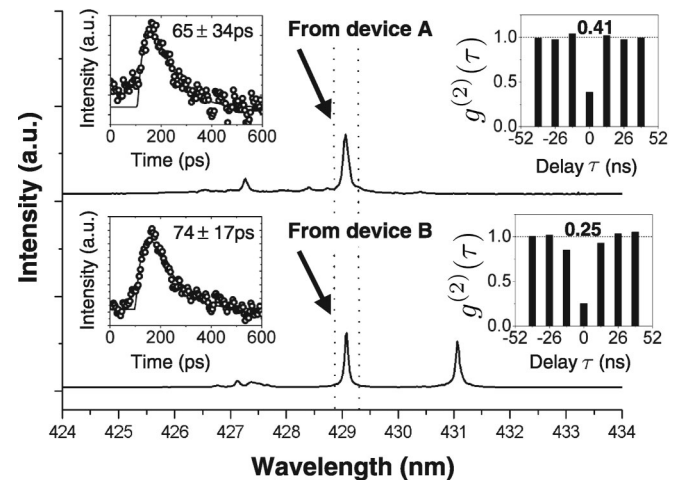


FIG. 2. Emission spectra of device A and device B under pulsed above-band-gap (410 nm) excitation. The arrow notes lines from both spectra showing the same wavelength 429.06 nm within the 0.01 nm resolution limit of the spectrometer. The arrows and dotted lines indicate the spectral window for the normalized photon correlation histogram (right insets) and time-dependent emission decay function with 5-point averaging (left insets).

This photon correlation histogram features a series of peaks separated by 13 ns, the repetition period of the pulsed laser. (The shape of each peak would be determined by the detector timing resolution, but we have integrated over this shape.) Due mostly to photon collection inefficiency, only one photon is counted per 10^4 pump pulses. The residual two-photon probability at zero delay is measured to be $g^{(2)}(0) = 0.41$ and 0.25 for devices *A* and *B*. This certainly indicates the emission of sub-Poissonian light from these devices (unlike laser or thermal sources), but there remain residual two-photon counts due to emission from the tail of the free-exciton spectrum as well as residual light from nearby devices. There is also slight antibunching evident at $\tau = \pm 13$ ns, likely due to a longer-lived nearby charge fluctuator [23].

Also in the left insets of Fig. 2 are the time-dependent decay functions for each device as measured by a streak camera, showing that each emitter has a lifetime of less than 100 ps. This is comparable to the lifetime of InAs QD excitons shortened by a DBR microcavity structure [9].

The experimental setup of the two-photon interference experiment is shown in Fig. 3. Many semiconductor nanostructure devices are fabricated in a matrix array configuration with a pitch of $10 \mu\text{m}$. The two particular devices *A* and *B* used in this study are separated by a distance of $42 \mu\text{m}$, which is within the $50\text{-}\mu\text{m}$ -diameter focus spot of the laser. The device array is cooled to 6 K in a coldfinger cryostat. The photons are collected by the same objective lens, but the light from one of the devices is separated using a cut mirror placed close to a Michelson-type interferometer. At the first polarizing beam splitter, the light beams from the two devices are recombined into the same spatial mode, but with orthogonal polarizations. This mode undergoes spectral filtering of a single emission line using a grating monochromator. When the half-wave plate is set to rotate the polarization by 45° , the two orthogonal incident polarizations are equally mixed at the second polarizing beam splitter, where quantum interference occurs. If the photons are identical, they will bunch into a single mode, reducing the probability of a coincidence photon count at

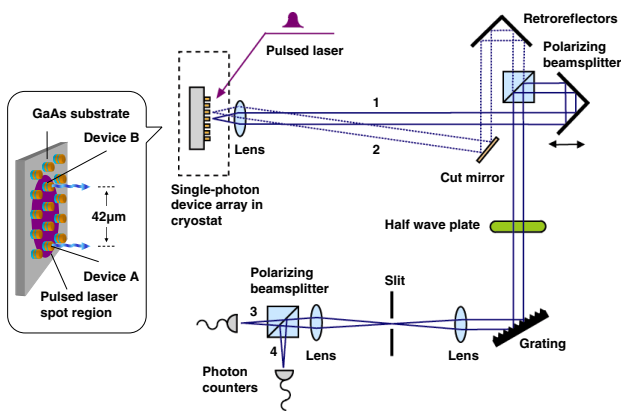


FIG. 3 (color online). Experimental setup. The inset shows the detailed configuration of the single-photon device matrix array.

the output. The outputs from this second polarizing beam splitter are detected using a pair of single-photon-counting avalanche photodiodes. The electronic signals from the photon counters are connected to a high-resolution counting card in a start-stop configuration, which generates a photon correlation histogram of the relative delay time between the arrival times of the photons on the detectors. This interferometer is different from a more typical apparatus using nonpolarizing beam splitters; however, this setup behaves equivalently while reducing the number of optical components for polarization and spectral filtering.

The experimental cross-correlation functions $g_{34}^{(2)}(\Delta t, \tau)$ (again with τ coarse grained over the timing resolution of the detectors) for devices *A* and *B* at two different interferometer delay times Δt are shown in Figs. 4(a) and 4(b). These functions are calculated by integration of each peak and normalizing by the average intensity. When the setup is in a 50/50 beam splitter configuration and the interferometer path delay Δt is zero, $g_{34}^{(2)}(0, 0)$ reaches a minimum of 0.68 with respect to the other peaks [Fig. 4(a)]. In contrast, when the path delay is 0.2 ns or larger, the two-photon coincidence probability $g_{34}^{(2)}(0.2 \text{ ns}, 0)$ is close to 1 [Fig. 4(b)]. We observe that the minimum $g_{34}^{(2)}(\Delta t, 0)$ is given when the interferometer delay time is zero with an accuracy of 3 ps. The observed visibility is 31%.

