Supporting Information

High-quality diamond microparticles containing SiV centers grown by chemical vapor deposition with preselected seeds

Tongtong Zhang,[a, #] Madhav Gupta,[a, #] Jixiang Jing,[a] Zhongqiang Wang,[b] Xuyun Guo,[c] Ye Zhu,[c] Yau Chuen Yiu,[a, d] Tony K.C. Hui,[d] Qi Wang,[b,*] Kwai Hei Li,[e, *] and Zhiqin Chu[f, *]

[a] Department of Electrical and Electronic Engineering, The University of Hong Kong, Pokfulam Road, Hong Kong, China

[b] Dongguan Institute of Opto-Electronics, Peking University, Guangdong, China

[c] Department of Applied Physics, The Hong Kong Polytechnic University, Hung Hom, Hong Kong, China

[d] Master Dynamic Ltd., Hong Kong, China

[e] School of Microelectronics, Southern University of Science and Technology, Shenzhen, China

[f] Department of Electrical and Electronic Engineering, Joint Appointment with School of Biomedical Sciences, The University of Hong Kong, Pokfulam Road, Hong Kong, China

[#] These authors contributed equally to this work.

[*] Correspondence and requests for materials should be addressed to Q.W. (wangq@pku-ioe.cn), K.H.L. (khli@sustech.edu.cn), and Z.Q.C. (zqchu@eee.hku.hk).

1. Experimental

1. 1 Salt-assisted air oxidation (SAAO) of NDs

0.5 g NDs with a mean particle size of 50 nm (HPHT, PolyQolor, China) were mixed with 2.5 g Sodium Chloride (NaCl, 99.5 %, Sigma-Aldrich), and they were heated at 500 °C for 1 hour in air. The resultant sample was dispersed in deionized (DI) water and sonicated for 1 hour, and the NDs were then purified with DI water three times by centrifugation. The purified NDs were re-dispersed in DI water and sonicated for 10 min to obtain well-dispersed NDs suspension for the CVD diamond growth.

1.2 Chemical vapor deposition (CVD) of diamond microparticles on Si substrate

The Si substrate was firstly treated with hydrogen plasma by the microwave-plasma assisted chemical vapor deposition (MPCVD) system (Seki 6350) for 10 minutes. Then the CVD diamond seeds, i.e., 50 nm HPHT SAAO NDs and commercially available DNDs (<10nm, TCI) were spin-coated on the standard single-crystal Si (100) wafers (2 inches). And the diamond samples were grown with a gas mixture of H_2/CH_4 (94/6) under fixed power and pressure conditions for 80 minutes. The plasma conditions were optimized to obtain a relatively large number of diamond microparticles with good crystalline quality.

1.2 Air oxidation of the CVD grown diamond microparticles

The as-grown CVD diamond samples (both SAAO ND- and DND-grown diamonds) were oxidized in air at 600 °C for 1 hour to improve the photoluminescence (PL) intensity of SiV centers.

2. Characterizations

The morphology of the diamond microparticles was analyzed by a scanning electron microscope (SEM, Hitachi S-4800) and a transmission electron microscope (TEM, FEI Tecnai G2 20 S-TWIN). The element compositions of the grown diamond were analyzed by the energy-dispersive X-ray spectrometer (EDX) equipped on the FEI Tecnai G2 TEM, and the electron energy loss spectrometer (EELS) equipped on a scanning TEM (STEM, JEM-2100F). The crystallinity of the grown diamond was analyzed using X-ray diffraction (XRD, D8 Advance).

The Raman spectra of the samples were measured by a Renishaw InVia Raman microscope, which is equipped with a Leica 100X NA0.85 objective lens focusing a 633 nm excitation laser onto the sample plane and collecting the backscattered Raman scattered light, and a 633 nm long pass filter blocking the backscattered 633 nm excitation laser. The total accumulated Raman spectra were obtained with the Raman Microscope operated in the sample scanning mode. In the sample scanning mode, the sample was in a two-dimensional scanning motion in the sample plane with respect to the stationary focused 633 nm excitation laser. In total, Raman spectra from 109 pixels × 109 pixels over a region of 54 μm × 54 μm on each sample were collected and summed up into one single spectrum for each sample. Therefore, our measurement is equivalent to the accumulated Raman signal over a large region under uniformed multibeam excitation, which would clearly suggest the averaged information (e.g., crystallinity) of the ensemble diamond microparticles grown within the selected region (54 μm × 54 μm) on the Si substrate.

