



Optical properties of implanted Xe color centers in diamond

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ABSTRACT

Optical properties of color centers in diamond have been the subject of intense research due to their promising applications in quantum photonics. In this work we study the optical properties of Xe related color centers implanted into nitrogen rich (type IIA) and an ultrapure, electronic grade diamond. The Xe defect has two zero phonon lines at ~ 794 nm and 811 nm, which can be effectively excited using both green and red excitation, however, its emission in the nitrogen rich diamond is brighter. Near resonant excitation is performed at cryogenic temperatures and luminescence is probed under strong magnetic field. Our results are important towards the understanding of the Xe related defect and other near infrared color centers in diamond.

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Luminescent defects in diamond are important for a variety of applications spanning quantum nanophotonics, bio labeling, light emitting diodes, superconductivity and spintronics [1–6]. Some of these defects — namely the nitrogen vacancy (NV) [7], the silicon vacancy (SiV) [8–11] and more recently the germanium vacancy (GeV) [12,13] have been subject to rigorous research owing to their relatively known crystallographic and electronic structures. Now, however, there is an increased interest in the study of color centers emitting further in the infra-red, partially, due to the potential integration with free space quantum key distribution and the telecommunications bandwidth [14,15].

An example of near infra-red color centers in diamond is the Xe related defect with two zero phonon lines (ZPLs) at 794 nm and at 812 nm [16–20]. The crystallographic structure of the Xe color center is still under debate, however it is believed to be an interstitial xenon splitting two vacancies along the $\langle 111 \rangle$ crystallographic direction [21]. This was supported by polarized luminescence measurements, which assign the high and low energy ZPLs respectively to σ - σ (XY–XY for absorption and emission, respectively) and σ - π (XY–Z) transitions at a $\langle 111 \rangle$ -oriented center [18]. A particularly promising aspect of this color center is the ability to form it via ion implantation. Furthermore, the Xe related center has a relatively low Huang Rhys factor of ~ 0.3 , indicating that most of the photons are emitted into the ZPL, which is

beneficial for efficient filtering [16–18]. Finally, there is an increasing interest to explore fluorescent defects that include heavy ions in their crystallographic structure, resulting in lower phonon-mediated orbital relaxation, thus enabling longer spin coherence times [22].

Earlier works showed that Xe related color centers could be formed using ion implantation [16–18]. In this work, we study in detail the optical properties of the Xe defect, and show that its intensity depends on the nitrogen concentration within the host material. A series of variable dose Xe ion implantations is performed to determine the emission brightness. Furthermore, the lifetime and polarization properties of the Xe related defect are measured and photoluminescence (PL) excitation at cryogenic temperatures is performed. Luminescence under increased magnetic field is also done to study the potential Zeeman splitting of the Xe related center.

To engineer the Xe related defects, ion implantation is performed into two separate diamonds – a type IIA chemical vapor deposition (CVD) diamond with 1 ppm nitrogen and an electronic grade CVD diamond with <1 ppb nitrogen (see Fig. 1(a)) – both manufactured by Element Six. Using these two samples enables to correlate between the PL intensity of the Xe emitters and the nitrogen dopant concentration in diamond. Ion implantation was performed at room temperature using a National Electrostatics Corporation 4UH ion accelerator with 500

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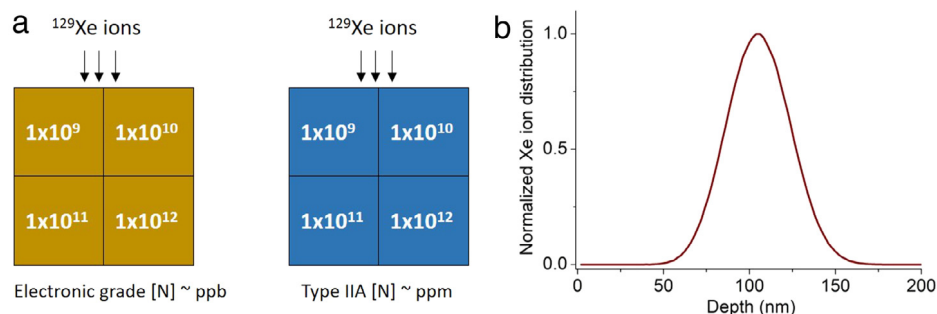


Fig. 1. (a) 500 keV ^{129}Xe ions were implanted into two separate diamonds — electronic grade with $\sim$ ppb nitrogen concentration and type IIA with $\sim$ ppm nitrogen concentration. The doses are given in ions/cm² (b) Implantation depth profile of 500 keV ^{129}Xe ions into single crystal diamond calculated using the SRIM software package. A single Gaussian fit to the implantation profile produces a peak depth of 105 nm and a FWHM of 26 nm.

keV $^{129}\text{Xe}^+$ ions to a total dose of 1×10^{12} , 1×10^{11} , 1×10^{10} , 1×10^9 Xe ions/cm² (Fig. 1(a)). The dose rate was $3 \times 10^{12} - 5 \times 10^{12}$ ions/cm²/s depending on the respective quadrant. Irradiations were performed at 7° off the [100] direction to minimize channeling effects. These doses have been used previously for various impurities (Si, N and others) to study dose dependent luminescence and identification of single quantum emitters [6]. The high implantation energy of 500 keV was chosen to avoid xenon ions in a close proximity to the diamond surface. The depth profile of implanted ^{129}Xe atoms under experimental conditions was calculated using the SRIM software package [23] (density = 3.515 g/cm³, displacement energy = 40 eV) (Fig. 1b). A single Gaussian fit to the implantation profile calculated by SRIM produces a peak at a depth of 105 nm and a half width at half maximum of 26 nm. After implantation, all samples were annealed at 1400 °C for one hour under a pure nitrogen atmosphere. Under these annealing conditions, nitrogen is not expected to diffuse into the diamond.

