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Genuine and faux single G centers in carbon-implanted silicon

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Among the wide variety of single fluorescent defects investigated in silicon, numerous studies have focused on color centers with a zero-phonon line around $1.28\mu\text{m}$ and identified to a common carbon-complex in silicon, namely the G center. However, inconsistent estimates regarding their quantum efficiency cast doubt on the correct identification of these individual emitters. Through a comparative analysis of their single-photon emission properties, we demonstrate that these single color centers are split in two distinct families of point defects. A first family consists of the genuine single G centers with a well-identified microscopic structure and whose photoluminescence has been investigated on ensemble measurements since the 60's. The remaining defects belong to a new color center, which we will refer to as G^* center, whose atomic configuration has yet to be determined. These results provide a safeguard against future defect misidentifications, which is crucial for further development of quantum technologies relying on G or G^* center quantum properties.

The recent observation of single color centers in silicon has opened a new exploration path for silicon-based quantum technologies [1–15]. In less than three years, advanced single-defect spectroscopy has fueled the detection of more than 10 families of individual defects in silicon, able to emit non-classical, antibunched radiation in the near infrared [1–7, 9]. Surprisingly, some of these defects were not previously referenced in the extensive literature about spectroscopic measurements on defect ensembles in silicon, likely because of their relative scarcity [3]. In view of applications in the fields of single photon sources or spin-photon interfaces, it is highly desirable for a color center to possess a high radiative quantum efficiency η . This value is defined as the probability after excitation to relax through photon emission rather than through a non-radiative channel. In this context, seemingly contradictory reports have been published for the G center in carbon-implanted silicon [1, 3, 8, 10–12], a defect with a zero-phonon line (ZPL) close to $1.28\mu\text{m}$ [16–19]. A remarkably high quantum efficiency $\eta > 0.5$ has been reported in [1], while other investigations at the single defect level [8, 11] or on ensembles of G centers in optical cavities [20] point at a much smaller η in the few percents range at most.

In this paper, we show that low- η and high- η color centers correspond to two different point defects, hereafter called G and G^* centers, respectively. Based on optical

experiments at the single defect level, we establish the specific fingerprints of the G and G^* centers, and provide simple guidelines to identify them unambiguously among the wide variety of carbon-based color centers in silicon.

The experimental setup is a home-made low-temperature confocal microscope built up in a He closed-cycle cryostat (MyCryoFirm). Optical excitation of the sample is performed using a continuous laser at 532 nm and with a microscope objective (Olympus, LCPLN100XIR) mounted inside the cryostat vacuum chamber. The sample photoluminescence (PL) is collected by the same objective and measured by superconducting nanowire single-photon detectors with a detection efficiency of 78% at $1.3\mu\text{m}$ (SingleQuantum). Unless otherwise indicated, all measurements were conducted at 30K.

Single defects are investigated in two different silicon-on-insulator (SOI) samples. The first sample (#1) underwent a carbon-implantation over its entire surface at a fluence of $5 \times 10^{13}\text{ cm}^{-2}$, followed by a rapid thermal annealing during 20 s at $1,000^\circ\text{C}$ [1]. The second sample (#2) was locally implanted with carbon ions, then annealed with the same parameters as Sample #1, and at last locally irradiated with protons [6]. In Sample #2, only the lowest doses, typically $\lesssim 1 \times 10^{11}\text{ cm}^{-2}$ for both carbon atoms and protons, lead to densities compatible with single color center isolation [6]. Both SOI samples have a (001)-oriented top surface. The silicon top layer has a thickness of 220 nm for Sample #1 and 60 nm for Sample #2.

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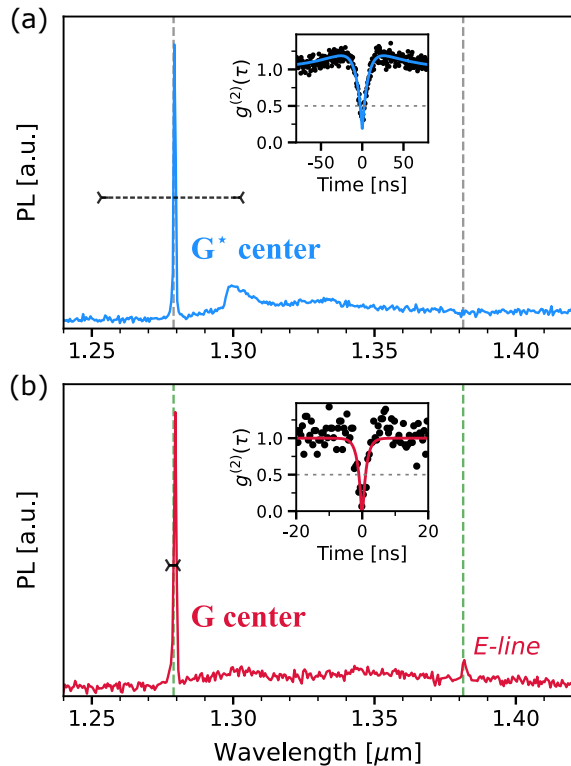


