

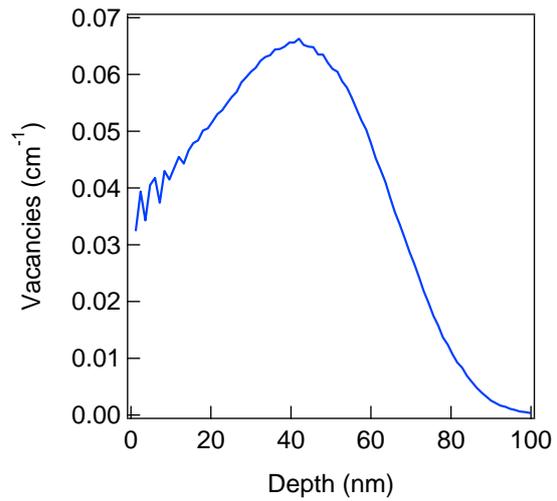
Supplementary material

Spin property improvement of boron vacancy defect in hexagonal boron nitride by thermal treatment

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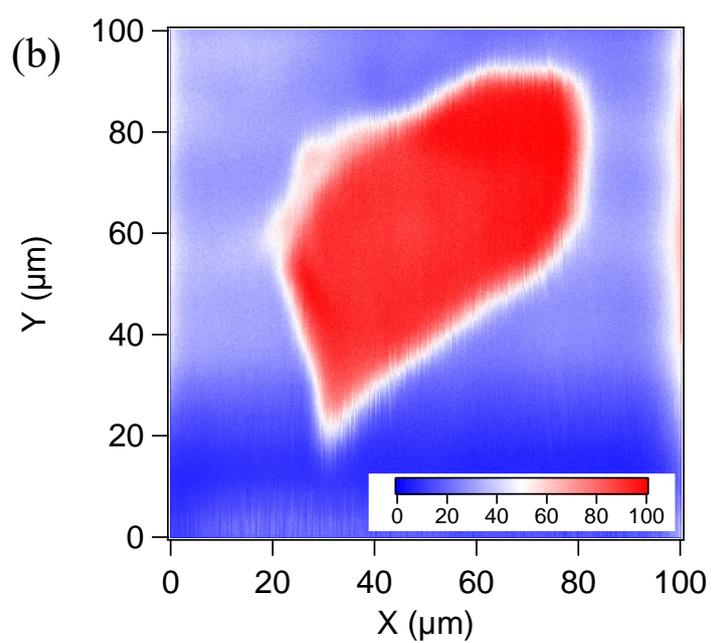
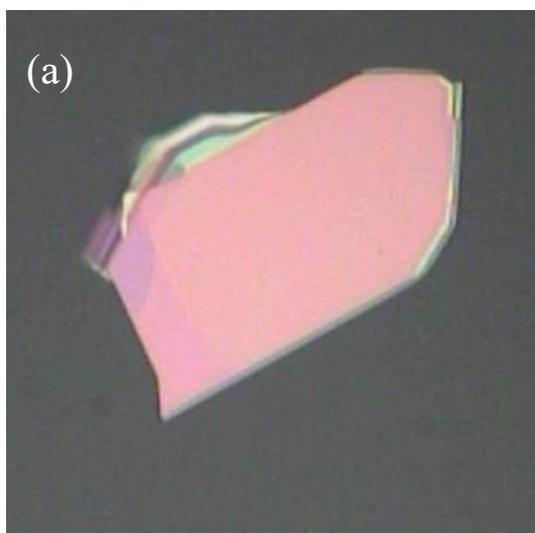
Supplementary Figures

