Triggered Source of Single Photons based on Controlled Single Molecule Fluorescence

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We use the method of adiabatic following to prepare a single molecule in its fluorescing excited state. Spontaneous emission from this state gives rise to a single photon. With our current experimental conditions, up to 74% of the sweeps lead to the emission of a single photon. Since the adiabatic passage is done on command, the molecule performs as a high rate source of triggered photons. The experimental results are in quantitative agreement with quantum Monte Carlo simulations.

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In addition to their fundamental interest [1], squeezed or controlled light states have many potential applications, such as communications, quantum computing, and quantum cryptography [2], for example, by using split single-photon states to generate encoding keys. Correlated or entangled photon states can be generated by an atomic cascade process [3] or by a parametric down-conversion process [4], where incident photons are converted into photon pairs (signal and idler). If an idler photon is detected at a certain time and position, we know that a signal photon exists at the conjugated position: This is equivalent to generating a heralded single-photon state. However, the generation time of this photon is completely random. For many applications, it would be desirable to determine the generation time while keeping a high rate of photon creation.

The ultimate quantum control of the single-photon generation process can only be achieved when a single photon is generated at an arbitrary, predetermined time, within a short time interval. This would result in heralded single photons with complete information about their generation times. Different schemes have been suggested to build such a photon source, for example, by means of the Coulomb blockade of electrons and holes tunneling in quantum confined p-i-n heterojunctions [5] or by means of highly nonlinear cavities [6]. A single fluorescent molecule or atom is a good potential source because it can emit only one photon at a time. The emitted light is antibunched [7] and consists of single photons separated by random time intervals which depend on the excited state lifetime and on the pumping rate. An even more regular stream of photons would be obtained if single photons were emitted periodically in time. Photon emission can be triggered with a short excitation pulse [8] or, as proposed here, with a suitable electronic signal [9]. Such a photon source would be quite similar to the locks and turnstiles [10] that were developed for single electrons. Here, we demonstrate a triggered one-photon source based on a single molecule.

To prepare the molecule in its fluorescing excited singlet state S_1 at a given time, one could excite the molecule with a short pi-pulse, for example, by means of a cw laser beam modulated with an electro-optical modulator [11]. Here, we use rapid adiabatic passage (or adiabatic following) [12] to excite the molecule. In the effective spin picture, the Bloch vector follows the driving effective field adiabatically if the frequency of the exciting cw laser is swept slowly through the resonance of the molecule. Since the passage inverts the effective field, the system goes from the ground to the excited state. Equivalently, in the dressed molecule picture, the system remains in one of the dressed eigenstates as the detuning is swept through the resonance. This excitation process is robust because it does not depend critically on the exact value of the laser intensity or on the specific shape of the frequency chirp. Because of relaxation, however, the sweeping cannot be too slow. Let T_{pass} be the passage time, i.e., the time it takes the laser frequency to cross the saturated molecular resonance. For the passage to be adiabatic, T_{pass} must be longer than the Rabi period, but shorter than the fluorescence lifetime. This requires that the Rabi frequency Ω be much larger than the homogeneous width Γ. To obtain a rapid adiabatic passage we sweep the molecular resonance at fixed laser frequency. This can be done easily by applying an electric field to the molecule [13], as was done to observe transient nutation [14]. The emitting structure can be made very compact, and it can be operated with low voltages. Earlier experiments have shown that the transition frequency of a single molecule can be modulated at high frequency [13], and its interaction with a laser field is very well described by the optical Bloch equations of a two-level system [15].

The system used for the present study is the highly fluorescent dibenzanthanthrene (DBATT) molecule (fluorescence yield close to unity) in a n-hexadecane matrix, at 1.8 K [16]. Most of the molecules in the wings of the inhomogeneous profile (centered at 589 nm) present a linear Stark effect [17], i.e., a difference between the static dipole moments of their ground and excited states. The optical design for the excitation of single molecules and the collection of fluorescence is of the lens-paraboloid-type [18] with a full silica parabolic mirror (see Fig. 1). A silica plate was covered with aluminum, except for a narrow stripe where the sample, a small drop of a DBATT solution in hexadecane has been deposited. The rf electric

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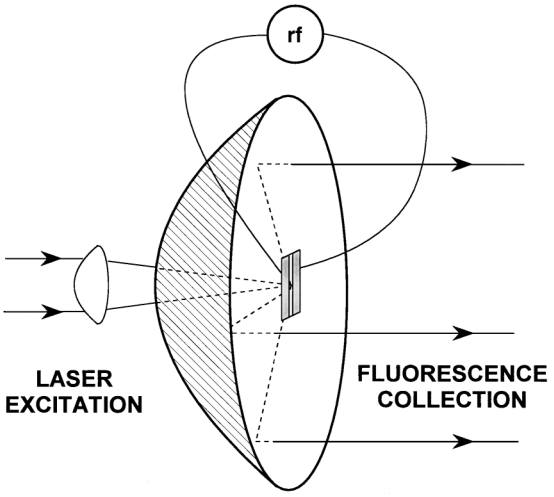