Optical characterizations of the sample were done using a lab-built widefield microscope (Fig. S2). A continuous-wave 532nm laser beam was focussed on the back focal plane of a high NA (0.95) objective (Olympus UPLXAPO40X), and the emitted fluorescence was detected by a compact CCD spectrometer (Thorlabs CCS175/M). A widefield lens (f = 30 mm) was placed before the objective, focusing the incident laser beam on the back focal plane of the objective. This resulted in a Gaussian beam with FWHM = 18 μm coming out of the

objective and shining on the diamond sample. The excitation laser and PL signal were separated using dichroic mirror (DM) with a cut-on wavelength of 605 nm, whereas a 650 nm long pass filter was inserted in the detection pass to filter out the excitation laser further. Notice that all the PL measurements were conducted under the same conditions (fixed laser power, the same integration time of spectrometer, etc.); therefore, the measured PL spectra from two samples (DND- and SAAO ND-grown diamond microparticles) could be used for direct comparison. The temperature of the diamond sample was stabilized using an electronic temperature-controlled system. The sample was glued to a metal ceramic heater (Thorlabs, HT24S) and a platinum thermistor (Thorlabs, TH100PT). The heater and the thermistor were both electronically connected to a proportional-integral derivative (PID) temperature controller (Thorlabs, TC200), stabilizing the sample temperature with a precision of ±0.1 °C. Notice that all of our optical measurements in the current study were for ensemble microparticles; thus, we could directly perform those measurements on as-grown ones (e.g., diamond layers on the Si substrate).

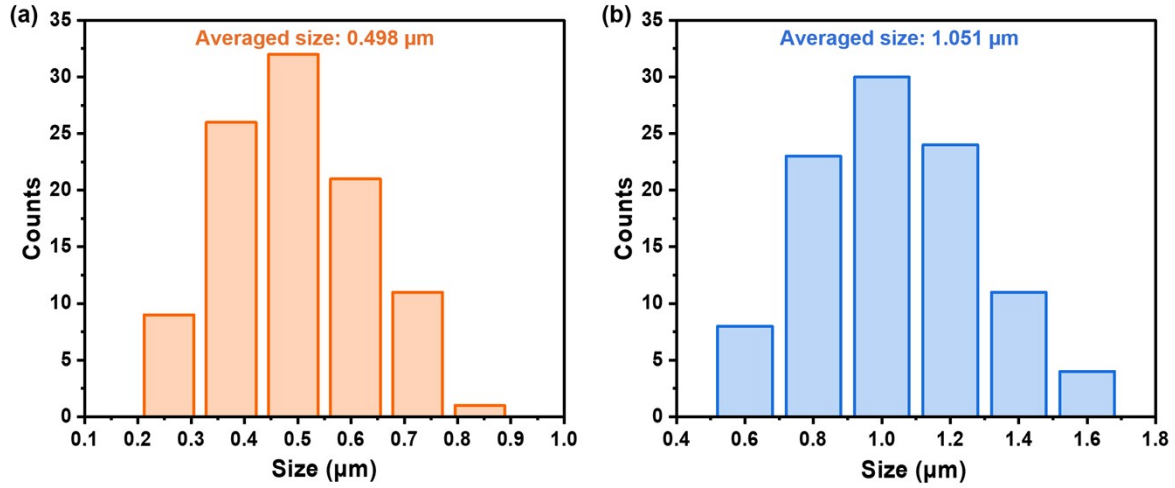

Fig. S1 The histograms of the size of (a) DND- and (b) SAAO ND-grown diamond microparticles obtained by randomly choosing 100 particles from their electron microscopy images for each sample.

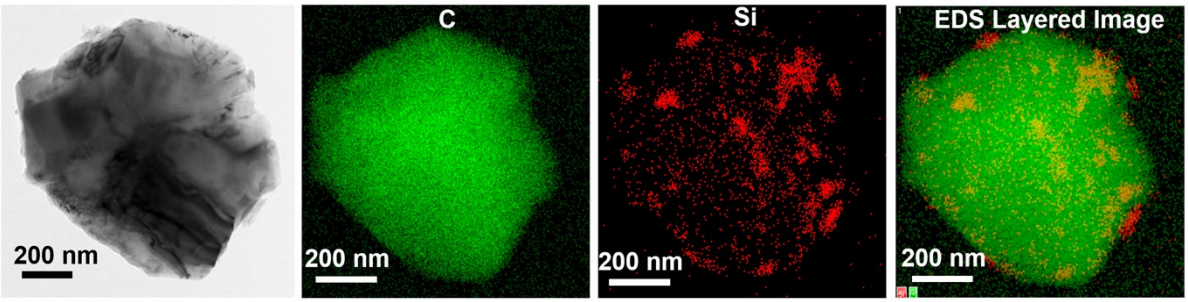

Fig. S2 TEM image and corresponding EDS elemental maps of the SAAO ND-grown diamond microparticles.