The HOM dip may be modeled in the following way. When the source successfully delivers a pair of photons from devices *A* and *B*, the two-photon state can be written as $|\psi\rangle = \int ds \int dt \alpha(s)\beta(t)a_A^\dagger(s)a_B^\dagger(t)|0\rangle$, where $a_A^\dagger(t)$ and $a_B^\dagger(t)$ are the photon creation operators for the two modes at time t , and $|0\rangle$ is the vacuum state. The time-dependent amplitude of the two-photon wave packets are defined by

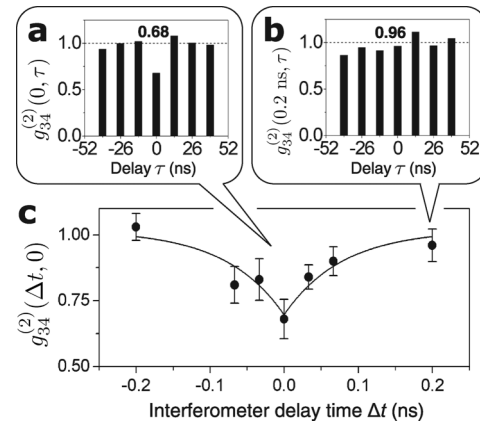


FIG. 4. Two-photon correlation functions $g_{34}^{(2)}(\tau)$ from the output of the interferometer. (a) Normalized photon correlation histogram of the time delays of the arrival of two photons on the photon counters, in the start-stop configuration, with interferometer path difference zero and (b) 0.2 ns. (c) Normalized two-photon coincidence count on the photon counters, plotted as a function of interferometer path-length difference. Error bars are based on the Poisson fluctuation in the number of counts on the uncorrected data.

the normalized complex-number functions $\alpha(t)$ and $\beta(t)$. As a function of interferometer path delay time, the normalized, coarse-grained coincidence count for the two detectors is given by [24]

$$g_{34}^{(2)}(\Delta t, 0) = \frac{1}{2} \left[1 - \text{Re} \int dt \int ds \langle \alpha^*(s) \beta^*(t + \Delta t) \times \beta(s + \Delta t) \alpha(t) \rangle \right] + g_{\text{back}}. \quad (1)$$

The last term g_{back} is the background coincidence count, which includes nonzero $g^{(2)}(0)$ for devices *A* and *B* and extra stray light from other sources [24], and does not depend on Δt . The brackets $\langle \dots \rangle$ average over many trials, resulting in an overlap term phenomenologically given as $I \exp(-|\Delta t|/\tau_c)$, where I is two-photon indistinguishability and τ_c is the two-photon correlation time. The indistinguishability I is upper-bounded by the ratio of the single-photon correlation time divided by twice the radiative lifetime. The correlation time τ_c is upper bounded by the spontaneous lifetime [24]. The observed indistinguishability and correlation times are in general reduced from these upper bounds by frequency and timing jitter [25].

The indistinguishability and correlation time are found by fitting Eq. (1) to the data in Fig. 4(c). This fit indicates that the excess background noise g_{back} is about 0.51 ± 0.06 . This is higher than the amount of two-photon events that would be observed from two sources showing the $g^2(0)$ values indicated by Fig. 2. The reason for the discrepancy is that the amount of filtering is different in the two experiments. The single-device experiments imaged each device onto a small pinhole to reduce background light from nearby devices to demonstrate single-photon operation. This spatial filtering is absent in the two-device experiment, increasing the number of two-photon events. (The spectral and polarization filtering are unchanged between the two experiments.) The spurious extra photons in the two-device experiment are not expected to show any interference, and so the reduction of the two-photon probability is attributed to the interference of those photons originating from the spectrally matched devices *A* and *B*.

The fit indicates a correlation time of $\tau_c = 74 \pm 38$ ps. This time is comparable to the measured spontaneous emission lifetimes of the devices *A* and *B*, suggesting nearly lifetime-limited sources; this suggests a potential for near-unity indistinguishability. However, the fit indicates an indistinguishability of $I = 65 \pm 13\%$, which is reduced from its ideal limit for several reasons. First, a small wavelength mismatch or wavelength jitter between the two sources may still exist, although it is unresolvable by our spectrometer. The indistinguishability is also reduced by random timing jitter resulting in part from the noisy relaxation from higher-energy states to the lowest bound-exciton state from which we collect emission [9]. The width of this jitter is estimated to be 40 ps from the fitting to the streak-camera data in Fig. 2, so this effect is small in comparison to dephasing time. The larger contri-

bution to the reduction of I is probably the imperfect overlap of the spatial modes of the two photons. In principle, this might be reduced by increased spatial filtering at the expense of the single-photon count rate.

In conclusion, we have experimentally demonstrated the two-photon quantum interference between indistinguishable photons emitted by independent semiconductor nanostructure devices with an observed visibility of 31% and a deduced indistinguishability of 65%. One route for improvement is the fabrication of microcavities based on DBR or PC structures, which can enhance the spontaneous emission rate and collection efficiency. Further integration of such cavities with on-chip waveguides may enable compact, solid-state technologies for efficient, scalable optical quantum computation and quantum communication.

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