Coupling Nitrogen Vacancy Centers in Diamond to Superconducting Flux Qubits: Supplementary Information

D. Marcos,[1] M. Wubs,[2, 3] J. M. Taylor,[4] R. Aguado,[1] M. D. Lukin,[5] and A. S. Sørensen[2]

[1] *Departamento de Teoría de la Materia Condensada,*
Instituto de Ciencia de Materiales de Madrid, CSIC, Cantoblanco 28049, Madrid, Spain
[2]*QUANTOP, The Niels Bohr Institute, University of Copenhagen, 2100 Copenhagen Ø, Denmark*
[3]*DTU Fotonik, Danish Technical University, 2800 Kgs. Lyngby, Denmark*
[4]*Joint Quantum Institute and the National Institute of Standards and Technology, College Park, MD 20742*
[5]*Department of Physics, Harvard University, Cambridge MA 02138*

In the main text we have omitted several details from the estimates of decoherence and the performance of the procedures. In the following we give more details on how these were obtained.

PACS numbers: 03.67.Lx, 85.25.Cp, 76.30.Mi

I. DECOHERENCE OF THE NV CENTERS

A. $\sqrt{\text{SWAP}}$ operation between individual NV centers

In the main text, we describe a method to perform an entangling operation between two NV centers near the same FQ using this as a quantum bus. The interaction between different NVs is mediated off-resonantly by the FQ and can be described starting from the Hamiltonian $\hat{H}_{\text{RWA}}$ in the main text, extended to the case of 2 NVs. Taking the mixing angle $\theta \to \pi/2$, such that the FQ is optimally biased with respect to low-frequency flux noise, and a rotating frame such that the FQ is detuned $-\delta$, rather than the NVs being detuned δ, this Hamiltonian reads

$$H = -\frac{\delta}{2}(\hat{\tau}_3 + 1) + \frac{1}{\sqrt{2}}\hat{\tau}_- \left(W^{FQ}_{\perp,1}\hat{\sigma}^{(1)}_+ + W^{FQ}_{\perp,2}\hat{\sigma}^{(2)}_+\right) + \text{H.c.} \tag{1}$$

In this picture, it becomes clear that there are two zero-energy eigenstates: $H|00\rangle|0\rangle_{FQ} = H|D\rangle|0\rangle_{FQ} = 0$, with the dark state $|D\rangle \equiv \frac{1}{\sqrt{2}G_2}\left(W^{FQ}_{\perp,2}|10\rangle - W^{FQ}_{\perp,1}|01\rangle\right)$ and the collective coupling constant for the two NVs $G_2 = \sqrt{(W^{FQ}_{\perp,1})^2 + (W^{FQ}_{\perp,2})^2}/\sqrt{2} = \sqrt{g_1^2 + g_2^2}$. There is a corresponding bright state $|B\rangle = \frac{1}{\sqrt{2}G_2}\left(W^{FQ}_{\perp,1}|10\rangle + W^{FQ}_{\perp,2}|01\rangle\right)$, and the state $|B\rangle|0\rangle_{FQ}$ is coupled to $|00\rangle|1\rangle_{FQ}$ with coupling strength G_2. Similarly the state $|11\rangle|0\rangle_{FQ}$ is coupled to $|B\rangle|1\rangle_{FQ}$ with coupling strength G_2.

Therefore, in second order perturbation theory, projecting onto the lower energy state of the flux qubit, we have

$$H_{\text{eff}} = \frac{G_2^2}{\delta}(|B\rangle\langle B| + |11\rangle\langle 11|) \ . \tag{2}$$

Here we will assume $W^{FQ}_{\perp,1} \approx W^{FQ}_{\perp,2}$ and thus $G_2 \approx \sqrt{2}g$. The probability of finding the system in the excited state is then zero for the two dark states and $2g^2/\delta^2$ for the two bright states of the system. Thus, FQ relaxation and Markovian dephasing, which occur directly to the FQ at a rate $1/T_2^{\text{FQ}}$, will lead to errors at a rate $\sim 2g^2/\delta^2 T_2^{\text{FQ}}$ for the two NV spins. Low- frequency noise enters in small variations of δ, which also contribute at order $2g^2/\delta^2$. Since such low-frequency noise is non-Markovian it will only give a quadratic contribution $\sim (g^2 t/(\delta^2 T_2^{\text{FQ}}))^2$, which will be less severe than the Markovian dephasing. We therefore take a worst-case scenario and assume the decoherence to be produced by Markovian dephasing.

