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Abstract of dissertation

Study of diamond crystal growth for highly sensitive quantum sensors

(高感度磁気センサに向けたダイヤモンドの結晶成長の研究)

Takeyuki Tsuji

Nitrogen-vacancy centers in diamond are attracting attention as a next-generation magnetoencephalography (MEG). Unlike existing MEG based on superconducting quantum interference devices (SQUIDs), which only operate at cryogenic temperatures and require a large cooling system, NV centers can operate at room temperature and are expected to significantly reduce the device size of MEG. However, the magnetic sensitivity of NV centers has been 15 $\mathrm{pT}/\sqrt{\mathrm{Hz}}$, lower than that of SQUIDs (ranging from 100 to 1 $\mathrm{fT}/\sqrt{\mathrm{Hz}}$). Therefore, enhancing the magnetic sensitivity of NV centers to under 1 $\mathrm{pT}/\sqrt{\mathrm{Hz}}$ is needed for the miniaturization of MEG.

The magnetic sensitivity of NV centers is enhanced by improving the contrast (C), number of NV centers (N) and spin dephasing time (T_2^*). Firstly, controlling the alignment of NV centers is needed to improve C. NV centers can be aligned in four directions and each NV center is sensitive only to the magnetic field component parallel to its orientation axis. Therefore, aligning all NV centers in a single specific direction leads to a four-fold increase in C. Secondly, increasing the thickness of diamond film can improve N. Thirdly, mitigating stress distribution in the diamond lattice can improve T_2^*. NV center can be formed using the Chemical Vapor Deposition (CVD). To achieve the required magnetic sensitivity for MEG, it is needed to realize diamond film containing aligned NV centers with a thickness of 100 μm or more and mitigating stress distribution.

This study focused on enhancing the magnetic sensitivity of NV centers by increasing the thickness and mitigating the stress distribution of diamond films with aligned NV centers using CVD. Firstly, the growth rate of CVD diamond film was improved by introducing high plasma power density CVD. This study introduced high plasma power density CVD, which enhanced gas decomposition efficiency. By reflectively concentrating the microwave plasma directly on the diamond substrate with a spherical chamber, the plasma power density in this study was 2.5 times higher than in previous studies. As a result, the growth rate of step-flow growth for 6.6 μm/h was achieved which was ten times higher than previous studies. Secondly, the misorientation-angle dependence of NV center characteristics was evaluated. To introduce step-flow growth required for realizing aligned NV centers, the (111) diamond surface needed to be

polished at an angle (=misorientation-angle θ_{mis}) in the $[\bar{1}\bar{1}2]$ direction. The maximum thickness of the CVD film increases as the θ_{mis} is increased. In this study, the θ_{mis} dependence (ranging from 0.5° to 10°) of NV center properties was evaluated, revealing that the product of NV center density and T_2^* was higher at larger θ_{mis}. In addition, a thick diamond film containing aligned NV centers with a thickness of 100 µm or more was successfully obtained using a diamond substrate polished at a high θ_{mis} of 10°. Thirdly, the stress distribution in the diamond film was mitigated to improve T_2^*. The T_2^* of NV centers is primarily limited by electron spin bath around the NV centers and the stress distribution. This study discovered that increasing the θ_{mis} on the diamond substrate from 2° to 10° resulted in a reduction of the stress distribution by more than 1/10 and T_2^* approached its value limited by the electron spin bath.

In conclusion, since these technologies contribute to the fabrication of diamond materials with high magnetic sensitivity, this study pave the way for the highly sensitive quantum sensors.