

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

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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 silicon-vacancy 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.

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Integration of solid-state quantum emitters with nanophotonic structures offers a scalable quantum photonics platform [1] that is essential for photonic quantum simulation [2], quantum metrology [3], quantum repeaters [4], and quantum networks [5,6]. However, despite significant progress in coupling single solid-state qubits with photons [7–10] and entangling two qubits [11–14], 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 [15], which limits their prospects in realizing many-body interactions through exchange of photons [5]. 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 interest in recent years is the negatively charged silicon-vacancy (SiV⁻) center in diamond. SiV⁻ centers possess narrow inhomogeneous broadening on the order of 1 GHz in high-quality diamond [16,17]. They also exhibit properties that make them promising as optically accessible quantum memories, including high spectral stability [16], large zero-phonon-line emission (>70%) [18], gigahertz coupling strength with nanocavities [13,19], as well as milliseconds spin coherence time [20]. Recent

experiments have demonstrated photon-mediated entanglement between two SiV⁻ centers in a bare waveguide [13], where Raman emissions with a tuning range of 10 GHz were employed to compensate the spectral inhomogeneity of SiV⁻ centers. However, there are two main limitations in using this approach towards realizing photon-mediated many-body interactions. First, once embedded in nanostructures, SiV⁻ centers typically display a much larger spectral inhomogeneity (>20 GHz) than bulk due to the difficulties in engineering a homogeneous strain distribution [21]. Second, the observed Raman emission is accompanied with a strong spontaneous emission from the same branch of the Λ system [13], 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 [22–24], where tuning of the emission frequency by ~ 100 linewidths has been achieved [25], 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 [26] and variable pulse shape [27,28]. However, the cavity-enhanced tuning range remains 2 orders of magnitude smaller compared with the spectral inhomogeneity of

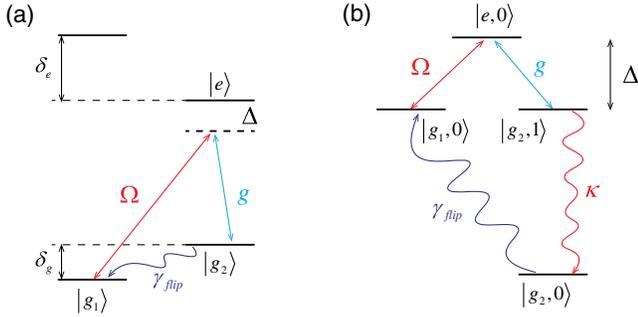


FIG. 1. (a) Energy level structure of a SiV^- center. (b) Relevant energy level structure of the emitter-cavity system for cavity-enhanced Raman emission.

quantum dots [29]. Here, we show that an optical cavity enables a frequency tuning range of 100 GHz for Raman emission from a single SiV^- 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^- 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 solid-state quantum emitters in an integrated nanophotonic platform.

Figure 1(a) shows the energy level structure of a single SiV^- center [30]. In the absence of a magnetic field, the SiV^- center contains two ground states separated by δ_g and two excited states separated by δ_e . The values of δ_g and δ_e are typically $\delta_g/2\pi = 50$ GHz and $\delta_e/2\pi = 260$ GHz, respectively [30], but they increase significantly in the presence of strain [31,32]. In Fig. 2(c) we will show that, for the specific emitter we measured, the ground state splitting is $\delta_g/2\pi = 544$ GHz. We utilize the Λ 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 Ω , 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 Δ) in order to achieve Raman resonance [22–24]. Note that, unlike the scheme of stimulated Raman adiabatic passage [33] that requires two lasers to drive both branches of the Λ 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^- 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 [34]. When accounting for ground state relaxation, this assumption corresponds to the condition $\kappa \gg \gamma_{\text{flip}}$, where κ is the cavity energy decay rate, and γ_{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 $\Omega_{\text{eff}} = \Omega g/\Delta$ [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 Ω_{eff} , which then decays to the state $|g_2, 0\rangle$ via emitting a photon through the cavity. The emission frequency is tunable with Δ 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 [31], because it requires absorption of a single phonon at the frequency $\delta_g/2\pi = 544$ GHz, which is much larger than the thermal energy $k_B T = 83$ 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 the Supplemental Material [34], we demonstrate that the cavity-enhanced Raman emission rate is given by $R_c = (\Omega_{\text{eff}}^2/\kappa) = (g^2/\kappa)(\Omega/\Delta)^2$, while the upper bound of the Raman emission rate without a cavity is given by $R_0 = [(\Omega/2)^2]/[\Delta^2 + (\Gamma/2)^2]\Gamma$, where Γ is the spontaneous emission rate of transition $|e\rangle \rightarrow |g_2\rangle$. In the limit where $\Delta \gg \Gamma$, the Raman emission rate is enhanced by a factor $(R_c/R_0) = (4g^2/\kappa\Gamma)$, 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^- center with a monolithic diamond nanobeam photonic crystal cavity [19]. 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. 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^- 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 [38].