To optimize the gate operation, we note that the overall time when the FQ is coupled is $t_X = \frac{\pi}{2}\frac{\delta}{2g^2}$. There is an additional wait time for the NV centers of $2t_W = t_X(1 - 4\phi/\pi)$, where $\phi = 2\sin^{-1}(1/\sqrt{8})$ as in the main text. For NV spins in the states $|00\rangle, |D\rangle$, the error is entirely from the total time $t_X + 2t_W$, as the FQ is not included, while for NV spins in the states $|11\rangle, |B\rangle$, both FQ and NV dephasing errors enter. We take the latter case to overestimate the error induced in the operation, and find an effective dephasing of the NV center $\exp[-(\frac{t_X+2t_W}{T_2^{\text{NV}}})^3]$, as expected from a dipole-dipole bath decorrelation, and an additional dephasing due to the admixture of the FQ as $\exp[-\frac{2t_X g^2}{\delta^2 T_2^{\text{FQ}}}]$. The only free parameter we can optimize over is the detuning δ. Rewriting the total fidelity as

$$F = \exp[-\frac{\delta^3}{\alpha^3} - \frac{\beta}{\delta}] \tag{3}$$

with $\alpha = \frac{2g^2 T_2^{\mathrm{NV}}}{\pi(1-2\phi/\pi)}$ and $\beta = \frac{\pi}{2T_2^{\mathrm{FQ}}}$, the optimum occurs for $\delta_* = (\alpha^3\beta/3)^{1/4}$, giving $F_* = \exp[-(\beta/\alpha)^{3/4}(3^{1/4}+3^{-3/4})]$. Substituting α and β in δ_* and the numbers given in the main text we find $F_* = \exp[-2.18(g^2 T_2^{\mathrm{FQ}} T_2^{\mathrm{NV}})^{-3/4}] \approx 0.98$ and $\delta_* = 0.96[g^6 (T_2^{\mathrm{NV}})^3 / T_2^{\mathrm{FQ}}]^{1/4} \approx 2\pi \times 3.6$ MHz.

An additional error arises when $W_{\perp,1}^{FQ} \neq W_{\perp,2}^{FQ}$, since then the unitary operation is not performing exactly the desired evolution; this leads to a reduced fidelity of the entangling operation by a factor $W_{\perp,1}^{FQ} W_{\perp,2}^{FQ}/g^2$. It is important to note, however, that this imperfection is not a decoherence effect and it may be possible to exploit the resulting unitary evolution even despite this imperfection, i.e., the resulting operation still resembles a $\sqrt{\mathrm{SWAP}}$.

B. Decoherence of the ensemble of NV centers

In the main article we described the transfer of an excitation from the FQ to the collective state of an ensemble of NV centers. In this section we estimate the dephasing of this collective state of the ensemble due to magnetic dipole-dipole interactions with other spins in the diamond crystal. The paramagnetic impurities in the diamond crystal consist of both other NVs centers with either the same or different orientation as well as unpaired electronic spins on substitutional Nitrogens. In most experiments the ratio of Nitrogen to NV centers is typically quite low, e.g. on the order of 1% to 10% [1]. Below we refer to this ratio as the conversion efficiency.

