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Alignment and Coherence Time of 2D Shallow NV Ensemble Fabricated from (111) Diamond

K. Kanehisa[1], T. Tatsuishi[1], T. Sonoda[1], K. Kawakatsu[1], Y. Hata[1], T. Tanii[1], S. Onoda[2], A. Stacey[3], M. Moshkani[4], R. Mildren[4], J. Isoya[5], S. Kono[6], H. Kawarada[1, 6]

[1]Waseda Univ., 3-4-1, Okubo, Shinjuku-ku, Tokyo 169-8555, Japan
Phone: +81-3-5286-3391 E-mail: kanehisa1216@ruri.waseda.jp
[2]QST, [3]Univ. of Melbourne, [4]Macquarie Univ. [5]Univ. of Tsukuba, [6]ZAIKEN

Abstract

We established the method to fabricate two-dimensional (2D) shallow NV ensemble. We realized nitrogen-terminated (111) diamond surface with nitrogen radical exposure and grew very thin (approximately 10 nm) diamond film on the surface by microwave plasma-enhanced chemical vapor deposition (MPCVD). We counted the number of NV ensembles aligned in [111] direction of the NV ensembles which we observed. The probability of NV ensemble being aligned in [111] was 42 %. On the other hand, we changed the surface condition of N-terminated (111) diamond by thermal annealing. Then, we grew 10 nm diamond film on the surface. As a result, the probability of NV ensemble being aligned in [111] rose to 60 %. Also, s-factor developed from 0.44 to 0.52.

1. Introduction

Negatively charged nitrogen-vacancy (NV) center in diamond lattice has unique attributes, for example, long spin coherent time T_2 up to a few ms at room temperature [1] and a spin-triplet state which can be manipulated and read out optically. NV center is expected to apply for magnetic sensor [2] or quantum computer [3]. Especially, shallow NV center is desirable for the nano-NMR detection, such as ^{1}H nuclear spin [4]. The magnetic sensitivity of NV center is proportional to the square root of the number of NV centers which act as sensor [5]. Thus, it is expected that two-dimensional shallow NV ensemble performs the nanoscale NMR detection more sensitively. The nitrogen delta-doped layer was fabricated by epitaxial diamond growth on a nitrogen-terminated (100) diamond surface [6]. The depth of NV ensemble can be controlled this way. However, it is difficult to be align the NV ensemble in the case of N-terminated (100) diamond because NV centers aligned in each of the four directions are generated equally. NV ensemble must be aligned because the NV centers not aligned may deteriorate the magnetic sensitivity [7]. On the other hand, it is easy to align NV ensemble in [111] direction in the case of N-terminated (111) diamond. Fig. 1 shows the ideal N-terminated (111) diamond surface [8] and 2D shallow NV ensemble aligned in [111] direction. One nitrogen bonded to three carbons forms three-coordinate N-termination (C_3-N). Actually, we fabricated 2D shallow NV ensemble aligned in [111] direction by epitaxial diamond growth on N-terminated (111) diamond surface last year.

We measured optical detected magnetic resonance (ODMR) spectrums under the bias magnetic field along [111] in order to conform if NV ensemble was aligned in [111] or not. Then, three types of ODMR spectrums were found. Fig. 4 shows that (a) NV ensemble is aligned in [111] direction, and (b) NV ensemble has both NV centers aligned in [111] and ones aligned in [1-1-1], [-11-1], or [-1-11] direction (other direction), and (c) NV ensemble is aligned in other direction. Of all observed NV ensembles (33 NV ensembles), the proportion of NV ensembles showing ODMR spectrum (a) (14 NV ensembles) was calculated. As a result, the probability of NV ensemble being aligned in [111] was 42 %. From this result, we expected that there were not only C_3-N but also amino termination (C-NH_2) on the N-terminated (111) diamond surface. Fig. 2 shows the presumed real N-terminated (111) diamond surface and NV ensemble including NV center aligned in other direction. C-NH_2 on the (111) diamond surface has one nitrogen bonded to one carbon. So, it converted into NV center aligned in other direction after diamond film growth. We expected that C-NH_2 was removed more easily by thermal energy than C_3-N because of the number of bonds between a nitrogen and a carbon. Thus, we introduced the thermal annealing into the process in order to improve both the probability of NV ensemble being aligned in [111] and the coherence time.

2. Material and Method

N-terminated (111) diamond was realized by the nitrogen radical exposure using the molecular beam epitaxy system [4]. This method can reduce the damage done to the diamond surface. Then, the N-terminated (111) diamond was annealed at 400 ℃ vacuum in order to remove amino group termination. Finally, a pure diamond film of approximately 10 nm was grown on each N-terminated surface by MPCVD. The nitrogen concentration of diamond film on the nitrogen surface is less than 1.0×10^{16} cm^{-3}, which is the detection limit of SIMS measurement.

3. Experiment and Result

Nitrogen coverage of N-terminated surface before (a) and after (b) thermal annealing was derived from XPS spectrums. As a result, the nitrogen coverage decreased from 0.64 ML to 0.41 ML and is shown in Fig. 3. Additionally, we observed NV ensembles through a confocal fluorescence microscope (CFM) and measured ODMR spectrums. Of all observed NV ensembles (68 NV ensembles), the proportion of NV ensembles showing ODMR spectrum (a) (41 NV ensembles) was calculated. As a result, the probability of NV ensemble being aligned in [111] was 60 %. For the above results, C-NH_2 may decrease by introducing 400 ℃ annealing into the process.

