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On-Demand Single Photon Source from NV Centers in Diamond at Room Temperature

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14. ABSTRACT

This document provides an overview of the work conducted under the NRL 6.1 Base program titled "On demand single photon source from NV centers in diamond at room temperature." The goals of this program were: (1) investigate room-temperature, on-demand single photon sources for quantum information processing using nitrogen vacancy (NV) centers in diamond films and single-crystal nanodiamond particles; (2) evaluate the use of laser processing techniques to synthesize diamond nanoparticles; and (3) implement instrumentation for measuring and characterizing single-photon emission from the NV-centers on diamond films.

15. SUBJECT TERMS

Nonlinear optics; Stimulated emission; Nitrogen Vacancies (NV) centers; NV-centers in nanodiamonds and diamond films; Single photon emission

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1. Background

The current state of the art for single photon sources falls short of what is necessary to establish either reliable quantum channels for communication or sensing applications. There are three conditions a single photon source must meet to be effective. First, each photon must be indistinguishable from the others. This means that each photon must have identical polarization, spatial mode, and energy. Second, the source must be able to produce photons 'on-demand' with zero probability of multi-photon events. Finally, the generation and detection of these photons must be done with high efficiency. A field-deployable light source that meets these conditions is critical for technologies that rely on the transfer of information between quantum systems. These include sending qubits through long-range fiber optic networks as well as optimizing the bandwidth and range of secure communications. A potential solution to these challenges is provided by defect states in wide-bandgap semiconductor materials. They can be fabricated with the pristine consistency that scalable devices demand, and they maintain emission coherence at room temperature. A handful of defect states in wide-bandgap semiconductors such as diamond and silicon carbide have been identified as promising single photon sources. Unlike quantum dot-based sources, which currently require cryogenic cooling, diamond defect centers boast a high level of photostability at room temperature as well as electrical properties that stand up to extreme electromagnetic, chemical, and thermal conditions. Furthermore, diamond's large bandgap (5.5 eV) shields the NV-centers from free electron interference and allows for efficient coupling of the system to an external cavity. However, to develop this technology, an optical means of controlling single photon emission needs to be implemented to ensure the generation of completely indistinguishable photons with no chance of a multi-photon event. The objective of this program is to demonstrate triggered single photon emission from a single NV defect site in diamond which can be tuned across a large portion of the optical spectrum. We also investigated the use of laser-based direct-write techniques for the generation of NV-centers in diamond with specific spatial distributions. Additionally, we present probability transition calculations in order to describe the polarization orientation between a stimulating photon and the resulting emission from the defect site.

2. Energy dynamics of the NV-center in diamond

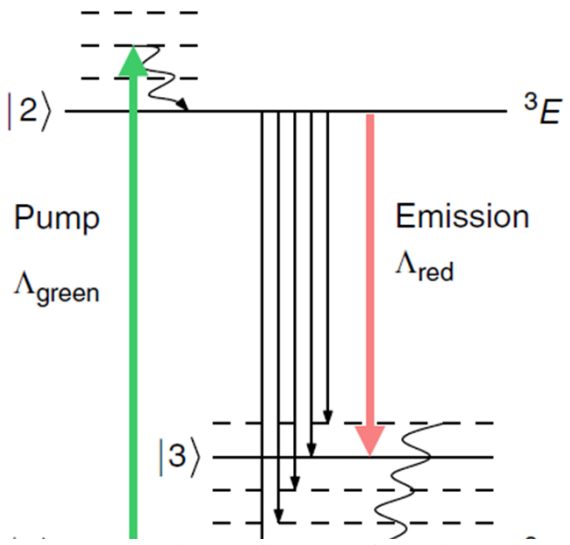

Fig. 1-- Energy diagram for a NV defect in diamond. Direct optical pumping into the 3E state relaxes into the metastable 1E state before radiatively decaying back to the ground state [1]

