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Tunable cavity coupling of the zero phonon line of a nitrogen-vacancy defect in diamond

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Abstract

We demonstrate the tunable enhancement of the zero phonon line of a single nitrogen-vacancy colour centre in diamond at cryogenic temperature. An open cavity fabricated using focused ion beam milling provides mode volumes as small as \(1.24 \mu m^3\) (4.7 \(\lambda^3\)) and quality factor \(Q \approx 3000\). In situ tuning of the cavity resonance is achieved with piezoelectric actuators. At optimal coupling to a TEM\(_{00}\) cavity mode, the signal from individual zero phonon line transitions is enhanced by a factor of 6.25 and the overall emission rate of the NV\(^-\) centre is increased by 40\% compared with that measured from the same centre in the absence of cavity field confinement. This result represents a step forward in the realisation of efficient spin-photon interfaces and scalable quantum computing using optically addressable solid state spin qubits.

1. Introduction

Coupling of fluorescence from nanoscale quantum systems to optical microcavities provides a means to control the emission process and can be an essential element of nanophotonic device applications. The negatively charged nitrogen-vacancy (NV\(^-\)) defect in diamond is an example of a solid state system that has gained significant attention in recent years as a quantum spin register [1, 2] and nanoscale sensor [3–5] due to its long spin coherence times and capacity for optical manipulation and readout. The recent demonstration of quantum error correction in an NV\(^-\) defect [6] provides a sound basis for using these systems in practical quantum information technologies. A well established strategy for a coherent spin photon interfaces is to significantly reduce interaction with phonons and enhance coupling to the ‘zero phonon line’ (ZPL) under conditions of minimised pure dephasing. The low Debye–Waller factor (DW \(\approx 0.04\)) of the NV\(^-\) centre imposes a significant limitation in this regard, since most spontaneously emitted photons are accompanied by a phonon emission event which leaks information to the environment. The enhancement of the ZPL and its efficient coupling to external optics are therefore important challenges to which microcavities are well suited. In particular is an essential requirement for generating large scale entangled states between spatially separated defects connected via photonic networks, for which proof of principle experiments have been achieved [8] but further development is hampered by low entanglement efficiency. Success in this endeavor may provide a route to the realisation of scalable quantum computers based on optical networks of electronic and nuclear spins [9, 10].

Resonant coupling of the NV\(^-\) ZPL to micro-ring resonators [11] and photonic crystal cavities [12–15] has been demonstrated to provide effective enhancements, but the monolithic structure of these cavities prevents positioning of the emitter at the heart of the cavity mode in situ. Tuning of the cavity mode to the ZPL resonance can be achieved in these systems using either progressive gas condensation in a cryostat (red tuning) or repeated thermal etching steps (blue tuning) but these procedures are difficult to optimise and have limited scope for use in device applications. Open cavities [16–19] on the other hand provide a flexible approach to cavity-coupled
devices that permit full in situ alignment and tuning using piezoelectric actuators, thus providing the rapid and precise device alignment necessary for more advanced experiments. An additional advantage is the efficient coupling to external optics, which is achieved using Gaussian TEM\(_{00}\) modes that leak from the cavity through one of the mirrors. Here we demonstrate control over the emission from a single NV\(^{-}\) centre by coupling its ZPL to the resonant mode of an open microcavity. The open cavity geometry allows direct comparison between the emission properties of the same defect in and out of the cavity, thus providing unambiguous evidence of the effect of the cavity mode. We demonstrate tunable enhancement of the ZPL emission and reduction of the fluorescence lifetime of the defect in a controlled manner, and analyze our results in terms of the Purcell effect acting on the ZPL and the phonon sideband (PSB). Our work builds on recent demonstrations of room temperature coupling of NV\(^{-}\) defects to open cavities [20–22] in which lifetime changes were not observed due to the far greater ZPL line width.

