Monodisperse Five-Nanometer-Sized Detonation Nanodiamonds Enriched in Nitrogen-Vacancy Centers

Journal Article

Author(s):
Terada, Daiki; Segawa, Takuya F.; Shames, Alexander I.; Onoda, Shinobu; Ohshima, Takeshi; Ōsawa, Eiji; Igarashi, Ryuji; Shirakawa, Masahiro

Supplementary Information

Mono-Disperse Five-Nanometer-Sized Detonation Nanodiamonds Enriched in

Nitrogen-Vacancy (NV⁻) Centers

Daiki Terada[1,2], Takuya F. Segawa*[1,3], Alexander I. Shames[4], Shinobu Onoda[2,5], Takeshi Ohshima[2,5], Eiji Ōsawa[6], Ryuji Igarashi[1,2], Masahiro Shirakawa[1,2]

1 Department of Molecular Engineering, Graduate School of Engineering, Kyoto University, Nishikyo-Ku, Kyoto 615-8510, Japan.

2 Institute for Quantum Life Science, National Institutes for Quantum and Radiological Science and Technology, 4-9-1, Anagawa, Inage-Ku, Chiba 263-8555 Japan.

3 Laboratory for Solid State Physics, Eidgenössische Technische Hochschule (ETH) Zürich, CH-8093 Zürich, Switzerland.

4 Department of Physics, Ben-Gurion University of the Negev, 8410501 Beer-Sheva, Israel.

5 Takasaki Advanced Radiation Research Institute, National Institutes for Quantum and Radiological Science and Technology, 1233 Watanuki, Takasaki, Gunma 370-1292, Japan.

6 NanoCarbon Research Institute, AREC, Shinshu University, Ueda, Nagano 386-8567, Japan.
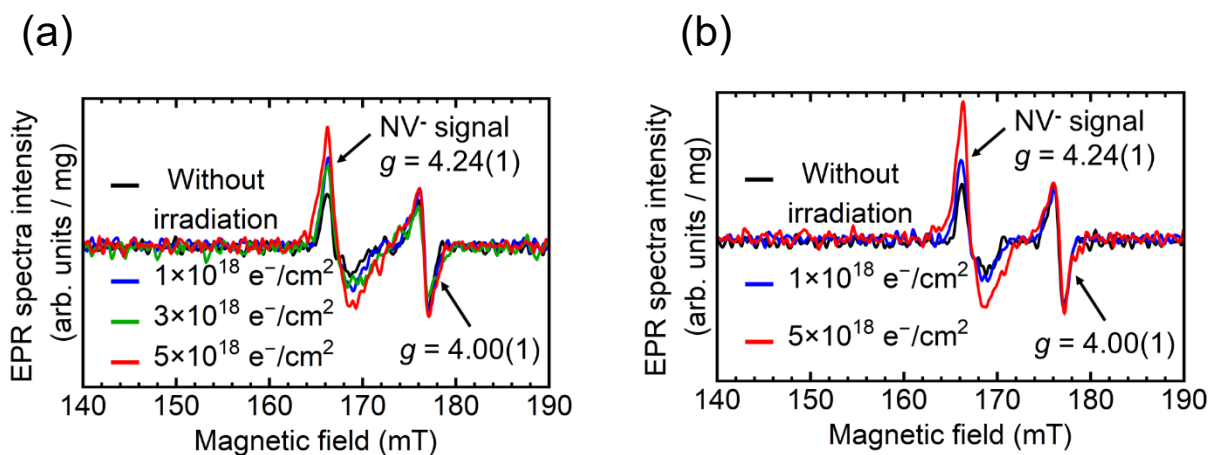
* E-mail: segawat@ethz.ch

Figure S1. All half-field EPR spectra with different irradiation fluences normalized per weight and after baseline correction. Non-irradiated (black), 1×10^{18} e^- /cm^2 (blue), 3×10^{18} e^- /cm^2 (green) and 5×10^{18} e^- /cm^2 (red) irradiated DND are shown without (a) or with annealing (b). The NV$^-$ signal appears at $g = 4.24(1)$ for the applied microwave frequency of 9.87 GHz.
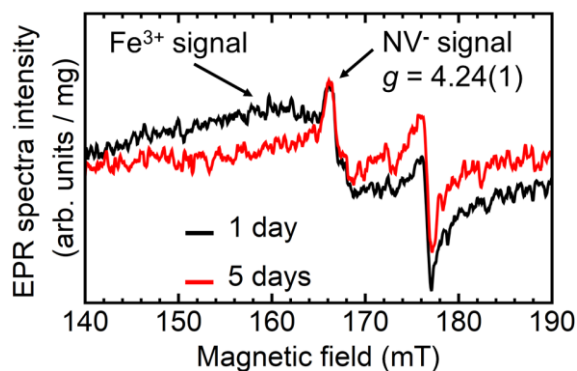

