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Observation of 1.2-GHz linewidth of zero-phonon-line in photoluminescence spectra of nitrogen vacancy centers in nanodiamonds using a Fabry-Perot interferometer

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Abstract: Photoluminescence (PL) spectra of single nitrogen vacancy (NV) centers in 50-nm diamond nanocrystals at the zero-phonon line (ZPL) were directly observed using a Fabry-Perot interferometer at cryogenic temperatures. The narrowest linewidth of ZPL was 1.2 GHz (1.9 ± 0.7 GHz on average), comparable to ZPL linewidths in PL spectra reported for NV centers in pure bulk diamond. This observation is important to the application of NV centers for use in quantum communication and computation devices, and in nano-sensing.

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OCIS codes: (300.0300) Spectroscopy; (300.2530) Fluorescence, laser-induced; (160.2220) Defect-center materials.

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1. Introduction

Nitrogen vacancy (NV) color centers in diamond have attracted many researchers in recent years. NV centers are robust, exhibit optical transitions with life-time limited linewidth, and have very long spin coherence times [1–4]. These properties render NV centers the most promising among various solid-state quantum nanoemitters, including quantum dots and molecules. From among these advantageous properties, the optical transitions have been extensively investigated both theoretically and experimentally [5–7]. These transitions give rise to a very narrow (life-time limited 13 MHz [5]) and intense zero-phonon line (ZPL), which is useful for future quantum information processing [8], photonic quantum memories/gates [9,10], and nano-magnetic sensors [11]. For example, such life-time limited linewidth of ZPL allows the generation of narrow-band, indistinguishable single photons [12] required for quantum computation [13], quantum repeaters [14], and quantum metrology [15].

Although top-down approaches for nanophotonic devices using bulk diamonds have been reported [16], NV centers in nanodiamonds have particularly attracted attention as portable robust nanoemitters that can be used in the bottom-up approach [17–22]. For example, highly efficient single photon sources have been demonstrated by coupling single NV centers in nanodiamonds to tapered optical fibers [21, 22]. However, it has been proposed that the optical coherence property of NV centers in nanodiamonds is much worse than that exhibited

in the bulk crystal [23,24]. Photoluminescence excitation (PLE) spectral measurements have reported that there is a large spectral diffusion of ZPL (or spectral jumping of ZPL frequency) from NV centers in nanodiamonds, compared with that observed in bulk diamonds [6,23,24]. In case of these PLE measurements, narrow-band tunable lasers were used to excite the NV centers and the intensity of fluorescence in phonon sideband was measured.

For the applications using photons emitted from the NV centers, photoluminescence (PL) spectra of ZPLs of NV centers can give more direct information of emitted photons than PLE spectra. When NV centers are used as triggered single photon sources [25,26], usually the NV centers are off-resonantly excited with high excitation intensity, and thus the actual coherence time of photons are very different from that estimated by PLE. Very recently, PL spectra of single NV centers in type IIa pure bulk diamonds have been reported [24,25], but PL spectra of NV centers in nanodiamonds have not been reported and thus, a study of high-resolution PL spectra of NV centers in nanodiamonds is necessary.

Here, we report the first measurement of high-resolution PL spectra of single NV centers in nanodiamonds at cryogenic temperatures, using a Fabry-Perot interferometer (resolution 0.1 GHz). In this work, nanodiamonds were spread on thermally oxidized silicon substrates where the phonon sideband of NV centers was suppressed [27,28]. The narrowest ZPL linewidth of a single NV center was 1.2 GHz, and the statistical mean for all the 7 NV centers observed was 1.9 ± 0.7 GHz. Our reported linewidths are comparable to the reported ZPL linewidths in PL spectra of NV centers in pure bulk crystals (from 0.5 to 8 GHz). We observed that the linewidth broadened to 5.4 GHz as the temperature increased upwards to 25 K. We believe that our observation of the ultra-narrow NV ZPL linewidths in nanodiamonds makes it possible to use such nanodiamonds by manipulating them into hybrid nanophotonic devices for quantum information and quantum sensing applications.

2. Experimental methods and setups

We used a type-Ib diamond monocrystalline suspension (Microdiamant, MSY 0–0.1 μm GAF) with a particle-size distribution of 0 to 100 nm. The suspension was purified by a centrifuge to remove fluorescent contaminants and was then dispersed into pure water. A small aliquot of this suspension was spin-coated onto thermally oxidized silicon substrates (2- μm SiO₂ layer on Si substrate) that were cleaned as described elsewhere [27,28]. The substrates were then attached to the cold finger of a liquid-helium flow cryostat (Oxford Instruments, Microstat HiRes-II, temperature stability was less than 0.01 K) for the optical experiments.

