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SCUOLA DI INGEGNERIA INDUSTRIALE E DELL'INFORMAZIONE

Executive Summary of the Thesis

Femtosecond Laser Writing in Material Platforms for Quantum Technologies

Master of Science in Engineering Physics - Photonics and Nano Optics

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1. Introduction

Quantum technology is one of the most promising emerging fields of engineering and physics, it refers to technologies that rely on quantum mechanical properties, particularly quantum entanglement, quantum superposition, and quantum tunneling. Emerging quantum technologies include quantum computing, sensors, cryptography, simulation, measurement, and imaging. Integrated quantum photonics is seen as an important step in developing useful quantum technology. A relevant role is being played by the direct inscription of photonic circuits by femtosecond lasers [1]. Concurrently, color centers in wide bandgap semiconductors have shown promise in becoming one of the best single-photon emitters for non-classical light sources and excellent qubits with desirable properties such as long coherence times, spin-photon interfaces and relatively high temperature of operation. This thesis investigated the potential of femtosecond laser micromachining for the fabrication of photonic devices relevant to quantum sensing, such as Y-Splitters in OG diamond and waveguides in NV doped diamond, as well as the deterministic placement of single color centers for quantum computing and communication in interesting materials such as diamond, silicon carbide, and hexagonal boron nitride. The goal of the thesis was to show that femtosecond laser fabrication can be a fast, reliable, and scalable technique for creating integrated quantum photonic devices comprised of active color centers interconnected by optical waveguides.

2. Background

The second chapter deals with the theoretical background regarding femtosecond laser micromachining and spin defects in semiconductors.

2.1. Femtosecond laser micromachining

The fundamental operating principle of the presented writing method is the fact that linear absorption cannot occur in transparent dielectrics and only higher-order absorption has the potential to happen through phenomena such as multiphoton absorption or tunneling ionization. Since nonlinear absorption processes are phenomena that depend on an intensity threshold, using of focusing objectives allows micrometer spot size at the focal volume and thus the possibility to inscribe 3D structures by moving the position of the focal volume. After the femtosecond laser pulse has been nonlinearly absorbed by the transparent material, the relaxation transfers the energy to the lattice at the focal volume, altering the material and leading to three distinctive material modifications depending on the laser energy, namely; smooth refractive index change, birefringent gratings and void creation. The first type of modification will be used to create waveguides and the third to create vacancy-related defects.

2.2. Solid-State Defects

Solid-state defects are either highly detrimental to crystalline growth, for example, or beneficial due to their ability to modulate and control material properties. When isolated they act as analogues of atomic systems in an effective “semiconductor vacuum”. In recent times, spin defects within solid-state materials have been rapidly applied to all three major fields of quantum science: sensing, computing, and communication. In addition to the properties of their host material, defects are defined by their spin, optical, and charge states. For defects in the solid state, quantum information is generally encoded in the electron spin of the orbital ground state of the defect. Controllable qubits with long relaxation and coherence times can be obtained by electron spins. They can be coupled to nuclear spins for long-lived quantum memories and advanced applications. The optical addressability of numerous spin defects, which provides a photonic interface for quantum applications and thus the possibility of isolating single defects, stimulates materials research for defects with the best optical properties. The emission wavelength is one of the fundamental properties, e.g. in order to minimize optical fiber losses for quantum communication, infrared photons are preferred. The charge properties of defects are fundamental to their reliable use, in fact a spin defect that corresponds to a specific charge state results in radically different spin and optical interfaces when it is in a different charge state. Lastly, spin defect qubit properties are interwoven with the intrinsic host material properties, including variations in crystallographic, dopant and nuclear-spin imperfections in their local environment. In summary, the selection of host materials should consider not only the properties of the spin defect but also the scalability, ease of fabrication and unique properties of the host.

3. Experimental Details

In this chapter the experimental details of laser fabrication and characterization of waveguides are discussed in detail.

3.1. Micromachining System

The Felis laboratory at the CNR-IFN facility located at the Politecnico di Milano, Lecco campus served as the primary laser micromachining station for this thesis' laser writing. It uses a Yb:KGW BlueCut all-fiber integrated fiber laser system that is designed for industrial use. The output is a 300 fs laser beam with a peak pulse energy of 10 μJ and a tunable repetition rate from 10 MHz to single pulse. The sample is positioned on an Aerotech ANT130-110-XY stage that enables translations along the x and y axes, or along the plane of the optical table. The laser light comes towards the microscope objective that is mounted on an Aerotech ANT130-L-ZS air bearing stage, which allows for vertical z-axis displacement of the objective. The Aerotech stages are programmed in the Aerobasic compiler provided by the manufacturer using the Aero-code language. The polarization and power of the laser can be adjusted by rotating an assembly composed of a half wave-plate and a polarizer.