Nitrogen vacancy centers are an established source of high-fidelity single photon sources that can operate in the optical spectrum at room temperature. The photon statistics from a properly prepared NV center are consistent with a shelving state [1]. This relationship is illustrated in Figure 1. Pump probe measurements show that these metastable energy states last for an average of 10ns before radiatively decaying back into the ground state. Two possible photo-luminescent charge states have been observed: a negative charge state with an emission peak at 638 nm (known as NV^-) and a neutral charge state with an emission peak at 575 nm (NV^0) [2]. These relatively long-lived excited states provide enough time to address the defect optically using an ultrafast laser system that operates on the order of femtoseconds. These lifetimes are illustrated by the fluorescence lifetime measurements taken on defects inside of

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nanodiamond powders with mean particle sizes of 60 nm nanometers, as shown in Figure 2. The primary advantage of diamond defects over other leading quantum photonic sources such as quantum dots is that they maintain photostability at room temperature. This means that diamond sources have greatly reduced bleaching or blinking behaviors at high excitation powers and minimized dephasing and spectral diffusion.

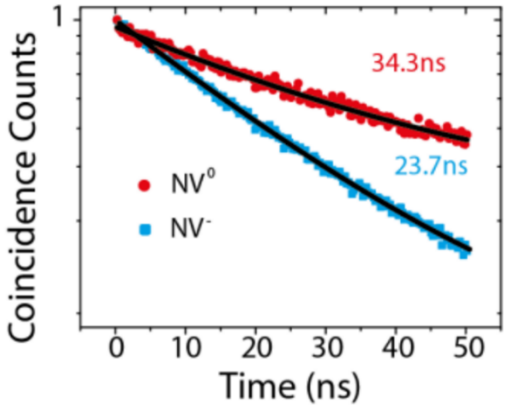

Fig. 2 -- Fluorescence lifetime measurements for NV^- and NV^0 centers obtained from nanodiamond powders excited with a 532 nm pulsed laser [2]

These advantages are offset by the challenges that the high refractive index and fabrication challenges that diamond presents. While the defects inside a diamond substrate can be placed with pristine accuracy and consistency, the diamond itself is difficult to grow on almost all substrate materials. Additionally, diamond's high refractive index generates total internal reflection events and spherical aberrations that reduce the total collection efficiency of single photon emission. Since most quantum-based optical applications demand extremely low error rates, this challenge is among the most important to address for generating devices that utilize an ultra-wide bandgap semiconductor as a quantum source.

3. Stimulated emission in semiconductor defects

Normally, stimulated emission is a fundamental process of laser operation that is used to amplify the number of photons oscillating in a cavity. It creates completely phase, and sometimes frequency, coherent photon streams in a single direction of emission. This project seeks to utilize only the coherence properties of the stimulated emission process in order to trigger the release of single photons that can later be compared to the stimulating electric field. The laser pulse in this instance can be seen as an idler pulse for all practical purposes. To better understand this relationship, we can use Fermi's golden rule, equation 1, in order to solve the linear case for stimulated emission:

$$\Gamma_{i\to v} = \frac{2\pi}{\hbar}|\langle v|H'|i\rangle|^2\rho(E_v) \tag{1}$$

$$\vec{E} = \hat{k}E_e\cos\omega(t - {}^{x}/_{c}) - \alpha \tag{2}$$

where H' is the perturbing Hamiltonian of the molecular system under the influence of a stimulating electric field E_v. And the indices i and v represent the initial and virtual states. By applying an external electric field, equation 2, we can see that the transition probability scales inversely with the energy difference between the stimulating photon and energy between the excited and ground states giving us the solution in equation 3:

$$\omega \approx \omega_0 = E_i - E_v \tag{3}$$

Hence, for the linear case, the incoming and resulting photon must not only be the same frequency but must also be completely indistinguishable from one another. This result is convenient for designing a laser cavity, but it means that building a single photon source in the linear regime using this mechanism is not feasible. Instead, there must be an intermediate state to stimulate the system into that breaks this degeneracy. In order to separate the exciting and stimulating laser from the single photon emission, we propose to use higher order absorption events coupled with optically gated signals to control the defect energy state. The goal is to achieve on-demand control over the polarization and energy state of the emitted photon as well as

increasing the efficiency of detection with the use of an external optical cavity and well-defined optical gate.

