New Delta Doping of Nitrogen for 2 Dimensionally Condensed NV Centers Using Nitrogen Terminated Diamond Followed by High Purity Diamond Growth

Tetsuya Tatsuishi¹, Takahiro Sonoda¹, Jorge J. Buendia¹, Taisuke Kageura^{1, 5}, Kazuto Kawakatsu¹, Yuki Hata¹, Kiro Nagaoka¹, Yu Ishii¹, Takashi Tanii¹, Moriyoshi Haruyama^{2, 3}, Keisuke Yamada², Shinobu Onoda², Wataru Kada³, Osamu Hanaizumi³, Alastair Stacey⁴, Tokuyuki Teraji⁵, Junichi Isoya⁶, Shozo Kono⁷, Hiroshi Kawarada^{1, 7}

¹ Waseda University, 3-4-1, Okubo, Shinjuku-ku, Tokyo 169-8555, Japan

Phone: +81-3-5286-3391 E-mail: tetsuya-mail-wu@fuji.waseda.jp

² National Institute of Quantum and Radiological Science and Technology, ³ Gunma University, ⁴ University of Melbourne,

⁵ National Institute for Materials Science, ⁶ University of Tsukuba,

⁷ Kagami Memorial Research Institute for Materials Science and Technology

Abstract

We have successfully made 2 dimensionally condensed NV centers from N-terminated (111) diamond with near monolayer coverage. The N on the surface has been embedded by high purity homoepitaxial (111) diamond film to form NV centers. The NV centers made by these techniques were highly aligned. This new method of fabricating NV centers is useful for application of NV centers to more sensitive quantum sensors.

1. Introduction

Nitrogen Vacancy (NV) center in diamond, which is consisted of a substitutional N and a vacancy adjacent to the N, is a promising candidate for quantum computing, quantum communication, and quantum sensor. One of the most promising application of NV center is magnetic sensor with high sensitivity even at room temperature. Since the sensitivity is inversely proportional to the cube of distance, near surface NV centers are preferable. However, appropriate surface termination is inevitable for the NV close to the surface to stabilize NV⁻. Oxygen (O) terminated surface is normally used because of its larger positive electron affinity (PEA, +1.3 eV) advantageous for the stability of NV⁻. Theoretically, N-terminated surface (Figure 1) possess strong PEA (+3.2 eV) [1]. Recently, we fabricated shallow and single NV centers followed by N-termination. The single NV center as magnetic sensor succeeded in detecting the signal from ¹H nuclear spins out of the diamond substrate with high stability [2, 3]. Generally the magnetic sensitivity limited by shot noise is expressed as equation below [4].

$$\delta B \simeq \frac{1}{g_e \mu_B C \sqrt{\eta} \sqrt{NT_2}} \tag{1}$$

where g_e is the NV electronic Lande factor, μ_B is the Bohr magnetron, *C* is the measurement contrast, η is the detection efficiency, *N* is the number of NV centers, and T_2 is the spin relaxation time. Therefore, to get better magnetic sensitivity, it is required to make NV center with longer spin coherence time or to increase the number of NV center. If the number of *N* is over 1, NV centers called as NV ensemble. In the case of NV ensemble, it is required to make NV center completely aligned with other NV, because non-aligned NV centers degrade the performance as high sensitive magnetic sensor [5]. It is difficult to fabricate highly aligned NV ensemble by ion implantation, because the random movement (diffusion) of vacancies in annealing cannot be controlled. Vacancies are positioned equally on 4 possible sites of N adjacent site and become to non-aligned NV center. On the other hand, highly aligned NV ensemble were fabricated by growth of N-doped diamond film on (111) diamond substrate [4] because 4 possible sites adjacent to N are not equal for vacancy. A top site is more preferable than other 3 equivalent sites to become vacancy. However, it is difficult to fabricate N-doped diamond film where density of NV center is beyond 1×10^{18} cm⁻³ because of both inefficient activation and the low sticking probability of N [6]. By the way, delta-doping of N was achieved by diamond growth on N-terminated (001) diamond and the estimated 2D concentration of NV center was only 1×10^{11} cm⁻² [6]. Of course, NV center in (001) delta doped layer would be not aligned because of 4 sites are equivalent. We propose more densified NV center by growth of diamond film on N-terminated (111) diamond (Figure 1) [1]. If the diamond film is grown on N-terminated (111) diamond as in Figure 2, NV centers highly aligned would be fabricated with a high 2D concentration. Recently, we partially succeeded to make truly delta doping of N from N-terminated diamond with half mono layer (ML) coverage embedded by high purity diamond film (111) growth.

2. Experimental Details

First, N-terminated (111) surface was fabricated N radical beam. It is a method generally used for molecular beam epitaxy (MBE) growth of III-nitride (III-N). To prevent unpredicted damage on the surface in this treatment, the N radical was irradiated far from the sample and N ions made by RF plasma were removed by ion canceller before irradiation [3]. After the fabrication of the N-terminated (111) diamond, thin layer (thickness is about 10 nm) of high purity diamond film were grown on the N-terminated (111) diamond. To avoid the incorporation of impurities such as N into diamond film during growth, we introduced gas purifiers of H₂ and CH₄ just before the gases get into the growth chamber. According to secondary ion mass spectroscopy (SIMS), density of N in the high purity diamond film was well below 1×10^{16} cm⁻³. A custom-built laser scanning confocal fluorescence microscope (CFM) was used to confirm the existence of NV center. It was confirmed by optical detection magnetic resonance (ODMR) whether NV centers we made were highly aligned or not.

3. Results and Discussions

The XPS spectrum of N-terminated (111) diamond fabricated by N radical exposure is shown in Figure 3. From this spectrum, the coverage of N was estimated to be 0.60 ML.

Figure 4(a) and 4(b) show the confocal PL-intensity mapping, obtained with 532 nm excitation laser. Figure 4(a) is the mapping before N radical exposure, which is the process to fabricate N-terminated diamond. Figure 4(b) is the mapping after diamond growth on N-terminated (111) diamond. Figure 4(b) has many spots where PL-intensity are over 100 kcps, but figure 4(a) has no point where PL-intensity is above 10 kcps. 100 kcps correspond to ten times of PL-intensity from single NV center. Therefore, NV ensemble which contains about 10 NV centers in square of 100nm×100nm was fabricated after diamond growth on N-terminated (111) diamond.

Figure 5 is ODMR spectrum when the static magnetic field was applied along the [111] axis ($\mathbf{B} \parallel [111]$). In this case, the ODMR spectrum of NV ensemble has only two dips (Figure 5), which might be derived from the NV center whose axis is aligned along [111]. On the other hand, non-aligned NV ensemble along [111] axis must have four dips [7].

4. Conclusions

N-terminated (111) diamond with high N coverage was fabricated by N radical exposure. NV ensemble was fabricated by diamond growth on N-terminated (111) diamond. In addition, NV ensemble was aligned along axis of [111]. Therefore, the new fabrication of NV center by growing diamond film on N-terminated (111) diamond is useful for application of NV center for highly sensitive magnetic sensor.

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Fig. 1 The structure of N-terminated (111) diamond.



Fig. 2 Image of NV centers fabricated by diamond growth on N-terminated (111) diamond



Fig. 3 XPS spectrum of N-terminated (111) diamond fabricated by radical beam exposure.



Fig. 4 The confocal PL-intensity mapping (a) before N radical exposure and (b) after diamond growth on N-terminated (111) diamond.



Fig. 5 ODMR spectrum when magnetic field applied along to [111] axis ($\mathbf{B} \parallel [111]$).