Study of Ion Damage Effects on the Quenching of NV Luminescent Centres in Diamond with Time Correlated Single Photon Counting Spectroscopy

D. Gatto Monticone[1,2,3], S. Borini[4], A. Lo Giudice[1,2,3,5], R. Mercatelli[6], P. Olivero[1,2,3,5], F. Quercioli[6], S. Soria[7], M. Vannoni[6], E. Vittone[1,2,3,5].

[1]Physics Department, University of Torino, Torino, Italy. [2]NIS Centre of Excellence, University of Torino, Torino, Italy. [3]INFN Sez. Torino, Torino, Italy. [4]Thermodynamics Division, Istituto Nazionale di Ricerca Metrologica (INRiM), Torino, Italy. [5]Consorzio Nazionale Interuniversitario per le Scienze Fisiche della Materia (CNISM), Sezione di Torino, Italy. [6]CNR, Istituto Nazionale di Ottica, Firenze, Italy. [7]CNR, Istituto di Fisica Applicata "Nello Carrara" (IFAC), Firenze, Italy.

INTRODUCTION

The NV luminescent centres in diamond attracted increasing interest among the scientific community due to its significant potential in advanced photonic applications such as quantum cryptography [1], quantum information processing [2] and optically-readout magnetometry [3]. The NV centre is based on a nitrogen-vacancy-complex defect incorporated inside the diamond lattice and is characterized – in high purity samples – by high quantum efficiency, long spin coherence time, spin preserving and magnetic-sensitive optical transitions. Ion implantation represents the key technology to deterministically produce NV centres with high spatial accuracy [4], therefore it is of paramount importance to assess the degree of luminescence quenching induced by collateral damage effects associated with ion implantation.

In the present work we report about the systematic investigation of the damage-induced decrease of luminescence efficiency of NV centers in MeV-ion-implanted single-crystal diamond.

ION IMPLANTATION AND SAMPLE PROCESSING

Artificial single-crystal diamond samples produced with the high-pressure-high-temperature technique (HPHT) by Element Six were employed. The samples size is 3×3×0.3 mm^3 and they are classified as type Ib, i.e. their native concentration of nitrogen is comprised between 10 and 1000 ppm and it is in single substitutional form (this introduces an absorption continuum of light above 1,7 eV, thus giving diamonds a yellow color).

The samples were implanted at the microbeam line of the AN2000 accelerator of the Legnaro National Laboratories with 2 MeV H^+ ions. Squared areas with lateral sizes ranging from 100 μm to 200 μm were implanted at different fluences by means of a raster-scanning ion microbeam in order to deliver a uniform fluence. The fluence was accurately monitored by connecting an electrometer to the sample chamber which is electrically insulated from the rest of the beamline, thus acting effectively as a Faraday cup. In order to avoid any surface charging effect, samples were previously coated with 50 nm thick Ag deposition.

The H^+ ion implantation has two principal effects on the optical properties of the luminescent centers in the material:

- inducing the formation of new NV centers by creating vacancies which can recombine with the native population of nitrogen in the sample at temperatures above 600 °C;
- degrading the quantum efficiency of the luminescent centers by introducing non-radiative decay channels through their coupling with structural defects that they creates in the lattice.

After ion implantation, the samples were annealed in vacuum at 800 °C for 2 hours with the purpose of enabling the formation of the optical centers through the thermally-activated recombination of ion-induced vacancies and native substitutional nitrogen impurities, as mentioned above. Figure 1 reports an optical micrograph of the as-implanted sample: areas irradiated at higher fluences appear as darker due to stronger damage effects, while areas implanted at low fluences are invisible.

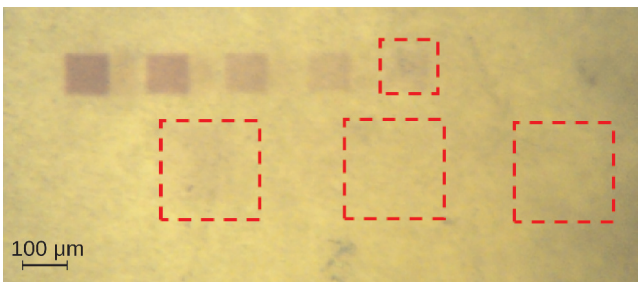

Fig. 1. Optical micrograph of the as-implanted sample; red squares highlight regions where low-fluence implantations have been performed.

The damage profile of 2 MeV H^+ ions is strongly non-uniform, being peaked at the ions end-of-range depth (i.e. ~24 μm), but the probing depth of the confocal mapping technique described below is limited to few μm below the sample surface, therefore in the following characterization the implanted regions will be investigated at depths at which the damage density can be correctly approximated as constant.

