Creating Single Color Centers in Nanodiamonds with Ion Implantation

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Abstract: We report the first demonstration of single silicon vacancy center creation in 20 nm nanodiamonds using silicon ion implantation combined with thermal annealing. Room-temperature single photon emission with linewidth below 10 nm is observed.

1. Introduction

The development of bright, stable quantum emitters is critical for the advancement of multiple areas across bioscience and quantum photonic technologies [1,2]. Specifically, the negatively charged silicon vacancy (SiV^-) center in diamond has received significant interest due to its narrow zero-phonon line (ZPL), a high Debye-Waller factor and interesting spin properties, making it promising for obtaining indistinguishable photons as well as hybrid quantum systems involving a spin-photon or spin-phonon interface [3]. Compared to bulk diamond, nanodiamonds (NDs) containing SiV^- centers possess unique advantages to realize the deterministic integration of emitters with photonic and/or plasmonic structures for quantum emission engineering as well as to be used for bio-imaging, drug delivery or other biomedical applications.

NDs containing SiV^- centers can be fabricated using several reported methods. One is synthesizing NDs chemically with the introduction of Si atoms during the synthesis. In the second method, NDs are obtained from the mechanical milling of bulk diamond that contains SiV^- centers. The third method involves NDs formed through detonation with Si-containing dopants. Recently, ion implantation has been demonstrated to create SiV^- ensembles in NDs. Compared to methods above, ion implantation offers the unique potential to control the number and positions of quantum emitters created. However, the creation of single color centers in NDs using this technique remains a challenge, likely due to the higher susceptibility of NDs to ion beam damage as well as their small sizes that complicate the optimization of implantation conditions. In this work, we report the creation of high-purity single SiV^-centers in NDs using silicon ion implantation. The photophysics of the resulting color centers and their inhomogeneous distribution of emission wavelengths are investigated. In addition, the impact and potential applications of the implantation method are discussed.

2. Results and Discussion

Non-fluorescence NDs with an average size of 20 nm were obtained from Adamas Nanotechnologies and drop-casted onto quartz coverslips with particles closely packed. For the Si ion implantation experiments, the ion energy of 12 kV was used, corresponding to an average Si ion penetration depth of 10 nm as obtained from the Stopping and Range of Ions in Matter (SRIM) simulation. To investigate the impact of ion fluence at room temperature, four different fluences, i.e., $2x10^{10}$, $1x10^{12}$, $5x10^{13}$ and $1x10^{15}$ ions/cm^2, were tested. Thermal annealing was performed following ion implantation as an essential step to form stable, bright color centers. The annealing procedure was adopted from Ref [4]. The samples were then checked with a home-built confocal microscope under the excitation of a 690 nm laser. It was found that only the sample implanted with $1x10^{15}$ ions/cm^2 contained stable emitters that gave well-defined, sharp emission lines. For example, Fig. 1(a) is a PL map scanned over one area on this sample, show isolated bright emission spots. The emitter noted as "SPE" was identified to possess a sharp emission peak centered at 764.7 nm, with a full width at half-maximum (FWHM) of 6.5 nm (Fig. 1(b)). It exhibited stable photoluminescence emission during the optical characterization without notable blinking or bleaching. Second-order autocorrelation measurement performed on this emitter revealed $g^{(2)}(0) = 0.01$ (Fig. 1(c)), indicative of high-purity single photon emission. Its excited-state lifetime was estimated to be 2.71 ns from the $g^{(2)}(t)$ measurement, agreeing

with the typically reported lifetimes of SiV^- centers at room temperature. The emitter possessed a brightness of ~70 kcps measured using an air objective configuration (Fig. 1(d)).