2. Experimental method

The open microcavities are of a plano-concave design, the concave features produced by focused ion beam milling of a fused silica substrate [18, 23]. These cavities support a Hermite–Gauss mode structure with TEM\(_{00}\) modes that can be effectively mode-matched to an external Gaussian beam. The radius of curvature of the concave mirror used here is 7.6 \(\mu\)m. The concave and planar dielectric mirrors have reflectivities of >99.99% and 99.7% respectively at the design wavelength of 637 nm, and reflection bands extending from 550 nm to 720 nm. The planar mirror is terminated with a low index layer to provide a field anti-node at its surface. The shortest cavity length achieved here has a mirror spacing of 1.11 \(\mu\)m providing an additional 3 field intensity maxima of the TEM\(_{00}\) mode between the mirror surfaces with \(\lambda = 637\) nm, as shown in figure 1(d). We label this mode with the longitudinal index \(q = 4\), or the set of indices \((q, m, n) = (4, 0, 0)\). The mode volume is calculated numerically to be 1.24 \(\mu\)m\(^3\).

The NV\(^{-}\) defects are located in nanodiamonds which were fabricated by the high pressure high temperature detonation synthesis method, and were subject to an acid clean to remove graphitic material before being spin cast onto the planar mirror. Registration marks on the planar mirror created using a focused ion beam allow individual defects to be identified and characterised both in and out of the cavity. The two experimental geometries used in this report are displayed in figure 1. The ‘out-of-cavity’ fluorescence is measured in the absence of a concave mirror and with the planar mirror as a substrate behind the nanodiamonds (figure 1a). For the ‘in-cavity’ experiments the planar mirror is inverted and both excitation of the NV\(^{-}\) centres (at \(\lambda = 532\) nm) and collection of fluorescence is carried out through the planar mirror. The microcavity assembly comprises a set of piezoelectric actuators that provide full control of the cavity length and relative position of the planar and concave mirrors at cryogenic temperature (figure 1(b)). All measurements are carried out at 77 K in a dry He exchange gas environment with the container supported in a liquid nitrogen bath cryostat [24]. Further details of the experimental apparatus can be found in section A of the supplementary information.
Figure 2. Characterisation of the NV$^{-}$ centre on the planar mirror. (a) Fluorescence image showing point-like emission from single NV$^{-}$ centres, and registration lines generated by focused ion beam Ga$^{+}$ implantation that allow re-location of the same NV defect in the two experimental configurations. (b) Photon correlation data from the NV$^{-}$ centre measured, (c) and (d) fluorescence spectra from the NV$^{-}$ defect at $T = 77$ K, and (e) polarisation plots of peak 2 (blue) and peak 3 (purple) of the zero phonon line doublet.

Figure 3. In-situ tuning of a cavity mode through the ZPL resonance. (a) Selected spectra (blue) recorded at five different cavity positions, with a simulation of each spectrum calculated using equation 5 (red), (b) 2D false colour-scale plot of the measured spectrum including all of the tuning steps recorded.
3. Results

A low temperature fluorescence image recorded in the out-of-cavity configuration reveals well-dispersed nanodiamonds (figure 2(a)) with some instances of single centres recorded by the Hanbury-Brown and Twiss method. The nanodiamond used in this study is circled, and its uncorrected photon intensity correlation data shown in figure 2(b), revealing $g^{(2)}(0) = 0.38$. Subtraction of background due to autofluorescence from the mirrors reduces this value to $g^{(2)}(0) = 0.07$ implying that $\approx 97\%$ of the fluorescence is from a single emitter.
Fluorescence spectroscopy of this NV⁻ centre reveals a ZPL spectrum that is dominated by a linearly polarised doublet (fitted peaks 2 and 3 in figure 2(d)) indicating a high level of strain that lifts the degeneracy of the orthogonal \(^3E_i, \text{and} \quad ^3E_f\) excited state dipoles of the defect (figure 2(c)). The line widths of the individual ZPL components are approximately 0.4 nm, with Gaussian line shapes suggesting that they are dominated by inhomogeneous broadening due to spectral diffusion. Weak additional lines are seen, potentially due to an additional NV centre in close proximity. The relative intensities of peaks 2 and 3 and the angle between their polarisations (figure 2(e)) indicate that the NV⁻ defect axis is oriented at 49° relative to the optical axis of the cavity, and the two orthogonal transition dipoles for lines 2 and 3 are at angles of 39° and 24° to the plane of the mirror respectively (see supplementary information section B). The ZPL measured has \(\Gamma_{\text{ZPL}} = 0.044\), as is typical for NV⁻ centres.