3.2. Waveguide Characterization

Certain general and specific characterizations need to be carried out to determine the performance of the laser-written photonic networks for the targeted application. The mode field diameter (MFD), i.e. diameter at which the power density is reduced to $1/e^2$ of its maximum value, is one of the primary characteristic parameters of any waveguided Gaussian beam. The simplest method for measuring this parameter is to fiber couple the input facet of the waveguide using a regular fiber connected to a laser emitting at the desired wavelength. The waveguide and fiber are positioned using high-resolution three-axis manual positioners. A vision system for better alignment can be used with an overhead microscope with CCD. The optical loss in transmitted power is another key parameter of any optical waveguide. The term insertion loss (IL) refers to the total loss caused by the insertion of the optical device. The waveguide insertion loss can be determined using the fiber-coupled characterization technique by measuring the power transmitted through the laser-written waveguide by butt-coupling a single mode fiber at both the input and the output facets of the waveguide and normalizing to the reference power measured by direct butt-coupling of the input fiber to the output fiber.

4. Diamond Quantum Photonics

In this section, photonic components are fabricated in diamond, a very powerful platform for quantum computing, sensing and communication.

4.1. Optical Waveguides in Diamond

Previous research focused on determining the ideal settings for laser writing single continuous lines and type II waveguides in diamond was carried out by Dr. Bharadwaj and reported in this subsection. The main parameters with a wide range of options were the laser wavelength, laser repetition rate, sample scan speed, NA of the focusing objective, laser power, and input beam polarization. Uniform and repeatable modifications in the majority of samples were determined to be created most effectively using the following parameters: 500 kHz repetition rate, 50 mW average power, 1.25 NA objective (RMS100X-O,100, Olympus), and 0.5 mm/s scan speed. Motivated by the good results on single line modifications, two closely spaced single line modifications were engraved into a waveguide using the type II modality of waveguide writing. The buried waveguide in diamond was inscribed for the first time in literature [2]. The insertion loss was determined to be 11 dB for the best waveguide with a waveguide length of 5 mm. The waveguides displayed polarization-dependent behavior by only supporting the TM mode.

4.2. Y-Splitters in Diamond

Motivated by evidence for a single-photon nonlinearity in a laser-written SiV–waveguide system [3], it was decided to engineer vertical Y-Splitters in order to increase the collection efficiency from defects centers, create path-entangled light fields and open the possibility for on-chip Hong-Ou-Mandel interference. Most of the writing parameters were fixed: 515 nm central wavelength, 500 kHz repetition rate, 1 ps pulse duration, 1.25 NA (100× objective), 60 μm depth of the input waveguide. The parameters that were varied were: the radius of curvature of the bent regions R, the separation between output waveguides, the writing pulse average power P. A total of 88 y-splitters were fabricated on a sample of optical grade diamond. Imaging the guided modes with the beam profiler, we observed that all the waveguides showed to have good mode shapes. We also noted that changing slightly the input coupling changed the coupling efficiency of the two output waveguides, pointing to the existence of different guiding paths within the waveguide sidewalls. Moreover, the shallow paths showed a higher attenuation with respect to the deep ones. In conclusion, the best splitter was found to be characterized by radius of curvature 175 mm, separation between output ports 20 μm and writing power of 50 mW. The insertion losses for upper and lower output ports respectively of 28,8 dB and 28,5 dB, so we were able to achieve a good splitting ratio but at the cost of very high losses.

