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Coherence length of photons from a single quantum system

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We present a methodology that allows recording the coherence length of photons emitted by a single quantum system in a solid. The feasibility of this approach is experimentally demonstrated by measuring the self-interference of photons from the zero-phonon line emission of a single nitrogen-vacancy defect in diamond at 1.6 K. The first-order correlation function has been recorded and analyzed in terms of a single exponential decay time. A coherence time of ~ 5 ps has been obtained, which is in good agreement with the corresponding spectral line width and demonstrates the feasibility of the Fourier-transform spectroscopy with single photons.

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The creation of single-photon states is essential for most applications in quantum computation [1], cryptography [2], and interferometric quantum nondemolition measurements [3]. The coherence properties of the single-photon source make it highly nontrivial to build an optical system for linear quantum computation schemes. Phase-sensitive entanglement techniques require long coherence length of single photons (longer than the phase difference in interferometers). There are many proposals for single-photon sources, ranging from faint laser pulses and parametric down-conversion [2], and recombination of electrons and holes in single quantum dots [4,5] to optically excited single molecules [6]. The interest in single molecules and quantum dots is based on their unique property, which is inherent in a single quantum system not being able to emit the next photon before being excited again and relaxed to the proper state. An important disadvantage of the sources based on single quantum systems, as reported to date, is the broad emission spectral bandwidth, which enhances the perturbation effect in the quantum channels. It will be vital to characterize and control the generation and the localization properties of the photons to be used.

The coherence length of the photon, which is emitted from a single quantum system, is given by the coherence time τ_c of the excited state, and is determined by Heisenberg's uncertainty relation, $\Delta E \tau_c \geqslant h/2\pi$. Here, τ_c can be considered as the dephasing time of the excited state, and, from the quantum optics point of view, as the duration of the wave train that is associated with the emitted photon. The length of the wave train as well as the uncertainty of the photon energy cannot be measured for a single photon. The operators of phase and number of photons do not commute. Therefore, the definition of a single photon as an oscillatory wave with a definite phase is meaningless. However, when experiments on an ensemble of quantum systems or on a single quantum system undergoing multiple excitation-emission cycles are performed, the frequency distribution of the emitted wave packet, ΔE, and its phase information become experimentally accessible [7,8]. By measuring the emission first-order autocorrelation function, we demonstrate in this paper that the Fourier-transform spectroscopy can be applied in order to characterize the coherence length of a single-photon source.

For most applications, the use of a single organic dye molecule is limited by its poor photostability at room temperature. In contrast, a promising candidate for a photon source is a nitrogen-vacancy (N-V) defect center in diamond [9]. A color center consists of a vacancy trapped by substitutional nitrogen in a diamond lattice [Fig. 1(b)], and exhibits unparalleled photostability even at room temperature [10]. Positron annihilation experiments [11] show that the charge state of the N-V defect is negative. It exhibits a strong dipole-allowed $E \rightarrow A$ optical transition with a zero-phonon line (ZPL) at 1.945 eV (637 nm), allowing individual centers to be observed optically by means of fluorescence detection [Fig. 1(a)] [12]. As a sample material we used diamond powder (De Beers) with a grain size <250 nm. Irradiation

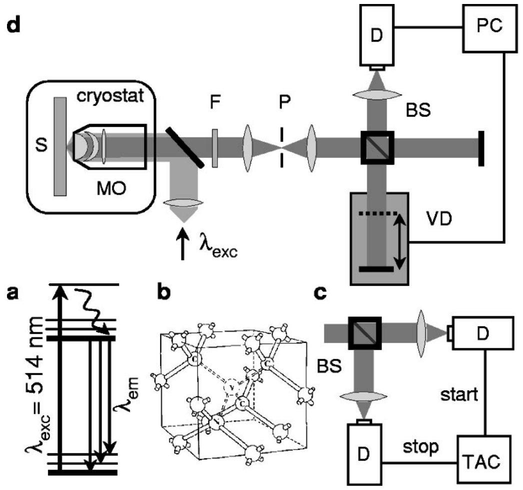

FIG. 1. Scheme of the experiment. Relevant energy-level diagram (a) and structure (b) of an N-V defect center in diamond. Schematic representation of the scanning confocal microscope setup used to characterize the single-center first (d) and second (c) -order coherence. (For details, see text.)

