

# CHCRUS

This is the accepted manuscript made available via CHORUS. The article has been published as:

## Cavity-Enhanced Raman Emission from a Single Color Center in a Solid

Shuo Sun, Jingyuan Linda Zhang, Kevin A. Fischer, Michael J. Burek, Constantin Dory, Konstantinos G. Lagoudakis, Yan-Kai Tzeng, Marina Radulaski, Yousif Kelaita, Amir Safavi-Naeini, Zhi-Xun Shen, Nicholas A. Melosh, Steven Chu, Marko Lončar, and Jelena Vučković Phys. Rev. Lett. **121**, 083601 — Published 21 August 2018 DOI: 10.1103/PhysRevLett.121.083601

### Cavity-enhanced Raman emission from a single color center in a solid

Shuo Sun<sup>1‡\*</sup>, Jingyuan Linda Zhang<sup>1‡</sup>, Kevin A. Fischer<sup>1‡</sup>, Michael J. Burek<sup>2</sup>, Constantin Dory<sup>1</sup>, Konstantinos G. Lagoudakis<sup>1</sup>, Yan-Kai Tzeng<sup>3</sup>, Marina Radulaski<sup>1</sup>, Yousif Kelaita<sup>1</sup>, Amir Safavi-Naeini<sup>1</sup>, Zhi-Xun Shen<sup>3,4,5</sup>, Nicholas A. Melosh<sup>4,5</sup>, Steven Chu<sup>3,6</sup>, Marko Loncar<sup>2</sup>, and Jelena Vucković<sup>1</sup>

<sup>1</sup>E. L. Ginzton Laboratory, Stanford University, Stanford, California 94305, USA
 <sup>2</sup>School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA
 <sup>3</sup>Department of Physics, Stanford University, Stanford, California 94305, USA
 <sup>4</sup>Geballe Laboratory for Advanced Materials, Stanford University, Stanford, California 94305, United States
 <sup>5</sup>Stanford Institute for Materials and Energy Sciences, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA
 <sup>6</sup>Department of Molecular and Cellular Physiology, Stanford University, Stanford, California 94305, USA
 <sup>\*</sup>These authors contributed equally
 \*Email: shuo@stanford.edu

#### PACS number(s): 03.67.-a, 42.50.-p

#### Abstract

We demonstrate cavity-enhanced Raman emission from a single atomic defect in a solid. Our platform is a single silicon-vacancy center in diamond coupled with a monolithic diamond photonic crystal cavity. The cavity enables an unprecedented frequency tuning range of the Raman emission (100 GHz) that significantly exceeds the spectral inhomogeneity of siliconvacancy centers in diamond nanostructures. We also show that the cavity selectively suppresses the phonon-induced spontaneous emission that degrades the efficiency of Raman photon generation. Our results pave the way towards photon-mediated many-body interactions between solid-state quantum emitters in a nanophotonic platform. Integration of solid-state quantum emitters with nanophotonic structures offers a scalable quantum photonics platform<sup>1</sup> that is essential for photonic quantum simulation<sup>2</sup>, quantum metrology<sup>3</sup>, quantum repeaters<sup>4</sup>, and quantum networks<sup>5,6</sup>. However, despite significant progress in coupling single solid-state qubits with photons<sup>7-10</sup> and entangling two qubits<sup>11-14</sup>, a scalable quantum photonic circuit consisting of many quantum emitters remains an outstanding challenge. One major obstacle towards this goal is the spectral inhomogeneity of solid-state quantum emitters<sup>15</sup>, which limits their prospects in realizing many-body interactions through exchange of photons<sup>5</sup>. The ability to tune the emission frequency of a solid-state quantum emitter across the full range of inhomogeneous broadening remains a key missing ingredient in developing scalable quantum photonic circuits.

Color centers in solids have recently shown great promise for applications in scalable quantum photonic circuits, largely owing to their narrow spectral inhomogeneity. One of the candidates that has attracted significant interests in recent years is the negatively charged silicon-vacancy (SiV<sup>-</sup>) center in diamond. SiV<sup>-</sup> centers possess narrow inhomogeneous broadening on the order of 1 GHz in high quality diamond<sup>16,17</sup>. They also exhibit properties that make them promising as optically accessible quantum memories, including high spectral stability<sup>16</sup>, large zero-phonon-line emission (>70%)<sup>18</sup>, gigahertz coupling strength with nano-cavities<sup>13,19</sup>, as well as milliseconds spin coherence time<sup>20</sup>. Recent experiments have demonstrated photon-mediated entanglement between two SiV<sup>-</sup> centers in a bare waveguide<sup>13</sup>, where Raman emissions with a tuning range of 10 GHz were employed to compensate the spectral inhomogeneity of SiV<sup>-</sup> centers. However, there are two main limitations in using this approach towards realizing photon-mediated many-body interactions. First, once embedded in nanostructures, SiV<sup>-</sup> centers typically display a much larger spectral inhomogeneity (>20 GHz) than bulk due to the

difficulties in engineering a homogeneous strain distribution<sup>21</sup>. Second, the observed Raman emission is accompanied with a strong spontaneous emission from the same branch of the  $\Lambda$ -system<sup>13</sup>, which fundamentally limits the efficiency of Raman photon generation and the fidelity of many-body interactions. To address both challenges requires selective enhancement of the Raman emission while suppressing the undesired spontaneous emission.

