Coupling spin ‘clock states’ to superconducting circuits

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A central goal in quantum technologies is to maximize $G T_2$, where $G$ stands for the rate at which each qubit can be coherently driven and $T_2$ is the qubit’s phase coherence time. This is challenging, as increasing $G$ (e.g. by coupling the qubit more strongly to external stimuli) often leads to deleterious effects on $T_2$. Here, we study a physical situation in which both $G$ and $T_2$ can be simultaneously optimized. We measure the coupling to microwave superconducting coplanar waveguides of pure (i.e. non magnetically diluted) crystals of HoW$_{10}$ magnetic clusters, which show level anticrossings, or spin clock transitions, at equidistant magnetic fields. The absorption lines give a complete picture of the magnetic energy level scheme and, in particular, confirm the existence of such clock transitions. The quantitative analysis of the microwave transmission allows monitoring the overlap between spin wave functions and gives information about their coupling to the environment and to the propagating photons. The formation of quantum superpositions of spin-up and spin-down states at the clock transitions allows simultaneously maximizing the spin-photon coupling and minimizing environmental spin perturbations. Using the same experimental device, we also explore the coupling of these qubits to a 11.7 GHz cavity mode, arising from a nonperfect microwave propagation at the chip boundaries and find a collective spin to single photon coupling $G_N \simeq 100$ MHz. The engineering of spin states in molecular systems offers a promising strategy to combine sizeable photon-mediated interactions, thus scalability, with a sufficient isolation from unwanted magnetic noise sources.

INTRODUCTION

Spins embedded in solid hosts are one of the simplest and most natural choices to realize qubits, the building blocks of quantum technologies.$^{12}$ Their quantized spin projections can encode the logic qubit states whereas operations between them can be induced via the application of microwave radiation pulses, using well-established magnetic resonance protocols. Among the different candidates, chemically designed magnetic molecules stand out for several reasons.$^3$ Besides being microscopic, thus fully reproducible and intrinsically quantum, they represent the smallest structure that remains tuneable. The ability to modify the relevant properties by properly choosing the molecular composition and structure allows engineering the qubit spin states and energies,$^{23}$ and enables scaling up computational resources within each molecule (e.g. by accommodating several different magnetic atoms in exquisitely defined coordinations,$^{24,10}$ or by making use of multiple internal spin states$^{11,14}$).

This approach faces the challenge of wiring-up different molecules into a scalable architecture. A promising technology is to use microwave photons, trapped in on-chip superconducting resonators to mediate molecule-molecule interactions.$^{15,16}$ Working with high-spin molecules helps maximizing the spin-photon coupling, as required for such applications.$^{13}$ However, it also tends to enhance decoherence, as their interactions with fluctuating hyperfine and dipolar magnetic fields also become stronger.$^{20,21}$

A way to reconcile a high degree of scalability, i.e. a high density of spin qubits, with sufficient isolation from noise sources is to encode the qubit states in a subspace that is robust against magnetic field fluctuations. In the case of molecular systems, this can be achieved by associating 0 and 1 to superpositions of spin-up and spin-down states that arise at ”spin-clock” transitions, relatively insensitive to magnetic field changes. A paradigmatic example is provided by the sodium salt of the cluster [Ho(W$_2$O$_{18}$)$_2$]$^{9−}$, hereafter referred to as HoW$_{10}$, which consists of a single Ho$^{3+}$ ion encapsulated by polyoxometalate moieties (Fig. 1). Its fourth-fold coordination symmetry gives rise to fourth-order off-diagonal terms in the spin Hamiltonian that strongly mix the $m_J=\pm 4$ projections of the ground electronic spin doublet. The large quantum tunneling gap $\Delta \simeq 9.1$ GHz generated by such terms, combined with the hyperfine interaction with the $I = 7/2$ spin of the Ho nucleus, gives rise to a set of level anti-crossings at which the spin coherence time $T_2$ becomes maximum.$^5$