FIG. 1. Schematic diagram of the optical setup used to excite single molecules and to collect their fluorescence. The exciting laser is focused by a lens through a hole in a parabolic collecting mirror onto the sample placed on the front face of the mirror. The Stark shifting rf field is applied via two aluminum electrodes 18 μm apart.

field with a frequency ν_{rf} of a few MHz is applied to the electrodes and shifts the single molecule's transition frequency sinusoidally with time. The excitation laser is tuned to the molecular resonance as it is in the absence of the rf, i.e., to the center of the frequency interval swept by the molecule. Every time the molecule's transition crosses the laser frequency in the center of the scan the molecule gets excited and subsequently emits fluorescence photons with an average delay of 8 ns, which is the lifetime of the S_1 state of DBATT. The homogeneous linewidth Γ is lifetime limited and equal to about 20 MHz. The single DBATT molecules we chose had high linear Stark coefficients, high photostability, and their lines were standing alone in wide frequency intervals of several GHz.

The arrival times of fluorescence photons were first recorded with a single detector [a photomultiplier tube (PMT)] and a time-to-amplitude converter triggered by the rf signal. The overall detection efficiency of the setup was $\eta_1 \approx 10^{-3}$. An example of a triggered recording of the fluorescence as a function of time is presented in Fig. 2. The inset shows that photons are emitted at regular time intervals, in bursts separated by half the rf period. The bursts are generated when the rf electric field vanishes, i.e., when the molecule is in resonance with the laser. The rf frequency is $\nu_{\mathrm{rf}} = 3$ MHz, the Rabi frequency is $\Omega = 2.6\Gamma$, and the total frequency interval Δ_0 swept by the molecule is 1.6 GHz, which is about 90 times the linewidth Γ. The detected photon rate is about 6300 counts/s and the data presented are accumulated over 480 s. Curve (*a*) of Fig. 2 shows the detailed shape of one of the bursts for a different set of parameters ($\Omega = 3\Gamma$, $\nu_{\mathrm{rf}} = 4$ MHz, and $\Delta_0 = 160\Gamma$). The peak presents a short rise time of the fluorescence, i.e., of the excited state population (about 5 ns, depending on the rf and the Rabi frequencies), which shows the rapid excitation of the molecule. The population decays exponentially with a time constant of approximately 8 ns, in agreement with the lifetime of the excited state. The oscillations in the exponential tail are a transient nutation effect. They appear because the conditions for a rapid adiabatic passage are not perfectly satisfied. The