In this Letter, we demonstrate cavity-enhanced Raman emission from a single color center. Cavity-enhanced Raman emission has been first demonstrated with single trapped atoms<sup>22-24</sup>. where tuning of the emission frequency by  $\sim 100$  linewidths has been achieved<sup>25</sup>, much larger than trap-induced linewidth broadening. In solid-state platforms, optical cavities have been utilized to enhance Raman emission from a single quantum dot, which enables generation of single-photons with large tuning bandwidth<sup>26</sup> and variable pulse shape<sup>27,28</sup>. However, the cavityenhanced tuning range remains two orders of magnitude smaller compared with the spectral inhomogeneity of quantum dots<sup>29</sup>. Here, we show that an optical cavity enables a frequency tuning range of 100 GHz for Raman emission from a single SiV<sup>-</sup> center in diamond, which is an order of magnitude larger than previously achieved with color centers and far exceeds the typical spectral inhomogeneity of SiV<sup>-</sup> centers in nanostructures. In addition, we provide a quantitative model to explain the undesired spontaneous emission by accounting for electron-phonon interactions, and show that the cavity can selectively suppress the spontaneous emission and only enhance the Raman photon generation. Our results represent an important step towards the implementation of scalable quantum circuits and quantum networks that involve multiple solidstate quantum emitters in an integrated nanophotonic platform.



FIG. 1. (Color online) (a) Energy level structure of a  $SiV^-$  center. (b) Relevant energy level structure of the emitter-cavity system for cavity-enhanced Raman emission.

Figure 1(a) shows the energy level structure of a single SiV<sup>-</sup> center<sup>30</sup>. In the absence of a magnetic field, the SiV<sup>-</sup> center contains two ground states separated by  $\delta_g$ , and two excited states separated by  $\delta_e$ . The values of  $\delta_g$  and  $\delta_e$  are typically  $\delta_g/2\pi = 50 \text{ GHz}$  and  $\delta_e/2\pi = 260 \text{ GHz}$  respectively<sup>30</sup>, but they increase significantly in the presence of strain<sup>31,32</sup>. In Fig. 2(c) we will show that for the specific emitter we measured, the ground state splitting is  $\delta_g/2\pi = 544 \text{ GHz}$ . We utilize the  $\Lambda$ -system formed by the lower excited state (labeled as  $|e\rangle$ ) and the two ground states (labeled as  $|g_1\rangle$  and  $|g_2\rangle$ ) to generate tunable Raman emission. We optically drive transition  $|g_1\rangle \leftrightarrow |e\rangle$  using a continuous-wave laser with a Rabi frequency given by  $\Omega$ , and couple transition  $|g_2\rangle \leftrightarrow |e\rangle$  to a cavity with a coupling strength given by g (vacuum Rabi frequency of 2g). We set the detuning between the driving laser and transition  $|g_1\rangle \leftrightarrow |e\rangle$  to be identical to the detuning between the cavity and transition  $|g_2\rangle \leftrightarrow |e\rangle$  (both are given by  $\Delta$ ) in order to achieve Raman resonance<sup>22-24</sup>. Note that unlike the scheme of stimulated Raman

adiabatic passage<sup>33</sup> that requires two lasers to drive both branches of the  $\Lambda$ -system, here we only need a single laser to drive one branch since the cavity will stimulate the emission from the other one.

To understand how we generate cavity-enhanced Raman emission, we illustrate the level structure in the interaction picture as shown in Fig. 1(b). We denote each state in the form  $|x, n\rangle$ , where  $x \in \{g_1, g_2, e\}$  is the state of the SiV<sup>-</sup> center, and  $n \in \{0, 1\}$  is the number of photons in the cavity. By truncating the infinite Jaynes-Cummings ladders, we implicitly assume that the system contains at most one excitation. This assumption is always valid in the absence of ground state relaxation<sup>34</sup>. When accounting for ground state relaxation, this assumption corresponds to the condition  $\kappa >> \gamma_{flip}$ , where  $\kappa$  is the cavity energy decay rate, and  $\gamma_{flip}$  is the ground state relaxation rate from  $|g_2\rangle$  to  $|g_1\rangle$ . We also assume that  $\Omega, g \ll \Delta$ , so that we can adiabatically eliminate the state  $|e,0\rangle$ , and treat the system as two-levels  $|g_1,0\rangle$  and  $|g_2,1\rangle$  driven by an effective Rabi frequency  $\Omega_{eff} = \Omega g / \Delta^{34}$ . Thus, if the system is initially in the state  $|g_1, 0\rangle$ , it will coherently rotate to the state  $|g_2,1\rangle$  with a Rabi frequency  $\Omega_{eff}$ , which then decays to the state  $|g_2,0
angle$  via emitting a photon through the cavity. The emission frequency is tunable with  $\Delta$ because it does not involve any real excitation of the state  $|e,0\rangle$ . We utilize the phonon-mediated ground state relaxation to reinitialize the state from  $|g_2,0\rangle$  back to  $|g_1,0\rangle$  after the Raman emission. Note that the reverse relaxation process from  $|g_1,0\rangle$  to  $|g_2,0\rangle$  is negligible as has been demonstrated recently<sup>31</sup>, because it requires absorption of a single phonon at the frequency  $\delta_g/2\pi = 544 \text{ GHz}$ , which is much larger than the thermal energy  $k_B T = 83 \text{ GHz}$  at the measurement temperature of 4 K.