The dipole-dipole interaction is described by the Hamiltonian [2]

$$H_{\mathrm{dip}} = \sum_{j\neq k} \frac{1}{2} \frac{\mu_0}{4\pi r_{jk}^3} \Big[(\vec{m}^{(j)} \cdot \vec{m}^{(k)} - 3(\vec{m}^{(j)} \cdot \vec{e}_{jk})(\vec{m}^{(k)} \cdot \vec{e}_{jk}) \Big], \tag{4}$$

where r_{jk} is the distance between the dipoles j and k, $\vec{e}_{jk}$ is a unit vector between them, and $\vec{m}^{(j)}$ and $\vec{m}^{(k)}$ are their magnetic moments. These can be expressed in terms of the spin operators of the impurities, but the quantization axis for the different spins will be different: NV centers will be aligned along the symmetry axis of the center whereas electron spins on Nitrogens will be quantized along the axis of the applied static field. To describe this situation we introduce spin operators $\hat{s}_z^{(j)}$, $\hat{s}_+^{(j)}$ and $\hat{s}_-^{(j)}$, defined relative to the quantization axis of each particular spin, and take the different orientations into account in the coupling constant describing the interaction between different spins.

For the NV centers we furthermore ignore the $m_S = -1$ state which is assumed to be shifted out of resonance by a magnetic field. All the spins are therefore two-level systems which can be described by Pauli matrices $\hat{\sigma}_z^{(j)}$, $\hat{\sigma}_+^{(j)}$ and $\hat{\sigma}_-^{(j)}$. The spin operators can then be expressed as

$$\hat{s}_z^{(j)} = \frac{1}{2}(\hat{\sigma}_z^{(j)} + l_j), \qquad \hat{s}_+^{(j)} = \sqrt{1+l_j}\,\hat{\sigma}_+^{(j)}, \tag{5}$$

where the quantity l_j, which is unity for NV centers and vanishes for Nitrogen spin, accounts for the fact that the Nitrogen has spin 1/2 whereas the two-level NV system is made from the $m_S = 0$ and $m_S = 1$ states of a spin-1 particle. The magnetic dipole Hamilonian is then given by

$$H_{\mathrm{dip}} = \sum_{j,k\neq j} a_{jk} \hat{\sigma}_+^{(j)} \hat{\sigma}_-^{(k)} + b_{jk} (\hat{\sigma}_z^{(j)} + l_j)(\hat{\sigma}_z^{(k)} + l_k), \tag{6}$$

where we have used the rotating-wave approximation to ignore terms which do not conserve energy. By this approximation, the first term vanishes between spins with different resonance frequencies, i.e., with different orientations. Notice, that if the quantization axes for spin j and k are different, then the angle between these axes can be contained in the coupling constants a_{jk} and b_{jk}. This is the case for the interaction between the NV and Nitrogen spins, whose quantization axes, determined by the crystal axis and the external magnetic field respectively, form an angle β between them. For the sake of simplicity, we here only present the case $\beta = 0$. This is a worst-case scenario for the T_2^* derived below, but the general situation $\beta \neq 0$ only leads to minor modifications. With parallel quantization axes, we have

$$\begin{aligned} a_{jk} &= \mu_{jk}^{(a)} \left[1 - 6 e_-^{jk} e_+^{jk} \right] = \mu_{jk}^{(a)} \left[1 - \frac{3}{2}\sin^2\Theta_{jk} \right], \\ b_{jk} &= \mu_{jk}^{(b)} \left[1 - 3(e_z^{jk})^2 \right] = \mu_{jk}^{(b)} \left[1 - 3\cos^2\Theta_{jk} \right], \end{aligned} \tag{7}$$

where Θ_{jk} is the angle between the vector $\vec{e}_{jk}$ and the crystal axis, $e_\pm^{jk} := (e_x^{jk} \pm i e_y^{jk})/2$, and

$$\mu_{jk}^{(a)} := \frac{1}{2}\frac{\mu_0}{4\pi} \frac{\mu_B^2 g_e^{(j)} g_e^{(k)} \sqrt{1+l_j}\sqrt{1+l_k}}{r_{jk}^3}, \qquad \mu_{jk}^{(b)} := \frac{1}{8}\frac{\mu_0}{4\pi} \frac{\mu_B^2 g_e^{(j)} g_e^{(k)}}{r_{jk}^3}, \tag{8}$$

with μ_B the Bohr magneton and $g_e^{(j)}$ the electron g-factor of the jth spin.

