Supplemental Material

A. Fundamental vibration mode of the carbon nanotube

The motion of a suspended nanotube can be described by the Euler-Bernoulli theory [S1, S2]. The Euler-Bernoulli equation for the static and dynamic displacement of a thin beam subjected to an external driving force reads

$$\rho A \frac{\partial^2}{\partial t^2}\phi(x,t) + E\mathcal{I}\frac{\partial^4}{\partial x^4}\phi(x,t) - T\frac{\partial^2}{\partial x^2}\phi(x,t) = F_{\rm ext}(x,t), \tag{S1}$$

where $\phi(x,t)$ is the lateral displacement in the y direction, ρ the mass density, A the beam cross section, E the Young modulus, $\mathcal{I}$ the moment of inertia, T the tension in the tube, and $F_{\rm ext}(x,t)$ a unit length force that accounts for the effect of the gate electrodes. The displacement amplitude increases remarkably when the tube is driven at an eigenfrequency of the system. These eigenfrequencies correspond to the bending modes of the nanotube, which we label by mode number n starting from zero for the fundamental mode. We here consider the eigenfrequencies of a perfect clamping nanotube resonator, in which case the tension T goes to zero at zero gate voltage $F_{\rm ext}(x,t) = 0$ [S3, S4]. In this case the Euler-Bernoulli equation reads

$$\rho A \frac{\partial^2}{\partial t^2}\phi(x,t) + E\mathcal{I}\frac{\partial^4}{\partial x^4}\phi(x,t) = 0. \tag{S2}$$

The eigenmodes ϕ_n and eigenfrequencies ω_n satisfy:

$$\omega_n^2 \rho A \phi_n = E\mathcal{I}\frac{\partial^4}{\partial x^4}\phi_n \tag{S3}$$

The solutions to this equation are

$$\phi_n(x) = C_1(\cos k_n x - \cosh k_n x) + C_2(\sin k_n x - \sinh k_n x). \tag{S4}$$

For a doubly clamped beam, the boundary conditions are $\phi_n(0) = \phi_n(L) = 0, \phi'_n(0) = \phi'_n(L) = 0$, and for a cantilever the boundary conditions are $\phi_n(0) = \phi'_n(0) = 0, \phi''_n(L) = \phi'''_n(L) = 0$. For the former case, the frequency equation is given by

$$\cos k_n L \cosh k_n L = 1. \tag{S5}$$

For the latter, the frequency equation is given by

$$\cos k_n L \cosh k_n L = -1. \tag{S6}$$

The first five nontrivial consecutive roots of these equations are given below

Mode n	Cantilever $k_n L$	Beam $k_n L$
0	1.875	4.730
1	4.694	7.853
2	7.855	10.996
3	10.996	14.137
4	14.137	17.279

The corresponding eigenfrequencies are

$$\omega_{\rm n} = k_n^2 \sqrt{\frac{E\mathcal{I}}{\rho A}}. \tag{S7}$$

Therefore, the fundamental vibrational mode of a nanotube has the vibration frequency $\omega_{\rm nt} \sim \frac{1}{L^2}\sqrt{\frac{E\mathcal{I}}{\rho A}}$ [S3, S4]. In table I we present the relevant parameters for the carbon nanotube resonator without gate voltages.

TABLE I. Parameters for the nanotube considered in this work.

Term	Value	Units
Length L	2	μm
Radius r	1.5	nm
Wall thickness t	0.335	nm
Mass density ρ	1350	kg/m^3
Effective mass m	7×10^{-21}	kg
Young modulus E	1	TPa
Fundamental frequency ω_0	$2\pi\times 2$	MHz
Current carrying capacity C	≥ 10	μA/nm^2
Current I	60	μA
Quality factor Q	10^5	/

B. Derivation of the spin-vibration interaction

The interaction of a single NV center located at $\vec{r}$ with the total magnetic field (external driving and from the nanotube) is

$$\hat{H}_{\rm NV} = \hbar D\hat{S}_z^2 + \mu_B g_s\ B_z\hat{S}_z + \mu_B g_s(\vec{B}_{\rm nt}(\vec{r}) + \vec{B}_{\rm dr})\cdot\hat{\vec{S}}. \tag{S8}$$