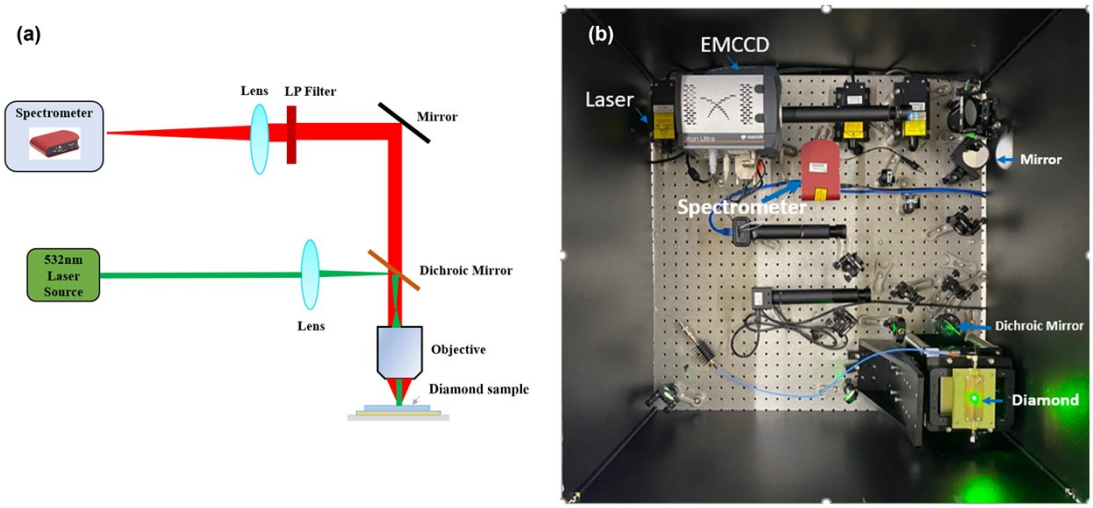

Fig. S3 (a) Schematic of the optical setup. (b) Experimental setup of the widefield microscope.

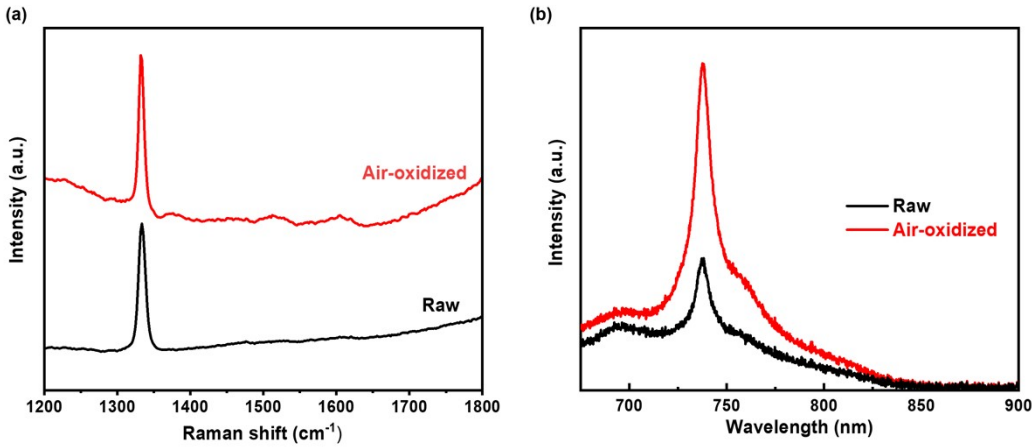

Fig. S4 (a) Raman and (b) PL spectra of the SAAO ND-grown diamond microparticles before and after air oxidation (600 °C, 1 hour), indicating the improved PL intensity of SiV centers after the air oxidation treatment, without any destruction to the diamond crystal.

