Decay rate enhancement of diamond NV-centers on diamond thin films: supplement

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Decay rate enhancement of diamond NV-centers on diamond thin films: supplemental document

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S1. Cathodoluminescence (CL) experimental setup

Our time-resolved cathodoluminescence system is based on a scanning electron microscope (CamScan CS3000) fitted with a LaB6 electron source, operated at 10 kV in fixed-spot mode in our measurements. The pulsed electron beam was created by a beam blanker driven by a waveform generator (ROHDE & SCHWARZ AM300 dual-channel Arbitrary / Function Generator). Wavelength and time resolved measurements of emitted photons were performed by a Horiba iHR320 spectrometer and a Horiba TBX picosecond photon detection module, respectively. A Horiba FluoroHub single photon counting controller provided the required synchronization for time-correlated single-photon counting. In our experiments, NV centres are excited by a pulsed e-beam. The electron pulse shape and the corresponding CL signal are shown in Fig. S1. The pulse has a duration of $t_D = 1.5$ µs. The pulse repetition rate is 500 kHz, which corresponds to 2 µs. Therefore, the switch-off time of the e-beam is $t_{off} = 500$ ns, which is sufficiently long to measure the NV centre lifetime.

![Fig. S1. Electron pulse shape (a) and corresponding transient CL signal from NV centres (b).](image)

S2. Dependence of NV lifetimes on ND cluster size and shape

The correlation between the recorded lifetimes and ND cluster geometry are examined in Fig. S2 for diamond nanoparticles placed on a diamond film or a bare Si substrate. The decay rates are presented as function of size (Fig. S2(a)) and shape (Fig. S2(b)). In both cases, the decay rate is nearly independent of both ND cluster size and cluster aspect ratio. Finally, we
NOTE THAT THE NV CENTER EMISSION SPECTRA EXHIBIT A MORE COMPLEX DEPENDENCE ON ND CLUSTER SIZE, IN PARTICULAR WITH RESPECT TO EMISSION LINEWIDTH. AN IN-DEPTH STUDY OF THESE EFFECTS LIES BEYOND THE SCOPE OF THE CURRENT WORK AND IS RESERVED FOR FUTURE STUDY.

Fig. S2. Experimentally measured NV center lifetime vs cluster size (a) and aspect ratio (b) for diamond nanoparticles deposited on a diamond thin film (top) or a silicon substrate (bottom). The correlation between lifetime and cluster size and aspect ratio is quantified by the slope and RMSE as obtained by a least-squares fit. The cluster size and aspect ratio are calculated as \((a+b)/2\) and \(a/b\), respectively, where \(a\) and \(b\) is the length of the cluster longer (shorter) (see inset to Fig. 1(c) in main text).

S3. Simulation of emission from a nanodiamond with NV center

Numerical simulations were performed by a commercial software (COMSOL 5.3a) based on the finite-element method. We set the refractive index of diamond and Si at the wavelength of 575 nm as \(n_{\text{diamond}} = 2.40\) [1] and \(n_{\text{Si}} = 4.00 + 0.03i\) [2], respectively. The simulation domain represents a cylinder with the radius of 1500 nm and height 3000 nm, terminated by scattering boundaries on all sides (see Fig. S3). The NV\(^0\) center excitation at \(\lambda = 575\) nm was introduced via a volume polarization density oscillating inside a 10 nm sphere. The total radiated power, \(P_{\text{tot}}\), is calculated as the integral of power flow over the surface of a 12 nm large sphere encapsulating the emitting center. The nanodiamond was modeled as a dielectric sphere with radius of 60 nm and with the NV\(^0\) center placed at its center. The lifetime of NV\(^0\) center on a substrate was given by \(\tau = \frac{\tau_{\text{bulk}}}{P_{\text{tot}}/P_{\text{bulk}}}\), where \(P_{\text{bulk}}\) is the power emitted by the modelled NV\(^0\) center in bulk diamond and \(\tau_{\text{bulk}}\) corresponding to a known bulk lifetime \(\tau_{\text{bulk}} = 19\) ns [3]. The total decay rate was calculated as \(\gamma_{\text{tot}} = \frac{1}{\tau} = \frac{P_{\text{tot}}}{\tau_{\text{bulk}}P_{\text{bulk}}}\). The radiative decay rate was obtained by \(\gamma_{\text{rad}} = \frac{P_{\text{air}}}{\tau_{\text{bulk}}P_{\text{bulk}}}\), where \(P_{\text{air}}\) is the power radiated to the far-field. The nonradiative decay rate was calculated as \(\gamma_{\text{non}} = \gamma_{\text{tot}} - \gamma_{\text{rad}}\). A summary of the simulated lifetimes and rates for different dipole orientations is shown in Table S1.
Fig. S3. Schematic illustration of the model used in lifetime calculations for a single ND on (a) Si and (b) diamond film on Si. The NV<sup>0</sup> center is introduced as a volume polarization density with the orientation along the x-axis (in the plane of the substrate) or z-axis (normal to the substrate) oscillating inside a sphere of radius \( r_0 = 5 \) nm.

Table S1. Summary of simulated lifetimes and rates of vertical and horizontal dipole emission and photon loss.

| Substrates | \( \tau_{\text{sim}} \) (ns) | \( \gamma_{\text{tot}} \times 10^6 \mu s^{-1} \) | \( \gamma_{\text{rad}} \times 10^6 \mu s^{-1} \) | \( \gamma_{\text{dual}} \times 10^6 \mu s^{-1} \) |
|------------|---------------------|---------------------|---------------------|---------------------|
| Si         | 130.55              | 7.66                | 5.30                | 2.36                |
| Diamond    | 105.15              | 9.51                | 7.47                | 2.04                |
| Si         | 38.21               | 26.17               | 17.04               | 9.12                |
| Diamond    | 49.31               | 20.28               | 16.33               | 3.95                |

S4. Collection efficiency of the parabolic mirror

The collection efficiency of the parabolic mirror in our TR-CL system depends on the orientation of the electric dipole of an NV<sup>0</sup> center, as shown in Fig. S4. In our experiments, the nanodiamonds are at the focus of the parabolic mirror. We define the quantities \( \eta_x, \eta_y, \eta_z \), where \( P_x, P_y, P_z \) are the radiated powers collected by the parabolic mirror for dipoles moments of NV<sup>0</sup> centers orientated along x, y, z directions, respectively. The average lifetime can then be calculated as:

\[
\tau_{\text{sim}} = \frac{1}{\tau_{\text{sim}}} = \frac{1}{(\eta_x^2 + \eta_y^2)\gamma_{\text{tot}}^2 + \eta_z^2 \gamma_{\text{tot}}^2}
\]  

(S2)
Fig. S4. (a) Schematic of the parabolic mirror used for collecting photons emitted by NV centers in our experiments. (b) Diagram of photon collection. The angle $\theta$ is $\sim 135^\circ$ according to the dimensions in (a).

| Substrates | $\eta_\parallel^1$ | $\eta_\parallel^2$ | $\eta_\perp$ |
|------------|--------------------|--------------------|--------------|
| Si         | 64.04%             | 18.29%             | 17.67%       |
| Diamond    | 48.73%             | 25.96%             | 25.31%       |

Table S2. Collection efficiencies of parabolic mirror for a dipole with x, y z-orientation

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