The coupling between the emitter and the cavity enhances the rate of the Raman emission. Here we define the Raman emission rate as the inverse of the average time it takes to emit a photon when the system is initially in the state  $|g_1,0\rangle$ . In Supplementary Materials<sup>34</sup>, we

demonstrate that the cavity-enhanced Raman emission rate is given by  $R_c = \frac{\Omega_{eff}^2}{\kappa} = \frac{g^2}{\kappa} \cdot \left(\frac{\Omega}{\Delta}\right)^2$ , while the upper bound of the Raman emission rate without a cavity is given by  $R_0 = \frac{(\Omega/2)^2}{\Delta^2 + (\Gamma/2)^2} \cdot \Gamma$ , where  $\Gamma$  is the spontaneous emission rate of transition  $|e\rangle \rightarrow |g_2\rangle$ . In the

limit where  $\Delta >> \Gamma$ , the Raman emission rate is enhanced by a factor  $\frac{R_c}{R_0} = \frac{4g^2}{\kappa\Gamma}$ , which is the

Purcell factor of the coupled emitter-cavity system. For  $SiV^-$  centers, the Purcell factor can be more than a factor of  $10^{19}$ , corresponding to at least an order of magnitude enhancement of the Raman emission rate.

We couple a single SiV<sup>-</sup> center with a monolithic diamond nanobeam photonic crystal cavity<sup>19</sup>. Figure 2(a) shows a scanning electron microscope image of the fabricated cavity. The device fabrication starts with homoepitaxial growth of a thin layer of diamond on a single-crystal diamond substrate using microwave plasma chemical vapor deposition (MPCVD). We place a silicon wafer underneath the diamond substrate to generate silicon atoms in the growth chamber through hydrogen plasma etching, which then form SiV<sup>-</sup> centers due to plasma diffusion. We then fabricate nanobeam photonic crystal cavities using electron beam lithography followed by angled etching of the bulk diamond to create a suspended nanobeam<sup>38</sup>.

We mount our sample in a closed-cycle cryostat and cool it down to 4 K. Supplemental Materials contain detailed descriptions of the measurement methods<sup>34</sup>. We first measure the bare cavity transmission spectrum using a supercontinuum source (Fig. 2(b)). By fitting the measured

data (blue circles) to a Lorentzian function (red solid line), we obtain a cavity energy decay rate of  $\kappa/2\pi = 53.7 \pm 0.4$  GHz (corresponding to a quality factor of 7600).



FIG. 2. (Color online) (a) Scanning electron microscope image of a fabricated nanobeam photonic crystal cavity in diamond. (b) Transmission spectrum of a bare cavity measured using a supercontinuum source. (c) Photoluminescence spectrum of the SiV<sup>-</sup> center we used in our experiment. (d) Lifetime measurement of the lower excited state of the SiV<sup>-</sup> center when the cavity is far detuned from the emitter (upper panel) and when the cavity is resonantly coupled with transition  $|g_2\rangle \leftrightarrow |e\rangle$  (lower panel). In both panels (b) and (d), blue dots show the measured data, the red solid lines show the numerical fit.

Figure 2(c) shows the photoluminescence spectrum of the SiV<sup>-</sup> center embedded in the cavity. To eliminate the effect of the cavity on the emission spectrum, we red detune the cavity by more than 40 linewidths from all transitions of the SiV<sup>-</sup> center. We observe four distinct peaks in the photoluminescence spectrum, labeled as A – D in the figure, corresponding to the four optical transitions of a single SiV<sup>-</sup> center. The peaks C and D correspond to transitions  $|g_1\rangle \leftrightarrow |e\rangle$  and  $|g_2\rangle \leftrightarrow |e\rangle$  respectively<sup>39</sup>. From the frequency splitting between the emission peaks C and D, we calculate that  $\delta_g/2\pi = 544$  GHz. This value is significantly larger than the value obtained in the bulk (50 GHz) using the same sample<sup>34</sup>, suggesting large residual strain in the nanobeam photonic crystal. Second order correlation measurements verify that the emissions from both peaks C and D exhibit clear anti-bunching and are therefore originated from a single SiV<sup>-</sup> center<sup>34</sup>. We attribute the weak emission peak near transition C to a different emitter.

