Ion Beam Fabrication of Graphitic Structures in Single-Crystal Diamond for Electrically-Stimulated Luminescence

S. Ditalia Tchernij[1,2], G. Andrini[2,3], P. Aprà[1,2], M. Campostrini[4], D. Carlucci[4], E. Corte[1], I. Degiovanni[2,5], M. Genovese[2,5], L. La Torre[4], C. Marinelli[6], E. Moreva[5], V. Prestopino[6], V. Rigato[4], P. Traina[5], V. Varzi[1], G. Verona[6], C. Verona Rinati[6], P. Olivero[1,2], J. Forneris[1,2], F. Picollo[1,2]

[1] Physics Department and Centro inter-dipartimentale "NIS", University of Torino, Torino, Italy.
[2] INFN section of Torino, Torino, Italy.
[3] Department of Electronics and Telecommunications (DET), Polytechnic di Torino, Torino, Italy.
[4]INFN, Nazional Laboratory of Legnaro, Legnaro (Padova), Italy.
[5] Nazional Instisute of Metrologic Research, Torino, Italy.
[6] Department of Industrial Engineering, University of Roma "Tor Vergata" (Roma), Italy

INTRODUCTION

The development of diamond devices based on the functionalities of sub-superficial graphitic micro-electrodes for applications in biosensing has been investigated in several works [1-3]. The fabrication method relies on the direct writing in the single-crystal diamond bulk of amorphized channels by the selective damage induction associated with the Bragg's peak of MeV ions. The crystal volume in which the radiation-induced vacancy density exceeds a threshold value converts to nano-crystalline graphite upon thermal treatment at temperatures above 900 °C, while the diamond lattice is partially recovered where the radiation damage density is lower [4].

In this work, we investigate the potential exploitation of the fabrication technique for the electrical excitation of color centers in diamond, aiming at the development of practical single-photon-emitting devices based on the efficiency and stability of the above-mentioned centers, for applications in quantum optics [5].

With this purpose, a diamond device with four independent buried graphitic electrodes was fabricated by means of MeV ion beam lithography. Differently from previous works, in which the electrical excitation of color centers is achieved by means of diodes and p-i-n structures [6], the device proposed in this work exploits the current flowing between buried graphitic electrodes, in order to obtain electroluminescence (EL) from the diamond region located in the inter-electrode gap. The light emission properties of the fabricated structure were investigated by means of luminescence mapping and the relevant spectra were analyzed to attribute the active defects in the device under investigation.

DEVICE FABRICATION

The device under test is based on a ~40 µm thick intrinsic diamond film homoepitaxially grown on a commercially available 4×4×0.4 mm^3 type-Ib single-crystal high-pressure high-temperature (HPHT) substrate, using a microwave plasma enhanced chemical vapor deposition process at the laboratories of Rome ''Tor Vergata'' University. A scanning 1.8 MeV He$^+$ ion microbeam (~10 µm of diameter spot size) was used to perform a deep ion beam lithography (DIBL) process at the AN2000 micro-beam line of the INFN National Laboratories of Legnaro. The fabrication methodology is extensively described in previous works [7,8].

Five graphitic channels were defined at a depth of ~3 µm below the diamond surface using a ion fluence of 1.5×10^{17} cm^{-2}, which, according to SRIM2011 simulations, is sufficient to achieve a vacancy density above the graphitization threshold at the end of range.

The implanted sample underwent a thermal annealing process for 2 h in vacuum, in order to promote the conversion of the amorphized regions at the end of the Bragg's peak to a graphitic phase and to concurrently recover the residual structural damage in the region comprised between the buried channels and the diamond surface. The resulting micro-fabricated structure (Fig. 1) consisted of four parallel, ~10 µm wide independent graphitic electrodes, spaced by ~12 µm, plus an additional horizontal electrode, which was not employed in the present work.

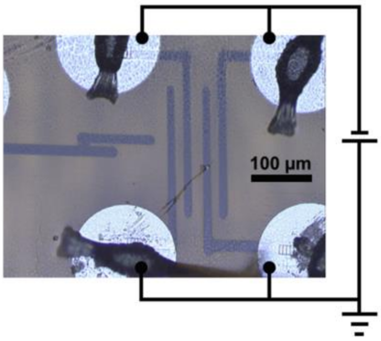

Fig. 1. Optical micrograph of the graphitic electrodes structure after annealing and wire bonding. The electrical connections are schematically presented.

Each graphitic channel was connected to the external circuitry by the deposition of 80 nm thick Cr/Al circular contacts (150 µm diameter), which were exploited as pads for the electrodes wire-bonding. As schematically represented in Fig. 1, the graphitic channels were shorted in pairs, defining a two-electrode inter-digitated geometry.

