Observation of dark states in a superconductor diamond quantum hybrid system

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The hybridization of distinct quantum systems has opened new avenues to exploit the best properties of these individual systems. Superconducting circuits and electron spin ensembles are one such example. Strong coupling and the coherent transfer and storage of quantum information has been achieved with nitrogen vacancy centres in diamond. Recently, we have observed a remarkably sharp resonance ($|B| \approx 1\text{MHz}$) at 2.878 GHz in the spectrum of flux qubit negatively charged nitrogen vacancy diamond hybrid quantum system under zero external magnetic field. This width is much narrower than that of both the flux qubit and spin ensemble. Here we show that this resonance is evidence of a collective dark state in the ensemble, which is coherently driven by the superposition of clockwise and counter-clockwise macroscopic persistent supercurrents flowing in the flux qubit. The collective dark state is a unique physical system and could provide a long-lived quantum memory.

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The last decade has seen that our understanding of the principles of quantum mechanics lead to the development of technologies underpinned by it. We are at the stage where real engineering of quantum devices is taking place. Hybridization of distinct quantum systems allows us to exploit the best properties of these individual systems without their weaknesses (refs 1–3 and references within). One well-known example of such hybridized quantum systems are superconducting circuits coupled to electron spin ensembles. These hybridized systems have seen many fundamental experiments taking place including for instance, the strong coupling and the coherent transfer and storage of quantum information between the superconducting circuit and an electron spin ensemble from nitrogen vacancy (NV) centres in diamond. Nitrogen vacancy centres were generally considered as an ensemble of two-level systems. Our characterization of this hybrid system has hinted at a fine spectroscopy feature near the 2.878-GHz resonance but these have not been really explored. In this work we investigate its nature.

Results

Characterization. Our characterization of this hybrid system begins with spectroscopic measurements (shown in Fig. 2) of two physically different ensembles (A and B) with NV centres density of $\sim 4.7 \times 10^{17}$ cm$^{-3}$ and $1.1 \times 10^{18}$ cm$^{-3}$, respectively. Both our samples (Fig. 2a,c) show significant hybridization near the optimal point $\Phi_{qb} \sim 1.5\Phi_0$, clearly indicating strong coupling. The splitting between the hybridized peaks is 28 MHz and 42 MHz, respectively. The energy spectrum shows a narrow peak located in the middle of the avoid crossing structure (Fig. 2a,c).

Figure 1 | Illustration of a hybrid-coupled system. The hybrid system is composed of a gap-tuneable flux qubit and an ensemble of NV centres in diamond. The flux qubit is composed of four junctions (represented by the shaded circles). Input lines to adjust and drive the qubit are shown coming from the edges. The ensemble (light red coloured object) sits on top of the qubit.

The model. To establish the mechanism causing this narrow central peak, we need to consider a Hamiltonian model of a driven flux qubit coupled to an ensemble of NV centres. The full description is presented in Supplementary Note 1, but this can be significantly simplified when we consider that our flux qubit will only drive a few excitations in the ensemble. In such a situation we can model the spin ensemble as a number of harmonic oscillators and so write the Hamiltonian after a rotating wave approximation as

$$H = \hbar \sqrt{\epsilon^2 + \Delta^2} \sigma^+ \sigma^- + \hbar D \hat{b}^+ \hat{b} + \hbar D \hat{d}^+ \hat{d}$$

where $\sigma^\pm$ are the raising/lowering operators of the flux qubit in the energy basis $|0\rangle_{qb}$ and $|1\rangle_{qb}$. $\Delta$ represents the flux qubit’s tunable splitting while $\epsilon$ is its energy bias. The ensembles are represented as two harmonic oscillators $D \hat{b}^+ \hat{b}$ and $D \hat{d}^+ \hat{d}$ where $\Delta = \hbar D / \sqrt{2}$ and $\Omega = \hbar D / \sqrt{2}$. The term $\hbar \Delta \sigma^+ \sigma^-$ couples the two harmonic oscillators together allowing exchange of excitations between them. It is in effect a direct current (DC) driving term whose magnitude can be varied by detuning the flux qubit $D \hat{b}^+ \hat{d}^+ \hat{d}$ harmonic oscillators energy levels. The term $J$ arises from both strain and Zeeman effects. The flux qubit couples only to one mode of this ensemble, which we describe by $G \sigma^+ \sigma^-$. It represents the flux qubit driving the ensemble with a single excitation. The coupling strength $G$ of this interaction is given by $G = \sqrt{N g_{qb} B_{ qb} / \sqrt{\epsilon^2 + \Delta^2}}$, where $N$ is the number of centres in the coupled ensemble, $g_{qb} B_{ qb}$ is the perpendicular component to the NV axis of the magnetic field generated by the flux qubit. This coupling strength can be tuned by moving the flux qubit off-resonance with the ensemble (non-zero $\epsilon$).