Supplementary Figure S1



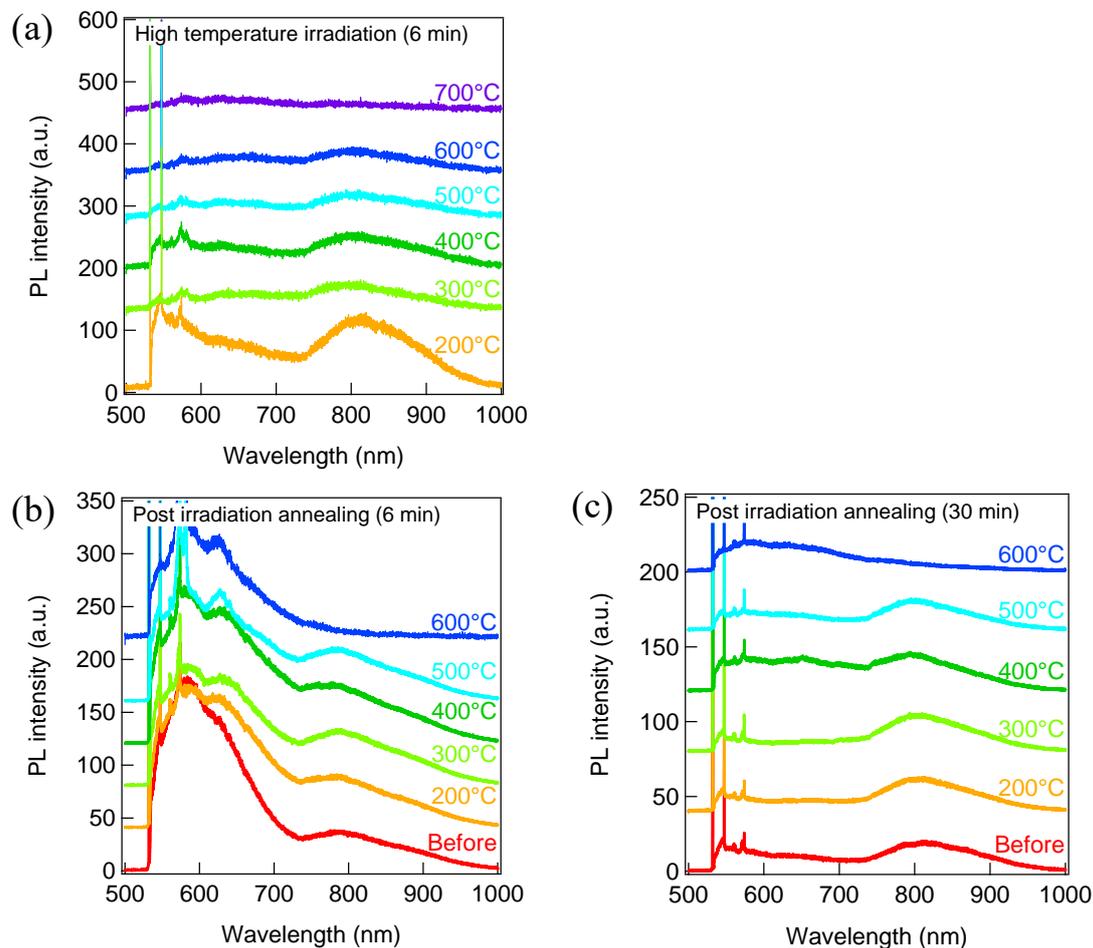
SRIM simulation result for irradiating 20 keV-N ion (equivalent to 40 keV-N₂ ion) on hBN layer. According to this simulation, V_B⁻ defects are formed within 100 nm from the surface of the hBN flake. The value on the vertical axis multiplied by the irradiation fluence is the actual defect concentration. In this experiment, the fluence was $1 \times 10^{15} \text{ cm}^{-2}$.

Supplementary Figures S2



(a) Optical microscope image of typical hBN flake. (b) PL mapping of hBN flake after ion irradiation. This indicates that V_B^- is formed uniformly in the flake.