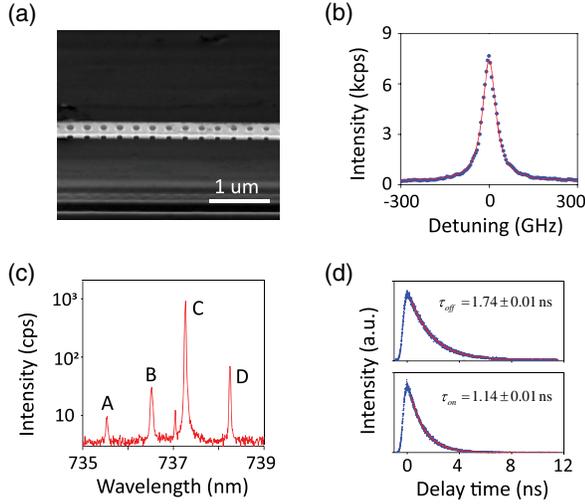


FIG. 2. (a) Scanning electron microscope image of a fabricated nanobeam photonic crystal cavity in diamond. (b) Transmission spectrum of a bare cavity measured using a supercontinuum source. (c) Photoluminescence spectrum of the SiV^- center we used in our experiment. (d) Lifetime measurement of the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

We mount our sample in a closed-cycle cryostat and cool it down to 4 K. The Supplemental Material contains detailed descriptions of the measurement methods [34]. 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).

Figure 2(c) shows the photoluminescence spectrum of the SiV^- 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^- 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^- center. The peaks C and D correspond to transitions $|g_1\rangle \leftrightarrow |e\rangle$ and $|g_2\rangle \leftrightarrow |e\rangle$, respectively [39]. 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 [34], 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 antibunching and are therefore originated from a single SiV^- center [34]. 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 $\tau_{\text{off}} = 1.74 \pm 0.01$ ns for the far detuned case and $\tau_{\text{on}} = 1.14 \pm 0.01$ 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 $(1/\tau_{\text{on}}) = 4g^2/\kappa + (1/\tau_{\text{off}})$. 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 Δ 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 [13,34]. 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 Δ . We observe two distinct peaks in the measured spectra, labeled as R and S, respectively. The emission peak R continuously redshifts as we increase the detuning Δ , 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

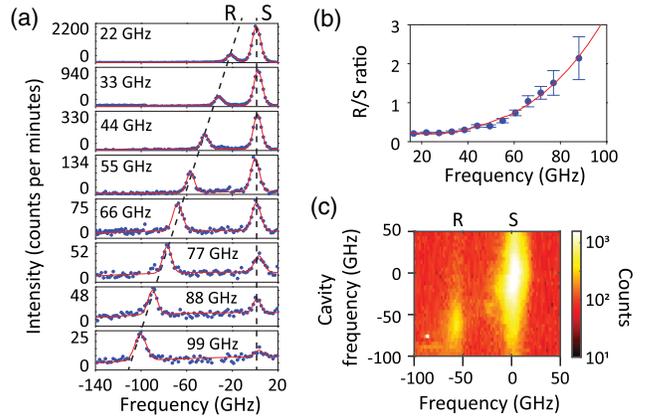


FIG. 3. (a) Cavity emission spectra as we vary the excitation detuning Δ . The blue dots show measured data, and the red solid lines show the numerical fits to a double Lorentzian function. The labels R and S represent the Raman and spontaneous emission peaks, respectively. (b) Ratio between the Raman and spontaneous emission intensity as we vary the excitation detuning Δ . 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. (a),(c) The frequency values are given in terms of detuning from transition $|g_2\rangle \leftrightarrow |e\rangle$.

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 [13]. 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.

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 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 tenfold 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 [13], 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^- 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 Λ system shown in Fig. 1(b), given by

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

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

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

In Eq. (2), \mathbf{k} is the wave vector of each phonon mode, $\omega_{\mathbf{k}}$ is the frequency of the phonon mode \mathbf{k} , and $\mathbf{b}_{\mathbf{k}}$ is the bosonic annihilation operator for the phonon mode \mathbf{k} . The interaction Hamiltonian between the SiV^- center and phonons could be written as

$$\hat{\mathbf{H}}_{\text{sys-bath}} = \sum_{\mathbf{k}} (\mathbf{b}_{\mathbf{k}} + \mathbf{b}_{\mathbf{k}}^\dagger) (p_{\mathbf{k}}|g_1, 0\rangle\langle g_1, 0| + q_{\mathbf{k}}|g_2, 1\rangle\langle g_2, 1| + r_{\mathbf{k}}|e, 0\rangle\langle e, 0|), \quad (3)$$

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 [34].