We implemented spin measurement of NV ensembles. Coherence time T_2 was measured by Hahn echo pulse sequence, and T_2 can be extended by dynamical decoupling

(DD) pulse sequences (CPMG-N, XY8-k) by more effectively decoupling NV^- spins from decoherence sources. Also, s-factor was calculated by the equation of $T_2^{DD} = N^s \cdot T_2^{Hahn}$. N is the number of π pulses in the sequence. Fig. 5 shows the relation between the number of π pulses and T_2. For the unannealed sample, T_2 which was 2.45 μs by the Hahn echo (N=1), was extended to 13.8 μs by the XY8-8 (N=64), and the s-factor was 0.44. For the annealed sample, T_2 which was 3.82 μs by the Hahn echo, was extended to 30.8 μs by the XY8-8, and the s-factor was 0.52.

4. Summary and Conclusion

We removed C-NH_2 from N-terminated (111) diamond surface by thermal annealing, and grew a diamond film on the surface in order to fabricate 2D shallow NV ensemble aligned in [111] direction. As a result, the probability of NV ensemble being aligned in [111] rose from 44 % to 60 %, and both the coherence time and s-factor developed after introducing 400 °C annealing. It can be said that the development of coherence time is one step to realize more sensitively nano-NMR detection.

Acknowledgement

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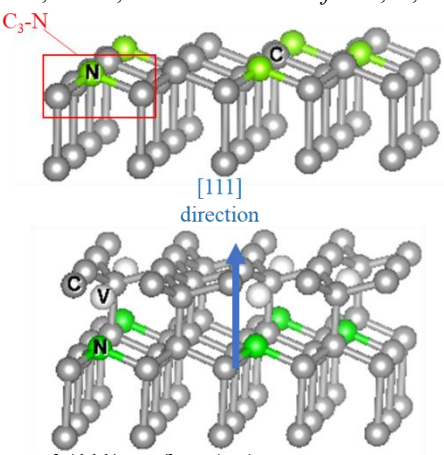

Fig.1.1 Ideal N-terminated (111) surface (up)
Fig.1.2 2D shallow NV ensemble aligned in [111] direction (down)

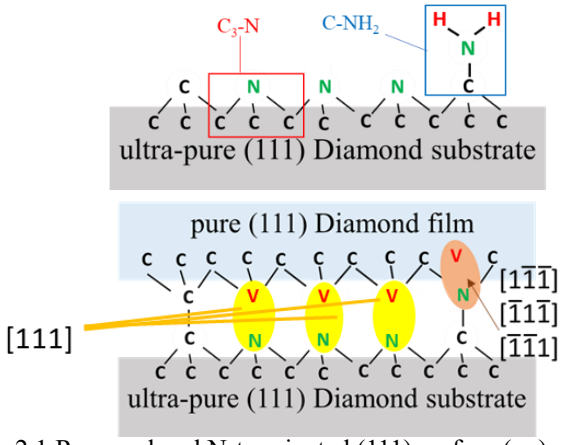

Fig.2.1 Resumed real N-terminated (111) surface (up)
Fig.2.2 NV ensemble including NV center aligned in other direction (down)

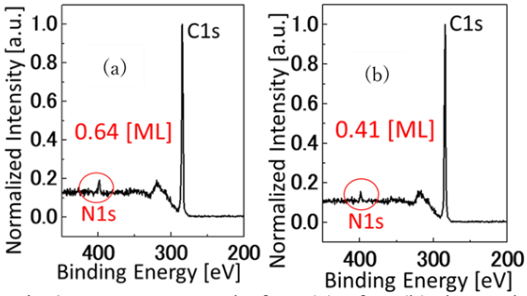

Fig.3 XPS spectrum before (a) after (b) thermal annealing N-term (111) diamond

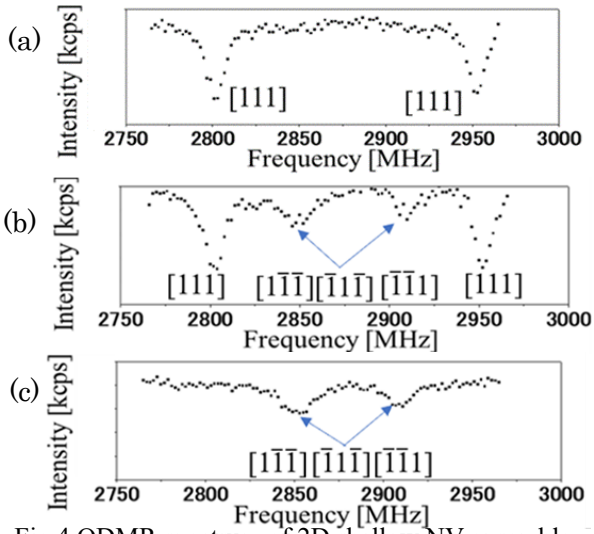

Fig.4 ODMR spectrum of 2D shallow NV ensemble

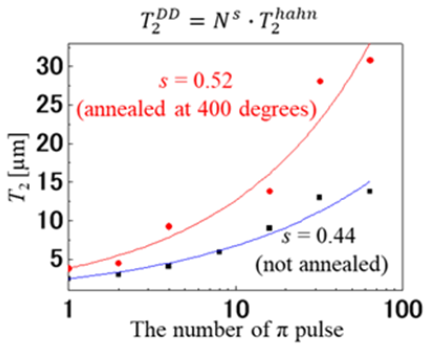

Fig.5 Relation between the number of π pulses and T_2