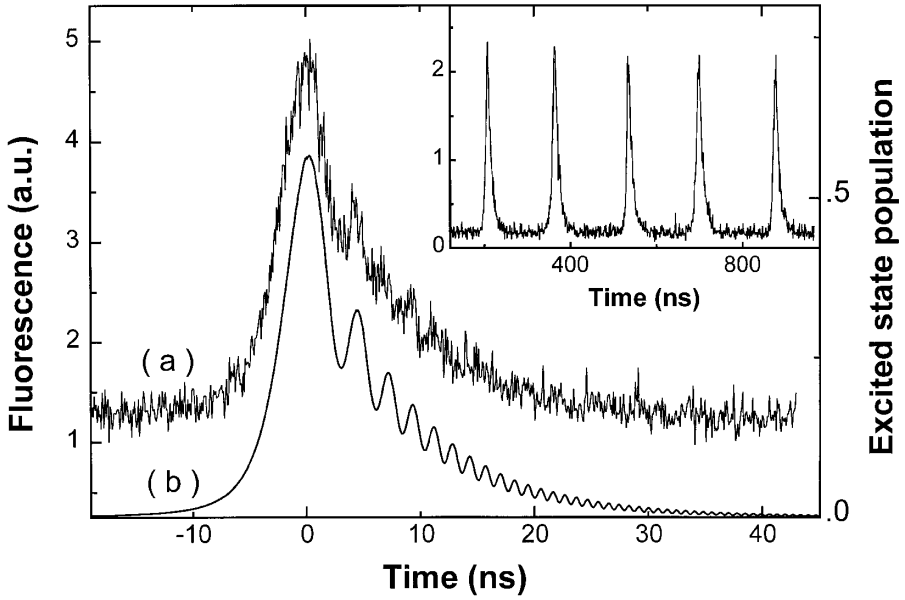

FIG. 2. Inset: fluorescence bursts of a single molecule under periodic linear Stark shifting by a sinusoidal rf wave, with cw laser excitation at a fixed frequency in the middle of the swept interval. Photon bursts are obtained whenever the molecular line crosses resonance with the laser. The rf frequency is 3 MHz and the Rabi frequency is $\Omega = 2.6\Gamma$. The molecular frequency sweeps $\Delta_0 = 90\Gamma$ ($\Gamma = 20$ MHz). The data were accumulated over 480 s. Curve (*a*) the figure shows the detailed shape of a fluorescence burst for a different set of parameters ($\nu_{\mathrm{rf}} = 4$ MHz, $\Omega = 3\Gamma$, and $\Delta_0 = 160\Gamma$). Curve (*b*) is the burst shape from a quantum Monte Carlo simulation with the same parameters.

theoretical curve (*b*) of Fig. 2 is a quantum Monte Carlo simulation [19] of the time dependence of the excited state population. The linewidth and Rabi frequency parameters for the simulation were determined from the optical saturation of the single molecule line, as discussed elsewhere [13,15], and the frequency sweep rate was taken from a dc Stark measurement [17]. Although there was no free parameter left, besides the background level and the overall intensity of the fluorescence, the simulation is in complete agreement with the measured signal.

Having checked that photons are delivered only when the molecule passes through resonance, we have to determine how many photons are emitted at each passage. The detected count rate of the inset of Fig. 2 corresponds roughly to the expected signal for the emission of a single photon per burst and for an overall detection yield of 10^{-3} at an emission rate of 6 MHz (twice the rf frequency). However, the emission rate alone does not allow us to prove that our source delivers single photons, because there can be bursts with zero, one, two, or more photons emitted. A complete determination of these probabilities would require an accurate measurement of the absolute detection yield, which is difficult. Instead, we decided to determine the number of bursts giving rise to two or more photons. By comparing the number of photon pairs to the average signal, we can eliminate the unknown quantum yield. We built a Hanbury-Brown and Twiss setup [7], with a beam splitter and two detectors, a PMT and an avalanche photodiode (APD). The histogram of the delays between detected photons is measured using a time-to-amplitude converter, in which the start signal (delayed for negative times) is given by the APD (overall detection efficiency $\eta_2 \approx 2 \times 10^{-3}$) and the stop signal by the PMT (overall detection efficiency $\eta_1 \approx 10^{-3}$). The left part of Fig. 3 shows the distribution of time intervals of consecutive photon pairs for three different values of Δ_0 ($\Delta_0 = 50\Gamma$, 88Γ, and 130Γ), under a 3 MHz rf field, and with a Rabi frequency of 3.2Γ. From the time pattern of the photon emission in Fig. 2, we expect the start-stop histograms to also show a peak pattern, as confirmed by Fig. 3. The central peak around zero time arises from photon pairs emitted during the same burst. It has a dip in its center, which arises from antibunching [7] since photons are emitted one by one. Although the dip does not quite reach zero due to the limited time resolution of the setup (a similar effect arises in the simulated histograms of Fig. 3), its large amplitude confirms that the fluorescence arises from a single molecule. For the Poissonian light states of a pulsed periodic coherent source (such as an attenuated laser), the central peak would be identical in intensity and shape to the lateral ones, but it should vanish altogether if a single photon was emitted in each burst. Therefore, the intensity ratio of the central peak to lateral ones [20] is directly related to the fraction of bursts giving two or more photons.

The right part of Fig. 3 shows quantum Monte Carlo simulations of the second order correlation function (i.e., the distribution function of delays) for the experimental data presented on the left. From these simulations, we deduced the probabilities $p(m)$ to have m photons emitted per burst. As expected, they are radically different from the values $e^{-n_{\mathrm{av}}} n_{\mathrm{av}}^m / m!$ corresponding to a pulsed coherent source with the same average number of photons n_{av}. The probabilities $p(m)$ enable us to calculate the ratio $R_{\mathrm{sim}} = A_0/A_1$ between the areas A_0 of the central peak and A_1 of one of the lateral peaks:

$$R_{\mathrm{sim}} = \frac{1}{n_{\mathrm{av}}^2} \sum_{m=2}^{\infty} m(m-1)p(m),$$

$$\text{where } n_{\mathrm{av}} = \sum_{m=0}^{\infty} mp(m).$$

We can now compare the simulated ratio R_{sim} to the experimental ratio R_{exp} deduced from Fig. 3, for fixed Rabi and rf frequencies and for three different values of the swept frequency interval, inversely proportional to the passage time. Table I shows that the agreement is very good. While the average number n_{av} of emitted photons increases with the passage time, we can see from Table I that the probability $p(1)$ of having a single-photon burst goes through a maximum close to 0.7. It is convenient to compare the distribution of emitted photons to a Poisson