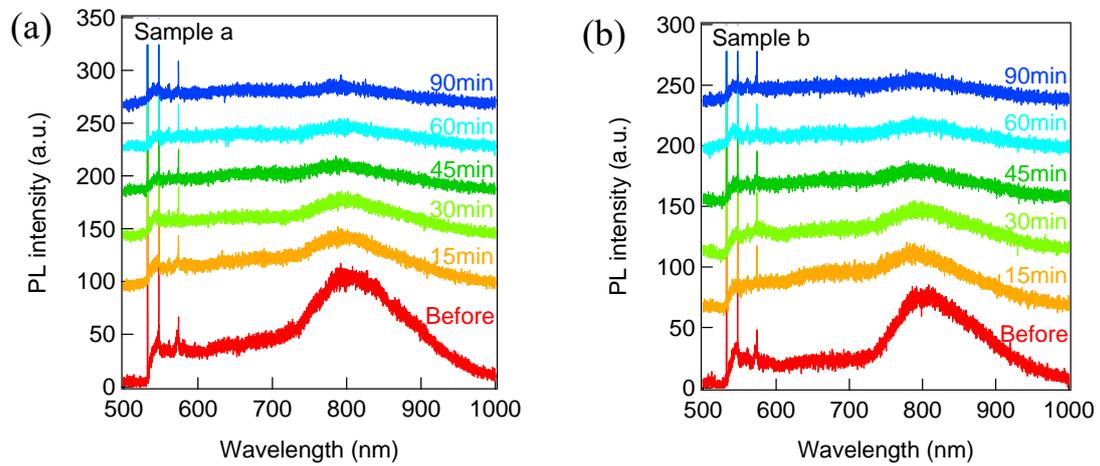
Supplementary Figures S3



(a)-(c) Temperature dependence of PL spectra. The broad peak at ~820 nm is PL emission from V_B^- . For Fig. 1, PL intensity is integration of a fitting line which is obtained by fitting with a Gaussian function after subtracting a tail of the peak at 500-700 nm fitted with an exponential function as a background. (a) Spectra for samples ion irradiated at 200-700 °C. Ion irradiation time is 6 min. (b)(c) Spectra for samples annealed at 200-600 °C after ion irradiation at room temperature. The annealing time for (b) is 6 min and for (c) is 30 min. As the temperature is increased, the PL peak is eventually not observed,

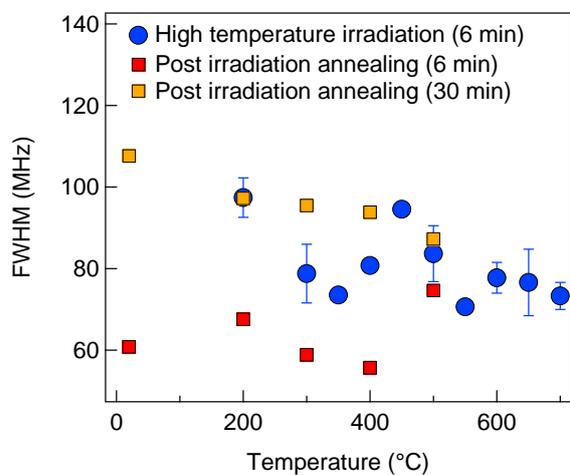
which indicates that the V_B^- defects are annihilated due to heat treatment. For post irradiation annealing, the origin of the PL signal in 500-700 nm may be assigned to $V_B C_N$. However, the signal intensity changed little before and after post annealing. Therefore, the formation of $V_B C_N$ is not major process for V_B annihilation.

Supplementary Figures S4



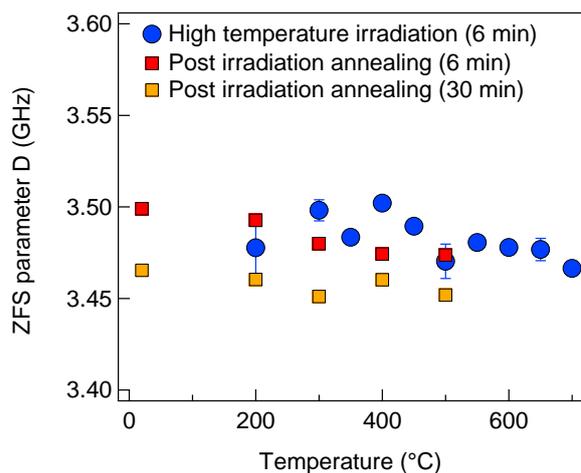
(a)(b) PL spectra as a function of annealing time (annealing temperature was 550 °C) for two samples (denoted by a and b) prepared by room temperature ion irradiation. PL emission at ~820 nm from V_B^- decreased with annealing time.

Supplementary Figure S5



Temperature dependence on the FWHM of different ODMR spectra. Regardless of the treatment method, the FWHM decreased slightly with increasing temperature, but overall, no significant change was observed.

Supplementary Figure S6



Temperature dependence of the ZFS parameter D . D is almost constant regardless of temperature and method of thermal treatments.

