Creation of High-Density Nitrogen-Vacancy Centers in CVD Diamond using High-Energy Photons from Ar+ Plasma

Prem Bahadur Karki,[1] Rupak Timalsina,[2] Mohammadjavad Dowran,[2] Ayodimeji E. Aregbesola,[1] Abdelghani Laraoui,[2,3] and Kapildeb Ambal[1†]

[1]Department of Mathematics, Statistics, and Physics, Wichita State University, 1845 Fairmount St. Wichita, KS 67260, USA.
[2]Department of Mechanical & Materials Engineering, University of Nebraska-Lincoln, 900 N 16th St. W342 NH. Lincoln, NE 68588, USA.
[3]Department of Physics and Astronomy and the Nebraska Center for Materials and Nanoscience, University of Nebraska-Lincoln, 855 N 16th St, Lincoln, Nebraska 68588, USA.
[†]Kapildeb.ambal@wichita.edu

Abstract: We use high-energy photons generated from Ar^+ plasma source to create a high-density and thick (up to a thickness of 150 μm) nitrogen-vacancy centers layer in a commercially available type-IIa CVD-grown diamond substrate.

The negatively charged nitrogen-vacancy (NV^-) centers are one of the leading solid state-based quantum sensors enabling ultra-sensitive magnetic field sensing for various scientific and commercial applications, including satellite-free positioning systems, magnetic gyroscopes [1], and precision magnetic field sensing [2]. High densities of NV^- centers are favored for precision magnetometry due to improved signal-to-noise and sensitivity from $1/\sqrt{N}$ enhancement (N is the number of spins) [2]. High-density NV^- centers provide high fluorescence intensity, enabling the utilization of regular photodetectors compared to complex detection schemes using single photon detectors.

We present an alternative method apart from common creation methods for a quick and cost-effective way to create high-density and thick (up to a thickness of 150 μm) NV^- centers layer in a commercially available type-IIa chemical vapor deposition (CVD)-grown diamond substrates (Fig. 1a) with as-grown nitrogen concentration of 1×10^{17} cm^{-3} (~ 0.6 ppm) by using high-energy photons generated from Argon (Ar^+) plasma source [3]. The direct absorption of the above bandgap photons creates a shower of electrons into the conduction band by exciting electrons from P_1 centers, other impurities, and the valence band. These electrons could travel through the conduction band of the diamond and recapture, thus converting NV^0 to a stable NV^- center.

The tri-acid (1 H_2SO_4: 1 HNO_3: 1 $HCLO_4$) cleaned and dry diamond substrate was first exposed to Ar^+ plasma (Fig. 1b) by using Trion Minilock-Phantom III Inductively Coupled Plasma (ICP.) Reactive Ion Etching (RIE) system for 30 s. The plasma process was performed with ICP power of 200 W, RIE power of 50 W, Ar gas flow of 5 sccm, and ICP-RIE chamber pressure of 10 mTorr respectively. After the plasma exposure, the diamond substrate was cleaned with the tri-acid mixture. The dry substrate was then annealed under vacuum (10^{-6} Torr) at 1100 ^{o}C for two hours with another subsequent triacid cleaning.

The optical and spin properties of the newly created NV^- centers in the diamond substrate (Fig. 1c) were measured using a custom-built confocal fluorescence microscope (Fig. 1d) consisting of a green laser (532 nm) for optical excitation of NV^- centers, a permanent magnet to apply a magnetic field for optical detected magnetic resonance (ODMR) measurements, a 100x microscope objective with a numerical aperture of 0.8 NA to focus light on the diamond substrate, and fluorescence collection.

Based on ODMR and fluorescence confocal microscopy measurements, we measured high density (~ 20,000 NV^- centers per diffraction-limited sample volume) of newly created NV^- centers distributed homogeneously over 150 μm deep (Fig. 1e) from the diamond surface facing the Ar^+ plasma source.

We obtain a shot noise-limited DC magnetic field sensitivity of ~ 104 $nT.Hz^{-1/2}$ and an AC magnetic field sensitivity of ~ 0.12 $pT.Hz^{-1/2}$ respectively. We also demonstrate real-time AC magnetic field sensing with a frequency of up to 90 Hz using the diffraction-limited active sample volume.

