Femtosecond laser writing of integrated photonic circuits in diamond

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Abstract. Integrated photonic circuits pave the way for next generation technologies for quantum information and sensing applications. Femtosecond laser writing has emerged as a valuable technique for fabricating such devices when combined with diamond's properties and its nitrogen vacancy color center. Such color centers are fundamental for sensing applications, being possible to excite them and read them out optically through the fabrication of optical waveguides in the bulk of diamond. We show how to integrate these building blocks in diamond, to develop proof-of-concept devices with unprecedented electric and magnetic field sensitivities.

1 Introduction

Femtosecond laser fabrication has already established itself as a powerful technique for application in the field of material processing for welding, cutting and drilling [1], and it is attracting a growing interest for its capability to directly write microstructures in the bulk of transparent materials [2]. Focused femtosecond laser pulses can be employed to induce a change in the refractive index of almost all transparent materials such as glasses, single crystals, and polymers, enabling the writing of 3D photonic structures. The locally modified refractive index profile depends on both material and laser processing parameters [3].

Typically in crystals, the irradiation of femtosecond laser pulses produces a decrease of the density at the focal volume due to the creation of lattice disorder, leading to a decrease of the refractive index. In this situation, guiding of light is usually achieved through the Type II waveguide (WG) scheme, consisting in the fabrication of two parallel lines in the bulk of the crystal, resulting in a relative refractive index increase between the two written tracks. Synthetic diamond is emerging as a convenient platform for the fabrication of three-dimensional photonic structures, not only for its excellent transparency, hardness and chemical resistance, but because of the existence of vacancy complexes behaving as color centers. These quantum emitters can be interfaced with photonic circuits written on the surface or in the bulk of the crystal for quantum sensing or information. The most appealing of such color centers is the Nitrogen Vacancy (NV) center, whose negatively charged state (NV$^-$) is characterized by a spin triplet ground state, which can be polarized under green excitation [4]. One can exploit the

NV$^-$ color center also for optical read out, as the fluorescence from one state is brighter than the other due to the existence of a non-radiative decay path. Finally, the NV$^-$'s spin states are sensitive to electric and magnetic fields, enabling room temperature quantum sensing with unprecedented spatial resolution and sensitivity.

The implementation of sensing devices or quantum information systems based on NV$^-$ centers and optical WGs in the bulk of diamond requires the ability to fabricate photonic circuits and color centers in a controlled manner. Both the formation of NV$^-$ centers and optical WGs in diamond can be carried out by the same technique, namely femtosecond laser writing. We will show how this laser fabrication method is a versatile and non-invasive approach capable of retaining the optical quality of the crystal and its color centers. We will demonstrate in this paper a first proof-of-concept room temperature sensing device, based on an ensemble of color centers and accessible through laser written waveguides in the bulk of diamond, which could allow unprecedented sensitivities for the detection of magnetic and electric fields.

2 Integration of NV$^-$ centers and WGs

The NV$^-$ center appears intrinsically in both natural and synthetic diamond and results when two adjacent sites of the diamond carbon lattice are replaced by a nitrogen and a vacancy. Interestingly, the ground state of such a complex is sensitive to electric and magnetic fields through the Stark and Zeeman effects, respectively [5]. This property makes of NV$^-$ centers a convenient room

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temperature detection complex for not only electric and magnetic fields by measuring the shift of the ground state spin transition frequencies under the influence of the fields themselves, but also changes in temperature through optically detected magnetic resonance measurements (ODMR) [6]. However, the possibility to develop real-life sensing devices is bound to the ability to laser-write waveguides and color centers in diamond in a systematic way and finally to integrate them in more complex photonics circuits.

2.1 Fabrication of WGs and single NV⁻s

In 2016, the first Type II WG was successfully written in diamond by focusing a femtosecond Yb laser (300 fs pulse duration, 500 kHz repetition rate, 515 nm central wavelength, 100 nJ pulse energy) into the bulk of the single crystal synthetic diamond at different depths through an objective lens with 1.25 numerical aperture (NA)[7]. Shortly thereafter, Chen et al. [8], demonstrated how a similar femtosecond laser system could induce the creation of on-demand and high-quality single NV⁻ centers in the bulk of quantum grade diamond. In this case, a single low intensity femtosecond laser pulse was focused beneath the surface of diamond to form an ensemble of vacancy defects within the focal volume. After a post high temperature annealing treatment (1000 °C), the vacancies were mobilized, enabling the recombination with nitrogen impurities within diamond to form NV⁻ centers. The next step in the development of quantum photonic circuits is the integration of the color centers with the optical waveguides. Our group has recently demonstrated such a result [9], showing the possibility to firstly write type II WGs and then induce the creation of NV⁻ color centers in between the laser written tracks with a spatial accuracy of ~500 nm, fabricating a proof-of-concept device in which precisely located NV⁻ centers can be externally accessed, excited and read out though the laser written WGs.

