

Electronic Supplementary Information for:

Magnetic field-induced enhancement of the nitrogen-vacancy fluorescence quantum yield

M. Capelli, P. Reineck, D. W. M. Lau, A. Orth, J. Jeske, M. W. Doherty, T. Ohshima, A.D. Greentree and B.C. Gibson

Focal spot size

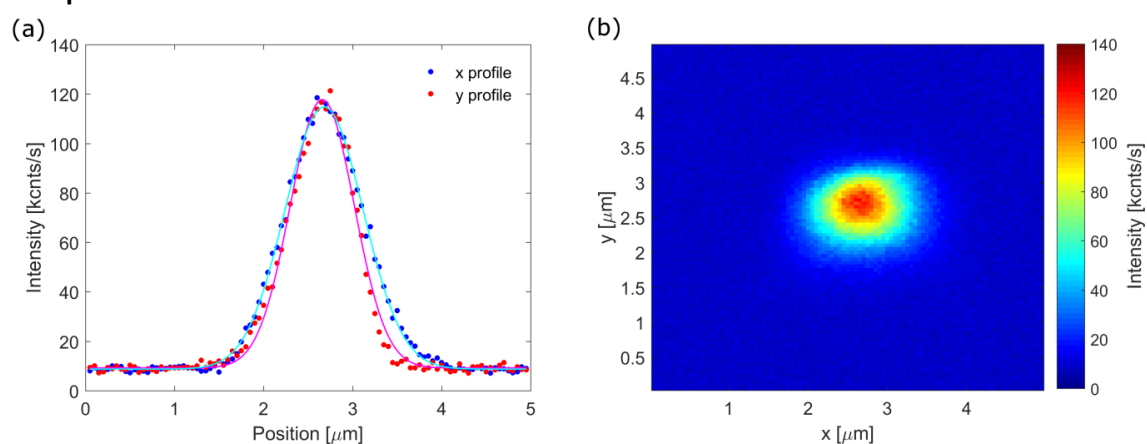


Fig. S1 Fluorescent emission (a) profiles and (b) map for a 120 nm nanodiamond imaged with a 0.1 NA objective. The profiles are fitted with Gaussian function $A \cdot \exp\left[-\frac{(x-x_0)^2}{\sigma_x^2}\right]$ where σ_x represents the semi-axis along x (the same applies for y). The focal spot area has been calculated as $\pi\sigma_x\sigma_y$, and its error propagated from the standard deviation of the parameters obtained from the fitting algorithm.

Experimental method and analysis

Each data point in Fig. 2 represents the averaged value of a recorded fluorescence time-trace. Each time-trace lasted 60 s with an acquisition frequency of 20 samples/s. At the same time, the laser beam intensity was recorded with power meter (Thorlabs PM100D) software. We took into account the average laser intensity to correct small variations on excitation intensity during the experiment. Each data point was then normalized to the value recorded at 0 mT.

High excitation power corrections to the NV centre emission

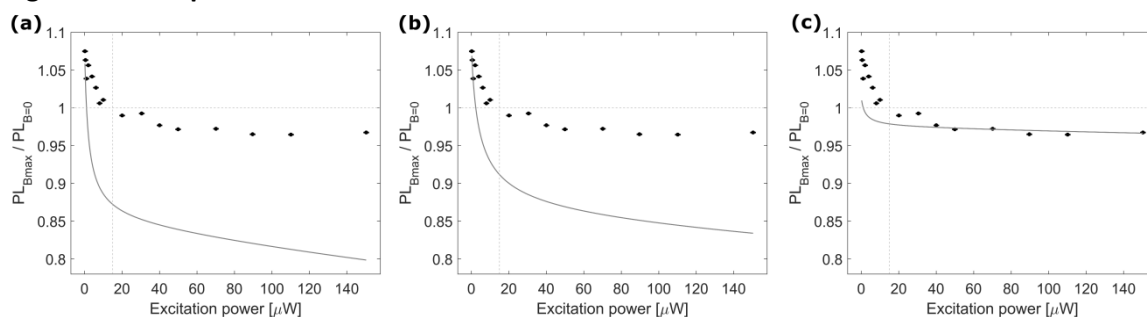


Fig. S2 (a) Ratio of the PL intensity at $B_{\text{max}}/B=0$ mT as a function of excitation intensity as reported in Fig. 2b. We modelled different correction to try to reduce the discrepancy. (b) Fit of the data considering Gaussian-distributed excitation intensity. In the initial fit, the excitation rate used was constant over the entire focal spot. In this fit, a Monte Carlo simulation has been used to reproduce the difference in excitation intensity for multiple NV centres scattered at random distances from the centre of the focal spot. Applying such correction, the model better represents the experimental data, but the discrepancy at high excitation power is not completely solved. (c) Fit of the data considering a constant, magnetic

