

# Journal Name

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### Supplemental Information: Photon bunching in cathodoluminescence induced by indirect electron excitation

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The short length scales associated with aloof excitations in diamond, together with the spatial resolution of the SEM and vibrations in the lab limited our ability to generate aloof  $g^2(\tau)$  maps for suspended nanodiamonds, but a prototypical bunching curve for a single nanodiamond with the electron beam held 50 nm from the nanodiamond over a hole in the TEM membrane is shown in Fig. S1. with the measured bunching for direct excitation of the same nanodiamond included for reference.

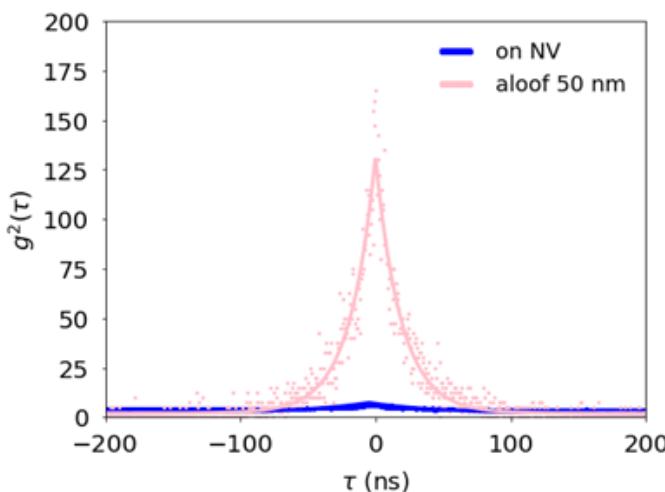


Fig. S1.  $g^2(\tau)$  for purely aloof excitation of nanodiamonds dropcast on holey SiN. The electron beam was positioned 50 nm away from the nanodiamond. A 5 kV, 7 pA beam was used. The fitted lifetime is 37 ± 1 ns.

CL  $g^2(\tau)$  maps for nanodiamonds located near the Al/SiO<sub>2</sub> interface are shown in Fig. S2.. Reduced secondary electron scattering is seen across that boundary, resulting in increased bunching approaching 10<sup>4</sup> far from the nanodiamond. As in the manuscript, at pixels where the bunching could not be extracted with sufficient statistical confidence, the bunching parameters were excluded from the plot.

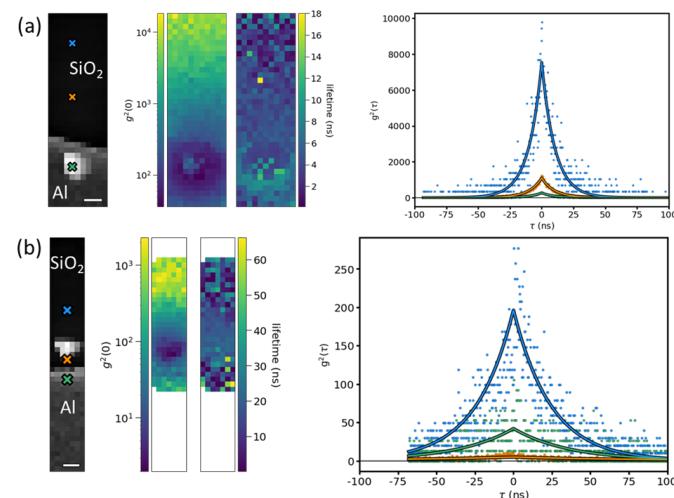


Fig. S2.  $g^2(\tau)$  CL maps for nanodiamond (bright white region) on (a) the edge of an aluminum film on an SiO<sub>2</sub> substrate and (b) on an SiO<sub>2</sub> substrate near the edge of an aluminum film. The left panels show the integrated CL counts at each pixel. The middle panels show the values of  $g^2(0)$  and bunching lifetime extracted from each pixel. The panel on the right shows  $g^2(\tau)$  at locations marked with a 'x' in the left panels. A 5 kV, 7 pA beam was used with 30 nm pixel resolution and 256 ps time bins.

The CASINO simulation of the number of secondary electrons that reach the nanodiamond as a function of incident electron beam distance for SiO<sub>2</sub> and Al substrates is shown in Fig. S3.. A greater number of secondary electrons reach the nanodiamonds on the SiO<sub>2</sub> at a constant distance, consistent with a higher effective electron beam current at the nanodiamond and thus with reduced observed CL bunching on the SiO<sub>2</sub> substrate.

The CASINO simulation of the number of secondary electrons that reach the nanodiamond as a function of incident electron beam energy for the Al substrate is shown in Fig. S4.. Critically, we see that, as the electron beam energy increases, the secondary electrons penetrate deeper into the substrate and a smaller number of secondary electrons escape the substrate at a fixed distance from the incident electron beam (resulting in a smaller effective current at the nanodiamond). This is consistent with the observed increase in bunching with increasing beam energy in Figure 4 of the manuscript.

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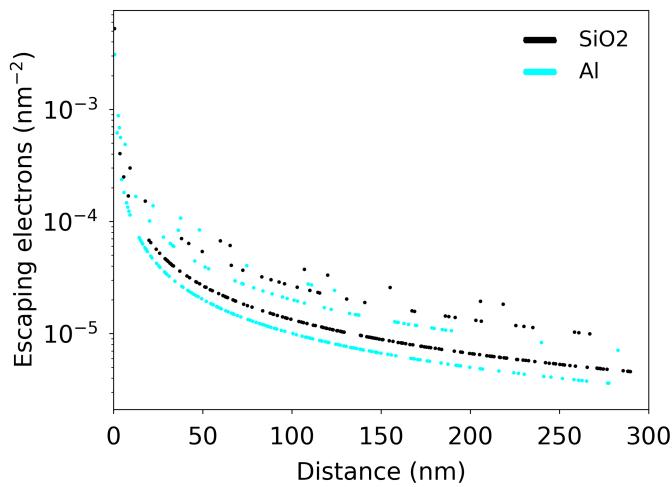


Fig. S3. CASINO Monte Carlo model for the number of secondary electrons that reach the nanodiamond as a function of incident electron beam distance for  $\text{SiO}_2$  and Al substrates.

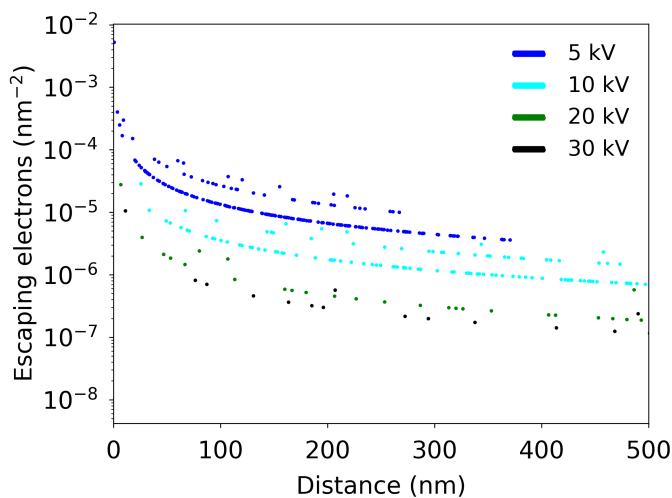


Fig. S4. CASINO Monte Carlo model for the number of secondary electrons that reach the nanodiamond as a function of incident electron beam energy on the Al substrate.