To evaluate the dephasing of the spin wave we will assume that we start out in a state $|00...0\rangle_{\rm NV}$ where all the NVs are initially prepared in their ground states, and that a superposition state $c_0|0\rangle_{\rm FQ}+c_1|1\rangle_{\rm FQ}$ is transferred into the collective spin wave, resulting in a state $(c_0+c_1J_+)|00...0\rangle_{\rm NV}$, with $J_+=(1/G)\sum_j g_j\hat{\sigma}_+^{(j)}$. We then evaluate the time evolution of the coherence

$$\langle J_-(t)\rangle = {\rm Tr}\big[e^{iHt}J_-e^{-iHt}(c_0+c_1J_+)|00...0\rangle_{\rm NV}\langle 0...00|(c_0^*+c_1^*J_-)\rho_{\rm B}\big], \tag{9}$$

where $\rho_{\rm B}$ is the initial density matrix of the bath. Since the Hamiltonian (6) conserves the number of excitations in the NVs, this expression can be simplified to

$$\langle J_-(t)\rangle = c_0^*c_1{\rm Tr}\big[e^{iHt}J_-e^{-iHt}J_+|00...0\rangle_{\rm NV}\langle 0...00|\rho_{\rm B}\big]. \tag{10}$$

Writing out the above equation in terms of single-spin operators we see that the dephasing is determined by the time evolution of the two-point correlation function $\langle\hat{\sigma}_-^{(j)}(t)\hat{\sigma}_+^{(k)}(t=0)\rangle$ in the state $|00...0\rangle_{\rm NV}\langle 0...00|\rho_{\rm B}$. Since the density of Nitrogen is much higher than the density of NVs, the dephasing of the spin wave will predominantly be due to the interaction with the electronic spins of Nitrogen atoms, and we shall therefore ignore the interaction among the NVs. In this approximation there is no longer a mechanism in the Hamiltonian which can transfer the excitation from one NV to another and the correlation function $\langle\hat{\sigma}_-^{(j)}(t)\hat{\sigma}_+^{(k)}(t=0)\rangle$ vanishes exactly for $j\neq k$. The dephasing of the spin waves thus reduces to the calculation of the single-spin dephasing averaged over the spin wave:

$$\langle J_-(t)\rangle = c_0^*c_1\sum_j\frac{|g_j|^2}{G^2}{\rm Tr}\Big[e^{iHt}\hat{\sigma}_-^{(j)}e^{-iHt}\hat{\sigma}_+^{(j)}|00...0\rangle_{\rm NV}\langle 0...00|\rho_{\rm B}\Big], \tag{11}$$

where the sum runs over all NVs in the spin wave. Assuming all NVs to be equivalent this can be simplified to the calculation of the dephasing of a single-spin in a bath

$$\langle J_-(t)\rangle = \langle J_-(t=0)\rangle{\rm Tr}\Big[e^{iHt}\hat{\sigma}_-^{(j)}e^{-iHt}\hat{\sigma}_+^{(j)}|0\rangle_{\rm NV}\langle 0|\rho_{\rm B}\Big]. \tag{12}$$

When considering a single NV, the action of the spin bath described by (6) essentially corresponds to a random magnetic field generated from the spin ensemble along the crystal axis. This effective field will be fluctuating in time because the dipole-dipole interaction among the Nitrogen spins introduces flip-flop processes. When the Nitrogen concentration is much higher than that of NVs, the distance between Nitrogens is comparable to the distance between NVs and the nearest Nitrogen. These flip-flop processes will therefore take place on a time scale which is comparable or only slightly faster than the NV dephasing time. For simplicity we here ignore the flip-flop processes and consider a static environment. This represents a worst-case scenario, since this approximation removes the time averaging of the field from these processes. The actual dephasing time T_2^* will therefore be slightly larger than what we predict here. Within this approximation the expression in Eq. (12) can be reduced considerably:

$$\langle J_-(t)\rangle = \langle J_-(t=0)\rangle{\rm Tr}\left[\prod_k e^{-i4b_{jk}\hat{\sigma}_z^{(k)}t}\rho_{\rm B}\right], \tag{13}$$

where the product is over the Nitrogen spins.