Expanding the magnetic field $\vec{B}_{\rm nt}(\vec{r})$ up to first order in $\hat{u}_y$, we have

$$\hat{H}_{\rm NV} = \hbar D\hat{S}_z^2 + \mu_B g_s[\ B_z + B_{\rm nt}(d)]\hat{S}_z + \mu_B g_s\vec{B}_{\rm dr}\cdot\hat{\vec{S}} + \mu_B g_s\hat{S}_z\partial_y B_{\rm nt}\hat{u}_y. \tag{S9}$$

In the basis defined by the eigenstates of $\hat{S}_z$, i.e., $\{|m_s\rangle, m_s = 0, \pm 1\}$, with $\hat{S}_z|m_s\rangle = m_s|m_s\rangle$, we get

$$\begin{aligned}\hat{H}_{\rm NV} = &\sum_{m_s}\{\langle m_s|[\hbar D\hat{S}_z^2 + \mu_B g_s[\ B_z + B_{\rm nt}(d)]\hat{S}_z]|m_s\rangle\}|m_s\rangle\langle m_s|\\ &+\sum_{m_s,m_s'}\{\langle m_s|\mu_B g_s\vec{B}_{\rm dr}\cdot\hat{\vec{S}}|m_s'\rangle\}|m_s\rangle\langle m_s'| + \sum_{m_s}\{\langle m_s|\mu_B g_s\hat{S}_z\partial_y B_{\rm nt}\hat{u}_y|m_s\rangle\}|m_s\rangle\langle m_s|.\end{aligned} \tag{S10}$$

Taking $\vec{B}_{\rm dr} = B_0\cos\omega_0 t\vec{e}_x = B_0/2(e^{i\omega_0 t} + e^{-i\omega_0 t})\vec{e}_x$, we have

$$\begin{aligned}\hat{H}_{\rm NV} = &\sum_{m_s}\{\hbar D m_s^2 + \mu_B g_s[B_z + B_{\rm nt}(d)]m_s\}|m_s\rangle\langle m_s|\\ &+\sum_{m_s,m_s'}\frac{1}{2}\mu_B g_s B_0(e^{i\omega_0 t} + e^{-i\omega_0 t})\langle m_s|\hat{S}_x|m_s'\rangle|m_s\rangle\langle m_s'|\\ &+\sum_{m_s}\mu_B g_s(\hbar/2m\omega_{\rm nt})^{1/2}\partial_y B_{\rm nt} m_s|m_s\rangle\langle m_s|(\hat{a}^\dagger + \hat{a}).\end{aligned} \tag{S11}$$

In the rotating-frame at the driving frequency ω_0 and under the rotating-wave approximation, we can obtain

$$\begin{aligned}\hat{H}_{\rm NV} = &(\hbar D + \mu_B g_s B_z + B_{\rm nt} - \hbar\omega_0)|+1\rangle\langle +1| + (\hbar D - \mu_B g_s B_z - B_{\rm nt} - \hbar\omega_0)|-1\rangle\langle -1|\\ &+\frac{\sqrt{2}}{4}\mu_B g_s B_0(|0\rangle\langle +1| + |+1\rangle\langle 0|) + \frac{\sqrt{2}}{4}\mu_B g_s B_0(|0\rangle\langle -1| + |-1\rangle\langle 0|)\\ &+\mu_B g_s(\hbar/2m\omega_{\rm nt})^{1/2}\partial_y B_{\rm nt}(|+1\rangle\langle +1| - |-1\rangle\langle -1|)(\hat{a}^\dagger + \hat{a}).\end{aligned} \tag{S12}$$