4.3. High Density NVs in DNV Diamond

Another interesting project was to write optical waveguides in a new commercial CVD diamond from Element 6 specifically designed for quantum sensing, having a uniform and high concentration of NV centers (4.5 ppm NV centers with T_2^* of 0.5 ms). Optical waveguides were laser written with our standard fabrication parameters of 515-nm wavelength, 300-fs pulse duration, 500-kHz pulse duration, 1.25-NA oil immersion objective, 0.5-mm/s scan speed, and 13-mm separation between waveguide sidewalls. The mode field diameter was 8.1 mm × 9.4 mm at 635-nm wavelength. The insertion loss was 11 dB for the 3-mm long waveguides. Confocal photoluminescence maps shown similar count rates were observed within the pristine and waveguide regions, while a dramatic reduction in signal within the laser-induced damage tracks which define the waveguide sidewalls was observed. Further characterization was done from Cardiff University. Both waveguide and pristine areas have the comparable ODMR contrast (1%) and dephasing coherence time (500 ns). By taking the PL rate of 3.2×1012 Hz, contrast of 4 %, T_2^* of 500 ns, the photon shot noise sensitivity is estimated at 257 $pT \cdot Hz^{-1/2}$.

5. Deterministic Placing of Color Centers

Ion beam implantation, in which high energy ions are bombarded onto the surface of the material, is the traditional technique for deterministically placing color centers. However, the method is incapable of creating defects in the bulk deeper than 5 μm with high accuracy and causes residual damage to the crystal lattice, resulting in undesirable strain and local unwanted defects, which reduces the spectral properties of the defects. The possibility of laser-induced vacancy creation by focusing femtosecond laser pulses with energies lower than the amorphization threshold is explored.

5.1. Laser Writing of NV Centers in Diamond

Previous work from Dr. Bharadwaj showed the ability to deterministically fabricate NV centers in bulk diamond using femtosecond laser pulses. Static exposures with pulse energies from 10 to 30 nJ and number of pulses N of 5 and 1 were performed on an ultrapure electronic grade diamond sample with the aim to create single NVs for quantum computing applications. The sample, following the static exposures was annealed at 1000° C for 3 h. It was seen that for a pulse energy of 24 nJ, 8 out of the 10 trials produced NV centers giving a NV center creation probability of 80% but when averaged over 5 different samples, the probability was about 50%. Photoluminescence maps measured from the static exposure showed an emission with a spatial confinement of about 1 μm^2 within the static exposure. This constituted a tremendous achievement compared to the resolution that can be achieved by ion implantation in the bulk. The antibunching $g^{(2)}(0)$ dip for the laser written NV centers was obtained to be well below 0.5, implying single photon emission from the NV created in the bulk of diamond using single femtosecond laser pulse irradiation followed by annealing [4].

5.2. Laser Writing of Vacancy Centers in SiC

SiC is a very interesting material for quantum technologies as it can host several optically active point defects that emit light in the spectral region from visible to near-infrared and can overcome many of the drawbacks of diamond such as compatibility with microelectronics technology, nanostructuring and n- and p-type doping. Motivated by the possibility to create laser-induced vacancies with focused femtosecond laser pulses, for this project we started a collaboration with the group of Prof. Castelletto from RMIT University towards this common goal. From previous research [5], they observed from PL confocal maps that laser pulse energies below 13 nJ did not provide any PL suggesting that the damage was not enough to produce vacancies or localization was not achieved. Using room temperature and 80 K spectroscopy, they determined that the broad emission of the fabricated areas at 920 nm is attributed to the V_{Si}. Encouraged by these results, we set up an initial experiment on a sample of High Purity Semi-Insulating 4H-SiC. We used a central wavelength of the pulses of 515 nm and a focusing objective with NA of 1.25 and 100x magnification. For statistical analysis, 6 trials of the same parameters were performed. The parameters being varied were: the average pulse energy, from 10 to 230 nJ ; the number of pulses per exposure, from 1 to 500; the focusing depth, from 7 to 25 μm. Unfortunately our femtosecond source turned out to have one of the diodes broken and the internal pulse compressor misaligned, showing an autocorrelation $>$ 1 ps. Nevertheless, the technique's potential to create active color centers in SiC is undeniably very promising.