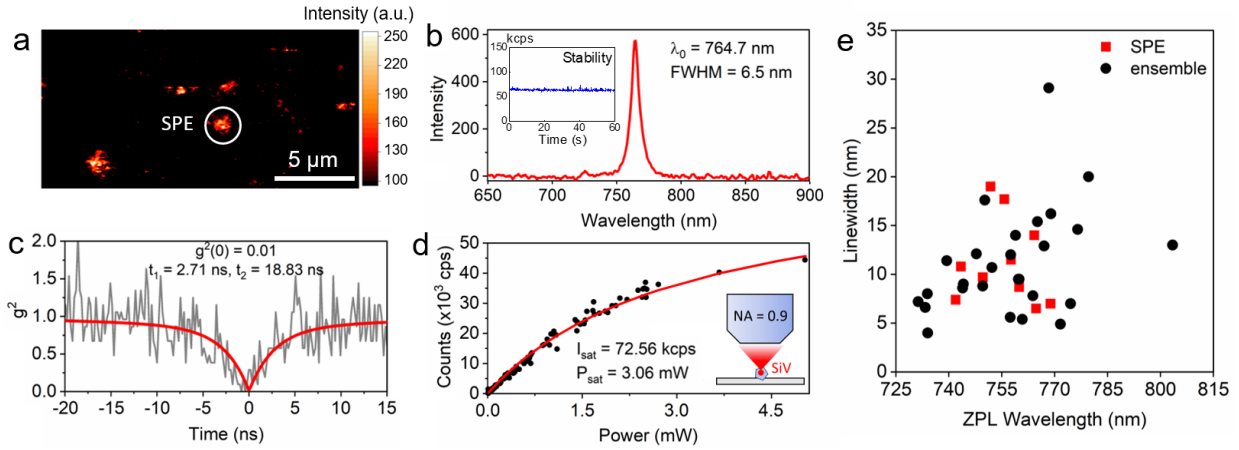

Fig. 1. (a) PL map scanned over a nanodiamond (ND) sample after ion implantation ($1x10^{15}$ ions/cm^2) and thermal annealing. The emitter circled in white, noted as "SPE" was selected for the following measurements. (b) – (d) Emission spectra and stability curve (b), second-order autocorrelation measurement using a three-level fitting model: $g^{(2)}(t) = 1-\alpha\cdot\exp(-|t|/t_1) + \beta\cdot\exp(-|t|/t_2)$, (c) and saturation curve d) of emitter "SPE" under the excitation of a 690 nm laser. Inset of (d) illustrates the collection configuration used for ND characterization. (e) Distribution of zero-phonon line (ZPL) wavelength and linewidth of both single-photon emitters (SPEs) and emitter ensembles observed in experiments.

The ZPL wavelengths and emission linewidths of identified emitters, including both single-photon emitters (SPEs) and ensembles, vary significantly. This is summarized in Fig. 1(e) with data collected from 37 stable emitters on the sample implanted at $1x10^{15}$ ions/cm^2. The ZPL wavelengths are observed to span over a broad spectral range from 730 nm up to 803 nm, while the ZPL linewidths are measured in the range of 4 nm and 29 nm. An important cause of the emission line shift and linewidth broadening is the higher strain or residual stress in NDs compared to bulk diamond. However, further investigations are still needed to determine if other mechanisms are involved and more complicated Si-related defects, other than SiV^- centers, are formed. Our successful fabrication of single color centers, that are free from the effects of center-center interaction and ensemble averaging, provides an excellent platform for future research on this topic.

Our work greatly expands the application of ion implantation in the field of quantum photonics. It opens up new possibilities to leverage NDs containing single narrowband color centers for quantum information technologies. Color centers in sub-50 nm NDs are uniquely compatible with nanoscale plasmonic cavities and hybrid plasmonic-dielectric resonators that can provide orders of magnitude higher Purcell enhancements than dielectric ones [5,6]. Integrating ND-based group IV color centers with nanophotonic structures promises ultrafast single photon emission of on-demand indistinguishable photons at cryo-free temperatures. Furthermore, considering the bio-compatibility of NDs and the SiV^- emission wavelength in the near infra-red regime, NDs with ion implantation-induced single SiV^- centers will contribute to the advancement of quantum-based bio-imaging.

3. References

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