Including the free Hamiltonian of the vibration mode, we have

$$\begin{aligned}\hat{H}_{\rm NV} = &\hbar\omega_{\rm nt}\hat{a}^\dagger\hat{a} + \hbar\Delta_+|+1\rangle\langle +1| + \hbar\Delta_-|-1\rangle\langle -1|\\ &+\hbar\Omega[|-1\rangle\langle 0| + |0\rangle\langle -1|] + \hbar\Omega[|+1\rangle\langle 0| + |0\rangle\langle +1|]\\ &+\hbar g(|+1\rangle\langle +1| - |-1\rangle\langle -1|)(\hat{a}^\dagger + \hat{a}).\end{aligned} \tag{S13}$$

with $\hbar\Delta_\pm = \hbar D \pm \mu_B g_s(B_z + B_{\rm nt}) - \hbar\omega_0$, $\hbar\Omega = \frac{\sqrt{2}}{4}\mu_B g_s B_0$, and $\hbar g = \mu_B g_s(\hbar/2m\omega_{\rm nt})^{1/2}\partial_y B_{\rm nt}$.

In the following we assume symmetric detunings $\Delta_+ = \Delta_- = \Delta$ for simplicity. We can define the bright and dark states for the NV spin states

$$
\begin{aligned}
|\mathcal{B}\rangle &= \frac{1}{\sqrt{2}}(|+1\rangle + |-1\rangle) \\
|\mathcal{D}\rangle &= \frac{1}{\sqrt{2}}(|+1\rangle - |-1\rangle).
\end{aligned} \tag{S14}
$$

Then we find that the microwave field couples the state $|0\rangle$ to the bright state $|\mathcal{B}\rangle$, while the dark state $|\mathcal{D}\rangle$ remains decoupled. In the dressed state basis $\{|\mathcal{G}\rangle = \cos\theta|0\rangle - \sin\theta|\mathcal{B}\rangle, |\mathcal{E}\rangle = \cos\theta|\mathcal{B}\rangle + \sin\theta|0\rangle\}$,with $\tan 2\theta = 2\sqrt{2}\Omega/\Delta$, Hamiltonian (S13) can be rewritten as [S5]

$$
\begin{aligned}
\hat{H}_{\mathrm{NV}} = &\hbar\omega_{\mathrm{nt}}\hat{a}^\dagger\hat{a} + \hbar\omega_{eg}|\mathcal{E}\rangle\langle\mathcal{E}| + \hbar\omega_{dg}|\mathcal{D}\rangle\langle\mathcal{D}| \\
&+\hbar(g_1|\mathcal{G}\rangle\langle\mathcal{D}| + g_2|\mathcal{D}\rangle\langle\mathcal{E}| + \mathrm{H.c.})(\hat{a}^\dagger + \hat{a}),
\end{aligned} \tag{S15}
$$

where $\omega_{eg} = \sqrt{\Delta^2 + 8\Omega^2}$, $\omega_{dg} = \frac{\Delta+\sqrt{\Delta^2+8\Omega^2}}{2}$, $g_1 = -g\sin\theta$, and $g_2 = g\cos\theta$. Under the condition $\Delta \gg \Omega$, one has $\sin\theta \simeq 0$, $\cos\theta \simeq 1$, $\omega_{eg} \simeq \Delta + \frac{4\Omega^2}{\Delta}$, $\omega_{dg} \simeq \Delta + \frac{2\Omega^2}{\Delta}$, and $|\mathcal{E}\rangle \simeq |\mathcal{B}\rangle$, which leads to

$$
\hat{\mathcal{H}}_q = \hbar\omega_{\mathrm{nt}}\hat{a}^\dagger\hat{a} + \frac{1}{2}\hbar\Lambda\hat{\sigma}_z + \hbar g(\hat{\sigma}_+ + \hat{\sigma}_-)(\hat{a}^\dagger + \hat{a}). \tag{S16}
$$

C. The two-qubit operations

1. *Strong spin-spin interactions mediated by phonons*

We consider two NV centers, separated by a distance $l \sim 1\mu m$, coupled to the same vibration mode of the nanotube, with Hamiltonian

$$
\hat{\mathcal{H}}_{2q} = \hbar\omega_{\mathrm{nt}}\hat{a}^\dagger\hat{a} + \sum_{i=1,2}\frac{1}{2}\hbar\Lambda_i\hat{\sigma}_z^i + \sum_{i=1,2}\hbar g_i(\hat{\sigma}_+^i + \hat{\sigma}_-^i)(\hat{a}^\dagger + \hat{a}). \tag{S17}
$$

