^{15}N-doped homoepitaxial diamond (100) free-standing crystal growth for characterizing nitrogen-related point defects

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1. Introduction

Research on weak magnetic field detection using NV centers in diamond has been intensively studied in the world. Controlling the nitrogen concentration [N] in the diamond crystal in the concentration range of 1-30 ppm is required to obtain higher magnetic sensitivity.[1–3] In this study, nitrogen-doped free-standing crystals were grown by microwave-plasma chemical vapor deposition (MPCVD) method, and the type and concentration of nitrogen-related point defects formed in these free-standing crystals were characterized by using various evaluation methods.

2. Experimental

Homoepitaxial diamond films were grown by using the MPCVD system developed at NIMS.[4] The HPHT-grown type-Ib (100) substrates were used for homoepitaxial film growth. In order to suppress the deterioration of crystallinity due to nitrogen doping, the growth condition of oxygen addition was applied.[5] ^{15}N enriched molecular nitrogen gas ($^{15}N_2$) was used as a nitrogen source. The CVD condition is as follows; the reaction pressure of 110 Torr, microwave power of 1.4 kW, methane concentration ratio (flow rate ratio of CH_4 to the total gas flow) of 10%, oxygen concentration (flow rate ratio of O_2 to the total gas flow) of 2% and substrate temperature of 1020–1090°C. The nitrogen-doped free-standing crystals with a thickness of 0.3 mm or more. The CVD layer and the substrate were separated by laser cutting from the side to obtain a free-standing diamond CVD plate. The type and concentration of nitrogen-related point defects were characterized by secondary ion mass spectrometry (SIMS), Fourier transform infrared (FTIR), and electron paramagnetic resonance (EPR).[6–7] In the SIMS analysis, the use of ^{15}N as the nitrogen source allowed the effects of doping (^{15}N signal) and external leakage (^{14}N signal) to be estimated separately. In the EPR measurement, signals of the negatively charged NV^- center with hydrogen (NVH^- center) and the substitutional nitrogen in neutral charge states (P1 center) were separated by using ^{15}N, resulting in accurate quantification of these defects.

3. Results and Discussion

We have found that (1) nitrogen is uniformly incorporated in the crystal (standard deviation is less than 10% of the concentration), (2) most of the nitrogen is isolated substitutional nitrogen, (3) hydrogen is incorporated into the crystal in approximately the same amount as nitrogen, (4) about 10% of the hydrogen is in the form of NV^- center with hydrogen (NVH^- center) and (5) both NVH center and NV center are mostly negatively charged. The nitrogen concentration in the diamond crystal has been controlled by changing the N/C gas ratio in a wide doping range from 10 ppb to 10 ppm.

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