5.3. Laser Writing of color centers in hBN

Hexagonal boron nitride (hBN) is a laminar van der Waals material, and thus very interesting for the purpose of researching semiconductor physics in two dimensions, a large number of defect states with internal optical transitions that produce color centers may be present in its wide bandgap. With the goal of creating single photon emitters in hBN using the technique of femtosecond laser micromachining, we started a collaboration with various research groups. The samples consisted in 1-10 layers of hBN flakes deposited on fused silica ($10 \times 10 \times 1$ mm^3) obtained by exfoliation and were provided by CIC nanoGUNE research center. Fabrications were carried out at Ulm University. The laser writing parameters were: 515 nm wavelength, 250 kHz repetition rate, 100× objective (without oil), NA of 0.9. Considering relevant samples, writing energies were varied between 4 and 80 nJ. The number of pulses per exposure ranged between one and 50 pulses. After the fabrication, images and spectra were acquired through confocal microscopy to inspect for presence of color centers. One of the sample showed plenty of possible emitters all over the examined spectral region (580-800 nm), even though the correlation between irradiated spots and emitters location was not clearly defined and many of them were unstable and blinking. In conclusion, strong evidence of fabrication of color centers in hBN through femtosecond laser pulses were produced, next steps could be to study of the type of emitters and their concentration as a function of laser writing parameters; measuring the second order correlation $g^{(2)}$ in order to check for the actual presence of single photon emitters.

6. Figures

A few important figures related to the present work are reported.

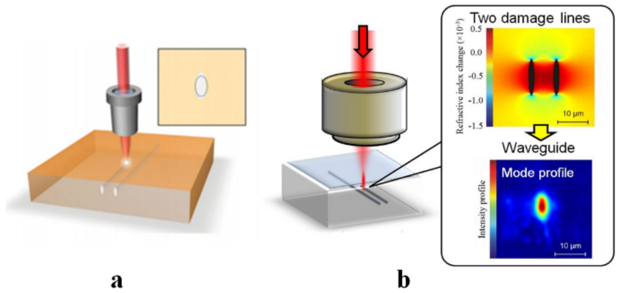

Figure 1: Schematic showing the two modalities of femtosecond laser writing of waveguides. (a) The type I modality, where a single line of increased refractive index modification acting as the core for the optical waveguiding is created. This is generally employed for laser writing in glasses. (b) The type II modality, where two lines of decreased refractive index are laser written close to each other creating a stressed central in the region between them and hence confining the optical mode between the two lines. This is generally employed for laser writing of optical waveguides in crystals.

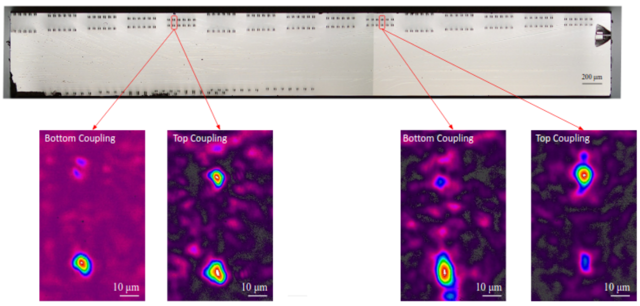

Figure 2: Cross sectional optical microscopy images of output ports of Y-splitters in OG diamond. In set images show the behavior of splitters 33 and 66 for different launching conditions. It can be seen that for splitter 66 there exists a condition for achieving splitting of the field onto the output ports.

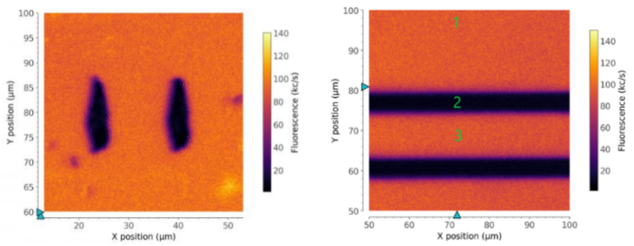

Figure 3: Confocal fluorescence microscopy scan with overhead (right) and cross sectional (left) views showing type II waveguide, with similar photon count rates in the pristine and waveguide regions. The waveguide depth (center of modification to surface) is approximately 30 mm. The air interface is just off screen at a Y position of 50 mm.