To characterize the coupling strength g between the cavity and transition  $|g_2\rangle \leftrightarrow |e\rangle$ , we measure the lifetime of the excited state  $|e\rangle$  both when the cavity is far detuned and resonant with the transition  $|g_2\rangle \leftrightarrow |e\rangle$ , as shown in the upper and lower panels of Fig. 2(d). By fitting the measured data (blue dots) to an exponential function (red solid line), we determine the lifetime of the excited state  $|e\rangle$  to be  $\tau_{off} = 1.74 \pm 0.01$  ns for the far detuned case, and  $\tau_{on} = 1.14 \pm 0.01$  ns for the resonant case. We thus calculate the coupling strength to be  $g/2\pi = 0.80 \pm 0.01$  GHz using the relation  $\frac{1}{\tau_{on}} = 4g^2/\kappa + \frac{1}{\tau_{off}}$ . We also estimate a lower-bound Purcell factor of  $20^{34}$ .

We now demonstrate cavity-enhanced Raman emission. We excite the transition  $|g_1\rangle \leftrightarrow |e\rangle$ using a continuous-wave laser with a variable detuning  $\Delta$ , and collect the emission from the cavity. To reject the direct reflection of the laser from the sample surface, we spatially separate the excitation and collection by irradiating the laser on a notch located at the end of the nanobeam, which is designed for coupling light from free-space to the waveguide<sup>13,34</sup>. We collect the far-field scattered signal from the cavity at the center of the nanobeam. We also use a double monochromator to further filter out the laser reflection and spectrally select the emission around transition  $|g_2\rangle \leftrightarrow |e\rangle$  within a bandwidth of 120 GHz. Figure 3(a) shows the measured emission spectrum as we vary the detuning  $\Delta$ . We observe two distinct peaks in the measured spectra, labeled as *R* and *S* respectively. The emission peak *R* continuously red shifts as we increase the detuning  $\Delta$ , corresponding to the cavity-enhanced Raman emission. The emission peak *S* remains centered around the natural frequency of  $|g_2\rangle \leftrightarrow |e\rangle$ , which is originated from incoherent excitation of the system into the state  $|e\rangle$ followed by spontaneous emission via transition  $|e\rangle \rightarrow |g_2\rangle$ . We are able to achieve a tuning range of 99 GHz for the Raman emission, which is an order of magnitude larger than the best value achieved previously for a color center<sup>13</sup>. Note that the demonstrated tuning range is only limited by the bandwidth of our spectral filter (120 GHz), and does not constitute a fundamental limit.



FIG. 3. (Color online) (a) Cavity emission spectra as we vary the excitation detuning  $\Delta$ . The blue dots show measured data, and the red solid lines show the numerical fits to a double Lorentzian function. The labels R and S represent the Raman and spontaneous emission peaks, respectively.

(b) Ratio between the Raman and spontaneous emission intensity as we vary the excitation detuning  $\Delta$ . The blue circles show measured values, and the red solid line shows numerically calculated ratios. (c) Cavity emission spectra as we tune the cavity across both the spontaneous and Raman emission peaks. In both panels (a) and (c), the frequency values are given in terms of detuning from transition  $|g_{1}\rangle \leftrightarrow |e\rangle$ .

Besides an unprecedented tuning bandwidth, the cavity also enables selective enhancement of Raman emission as we spectrally detune the Raman emission away from the emitter resonance. To quantitatively show this effect, we extract the ratio between the Raman and spontaneous emission intensity (referred as the R/S ratio) at each detuning, as shown in Fig. 3(b). The R/S ratio increases by a factor of 10 when we increase the detuning from 15 GHz to 88 GHz. The R/S ratio achieves even higher value at 99 GHz, but we cannot accurately calculate the ratio at this condition due to the vanishing spontaneous emission peak that is too close with the noise floor.

We now verify that the selective enhancement at large detuning originates from the cavity. We fix the excitation detuning at  $\Delta/2\pi = 55$  GHz, and finely tune the cavity frequency across both the Raman and spontaneous emission peaks. If the improvement of R/S ratio at large detuning is not related with the cavity, we should observe no dependence of the R/S ratio as we sweep the cavity frequency. In contrast, as shown in Fig. 3(c), when the cavity is resonant at the Raman emission frequency (-55 GHz), we observe at least 10-fold enhancement of the Raman emission intensity compared with the case when the cavity is detuned 100 GHz away from the Raman emission. The cavity can also enhance the spontaneous emission, but at a different frequency (~0 GHz). These results confirm that the selective enhancement of the Raman emission is enabled by the cavity.