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with 2-MeV electrons resulted in a defect concentration of one center per nanocrystal.

The experimental implementation of the photon detection schemes is schematically depicted in Figs. 1(c) and 1(d). Diamond nanocrystals were spin coated on a quartz substrate, and single nanocrystals were observed with a home-built low-temperature confocal microscope [12]. The excitation beam (514 nm line of a cw Ar^+ laser) was focused onto the sample *S*, which was placed in a helium cryostat (Janis), by a microscope objective lens (MO, Melles Griot, numerical aperture of 0.85). Fluorescence photons were collected by the same objective, spectrally isolated from the residual excitation light by a holographic notch filter (Kaiser) and an interference band-pass filter (F, Omega), and passed through a confocal pinhole (P, diameter: 100 μm). In order to measure the second-order coherence, the emitted fluorescence was sent into a Hanbury-Brown and Twiss interferometer detection setup [Fig. 1(c)], consisting of a 50-50 beam splitter (BS) and two (start and stop) silicon avalanche photodiodes D. The histogram of the time lags between detected photons was measured using a time-to-amplitude converter (TAC, Becker and Hickl, SPC-630). The first-order coherence was monitored using a Michelson interferometer [Fig. 1(d)]. The path length of one of the arm of the interferometer was scanned by a computer-controlled translation stage (VD, Physik Instrumente).

Figure 2(a) shows the fluorescence spectrum of a single N-*V* defect center in a diamond nanocrystal at 1.6 K upon 514-nm excitation. The spectrum consists of the zero-phonon line at 637 nm and the broad redshifted phonon sideband. In order to guarantee the quality of the single-photon source, the emitted photons were first investigated by means of their photon arrival times, and analyzed in terms of the interphoton time distributions and the second-order fluorescence intensity correlation functions. The normalized histogram of interphoton times, as displayed in Fig. 2(b), represents the second-order correlation function $g^{(2)}(\tau)$. It shows a dip between consecutive photon detection events at zero time delay. This photon antibunching effect indicates the characteristic sub-Poissonian photon statistics in the fluorescence from a single quantum system, which cannot be described in classical terms. The photon detection event projects the emitting system into its ground state, resulting in a zero probability to detect the second photon simultaneously. This is confirmed for a single N-*V* defect center by the experiment, where the observed zero-delay value of $g^{(2)}(\tau)$ amounts to 0.45 ± 0.02. Note that the zero-delay values lower than 0.5 guarantee an existence of a single emitting state [6]. For increasing interphoton times τ, the probability distribution curve follows an exponential rise with a rate constant that is determined by the lifetime of the excited state. The fit yields a rise time of 10 ns, which is in a good agreement with the fluorescence lifetime observed previously for an ensemble of N-*V* centers [13].

Having verified the single-photon nature of the fluorescence from a single N-*V* defect, the photon stream was directed into a Michelson interferometric detection setup [see Fig. 1(d)]. The phase properties of the spontaneous emission from a single defect center reveal themselves after passing the interferometer. Fig. 2(c) displays the measured interferogram $I(\tau)$ that is the photon-count rate of the N-*V* defect fluorescence integrated over the entire emission spectrum as a function of the delay time τ. The latter is introduced by the path difference of the interferometer, $\Delta=c\tau/2$, with *c* being the vacuum speed of light. Interference fringes with a period of ~700 nm are observed, which is in agreement with the mean wavelength of the emission of an N-*V* defect [Fig. 2(a)]. The visibility of interference fringes, $V(\tau)$, is defined as