Energy levels and their associated states. Tuning the energy gap of the flux qubit ($\Delta$) to $D$, we can straightforwardly determine the eigen energies and eigenstates of the hybrid system using equation (1). We choose for simplicity $\theta = \pi/2$ to illustrate this (in Supplementary Note 1, we relax this constraint). The eigen energies are plotted against $J$ in Fig. 3a. The lowest energy level labelled by $L_0$ corresponds to the ground state $|0\rangle_{qb} |0\rangle_{ens}$ where $|0\rangle_{ens} = |0\rangle_{NV_1} |0\rangle_{NV_2} \cdots |0\rangle_{NV_n}$. The energy level $L_++L_-$ (L+) corresponds to the single-excitation dressed bright states

$$|B^{(+)\pm}\rangle = \frac{1}{\sqrt{2}} |0\rangle_{qb} |0\rangle_{ens} \pm \frac{i}{\sqrt{2}} |\pm\rangle \propto \frac{1}{\sqrt{2}} |0\rangle_{qb} |0\rangle_{ens}.$$  

The transitions between $|0\rangle_{qb} |0\rangle_{ens} \leftrightarrow |B^{(+)\pm}\rangle$ corresponds to the two broader peaks shown in Fig. 2a,c near $\Phi_{qb} \sim 1.5\Phi_0$. These transitions are being directly driven.
avoided crossing structure. In the NV centre peak part is illustrated. The sharp peak shows a small flux bias as 28 MHz for sample A and 42 MHz for sample B. Both spectroscopic measurements superconducting circuit with the (001) surface facing the flux qubit (a, b). The small in-plane magnetic field splits the degenerate excited states $|\psi\rangle$. The normalized switching probability of the central peak is shown for several small in-plane magnetic fields (our diamond crystal was attached on top of the ensemble as a function of the temperature the zero-field splitting increases to $2.878$ GHz. There are many subradiant states $|\psi\rangle$ that can be broadened significantly by strain variation and inhomogenous broadening (light red). In (b), the energy-level diagram for the superconducting flux qubit—NV $^−$ centre ensemble is given, where the bright states are ($|B\rangle$) and the dark state ($|D\rangle$) energy is seen at 2.878 GHz. There are many subradiant states $|\psi\rangle$ that can be broadened significantly by strain variation and inhomogenous broadening (light red). In (b), the optically detected magnetic resonance spectrum of the NV $^−$ ensemble is shown. For the bright states ($|B\rangle$) the zero-field splitting increases to 2.878 GHz while at the low temperature the zero-field splitting increases to $D/2\pi \sim 2.878$ GHz while at the low temperature the zero-field splitting increases to $D/2\pi \sim 2.878$ GHz.
Next in our diagram are energy levels associated with the subradiant states. These are composed of states having only a single excitation in the ensemble. These states have no \( |1\_\mathrm{qb}\rangle_\mathrm{ens} \) component and so cannot be detected using flux qubit spectroscopic measurements. For finite \( J \), these subradiant states are split either above or below the 2.878-GHz resonance (Fig. 3a) and in the limit \( g_\mathrm{e}\mu_\mathrm{B}B_\mathrm{qb} \ll J \) be written as the spin waves \( (k>0) \)

\[
|S\rangle^{R(k)} \approx \frac{1}{\sqrt{N}} \left( \sum_{j=1}^{N} e^{2\pi(j-1)k/N} |A_\pm \rangle_\mathrm{NV} \langle 0 | \right) |0 \rangle_\mathrm{ens}
\]

with an energy of \( E_{S\rangle^{R(k)}} \sim D \pm J \). These spin waves are orthogonal to the \( k=0 \) mode and so do not couple to the flux qubit.