Finally, we investigate the origin of the strong spontaneous emission, especially at small detuning. In fact, previous studies have observed similar spontaneous emission<sup>13</sup>, but the

physical mechanism for this observation has not been explored thoroughly. We quantitatively explain the spontaneous emission by accounting for interactions between the SiV<sup>-</sup> center and a phonon reservoir. Specifically, we derive a microscopic model that quantifies how the state  $|e\rangle$ is excited by absorbing both a photon from the driving field and a phonon from the reservoir, leading to the spontaneous emission.

We start with the Hamiltonian of the driven  $\Lambda$ -system shown in Fig. 1(b), given by

$$\hat{\mathbf{H}}_{sys} = \Delta |e,0\rangle \langle e,0| + \left(\frac{\Omega}{2}|e,0\rangle \langle g_1,0| + g|e,0\rangle \langle g_2,1| + h.c.\right).$$
(1)

We model the phonons as a bath of harmonic oscillators, given by

$$\hat{\mathbf{H}}_{bath} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \mathbf{b}_{\mathbf{k}}^{\dagger} \mathbf{b}_{\mathbf{k}}.$$
(2)

In Eq. (2), **k** is the wavevector of each phonon mode,  $\omega_{\mathbf{k}}$  is the frequency of the phonon mode **k**, and **b**<sub>k</sub> is the bosonic annihilation operator for the phonon mode **k**. The interaction Hamiltonian between the SiV<sup>-</sup> center and phonons could be written as

$$\hat{\mathbf{H}}_{sys-bath} = \sum_{\mathbf{k}} \left( \mathbf{b}_{\mathbf{k}} + \mathbf{b}_{\mathbf{k}}^{\dagger} \right) \left( p_{\mathbf{k}} \left| g_{1}, 0 \right\rangle \left\langle g_{1}, 0 \right| + q_{\mathbf{k}} \left| g_{2}, 1 \right\rangle \left\langle g_{2}, 1 \right| + r_{\mathbf{k}} \left| e, 0 \right\rangle \left\langle e, 0 \right| \right), \tag{3}$$

where  $p_{\mathbf{k}}$ ,  $q_{\mathbf{k}}$ , and  $r_{\mathbf{k}}$  are the deformation coupling strength between the phonon mode  $\mathbf{k}$  and the electronic states  $|g_1\rangle$ ,  $|g_2\rangle$ , and  $|e\rangle$  respectively. Note that here we do not include the phonon-induced ground state relaxation since this process only determines the number of excitation and emission cycles per second and does not affect the R/S ratio. We will add this term phenomenologically in the final master equation<sup>34</sup>.

We now derive the electron-phonon interactions in the form of Lindblad operators following a similar formalism used for semiconductor quantum dots<sup>40,41</sup>. To derive the Lindblad operators,

we first transform the interaction Hamiltonian  $\hat{\mathbf{H}}_{sys-bath}$  into the diagonal basis of  $\hat{\mathbf{H}}_{sys}$  (Eq. (1)), and then write it in the rotating reference frame with respect to  $\hat{\mathbf{H}}_{sys} + \hat{\mathbf{H}}_{bath}$ . The final master equation is given by  $d\rho_{sys}/dt = -i[\hat{\mathbf{H}}_{sys}, \rho_{sys}] + L_{phonon}(\rho_{sys})$ , where  $\rho_{sys}$  is the density matrix of the system, and  $L_{phonon}(\rho_{sys})$  is the phonon dissipator, given by

$$L_{phonon}\left(\rho_{sys}\right) = \frac{g^{2} + (\Omega/2)^{2}}{\Delta^{2}} J_{1}(\Delta) \Big[ n_{th}(\Delta) D(|+\rangle\langle-|) + (1 + n_{th}(\Delta)) D(|-\rangle\langle+|) \Big]$$

$$+ \frac{g^{2} + (\Omega/2)^{2}}{\Delta^{2}} J_{2}(\Delta) \Big[ n_{th}(\Delta) D(|+\rangle\langle d|) + (1 + n_{th}(\Delta)) D(|d\rangle\langle+|) \Big],$$
(4)

where  $D(\hat{\mathbf{O}})\rho_{sys} = \hat{\mathbf{O}}\rho_{sys}\hat{\mathbf{O}}^{\dagger} - \frac{1}{2}\hat{\mathbf{O}}^{\dagger}\hat{\mathbf{O}}\rho_{sys} - \frac{1}{2}\rho_{sys}\hat{\mathbf{O}}^{\dagger}\hat{\mathbf{O}}$  is the general Lindblad superoperator for

the collapse operator  $\hat{\mathbf{O}}$ . Note that here we only elaborate the phonon mediated dissipation for the convenience of discussion. The Supplementary Materials contain the complete master equation and detailed derivations<sup>34</sup>. In Eq. (4), the states  $|+\rangle$ ,  $|-\rangle$ , and  $|d\rangle$  are eigenstates of  $\hat{\mathbf{H}}_{sys}$ , given by