For simplicity, we assume $\Lambda_1 \simeq \Lambda_2 = \Lambda$ and $g_1 \simeq g_2 = g$, and consider the dispersive regime $|\Lambda - \omega_{\mathrm{nt}}| \gg g$, when two NV centers are far detuned from the resonator but in resonance with each other. After the use of a Schrieffer-Wolff transformation, this will lead to an effective nonlocal spin-spin interaction via the exchange of virtual phonons,

$$
\hat{\mathcal{H}}_{\text{s-s}} = \hbar\lambda_{\mathrm{eff}}(\hat{\sigma}_+^1\hat{\sigma}_-^2 + \hat{\sigma}_-^1\hat{\sigma}_+^2), \tag{S18}
$$

with the coupling strength $\lambda_{\mathrm{eff}} = g^2/|\Lambda - \omega_{\mathrm{nt}}|$. This interaction can extend over distances on the order of the nanotube's length, which allows us to coherently control the interactions between distant NV spins.

We now proceed to discuss the coherence length l_c of the phonon mediated NV spin coupling, which is essential to evaluate the application potential of this device. In particular, it is a very important issue when propagating phonons are considered rather than the confined one used in this work. At ambient temperature, phonon scattering inside solid state materials results in harmful decoherence processes. An important figure of merit that is used to quantitatively characterize all the phonon dissipation mechanisms, including the decay of vibrations into the support as well as intrinsic damping mechanism within the nanotube, e.g. due to scattering from surface defects, is the mechanical quality factor Q, defined as the ratio of the resonant frequency over the linewidth. It is interesting to estimate a sort of coherence length for the nanoresonator mode. To do so one can think of the fundamental bending mode as a traveling wave, which is reflected at the ends of the nanotube. Thus, the coherence length can be estimated as the effective mean phonon free path $l_c \simeq v\tau$ [S6, S7], where v is the effective speed of sound, and τ is the relaxation time (mechanical damping rate $\gamma_m = \tau^{-1}$). The effective speed of sound v can be found from the relation as $\omega_{\mathrm{nt}} = vk_0$ [S8, S9], while the relaxation time is related to the quality factor of the mechanical mode $\frac{1}{\tau} = \omega_{\mathrm{nt}}/Q$. Thus, we can estimate the coherence length of the phonon mediated NV spin coupling as $l_c \sim Q/k_0 \sim QL$. It can extend over distances on the order of several centimeters, much larger than the distance between two NV spins. Therefore, for a high-Q mechanical resonator, phonons can coherently propagate inside it back and forth for quite a long distance before they finally dissipate. This phenomenon has a direct analogy to photons bound in a high-Q micro-cavity. Thus, in this scheme, we can safely ignore the harmful effect of phonon scattering inside solid state materials, provided that the mechanical resonator possesses a very high quality factor at low temperature. We need to emphasize that the recent fabrication of carbon nanotube resonators can possess a quality factor exceeding 10^5 [S10], which ensures that phonon losses do not severely limit our scheme. The minimum requirement of this work is that the length scale of the phonon mediated NV spin coupling is on the order of the tube's length, which would allow us to coherently control the NV spin interactions, and facilitate potential applications of this hybrid device.

2. *Dynamics of the two coupled NV spins*

To implement this protocol, we need a microwave to drive the transition between the qubit state $|0\rangle_j$ and the bright state $|\mathcal{B}\rangle_j$ in each qubit with Rabi frequency Ω_j and frequency detuning δ_j. The dynamics of the entire system is described by

$$\hat{\mathcal{H}} = \sum_j \hbar\delta_j |\mathcal{B}\rangle_{jj}\langle\mathcal{B}| + \sum_j [\hbar\Omega_j |\mathcal{B}\rangle_{jj}\langle 0| + \text{H.c.}] + \hat{\mathcal{H}}_{\text{s-s}}. \tag{S19}$$

