Coherent coupling of a superconducting flux-qubit to an electron spin ensemble in diamond

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Electron-spin nitrogen-vacancy color centers in diamond are a natural candidate to act as a quantum memory for superconducting qubits because of their large collective coupling and long coherence times. We report here the first demonstration of strong coupling and coherent exchange of a single quantum of energy between a flux-qubit and an ensemble of nitrogen-vacancy color centers.

During the last decade, research into superconducting quantum bits (qubits) based on Josephson junctions has made rapid progress [1]. Many foundational experiments have been performed [2–10] and superconducting qubits are now considered one of the most promising systems for quantum information processing. However, the experimentally reported coherence times are likely to be insufficient for future large-scale quantum computation. A natural solution is a dedicated engineered quantum memory based on atomic and molecular systems. Since macroscopic quantum coherence was first demonstrated in Josephson junction circuits [2], the question of whether or not coherent quantum coupling between a single macroscopic artificial atom and an ensemble of natural atoms or molecules is possible has attracted significant attention [11–13]. In this Letter, we present for the first time evidence of coherent strong coupling between a single macroscopic superconducting artificial atom (a flux qubit) and an ensemble of electron-spin nitrogen-vacancy color centers (NV− centers) in diamond. Furthermore, we have observed coherent exchange of a single quantum of energy between a flux qubit and a macroscopic ensemble consisting of ∼3 × 107 of NV− centers. This provides a foundation for future quantum memories and hybrid devices coupling microwave and optical systems.

With the early successes of single atom quantum state manipulation [14], research in quantum information processing with atomic and solid-state systems has progressed largely in a separate fashion. In recent years, significant effort has been devoted to coupling atomic and molecular systems to solid-state qubits to form hybrid quantum devices [11–13]. Hybrid devices involving the integration of an atomic system with a superconducting transmission line resonator have been realized [16–18]. Such schemes have the potential to couple superconducting solid-state qubits to optical fields via atomic systems, thus enabling quantum media conversion. The coupling strength g of an individual atomic system to one electromagnetic mode in a resonator circuit is usually too small for the coherent exchange of quantum information. However, the coupling strength of an ensemble of N such atomic systems will be enhanced by a factor of √N [19], allowing one to reach the strong coupling regime (g√N ≫ κ, γ, where κ and γ are the damping rates of resonator circuit and atomic system).

Of the many possible hybrid systems, coupling a flux-qubit to an (NV−) center in diamond is particularly appealing. Firstly, the magnetic coupling between a flux-qubit and a single NV− center can be three orders of magnitude larger than that associated with a superconducting transmission line resonator [14]. Second, the ground state of an NV− center is a spin 1 triplet due to its C3v symmetry (Figure 1b). The S = 1 spin triplet |ms = 0⟩ state is separated by 2.88 GHz from the near degenerate excited states |ms = ±1⟩ under zero magnetic field (Figure 1c). This energy separation is ideal for a gap tunable flux-qubit to be brought on and off resonance with it.

In this Letter, we report on the first observation of vacuum Rabi oscillations between a flux-qubit and an ensemble of approximately three million NV− centers in diamond. This demonstrates strong coherent coupling between two dissimilar quantum systems with an effective collective coupling constant of gens ∼ 70 MHz.

We begin by describing our experimental setup as depicted in (Figure 1). An NV− diamond sample was prepared by ion implantation of 12C2+ at 700 keV under high vacuum into high-pressure, high-temperature (HPHT)-synthesized type Ib (001) surface orientation single crystal diamond. The 12C2+ ions, with a dose condition of 3 × 1013 cm−2, were stopped at a depth of 600±50 nm. This generated on the order of 5 × 1018 cm−3 vacancies over a depth of ∼0.7 μm. After implantation, the crystals were annealed at 900°C under vacuum for 3 hours. This high dose carbon implantation method en-
the zero-field splitting (2.878 GHz), $E$ the strain-induced splitting (<1 MHz), the N-V Landé factor $g_{NV} = 2$, and $\mu_B = 14$ MHz/nT. The last term represents the Zeeman splitting, which is negligible in our case as the magnetic field applied perpendicular to the surface of the chip to prepare the flux qubit is less than 0.1 mT.

