Photon antibunching in the fluorescence of individual color centers in diamond

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We observed photon antibunching in the fluorescent light emitted from a single nitrogen-vacancy center in diamond at room temperature. The possibility of generating triggerable single photons with such a solid-state system is discussed. © 2000 Optical Society of America

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The security of quantum cryptography is based on the fact that each bit of information is coded on a single quantum object, namely, a single photon. The fundamental impossibility of duplicating the complete quantum state of a single particle prevents any potential eavesdropper from intercepting the message without the receiver's noticing.[1] In this context, the realization of an efficient and integrable light source that delivers a train of pulses that contain one and only one photon is an extremely challenging goal.[2] To reach it we need to address many issues, which range from achieving full control of the quantum properties of the source to easy handling and integrability of these properties in a practical quantum cryptography setup.

Several pioneering experiments have been made with a goal of obtaining single photon sources.[3–7] References 3 and 4 are based on the spontaneous emission of twin photons. One of the photons is used to open a time gate during which the twin photons can be observed. In these types of setup the emission time of the photon is random. In quantum cryptography–based communication systems it is desirable to deal with single photons synchronized to an external clock, namely, triggerable single photons.[5–7] In the experiment of Kim *et al.*[6] the regulation of the electron flux at the level of a single electron in a light-emitting mesoscopic $p-n$ junction is used. Experiments by de Martini *et al.*[5] and Brunel *et al.*[7] were based on the collection of fluorescent light emitted by a single dye molecule. The experiments reported in Refs. 6 and 7 require cryogenic apparatus (liquid-He temperature). The experiment reported in Ref. 5 may run at room temperature, but then the molecules are rapidly destroyed by photobleaching and must be renewed, e.g., by being immersed in a liquid solvent.[8] Another general limitation in these experiments is their very small collection efficiency ($\eta < 10^{-3}$), which means that fewer than 0.1% of the emitted photons are actually detected.

Generally speaking, a single emitting dipole will emit only one photon at a time. Using an appropriate pulse-excitation scheme, one can then produce only one photon per pulse.[7,9] In this Letter we report on the use of single color centers in diamond as single quantum emitters.[10] By investigating the quantum statistics of the fluorescent light we observed photon antibunching, which is a clear signature of the unicity of the emitting dipole. Photon antibunching has been observed in many experiments that involve, e.g., a single atom,[11] a single trapped ion,[12] and a single molecule.[13] With respect to these experiments we point out that our setup is particularly simple because it involves bulk diamond at room temperature and nonresonant excitation from an argon-ion laser at 514 nm, with a typical power of 10 mW. The collection efficiency that is now available is still low (see the discussion below), but straightforward improvements are possible. We consider thus that the present result is a first step toward the realization of a simple and efficient solid-state source for single photons.

The color center that we used is the nitrogen-vacancy (NV) defect center in diamond, with a zero phonon line at a wavelength of 637 nm. The defect consists of substitutional nitrogen and a vacancy in an adjacent site. A simplified level structure is a four-level scheme with fast nonradiative decay within the two upper states and within the two lower states. The excited-state lifetime is $\tau = 11.6$ ns.[14] We used 0.1 mm $\times$ 1.5 mm $\times$ 1.5 mm single [110] crystals of synthetic *Ib* diamond from Drukker International. Nitrogen is present in the crystal as an impurity. Vacancies are created by irradiation with 2-MeV electrons at a dose of $3 \times 10^{12}\ e^-/\mathrm{cm}^2$. The irradiation dose is chosen such that the density of vacancies is of the order of 1 $\mu\mathrm{m}^{-3}$. After irradiation, the crystals are annealed in vacuum at 850 °C for 2 h to form the NV centers. A remarkable property of these centers is that we could not observe any photobleaching: The fluorescence level remained unchanged after several hours of continuous laser irradiation of a single center in the saturation regime. A limitation of the system is the existence of shelving in a metastable singlet state. Shelving leads to the observation of photon bunching for a time scale longer than the lifetime of the center[8,13,15] and to a decrease of the fluorescence count rate, owing to the time spent by the centers in this long-lived state. Various techniques for laser-assisted deshelving have been proposed in the literature.[16] Such mechanisms should be considered in relation to a pulse-excitation scheme that will be required for true single-photon generation.