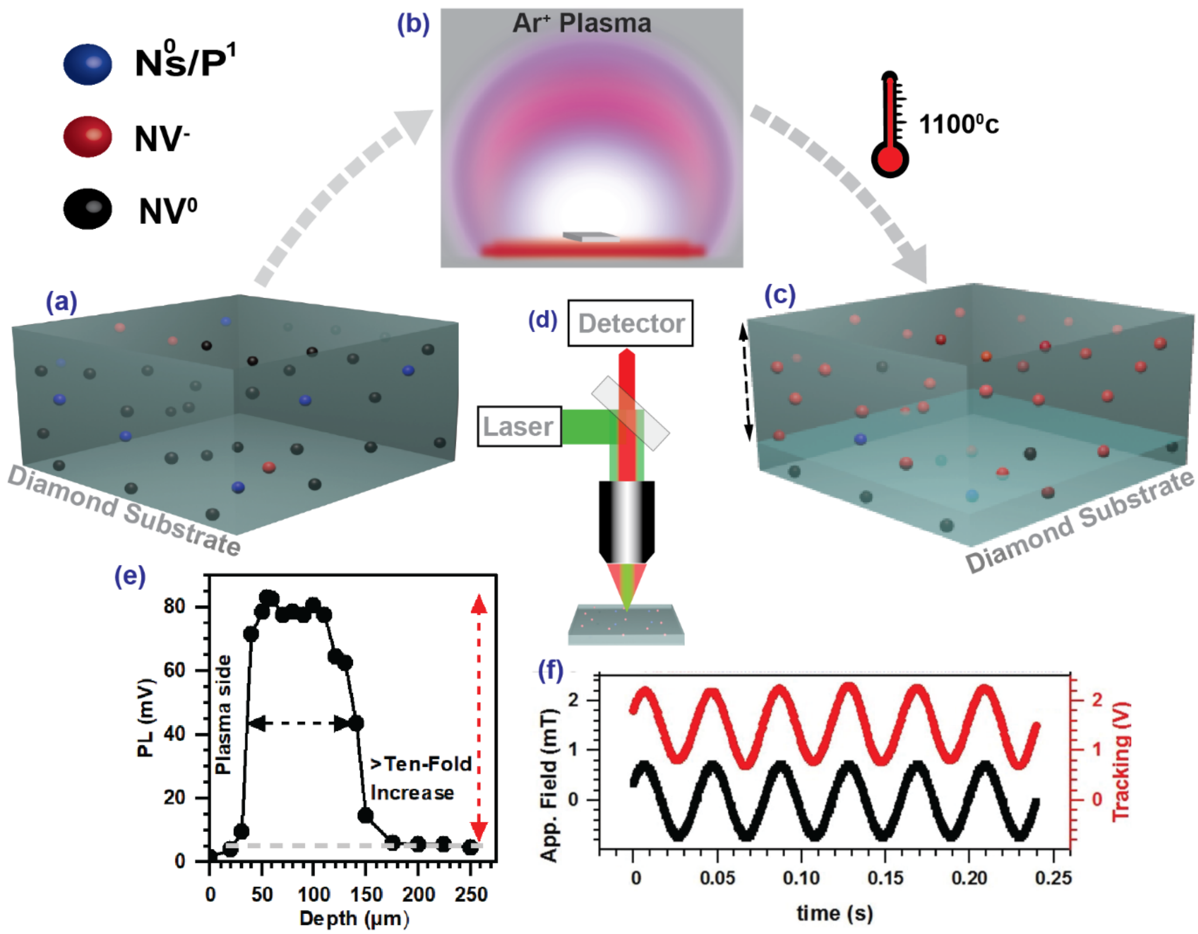

Fig. 1. Representation of the process and characterization of high-density NV^- centers layer using Ar+ plasma photons. (a), (b) and (c) The schematic of the process in creating high-density NV^- centers layer. (d) A schematic of the custom-made confocal optical microscope used to characterize the high-density NV^- centers. (c) The distribution of the created NV^- centers as a function of the depth from the side facing the plasma measured on 0.25 mm substrate. (f) Real-time AC magnetic field measured using the NV^- centers created by high-energy photons from Ar^+ plasma over the diffraction-limited sample volume.

KA would like to acknowledge the support of the National Science Foundation/EPSCoR R.I.I. Track-4 Award OIA-2033210 and NSF Award 2328822. The research was partially performed in the Nebraska Nanoscale Facility, which is supported by the National Science Foundation under Award ECCS: 2025298 and the Nebraska Research Initiative. PBK would like to acknowledge the support of the Wichita State University Convergence Science Initiative Program. A. L. would like to acknowledge the support of the National Science Foundation/EPSCoR R.I.I. Track-1: Emergent Quantum Materials and Technologies (EQUATE) Award OIA-2044049, and NSF Award 2328822.

[1] A. Jarmola, S. Lourette, V. M. Acosta, A. G. Birdwell, P. Blümler, D. Budker, T. Ivanov, and V. S. Malinovsky, *Demonstration of Diamond Nuclear Spin Gyroscope*, Science Advances **7**, eabl3840 (2021).

[2] C. L. Degen, F. Reinhard, and P. Cappellaro, *Quantum Sensing*, Rev. Mod. Phys. **89**, 035002 (2017).

[3] P. B. Karki, R. Timalsina, M. Dowran, A. E. Aregbesola, A. Laraoui, and K. Ambal, *An Efficient Method to Create High-Density Nitrogen-Vacancy Centers in CVD Diamond for Sensing Applications*, Diamond and Related Materials **140**, 110472 (2023).