2.2 High density NV⁻ ensembles for sensing applications

Single-NV⁻ centers are characterized by a nanometric spatial resolution due to the atomically small size of single isolated complexes, but by a relatively low sensitivity which can be improved by averaging over many measurements. If higher sensitivity (or speed) is required and the nanometric spatial resolution is not necessary, single NV⁻ centers can be replaced by high density NV⁻ centers (ensembles) at the expense of reduced overall spatial resolution. For this purpose, it is critical to be able to deterministically create the ensembles with a required density and to interact with them through the optical WGs. It is known that the sensitivity (η) of an ensemble of NV⁻ centers can be described as:

$$\eta = \frac{1}{k} \ \frac{1}{C\sqrt{M\Gamma}} \ \frac{1}{T_2^*} \qquad (1)$$
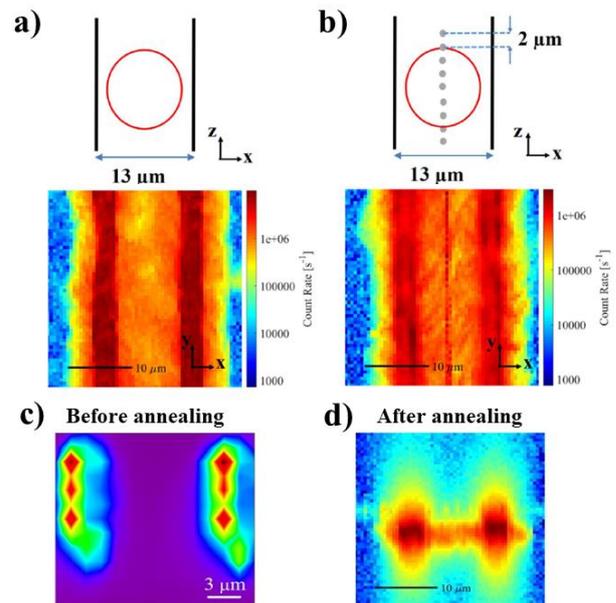

Fig. 1. Schematic cross-section of femtosecond laser written 'empty' (a, above) and 'static exposure' (b, above) waveguides, with modification lines in black and the expected mode field diameter of the waveguide mode outlined in red, with integrated static exposures marked in gray. Photoluminescence confocal maps of 'empty' (a, below) and 'static exposure' (b, below) waveguide and modification lines written in HPHT sample after annealing. PL Photoluminescence confocal cross-section of 'empty' waveguide and modification lines written in HPHT sample before (c) and after (d) annealing.

where k represents the frequency shift of the NV⁻ center's spin transitions under magnetic (k_m = 28 GHz T⁻¹) or electric fields (k_e = 17 Hz cm V⁻¹)[10], C is the ODMR contrast, M is the number of NV⁻ centers in the ensemble measurement, Γ is the detected count rate for each NV⁻ center, and T_2^* is the inhomogeneous NV⁻ coherence time [11]. Following equation (1), the sensitivity of such devices based on ensembles of color centers scales as $\sqrt{M}$. This can be achieved by either increasing volume of NV⁻ centers or the density of NV⁻ centers sampled, while paying attention not to compromise their coherence time. Our group managed to write high density NV⁻ center ensembles in HPHT diamond through single static laser exposures followed by annealing, and their integration within a waveguide has also been studied. Arrays of 'empty' type II waveguides with 13 μm separation between the sidewalls and 10 μm estimated waveguide mode field diameter at 532 nm (Fig. 1(a), above), together with 'static exposure' ones, containing arrays of nine single 100 nJ static exposures separated by 2 μm axially (Fig. 1(b), above), were written in HPHT diamond at 18 μm depth. Fig.1(a-b, below) shows PL confocal maps in overhead view of the 'empty' and 'static exposure' waveguides, where it can be noticed how despite some brighter spots are visible in the 'static exposure' WG, a diffused bright fluorescence is observed in both cases. This is due to the fact that during the WGs fabrication, vacancies are created in the laser written sidewalls, which can migrate during the annealing treatment and combine
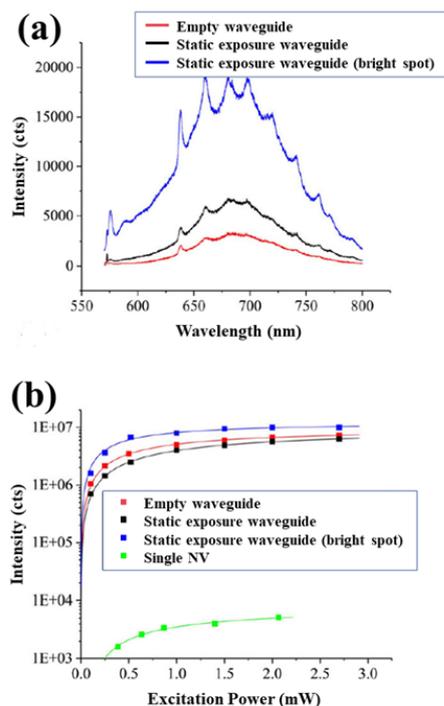
Fig. 2. PL spectra (a) and Power dependent PL (b) from 'empty' waveguide, 'static exposure' waveguide and from a bright spot (static exposure).

