

Light emission from color centers in phosphorus-doped diamond

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Abstract. Light emission from color centers in diamond is being extensively investigated for developing, among other quantum devices, single-photon sources operating at room temperature. By doping diamond with phosphorus, one obtains an n-type semiconductor, which can be exploited for the electrical excitation of color centers. Here, we discuss the optical properties of color centers in phosphorus-doped diamond, especially the silicon-vacancy center, presenting the single-photon emission characteristics and the temperature dependence aiming for electroluminescent single-photon emitting devices.

1 Introduction

Single-photon emitters in the solid state represent an essential ingredient for developing quantum photonics technologies, such as quantum cryptography and linear optical quantum computing [1,2]. Color centers in diamond are promising in this regard because they exhibit stable emission characteristics, and they can be operated at room temperature. In particular, the silicon-vacancy (SiV) center provides most of its emission in a narrow zero-phonon line (ZPL) [3] and it withstands large temperatures [4].

For device miniaturization and energy efficiency, it is desirable to excite the color center electrically. This requires electron-hole recombination at the color center, hence charge transport in diamond. Early work demonstrated emission from nitrogen vacancy and SiV centers in p-i-n diodes [5,6]. However, growing p-i-n diamond structures is still quite challenging, and the devices are poorly reproducible. An alternative approach relies on Schottky diodes, where the color centers are placed in n-type diamond and holes are injected through the metal contact, if the Schottky barrier is reasonably small. This would only require an n-type diamond layer and the deposition of metal contacts, hence greatly simplifying the device structure.

2 Silicon-vacancy centers in n-type diamond

2.1 Sample fabrication

To obtain n-type diamond, the material can be doped with phosphorus (P) during microwave (MW) plasma enhanced (PE) chemical vapor deposition (CVD) growth. Specifically, one P-doped single-crystal diamond film (sample A) was grown on a diamond substrate utilizing an in-house built 2.45 GHz MWPECVD reactor. The hydrogen (H₂) rich plasma contained 0.09% of methane (CH₄) with a PH₃/CH₄ ratio of 4300 ppm. Two more P-doped homo-epitaxial samples were grown similarly by MWPECVD in a 2.45 GHz ASTeX PDS17 reactor with a water-cooled molybdenum substrate holder. The H₂ plasma contained 0.15% CH₄. One of the samples (sample B) was grown with a P-gradient, with the PH₃/CH₄ ratio adjusted in steps of 0, 250, 500, 1000, 2500, 5000, 10,000, and 20,000 ppm. The other sample (sample C) was grown with a constant 5000 ppm PH₃/CH₄. The H₂ and CH₄ gasses are filtered to <1 ppb (9 N) purity, and the PH₃ is used from a source diluted to 200 ppm in H₂.

The three samples underwent Si-ion implantation based on a 3 MV Tandatron accelerator (High Voltage Engineering Europe) equipped with a HVEE 860 Negative Sputter Ion Source. Aluminum (Al) metal foils were used to decrease the ion energy down to a few tens of keV for shallow implantation (within 200 nm from the surface). A custom designed furnace (1200 °C in high-vacuum conditions ~ 10⁻⁷ mbar) enables the activation of the SiV color centers in P-doped samples. The samples have been Si-implanted with five different fluences (~10¹⁴

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cm^{-2} , $\sim 10^{13} \text{ cm}^{-2}$, $\sim 10^{12} \text{ cm}^{-2}$, $\sim 10^8 \text{ cm}^{-2}$ and $\sim 10^7 \text{ cm}^{-2}$). The expected implantation depth is $\leq 200 \text{ nm}$. Details on the implantation process can be found in Ref. [7].

2.2 Optical properties

Since the inclusion of phosphorous modifies the host environment, the first important question is whether SiV centers exhibit optical properties comparable with intrinsic diamond and if it is possible to obtain single-photon emission [5].

We investigated the three diamond samples with different phosphorus doping profiles. Firstly, we clarified that co-doping with nitrogen causes a significant fluorescence background related to nitrogen vacancy (NV) centers that prohibits the observation of single SiV centers. Secondly, we observed that for samples implanted with low silicon ion fluences the emission background is significantly reduced. Figure 1 illustrates the emission spectrum under different ion fluences.

For low implantation fluences on samples with low nitrogen content (sample C) we finally observed single-photon emission from SiV centers [7]. While the lifetime is comparable to that of SiV centers in intrinsic single-crystal diamond, the ZPL is broader, probably because the host environment is more defective than intrinsic diamond. In this regard, low-temperature measurements would be needed to better understand the broadening effect.

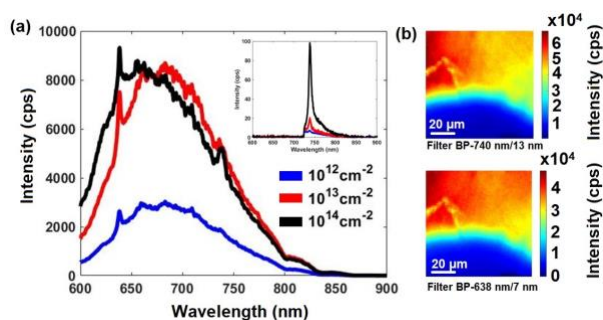


Fig. 1. Photoluminescence spectra in the regions of sample A implanted with a fluence of 10^{12} cm^{-2} , 10^{13} cm^{-2} and 10^{14} cm^{-2} using 532 nm CW laser show NV-related background. Inset: The background due to NV color centers is reduced by exciting the SiV color centers using 690 nm CW laser, as NV complexes have less absorption in this spectral range. (b) Spatially- and spectrally resolved confocal images indicate that the NV color centers (below) are mainly created at the location where the Si ions are implanted (top) (Figure from Ref. [7]).

2.3 Temperature studies

Since free carriers in P-doped diamond suffer from the relatively high activation energy of donor levels ($\sim 0.6 \text{ eV}$), efficient electrical operation requires room or higher temperatures. We thus performed optical studies of SiV centers as a function of temperature, finding that the SiV center remains photostable up to the experimentally considered temperatures ($100 \text{ }^\circ\text{C}$ in ambient conditions). Furthermore, the temperature dependence of the ZPL in

intrinsic diamond and in P-doped diamond differ. This is likely to be attributed to the different crystalline environment between undoped and doped diamond. Details on the experiments can be found in Refs. [8].

3 Conclusions and outlook

We successfully created and characterized single SiV centers in P-doped (n-type) diamond in view of developing optoelectronics light emitting devices. While low-temperature studies would be necessary to more deeply understand the properties of SiV centers in n-type diamond for quantum applications, the temperature studies indicate that for classical light emission the system is promising. For instance, while conventional light-emitting devices (LEDs) degrade their performances when temperature increases, diamond-based LEDs would exhibit better performances under large temperatures [9] and operate in a much wider temperature range, thus offering a unexplored applications for LEDs.

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