## Supplementary Materials

## Electrical tailoring of the photoluminescence of silicon-vacancy

## centers in diamond/silicon heterojunctions

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**Figure S1**. The PL spectrum of the as-deposited diamond film excited by a 785 nm laser at liquid nitrogen temperature (77 K). A clear peak located at 948 nm indicates the presence of neutrally-charged  $SiV^0$  centers. The inset shows the Raman spectra of the diamond film under the same excitation laser.



**Figure S2**. The SEM images of the diamond films annealed at various temperatures and durations: (a) 600°C-30 min; (b) 600 °C-60 min; (c) 700 °C-30 min.



**Figure S3**. XPS spectra of the C1 core carbon of the as-deposited samples before and after air annealing treatment: (a) the as-deposited sample; (b) the 600-30 sample; (c) the 600-60 sample; (d) the 700-30 sample. Due to the insulting nature of diamond, all the spectra are calibrated by aligning the sp<sup>3</sup> C-C peak to the known literature value of 284.8 eV.



**Figure S4**. The PL spectra of the as-annealed sample before and after acid treatment. In this process, the as-annealed (600-30) sample was soaked in mixed acid ( $H_2SO_4$ :  $HNO_3 = 3:1$ ) at a temperature of 130°C for 120 min, in order to etch the surface sp<sup>2</sup>-C introduced in the annealing process. The PL result clearly shows that the surface graphitization plays a negligible role on the PL intensity of SiV<sup>-</sup> centers in the whole depth.



**Figure S5**. The water contact angle of the as-deposited samples before and after air annealing treatment: (a) the as-deposited sample; (b) the 600-30 sample; (c) the 600-60 sample; (d) the 700-30 sample.



Figure S6. The PL signals collected at the region about 2 mm away from the electrodes with applying bias voltages on the diamond/  $n^+$ -Si heterojunction.



**Figure S7**. The PL spectra of the diamond/n<sup>+</sup>-Si heterojunctions at different forward bias voltages.



**Figure S8**. UPS valence band spectrum of the as-deposited H-terminated diamond film, exhibiting the energy difference from vacuum level ( $E_{VAC}$ ) to valence band maximum ( $E_V$ ) is estimated to 21.2 eV - 15.9 eV = 5.3 eV. The electron affinity of diamond ( $\chi$ ) was derived by:  $\chi = E_{VAC} - E_C = E_{VAC} - E_V - E_g^{dia} = 5.3 - 5.47 = -0.17 eV$ . All energies are measured relative to the common Fermi level that is determined from a reference gold foil.