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Nonclassical radiation from diamond nanocrystals

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The quantum properties of the fluorescence light emitted by diamond nanocrystals containing a single nitrogen-vacancy (NV) colored center are investigated. We have observed photon antibunching with very low background light. This system is therefore a very good candidate for the production of single photon on demand. In addition, we have measured a larger NV center lifetime in nanocrystals than in the bulk, in good agreement with a simple quantum electrodynamical model.

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Light sources able to emit individual photons on demand would be of great potential use for quantum cryptography [1,2]. A quantum computation scheme requiring such sources has also been proposed recently [3]. Considerable activity is thus dedicated to designing and implementing efficient, robust, room-temperature sources delivering a periodic train of pulses containing one and only one photon. These sources are based on the property of a single emitting dipole to emit only one photon at a time. When excited by a short and intense pulse, such an emitter delivers one and only one photon [4,5]. After pioneering experiments demonstrating photon antibunching [6–8] and conditional preparation of single-photon states [9,10], followed by first attempts to build triggered single photon sources [4,11,12], the present generation of experiments is concentrating on solid-state schemes better suited for practical use, such as single organic molecules [13–16], self-assembled semiconductor quantum dots [17,18], or semiconductor nanocrystals [19]. The successful candidate should work at room temperature, and be photostable.

A promising system for a robust single photon source is provided by individual nitrogen-vacancy (NV) color centers in diamond [20], that already permitted to observe photon antibunching under continuous excitation conditions in bulk crystals [21–23]. These color centers have the great advantage of being photostable and do not exhibit any photoblinking. The setup in these experiments is particularly simple, since it involves diamond samples at room temperature, and nonresonant excitation from a laser at 532 nm, with a typical power in the milliwatt range. However, a significant limitation in bulk diamond is that the light is emitted in a high-index material ($n_d=2.4$), that makes its efficient extraction difficult. Refraction at the sample interface leads to a small collection solid angle and to aberrations. A similar problem arises in semiconductor light-emitting devices [24].

In this paper, we show that diamond nanocrystals (typical size 40 nm) containing a single NV center offers a very efficient solution to circumvent these problems [25]. The subwavelength size of these nanocrystals renders refraction irrelevant. One can simply think of the nanocrystal as a point source emitting light in air. Furthermore, the small volume of diamond excited by the pump light yields very small background light. This is of crucial importance for single photon sources, since background light contributes to a nonvanishing probability of having two or more photons within the light pulse.

In addition, the width of the dip of the fluorescence intensity autocorrelation function $g^{(2)}(t)$ gives information about the lifetime of the emitters. Using this technique, we have observed an increase of the lifetime of a NV center in a nanocrystal compared to the bulk value [26]. This effect arises from the fact that, in a nanocrystal, the center can be considered as radiating in air, whereas it radiates in a medium of refractive index $n_d=2.4$ in bulk [27]. We will make the assumption, that is consistent with our observations, that the local field experienced by the NV center is the same in the bulk and in a nanocrystal. Our observed change in lifetime would thus be independent of local field corrections, that have been a controversial issue during the last decade [28–30].

The color center used in our experiments is the NV defect center in synthetic diamond, with a zero phonon line at a wavelength of 637 nm [20]. The defect consists in a substitutional nitrogen and a vacancy in an adjacent site. A simplified level structure is a four-level scheme with fast nonradiative decays within the two upper states and within the two lower states. The excited state lifetime in the bulk is $\tau_b = 11.6$ ns [31]. The nanocrystals come from synthetic diamond powder bought from de Beers. The defects are created by irradiation with 1.5 MeV electrons at a dose of $3 \times 10^{17} e^-/\text{cm}^2$, and annealing in vacuum at 850 °C during 2 hours. The density of NV centers created is then estimated to be of one in a 30-nm diameter sphere [20]. The nanocrystals are dispersed by sonification in a solution of polymer (Polyvinylpyrrolidone at 1 wt.% in propanol). This allows the disaggregation of the particles and their stabilization in a colloidal state. Centrifugation at 11 000 rpm for 30 mn allows us to select nanocrystal sizes of $d_0=90 \pm 30$ nm (measured by dynamical light scattering). The average number of NV centers in a nanocrystal has been evaluated to 8 (see below). Nanocrystals containing a single NV center should therefore have a size around $d_0/2=45$ nm, that lies in the lower wing of the size distribution. The nanocrystal solution is then spin coated at 3000 rpm on thin fused silica substrates. Evaporation of the solvent leaves a 30-nm thick film of polymer with the nanocrystals well dispersed on the surface. Their density was estimated to be around $0.02 \mu\text{m}^{-2}$. In most experiments we look at the centers from the other side of the plate, that is in contact with the oil of an immersion microscope lens (Nachet 004279, N.A.=1.3).