FIG. 1. PL spectra measured for (a) a single G^* center and (b) a single G center in silicon, acquired at 11K and 30K respectively. The vertical dashed lines indicate the wavelengths of the ZPL and E-line of the G center in silicon in PL reported in the literature [16–19]. The dashed horizontal bars indicate the ZPL dispersion range on 41 and 39 defects, respectively. Inset: autocorrelation function recorded on each defect, evidencing the single-photon emission with an antibunching at zero delay $g^{(2)}(0) < 0.5$ [21].

We start by examining the photoluminescence (PL) spectra of single defects evidenced by antibunching $g^{(2)}(0) < 0.5$ (see Fig. 1 Insets). The spectral emission of a single color center from Sample #1 is shown in Figure 1(a). It displays a strong ZPL at 1279 nm, corresponding to the 969.45 meV referenced ZPL energy of the G center in silicon [17, 19]. A broad phonon replica with an energy of $\simeq 14.5$ meV is also found in the PL spectrum [3]. This type of color center manifests a strong wavelength dispersion between defects: the ZPL position fluctuates in the range [1253–1303] nm (horizontal dashed line in Fig. 1(a), for a set of 41 defects, with an average value of 1273 nm and a standard deviation of 12 nm. It should be noted that, although predominant, this family of fluorescent defects coexists with other unidentified single color centers in the Sample #1 [3]. On the contrary in Sample #2, 98% of the fluorescent defects belong to the same family, connected to the typical spectrum displayed in Figure 1(b) (the remaining 2% have a broad spectrum with no detectable ZPL, similar to SD-6 defects in [3]). This emission from an individual defect also

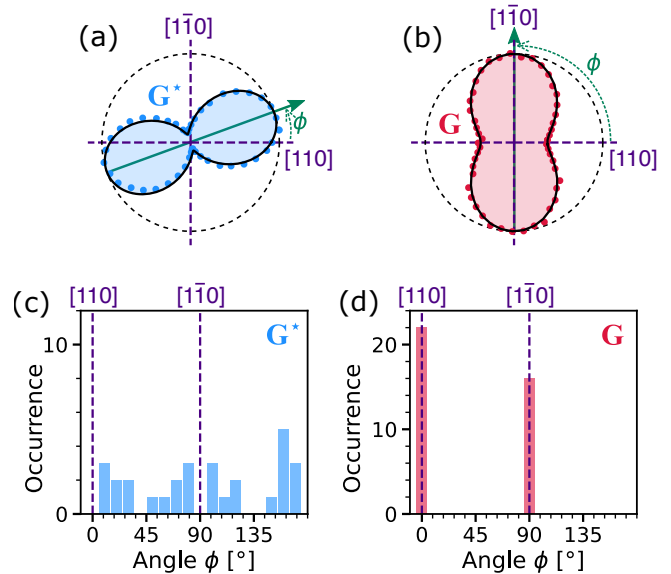


FIG. 2. Emission polarization diagrams recorded on (a) a single G^* center and (b) a single G center. Solid lines represent data fitting using the function: $1 - V + V \cos^2(\phi)$, where extracted visibilities are respectively $V^{(G^*)} = 90 \pm 2\%$ and $V^{(G)} = 62 \pm 2\%$. Histograms of the diagram orientation angle ϕ for a set of (c) 30 single G^* defects and (d) 38 individual G centers.

shows an intense ZPL at the reference wavelength of the G center. However, here the zero-phonon line variations between centers are much smaller. Indeed, for 39 defects, the ZPL is only found between 1277 and 1280 nm, with a mean value of 1279 nm and a standard deviation of 0.5 nm. Furthermore, an extra line is visible in the PL spectrum at 1382 nm, hence $\simeq 72$ meV lower than the ZPL energy. This emission line matches the E-line from the G center in silicon, associated to a local vibration mode (LVM) with a phonon energy of 71.9 meV reported in PL spectra on G -ensembles [16, 17]. It can be seen on Figure 1(a) that this E-line is not present in the PL spectrum of the individual defect from Sample #1. As a consequence, we can conclude that single color centers from Sample #2 are genuine single G centers in silicon (Fig. 1(b)). Because of their markedly different spectral signatures, namely a strong ZPL dispersion and the lack of an E-line, single defects from Sample #1 are not genuine G centers, and will be called G^* centers instead (Fig. 1(a)).