$$\left|+\right\rangle = \frac{\Omega}{2\Delta} \left|g_{1},0\right\rangle + \frac{g}{\Delta} \left|g_{2},1\right\rangle + \left|e,0\right\rangle,\tag{5}$$

$$|-\rangle = \frac{\Omega/2}{\sqrt{g^{2} + (\Omega/2)^{2}}} |g_{1},0\rangle + \frac{g}{\sqrt{g^{2} + (\Omega/2)^{2}}} |g_{2},1\rangle - \frac{\sqrt{g^{2} + (\Omega/2)^{2}}}{\Delta} |e,0\rangle.$$
(6)

$$|d\rangle = \frac{g}{\sqrt{g^{2} + (\Omega/2)^{2}}} |g_{1},0\rangle - \frac{\Omega/2}{\sqrt{g^{2} + (\Omega/2)^{2}}} |g_{2},1\rangle.$$
(7)

The parameters  $J_1(\Delta)$  and  $J_2(\Delta)$  are the spectral density of phonons that couple with the transition  $|+\rangle \leftrightarrow |-\rangle$  and  $|+\rangle \leftrightarrow |d\rangle$  respectively. The parameter  $n_{th}(\Delta)$  is the number of

phonons per mode, which follows the Bose-Einstein distribution given by  $n_{ih}(\Delta) = \left[\exp(\Delta/k_B T) - 1\right]^{-1}.$ 

The phonon dissipator in the form of Eq. (4) has a clear physical intuition. It shows how the system can be populated incoherently into the dressed state  $|+\rangle$  from the states  $|-\rangle$  or  $|d\rangle$  by absorption of a single phonon from the reservoir. Since  $|+\rangle \approx |e\rangle$  in the limit  $\Omega, g \ll \Delta$ , the incoherent population transfer into the state  $|+\rangle$  leads to spontaneous emission from the excited state. Eq. (4) also includes the reverse process where the state  $|+\rangle$  decays to the states  $|-\rangle$  or  $|d\rangle$  by emitting a phonon, but this process has a minor effect since its rate is typically much slower than other decay mechanisms of the excited state  $|+\rangle$ .

We numerically solve the master equation of the system, and calculate the cavity emission spectrum using the quantum regression theorem<sup>34</sup>. We set all the parameters using experimentally measured values, except for the phonon spectral densities  $J_1(\Delta)$  and  $J_2(\Delta)$ . The exact form of  $J_1(\Delta)$  and  $J_2(\Delta)$  depends on many parameters such as the strain susceptibility of each electronic state of the SiV<sup>-</sup> center, the local strain of each phonon mode, and the phonon frequency dispersion, which is difficult to derive from the first principles. Here, we qualitatively assume a phonon spectral density function of the form  $J_{1,2}(\Delta) = \alpha_{1,2}\Delta^n$ , where  $\alpha_{1,2}$  is a trivial scalar, and *n* represents a geometric scaling factor that is determined by the structure<sup>31</sup>. For example, for phonons in the bulk n=3, but for surface phonons n=2. The red solid line in Fig. 3(b) shows the calculated R/S ratio using our model. For the best fit, we obtain  $n = 0.31 \pm 0.24$ . This value is much smaller than the bulk value of 3, suggesting that the nanobeam strongly modifies the phonon spectral density.

In conclusion, we have demonstrated cavity-enhanced Raman emission from a single SiV center. The cavity enables an unprecedented frequency tuning range of 99 GHz, which significantly exceeds the typical spectral inhomogeneity of SiV<sup>-</sup> centers in nanostructures. We also demonstrate that the cavity selectively enhances only the Raman emission, which is critical for achieving high-fidelity photon-mediated many-body interactions. In our current experiment, we employed two orbital ground states to form a  $\Lambda$ -system, which have short lifetimes<sup>37</sup> and thus limit our capability to generate single photons due to fast re-excitation. In order to obtain pure single photons from the Raman emission, we could utilize the spin sublevels of SiV<sup>-</sup> centers, which have lifetimes of milli-seconds at cryogenic temperature<sup>42,43</sup> and seconds at milli-Kelvin temperature<sup>20</sup>. The long coherence time of the electron spin may further enable quantum state transfer between single spins and photons through cavity stimulated adiabatic Raman passage<sup>22</sup>. Another important property for photon-mediated many-body interactions is the photon coupling efficiency. In our current device, the input and output photon coupling are achieved through freespace scattering from either the cavity or the notches at the end of the waveguide, which has a limited efficiency on the order of 1%<sup>13</sup>. Such coupling efficiencies can be significantly improved by using an adiabatic tapered fiber to directly couple with the nanobeam<sup>44,45</sup> or by adopting optimized grating couplers for efficient coupling from free-space to on-chip structures<sup>46</sup>. In addition, we notice that the spontaneous emission process accompanied with the Raman emission offers rich information about electron-phonon interactions that are worth future study, including applications in laser cooling of mechanical resonators<sup>47</sup> and generating entangled photon-phonon pairs. Ultimately, our results represent an important step towards developing chip-integrated quantum circuits and quantum networks that employ multiple solid-state qubits mediated by single photons in a nanophotonic platform.