Figure 3 shows the measured fluorescence spectrum for the in-cavity configuration as a TEM\(_{00}\) cavity mode is tuned through resonance with the ZPL. Clear enhancement of the ZPL emission is observed, providing a fully saturated photon count rate of 15 kc s\(^{-1}\). This represents an experimental enhancement of the total ZPL signal by a factor of 2.5 compared with that recorded from the defect with the planar mirror alone (see supplementary information section C). Since this intensity primarily corresponds to resonant tuning to peak 3 in the recorded spectrum, which contains about 40\% of the out-of-cavity ZPL photons, the enhancement factor of this peak is estimated as 6.25.

Figure 4 shows a side-by-side comparison of the optical properties of the NV⁻ centre out of the cavity, with that observed in the cavity at optimal tuning to the ZPL. Figure 4(a) shows the spectra over the full range of NV-emission, illustrating the extent to which the coupled ZPL dominates the measured fluorescence, a result of the cavity having no other modes in this range that couple efficiently into the objective lens. Figure 4(b) shows a comparison between the fluorescence decay characteristics. The lifetime of the out-of-cavity defect is measured as \((30.8 \pm 0.6)\) ns while that in the cavity is \((22.1 \pm 0.4)\) ns, corresponding to a \((39.5 \pm 0.7)\) % increase in the emission rate. The slight deviation from a single exponential decay in the in-cavity data at longer delay time is suspected to be due to spectral instability of the cavity mode leading to inhomogeneous broadening. Figure 4(c) shows the photon autocorrelation data, which reveal a reduced \(g^{(2)}(0)\) correlation value of 0.28 suggesting a slight improvement in the isolation of the single emitter when only the cavity-coupled ZPL is measured.

**4. Discussion**

In this section we develop a simple analysis of the measured data. The relative magnitudes of the cavity leakage rate, \(\kappa \sim 4\) THz, NV⁻ centre out-of-cavity decay rate, \(\gamma \sim 30\) MHz, and NV-cavity coupling rate \((g \sim 6\) GHz\) place these experiments firmly in the weak coupling regime of cavity quantum electrodynamics whereby classical treatment of the modified local optical density of states provides an adequate description of the cavity-induced changes to the emission process. While the spectrally narrow ZPL couples almost exclusively to a single TEM\(_{00}\) mode of the cavity, the much broader PSB couples to several other resonant and leaky modes that require numerical simulation. We therefore choose a simple Purcell model that can be adapted to the complexity of the cavity mode structure rather than adopt a full quantum treatment.

A complication in the theoretical modelling of these results is the presence of inhomogeneous broadening, both in the cavity line width and in the ZPL transition lines. The cavity resonance is subject to mechanical vibrations in the cavity assembly that modulate the cavity length on time scales of order milliseconds, while the ZPL emission line is affected by local electric field fluctuations experienced by the NV⁻ centre due to residual charge movement on time scales from nanoseconds to seconds. Within the total measured line widths of 0.7 and 0.4 nm for the cavity mode and ZPL transition respectively, the relative magnitudes of homogeneous and inhomogeneous broadening are difficult to quantify. Nominally identical cavities have been measured to provide homogeneous line widths of 0.2 nm however, and the minimum homogeneous line width of an NV⁻ centre in bulk diamond at 77 K is known to be \(\sim 0.1\) nm [27]. We therefore estimate that inhomogeneous effects account for a maximum of 0.5 nm of additional broadening in the cavity mode and a maximum 0.3 nm in the ZPL. The PSB emission has a homogeneous line width of 70 nm so inhomogeneous effects can be neglected.

