Realization of two Fourier-limited solid-state single-photon sources

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Abstract: We demonstrate two solid-state sources of indistinguishable single photons. High resolution laser spectroscopy and optical microscopy were combined at $T = 1.4 \, \text{K}$ to identify individual molecules in two independent microscopes. The Stark effect was exploited to shift the transition frequency of a given molecule and thus obtain single photon sources with perfect spectral overlap. Our experimental arrangement sets the ground for the realization of various quantum interference and information processing experiments.

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Single-photon sources have been demonstrated in the solid state with molecules \[1\] and quantum dots \[2\], quantum color centers in diamond \[4\] as well as with atoms \[5\] and ions \[6\] in the gas phase. Single photons have been successfully used for applications in quantum cryptography \[7, 8\], but complex schemes of quantum information processing require a large number of indistinguishable photons \[9, 10\]. The first efforts in this direction have used consecutively emitted photons by single quantum dots, atoms in cavities or molecules \[11\] \[12\] \[13\]. More recently experiments with two independently trapped atoms or ions have been also reported \[14\] \[15\]. Here, we show that two molecules embedded in solid samples can also be used to generate lifetime-limited photons for use in the investigation of photon interference and correlation effects \[16\]. The solid-state aspect of our system offers many advantages including well defined polarization and a nearly indefinite measurement time using the same single emitter.

The experimental arrangement is sketched in Fig. 1 (a). Two independent low temperature microscopes are separated by an opaque wall in a liquid helium bath cryostat at \( T = 1.4 \) K. Light from a tunable dye laser (Coherent 899-29, \( \lambda = 590 \) nm, linewidth 1 MHz) is coupled into the microscopes via galvo-optic mirror scanners and telecentric lens systems. Each sample is prepared by sandwiching a solution of dibenzanthanthrene (DBATT) in \( n \)-tetradecane (concentration of \( 10^{-7} / \text{mol} \)) between a glass substrate that contains interdigitating gold electrodes (spacing 18 \( \mu \text{m} \)) \[17\] and a hemispherical cubic zirconia solid immersion lens (SIL) \[18\]. The electrodes can be used to apply electric fields of up to \( 5 \times 10^5 \) V/m resulting in a Stark shift of more than 5 GHz for the molecular resonance. The SILs combined with aspheric lenses of high numerical aperture (NA=0.55) provide an effective NA of 1.12 \[19\], allowing a high spatial resolution and an enhanced fluorescence collection efficiency \[20\]. Three piezoelectric slider systems in each microscope make it possible to position the sample/SIL unit precisely with respect to the aspheric lens. The collected photons of the microscopes can be sent either to an avalanche photodiode (APD), a spectrometer with a resolution of 0.3 nm, an imaging CCD
camera or a Hanbury Brown and Twiss (HBT) type photon correlator. Long-pass cut-off and band-pass filters are used to block undesired photons.

The energy spectrum of a dye molecule embedded in a solid at low temperatures is composed of electronic and vibrational levels. In what follows, we denote the states corresponding to the electronic ground and the first excited states of DBATT by $S_{0,v}$ and $S_{1,v}$, respectively (see fig. 1(b)). It turns out that each vibrational transition is composed of a narrow zero-phonon line (ZPL) and a phonon wing that stems from the vibrational coupling of the molecule with the host matrix. The ratio of the photons emitted into the ZPL to the total number of photons emitted into that particular vibronic transition is given by the Debye-Waller ($\alpha_{DW}$). The ZPL of the transition between $S_{0,0}$ and $S_{1,0}$ (from now on referred to as 0-0 ZPL) in DBATT becomes natural linewidth limited at $T \leq 2\, K$, providing an ideal source of Fourier-limited single photons.

In standard fluorescence excitation spectroscopy, a narrow-band laser resonantly excites the molecule via the 0-0 ZPL. The molecular excitation decays consequently via the vibrational levels $S_{0,v}$, emitting Stokes-shifted photons (figure 1(b)). To achieve a high signal-to-noise ratio and a low background in detecting single molecules, however, the 0-0 ZPL emission is cut off by using filters in detection. Figure 2(a) displays an excitation spectrum from a single DBATT molecule. The full width at half-maximum (FWHM) of 17±1 MHz corresponds to the excited state lifetime of 9.4±0.5 ns.