Figure 1 shows an experimental setup of our in-house assembled confocal microscope for cryogenic experiments. A continuous-wave He-Ne green laser ($\lambda = 543.5$ nm) was used for the excitation. The laser polarization was converted from linear to circular by a quarter-wave plate. The beam was then focused through an objective lens (Olympus, LCPLFLN 50xLCD) with a numerical aperture of 0.7. The focused laser spot was almost 800 nm in diameter, nearly diffraction limited. The fluorescence from NV centers was separated from the laser using a dichroic mirror and additional filters. A pinhole with a diameter of 20 μm was used to increase the signal-to-noise ratio. The excitation power of 0.8 mW ($\sim 1 \times 10^5$ [W/cm²]) was used in all of the following measurements.

The fluorescence was then analyzed by a Hanbury-Brown and Twiss (HBT) interferometer, a spectrometer (Andor, DU420-OE & Oriel, MS257), and a Fabry-Pérot (FP) interferometer in order to obtain second-order photon correlation histograms [$G^{(2)}(\tau)$], whole PL spectra, and high-resolution PL spectra of ZPLs, respectively. The HBT interferometer consisted of two single-photon counting modules (Perkin Elmer, SPCM-AQRH-14) and a time-correlated single photon counter (Becker & Hickl, SPC-130). The wavelength resolution of the spectrometer was 2 nm. The FP interferometer was equipped with a band-pass filter (14 nm bandwidth at $\lambda_{\text{center}} = 640$ nm) to remove the phonon sideband, and with a pair of lenses to match the fluorescence beam to the focal length of the confocal scanning FP interferometer (Thorlabs, SA210-5B). The FP interferometer had the free spectral range (FSR) of 10 GHz

and the finesse of 100, which corresponds to the frequency resolution of 100 MHz. The finesse was experimentally determined by measuring the 200-kHz-bandwidth laser. The throughput of the FP interferometer was 18%. Because the throughput depends upon the alignment and the transverse mode of the light, the relatively small throughput was likely due to the confocal focusing of the cavity. We also measured a throughput of 18% when using a narrow band laser emitted from a single-mode fiber end.

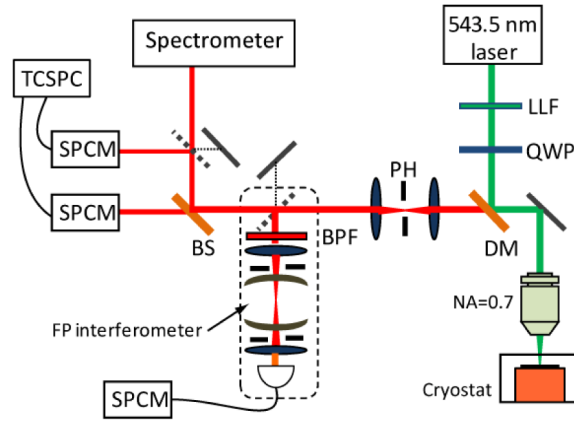


Fig. 1. A schematic picture of the experimental setup. LLF: laser line filter, QWP: quarter wave plate, DM: dichroic mirror, PH: pinhole, BS: 50/50 beam splitter, SPCM: single photon counting module, TCSPC: time-correlated single photon counter, BPF: band pass filter.

3. Results and discussion

A confocal scanning image of nanodiamonds measured at 4 K is shown [Fig. 2(a)]. We observed several well-defined bright spots. The PL spectrum of the bright spot, indicated by an arrow, is shown [Figs. 2(a) and 2(b)]. The PL spectrum exhibits an intense ZPL peak at around 637 nm and a broad phonon sideband in the range of 650–750 nm, and these observations are typical spectral features of negatively charged NV centers. The Debye-Waller factor (F_{DW}) of this PL spectrum was $F_{DW} = 11.7\%$, which indicates a high fraction of ZPL emission by the phonon sideband suppression effect from thermally oxidized silicon substrates, an observation that is consistent with those previously reported [27,28]. Fluorescence observed in the range below 637 nm is phonon sidebands of neutral NV centers (see ref. 27 for details).