The authors would like to acknowledge fruitful discussions with Srujan Meesala. This work is supported by Department of Energy (DOE), Laboratory Directed Research and Development program at SLAC National Accelerator Laboratory (contract DE-AC02-76SF00515), Army Research Office (ARO) (W911NF1310309 and W911NF-18-1-0062), Air Force Office of Scientific Research (AFOSR) MURI Center for Quantum Metaphotonics and Metamaterials and MURI for attojoule optoelectronics (Award No. FA9550-17-1-0002), National Science Foundation (NSF) (ECS-9731293 and DMR-1503759), Stanford Nano Shared Facility, ONR MURI on Quantum Optomechanics (Award No. N00014-15-1-2761), National Science Foundation (NSF) EFRI ACQUIRE program (Award No. 5710004174), and the Army Research Laboratory CDQI (W911NF1520067). Device fabrication is performed in part at the Center for Nanoscale Systems (CNS) at Harvard University, a member of the National Nanotechnology Infrastructure Network (NNIN), which is supported by the National Science Foundation under NSF award No. ECS-0335765.CNS. C.D. acknowledges support from the Andreas Bechtolsheim Stanford Graduate Fellowship.

#### References

- 1 O'brien, J. L., Furusawa, A. & Vučković, J. Photonic quantum technologies. *Nature Photonics* **3**, 687 (2009).
- 2 Aspuru-Guzik, A. & Walther, P. Photonic quantum simulators. *Nature Physics* **8**, 285 (2012).
- 3 Giovannetti, V., Lloyd, S. & Maccone, L. Advances in quantum metrology. *Nature Photonics* 5, 222 (2011).
- 4 Briegel, H.-J., Dür, W., Cirac, J. I. & Zoller, P. Quantum repeaters: the role of imperfect local operations in quantum communication. *Physical Review Letters* **81**, 5932 (1998).
- 5 Cirac, J. I., Zoller, P., Kimble, H. J. & Mabuchi, H. Quantum state transfer and entanglement distribution among distant nodes in a quantum network. *Physical Review Letters* **78**, 3221 (1997).
- 6 Kimble, H. J. The quantum internet. *Nature* **453**, 1023 (2008).
- 7 Togan, E. *et al.* Quantum entanglement between an optical photon and a solid-state spin qubit. *Nature* **466**, 730 (2010).
- 8 De Greve, K. *et al.* Quantum-dot spin-photon entanglement via frequency downconversion to telecom wavelength. *Nature* **491**, 421 (2012).
- 9 Gao, W., Fallahi, P., Togan, E., Miguel-Sánchez, J. & Imamoglu, A. Observation of entanglement between a quantum dot spin and a single photon. *Nature* **491**, 426 (2012).
- 10 Sun, S., Kim, H., Solomon, G. S. & Waks, E. A quantum phase switch between a single solid-state spin and a photon. *Nature Nanotechnology* **11**, 539-544 (2016).
- 11 Bernien, H. *et al.* Heralded entanglement between solid-state qubits separated by three metres. *Nature* **497**, 86 (2013).
- 12 Delteil, A. *et al.* Generation of heralded entanglement between distant hole spins. *Nature Physics* **12**, 218 (2016).
- 13 Sipahigil, A. *et al.* An integrated diamond nanophotonics platform for quantum optical networks. *Science* **354**, 847 (2016).
- 14 Stockill, R. *et al.* Phase-tuned entangled state generation between distant spin qubits. *Physical Review Letters* **119**, 010503 (2017).
- 15 Aharonovich, I., Englund, D. & Toth, M. Solid-state single-photon emitters. *Nature Photonics* **10**, 631 (2016).
- 16 Rogers, L. J. *et al.* Multiple intrinsically identical single-photon emitters in the solid state. *Nature Communications* **5**, 4739 (2014).
- 17 Sipahigil, A. *et al.* Indistinguishable photons from separated silicon-vacancy centers in diamond. *Physical Review Letters* **113**, 113602 (2014).
- 18 Neu, E. *et al.* Single photon emission from silicon-vacancy colour centres in chemical vapour deposition nano-diamonds on iridium. *New Journal of Physics* **13**, 025012 (2011).
- 19 Zhang, J. L. *et al.* Strongly Cavity-Enhanced Spontaneous Emission from Silicon-Vacancy Centers in Diamond. *Nano Letters* **18**, 1360–1365 (2018).
- 20 Sukachev, D. D. *et al.* Silicon-Vacancy Spin Qubit in Diamond: A Quantum Memory Exceeding 10 ms with Single-Shot State Readout. *Physical Review Letters* **119**, 223602 (2017).
- 21 Evans, R. E., Sipahigil, A., Sukachev, D. D., Zibrov, A. S. & Lukin, M. D. Narrow-linewidth homogeneous optical emitters in diamond nanostructures via silicon ion implantation. *Physical Review Applied* **5**, 044010 (2016).
- 22 Hennrich, M., Legero, T., Kuhn, A. & Rempe, G. Vacuum-stimulated Raman scattering based on adiabatic passage in a high-finesse optical cavity. *Physical Review Letters* **85**, 4872 (2000).
- 23 Kuhn, A., Hennrich, M. & Rempe, G. Deterministic single-photon source for distributed quantum networking. *Physical Review Letters* **89**, 067901 (2002).
- 24 Keller, M., Lange, B., Hayasaka, K., Lange, W. & Walther, H. Continuous generation of single photons with controlled waveform in an ion-trap cavity system. *Nature* **431**, 1075 (2004).
- 25 Mücke, M. *et al.* Generation of single photons from an atom-cavity system. *Physical Review A* **87**, 063805