With expression (12) we can evaluate the dephasing for a given bath. For a given density n, the typical strength of the spin-spin interaction is $\mu_0\mu_B^2g_e^2n/4\pi$, of the order of ~ 25 μK for high Nitrogen densities $n_{\rm N}\sim 10^{19}$ cm^{-3}. This is much smaller than the typical operating temperature of the FQ (tens of mK), and we can therefore neglect the interaction among the spins for determining the initial density matrix of the bath $\rho_{\rm B}$. Furthermore any non-vanishing mean value $\langle\hat{\sigma}_k^{(j)}\rangle\neq 0$ only results in a mean shift of the resonance frequency of the ensemble. The dephasing will thus be determined by spin fluctuations. A worst-case scenario can be obtained by assuming that the mean value vanishes $\langle\hat{\sigma}_k^{(j)}\rangle=0$, in which case the variance is maximal. The evolution of the coherence is then given by

$$\langle J_-(t)\rangle = \langle J_-(t=0)\rangle\prod_k\cos(4b_{jk}t). \tag{14}$$

To simplify this expression we expand it in time and find

$$\langle J_-(t)\rangle = \langle J_-(t=0)\rangle\Big(1-2t^2n_{\rm N}\int d^3\vec{r}_kb_{jk}^2\Big), \tag{15}$$

where we have replaced the sum by an integral and introduced the Nitrogen density n_{N}. Using Eq. (7) this expression reduces to

$$\langle J_-(t)\rangle = \langle J_-(t=0)\rangle \left[1 - t^2 \left(\frac{\mu_0}{4\pi}\frac{\mu_B^2 g_e^{\mathrm{N}} g_e^{\mathrm{NV}}}{8}\right)^2 4\pi n_{\mathrm{N}} \int_0^{\pi} \sin\Theta(1-3\cos^2\Theta)^2 d\Theta \int \frac{1}{r^4} dr\right]. \tag{16}$$

The angular integral gives 8/5 but the radial integral has a strong divergence at $r \to 0$. Since the integral is an integral over the distance between the NV and the Nitrogen impurity, this divergency represents the very fast dephasing of NV centers which happen to have a nearby Nitrogen spin. Such NV centers with a nearby Nitrogen spin will, however, also be far out of resonance during the interaction with the Flux qubit. This interaction takes a time $\sim 1/G$, and therefore any NV with a dipole interaction stronger than G will effectively not participate in the spin wave. We exclude these NVs by truncating the integral at a distance r_{min} when the interaction strength reaches the value of the coupling constant G, i.e.,

$$G = \frac{1}{8}\frac{\mu_0}{4\pi}\frac{\mu_B^2 g_e^N g_e^{NV}}{r_{\mathrm{min}}^3}. \tag{17}$$

This truncation of the integral gives

$$\langle J_-(t)\rangle = \langle J_-(t=0)\rangle \left[1 - t^2 \frac{\mu_0 \mu_B^2 g_e^{\mathrm{N}} g_e^{\mathrm{NV}}}{15\eta} n_{\mathrm{NV}} G\right], \tag{18}$$

where η is the Nitrogen-to-NV conversion efficiency. The result (18) corresponds to the first term in an expansion of an exponential decay $\exp(-(t/T_2^*)^2)$ with coherence time

$$T_2^* = \frac{1}{\sqrt{\frac{\mu_0 \mu_B^2 g_e^{\mathrm{N}} g_e^{\mathrm{NV}}}{15\eta} n_{\mathrm{NV}} G}}. \tag{19}$$