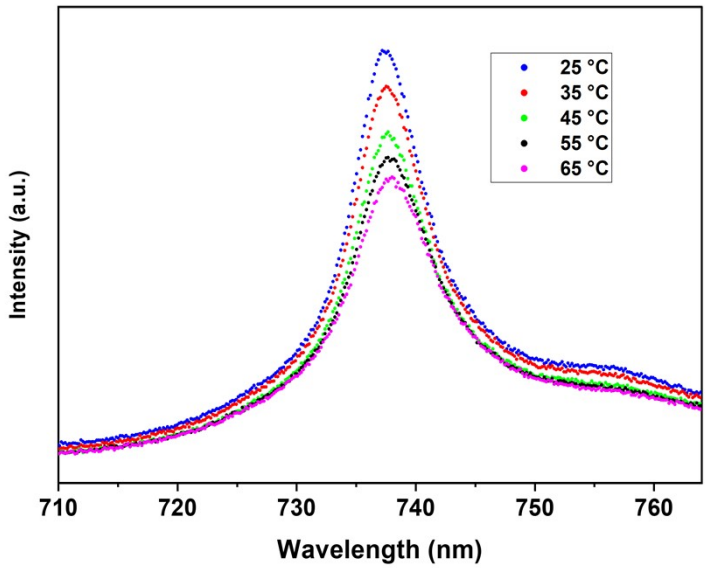

Fig. S5 The PL spectra of the SiV centers in SAAO ND-grown diamond microparticles at different temperatures. A clear red-shift of the optical ZPL with increasing sample temperature

can be seen, along with increasing linewidths (FWHM) and decreasing DWF. A larger temperature range (25–65 °C) has been chosen for this measurement to give a clear visual demonstration of the corresponding ZPL parameters shifting with increasing temperature.

3. Data Analysis

The measured spectra are fitted to the equation below: a sum of two Lorentzian curves from ZPL and phonon sideband (PSB), and a background noise $B(\lambda)$.[1]

$$\Phi(\lambda) = R_0\left[A_{zpl}\frac{\gamma^2}{4(\lambda-\lambda_{zpl})^2+\gamma^2} + \frac{\Gamma^2}{4(\lambda-\lambda_{psb})^2+\Gamma^2} + B(\lambda)\right]$$

The relevant extracted parameters from the fitting are:

(1) SiV ZPL Position: λ_{zpl}.

(2) SiV Linewidth (FWHM): γ.

(3) DWF (Ratio of ZPL and PSB intensity): A_{zpl}.

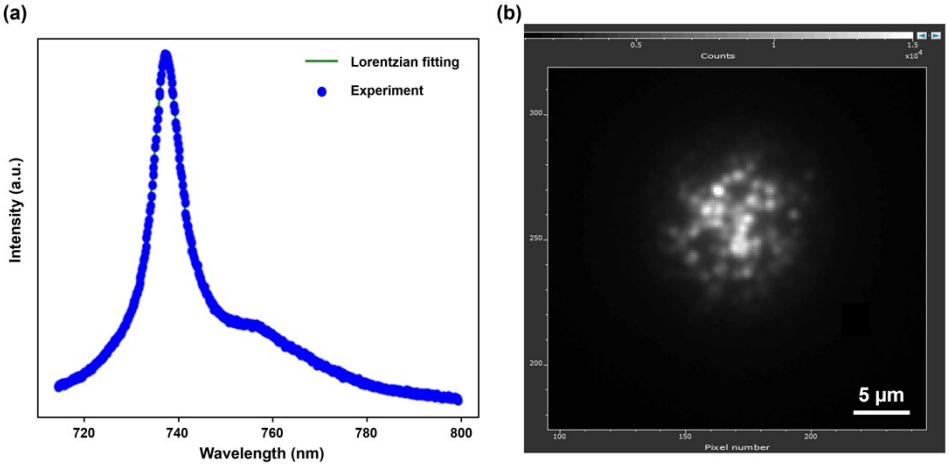

Fig. S6 (a) PL spectrum of the SiV centers in SAAO ND-grown diamond microparticles at 25 °C shown with relevant Lorentzian fitting. (b) EMCCD Image using 532 nm laser excitation source with a beam spot FWHM = 18 µm.