Finally, in terms of single excitation energy levels is the eigenstate at 2.878 GHz, which corresponds to

\[
|D\rangle = \left( \frac{G}{\sqrt{G^2 + J^2}} d^\dagger - \frac{iJ}{\sqrt{G^2 + J^2}} \sigma^+ \right) |0 \rangle_\mathrm{qb} |0 \rangle_\mathrm{ens}.
\]

This state can further be written in the form \( |D\rangle = (G/(\sqrt{G^2 + J^2})) |0 \rangle_\mathrm{qb} |W^{(k=0)} \rangle - (ij/(\sqrt{G^2 + J^2})) |1 \rangle_\mathrm{qb} |0 \rangle_\mathrm{ens} \) where \( |W^{(k=0)} \rangle = (1/\sqrt{N}) \left( \sum_{j=1}^{N} |A_\pm \rangle_\mathrm{NV} \langle 0 | \right) |0 \rangle_\mathrm{ens} \). The form of dark state \( |D\rangle \) is important because it contains both components of an excitation in the flux qubit \( |1\_\mathrm{qb}\rangle_\mathrm{ens} \) and the ensemble. The component \( |1\_\mathrm{qb}\rangle_\mathrm{ens} \) implies that a signal will be seen at 2.878 GHz in the spectroscopic measurements (observed in Fig. 2). Next, we tune the flux qubit away from the resonance point using the energy bias \( \epsilon \), we would expect to see a slight quadratic \( \Phi_\mathrm{qb} \) dependence in the observed resonance frequency (Fig. 2d) owing to the small \( |1\_\mathrm{qb}\rangle_\mathrm{ens} \) component in \( |D\rangle \). When we apply an in-plane magnetic field, we effectively strengthen the driving field \( J \) and so we increase the contribution of the \( |1\_\mathrm{qb}\rangle_\mathrm{ens} \) component to \( |D\rangle \). The population of this component is proportional to \( J \) and so we should expect to observe a quadratic dependence in the observed line width (observed in Fig. 2e). We can also understand the power-dependent measurements in (Fig. 2b). Given that the \( J \) field is not changing during these experiments, we would expect the area of the central peak to decrease at approximately the same linear rate as the outer broader peaks as the pump field driving the flux qubit decreases. This is clearly what is seen even at very low pump powers (Fig. 4) and clearly shows this is not a two-photon process. The amplitude of the centre peak being higher than the outer peaks is also an indicator.

**Coherence properties.** The line width of these peaks depends on the coherence properties of our hybrid system. Surprisingly, the line width of the central peak, 1.0 MHz for sample A and 2.2 MHz for sample B, seems quite narrow compared with the usual coherence properties of the system. The measured flux qubit coherence properties at the optimal point are \( T_1 \approx 300 \) ns (relaxation), \( T_2 \approx 150 \) ns (dephasing)\(^{16}\). However, in the spectroscopic measurements, power broadening of \( \sim 4 \) MHz makes these much worse. Further, the coherence properties also decrease rapidly as we move away from the optimal point. As \( |D\rangle \) is mainly composed of the state \( |0 \rangle_\mathrm{qb} |W^{(k=0)} \rangle \), it is now easy to understand why the dark states’ line width does not change much as the flux qubit moves away from the optimal point (by increasing \( \epsilon \)). Although the flux qubit line width increases, it has a minimal contribution to \( |D\rangle \) (proportionally to \( J^2/(G^2 + J^2) \)). Next, the electron spin ensemble \( T_1^\mathrm{NV} \) has been measured to be \( \sim 40 \) s (ref. 15); however, \( T_1^\mathrm{NV} \) is much shorter (tens of nanoseconds) in our case\(^{17}\). Inhomogenous broadening due to random magnetic fields caused by nitrogen (P1) and other centres in the diamond sample A (B) is likely to be the primary cause of this and it has been derived from the fitting to the spectrum data at \( \sim 3.1 \) MHz (7.8 MHz; see Supplementary Notes 1 and 2). The distribution of the strain field is \( \sim 4.4 \) MHz (7.6 MHz) while fluctuations in the zero-field splitting \( D \) are 0.45 MHz (0.75 MHz). An always-on hyperfine coupling to thermalized nitrogen nuclear spins is \( \sim 2.3 \) MHz (ref. 17). These values are consistent with the previous experimental results\(^{16,17}\). The measured line width of the central peak is narrower than all these (apart from \( D \) fluctuations). Our modelling of these system indicates that this central peaks is not limited by the effects of the strain field or inhomogenous broadening. Instead, it seems currently limited by qubit power broadening and zero-field fluctuations.