CHARACTERIZATION WITH TIME CORRELATED SINGLE PHOTON COUNTING SPECTROSCOPY

The time-correlated single photon counting spectroscopy technique available at the laboratories of the National Institute of Optics was employed to map with high spatial resolution the luminescence lifetime of the luminescent centers in the implanted sample within the spectral region of interest (i.e. at $\lambda = 700 \pm 8$ nm and $\lambda = 595 \pm 8$ nm for the NV^- and NV^0 emissions, respectively), thus accounting for changes in emission efficiency in regions implanted at different fluences. The basic principles of operation of the technique are schematically summarized in figure 2.

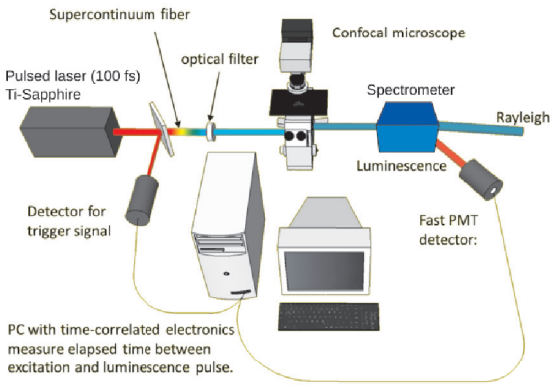

Fig. 2. Schematic representation of the basic operation principles of the time correlated single photon counting technique.

A typical photoluminescence decay chronogram for the NV^- emission from an area implanted at a fluence of 10^{15} cm^{-2} is reported in figure 3. In the inset of the graph we report the PL intensity micrograph highlighting the area from which the lifetime signal has been collected and mediated (i.e. the inner portion of the implanted region).

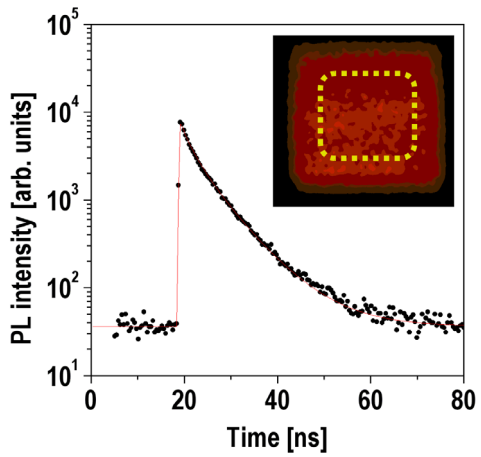

Fig. 3. Photoluminescence lifetime chronogram relevant to the are highlighted in the PL intensity micrograph reported in the inset.

In general the luminescence lifetime chronograms display an excellent consistency with the Förster resonant energy transfer theory [5]. This theory predicts that the luminescent centers under examination can transfer their excitation energy to other defects in the crystal (such as damage-induced structural defects, impurities or other luminescent centers), thus introducing a non-radiative decay rate that limits their quantum efficiency. According to the model, the decay rate can be expressed by the following formula:

$$I(t) = I(0) \cdot \exp\left[-\frac{t}{\tau} - c \cdot \left(\frac{t}{\tau}\right)^{1/2}\right]; \quad (1)$$

where τ is the intrinsic radiative decay time of the luminescent center and c is a parameter proportional to the concentration of quenching defects in the crystal. It is worth stressing that from the value of c it is possible to derive the quantum efficiency η. The intrinsic radiative decay time τ is known from literature: (12 ± 1) ns for the NV^- center and (30 ± 5) ns for the NV^0 center.

The variation of the quantum efficiency was systematically studied as a function of implantation fluence for both NV^- and NV^0 emissions, thus obtaining important information about the quantum efficiency for these centers in presence of different defect concentrations (see figure 4).

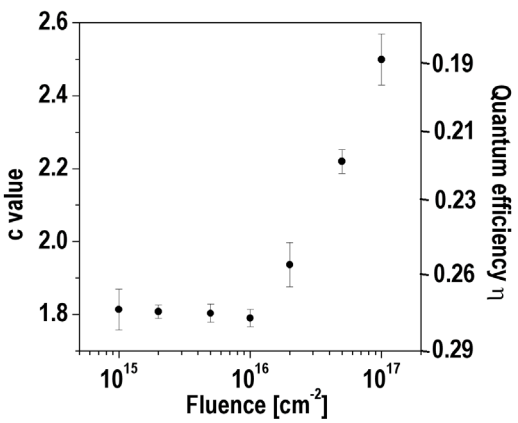

Fig. 4. Systematic variation of c (left axis) and η parameters as a function of implantation fluence.

CONCLUSIONS

Time correlated single photon counting PL spectroscopy demonstrated to be a powerful technique in the investigation of the optical properties of luminescent centers formed in diamond with MeV ion implantation, with promising applications in the engineering of color centers in this material for quantum optics applications.

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