Another way to identify if a single emitter is a G or a G^* center is to analyze the polarization of its single photons. To this end, the emission signal of single centers is measured versus the angle of a polarizer installed before the PL collection fiber. As shown on Figure 2, two notable differences are seen on the emission polarization diagrams recorded on single G and G^* defects. First, the visibility of the G^* diagram is close to unity [1], while the one of the G diagram is typically of $62 \pm 2\%$ (Fig. 2 (a,b)). Consequently the PL of G^* centers comes from a

single emission dipole [1], whereas the one from G centers is a result of the combination of several emission dipoles [22]. Secondly, a statistical analysis performed on individual G defects reveals that the maximum intensity is always oriented along either [110] or $[1\bar{1}0]$ crystal directions (Fig. 2 (d)). These findings are in line with former spectroscopic reports from the 80's predicting an electric dipole along the $\langle 110 \rangle$ axis [16, 23]. On the contrary, a similar study carried out on single G^* centers shows that their emission dipole can point in a large number of directions, except crystal axes [110] and $[1\bar{1}0]$ (Fig. 2 (c)) [1]. The orientation distribution of the emission polarization diagrams of G and G^* defects are thus non-overlapping, allowing to directly associate the emitters to one or the other types of defect.

Measuring the excited-state lifetime is an additional way to discriminate between G and G^* defects. Time-resolved measurements on single G centers show that they relax with a short timescale of roughly $\tau^{(G)} = 4.9 \pm 0.3$ ns (Fig. 3(a)). This lifetime is very close to that reported on various ensembles of G centers in silicon [6, 19]. On the contrary, single G^* centers exhibit a much longer lifetime, typically $\tau^{(G^*)} \geq 30$ ns, as shown in Figure 3(b) (see also Ref. [1]).

Almost one order of magnitude is also observed in the single-photon count rates between the two types of color centers. Figure 3 (b) displays the typical saturation curves recorded under continuous pumping for single G and G^* defects. While the G^* center count rate reaches $\simeq 65$ kcounts. s^{-1} for our experimental setup, the G center saturates around 7 kcounts. s^{-1} . In spite of their longer lifetime, single G^* defects are thus almost 10 times brighter at saturation than the genuine single G centers. We also note that the saturation powers follow the same trend. Indeed, the signal of the G center starts to saturate for an excitation power $\simeq 1\mu W$, while for the G^* defect, this occurs at $\simeq 12\mu W$ (Fig. 3 (b)).

Comparing excited state lifetimes and photon count rates at saturation enables to assess the ratio of quantum efficiencies of individual G and G^* centers. By assuming that under continuous excitation at saturation all populations are prepared in the excited state for both defects, their respective quantum efficiencies, $\eta_{QE}^{(G)}$ and $\eta_{QE}^{(G^*)}$, are related by the formula:

$$\eta_{QE}^{(G)} = \eta_{QE}^{(G^*)} \frac{I_{sat}^{(G)}}{I_{sat}^{(G^*)}} \frac{\tau^{(G)}}{\tau^{(G^*)}} \frac{\eta_{coll}^{(G)}}{\eta_{coll}^{(G^*)}}, \quad (1)$$

where $I_{sat}^{(G)}$, $I_{sat}^{(G^*)}$ are intensities at saturation and $\eta_{coll}^{(G)}$, $\eta_{coll}^{(G^*)}$ collection efficiencies for single G and G^* centers, respectively (Fig. 3(b)). For a maximally collected dipole in the (001) plane, the collection efficiency varies with depth between 0.5% and 2% for the 220-nm silicon layer of Sample #1 [1] [2.5% and 4% for the 60-nm silicon layer of Sample #2]. Considering that the brightest single defects have been selected in our experiments, the maximal value of the collection efficiency can be taken in both cases. Since the quantum efficiency of a single emitter