Notice that in order to be applicable, our regularization procedure requires r_{min} to be much smaller than the typical distance between spins $1/\sqrt[3]{n_{\mathrm{N}}}$, since we require that only a small fraction of the NVs are excluded. This condition is fulfilled in the interesting regime $GT_2^* \gg 1$. Taking a conversion efficiency $\eta = 0.05$, we find $T_2^* \approx 0.3$ μs for a FQ with $L = 5$ μm and a density $n_{\mathrm{NV}} = 10^{18}$ cm^{-3}, corresponding to $G \approx 2\pi \times 15$ MHz. For a full transfer of the state from the FQ to the spin wave and back we need a transfer time $t = \pi/G$, corresponding to an infidelity of the order of $1 - F \sim (t/2T_2^*)^2 < 0.5\%$ (the factor of 2 accounting for the fact that the excitation only spends half of the transfer time in the spin wave). For different densities the infidelity scales as $1 - F \propto \sqrt{n_{\mathrm{NV}}}$, becoming smaller at lower densities due to the reduced dipole-dipole interaction, e.g., at $n_{\mathrm{NV}} = 10^{16}$ cm^{-3} the error is reduced by an order of magnitude but then we are approaching the limit where the FQ decoherence becomes important. The transfer of excitations from the FQ to the spin wave is thus feasible in the regime 10^{16} cm$^{-3} \lesssim n_{\mathrm{NV}} \lesssim 10^{18}$ cm^{-3}. In order for the spin system to be useful as a long-term memory the coherence time should, however, also be sufficiently long to allow for the transfer to the nuclear spin for long-term storage. Since this can at best be achieved on a time scale set by the hyperfine interaction (~ 5 MHz), this would exclude working at the highest densities in this interval. Working at a density of $n_{\mathrm{NV}} \sim 10^{17}$ cm^{-3} leads to $T_2^* \approx 1.8\mu$s, which is sufficient to allow a transfer of the excitation from the electron spin to the nuclear spin with an infidelity $1 - F$ at the percent level.

The estimates above indicate that it is realistic to achieve a transfer of excitations from the FQ to the spin wave and back even without extending the coherence time by spin-echo techniques. These may, however, be desirable in order to achieve even longer coherence times. In particular T_2 may be extended if the NVs can be flipped by an external AC field on a time scale faster than the Nitrogen flip-flop processes. One should, however, be careful about applying spin-echo to the collective NV spin since, e.g. errors in the pulse area may give rise to collective decoherence processes. These can be more detrimental to quantum states stored in collective degrees of freedom than in individual spins (for instance flipping the $|0\rangle - |1\rangle$ transition would require control of the pulse area to an accuracy better than $1/\sqrt{N}$ in order to preserve the collective state). Since the dephasing of the NV spin wave is dominated by Nitrogen spins a more desirable solution could be to apply the external driving field to the electron spins on the Nitrogen atoms. If these spins are flipped on a time scale much faster than the flip-flop processes it would lead to an increased coherence time of the NV spin wave. Since Nitrogen spins have a considerably different resonance frequency, this could be achieved with little influence on the NV spin wave where the quantum information is stored.

II. DECOHERENCE OF THE FLUX QUBIT DUE THE SPIN BATH

A different concern is the extent to which the diamond crystal with a high density of spins may cause dephasing of the flux qubit, of particular importance for the coupling with an ensemble of NVs. As discussed above, the paramagnetic impurities in the diamond crystal consist primarily of NV centers and unpaired electron spins on Nitrogen atoms which were not converted into NVs centers during the annealing process. Due to the typically low conversion efficiency from Nitrogen to NV centers, we shall first consider the effect of Nitrogen impurity spins and then discuss the role of the NV centers. The coupling of the FQ to paramagnetic impurities can be described by a Hamiltonian similar to Eq. (1) in the main text:

$$\hat{H} = \hat{H}_{\text{spin}} + \varepsilon\hat{\tau}_3/2 + \lambda\hat{\tau}_1 + \hat{\tau}_3\sum_j \vec{W}^{\text{FQ}}(\vec{r}_j)\cdot\vec{S}^{(j)}, \tag{20}$$

where the spin Hamiltonian $\hat{H}_{\text{spin}} = \sum_j \Delta_j S_z^{(j)}/2 + \hat{H}_{\text{int}}$ describes an energy splitting Δ_j of the individual spins and the interaction between them, encapsulated in $\hat{H}_{\text{int}}$. Changing to the dressed-state picture of the flux qubit, this Hamiltonian is transformed into

$$\hat{H} = \hat{H}_{\text{spin}} + \omega\hat{\tau}_3/2 + (\cos\theta\hat{\tau}_3 - \sin\theta\hat{\tau}_1)\sum_j \vec{W}^{\text{FQ}}(\vec{r}_j)\cdot\vec{S}^{(j)}. \tag{21}$$

In the rotating frame with respect to $\omega\hat{\tau}_3/2$, the operator $\hat{\tau}_3$ remains stationary whereas $\hat{\tau}_1$ oscillates at a frequency ω – much higher than any time scale of the bath, provided that there are no near resonance impurities in the diamond sample. This is the case for the Nitrogen spins, whose splitting is determined by the applied field. The NVs are near resonance and will be dealt with below. The slowly varying and rapidly oscillating terms have different qualitative behavior so we will consider them separately.

Let us first consider the slowly-varying contribution. As we will show now this contribution vanishes if we work close to the degeneracy point of the FQ $\cos\theta \approx 0$ where the left- and right-circulating current states are degenerate. The slowly-varying contribution is described by

$$H_{\text{slow}} = \hat{\tau}_3 W_{\text{eff}}^{FQ}, \tag{22}$$

with

$$W_{\text{eff}}^{FQ} \equiv cos(\theta)\sum_j \left[W_z^{FQ}(\vec{r}_j)\cos(\beta_j) - W_\perp^{FQ}(\vec{r}_j)\sin(\beta_j)\right] S_3^{(j)}, \tag{23}$$

and β_j being the angle between the quantization axis the jth spin and the crystal axis of the NV (recall that the z-axis is defined by the crystal axis). For simplicity, we define

$$\kappa_j := W_z^{FQ}(\vec{r}_j)\cos(\beta_j) - W_\perp^{FQ}(\vec{r}_j)\sin(\beta_j). \tag{24}$$

Since the interaction of the flux qubit with an individual spin is on the order of g, the former has little influence on the state of an individual spin for the duration of the interaction $\sim 1/G \sim 1/g\sqrt{N}$. Furthermore the coupling of the FQ to collective degrees of freedom in the spin bath, will only have a limited influence on the state of an unpolarized bath, since the FQ can flip at most a single-spin. We can therefore ignore the influence of the FQ on the spin bath and consider W_{eff}^{FQ} as a fluctuating external field. Any mean value of this field will merely give rise to a shift of the resonance frequency which can be compensated and we thus need to consider the fluctuations. The root mean square of the fluctuating field is given by

$$\delta W_{\text{eff}}^{FQ} \;=\; \cos(\theta)\Big[\sum_{j,k}\kappa_j\kappa_k\langle\langle S_3^{(j)}S_3^{(k)}\rangle\rangle\Big]^{1/2}, \tag{25}$$

where we have used the cumulant notation $\langle\langle ab\rangle\rangle := \langle(a-\langle a\rangle)(b-\langle b\rangle)\rangle$. As discussed in the previous section, the temperature is typically high compared to the dipole-dipole interaction energy. We can therefore ignore correlations of different spins and consider only the $j = k$ contribution, giving

$$\delta W_{\text{eff}}^{FQ} \;\approx\; \cos(\theta)\Big[\sum_j \kappa_j^2\langle\langle(S_3^{(j)})^2\rangle\rangle\Big]^{1/2}. \tag{26}$$