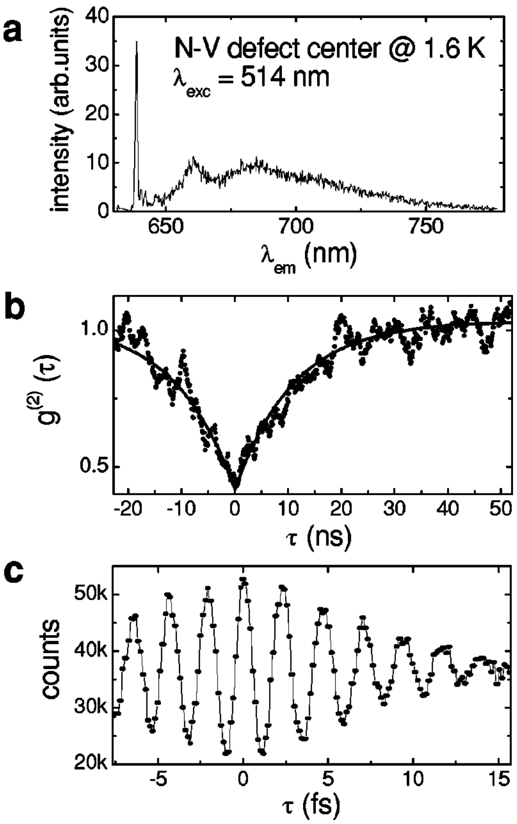

FIG. 2. (a) The fluorescence spectrum of a single N-*V* defect center in a diamond nanocrystal at 1.6 K upon 514-nm excitation. The spectrum consists of the zero-phonon line at 637 nm and the broad redshifted phonon sideband. (b) The second-order fluorescence autocorrelation function of the single N-*V* defect. The fit to an exponential law yields a contrast of $55\pm2\%$ at zero-delay time indicating that emission is originating from the single quantum system. The rise time amounts to 10 ns, which is in good agreement with the fluorescence lifetime observed for an ensemble of N-*V* centers previously. (c) The first-order field-field correlation function of the total fluorescence from a single N-*V* defect center. The detuning of the interferometer path length is presented in time scale.

$$V(\tau)=\frac{I_{max}(\tau)-I_{min}(\tau)}{I_{max}(\tau)+I_{min}(\tau)}, \tag{1}$$

where $I_{max}(\tau)$ and $I_{min}(\tau)$ represent the fringes maxima and minima, respectively. A measured maximum visibility of 0.45 at zero-delay time, which is below the value for pure coherence, can be attributed to the distortion of the photon wave front when the light is propagating through the diamond crystal. This experiment demonstrates two important aspects of interference. First, it reveals that a single photon from an N-V center exists in a superposition of states, including both paths of the interferometer. Second, the bandwidth of the single-photon emitter can be directly deduced from the decay of the visibility in the interferogram.

Assuming a Lorentzian shape of the emission line and perfect wave-front matching conditions, the observed interference signal intensity in the interferogram as a function of the delay time is given by the following expression [14]:

$$I(\tau)=2R^2T^2\langle I(t)\rangle[1+\cos(\omega_0\tau)e^{-\tau/\tau_c}]. \tag{2}$$

Here, R and T are the reflection and transmission coefficients of the beam splitter, respectively. $\langle I(t)\rangle$ is the time-averaged fluorescence intensity that is delivered to the Michelson interferometer. τ_c and ω_0 are the coherence time and frequency of the input field, respectively. The second term in the bracket is the interference term. In order to infer the coherence properties from the interferogram, we studied the visibility of interference fringes. The dependence of the visibility on τ can be written in terms of the first-order field coherence $g^{(1)}(\tau)$,

$$V(\tau)=e^{-\tau/\tau_c}=g^{(1)}(\tau). \tag{3}$$

Thus, by recording the visibility of the interference fringes as a function of interferometer detuning, one gains access to decoherence processes in the single quantum system. The measured coherence length reflects the photon emission process and, therefore, is dependent on the intramolecular and intermolecular dynamics of the emitting single defect occurring on the sub-picosecond time scale.