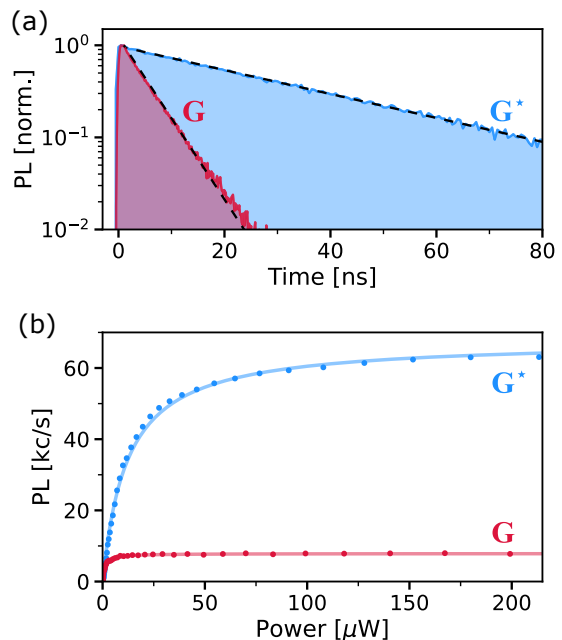


FIG. 3. (a) Excited-state lifetime measured on single G and G^* centers, under 150-ps pulsed laser excitation at 532 nm. A fit by a mono-exponential function (dashed lines) gives the following decay times: $\tau^{(G)} = 4.9 \pm 0.3$ ns and $\tau^{(G^*)} = 33.4 \pm 0.5$ ns. (b) Saturation curves recorded under continuous pumping for individual G and G^* defects. The solid lines show the data fitting with a standard saturation function $I_{sat}/(1 + P_{sat}/P)$, leading to intensities at saturation $I_{sat}^{(G)} = 7.9 \pm 0.1$ kcounts. s^{-1} and $I_{sat}^{(G^*)} = 68 \pm 1$ kcounts. s^{-1} , and to saturation powers $P_{sat}^{(G)} = 1.1 \pm 0.1 \mu W$ and $P_{sat}^{(G^*)} = 12 \pm 1 \mu W$.

cannot exceed 100% by definition, hence $\eta_{QE}^{(G^*)} \leq 100\%$, it follows immediately that the quantum efficiency of G centers is not greater than 1%. This low value is in agreement with previous studies [8, 20] and explains the difficulty to observe a lifetime reduction induced by the Purcell effect for these emitters [11, 20]. The quantum efficiency of single G^* centers is much greater. It has indeed been estimated to be at least 50% [1].

In conclusion, we have highlighted the existence of two families of single fluorescent defects in carbon-implanted silicon that emit with a ZPL around $1.28 \mu m$. A first family consists of genuine single G centers in silicon, associated with the atomic configuration $C_{(s)}-Si_{(i)}-C_{(s)}$ [24–26]. Individual G centers are unambiguously identifiable by (i) an emission spectrum containing the E-line at 1382 nm, (ii) a short excited state lifetime of $\simeq 5$ ns, (iii) a low quantum efficiency $\leq 1\%$ and (iv) a partial linear polarization but with well defined main polarization axis oriented either along [110] or $[1\bar{1}0]$ crystal axes. On the other hand, individual G^* centers are characterized by (i) no E-line in their PL spectrum, (ii) an excited state lifetime typically above 30 ns, (iii) a bright emission connected to a high quantum efficiency $> 50\%$ and

(iv) a single emission dipole whose orientation deviates from [110] or $\bar{1}\bar{1}0$. Following these criteria, the single emitters investigated in [1–3, 12] are likely G^* centers, whereas the genuine G centers are observed in Ref. [6–8, 10, 11]. Note that the fingerprints given here for G and G^* centers can vary depending on their environment. For instance, a thicker top silicon layer can reduce the inhomogeneous linewidth of G^* centers [2] compared to our findings, and cavity integration can favor one emission dipole of the G centers as in [11].

The G center has a head start since its microscopic structure has been identified [24–26] and it can be selectively fabricated at single-defect scale through ion implantation [6, 7]. Moreover, it could host a spin qubit since a spin resonance linked to a metastable spin triplet has been detected optically on ensembles of G centers in silicon [24, 27]. In contrast, the atomic configuration of the G^* center is currently unknown, as well as its spin properties, and no selective fabrication method is known to date for such centers [3]. Nevertheless, its high quantum efficiency combined with single-dipole emission

makes it a very promising color center to fabricate deterministic telecom single-photon sources integrated in silicon. Further investigations, including defect engineering to look for isotope shifts and *ab initio* calculations, are required to elucidate the G^* center origin and its potential connection to the G center in silicon.

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