FUNCTIONAL CHARACTERIZATION

An EL map acquired at a bias voltage of 450 V is shown in Fig. 2. The encoded color scale displays for clarity a maximum emission rate value of 5×10^4 counts per second (cps), each count corresponding to the detection of a single photon; however, the values recorded at the center of the bright spot reached values larger than 10^6 cps. The map clearly shows that the electroluminescent region corresponds to a localized conduction path connecting the buried electrodes (highlighted by the dashed black lines).

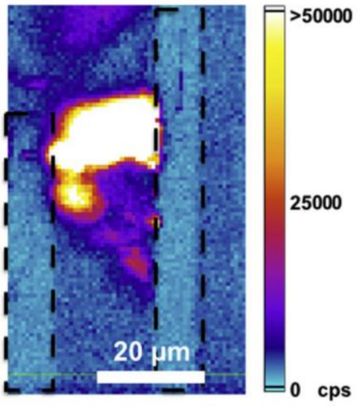

Fig. 2. EL map acquired from the device at an applied voltage of 450 V. The dashed black lines identify the relative position of the graphitic electrodes.

A typical EL spectrum is shown in Fig. 3. The two dips at 428 nm and at 633 nm are instrumental artifacts caused by the presence of a notch filter mounted in the spectrometer, which attenuates the laser excitation when the setup operates in its usual configuration.

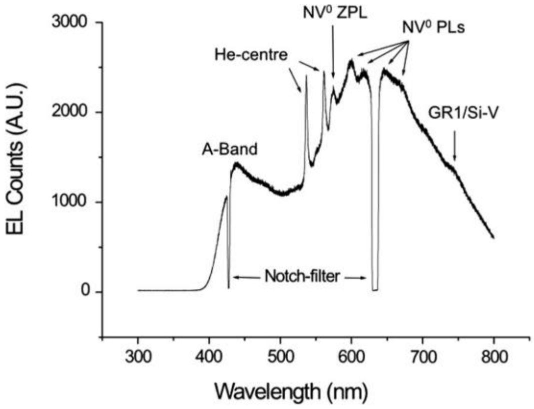

Fig. 3. EL spectrum from the active region. The following features are highlighted: A-band; He-center; NV0 and (tentatively) GR1 or Si-V. A notch filter present in the spectrometer creates two dips at 428 and 633 nm.

Firstly, a ~100 nm broad peak centered at 435 nm is visible. The broad peak is usually referred as the A-band and its origin is commonly attributed to radiative carriers recombination at extended lattice dislocations. Secondly, the two sharp peaks at 536.3 nm and 560.5 nm are attributed to a helium-related defect that was extensively characterized in a previous work [9], consistently with the fact that He^+ was employed for the micro-fabrication process and stray ions could easily have been implanted in the inter-electrode gap region. Thirdly, the peak at 575 nm and its replica at higher wavelengths respectively correspond to the zero-phonon-line and phonon sidebands of the well-known NV^0 center, as extensively observed in cathodoluminescence and EL measurements. Finally, a small peak emerging from the background at ~738–740 nm can be tentatively attributed to the Si-V color centers, since Si could be present as a contaminant in the CVD chamber, and it has been reported to be active in EL in previous works [11]. Alternatively, the peak could be ascribed to the general radiation center GR1 (741 nm), which is associated with radiation-induced vacancies. Although a direct observation in electroluminescence has not been reported so far to our knowledge, the emission from GR1 has been observed in cathodoluminescence regime [12] where a similar excitation process through the injection of electrodes in the diamond dielectric is taking place.

CONCLUSIONS

The reported results demonstrate the possibility of electrically exciting He-related defects, which are characterized by appealing photophysical properties, i.e. high emission rates, sharp spectral emission, negligible phononic sidebands. The possibility to identify and electrically stimulate isolated He-related defects might open to the study of new electroluminescent single-photon sources in diamond for applications in photonics and quantum information.

---

[1] P. Aprà et al., LNL Annual Report 2018 (2019) 1828.
[2] G. Peroni et al., LNL Annual Report 2019 (2020) 126.
[3] F. Picollo et al., Nano Letters 20 (2020) 3889.
[4] R. Kalish et al., Nucl. Instr. Meth. B 106 (1995) 492.
[5] N. Mizuochi et al., Nat. Photonics 6 (2012) 299.
[6] A. Lohrmann et al., Appl. Phys. Lett. 99 (2011) 251106.
[7] F. Picollo et al., New J. Phys. 14 (2012) 053011.
[8] J. Forneris et al., Nucl. Instr. Meth. B 306 (2013) 181.
[9] G. Prestopino et al., Appl. Phys. Lett. 111 (2017) 111105.
[10] J. Forneris et al., Nucl. Instr. Meth. B 306 (2013) 181.
[11] A.A. Melnikov et al., in: Advances in New Diamond Science and Technology, MYU, Tokyo, 1994.
[12] A.A. Melnikov et al., in: Advances in New Diamond Science and Technology, MYU, Tokyo, 1994.
[13] A.M. Zaitsev, Optical Properties of Diamond – Data Handbook, Springer, New York, 2001.