In this work, we explore the coupling of single crystals of HoW$_{10}$ molecules to photons propagating via superconducting co-planar waveguides. These experiments provide a method to investigate in detail how
The superconducting coplanar waveguides employed in this work consist of a 600µm wide central transmission line separated from two ground planes by 300µm wide gaps. They are fabricated by optical lithography of 100 nm thick Nb films deposited by sputtering onto single-crystalline sapphire substrates. The size of the central line and its meander shape were designed in order to best match the dimensions (ca. 10 × 4 × 1 mm³) of the large HoW₁₀ single crystals used in the experiments (an optical microscopy image of a chip can be seen in Fig. 3 of the SI).

Microwave transmission experiments were performed, at 4.2 K and between 0.01 GHz and 14 GHz, by immersing the chips in a liquid He cryostat and connecting their input and output lines to a vector network analyzer that measures the transmission, $S_{12}$ and $S_{21}$, and reflection, $S_{11}$ and $S_{22}$, coefficients. A dc magnetic field, whose amplitude $\mu_0 H$ ranged from 0 to 0.4 T, was applied along different directions within the plane of the device by means of a 1T×1T×9T superconducting vector magnet whose three axes define X, Y and Z of the laboratory frame. The crystals were attached onto the transmission line with apiezon N grease. The crystallographic b axis was oriented nearly parallel to Z and to the axis of the device, which corresponds also to the orientation of the microwave magnetic field seen by the majority of molecules in the crystal (Fig. 1 and Fig. 3 of the SI).

In order to compensate for the decay of the waveguide transmission with increasing frequency and to enhance the contrast of those effects associated with its coupling to the spins, $S_{21}$ and $S_{12}$ were normalized by a method similar to that proposed in reference [24]. The normalized transmission $t$ at magnetic field $H_1$ and frequency $\omega_1$ is given by

$$
t(H_1, \omega_1) = \frac{S_{21}(H_1, \omega_1) - S_{21}(H_2, \omega_1)}{S_{21}^{(0)}(\omega_1)}$$

where $H_2 > H_1$ and $S_{21}^{(0)}$ is the transmission of the ‘empty’ transmission line. In practice, $S_{21}^{(0)}$ is measured at a magnetic field for which all spin excitations are outside the accessible frequency region. For $H_2$ very close to $H_1$ (i.e. closer than the magnetic field width of a given absorption line), $t$ approximately corresponds to the derivative of the normalized transmission, similar to the signal detected by conventional Electron Paramagnetic Resonance (EPR) systems. The actual transmission can also be obtained, by choosing $H_2$ sufficiently far from $H_1$, but at the cost of deteriorating the signal-to-noise ratio.
RESULTS

A. Broad-band spectroscopy: field-tuned clock transitions

Figure 2 shows a two-dimensional map of the transmission derivative measured as a function of magnetic field and frequency. The data neatly show lines associated with the resonant absorption of microwave photons by the spins. Each of them corresponds to an allowed transition between two states having a different electronic spin wave function and the same nuclear spin state, such as those marked by vertical arrows in Fig. 1. These resonance lines then provide a complete picture of the low-lying magnetic energy levels in HoW$_{10}$. In particular, they show the presence of a finite gap $\Delta \approx 9.1$ GHz in the excitation spectrum associated with 4 different level anti-crossings. The position of these lines can be simulated using the spin Hamiltonian

$$\mathcal{H} = B_{20}\hat{O}_x^0 + B_{40}\hat{O}_x^4 + B_{60}\hat{O}_x^6 + B_{44}\hat{O}_x^{14} + g_J \mu_B \vec{H} \cdot \vec{J} + A J_z I_z$$

(2)

that includes the crystal field terms, the Zeeman interaction with the external magnetic field and the hyperfine interaction. As shown in Fig. 2, we find a very good agreement with the same parameters that were determined from EPR experiments performed on magnetically diluted samples (i.e. samples in which a $1 - x$ fraction of Ho$^{3+}$ ions were replaced with nonmagnetic Y$^{3+}$). The same parameters account also rather well for the specific heat and the magnetic response (see Supplementary Figs. 1 and 2). These results show that concentrated crystals retain the same magnetic anisotropy and confirm that the strong spin tunneling, and the associated energy gap, are genuine properties of each molecule.