The spin-spin interaction can be diagonalized with the states $|\pm\rangle_q = 1/\sqrt{2}[|\mathcal{B}\rangle_1|\mathcal{D}\rangle_2 \pm |\mathcal{D}\rangle_1|\mathcal{B}\rangle_2]$, leading to

$$\hat{\mathcal{H}}_{\text{s-s}} = \hbar\lambda_{\text{eff}}|+\rangle_{qq}\langle +| - \hbar\lambda_{\text{eff}}|-\rangle_{qq}\langle -|. \tag{S20}$$

To implement a SWAP gate and quantum information transfer between two qubits, we encode quantum information in the two spin states as $|0\rangle_q = |0\rangle$ and $|1\rangle_q = |\mathcal{D}\rangle$. The entire system is described by

$$\hat{\mathcal{H}} = \sum_j \hbar\delta_j |\mathcal{B}\rangle_{jj}\langle\mathcal{B}| + \sum_j [\hbar\Omega_j |\mathcal{B}\rangle_{jj}\langle 0| + \text{H.c.}] + \hbar\lambda_{\text{eff}}|+\rangle_{qq}\langle +| - \hbar\lambda_{\text{eff}}|-\rangle_{qq}\langle -|. \tag{S21}$$

To gain more insight into the dynamics of the coupled system, we write the Hamiltonian Eq. (S21) in the space S spanned by the state vectors $\{|0,1\rangle_q, |+\rangle_q, |-\rangle_q, |1,0\rangle_q, |0,0\rangle_q, |1,1\rangle_q, |\mathcal{B},\mathcal{B}\rangle_q, |0,\mathcal{B}\rangle_q, |\mathcal{B},0\rangle_q\}$,

$$\hat{\mathcal{H}} = \hbar \begin{bmatrix} 0 & \bar{\Omega}_1 & \bar{\Omega}_1 & 0 & 0 & 0 & 0 & 0 & 0 \\ \bar{\Omega}_1 & \delta_+ & 0 & \bar{\Omega}_2 & 0 & 0 & 0 & 0 & 0 \\ \bar{\Omega}_1 & 0 & -\delta_- & -\bar{\Omega}_2 & 0 & 0 & 0 & 0 & 0 \\ 0 & \bar{\Omega}_2 & -\bar{\Omega}_2 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & \Omega_2 & \Omega_1 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & \delta_1+\delta_2 & \Omega_1 & \Omega_2 \\ 0 & 0 & 0 & 0 & \Omega_2 & 0 & \Omega_1 & \delta_2 & 0 \\ 0 & 0 & 0 & 0 & \Omega_1 & 0 & \Omega_2 & 0 & \delta_1 \end{bmatrix} \tag{S22}$$

From the matrix form for the Hamiltonian Eq. (S21), we see that the space S can be decomposed into two independent subspaces $S_1 = \{|0,1\rangle_q, |+\rangle_q, |-\rangle_q, |1,0\rangle_q\}$ and $S_2 = \{|0,0\rangle_q, |1,1\rangle_q, |\mathcal{B},\mathcal{B}\rangle_q, |0,\mathcal{B}\rangle_q, |\mathcal{B},0\rangle_q\}$, i.e., $S = S_1 \oplus S_2$. Thus we can find that if the two qubits are initially prepared in the state $|0\rangle_q^1|1\rangle_q^2$ or $|1\rangle_q^1|0\rangle_q^2$, then the dynamics of the system will be confined in the subspace S_1 governed by the Hamiltonian

$$\begin{aligned}\hat{\mathcal{H}} = {} & \hbar\delta_+|+\rangle_{qq}\langle +| - \hbar\delta_-|-\rangle_{qq}\langle -| + \hbar\bar{\Omega}_1|+\rangle_{qq}\langle 0,1| \\ & + \hbar\bar{\Omega}_1|-\rangle_{qq}\langle 0,1| + \hbar\bar{\Omega}_2|+\rangle_{qq}\langle 1,0| - \hbar\bar{\Omega}_2|-\rangle_{qq}\langle 1,0| + \text{H.c.}\end{aligned} \tag{S23}$$

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