Figure S2. Half-field EPR spectra of the NV$^-$ signal of non-irradiated DND after 1 day (black) and 5 days of boiling acid treatment (red). This illustrates the decrease of the broad underlying Fe^{3+} signal, which is overlapping with the NV$^-$ signal.
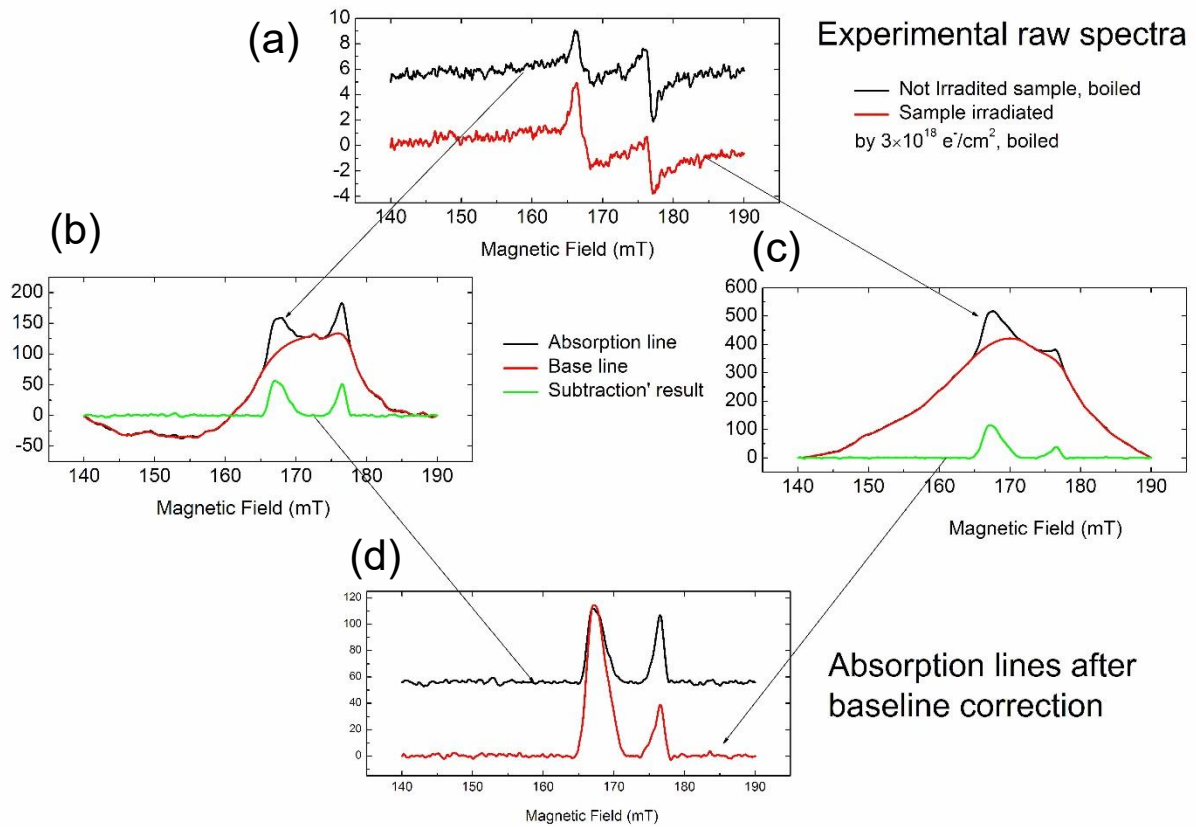
Figure S3. Illustration of the processing of the half-field EPR spectra. (a) Raw EPR spectra of non-irradiated (black) and 3×10^{18} e$^-$/cm^2 irradiated (red) DND samples after the boiling acid treatment. As common for CW EPR spectra, they are plotted as a first derivative with respect to the magnetic field. (b,c) This is followed by the integration of the EPR spectra (black). A cubic spline function is created to extrapolate the background signal from Fe^{3+} ion containing impurities as well as other background contributions (red). This is subtracted from the integrated signal resulting in the corrected absorption spectra (green). Fig. (b) shows this for the non-irradiated, Fig. (c) for the 3×10^{18} e$^-$/cm^2 irradiated samples. (d) The integrals of the peak at $g = 4.24(1)$ are proportional to the total number of NV$^-$ defects in the measured sample.
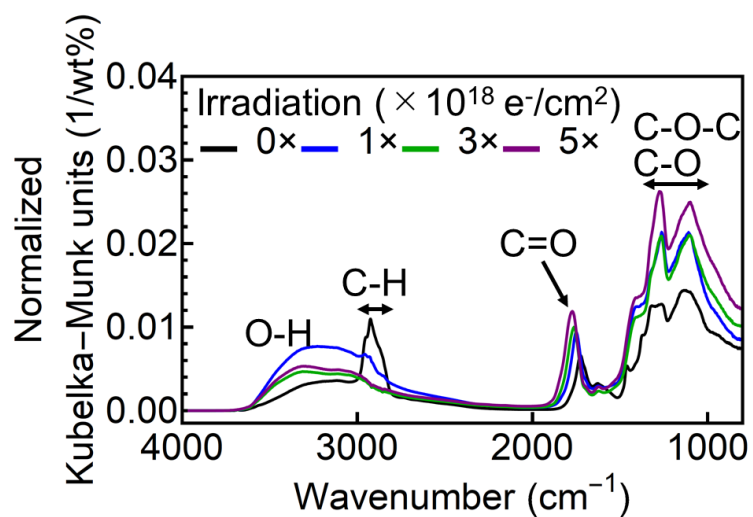
Figure S4. DRIFT spectra of non-irradiated (black), 1×10^{18} e⁻/cm² (blue), 3×10^{18} e⁻/cm² (green) and 5×10^{18} e⁻/cm² irradiated (purple) DND samples. While the C-H stretching peak (2800-3000 cm⁻¹) disappears after initial irradiation, C=O (1770 cm⁻¹) and C-O or C-O-C bands (1000-1350 cm⁻¹) become more intense with increasing electron irradiation fluences.