The experimental setup has been described in detail elsewhere [22]. It is based upon a homemade scanning confocal microscope, where the sample is excited using continuous-wave frequency-doubled YAG laser ($\lambda = 532$ nm). The fluorescence light (wavelength between 637 and 800 nm) is collected by the same objective and separated from the excitation laser by a dichroic mirror and filters. It can be sent either to a spectrometer, or to a standard intensity correlation setup using two avalanche photodiodes (EG&G, model SPCM-AQR 13), a time to amplitude converter (TAC), and a computer data acquisition board. The time bin is 1 ns, and a delay of 50 ns in one TAC input allows us to get data for negative time. A slow (8 s response time) x-y-z computerized servo-lock is used to maintain the fluorescence on its maximum for the observed center.

We have checked that the emission spectrum of a NV center in a nanocrystal at room temperature is identical to that in bulk to within our experimental accuracy (resolution 8 nm, signal to noise ratio larger than 10). It is also worth pointing out that the remarkable photostability of NV centers in bulk [20–22] is preserved in nanocrystals. Fluorescence has been observed in the saturation regime for hours without any photobleaching nor blinking.

Figure 1(a) displays a two-dimensional scan of a nanocrystal containing a single NV center. The resolution of 500 nm is that of the confocal microscope. The line scan in Fig. 1(b) shows that the signal (S) to background (B) ratio is very good with a value $S/B=20$. Note that B is the count rate measured about 2 μm away from a nanograin.

Figure 2 shows the fluorescence rate of a NV center in a nanocrystal and in bulk diamond as a function of the pump power. Slightly decreasing rate for high pump power is attributed to the presence of an additional shelving state [23,32]. It can be seen that the contribution of the background is greatly reduced in the nanocrystal configuration. The count rate in the nanocrystal is not as high as expected. However, a fair comparison with the bulk should take into account the $\tau_{nc}/\tau_b=2.2$ factor increase of the NV center lifetime in a nanocrystal (see below). The number of photons emitted in a lifetime is then larger in the nanocrystal.

The raw coincidences $c(t)$ (right axis) and autocorrelation function $g^{(2)}(\tau) = \langle I(t)I(t+\tau) \rangle / \langle I(t) \rangle^2$ (left axis) are represented in Fig. 3. For evaluating the intensity correlation function $g^{(2)}(t)$ of the NV center, the raw correlation data $c(t)$ is normalized and corrected in the following way. The raw coincidence rate $c(t)$ counted during a time T within a time bin of width w is first normalized to that of a Poissonian source according to the formula $C_N(\tau) = c(\tau) / (N_1 N_2 w T)$, where $N_{1,2}$ are the count rates on each detector. Then the normalized coincidence rate $C_N(\tau)$ is corrected for the background light, and we obtain $g_c^{(2)}(\tau) = [C_N(\tau) - (1 - \rho^2)] / \rho^2$, where $\rho = S / (S + B)$ is related to the signal to background ratio, that is measured independently in each experimental run. Note that we have checked experimentally that the background light has a Poissonian statistics. It can be seen in Fig. 3 that $g^{(2)}(0) \sim 0$, where the slight difference with zero is attributed to remaining background light emitted by the nanograin. This almost vanishing value of $g^{(2)}(0)$ is the signature of the presence of a single emitter in the observed nanocrystal. In