The interferogram in Fig. 2(c) was obtained by collecting the single N-V defect fluorescence over its entire frequency spectrum [see Fig. 2(a)]. It exhibits a fast decay of the visibility, which is shown in Fig. 3. The visibility data were fitted to an exponential decay, resulting in a decay time of 13 fs. Since the spectrally broad phonon sideband consists of several phonon replicas, and it is not homogeneously broadened, the measured decay time cannot be assigned to the phonon lifetime. When just the spectral interval corresponding to the ZPL transition was isolated for detection, the decay time was increased by more than two orders of magnitude. The time dependence of the visibility for the ZPL emission from a single N-V center is also shown in Fig. 3. The fit to Eq. (3) results in a value for the coherence time of $\tau_c=5$ ps, which is in agreement with the linewidth obtained using fluorescence excitation spectroscopy on single centers [15]. In comparison with values in the nanosecond range expected from spectral hole burning measurements [16], our experimental result is more than two orders of magnitude smaller. In order to rationalize such a fast decoherence of the system, spectral diffusion and dephasing have to be taken into account. The fact that the exponential decay time of the visibility is in good agreement with the Lorentzian shape of the emission line supports the occurrence of pure dephasing processes. A possible explanation is that dephasing is induced by the Stark effect due to the interaction of the N-V center with the electrons in the diamond conduction band. Atomic nitrogen is present in high concentrations (200 ppm) in type-1b diamond. Nitrogen is a shallow electron donor, 2 eV below the conduction band. It is known, however, that an electron of atomic nitrogen may be removed, giving rise to charge fluctuations. Since the N-V center is charged (N-V^-), the excited-state coherence is extremely sensitive to such field fluctuations in the vicinity of the defect that may induce decoherence.

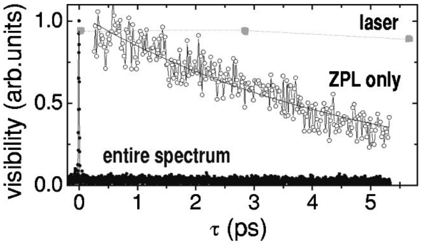

FIG. 3. Visibility of Michelson interferometric fringes as a function of the path detuning for the entire fluorescence emission (filled circles), and for the spectrally selected zero-phonon line (open circles). The precise coherence length was determined by fitting the visibility to an exponential decay (solid line), resulting in a coherence time of $\tau_c=(4.9\pm0.4)$ ps for the isolated zero-phonon line. For the entire spectrum, the observed decay time of (13.0 ± 0.1) fs is assigned to the spectral width of the phonon sideband. For comparison, the constant visibility of interference fringes from a narrow-band reference laser is shown (gray dotted line), indicating the stability of the interferometer over the full scan range.

We have demonstrated experimentally the decay of coherence in single-photon interference from a single quantum system in a solid. Since coherence is an important part of quantum computation schemes based on single photons and linear gates, our results provide the basic requirements for such methods. It should be noted that the concept of the present work might be also important for single-molecule spectroscopy. Earlier linewidth measurements of a single fluorescent molecule using fluorescence excitation spectroscopy techniques have demonstrated that single-molecule spectroscopy is able to overcome inhomogeneous broadening and resolve homogeneous linewidths of impurities in solids [17]. However, this approach is limited to systems with an intense Stokes-shifted vibrational band. The interferometric technique described here allows studies with a comparable spectral resolution, but using nonselective excitation. The spectral resolution limit is only given by the maximal scan length of the interferometer, a fact that makes the conventional Fourier-transform spectroscopy very powerful in comparison with diffraction spectrometers. This approach is particularly useful for the study of the zero-phonon linewidth of single quantum dots, where fluorescence occurs mainly in the zero-phonon line region.

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