Optical measurements were performed at room temperature using a standard confocal microscope with a high numerical aperture objective (0.7 N.A.). Green (532 nm) and red (633 nm) lasers were used for off resonant excitation, and a pulsed 675 nm (40 MHz, 80 ps pulses) laser for lifetime measurements. For the resonant excitation (PLE) measurements a Ti:Sapphire laser ($\sim$ sub 5-MHz linewidth) that is tuned to the high energy ZPL ($\sim$ 791 nm) was used. The PLE scan experiment was performed by scanning a piezo etalon within a mode hop-free range of $\sim$ 50 GHz and detecting the off resonant phonons or the 2nd ZPL ($\sim$ 812 nm). A wave meter was used to monitor the wavelength of the excitation laser, and provide feedback to stabilize the laser at a particular wavelength.

First, we studied the formation of Xe related color centers in the implanted diamonds. Fig. 2(a) shows luminescence spectra from the electronic grade sample using green (532 nm) excitation at room temperature. The Xe related ZPL doublet is clearly observed in the higher implantation dose region (1×10^{12} Xe ions/cm²) and is still observed at the lower dose of 1×10^{11} Xe ions/cm². However, no signature of the Xe related defects was observed from the lowest implantation doses (1×10^9 , 1×10^{10} Xe ions/cm²), even with high resolution confocal scanning. Notably, in the 1×10^{10} Xe ions/cm² region, single nitrogen vacancy (NV) emitters were found (as expected from the electronic grade diamond after ion irradiation and annealing). This implies that the quantum efficiency of the Xe related color center is at least an order of magnitude lower than that of the NV when pumped at room temperature using a 532 nm laser.

To gain increased information about the Xe related defects, we compared the fabricated emitters in the electronic grade and type IIA diamond samples. Fig. 2(b) shows a comparison of the emitters excited by the 633 nm laser. We note that this comparison is not possible using 532 nm excitation since emission spectra of the type IIA sample under this condition is dominated by NV fluorescence (not shown). The intensity of the Xe related emission in the nitrogen rich sample is an order of magnitude higher than in the ultra-pure material. This suggests

that the Xe center is negatively charged, since the presence of nitrogen is likely to provide excess electrons for defect formation. A similar effect has also been reported for the negatively charged silicon vacancy (SiV) defect in diamond, which displays an increased intensity in the presence of nitrogen [24]. Despite the higher efficiency in nitrogen rich diamond, we were not able to identify single Xe related centers in the regions of lowest implantation doses even in the type IIA sample.

Next, we measured the fluorescence lifetime of an ensemble of Xe emitters (Fig. 3). A very short excited state lifetime of $\sim$ 0.73 ns and $\sim$ 0.77 ns was measured for the Xe related color centers in type IIA and the ultrapure diamond, respectively. The laser decay is shown in comparison to elucidate the system response ($\sim$ 0.43 ns). Further analysis (Fig. 3c) shows that the lifetime in the 1×10^{12} Xe ions/cm² implanted region is slightly shorter than the emitters' lifetime in the 1×10^{11} Xe ions/cm² region (for the ultra pure sample). This is most likely associated with more non radiative channels due to an increased irradiation dose. The extremely short lifetime further supports the presence of non-radiative relaxation pathways from the excited state via phonon transitions. The observed lifetime is nevertheless comparable to the fluorescence lifetime of the SiV defect, which despite being a very dim emitter, can indeed be isolated as a single site after fabrication by ion implantation [25].

While isolation of single Xe based emitters was not successful using off resonant excitation at room temperature, the optical properties of Xe emitters at cryogenic temperatures can still be investigated. For these experiments we choose the sample with the brighter Xe emission, namely the type IIA diamond. Fig. 4a shows an off resonant excitation of an ensemble of Xe emitters recorded at 4 K. Both the high energy ZPL ($\sim$ 791 nm) and the low energy ZPL ($\sim$ 811 nm) are visible at cryogenic temperature. The full width at half maximum (FWHM) of both ZPLs were reduced to $\sim$ 0.88 nm and $\sim$ 0.81 nm, respectively. The inset of Fig. 4a reports on the emission polarization measurement of 811 nm ZPL that confirms the probed Xe centers are fully polarized. As shown in Fig. 4b, the Xe related center has two excited states. Due to the fast phonon relaxation passage (indicated by the thick dash down arrow), population in the upper excited state is lower than that of the lower ground state. Hence the 811 nm ZPLs is stronger than the 794 nm ZPL at low temperature, suggesting the thermalization of the 794 nm line.

To further demonstrate resonant excitation, we excited the high energy ZPL at 791.5 nm and collected the second ZPL and the associated phonon side bands as shown in Fig. 4(c). We note that the implanted Xe color centers exhibit some inhomogeneity and hence the ZPL can shift by several nm from its maximum at 794 nm. The observed spectra reveals several narrow lines, that are spectrometer limited ($\sim$ 120 GHz) and a weak phonon side band. While these lines are likely to correspond to single emitters, we were not able to record an autocorrelation measurement from an individual line.

Fig. 4(d) shows the lower energy ZPL under resonant excitation of the 791.5 nm line. The line is broadened due to strain or ultrafast spectral diffusion. We tune the excitation wavelength across 791.5 nm,

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