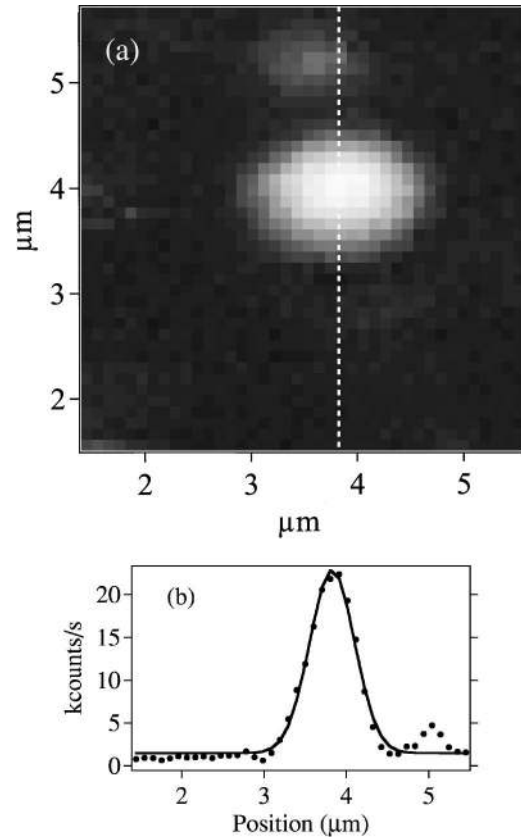


FIG. 1. (a) Confocal microscopy raster scan ($5 \times 5 \mu\text{m}^2$) of a diamond nanocrystal containing a single NV center. The count rate corresponds to one detector only. The size of a pixel is 100 nm and the integration time per pixel is 32 ms. The laser intensity impinging on the sample is 2.7 mW. In (b), a linescan along the dotted line of (a) is displayed, together with a Gaussian fit, that is used to evaluate the signal and background levels. Here we obtain $S/B = 20$.

the case of the presence of p centers within an unresolved peak, the value of the zero-time antibunching is $1 - 1/p$. This is actually how we estimate the number of NV centers in a nanocrystal. It should also be mentioned that $g^{(2)}(\tau)$ reaches values greater than unity for $\tau \neq 0$. This bunching effect is due to the presence of a third state in which the system can be shelved [23,32].

Obviously, for the ultimate achievement of a true single photon source, no background subtraction is possible and the meaningful quantity is the experimentally measured $C_N(0)$. Indeed, when the center is excited by a short intense pulse, the probability p_2 of having more than 2 photons in a pulse is given by (assuming $p_2 \ll 1$)

$$p_2 = C_N(0) p_1^2 / 2, \quad (1)$$

where p_1 is the probability of having a single photon. Note that for Poissonian light $C_N(0) = 1$ and Eq. (1) gives the photon probability distribution of an attenuated coherent pulse. In our case $C_N(0) = 0.17$ at the fluorescence rate maximum (input power of 2.7 mW), where the best value in bulk was 0.26 [21,23]. This would yield to significant im-

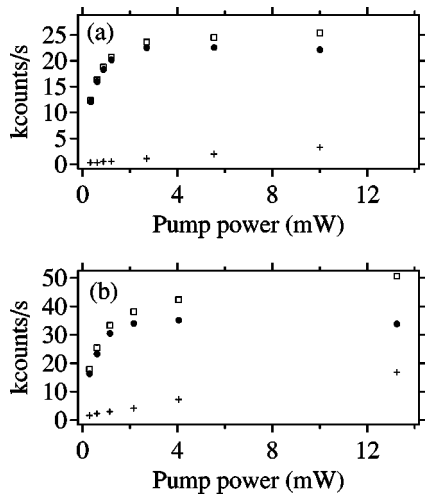


FIG. 2. Fluorescence rate of an NV center in a nanocrystal (a) and in bulk diamond (b) as a function of the pump power. The count rate corresponds to one factor only. The crosses, empty squares, and black circles represent, respectively, the background B , the total count rate $T=S+B$, and the signal from the center $S=T-B$. The maximum number of photons emitted in a lifetime is 5.5×10^{-4} for the nanocrystal (lifetime $\tau_{nc}=25$ ns) and 3.7×10^{-4} in the bulk (lifetime $\tau_{nc}=11.6$ ns). The data for the nanocrystal correspond to the center of Fig. 1.

provement compared to attenuated coherent pulses that are usually used in quantum cryptography experiments [1].

The central dip in the antibunching traces can be fitted by an exponential function of argument $-(r+\Gamma)|\tau|$, where r and Γ are, respectively, the pumping rate and the NV center decay rate. Such fits have been performed for different pumping powers. The inverse lifetime $\Gamma=1/\tau_{nc}$ of an NV center in a nanocrystal can then be inferred by extrapolating the value of the time constant for vanishing pump power. Figure 4 shows a lifetime of $\tau_{nc}=25$ ns for the observed NV center, that is significantly longer than the lifetime of a NV center in synthetic bulk diamond, $\tau_b=11.6 \pm 0.1$ ns [31]. This effect can be attributed to the strong change in the re-

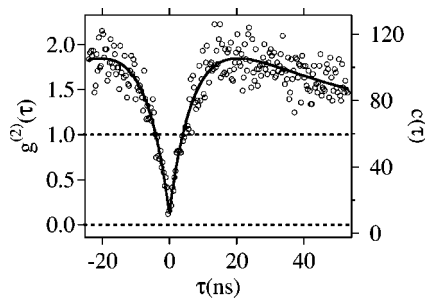


FIG. 3. Autocorrelation function $g^{(2)}(t)$ (left) and raw coincidence rate (right). The time bin is $w=0.3$ ns, the total integration time is 323 s, and the laser intensity impinging on the sample is 2.7 mW. Count rates on each photodiode are $N_1=22\,500$ s $^{-1}$ and $N_2=24\,500$ s $^{-1}$. The actual number of coincidences is indicated on the right. The zero-time value of the uncorrected normalized correlation function is $C_N(0)=0.17$. The fit is performed with the model used in [23]. The data correspond to the center of Fig. 1.