The results shown in Fig. 2 were measured with the magnetic field applied along the $Z$ laboratory axis (the main axis of the device, as shown in Fig. 3 of the SI). A series of similar experiments were carried out for different orientations of $\vec{H}$ in the $XZ$ plane (the plane of the chip, to minimize effects associated with the excitation and motion of superconducting vortices). The results are shown in SI Figs. 4-6. They enable us to determine the orientation of the magnetic anisotropy axis $\vec{z}$ with respect to the crystal and the laboratory frames. It turns out that $\vec{z}$ points along the long molecular axis (Fig. 1), as expected.

For all orientations, the positions of the resonance lines agree very well with those derived from Eq. (2). This result confirms also the very strong uniaxial magnetic anisotropy of HoW$_{10}$: at any $\vec{H}$, the positions of the resonance lines, and of the clock transitions, are almost fully determined by the projection $H_z$ of the magnetic field along the anisotropy axis. The photon magnetic field $\vec{b}$ lies at about 45 degrees from $\vec{z}$. This means that the matrix element that determines the net absorption at each resonance includes almost equal weights of longitudi-

B. Absorption line shapes: enhanced spin-photon coupling near clock transitions

Whereas the positions of the resonance lines give access to the energy level scheme, their intensities provide useful information on the wavefunctions of the involved states and about how HoW$_{10}$ spins interact with their environment, as we discuss in this section. An important advantage of working with open waveguides is that both frequency and magnetic field can be varied independently of each other. It is therefore possible to monitor how the absorption line shapes vary as a function of $H$.

Figure 3 shows the normalized transmission measured...
near the fourth clock transition, taking place at $\mu_0 H_{z,4} \approx 0.168 \, \text{T}$. The absorption peak corresponds to a photon induced transition between states $|\psi_1\rangle$ and $|\psi_2\rangle$ with (approximate) nuclear spin projection $m_I = \pm 7/2$. For $H_z > H_{z,4}$, this resonance is conveniently isolated from other lines. The results (Figs. 2 and 3) show that the absorption lines fade away, i.e., that the resonant absorption decreases and the linewidth increases, as $H_z$ moves away from the position of the level anticrossing.

We have analyzed these results using input-output theory adapted to our experimental conditions, i.e., to a close to 1-D coplanar waveguide and for finite temperatures. The line shape of a given resonance can then be described with the following expression (see SI for details)

$$\frac{S_{2,1}}{S_{2,1}^{(0)}} = \frac{1}{1 + \frac{\Gamma \Delta P_{1,2}}{\gamma + (\omega_2 - \omega)}}$$

where $\Gamma$ is the photon-induced transition rate, $\Delta P_{1,2} = (\exp (-E_1/\kappa T) - \exp (-E_2/\kappa B T))/Z$ is the difference in thermal populations of the two levels, $Z$ is the partition function, $\gamma$ is the line width, which mainly reflects the intrinsic spin decay rate, and $\omega_2$ is the resonance frequency at the given magnetic field. Figure 3 shows that $\Gamma$, i.e., the coupling of spins to the transmission line, becomes maximum whereas the width $\gamma$ becomes minimum at the clock transition.

Given the strong magnetic anisotropy of HoW$_{10}$, the matrix elements of transverse angular momentum operators nearly vanish. The transition rate $\Gamma$ is then approximately proportional to $|\langle \psi_1 | J_z | \psi_2 \rangle|^2$. Under these conditions, $\Gamma$ monitors the overlap between the two wavefunctions. The maximum absorption observed at the clock transition therefore provides experimental evidence, independent from the spectroscopic observation of the ensuing gap, for the formation of quantum superpositions of spin-up and spin-down states.