The total emission rate of the NV⁻ centre can be expressed as the sum of the emission rates into the ZPL and PSB, plus a non-radiative relaxation rate \(\gamma_{\text{nr}} = \gamma_{\text{ZPL}} + \gamma_{\text{PSB}} + \gamma_{\text{nr}}\), so we are able to treat the effect of the cavity on the ZPL and PSB of the emission separately. Within this model we can define the Debye–Waller factor, \(D_W\), as the fraction of the radiative emission into the zero phonon line \(D_W = \frac{\gamma_{\text{ZPL}}}{\gamma_{\text{ZPL}} + \gamma_{\text{PSB}}}\), and the quantum efficiency of the NV⁻ centre as the ratio of the radiative rate to the total relaxation rate \(\eta = \frac{\gamma_{\text{ZPL}} + \gamma_{\text{PSB}}}{\gamma_{\text{nr}}}\). We measure \(D_W = 0.044\) from the out-of-cavity fluorescence spectrum in figure 2(c). (Note that it is not possible to establish a modified \(D_W\) for the in-cavity case directly from the measured spectra because the different cavity modes couple in to the collection optics with very different efficiency. That the TEM\(_{00}\) mode couples far more efficiently into the collection optics than the other modes is evidenced by the measured in-cavity fluorescence shown in
figure 4a, in which the PSB is barely visible relative to the ZPL.) The ZPL and PSB relaxation rates will be modified by the presence of the cavity—we define these rate modifications as effective Purcell factors $F'_{P1}$ and $F'_{P2}$ respectively such that we can write the total cavity-coupled emission rate as $\gamma_{cav} = F'_{P1} \gamma_{ZPL} + F'_{P2} \gamma_{PSB} + \gamma_{nr}$.

This can in turn be expressed as a change in the overall emission rate

$$F'_P = \frac{\gamma_{cav}}{\gamma_{tot}} = 1 + \eta \left( F_{ZPL} + F_{PSB} \right),$$

where $F_{ZPL} = (F'_{P1} - 1)$. DW and $F_{PSB} = (F'_{P2} - 1)$. (1 − DW) are the contributions from the ZPL and PSB to the overall change in the radiative decay rate.

We separate the analysis of our results into two parts—a semi-analytic treatment of the ZPL emission interacting with a single TEM$_{00}$ mode to estimate $F_{ZPL}$ and a numerical treatment to estimate $F_{PSB}$. For the ZPL coupling we begin by expressing the wavelength dependent enhancement of the emission intensity resulting from a dipole $\mu$ coupling to a single cavity mode centred at wavelength $\lambda_{cav}$ as:

$$F_\mu(\lambda) = \frac{1}{1 + 4Q^2 \left( \lambda / \lambda_{cav} - 1 \right)^2},$$

Here $F^\text{max} = \frac{3}{4\pi^2} \left( \frac{\lambda_{cav}}{n} \right)^3 \frac{Q}{V_{\text{mode}}}$ is the maximum rate enhancement assuming perfect spatial alignment and orientation, $\xi_\mu = \left( \frac{|\mu \cdot E|}{|\mu| |E|_{\text{max}}} \right)$ is the spatial overlap and orientation factor between the dipole and the cavity electric field $E$ and $Q$ is the cavity quality factor. As we are able to position the emitter at the electric field maximum we assume that $E = E_{\text{max}}$ so that $\xi_\mu = \cos^2(\theta)$, where $\theta$ is the angle between the dipole and the plane of the mirror. For peaks 2 and 3, $\theta = 39^\circ$ and $\theta = 24^\circ$ respectively giving $\xi_2 = 0.60$ and $\xi_3 = 0.83$.

We begin by simulating the cavity coupled spectra shown in figure 3(a). Since the Purcell enhancement is linear and the spectra represent the time average of the inhomogeneous emitter–cavity coupling, only the total line widths are important, and we can calculate the enhancement in the intensity of the ZPL by simply integrating equation 2 over the out-of-cavity emission spectra of the dipoles collected over all directions, which we label $S_\mu$:

$$F_{ZPL} = \sum_\mu n_\mu \int d\lambda S_\mu(\lambda) F_\mu(\lambda).$$

Here $n_\mu$ are the branching factors of the two dipoles in the excited state: $n_\mu = 0.44$ and 0.56 for peaks 2 and 3 of the ZPL doublet respectively (see supplementary information section B). We can rewrite equation 3 as:

$$F_{ZPL} = F^\text{max} \sum_\mu n_\mu \xi_\mu \int d\lambda \frac{1}{1 + 4Q^2 \left( \lambda / \lambda_{cav} - 1 \right)^2} S_{\text{axial}}(\lambda),$$

where

$$S_{\text{axial}}(\lambda) = \frac{\sum_\mu n_\mu \xi_\mu S_\mu(\lambda)}{\sum_\mu n_\mu \xi_\mu}$$

is the spectrum emitted along the cavity axis, to which the spectrum shown in figure 2(d) is a good approximation when scaled to normalise the total measured intensity. The spectrum emitted into the cavity mode now reads

$$S_{\text{cav}}(\lambda) \propto \frac{S_{\text{axial}}(\lambda)}{1 + 4Q^2 \left( \lambda / \lambda_{cav} - 1 \right)^2}.$$