In order to capture the emission on the narrow 0-0 ZPL transition, we apply a second version of fluorescence excitation spectroscopy by exciting the molecule to $S_{1,1}$ at 242 cm$^{-1}$ higher frequency than the 0-0 ZPL. This state quickly relaxes into $S_{1,0}$ where it decays further to the electronic ground state and emits a visible photon as in the previous case (figure 1(b)). Figure 2(b) shows such an excitation spectrum for the same molecule studied in Fig. 2(a). The observed FWHM of 28 GHz is about three orders of magnitude broader than the ZPL transition and is caused by the short lifetime of about 5 ps for the $S_{1,1}$ state.

The large linewidth of the 0-1 transition implies its low absorption cross section. As a result, more than two orders of magnitude higher pump power (100 nW) is needed to obtain a signal comparable to the conventional excitation scheme used in Fig. 2(a). This high laser intensity increases the background fluorescence because together with broad resonance lines it facilitates the excitation of neighboring molecules, reducing the spectral selectivity that has been
the central asset in cryogenic single molecule detection. To get around this problem, we have worked with samples that are 100 times more dilute than commonly studied in single molecule spectroscopy. Furthermore, we have exploited the high resolution provided by SILs to identify single molecules spatially. The insets in Figs. 2(a) and (b) show confocal scan images recorded when the laser frequency was set to the resonances shown in each figure. The FWHM of 290 nm is very close to the diffraction limited value of 270 nm expected from NA=1.12 and allows us to verify that the same molecule is the source of signals in both cases.

The next step toward the preparation of a Fourier-limited single photon source is to filter the 0-0 phonon wing and the Stokes shifted emission to extract the 0-0 ZPL. Figure 3(a) shows the emission spectrum of a single DBATT molecule obtained by a 0-1 excitation. The dominant line at 590 nm represents the 0-0 ZPL. Several emission lines and the phonon wings (see also Fig. 3(b)) are clearly visible. The recorded spectrum is in good agreement with ensemble measurements reported in the literature [25]. By comparing the different areas under the measured spectrum, we can estimate the Franck-Condon and the Debye-Waller factors to be 0.4 and
To verify the realization of a single photon source, we have measured the second order auto-correlation function $g^{(2)}(\tau)$ on the 0-0 ZPL isolated according to the above-mentioned procedure. As shown in Fig. 4(a), a strong antibunching is observed with $g^{(2)}(0) = 0.18$. We attribute the deviation from the ideal value of 0 to a limited time resolution of the single photon counters. The antibunching characteristic time of 5.5 ns is shorter than the decay time of 9.4 ns for the $S_{1,v=0}$ state because of the strong pumping of the population out of the ground state [26]. To ensure that the emitted 0-0 ZPL photons are nevertheless natural linewidth limited, we sent the emitted photons through a Fabry-Perot cavity with a finesse of 200 and a free spectral range of 1.5 GHz. Figure 4(b) clearly shows a single peak within the free spectral range of the spectrum analyzer. The zoom in the inset highlights the narrow linewidth when accounting for the instrument FWHM of 7.5 MHz.

Having demonstrated the generation of narrow-band single photons from single DBATT molecules, we searched for two molecules, one in each microscope, that had 0-0 ZPLs within a few GHz of each other. By applying a constant electric field to the electrodes embedded in the glass substrates, we could tune the 0-0 ZPLs of the molecules independently via the Stark effect. Figure 5 shows the 0-0 fluorescence excitation spectra from two molecules recorded on one APD as the voltage applied to one sample was varied. The 0-0 ZPL of one molecule shifts with increasing voltage and sweeps through the resonance of the second molecule. At an applied field of about $1.2 \times 10^6$ V/m the spectra of the two molecules can be no longer distinguished. Given the well-defined orientation of the transition dipole moment in DBATT, the emission of single molecules can be linearly polarized with an extinction ratio of the order of 300:1 [19], making it a simple task to obtain the same polarization state for both single photon sources. Overlapping Fourier-limited spectral lines and polarization states establish the two molecules as sources of indistinguishable single photons.

In conclusion, we have combined cryogenic optical microscopy and spectroscopy to identify and prepare two single molecules that generate indistinguishable photons. A spectral analysis using a Fabry-Perot cavity and antibunching measurements gave direct proof for the generation of lifetime-limited single photons. The solid-state arrangement in this experiment using SILs makes it possible to miniaturize each single-photon source and to integrate several of them on one chip. Together with the nearly indefinite photostability and the long coherence times of dye molecules, these features make our approach promising for performing complex quantum
Fig. 5. (a) Fluorescence excitation spectra of two molecules. Molecule (B) is shifted with respect to molecule (A) with increasing voltage. (b)-(d) cross sections as indicated in (a). At $V = 21$ V, the two spectra are fully overlapped.

operations. We are currently working to trigger the emission of single photons by incorporating a pulsed laser for the 0-1 excitation.

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