(2013).

- Sweeney, T. M. *et al.* Cavity-stimulated Raman emission from a single quantum dot spin. *Nature Photonics* 8, 442 (2014).
- 27 Pursley, B., Carter, S., Yakes, M., Bracker, A. & Gammon, D. Picosecond pulse shaping of single photons using quantum dots. *Nature Communications* **9**, 115 (2018).
- 28 Lee, J. *et al.* Multi-dimensional photonic states from a quantum dot. *Quantum Science and Technology* **3**, 2 (2018).
- 29 Lodahl, P., Mahmoodian, S. & Stobbe, S. Interfacing single photons and single quantum dots with photonic nanostructures. *Reviews of Modern Physics* **87**, 347 (2015).
- 30 Hepp, C. *et al.* Electronic structure of the silicon vacancy color center in diamond. *Physical Review Letters* **112**, 036405 (2014).
- 31 Sohn, Y.-I. *et al.* Controlling the coherence of a diamond spin qubit through its strain environment. *Nature Communications* **9**, 2012 (2018).
- 32 Meesala, S. *et al.* Strain engineering of the silicon-vacancy center in diamond. *Physical Review B* **97**, 205444 (2018).
- 33 Vitanov, N. V., Rangelov, A. A., Shore, B. W. & Bergmann, K. Stimulated Raman adiabatic passage in physics, chemistry, and beyond. *Reviews of Modern Physics* **89**, 015006 (2017).
- 34 See Supplemental Material at [URL will be inserted by publisher] for detailed theoretical analysis and supporting experimental results, which includes Refs. [13, 30-32, 35-37].
- 35 Brion, E., Pedersen, L. H. & Mølmer, K. Adiabatic elimination in a lambda system. *Journal of Physics A: Mathematical and Theoretical* **40**, 1033 (2007).
- 36 Müller, T. *et al.* Optical signatures of silicon-vacancy spins in diamond. *Nature communications* **5**, 3328 (2014).
- 37 Jahnke, K. D. *et al.* Electron–phonon processes of the silicon-vacancy centre in diamond. *New Journal of Physics* **17**, 043011 (2015).
- 38 Burek, M. J. *et al.* High quality-factor optical nanocavities in bulk single-crystal diamond. *Nature Communications* 5, 5718 (2014).
- 39 Rogers, L. J. *et al.* Electronic structure of the negatively charged silicon-vacancy center in diamond. *Physical Review B* **89**, 235101 (2014).
- 40 Gauger, E. M., Benjamin, S. C., Nazir, A. & Lovett, B. W. High-fidelity all-optical control of quantum dot spins: Detailed study of the adiabatic approach. *Physical Review B* 77, 115322 (2008).
- 41 McCutcheon, D. P. & Nazir, A. Quantum dot Rabi rotations beyond the weak exciton–phonon coupling regime. *New Journal of Physics* **12**, 113042 (2010).
- 42 Pingault, B. *et al.* All-optical formation of coherent dark states of silicon-vacancy spins in diamond. *Physical Review Letters* **113**, 263601 (2014).
- 43 Rogers, L. J. *et al.* All-optical initialization, readout, and coherent preparation of single silicon-vacancy spins in diamond. *Physical Review Letters* **113**, 263602 (2014).
- 44 Tiecke, T. *et al.* Efficient fiber-optical interface for nanophotonic devices. *Optica* **2**, 70-75 (2015).
- 45 Burek, M. J. *et al.* Fiber-Coupled Diamond Quantum Nanophotonic Interface. *Physical Review Applied* **8**, 024026 (2017).
- 46 Gould, M., Schmidgall, E. R., Dadgostar, S., Hatami, F. & Fu, K.-M. C. Efficient extraction of zerophonon-line photons from single nitrogen-vacancy centers in an integrated GaP-on-diamond platform. *Physical Review Applied* **6**, 011001 (2016).
- 47 Wilson-Rae, I., Zoller, P. & Imamo lu, A. Laser cooling of a nanomechanical resonator mode to its quantum ground state. *Physical Review Letters* **92**, 075507 (2004).