Similar experiments have recently been published using semiconductor quantum dots that support a single bi-exciton [3]. By stimulating the bi-exciton into an intermediate or 'virtual' state, the remaining energy collapsed immediately and was efficiently collected into a single photon detector system. Virtual states are a name given to the necessary intermediate states in most nonlinear optical interactions. They are not eigenstates of the system and, therefore, they exist for an immeasurably small amount of time before spontaneously emitting. The resulting photon from the collapse of this virtual state is still well correlated with the stimulating event. Conservation of momentum and energy dictates that it has a deterministic polarization and phase with respect to the incoming optical pulse. Stimulated single photon emission from quantum dots has been observed with a precise control over the polarization and wavelength of the single photon state.

4. Laser Synthesis of Nanodiamonds

Nanodiamonds can be synthesized by a wide variety of techniques such as detonation of explosives, ball-milling at high pressures and temperatures, chemical vapor deposition (CVD), ion or electron beam irradiation of carbon materials, chlorination of SiC, cold compression of carbon phases, ultrasound cavitation of hydrocarbons, laser ablation of a carbon-containing target in a liquid, and laser-induced plasma dissociation of liquid ethanol.

A laser-induced plasma inside a liquid environment can produce non-equilibrium conditions at the plasma-liquid interface where highly reactive physical and chemical processes can occur. In addition, synthesis of nanoparticles within a liquid environment minimizes any contamination or agglomeration that may occur with other chemical or physical methods. Filamentation of laser beams in transparent media occurs when self-focusing is balanced by defocusing of the beam caused by the generated plasma column. This balance results in a maximum peak intensity within the column and is called "intensity clamping." The column is actually a series of finely-separated self-foci and regions of lowered electron density that is perceived by the eye as a single filament. Interest in filamentation research increased with the availability of intense femtosecond (fs) lasers that avoided the optical breakdown problems observed with longer pulse lasers (i.e. greater than a picosecond). Filamentation occurs in all transparent media (gas, liquid, solid) but the electron density generated in solids or liquids is more than two orders of magnitude higher than that in gases. Code 6364 has successfully used pulses from an infrared (IR) fs laser (λ=1026 nm, 160 fs, 1 kHz) in their ultrafast-laser-materials-processing lab to generate nanodiamonds particles. Code 6364 has also synthesized nanodiamond samples with a range of chemical purity and crystalline quality by laser ablation of carbon targets immersed in a liquid or by laser-induced plasma dissociation of liquid ethanol [4].

TEM analysis showed the nanodiamonds were well segregated, a few nm in size and distributed within a narrow size range as seen in Figure 3(a). A high-resolution image confirmed a lattice spacing of 2.06 Å, which is characteristic of the 111 phase of diamond (Figure 3(b) top image). The sampled region in Figure 3(b), center image corresponding to the area circled in red was analyzed via selected area electron diffraction (SAED). The analysis revealed spots that corresponded to the diamond [111] and [220] planes (Figure 3(b) bottom image). Electron energy loss spectroscopy (EELS) analysis of nanodiamond particles produced with 300 μJ laser pulses showed the characteristic nanodiamond signatures with the 35 eV edge in the low loss region (top graph in Figure 3(c)) and the sharp 290 eV onset at the carbon K-edge, as seen in the bottom graph in Figure 3(c) [5].