The line broadening $\gamma$ sets an upper bound for the spin decoherence rate $T_2^{-1} \leq 125 \, \text{MHz}$ (or equivalently, $T_2 \geq 8 \, \text{ns}$). This bound is compatible with the results of pulse-EPR experiments, which fail to detect any spin echo on fully concentrated HoW$_{10}$ samples, thus suggesting that $T_2 \leq 20 \, \text{ns}$. The strong suppression of spin coherence, even at the clock transition, with increasing Ho concentration (see Fig. 7 of the SI) can be associated with resonant flip-flop transitions of two neighbouring spins, mediated by their mutual dipole-dipole interactions.

The increase of the linewidth with $|H_z - H_{z,4}|$ suggests that additional perturbations are then activated, or at least enhanced. It is plausible to associate such perturbations to bias dipolar fields: their fluctuations can decrease $T_2$, thus enhance the homogeneous line broadening, whereas the static components give rise to an inhomogeneous broadening. These effects are largely suppressed at the clock transitions by the vanishing derivative of the levels’ energies with respect to $H_z$ (or, equivalently, by the insensitiveness of the two states involved to bias magnetic fields) but become gradually more important as their wavefunctions become more and more localized (see Fig. 4). We have numerically calculated the distribution of magnetic dipolar fields in HoW$_{10}$. The results are shown in Fig. 8 of the SI. We find a distribution width $\approx 6 \, \text{mT}$ that corresponds to a maximum energy broadening (for $m_I = \pm 4$) of about 900 MHz, thus enough to account for the observed increase in the line broadening.

Using all this information, it is possible to perform a quantitative simulation of the transmission curves as a function of magnetic field and frequency. For this, we have used Eq. 3, with $\Gamma$ given by (see SI for details)

$$\Gamma = 2\pi g^2 (\omega_{12}) |\langle \psi_1 | J_z | \psi_2 \rangle|^2 [n(\omega_{1,2}) + 1]$$

where $g(\omega)$, the only fitting parameter, is a spin-photon coupling density, which depends on the mode density in the transmission line and on geometrical factors (mainly the location of all spins with respect to the circuit and the latter geometry), and $n(\omega_{1,2}) = [\exp (\hbar \omega_{1,2}/k_B T) - 1]^{-1}$ is the bosonic occupation number. In order to account for the line width, we have considered homogeneous and inhomogeneous broadening. The former gives $\gamma = 1/T_1 + 1/T_2$. We have fixed $T_1 = 20 \, \mu\text{s}$, as reported in reference 5 and introduced the field dependence of $T_2$ via the following simple phenomenological expression

$$T_2(H_z) = T_2(H_{z,4}) \frac{\omega_{1,2}(H_z)^2}{\omega_{1,2}(H_{z,4})^2}$$
FIG. 4. Experimental (top) and simulated (bottom) normalized transmission near the cavity mode with \( \omega_r \simeq 11.7 \text{ GHz} \) as a function of the applied field and the frequency. The simulation has been performed using Eq. (6) with \( T_2(H_z, 4) \sim 8 \text{ ns} \) and \( \omega_{1,2}(H_z, 4) \simeq 9.1 \text{ GHz} \) being, respectively, the spin coherence time and the resonance frequency at the clock transition. The results have then been averaged over the distribution of dipolar bias fields obtained numerically at each magnetic field (see Fig. 8 of the SI) and normalized as described by Eq. (1), in order to mimic the experimental procedure.

The results of these calculations are compared to the experimental ones in Figs. 2 and 3. They account well for the main features, in particular for the stronger spin-photon coupling and the reduced line broadening that are observed at the clock transitions. The experiments show also an intensity “revival” for \( \omega \simeq 11.7 \text{ GHz} \) that is not predicted by this model. The origin of this enhanced spin-photon coupling is discussed in the next section.

