

## Systematic study of defect-related quenching of NV luminescence in diamond with time-correlated single-photon counting spectroscopy

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We report on the systematic characterization of photoluminescence (PL) lifetimes in  $NV^-$  and  $NV^0$  centers in 2-MeV  $H^+$ -implanted type Ib diamond samples by means of a time-correlated single-photon counting (TCSPC) microscopy technique. A dipole-dipole resonant energy transfer model was applied to interpret the experimental results, allowing a quantitative correlation of the concentration of both native (single substitutional nitrogen atoms) and ion-induced (isolated vacancies) PL-quenching defects with the measured PL lifetimes. The TCSPC measurements were carried out in both frontal (i.e., laser beam probing the main sample surface along the same normal direction of the previously implanted ions) and lateral (i.e., laser beam probing the lateral sample surface orthogonally with respect to the same ion implantation direction) geometries. In particular, the latter geometry allowed a direct probing of the centers lifetime along the strongly nonuniform damage profiles of MeV ions in the crystal. The extrapolation of empirical quasiexponential decay parameters allowed the systematic estimation of the mean quantum efficiency of the centers as a function of intrinsic and ion-induced defect concentration, which is of direct relevance for the current studies on the use of diamond color centers for photonic applications.

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### I. INTRODUCTION

The study of negatively charged single nitrogen-vacancy ( $NV^-$ ) luminescent centers in diamond has attracted a growing interest in recent decades, due to the opportunities they offer in the coherent manipulation of quantum states at room temperature, as well as in the efficient and high-rate emission of single photons on demand. Such unique properties make these centers appealing not only in fundamental quantum optics<sup>1–3</sup> but also in advanced applications, such as quantum computing,<sup>4–6</sup> single spin-based magnetic, electrical, and biological sensing,<sup>7–9</sup> quantum cryptography,<sup>10–12</sup> and quantum nanomechanics.<sup>13–15</sup>

A key advantage of single luminescent centers in diamond such as the  $NV^-$  complex is based on the fact that since they usually consist of deep defects in a wide band-gap material, they can be suitably considered as the solid-state analog of trapped atoms inside a spin-free environment characterized by a broad optical transparency. This is, of course, only true in an ideal crystal, and several works were devoted to assessing the variation of the spectral and spin coherence properties of the  $NV^-$  center, depending on the concentration in the hosting crystal of structural defects,<sup>16,17</sup> isotopic  $^{13}C$  impurities,<sup>18–20</sup> and foreign substitutional atoms such as nitrogen.<sup>21,22</sup>

The important issue of achieving single  $NV^-$  centers in bulk single-crystal diamond with minimum interaction with the surrounding crystal was addressed in a series of works with two approaches: optimizing the ion implantation and

postimplantation processing<sup>23–30</sup> and manipulating the  $NV^-$  centers with suitable noise-correcting procedures.<sup>31–37</sup>

Luminescence lifetime is an effective tool to directly study the nonradiative decay channels in color centers, thus giving significant information on their quantum efficiency, as much as on their interaction with the surrounding crystal environment.<sup>38,39</sup> Luminescence lifetime studies have been applied in the characterization of defect interactions for various luminescent centers in diamond, such as the H3 (Refs. 40 and 41) and N3 (Ref. 42) centers. In both of the above-cited works, a significant decrease in lifetime was observed in samples characterized by high nitrogen concentrations.

In early works, the lifetime of  $NV^-$  centers was evaluated as  $(13 \pm 0.5)$  ns, although a relation between the centers lifetime and the crystal quality in synthetic samples was identified.<sup>43,44</sup>

The  $NV^-$  center is characterized by a triplet ( $S = 1$ ) spin state, with different spin-projection states ( $m_S = 0$  and  $m_S = \pm 1$ ) being characterized by the same oscillator strength. However, intersystem crossing processes involving nonradiative transitions through intermediate metastable singlet states determine different lifetimes (13.7 and 7.3 ns, respectively<sup>45</sup>) for the  $m_S = 0$  and  $m_S = \pm 1$  spin-projection states of the defect, as found in lifetime measurements combined with MW manipulation of the excited states.<sup>45,46</sup>

Photoluminescence (PL) lifetime mapping measurements with a time-correlated single-photon counting (TCSPC) technique proved to be an effective tool to investigate the quenching mechanisms of  $NV^-$  and  $NV^0$  centers.<sup>40,47</sup>