**Discussion**

The energy diagram (Fig. 3a) illustrates why this collective state has such a narrow line width (and hence long lifetime). The state \( |D\rangle \) would couple with other subradiant states and dephase quickly if a significant number of them existed near 2.878 GHz.
Fortunately, the existence of inhomogenous broadening and the strain field induces an energy shift from 2.878 GHz on the subradiant states leaving this collective dark state well isolated.

Since the dephasing process is induced by low-frequency noise, such an energy gap can suppress unwanted transitions between \( |\pm 1\rangle \) and environmental states \( |SR\rangle \). This is quite different from what happens with general atom-based dark states. We have simulated the effects of strain variation and inhomogeneous broadening using the full Hamiltonian (detailed in Supplementary Note 1) along with decoherence on the flux qubit, enabling us to confirm our analytical results described above. Moreover, the room temperature detected magnetic resonance spectrum measurements\(^\text{23-25}\) support our interpretation that our subradiant states’ energies are sparsely occupied near the central peak in Fig. 3b. At room temperature, this reduction in population is at 2.87 GHz but shifts to 2.878 GHz at mK temperatures\(^\text{26}\). Thus, we have strong evidence for the existence of this collective dark state.

Generally with dark states, if one turns off the control field, the dark state disappears. In our situation, here we cannot turn off the DC field but by applying a large magnetic field we can split the \( |\pm 1\rangle \) levels by nearly 100 MHz. The coupling field is then effectively off-resonance because of the energy splitting. Experimentally, only double peaks showing the hybridization are seen either resonant with the \( |0\rangle \leftrightarrow |1\rangle + |2\rangle \) transition or the \( |0\rangle \leftrightarrow |1\rangle - |2\rangle \) transition. No fine feature is seen at 2.878 GHz. This further evidence of our collective dark state (in Supplementary Note 3 where we show how the observed phenomenon is inconsistent with an ensemble of two-level systems).

To summarize, we have experimentally observed a narrow peak in the avoided crossing structure of the energy spectrum in the flux qubit NV\( ^-\) diamond hybrid system. Our analysis indicates that this is the signature of a collective dark state, which should be long lived and could provide an alternative approach for quantum memories. The lifetime of this peak can further be increased in the near future with higher quality diamond samples that possess a smaller number of nitrogen centres and that have reduced homogenous broadening and zero-field fluctuations. This can be partially achieved by using electron beam irradiation to form the NV\(^-\) centres.

Methods

Measurements. Spectroscopic measurements were achieved by initializing the hybrid system in its ground state through relaxation. A 1-µs-long microwave pulse was applied to the flux qubit followed immediately by a current pulse to the DC-superconducting quantum interference device for detecting the state of the qubit. By repeating the measurement several thousand times, we obtain the switching probability \( P_{\text{sw}} \), which is proportional to the qubit excited-state population.

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Author contributions

All authors contributed extensively to the work presented in this paper. X.Z., R.A. carried out measurements and a analysis on the coupled flux qubit/NV\(^-\) ensemble. T.S., N.M. and K.S. prepared and characterized the NV\(^-\) diamond crystals. X.Z. and S.S. designed and fabricated the flux qubit and associated devices while S.S. and K.K. designed and developed the flux qubit measurement system. Y.M., K.N. and W.J.M.
provided theoretical support and analysis. X.Z., W.J.M., Y.M. and S.S. wrote the
manuscript, with feedback from all authors. K.S., W.J.M. and S.S. supervised the project.

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