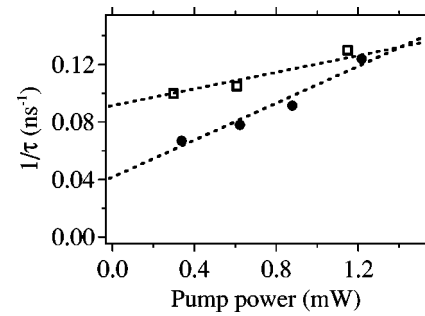


FIG. 4. Width of the exponential dip of antibunching traces obtained at different pump power. The extrapolation for vanishing pump power gives the lifetime. The “●” correspond to the data of Fig. 1 and yield $\tau_{nc}=25$ ns. The “□” correspond to a NV center in bulk diamond ($\tau_b=11.6$ ns). The slope in the nanocrystal is twice as large as the one in bulk. This is consistent with the lifetime increase, since the NV center in a nanocrystal can absorb twice as many photons during its lifetime.

fractive index of the surrounding medium, when going from bulk diamond to nanocrystals. In a simple approach [27], the squared amplitude of the one-photon electric field should be divided by the relative susceptibility $\epsilon_r=n^2$, while the mode density, that is proportional to the elementary volume in the wave-vector space, should be multiplied by n^3 . Since the inverse lifetime is proportional to the product of these two quantities, one obtains the simple formula [27]

$$\Gamma_n = n\Gamma_v, \quad (2)$$

relating the spontaneous emission rates Γ_n in the material and Γ_v in the vacuum. However, this formula does not take into account the local field effects due to the possible modification of the immediate surroundings of the emitting dipole, and the correct formula turns out to be $\Gamma_n = nl^2\Gamma_v$, where l is the local-field enhancement factor. Different models leading to different local-field correction factors have been proposed, and this topic is actively discussed in the literature [28–30].

However, our experimental results agree well with Eq. (2), and thus seem insensitive to the local field correction l . This may be explained by noticing that the immediate environment of the NV center at a scale of ~ 1 nm (i.e., a few crystalline periods) is unchanged. Therefore, the local field experienced by the NV center should be the same in the bulk and in a nanocrystal, so that local-field corrections cancel out, and the simple quantum electrodynamical analysis of Eq. (2) is valid. We note that NV defects located very close to the nanocrystal surface would have different optical properties than true NV centers owing to the modification of local symmetry. In particular their spectrum is expected to be modified, which is not what we have observed.

In order to evaluate the value of n that appears in Eq. (2), we consider that (outside the local field area) the NV center in bulk diamond emits within an medium of index $n_d=2.4$. For the center in a subwavelength nanocrystal, everything happens as if it were emitting in air for one half of the space, and in fused silica (refractive index $n_s=1.45$) for the other half. By using Eq. (2) in each half space we obtain $1/\tau_{nc}$

$= (1/2)[1/(n_d\tau_b) + n_s/(n_d\tau_b)]$, giving $\tau_{nc} = 22.7$ ns in good agreement with the experimental values. We note that this value is independent from the nanocrystal size, provided that this size is much smaller than the optical wavelength. On the other hand, by looking at 10 different nanocrystals, we have found a dispersion of ± 4 ns in the lifetime values. This dispersion may be attributed to the position of the NV center relative to the polymer/air interface, and to the randomness of the dipole orientation [33].

As a conclusion, we have observed almost background-free photon antibunching from single NV centers in diamond nanocrystals at room temperature. The photostability of NV centers in bulk diamond is preserved, allowing us to lock the laser beam on a single center during several hours. Manipulation of nanocrystals is a lot more flexible than bulk crystals. Straightforward improvement of the light collection efficiency should be possible by letting the nanocrystal sit on a metallic mirror, or inserting it in a microcavity [34–36]. It

should be stressed that in bulk diamond individual NV centers cannot be observed close to the surface, owing to excessive stray light [21,22]. Nanocrystals have thus a clear advantage for inserting NV centers in microcavities. These results show that diamond nanocrystals offer all the required properties for the realization of efficient single photon sources for quantum information systems. In addition, we found conclusive evidence that the lifetime of a NV center is larger in nanocrystals than in bulk, owing to change of the surrounding refractive index. Our data supports the conclusion that the local field effects do not contribute to the change in lifetime. This lifetime modification may then be interpreted as a simple quantum electrodynamics effect.

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