Using the measured time-averaged cavity line width $\delta \lambda_{\text{cav}} = 0.7$ nm to evaluate $Q$ in equation 6 generates the result shown in figure 3(a), reproducing well the measured spectra. The same value of $\delta \lambda_{\text{cav}}$ used in equation 5 gives $F_{ZPL} = 0.25$ corresponding to $F_{P1} = 6.7$. Based on the the branching factor for peak 3 of 0.56, the cavity-induced enhancement of emission from peak 3 is therefore found to be about a factor of 12.

It is instructive to compare this calculated ZPL intensity enhancement with a simple analytic treatment of the coupling to peak 3 alone. In a standard treatment of the Purcell enhancement applied to a broad emitter [28], the cavity $Q$ factor in equation 2 is simply replaced by an effective $Q$ factor: $Q_{\text{eff}} = \lambda / (\delta \lambda_{\text{cav}} + \delta \lambda_{\text{em}})$, where $\delta \lambda_{\text{em}}$ is the emission line width. Using $\delta \lambda_{\text{em}} = 0.4$ nm and $\delta \lambda_{\text{cav}} = 0.7$ nm gives a value of 9.2, in reasonable agreement with the result above. These values also compare reasonably well with the experimentally measured increase in intensity of peak 3 of 6.25.

Modeling of the measured change in the emission rate requires a different approach, as the inhomogeneous broadening does not lead to a simple averaging as described above. Temporal modulation of the detuning will produce fluctuations in the cavity-coupled fluorescence decay rate so that the time-averaged, measured decay
will contain a range of lifetime components. We note that the cavity-coupled decay data shown in figure 4(b) are suggestive of this effect: they are not fitted as well by a single exponential decay curve as are the out-of-cavity data. The fastest rate that appears in the decay data immediately after the excitation pulse thus corresponds more closely to the resonantly tuned, homogeneously broadened case than a simple average of the decay rates would indicate.

We include this effect in the analytical calculations by convoluting the mode line width with a Gaussian function $g(\lambda_{\text{cav}})$ to represent the distribution of cavity mode positions over the measurement integration time. The fractional change in the measured decay rate immediately after the excitation pulse as a result of ZPL coupling is then given by (supplementary information section E):\begin{equation}
F_{\text{ZPL, inhom}} = \frac{\int d\lambda_{\text{cav}} \ g(\lambda_{\text{cav}}) \left(1 + F_{\text{ZPL}}(\lambda_{\text{cav}})\right) F_{\text{ZPL}}(\lambda_{\text{cav}})}{\int d\lambda_{\text{cav}} \ g(\lambda_{\text{cav}}) F_{\text{ZPL}}(\lambda_{\text{cav}})}.
\end{equation}

Assuming the maximum degree of inhomogeneous broadening of the cavity mode i.e., using a cavity line width of $\delta\lambda_{\text{cav}} = 0.2$ nm to establish $F_{\text{ZPL}}$ and a Gaussian distribution of width 0.5 nm, gives $F_{\text{ZPL, inhom}} = 0.36$.

It is not possible to construct a model of the modified emission that includes inhomogeneous broadening of the emitter using the measured spectrum $S_{\text{axial}}$, and so the most straightforward way to estimate $F_{\text{ZPL}}$ assuming maximum inhomogeneous broadening of both the cavity and the emitter is to apply equation (2) with the effective $Q$ factor as described above (using $\delta\lambda_{\text{cav}} = 0.2$ nm and $\delta\lambda_{\text{em}} = 0.1$ nm gives $Q_{\text{eff}} = 2100$). Using the appropriate values of $\xi_3$ and $n_3$ generates a value of $F_{\text{ZPL}} = 0.69$.

We now turn to calculation of $F_{\text{P2}}$, for which we use finite difference time domain (FDTD) simulations that allow us to input the full cavity structure to accurately represent the dielectric environment of the NV$^-$ centre (see supplementary information section D for details). These calculations allow confirmation of the cavity parameters by matching the simulated mode spectrum to that measured, and a direct prediction of the change in

Figure 5. Comparison between (a) the measured in-cavity spectrum with the (5, 0, 0) mode optimally coupled to the ZPL, (b) the FDTD simulation of the Purcell enhancement, and (c) the semi-empirical calculation of the emitted power density spectrum. Modes are identified by their $(q, m, n)$ for longitudinal index $q$ and transverse indices $m, n$. 