A second-order photon correlation histogram of the photoluminescence from these nanocrystals is shown and the results indicate anti-bunching at time zero [Fig. 2(c)]. The red curve is a theoretical fit to the data by assuming a three-level electronic system of NV centers [Fig. 2(c)]. We obtained $G^{(2)}(0) = 0.29$, a value less than 0.5 which indicates that the NV center was single. To obtain a functional working distance, we used a microscope objective with a numerical aperture of 0.7, one that cannot allow for efficient collection of fluorescence from NV centers. The nonzero value of $G^{(2)}(0)$ possibly originates from the large background fluorescence relative to that of NV.

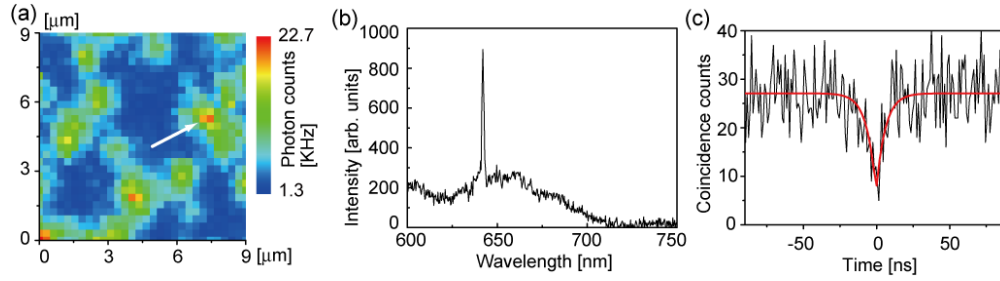


Fig. 2. (a) A scanning confocal image of diamond nanocrystals on the thermally oxidized silicon substrates. (b) The entire PL spectrum of the NV center indicated by an arrow in Fig. 2(a). (c) The second-order photon correlation histogram of this NV center, showing $G^{(2)}(0) = 0.29$.

A high-resolution PL spectrum of the ZPL of this single NV center as measured by the FP interferometer is shown (Fig. 3). The Gaussian fit to the data shows a frequency linewidth of 1.2 GHz. Use of a Gaussian fit is supported by considering the long acquisition time of the high-resolution PL spectrum. Each data point in Fig. 3 expresses photon counts in 10-second intervals such that the total accumulation time was about 10 min. In such a long acquisition time, it is known that the spectral shape becomes Gaussian due to the large spectral diffusion of NV ZPL peak (an inhomogeneous process), despite that ZPL of NV centers show a lifetime limited homogeneous linewidth at a moment in a PLE scanning measurement [23,29]. The frequency lateral axis was calibrated to the 10-GHz FSR by measuring the separation of two cavity resonance lines in the data. The instability of the FP interferometer caused the linewidth fluctuation of ± 0.07 GHz (vide infra), negligibly small for the current linewidth determination.

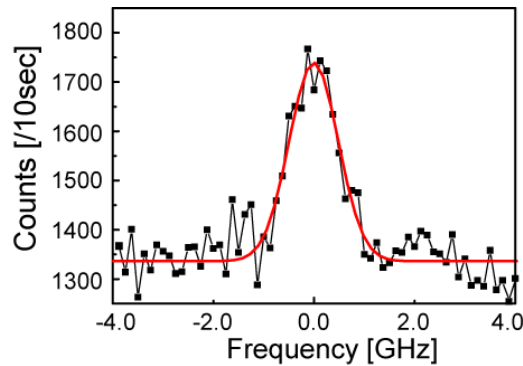


Fig. 3. The high-resolution PL spectrum of the single NV center incorporated in the nanodiamond in Fig. 2, measured at 4 K. The red curve is a Gaussian fit to the data.

We investigated many other nanodiamonds containing single NV centers. NV centers in each of these nanodiamonds showed very different characteristics due to their differences of inner stress, impurities and surface trapping states. To obtain the ZPL fine structure, it was necessary to preselect nanocrystals. The measurement of both the whole PL spectra and $G^{(2)}(\tau)$ histograms makes it easy for us to identify NV centers that show ZPL fine structures in FP interferometer experiments. Specifically, we selected nanodiamonds that incorporated single NV centers that showed large Debye-Waller factors ($F_{DW} > 8\%$). We measured the ZPL peaks in FP experiments for 6 nanocrystals out of several hundreds of nanocrystals, all showing more than 8-% Debye-Waller factors and $G^{(2)}(0)$ less than 0.5. The optical properties of these 6 nanocrystals are summarized in Table 1. These crystals exhibited frequency linewidths from 1.2 to 2.8 GHz, rendering a statistical value of 1.9 ± 0.7 GHz [30]. The other many nanocrystals did not show ZPL peaks in FP experiments despite their single, well-

defined ZPL peaks in the entire (low-resolution) PL spectra. These nanodiamonds show linewidths that are much larger than the 10-GHz FSR that were measured with the FP interferometer.