The PL spectrum was measured at each temperature for an integration time of 2.5 seconds, and the fitted parameters were recorded. Each parameter (ZPL position, ZPL FWHM, DWF) extracted by the Lorentzian data fitting of the PL spectrum could be used to perform temperature measurements independently. And these parameters were measured multiple times and averaged to improve the precision of the measurements. Most importantly, the measurement protocol was kept exactly the same for the SAAO ND- and DND-grown samples, so that a fair comparison could be performed. Fig. 3a–c of the main text shows the temperature dependence of each spectrum parameter. The standard deviation (σ) was calculated by repeating the above procedure 10 times and was added as error bars in the plots.

Hence, a separate noise floor (η) could be calculated independently for each spectrum parameter. We have chosen SiV ZPL position as the parameter to calculate the noise floor of our SAAO ND- and DND-grown samples, since this parameter has the lowest standard deviations, resulting in a lower noise floor. This is a common practice followed in the previously published paper as well.[2]

To elaborate on the calculation procedure, the standard deviation of the SiV ZPL position was measured for different integration times of the spectrometer. The measured

standard deviation was converted to temperature precision, by multiplying it with the thermal susceptibility calculated in Fig. 3a (0.0124 nm/°C). The obtained temperature precision *vs.* integration time curve was then fitted to the equation $\sigma = \eta/(t^{1/2})$,[3] corresponding to the shot-noise limit. And the noise floor (η) was extracted from the performed data fitting.

For the photostability measurement, photon counts were monitored for 1 hour in a region of interest of 60 pixels × 60 pixels (18 μm × 18 μm), using an exposure time of 20 ms for each image. The readings were then multiplied by 50 to scale up the measurements to counts per second (cps), followed by a spatial average to calculate the mean photon counts per second per pixel, which has been shown in Fig. 3f.

4. Temperature measurement using DND-grown diamond

To further demonstrate the superior properties of our SAAO ND-grown diamond, we conducted the temperature measurement using conventional DND-grown diamond and performed a direct comparison. The DND-grown sample shows higher inhomogeneous linewidths (~9.8–10 nm) with a significantly higher temperature noise floor (3.42 °C $Hz^{-1/2}$)

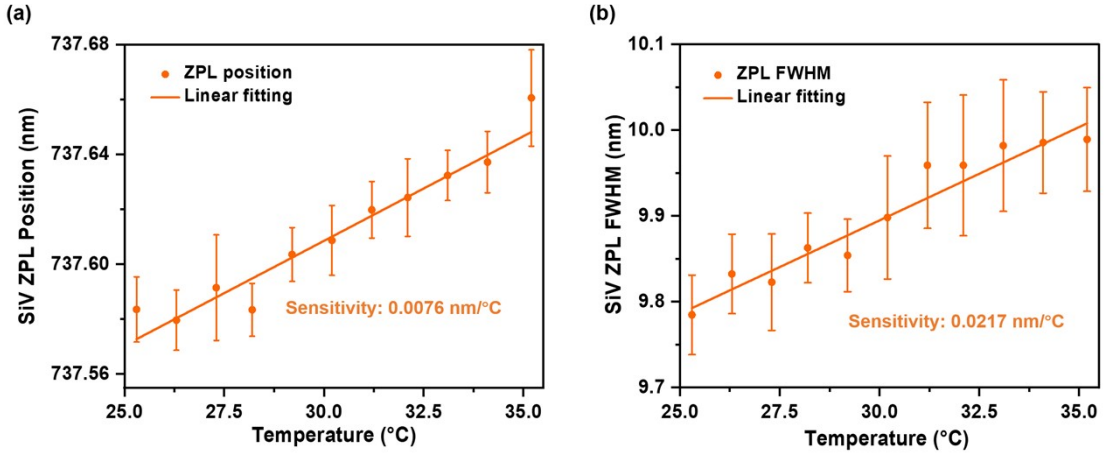

Fig. S7 Temperature measurement using the DND-grown diamond. Laser Power = 60 mW, Integration Time = 2.5 seconds (a) SiV ZPL Peak Position *vs.* Temperature. A sensitivity of 0.0076 nm/°C is extracted. (b) SiV ZPL FWHM *vs.* Temperature. A sensitivity of 0.0217 nm/°C is extracted.

References

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2. C. T. Nguyen, R. E. Evans, A. Sipahigil, M. K. Bhaskar, D. D. Sukachev, V. N. Agafonov, V. A. Davydov, L. F. Kulikova, F. Jelezko and M. D. Lukin, *Appl. Phys. Lett.*, 2018, **112**, 203102.
3. M. Fujiwara and Y. Shikano, *Nanotechnology*, 2021, **32**, 482002.