field independent background fluorescence. Considering another emitter E_0 distributed with a density n_{E_0} over the sample and with a brightness b -times the brightness of an NV centre, the correcting factor was proportional to $\frac{n_{E_0}}{n_{NV}} \cdot b$. Applying such correction, the model better represents the experimental data at high excitation power but at the cost of creating a discrepancy at low excitation power. Moreover, to obtain the result shown in (c) a correction factor of 5 needs to be used, which we consider unlikely in our experiment for both the emitter density (n_{E_0}) and brightness (b).

Spectral analysis

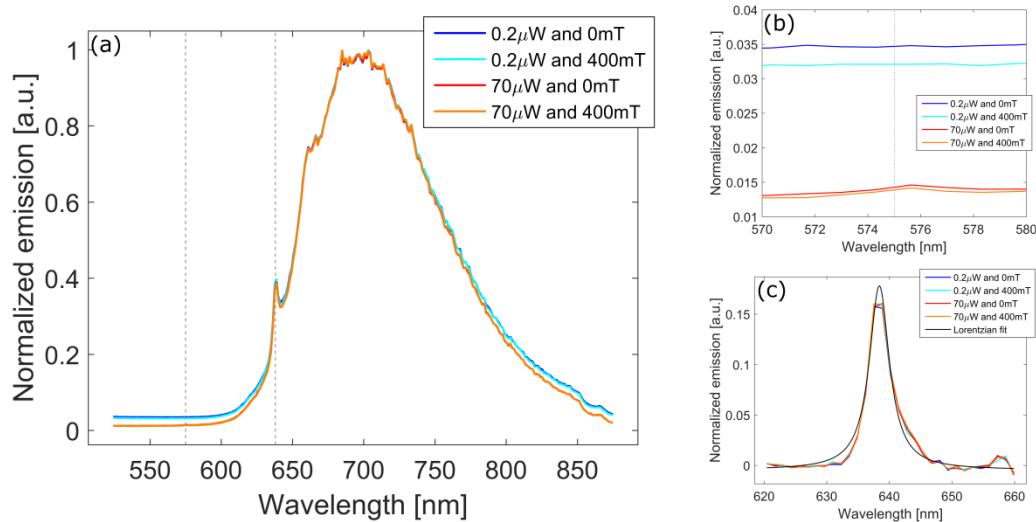


Fig. S3 (a) Spectra of ensembles of NV centres in HPHT single crystal diamond. The typical spectrum of the negatively charged NV centre (NV^-) is shown, with the characteristic zero phonon line (ZPL) at 638 nm (right dashed line) and a broad phonon sideband emission with a maximum around 700 nm. Our sample does not show a measurable presence of the less fluorescent neutrally charged NV centre (NV^0), whose ZPL should appear at 575 nm (left dashed line),¹ as shown in the magnified spectra in (b). The four spectra shown correspond to the four combinations of maximum and minimum magnetic field and excitation power used in our experiments, filtered only by a notch filter around 532 nm. At a constant excitation power, the spectrum does not change with the magnetic field. The slight offset of the spectra at the different excitation power is caused by a greater integration time used to take the measurement at 0.2 μ W which increased the background collected. (c) Magnification of the NV^- ZPL (638 nm) for all four combinations investigated alongside the best fit of a single Lorentzian curve. The ZPL position does not show the blue-shift associated with the presence of strain,² with an average value of 638.388 ± 0.016 nm in all conditions. Additionally, we found an average width of 2.74 ± 0.04 THz for the ZPL peak which implies that any strain-induced splitting³ would be smaller than the thermal energy at room temperature (6.5 THz). Since the strain-induced splitting is not visible, we can conclude that the orbital splitting of the triplet excited state caused by strain on the NV centre is averaged out at the temperature used in our experiment.⁴ This conclusion demonstrates a negligible strain in our experiment which does not impact the intersystem crossing and thus the fluorescent emission. Additionally, it justifies our use of a three-level orbit-free spin Hamiltonian model for the excited state model.