Table 1. Optical properties of the 6 nanocrystals. $\Delta\nu$: ZPL linewidth, F_{DW} : Debye-Waller factor. The nanocrystal #1 is the one shown in Figs. 2 and 3.

	#1	#2	#3	#4	#5	#6	Statistics
$\Delta\nu$ [GHz]	1.2	1.4	1.4	2.2	2.4	2.8	1.9 ± 0.7
F_{DW} [%]	11.7	10.0	10.3	9.2	9.6	10.9	10.3 ± 0.9
$G^{(2)}(0)$	0.29	0.40	0.29	0.23	0.21	0.30	0.29 ± 0.07

The ZPL linewidth of single NV centers in different types of diamonds is shown (Table 2). Single NV centers in our type Ib nanodiamonds showed a frequency linewidth almost comparable to the linewidths observed in type IIa bulk diamonds [24,25]. This result is in striking contrast to the fact that the optical properties of the NV centers in type Ib nanodiamonds are considered to be inferior to the optical properties of high-purity type IIa bulk diamonds. Indeed previous PLE measurements on type Ib nanodiamonds reported a large spectral diffusion of the ZPL peak, which can estimate the resultant linewidth to be tens of GHz, if the PL spectra is measured directly [6].

Table 2. Comparison of ZPL linewidths of single NV centers in different type of diamonds as measured by direct PL measurement at cryogenic temperatures.

Diamond type	Temperature	Linewidth	Ref.
Bulk type IIa	9 K	~ 0.5 GHz	[25]
Bulk type IIa	< 10 K	8 GHz	[24]
Nanodiamond type Ib	4 K	1.2 GHz (1.9 ± 0.7 GHz in average)	This work

We also measured the temperature dependence of ZPL peaks of single NV centers in diamond nanocrystals. We tracked the NV centers against the positional drift of the NV centers due to a change in temperature. A series of the ZPL spectra measured in a given temperature range (4–25 K) and the ZPL linewidth as a function of temperature are shown [Figs. 4(a) and 4(b)]. The ZPL spectrum of this single NV center had a linewidth of 1.36 GHz at 4 K. Interestingly, even while temperature increased from 4 to 10 K, the ZPL linewidth showed a very small increase, from 1.36 to 1.42 GHz. When the temperature was further increased to 20 K, the linewidth increased gradually but remained less than 2.0 GHz. The linewidth was rapidly increased with temperatures above 20 K, rendering a ZPL linewidth of 5.4 GHz at 25 K. The next cavity resonance line partly appeared in the range 10–12 GHz at 25 K [Fig. 4(a)].

The stability of the FP interferometer has been characterized independently by measuring the cavity transmission line of an external cavity laser diode (emission wavelength: 780 nm, linewidth: 200 KHz), under the exact conditions used for the above measurements. We found that the maximum central frequency shift of the cavity resonance line was ± 0.84 GHz, and that the instability of the FP interferometer induced the maximum linewidth fluctuation of ± 0.07 GHz. These additional data show that the temperature dependence of the linewidth of the ZPL is not affected by instabilities inside the cavity.