\section*{Supplementary information}
emission rate that will occur between the in-cavity and out-of-cavity experimental configurations. Figure 5 shows semi-logarithmic plots of three cavity-coupled spectra with the (5,0,0) mode tuned to the ZPL. This measured spectrum (figure 5(a)) is chosen as it reveals the positions of other modes coupling to the PSB and therefore provides a good reference by which to verify the geometrical parameters of the cavity. The relative Purcell factor (figure 5(b)) and the resultant semi-empirical prediction for the emission power density spectrum of the NV$^-\text{ centre}$ in the cavity (figure 5(c)) are also shown. The PSB emission couples to cavity modes with longitudinal index $q = 4$ and transverse indices 0, 2, and 4. The absence of observed coupling to modes with odd transverse indices suggests that the NV$^-\text{ centre}$ is well positioned on the cavity axis of symmetry where the electric field intensities of these modes drops to zero. Integrating the emitted power density spectrum between 640 and 740 nm reveals the Purcell enhancement of the full PSB emission to be $F_{P2} = 0.93$, corresponding to $F_{PSB} = 0.86$ This represents a 7% reduction of the PSB emission rate relative to that in the out-of-cavity geometry, as a result of the reduced optical density of states in the cavity within this spectral region.

We are now in a position to generate simulated values of the effective Purcell factor using equation (1). Assuming a unity quantum efficiency for the NV$^-\text{ centre}$ gives values of $F'_P = 1.18, 1.30, \text{ and } 1.62$ for the three different treatments of the ZPL described above (including no inhomogeneous broadening and using equation (4), including maximum inhomogeneous broadening of the cavity only and using equation (7), and including maximum inhomogeneous broadening of both cavity and ZPL and using equation (2) directly). Compared with the measured value of $F'_P = 1.40$ it is clear that inhomogeneous broadening of both cavity and ZPL must be considered. Reducing $\eta$ lowers the simulated values of $F'_P$, such that we can achieve good agreement with the measured data by assuming maximum inhomogeneous broadening in both cases and using $\eta \approx 0.62$.

In figure 6 we extend this comparison between simulated and measured enhancement factors by showing how key parameters vary as the cavity length is varied from $q = 4$ to $q = 7$, tuning successive TEM$_{00}$ modes into resonance with the ZPL. The PSB data points show that the increased suppression of emission into the sideband is not monotonic as the cavity shortens, a result of the tuning of the different cavity modes through the PSB spectral region. Figure 6(b) compares the the measured values $F'_P$ with the simulated values produced with the
different approaches to the ZPL described above. The simulation points that include maximum inhomogeneous broadening of both ZPL and cavity, and assume a quantum efficiency of 0.62, provide good agreement across the range of cavity lengths.

5. Conclusion

In summary we have shown the controlled coupling of the ZPL of a single NVcentre in nanodiamond to an open cavity at cryogenic temperatures, revealing a significant increase in ZPL emission and a 40% reduction in the measured lifetime. Modelling of these results suggests that inhomogeneous broadening is present, although it is difficult to quantify without independent measurements. This work is nevertheless informative in the pursuit of efficient coupling between NVcentres and optical networks. To conclude we comment briefly on the maximum coupling efficiency that may be anticipated using open cavities in such systems once all of the parameters are optimised. Open cavity Q factors exceeding $10^4$ have been demonstrated [19], and single NV defects implanted into bulk diamond at depths of 100 nm can offer ZPL line widths as narrow as 27 MHz at a temperature of 4.2 K [30], such that resonant coupling would result in enhancement of the emission rate into the ZPL by a factor of order $10^3$ and enhanced Debye–Waller factors of around 0.98. The use of NV defects embedded in membranes would also provide access to electron spin coherence times $>100$ μs that can be utilised as quantum registers. Such projections suggest that the experimental configuration demonstrated here is an attractive route towards efficient spin–photon interfaces that might be employed in the construction of scalable quantum processors.

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