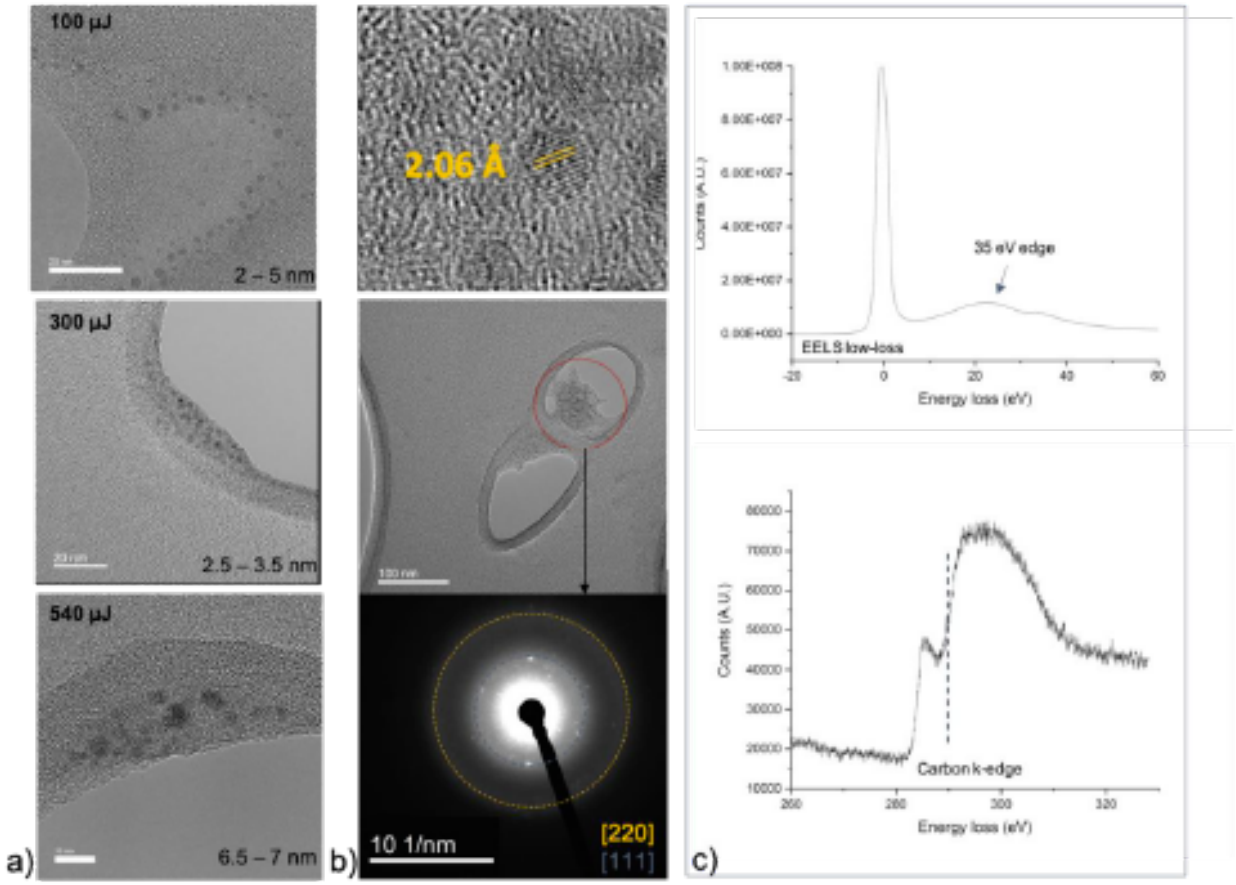

Fig. 3 -- (a) Bright-field TEM images of nanodiamond particles and size range vs. laser pulse energy (100 – 540 µJ). (b) Lattice spacing from high-resolution TEM analysis (top image), sample region (center image) for selected area electron diffraction (SAED) of nanodiamond grain (bottom image). (c) Electron energy loss spectroscopy of nanodiamond in the low-loss and carbon K-edge region. Analysis of (b) and (c) were conducted on particles synthesized using 300 µJ pulses [5]

The use of laser techniques for materials processing and modification has been investigated for many types of films and bulk materials by Code 6364. For the synthesis of nanodiamond particles, ultra-fast (fs) laser processing of ethanol was demonstrated under this program. In future work, we also plan to investigate the use of these lasers with fs pulses for generating nitrogen-vacancy defects on diamond thin films via direct-write techniques. The use of fs laser direct-write allows the delivery of the laser energy within much smaller volumes with spatial 3D placement resolutions of the order of 1 cubic micron or less. Such resolution is required to synthesize individual NV-centers at selected locations in a diamond sample. It is expected that further progress in laser direct-write with ultrashort pulses to first create the defect and then facilitate its diffusion to a desired location inside any type of diamond sample (i.e. single crystal bulk or thin film), will provide unique tools for the laser fabrication of materials and structures for quantum technologies as the ones investigated under this program.