Spin lattice relaxation time

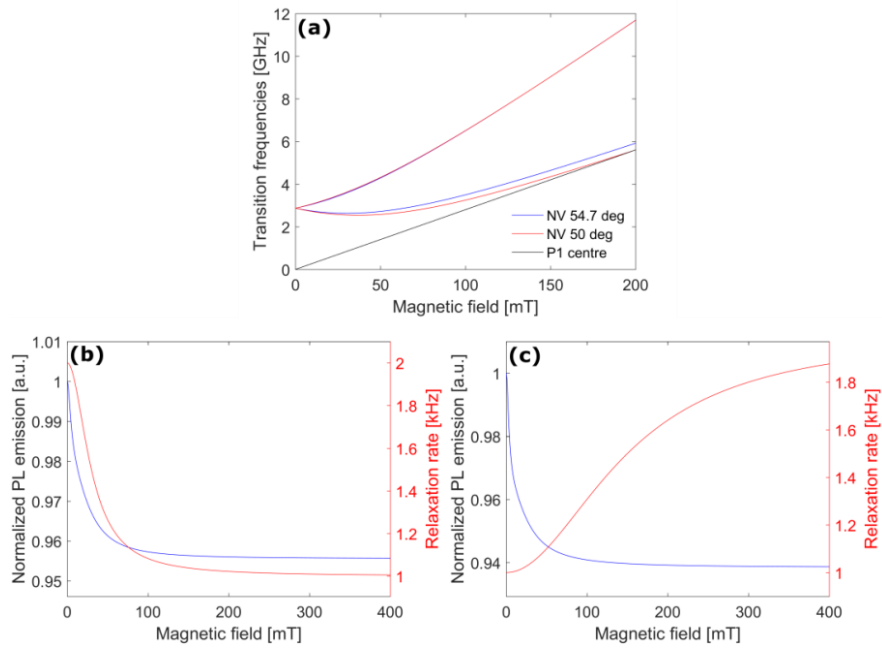


Fig. S4 (a) Transition frequencies as a function of magnetic field for NV centres misaligned by 54.7 degrees with the magnetic field vector (blue curve), NV centres misaligned by 50 degrees (red curve), P1 centres (electron spin of a single nitrogen atom, black curve). (b, c) NV emission intensity as a function of external magnetic field with constant triplet-to-singlet rate ($k_{es} = 0.04 \text{ ns}^{-1}$) and changing spin relaxation rate (k_{T_1}) as a function of magnetic field. In both simulations, a low excitation rate ($\lambda = 5.4 \text{ kHz}$) is used. The simulation (b) shows the effect of a decreasing relaxation rate with increasing magnetic field. The relaxation rate decreases when an external magnetic field is applied to an ensemble of NV centre with different orientations because it removes the resonant transition at $B=0 \text{ mT}$ (a difference of 5 degree is enough to reduce the initial relaxation rate by half).⁵ The simulation (c) shows the effect of an increasing relaxation rate with increasing magnetic field. The relaxation rate increases when the external magnetic field splits the electron spin energy levels of the surrounding nitrogen atoms (P1 centre), allowing a resonant transition between the NV centre and the P1 centre to occur (this resonance would happen at a specific magnetic field value if the NV centre were aligned to the magnetic field).⁶ Neither of the two changes in the spin relaxation rate have significant effect on the NV emission, even at low excitation rate.

Rate functions comparison

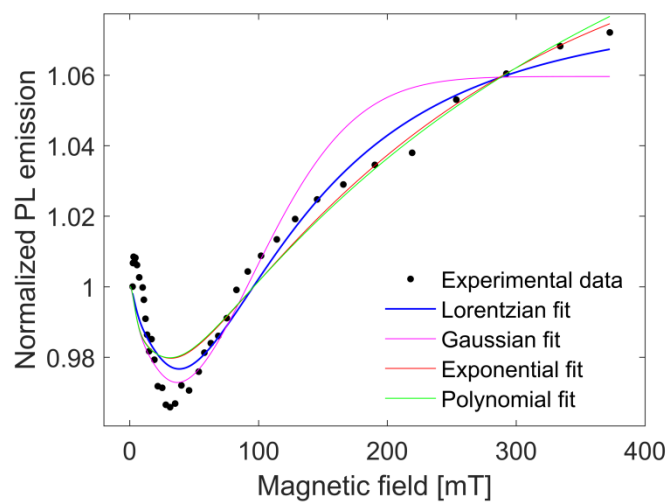


Fig. S5 Black dots represent the same photoluminescence (PL) emission at $0.2 \mu\text{W}$ reported in Fig. 2a. The 4 curves show the fit of the model to the data, each of them using a different decaying function for the transition rate change with

increasing magnetic field. Using the normalized residuals as a criterion, the Lorentzian fit emerges as the best representation of our data.

References

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