We now derive the electron-phonon interactions in the form of Lindblad operators following a similar formalism used for semiconductor quantum dots [40,41]. To derive the Lindblad operators, we first transform the interaction Hamiltonian $\hat{\mathbf{H}}_{\text{sys-bath}}$ into the diagonal basis of $\hat{\mathbf{H}}_{\text{sys}}$ [Eq. (1)], and then write it in the rotating reference frame with respect to $\hat{\mathbf{H}}_{\text{sys}} + \hat{\mathbf{H}}_{\text{bath}}$. The final master equation is given by $d\rho_{\text{sys}}/dt = -i[\hat{\mathbf{H}}_{\text{sys}}, \rho_{\text{sys}}] + L_{\text{phonon}}(\rho_{\text{sys}})$, where ρ_{sys} is the density matrix of the system, and $L_{\text{phonon}}(\rho_{\text{sys}})$ is the phonon dissipator, given by

$$L_{\text{phonon}}(\rho_{\text{sys}}) = \frac{g^2 + (\Omega/2)^2}{\Delta^2} J_1(\Delta) [n_{\text{th}}(\Delta) D(|+\rangle\langle -|) + (1 + n_{\text{th}}(\Delta)) D(|-\rangle\langle +|)] + \frac{g^2 + (\Omega/2)^2}{\Delta^2} J_2(\Delta) [n_{\text{th}}(\Delta) D(|+\rangle\langle d|) + (1 + n_{\text{th}}(\Delta)) D(|d\rangle\langle +|)], \quad (4)$$

where $D(\hat{\mathbf{O}})\rho_{\text{sys}} = \hat{\mathbf{O}}\rho_{\text{sys}}\hat{\mathbf{O}}^\dagger - \frac{1}{2}\hat{\mathbf{O}}^\dagger\hat{\mathbf{O}}\rho_{\text{sys}} - \frac{1}{2}\rho_{\text{sys}}\hat{\mathbf{O}}^\dagger\hat{\mathbf{O}}$ 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 Supplemental Material contains the complete master equation and detailed derivations [34]. In Eq. (4), the states $|+\rangle$, $|-\rangle$, and $|d\rangle$ are eigenstates of $\hat{\mathbf{H}}_{\text{sys}}$, given by

$$|+\rangle = \frac{\Omega}{2\Delta}|g_1, 0\rangle + \frac{g}{\Delta}|g_2, 1\rangle + |e, 0\rangle, \quad (5)$$

$$|-\rangle = \frac{\Omega/2}{\sqrt{g^2 + (\Omega/2)^2}} |g_1, 0\rangle + \frac{g}{\sqrt{g^2 + (\Omega/2)^2}} |g_2, 1\rangle - \frac{\sqrt{g^2 + (\Omega/2)^2}}{\Delta} |e, 0\rangle. \quad (6)$$

$$|d\rangle = \frac{g}{\sqrt{g^2 + (\Omega/2)^2}} |g_1, 0\rangle - \frac{\Omega/2}{\sqrt{g^2 + (\Omega/2)^2}} |g_2, 1\rangle. \quad (7)$$

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 $n_{\text{th}}(\Delta)$ is the number of phonons per mode, which follows the Bose-Einstein distribution given by $n_{\text{th}}(\Delta) = [\exp(\Delta/k_B T) - 1]^{-1}$.

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. Equation (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 [34]. 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⁻ 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 [31]. 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⁻ 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 Λ system, which have short lifetimes [37] and thus limit our capability to generate single photons due to fast reexcitation. In order to obtain pure single photons from the Raman emission, we could utilize the spin sublevels of SiV⁻ centers, which have lifetimes of milliseconds at cryogenic temperature [42,43] and seconds at millidegrees Kelvin temperature [20]. 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 [22]. 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 free-space scattering from either the cavity or the notches at the end of the waveguide, which has a limited efficiency on the order of 1% [13]. Such coupling efficiencies can be significantly improved by using an adiabatic tapered fiber to directly couple with the nanobeam [44,45] or by adopting optimized grating couplers for efficient coupling from free-space to on-chip structures [46]. 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 [47] 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.

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