Considering a Nitrogen-to-NV conversion of $\eta \sim 5\%$, the sum over the impurities in the ensemble will be dominated by the Nitrogen impurities. An estimate of this expression can be obtained by noting that the coupling to the Nitrogen spin κ is comparable to the coupling g to the NVs. The difference between this sum and the sum leading to the collective coupling constant G only differ by the number of terms in the sum and we obtain

$$\delta W_{\text{eff}}^{FQ} \sim \frac{\cos(\theta)}{\sqrt{\eta}} G. \tag{27}$$

A more accurate treatment taking into account the full spatial distribution of the field over a sample of the dimensions considered in the text only changes the estimate by a factor of less than 2. Eq. (27) quantifies how close to the degeneracy point we need to be so that the dephasing of the FQ induced by the bath of spins is negligible. In particular, from (27) we derive that we can safely neglect this dephasing on a time scale $1/G$ for a conversion efficiency $\eta = 0.05$ if $\cos(\theta) \lesssim 0.01$, for which $\delta W_{\text{eff}}^{FQ}/G \lesssim 0.05$.

Next we turn to the rapidly oscillating part of the coupling, described by the term containing $\hat{\tau}_1$, and assume that we are near the degeneracy point, that is $\sin\theta \approx 1$. We will now argue that since $\hat{\tau}_1$ is oscillating rapidly, the slowly-varying dynamics of the spin bath will only have a very weak influence on the FQ. Specifically the spin bath can influence the FQ either through direct transitions to the spin bath or through a slow dephasing. The direct coupling can be excluded by noting that the coupling constant to collective excitations of the bath is limited by $G/\sqrt{\eta}$, but for reasonably low applied magnetic fields, $B^{\text{ext}} \lesssim 10$ mT, the detuning $\omega - \Delta_j$ will be of the order of the NV zero-field splitting (a few GHz). The probability to transfer the excitation is $\sim G^2/(\omega - \Delta_j)^2\eta$, and can thus safely be neglected since $G \sim$ MHz. The dephasing of the FQ caused by the bath can be estimated by calculating the energy shift of the FQ in second order perturbation theory. Assuming that apart from the free precession, the spins change slowly on a time scale set by $\omega - \Delta_j$, the effective interaction is given by

$$H_{\text{eff}} = \hat{\tau}_3 \sum_l \frac{W_z^{\text{FQ}}(\vec{r}_l)^2}{4\omega} + \frac{W_\perp^{\text{FQ}}(\vec{r}_l)^2\omega}{4(\omega^2 - \Delta_j^2)} - \frac{W_\perp^{\text{FQ}}(\vec{r}_l)^2\Delta_j}{2(\omega^2 - \Delta_j^2)} S_z^{(j)}(t). \tag{28}$$

Here the first two terms are independent of the state of the impurities, and therefore merely lead to a mean shift of the energy, which can be compensated by a magnetic field. It is thus only the last term which leads to dephasing. Again, we can estimate the root mean square value of this term by neglecting the correlations among the impurities and we find a typical energy shift

$$\Delta E \sim \frac{G}{\sqrt{\eta}} \frac{g\Delta_j}{\omega^2 - \Delta_j^2}, \tag{29}$$

which we can safely neglect since the single-spin coupling $g \sim$ kHz is much smaller than any other quantities in the system.

A remaining problem is the influence of the NVs with different orientations. The interaction with these centers is similar to that with the resonant subensemble and of order $\sim G$. We assume that we select a single orientation by applying a magnetic field with a component along the axis of the NV center. This will shift all other centers out of resonance by an amount $\delta\omega \sim g_e\mu_B B^{\text{ext}}$, such that the resulting error can be estimated to be $G^2/\delta\omega^2$. For a reasonable applied field, e.g., $B^{\text{ext}} \sim 10$ mT, this error is negligible.

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