A diamond crystal was glued on top of the superconducting circuit with the $^{13}$C implanted surface facing the flux-qubit (Figure 1a). We used a gap tunable flux-qubit (Figure 1d, 1e) where the smallest junction of the three Josephson junction qubit is replaced by a low inductance superconducting quantum interference device (dc-SQUID) loop (the magenta loop in Figure 1e). The flux qubit - NV ensemble coupled system is measured by the qubit state using a built-in dc-SQUID (the biggest ZPL

FIG. 1. Experimental set-up of an NV-diamond sample attached to a flux-qubit system. (a) A diamond crystal is glued on top of a flux-qubit and its superconducting circuits (under the red box) with the diamonds $^{12}$C implanted (001) surface facing the chip. The distance between the flux-qubit and surface of the diamond crystal is carefully adjusted to be less than a micrometer using 100 nm height mesa structure on the diamond surface, a circle adjacent to the red square, and the optical interference pattern (Newton’s ring). (b) A sketch of a NV color center of diamond with its vacancy (V) and nitrogen (N) atom, as well neighboring carbon atoms. Four equivalent NV-axis exist depicted in purple color dangling bonds, all making the same angle with [001] direction to which the magnetic field generated by the flux-qubit points. (c) Energy diagram of the NV center, with the spin triplet $|m_s = 0\rangle$ ground state separated by 2.88 GHz from the degenerated $|m_s = \pm 1\rangle$ excited states under zero magnetic field. (d) Optical micrograph and the circuit scheme of the aluminum made flux-qubit, the magnified view of the chip under the red box region shown in Fig1a. The central M shaped circuit contains a flux-qubit and a SQUID detector. Two high-bandwidth (20 GHz) MW-control lines located both sides of the qubit circuit. (e) The H-shaped gap tunable flux-qubit and the edge-shared SQUID used as a switching qubit state detector. The flux-qubit contains two loops, the main loop (blue) and the $\alpha$-control loop (magenta) which controls tunneling energy gap of the flux-qubit. Magnified view of Josephson junctions are also shown. The mutual inductance of control line-1 to the $\alpha$-loop and main loop are 90 fH and 256 fH, and those of control line-2 are 0.5 fH and 549 fH. The magnetic flux penetrating these two loops can be controlled in ns time scale by applying synchronized current pulses to these control lines in situ.

Hances the yield of generated NV$^-\,$ centers [20]. Photoluminescence (PL) optical spectroscopy (shown in Figure 2) established that NV$^-\,$ centers were generated with a density of $\sim 1.1 \times 10^{18}$ cm$^{-3}$ over a 1 nm depth. We can describe the ground state of a single NV$^-\,$ center by the Hamiltonian [21],

$$H_{NV} = hD S^2_{x} + hE(S^2_{y} - S^2_{x}) + h g_{NV} \mu_B B \cdot S,$$  

(1)

where $S_x, S_y, S_z$ are the usual Pauli spin 1 operators, $D$ the Photoluminescence spectra of (a) ensembles of color centers in the highly carbon implanted sample and (b) a single NV- center in pure diamond at room temperature. Their signal intensities are normalized for the comparison of the spectra. In the spectrum of the single NV center, the contributions of phonon Raman scattering from bulk diamond at 573 and between 600 and 620 nm are subtracted [20]. The zero phonon line of NV- at 637 nm [20,30] is clearly observed in both spectra. Broad spectrum of phonon replicas is also very similar to each other and to the reported ones [20,30]. These indicate that the NV- center is produced as a major color center in the highly carbon implanted sample. The signal intensity of the ensemble is about $6.5 \times 10^4$ times stronger than that of the single one. From this result, the concentration of the NV- center in the highly carbon implanted sample was estimated to be $1.1 \times 10^{18}$ cm$^{-3}$.
nA is the persistent current in the qubit, $\Phi_{\text{ex}}$ is the external flux threading the qubit loop, and $\Phi_0 = h/2e$ is the flux quantum), and $\Delta$ is the tunnel splitting. The energy splitting of the gap tunable flux-qubit is

$$hF = h\sqrt{\epsilon^2 + \Delta^2}$$

where $\epsilon$ and $\Delta$ can be controlled independently by the external magnetic flux threading the two loops. This type of flux-qubit can be tuned into resonance with an NV$^-$ ensemble in situ at a base temperature of $\sim$12 mK while keeping the qubit at its optimum flux bias (degeneracy point). The total Hamiltonian of the coupled system is

$$H = \frac{h}{2} (\Delta \sigma_z + \epsilon \sigma_x) + \hbar \sum_i [DS_{z,i}^2 + E(S_{x,i}^2 - S_{y,i}^2)] + \hbar \sum_i g_i \sigma_z S_{x,i},$$

(3)

where $i$ runs over the NV$^-$ centers which couple to the flux qubit. The corresponding coupling constant can be estimated using the Biot-Savart law at $g_i \sim 8.8$ kHz. In our situation here, the $|\pm 1\rangle_i$ states of the NV$^-$ electronic spin are near degenerate and so our flux-qubit couples to both the $|0\rangle_i = |1\rangle_i$ and $|0\rangle_i = |-1\rangle_i$ transitions. This results in an effective coupling constant $\sqrt{2}g_i$ larger that generally anticipated.