The experimental setup is depicted in Fig. 1. It is based on a homemade scanning confocal microscope. The green line ($\lambda = 514$ nm) of an Ar^+ cw laser is

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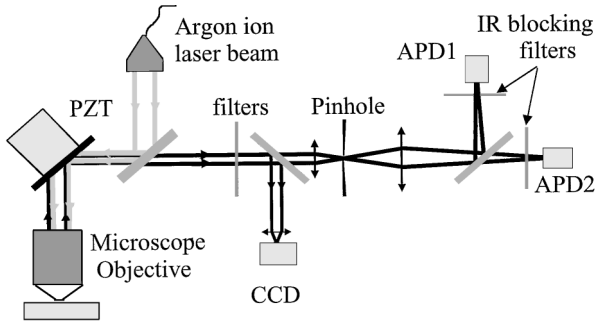

Fig. 1. Experimental setup. The sample fluorescence is excited and collected with a confocal-microscope setup. The intensity correlations are measured with two avalanche photodiodes (APD1 and APD2), a time-to-amplitude converter, and a multichannel analyzer. PZT, piezoelectric transducer.

focused on the sample by a high-numerical-aperture (1.3) immersion objective (Nachet 004279). A piezoelectric transducer–mounted mirror located just before the objective allows for an $x-y$ scan of the sample, and fine z scan is obtained with another piezoelectric transducer. The fluorescence (wavelength from 637 to 800 nm) is collected by the same objective and is separated from the excitation laser by a dichroic mirror. High-rejection-rate (10^{15}) long-pass filters are used to remove any green or blue light. We achieve confocal operation by imaging the sample onto a 50-μm pinhole with a 100× magnification. To investigate the fluorescence intensity correlation we use a Hanbury Brown–Twiss setup with two avalanche photodiodes (EG&G Model SPCM-AQR 13) and a 50/50 beam splitter. An IR filter and a pinhole are set in front of each detector to prevent optical cross talk between the detectors.[15] The time delay between pulses from the two photodiodes is converted by a time-to-amplitude converter into a voltage amplitude that is digitalized by the data-acquisition card of a computer.

Figure 2 shows a scan of the sample. Individual bright spots that correspond to NV centers appear clearly on the scan, with an apparent lateral resolution of 500 nm. However, the scan was obtained at a depth of 10 μm below the diamond surface, where the spherical aberration caused by the diamond–oil interface is not negligible, and reduces the focusing and the collection efficiencies. For an input power of 15 mW the typical signal from an individual center is $S_{\mathrm{det}} = 2000\ \mathrm{s}^{-1}$ on each photodiode. The evaluated overall detection efficiency is $\eta_{\mathrm{tot}} = 0.0014$, which can be split into $\eta_{\mathrm{geom}} = 0.08$ (geometrical collection angle), $\eta_{\mathrm{ab}} = 0.2$ (aberrations owing to the diamond–oil interface), $\eta_{\mathrm{opt}} = 0.25$ (optical transmission), $\eta_{\mathrm{BS}} = 0.5$ (beam splitter), and $\eta_{\mathrm{det}} = 0.7$ (detector quantum efficiency). The inferred emission rate from the center is thus $S_{\mathrm{det}}/\eta_{\mathrm{tot}} = S_{\mathrm{em}} = 1.4 \times 10^6\ \mathrm{s}^{-1}$. The difference between S_{em} and the fully saturated value $S_{\mathrm{rad}} = 9 \times 10^7\ \mathrm{s}^{-1}$ is attributed to the joint effects of shelving and nonsaturated excitation. It can be seen from Fig. 2(b) that peaks appear above a background level with a typical value $B = 4000\ \mathrm{s}^{-1}$. When the incident laser power is increased (in the range 2–20 mW), a saturation behavior appears on the signal fluorescence, whereas the background level increases linearly. The intensity correlation from the signal and the background also behaves differently, as we discuss in more detail below.

The raw coincidences $c(t)$ (right axis) and the correlation function $g^{(2)}(t)$ (left axis) are represented in Fig. 3 as functions of the delay t between two joint photodetection events. We used a slow (8-s response time) $x-y-z$ computerized servo lock to maintain the

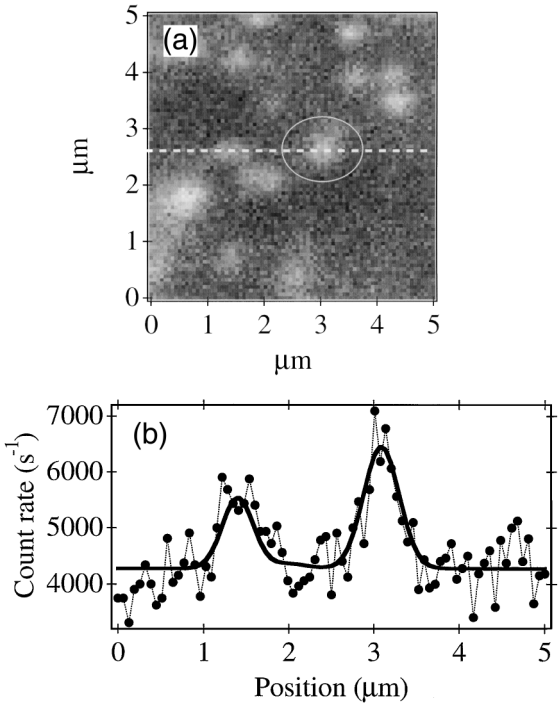

Fig. 2. (a) Confocal microscopy raster scan (5 μm × 5 μm) of the sample made ~10 μm below the diamond surface. The size of a pixel is 60 nm, the integration time per pixel is 32 ms, and the laser intensity impinging upon the sample is 15 mW. (b) Line scan along the dashed line in (a). The data are shown together with a Gaussian fit, which we use to evaluate the signal and background levels. Here we obtain $\rho = S/(S + B) = 0.34$.