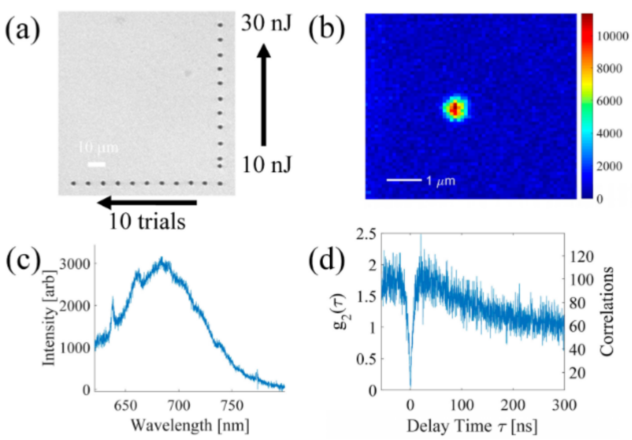

Figure 4: (a) Overhead microscope image of laser induced modification for pulse energies of 10, 12, 14, 16, 18, 20, 22, 24, 26, 28 and 30 nJ with pulse number of 1 at a depth of 25 μm. The visible dots are marker dots written with a pulse energy of 100 nJ and (N) = 25 pulses. (b) Overhead photoluminescence map of static exposure with a pulse energy of 24 nJ. (c) Photoluminescence measurement of the 24 nJ static exposure NV. (d) Intensity autocorrelation (corrected for background on left y-axis, raw uncorrected correlations counts on the right y-axis) revealing single photon emission.

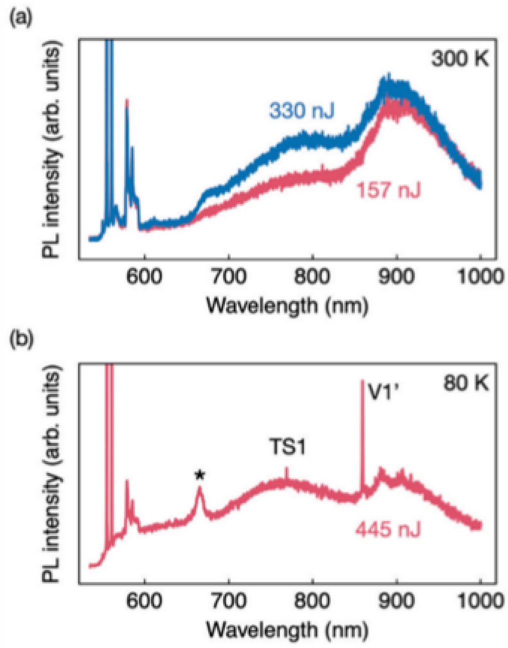

Figure 5: Spectroscopy of the (a) HPSI 4H-SiC laser written area corresponding to the highest energy dots of 445 and 330 nJ showing a broad emission centered at 920 nm and an emission at 770 nm. (b) Here, 80 K spectroscopy of a dot at 445 nJ. V1' is at 859 nm corresponding to the hexagonal ZPL of the VSi in the 4H-SiC. The emission at 769 nm shows a ZPL at low temperature, TS1, previously observed in proton irradiated 4H-SiC annealed at high temperature, however, the origin is unknown. A peak indicated with (*) is also an unknown emission and it is due to laser irradiation.

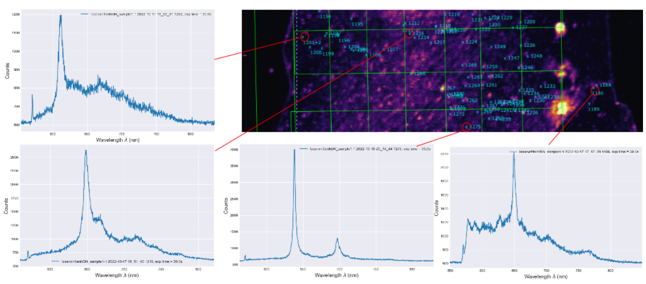

Figure 6: Confocal microscopy image of one of the most interesting flakes after irradiation and emission spectra of possible emitters.

7. Conclusions

Femtosecond laser writing has been confirmed to be a viable fabrication technique for laser writing photonic circuits in bulk transparent crystals. To improve the NV centers concentration for quantum sensing applications, waveguides in NV doped diamond were fabricated and characterized, showing promising results in terms of sensitivity. The ability to create cavities for multiple excitations and improve the coupling efficiency from active color centers was one of the goals of the project, so the fabrication of Y-Splitters focused on the optimization of the splitting ratio as a priority. The next steps would be to find the parameters that improve the losses of the device. Using the same femtosecond laser workstation for the writing of optical waveguides, NV centers in diamond have been previously created and attempts to create vacancy centers in SiC and to discover useful spin defects in hBN have been carried out . In conclusion, the proposed technique has been demonstrated to be highly effective as a potentially scalable method of fabricating integrated quantum photonic devices, although there is still much room for improvement, this work constitutes an effective proof-of-concept.

8. References

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