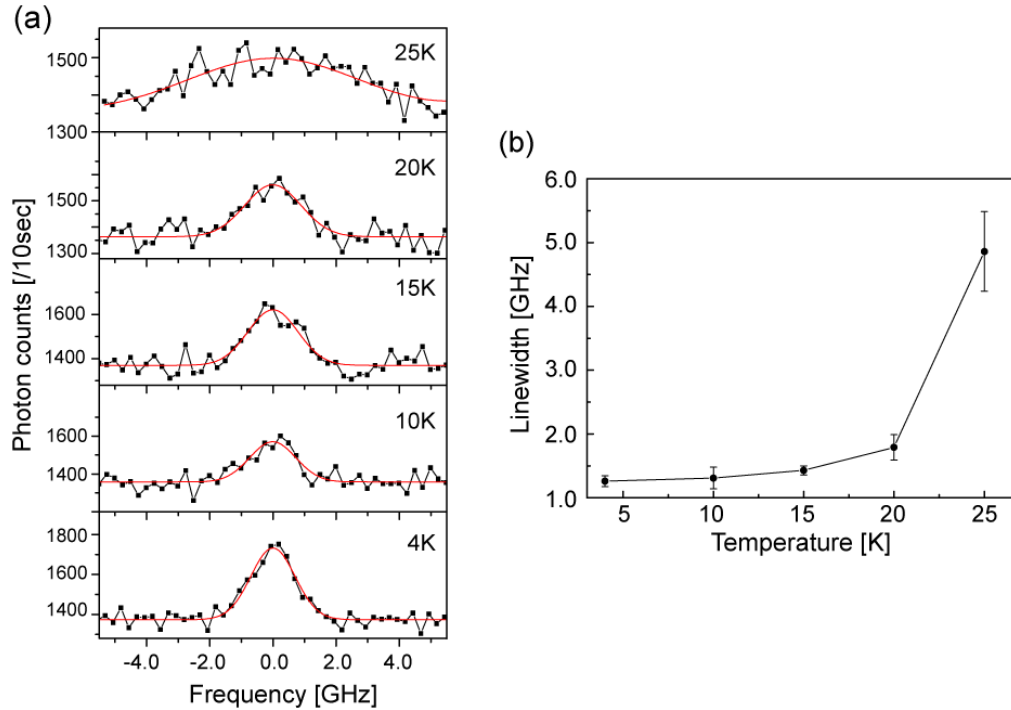


Fig. 4. (a) The high-resolution PL spectra of the ZPL at temperature from 4 to 25 K. The line connecting the dots is the guide for the eye. The red curve is a Gaussian fit to the data. The fitting error is added as the error bar. (b) The ZPL linewidth as a function of the temperature.

The observed ZPL linewidths in our PL spectra were somewhat broader than the previously reported values of the long-timescale linewidth of the PLE spectra of single NV centers in nanodiamonds [6, 31]. Such a linewidth difference between the PL and PLE was also reported in bulk diamonds [24, 25]. Nevertheless the clear mechanism has not been known. We therefore point out possible two reasons of this difference here. (1) First, the measurement time-scale is different between these two methods. In the PL measurement, the acquisition time for the one data and the overall measurement time are very long; 10 seconds for the one data and 10 minutes for the overall time in our case. In contrast the PLE measurement usually acquires a single scan spectrum for several seconds and the long-timescale data was about a minute. This large difference of the measurement timescale may cause the linewidth broadening in the PL spectra. (2) Second, the excitation mechanism is different. The PL measurements use very high intense green laser to off-resonantly excite the NV centers to directly see the ZPL of the PL, whereas the weak narrow-linewidth laser directly excites the NV center at the ZPL in the PLE measurement. Such a different excitation mechanism may cause the difference between these two methods too.

It should be noted that the existence of nanodiamonds with single NVs exhibiting outstanding optical properties presented here is very useful for the integration of nanophotonic devices in a hybrid approach. Once we find a nanodiamond having a single “excellent” NV center by PL measurement, we are able to manipulate that nanodiamond by so-called Pick-&-Place technique and are able to couple it with high-Q microcavities or other nanophotonic structures [17]. In this case, the small population of such “excellent” nanodiamonds is not a critical problem, even though the improvement of the ensemble-average property of NV centers is preferred. Thus our first observation of ultra-narrow ZPL linewidths of NV centers in nanodiamonds will stimulate further researches in hybrid diamond nanophotonics.

4. Conclusion

In summary, this work reports narrow ZPLs in PL spectra of single NV centers in diamond nanocrystals, in which phonon sidebands of the PL spectra were suppressed. Using a Fabry-Perot interferometer at cryogenic temperatures, we observed the narrowest ZPL linewidths of 1.2 GHz. The temperature dependence of the ZPL linewidth was also measured and the observed values are comparable to the ones reported for NV centers in pure bulk diamond. Our observations of such ultra-narrow ZPL linewidths of NV centers in nanodiamonds gives rise to the possibility of utilizing nanodiamonds in hybrid photonic nanodevices for quantum information and quantum sensing applications.

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