5. Diamond substrate design

To isolate single photon emission, it is important to use diamond substrates that have a sufficiently low density of defects such that multiple excitations do not occur within a single optical pulse. To this end we sourced electronic grade diamond substrates from Applied Diamond Inc. that were ground down to a 10µm thickness. This resulted in two important optical advantages. The optical cross section of the thinner

substrate was drastically lower causing the average number of defects in a given beam spot diameter to be closer to 1, making it easier to couple out single photon emission from the NV center. Since defect emissions are close to isometric, a thinner substrate means that a smaller solid angle of emission is reflected off from the diamond surface and never reaches the detectors, regardless of the size of the numerical aperture of the collection optics.

6. Optically addressing single defects

In order to determine the emission statistics from the 10μm diamond substrate, we constructed a Hanbury-Brown-Twiss (HBT) interferometer designed to measure the average distance between detection events across two separate single photon avalanche photodiodes as shown in Figure 4. A successful demonstration of a single photon source would result in average separation between coincidences across detectors A and B equivalent to the lifetime of the NV centers as governed by spontaneous emission. Should this emission, instead, be altered by premature relaxation into the ground state as a result of stimulated emission, this distribution would change to reflect a completely different lifetime. The average separation of these photons would be governed by the repetition rate of the stimulating laser pulses and the strength of the initial exciting field. These results remain to be seen, and so an upper limit on the excitation rate of this system has not yet been determined.

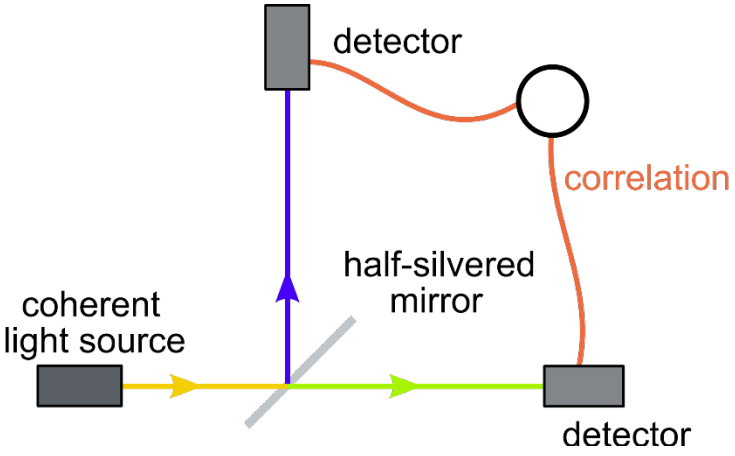

Fig. 4 -- A Hanbury-Brown-Twiss interferometer capable of measuring the coincidence distributions associated with single photon sources [6]

We have also begun designing a means of coupling NV centers in solution to a hollow core fiber that may prove to be a more useful means of addressing these defects as single photon sources. Ultrafast microscopy is in place in ultrafast optics laboratory in code 6364 to be able to address these systems.

7. Summary

This work focused on designing and modeling the coupling of NV center defects in diamond to photonic crystal cavities. We have made significant progress in the calculation of transition probabilities between eigenstates of NV centers and virtual states in the event of stimulated emission. These calculations demonstrate the means of polarization control via the relationship between incoming and stimulated photons. This relationship informs the design of the photonic crystal cavity, such that specific emission wavelengths will be preferred. There still remains a great deal of work in characterizing stimulating emission in NV centers and how it can drive the generation quantum photonic states in the future. We believe this still remains a promising route for the scalable generation of more sophisticated quantum light sources under operating conditions where expensive infrastructure components such as cryogenic cooling systems are simply not an option.

Acknowledgements

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This report is dedicated to Mr. Ray Auyeung from Code 6364 who served as Co-PI on this project until his untimely passing in January 2023.

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