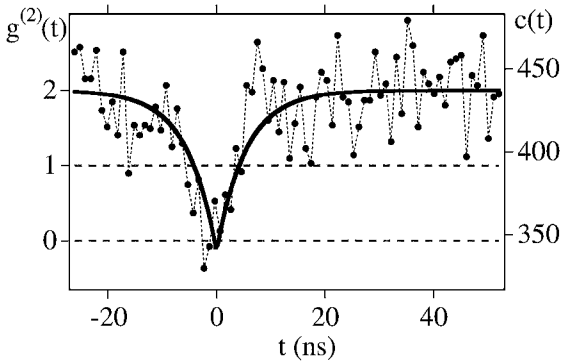

Fig. 3. Normalized correlation function $g^{(2)}(t)$ corrected for the random coincidences from the background (ρ = 0.34; see text). The data are for the NV center circled in Fig. 2(a). The actual number of coincidences is indicated at the right. The time bin is $w = 1$ ns, the integration time is $T = 11450$ s, and the single count rates are $N_1 = 5780\ \mathrm{s}^{-1}$ and $N_2 = 5990\ \mathrm{s}^{-1}$. Solid curve, an exponential fit to the data with the model described in Ref. 8.

fluorescence on the maximum of the NV center under study. Short-term and long-term drifts of the laser intensity are less than 10%. For a Poissonian light source the coincidence rate (in inverse seconds) in a time bin of width w is $N_1 N_2 w$, where $N_{1,2}$ are the count rates on each detector. The raw coincidence number $c(t)$ is thus normalized to that of a Poissonian source according to the formula $C_N(t) = c(t)/(N_1 N_2 w T)$. By stopping the scan at several centers (typically 1 μm away from a center of interest), we verified experimentally that the normalized coincidence rate from the background light is flat and equal to unity. We thus make the reasonable assumption that the photons emitted from the background are uncorrelated with the photons emitted from the center at the same location. This procedure allows us to correct for the random coincidences caused by the background light and obtain the $g^{(2)}(t)$ correlation function of the NV center:

$$g^{(2)}(t) = [C_N(t) - (1 - \rho^2)]/\rho^2, \qquad (1)$$

where $\rho = S/(S + B)$ is related to the signal-to-background ratio, which is measured independently in each experimental run. After this substraction procedure, the $g^{(2)}(t)$ correlation function shown in Fig. 3 goes to zero for $t = 0$, which is consistent with our claiming that there is only a single NV center in the fluorescence peak selected in Fig. 2. When n spatially unresolved centers are present, the value of the zero-time antibunching is $1 - 1/n$. Experimental runs with $g^{(2)}(0) > 0$ were also commonly observed and are typical of the brightest spots seen in Fig. 2. In Fig. 3 the nonunity value of $g^{(2)}(t)$ for $t > \tau$ is the bunching effect[8,13,15] that is due to the presence of a metastable singlet state.[16] We have observed experimentally that this bunching effect depends on the laser power, indicating that the 514-nm excitation light may also contribute to deshelving the metastable state. A quantitative study of these effects is under way.

In the present experiment the background level ($\rho = 0.34$) is too high for efficient use in quantum cryptographic systems. Spectrographic analysis of the fluorescent light reveals that one should obtain significant improvement by optimizing the spectral filters and dichroic mirrors to eliminate stray fluorescence near 600 nm, which does not arise from the NV center. Preliminary measurements show that the fraction of useful signal ρ can then reach values above 0.8. Other improvements can also be expected from a nonaberrating collection optics, from truly fluorescence-free immersion oil, and from the optimal choice of the diamond sample.

As a conclusion, we have observed photon antibunching from a simple setup involving a diamond crystal at room temperature. The total absence of photobleaching allowed us to lock the laser beam onto a single center during several hours. Color centers are included in a solid matrix, are easy to handle, and appear to be good candidates for realizing single-photon sources for quantum cryptography. We should obtain the ultimate efficiency for such a source by coupling the emitting dipole to a microcavity, so it will emit light in a single mode.[7] We point out, however, that a possible alternative method for achieving overall efficiencies in the range 10–20% may by the use of optimized wide-aperture collection optics.

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