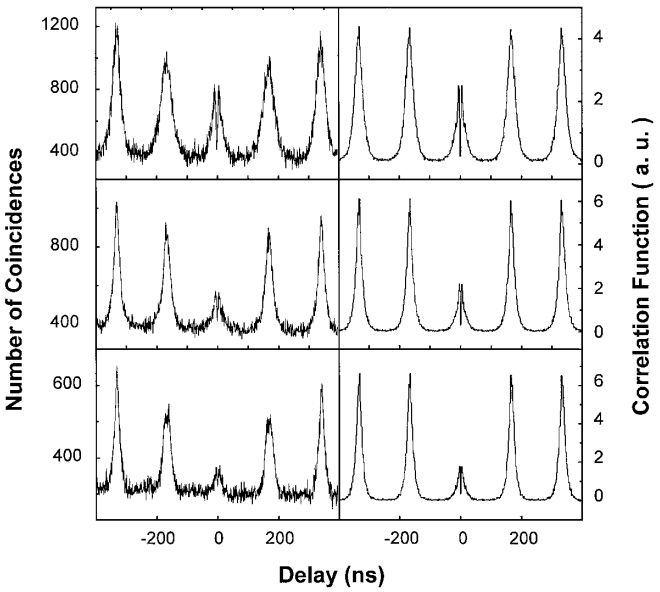

FIG. 3. Left: histograms of time delays in a start-stop experiment for three different values of the swept frequency interval ($\Delta_0 = 50\Gamma$, 88Γ, and 130Γ, from top to bottom). The rf frequency is 3 MHz, the Rabi frequency is 1.2Γ, and the histograms are normalized with respect to an accumulation time of 1000 s. The split central peak around zero delay results from bursts yielding two or more photons (the dip arises from antibunching). The ratio of the areas of the central peak to that of the lateral ones is given in Table I. For the coherent states of a pulsed laser source, the central peak would have the same area and shape as the lateral ones. For the second recording, 68% of the sweeps give a single photon. Right: simulated correlation functions for the same parameters.

TABLE I. Experimental values R_{exp} of the ratio between the central structure and a lateral structure of the coincidence pattern of Fig. 2, for three different values of the swept frequency interval Δ_0. Simulated values of the same ratio R_{sim}, the average number n_{av} of emitted photons, the probabilities $p(m)$ of the emission of m photons per burst, and the Mandel parameter Q_E of the source.

Δ_0	R_{exp}	R_{sim}	n_{av}	$p(0)$	$p(1)$	$p(2)$	Q_E
50Γ	0.56	0.60	1.53	0.02	0.56	0.31	-0.59
88Γ	0.42	0.43	1.12	0.11	0.68	0.18	-0.65
130Γ	0.32	0.33	0.91	0.22	0.66	0.10	-0.61

distribution by means of the Mandel parameter $Q = (\Delta n - n_{\text{av}})/n_{\text{av}}$, where Δn is the average squared number of photons [21]. The simulated parameters Q_E are characteristic of our source. They are negative, which means that the distribution of emitted photons is subPoissonian, and their value is not far from -1, which is the value for the single photon Fock state. (Of course, the Mandel parameter Q_D of the detected photocounts, being affected by the light detection efficiency of the setup, is much smaller. It can be deduced from $Q_D = \eta_1 Q_E$ [22] and is about -0.6×10^{-3}.) Using higher laser intensity and higher rf frequency will improve the conditions of the adiabatic following and thus the quality of this single-photon source. The limits of our setup are the background counts for high laser intensity and the excited state lifetime for short rf periods. With another molecule at $\Omega = 5\Gamma$, $\nu_{\text{rf}} = 3$ MHz, and $\Delta_0 = 160\Gamma$ (data not shown), we were able to obtain a probability of 74% to have a single photon emitted per burst.

We have built a triggered single-photon source and have demonstrated photon states with statistics that differ radically from those of coherent states. For quantum optical applications, the photons have to be collected and detected with higher efficiency [23], and their spatial, temporal, spectral, and polarization characteristics must be as well defined as possible. Here, the spatial mode was defined by the collecting paraboloid, the polarization lay along the dipole moment of the molecule, and the spectrum was that of the spontaneous fluorescence [16]. Coupling the molecule to a resonant cavity would be an elegant solution to narrow its emission spectrum, e.g., by selecting a single vibronic line.

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Note added.—After submitting this paper, we learned about another triggered single-photon source based on a p-n junction fed by a turnstile single-electron device [24]. The triggered single photon recording (Fig. 3 of Ref. [24]) is very similar to the inset of our Fig. 2. The photon coincidence measurement in our Fig. 3 is more quantitative, as it enables determination of probabilities of zero-, one-, and two-photon events.

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