with the nitrogen in the diamond forming the NV⁻ centers, as shown from the transverse confocal microscope images of Fig.1(c-d). To validate the formation of NV⁻ centers after annealing, photoluminescence (PL) spectra have been taken from the 'empty', 'static exposure' WGs and from a bright spot corresponding to static exposure in the waveguide (Fig.2(a)). In all three cases, the characteristic zero phonon line (ZPL) of NV⁻ centers at 637 nm is observed, confirming that the created vacancies migrated in between the walls of the waveguide and created the color centers. The spectra taken from the static exposures are brighter than the surrounding area, presenting also additional structures with lines at 660, 680, 697.6, 720.7, 741.1 and 760.7 nm. A much stronger contribution from a line at 575 nm is also shown corresponding to the neutral NV^0 ZPL.

To optimize the sensitivity of the device one has to be able to evaluate the density of the created NV⁻ ensemble. This can be done by performing power dependent PL saturation measurements of both the "empty" and "static exposure" waveguides and comparing them to the total signal collected by a single NV⁻ center in another sample measured using the same conditions (Fig.2(b)). By approximating the confocal microscope's spread function through an estimation of the collection volume of the confocal microscope itself using confocal images of a single NV⁻, it was possible to determine the ensemble densities. We found densities of 1.1×10^{15} cm⁻³ (or 6 ppb) in the 'empty' waveguide, and densities up to 1.4×10^{15} cm⁻³ (or 8 ppb) in the 'static exposure' ones. Although the coherence times of the generated color centers must be quantified, a first approximation of the achievable electric and magnetic sensitivities can be performed. We can
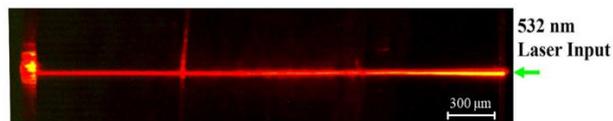

Fig. 3. Overhead microscope view of 'static exposure' waveguide showing the streak of fluorescence emission along the length of the waveguide.

consider a simple sensing device in which excitation and detection are performed through the coupling to a laser-written waveguide as shown in Fig. 3, where the sample was excited with a green laser end-fire coupled into the waveguide from the right-hand side, while the light scattering from the waveguide was filtered using a 532 nm notch filter from the top of the sample and recorded on a CCD. Assuming a 3 mm long waveguide having a 10 μm mode field diameter, and NV⁻ density of 1.1×10^{15} cm⁻³ we can expect to have $M = 2.6 \times 10^8$ NV⁻ centers contributing to the sensing signal, with a photon collection efficiency of $\Gamma = 900$ Hz per NV⁻ and a contrast $C = 0.05$. From this, we estimate a magnetic field sensitivity of 1.5 nT Hz$^{-1/2}$ and an electric field sensitivity of 2.4 V cm⁻¹ Hz$^{-1/2}$. These results can already be compared with the state-of-the-art systems, which generally are characterized by lower sensitivities, ≈ 290 pT Hz$^{-1/2}$ for magnetic fields [12] and ≈1.6 V cm⁻¹ Hz$^{-1/2}$ for electric fields [10]. Further optimization of laser writing parameters and nitrogen impurity levels within the diamond are anticipated to lead to record high sensitivities.

3 Conclusions

We have shown how femtosecond laser writing in diamond can be applied for the fabrication of next generation technologies, in particular for quantum optics and information. We have demonstrated how femtosecond lasers enable the formation of optical waveguides and color centers in an easy and repeatable fashion and how it is possible to integrate the two components for prototype devices exceeding the performance of the existing systems based on conventional technologies. In particular, we studied a room temperature sensing device based on ensembles of NV⁻ centers, which is characterized by unprecedented high sensitivities. It may be possible to improve the sensitivity even further by integrating femtosecond laser written Bragg reflectors into the waveguides to increase the effective light-NV⁻ center interaction length through multiple passes of the excitation laser [13]. Moreover, since the strain created between the lines confining the waveguide structure is equivalent to an electric field, the NV⁻ centers located in the middle of the waveguide and experiencing such a strain are affected by a constant bias electric field. Because of this bias field these NV⁻ centers are less sensitive to stray magnetic fields, while more sensitive to extremely weak electric fields, making them particularly attractive for the detection of such fields. Finally, further studies with the versatile 3D femtosecond laser writing method will improve the functionality and

quality of the fabricated quantum devices, allowing for more complex systems capable of advanced functionalities [14].

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