From the spectroscopic measurements, a clear anti-crossing was observed (Figure 3a) near the degeneracy point of the flux-qubit, while no gap was observed in the same flux-qubit prior to the mounting of the ensemble (inset in Figure 3a). We also note a narrow resonance at 2.878 GHz of less than 1 MHz width near these anti-crossings. This can be ascribed to the near degenerate excited states of the NV$^-$ ensemble and so indicates a strain-induced zero-field splitting coefficient $E$ of less than 1 MHz. From the fine scan spectroscopy shown in (Figure 3b), a vacuum Rabi splitting near $g_{\text{ens}} \sim 70$ MHz was clearly observed confirming strong coupling between the flux-qubit and the NV$^-$ ensemble. Next from the measured vacuum Rabi splitting and our calculated value of $g_i$ we can estimate the number of NV$^-$ centers in the ensemble at $N = g_{\text{ens}}^2/2g_i^2 \approx 3.2 \times 10^7$, where the factor of 2 in the denominator is due to the two-fold degeneracy of the excited $|\pm 1\rangle_i$ states of an NV$^-$ center. This estimate is consistent with the density of NV$^-$ centers measured by PL spectroscopy in the whole sample ($1.1 \times 10^{18}$ cm$^{-3}$) multiplied by the volume of centers coupling to the flux qubit (area 40 $\mu$m$^2$ x effective thickness 0.7 $\mu$m). The PL spectroscopy approach gives the number of coupled centers as $\approx 3.1 \times 10^7$.

Next, we investigated the dynamics of our system in the time domain using a similar measurement cycle to that performed in qubit-LC resonator coupled systems [24]. We first excited the flux-qubit and then brought it into resonance with the NV$^-$ ensemble. Single energy quantum exchange between the flux-qubit and NV$^-$ ensemble at resonance manifests itself as the vacuum Rabi oscillations

$$|1\rangle_{\text{qb}}|0\rangle_{\text{ens}} \Rightarrow |0\rangle_{\text{qb}}|1\rangle_{\text{ens}}$$

(4)

where $|1\rangle_{\text{ens}} = \frac{1}{\sqrt{N}} \sum_i S_{+,i} |00\cdots 0\rangle$, with $S_{+,i} = |1\rangle_i \langle 0|_i + |-1\rangle_i \langle 0|_i$ being the raising operator of the $i$-th NV$^-$ spin to both the $|\pm 1\rangle$ states. (Figure 4a) clearly shows vacuum Rabi oscillations between the flux-qubit.
and ensemble of electronic spins at the 2.878 GHz resonance. The decay time of the oscillations however is approximately 20 ns. This is much shorter than the relaxation time of both the flux-qubit ($T_{1,qb} \sim 150$ ns) and the NV$^-$ ensemble ($T_{1,NV} \gg 10$µs). As we tune the flux-qubit away from the 2.878 GHz resonance, the decay time associated with vacuum Rabi measurement becomes significantly longer (Figure 4b). From these results, one must conclude that a source of strong dephasing of unknown origin exists in the system near resonance. There are several likely sources. The most probable is a large electron spin $\frac{1}{2}$ bath from the P1 (nitrogen atom substituting a carbon atom) centers present in our HPHT Ib-type diamond crystal used to prepare the NV$^-$ ensemble. In our situation, where there is no external magnetic field, the NV$^-$ centers and P1 centers naturally couple. Hanson et. al [29] have shown an enhanced decay may result. The P1 center issue can be eliminated to a large extent by applying an external magnetic field to split the $|\pm1\rangle$ NV$^-$ states. A 1 mT field could split these by approximately 15 MHz, detuning the P1 centers and thus significantly improving the dephasing time of the coupled system. We can also decrease the number of P1 centers in the sample (from 100 ppm to 1 ppm) by using different synthesized diamond crystals. In addition, by using non HPHT Ib-type crystals we can remove the effect of other natural defects that may be present. Finally there is also a strong hyperfine interaction ($\sim$100 MHz) between the NV$^-$ electron spin and $^{13}$C nuclear spins. Without the nuclear spins being initially polarized, unwanted dephasing will result. By polarizing the nuclear spins this source of dephasing can be removed. This should allow us to observe vacuum Rabi oscillations where we are limited by $T_2$ of the flux-qubit.

In conclusion, we have experimentally demonstrated strong coherent coupling between a flux-qubit and an ensemble of nitrogen-vacancy color centers in single crystal diamond. Furthermore, we have observed, via vacuum Rabi oscillations, the coherent exchange (transfer) of a single quantum of energy. This is the first step towards the realization of a long lived quantum memory for condensed matter systems with an additional potential future application as an interface between the microwave and optical domains.

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