High-cooperativity coupling of rare-earth spins using yttrium orthosilicate as a substrate

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Yttrium orthosilicate (Y2SiO5, or YSO) has proved to be a convenient host for rare-earth ions used in demonstrations of microwave quantum memories and optical memories with microwave interfaces, and shows promise for coherent microwave-optical conversion owing to its favourable optical and spin properties. The strong coupling required by such microwave applications could be achieved using superconducting resonators patterned directly on Y2SiO5, and hence we investigate here the use of Y2SiO5 as an alternative to sapphire or silicon substrates for superconducting hybrid device fabrication. A NbN resonator with frequency 6.008 GHz and low power quality factor Q ≈ 400 000 was fabricated on a Y2SiO5 substrate doped with isotopically enriched 145Nd. Measurements of dielectric loss yield a loss-tangent tan δ = 4 × 10−6, comparable to sapphire. Electron spin resonance (ESR) measurements performed using the resonator show the characteristic angular dependence expected from the anisotropic 145Nd spin, and the coupling strength between resonator and electron spins is in the high cooperativity regime (C = 25). These results demonstrate Y2SiO5 as an excellent substrate for low-loss, high-Q microwave resonators, especially in applications for coupling to optically-accessible rare earth spins.

I. INTRODUCTION

Rare-earth ions (REIs) in crystals are promising candidate systems for quantum information applications, possessing electron and nuclear spins as well as optical transitions at telecom wavelengths. Coherence times in these systems range from milliseconds1 for microwave excitations stored in an electron spin to hours2,3 for optically accessible nuclear excitations, and such properties have led to proposals for using rare-earth ions as a photonic memory for a quantum repeater4, or as a microwave–optical transducer5,6 for use in quantum networks.

Yttrium orthosilicate (Y2SiO5) is a widely used crystalline host for REIs in such quantum information applications, as its constituent elements provide an environment with a low background of nuclear magnetic moments that would otherwise contribute to spin decoherence and inhomogeneous broadening. The narrow homogeneous linewidths, for example down to 3.5 kHz for the 883 nm transition7 in Nd3+:Y2SiO5, have been exploited as an optical quantum memory8,9 enabling storage of entangled states10 and teleportation with 93% fidelity11.

While optical storage experiments using REIs in Y2SiO5 can make use of a suitably optically dense medium, for microwave storage a cavity is employed to achieve strong coupling between an ensemble of REIs and microwave cavity field, so that excitations can be coherently exchanged between the two. This can be achieved with 3D microwave cavities12 which offer homogeneous B1 fields and spatial mode-matching between microwave and optical fields, or planar superconducting resonators which can reach quality factors over 105 yielding high sensitivities13 and act as an interface between superconducting qubits and spin ensembles14.

Common substrates for superconducting resonators include silicon and sapphire (Al2O3)16. For devices susceptible to dielectric losses, sapphire is often preferred for its low loss-tangent15 tan δ < 10−5. Experiments coupling superconducting resonators to rare-earths have then typically used a flip–chip approach with the doped sample glued or mechanically pressed onto the resonator chip17,18. This has the drawback of complicating the fabrication process, creates an additional interface layer between the resonator and sample increasing dielectric losses from spurious two-level systems (TLS) in the in-
terface [19], as well as a variable gap between device and spins which is less controllable than fabrication on the substrate itself.

In this work we investigate the suitability of using Y$_2$SiO$_5$ itself as a substrate for fabrication of planar superconducting devices. We fabricate a resonator on the polished surface of a Nd-doped Y$_2$SiO$_5$ sample, and find the device has a Q-factor $> 10^8$ at low powers due to its low loss-tangent $\tan \delta = 4 \times 10^{-6}$. We observe an anisotropic ESR spectrum matching that from simulations, and measure the coupling strength between resonator and electron spin to be in the high-cooperativity regime. These results suggest fabrication on doped Y$_2$SiO$_5$ is compatible with high-Q devices while simultaneously enabling a coupling to crystals doped with rare-earth ions, and could be a promising route toward scalable hybrid superconductor–spin quantum circuits.