C. Collective coupling to a cavity photon mode

Imperfections in the coupling of the transmission guide to input and output lines give rise to spurious reflections and the formation of cavity modes, seen as horizontal lines in Fig. 2. Although in principle unwanted, the presence of such imperfections allows studying, in the same device, the coupling of the spins to cavity modes. One such mode, centered at \( \omega_r \simeq 11.7 \text{ GHz} \) with a very modest quality factor \( Q \sim 100 \), lies just at the middle of the magnetic excitation spectrum of HoW\(_{10}\). The resonant coupling of some of these excitations to this mode leads to an enhancement of the absorption near \( \omega_r \) and to a broadening of the resonant mode, as shown in Figs. 4 and 5, respectively.

In order to characterize these effects and estimate the collective coupling strength \( G_N \) to this confined mode, we have fitted the field dependence of the resonance width \( \tilde{\kappa} = Q/\omega_r \) using the following expression (see SI for a derivation from input-output theory)

\[
\tilde{\kappa} = \kappa + \frac{\gamma G_N^2}{(\omega - \omega_r)^2 + \gamma^2},
\]

where \( \kappa \) is the intrinsic width of the cavity mode decoupled from the spins. While \( G_N \) remains between 5 and 10 times smaller than the intrinsic spin decoherence rate \( \gamma \sim T_2^{-1} \), mainly because we are dealing here with a fully concentrated sample, it is nevertheless of the order of the

FIG. 5. Top: Variation of the width \( \tilde{\kappa} \) of the cavity mode observed at \( \omega \simeq 11.7 \text{ GHz} \) with respect to the value \( \kappa \) measured beyond any resonances with spin excitations (\( \Delta \kappa \equiv \tilde{\kappa} - \kappa \)). The solid red lines are least-square fits made with a Lorentzian line shape Eq. (6). Bottom: Field dependence of the spin-photon coupling \( G_N \), the spin decoherence rate \( \gamma \), and the cavity decay rate \( \kappa \) obtained from these fits.
cavity decay rate $\kappa$ and reaches nearly 100 MHz. It is remarkable that such a sizeable coupling can be achieved in such far from ideal conditions and, in particular, with thermally populated spin levels. As before, it is possible to simulate the normalized transmission. Results of such simulations, performed with Eq. (6) and the same parameters used for the simulation of the transmission through the open line, are shown in Fig. [4]. They account well for the experimental results.

Taking into account the enhancement in $T_2$ (up to a few $\mu$s) that can be achieved by magnetically diluting the HoW$_{10}$ molecular spin qubits (see Fig. 7 in the SI and reference [5], and the fact that $G_N$ scales as the square root of spin concentration $x$, these results show that the strong coupling limit can be achieved for $x \approx 0.1 - 0.2$. This is another consequence of the special character (reduced sensitivity to magnetic fluctuations, relatively large overlap) that spin states involved in each clock transition have. As we have seen in the previous section, these properties allow optimizing the spin-photon coupling while, at the same time, shielding the spins from unwanted sources of decoherence.

CONCLUSIONS

We have explored the full Zeeman diagram of magnetic energy levels and have been able to establish the nature of the spin wave functions, finding Schrödinger ‘cat states’ at the anticrossings. In these optimal regions, we have for the first time obtained evidence for a stronger coupling to photons that is accompanied by a minimum decoherence, due to an efficient shielding from fluctuating magnetic fields. These results suggest that spin clock transitions can enable attaining coherent coupling in what would otherwise be far from optimum conditions: concentrated spin systems, thus strong dipole-dipole interactions, high $T$, and thus very small spin polarization, a poor photon mode, etc. Coupling spin ‘clock states’ to superconducting circuits therefore opens a promising route towards achieving fault-tolerant scalability with chemically engineered spin qubits.

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