II. DEVICE

The device is a lumped-element superconducting resonator, fabricated on a Czochralski-grown single crystal of Y$_2$SiO$_5$ [20] doped with 10 ppm isotopically purified $^{145}$Nd. The crystal was cut along the principal dielectric axes (D$_1$, D$_2$, b) and a face perpendicular to b was polished for thin-film growth. Y$_2$SiO$_5$ has two inequivalent crystal sites where a Y$^{3+}$ ion can be substituted by a rare-earth RE$^{3+}$ ion. The large ionic radius of Nd$^{3+}$ results in the larger crystal Site 1 being preferentially populated [1], resulting in a stronger signal from Site 1 over Site 2. Due to the crystal’s $C_{2h}^5$ (C$_2$/c) space group, each site has two orientations related by a $\pi$ rotation around the crystal b axis. These two orientations are termed sub-sites, and their ESR properties are degenerate for B$_0$ fields applied in the D$_1$–D$_2$ plane or parallel to the b axis.

Fabrication consisted of 40 nm of sputtered NbN being patterned by photolithography and a SF$_6$/Ar reactive ion etch process. A 2 $\mu$m wire functions as an inductor due to the constriction increasing the contribution of kinetic inductance. This wire is shunted by adjacent capacitative arms, forming the lumped-element design seen in the micrograph in Fig. 1 which generates an oscillating B$_1$ field to drive ESR transitions. The design and properties of this thin-ring resonator will be described in more detail in a future publication.

The patterned chip was enclosed within a 3D copper cavity (Q $\approx$ 100) to suppress spontaneous emission from the resonator to the environment [13]. This was installed in the bore of a vector magnet in a dilution refrigerator at 10 mK and probed using a vector network analyser (VNA).

The lumped-element resonator has a frequency of 6.008 GHz and an asymmetric lineshape due to interference with the background transmission of the 3D cavity. Fitting with a Fano resonance [21] yields a quality factor $Q \approx 400 000$ in the low-power limit in Fig. 1. By tracking the centre frequency versus temperature we find the dielectric losses in the device are comparable to resonators fabricated on sapphire [15][22] with a loss-tangent $\tan \delta = 4 \times 10^{-6}$, where the filling factor $F \approx \frac{1}{2}$ for this resonator geometry on a bulk-doped sample has been factored out.

This demonstrates that Y$_2$SiO$_5$ is well-suited for...
devices incorporating resonators and superconducting qubits which are typically susceptible to dielectric losses from TLSs [23], while also incorporating doped spins for cavity QED.

III. ELECTRON SPIN RESONANCE

A. ESR spectrum

A magnetic field \( B_0 \) was applied in the plane of the superconducting thin film, roughly perpendicular to the \( \text{Y}_2\text{SiO}_5 \) b crystal axis. The field was swept up to 360 mT and the resonator was used to observe ESR transitions, monitoring changes to its centre frequency tracked with a VNA. By this method a series of spectra were taken as a function of \( B_0 \) orientation in the \( \text{D}_1–\text{D}_2 \) plane to extract the angular dependence, or “roadmap”, of ESR transitions, and the resonator was used to observe ESR transitions of \( \text{Nd} \) nuclear spin superconducting thin film, roughly perpendicular to the spin species are routinely performed on bulk samples, but to do this with a superconducting resonator as shown here requires a device resilient to in-plane magnetic field. These measurements enable ESR transitions to be positively identified by comparing their angular dependence to simulations [25] and bulk ESR data, ensuring further analysis of spin–resonator coupling properties can be clearly linked to a particular spin species.

For an electron spin \( S \) coupled to a nuclear spin \( I \) the Hamiltonian accounting for the electron Zeeman and hyperfine terms is \( H = \mu_B B \cdot g S + S^z A I \). The anisotropy of \( \text{Y}_2\text{SiO}_5 \) results in the parameters \( g \) and \( A \) being tensors, previously calculated from spectroscopic studies of site 1 of \( ^{145}\text{Nd}:\text{Y}_2\text{SiO}_5 \) at 9.4 GHz [23] as

\[
g = \begin{pmatrix} 1.30 & 0.62 & 0.22 \\ 0.62 & -2.07 & 1.62 \\ 0.22 & 1.62 & -2.86 \end{pmatrix}_{(\text{D}_1, \text{D}_2, \text{b})} \\
A = \begin{pmatrix} -37.1 & -99.9 & -83.4 \\ -99.9 & -589.2 & 169.4 \\ -83.4 & 169.4 & -678.4 \end{pmatrix}_{(\text{D}_1, \text{D}_2, \text{b})} \text{MHz}
\]

A characteristic ESR spectrum (\( B_0 \) along the \( \text{D}_1 \) axis) is plotted in Fig. 2(b). We observe a series of eight ESR transitions indicated in Fig. 2(a), corresponding to the \( ^{145}\text{Nd} \) nuclear spin \( I = \frac{7}{2} \) in Site 1. A small misalignment between the \( \text{D}_1–\text{D}_2 \) and superconductor planes breaks the degeneracy of the sub-sites and causes the observed splitting of each line into two.

The roadmap of ESR spectrum with respect to \( B_0 \)-field angle is plotted in Fig. 2(c), with transitions marked by circles. The angular dependence seen in these spectra matches well simulations for Nd spins in Site 1 (blue) while we also observe a signal at lower magnetic fields (higher g-factor) which we attribute to Site 2 (red). The full spin Hamiltonian of \( ^{145}\text{Nd} \) in Site 2 including hyperfine terms has not been reported, therefore our simulations only account for Zeeman term (i.e. \( g \) tensor). Additional unidentified impurities are evident from the different angular dependences of 2–3 transitions marked in green and black.

ESR studies on the angular dependence of anisotropic spin species are routinely performed on bulk samples, but to do this with a superconducting resonator as shown here requires a device resilient to in-plane magnetic field. These measurements enable ESR transitions to be positively identified by comparing their angular dependence to simulations [25] and bulk ESR data, ensuring further analysis of spin–resonator coupling properties can be clearly linked to a particular spin species.

B. High-cooperativity coupling

The highest intensity ESR transition for \( ^{145}\text{Nd} \) in Site 1 with \( m_I = \frac{7}{2} \) at \( B_0 = 216 \text{ mT} \) along \( \text{D}_1 \) was selected for an evaluation of the spin–resonator coupling strength. Measuring the resonator \( S_{21} \) with a VNA while sweeping magnetic field strength \( B \), we observe the onset of an avoided crossing shown in Fig. 3. Fitting the resonator frequency \( \omega \) and linewidth \( \kappa \) at each field point [26] allows us to extract the coupling strength \( g_{\text{ens}} \), inhomogeneous spin ensemble half-width \( \gamma_s \), and resonator half-width \( \kappa_c \) with

\[
\omega = \omega_c - g_{\text{ens}}^2 \Delta / (\Delta^2 + \gamma_s^2) \\
\kappa = \kappa_c - g_{\text{ens}}^2 \gamma_s / (\Delta^2 + \gamma_s^2)
\]

where \( \Delta = m_0 (B - B_0) / h \) is the field detuning calculated from the spin magnetic moment \( m_0 = h \frac{d\sigma}{d\sigma} \). This accounts for cases where \( \frac{d\sigma}{d\sigma} \) varies with \( B \), as is the case for mixed spin systems in the low-field limit or near zero first-order Zeeman (ZEFOZ) points.

![Figure 3](image-url)

**FIG. 3.** Left: onset of an avoided crossing when sweeping B-field through resonance. Right: resonator frequency shift and quality factor as a function of B-field strength when sweeping through resonance. Extracting quality factor at the centre of the resonance proved unreliable as the prominence of the resonance diminished; these points are indicated with a dashed line. Fit to theory (red) indicates high-cooperativity coupling \( C = 25 \).
From this we extract $g_{\text{ens}} = 1.2 \text{ MHz}$, $\gamma_s = 4.6 \text{ MHz}$, and $\kappa = 13.2 \text{ kHz}$. This corresponds to a high cooperativity $C = \frac{g_{\text{ens}}^2}{\kappa \gamma} = 25$.

For a bulk doped sample there is no clear single-spin coupling rate as the $\frac{1}{2}$ decay of the field strength from the wire results in a $B_1$ inhomogeneity spanning orders of magnitude while all spins in the sample contribute to the signal. However, a characteristic rate representing the average coupling strength over the spins contributing 50% of the measured signal can be calculated. This corresponds to a region within 4 μm of the wire with $N \approx 6 \times 10^6$ resonant $^{145}\text{Nd}$ spins in the same sub-site, accounting for the thermal population in the $m_1 = \frac{1}{2}$ ESR transition at 10 mK. From this we derive a characteristic single-spin coupling rate $g_0 = \frac{g_{\text{ens}}}{\sqrt{N}} \approx 50 \text{ Hz}$ leading to an expected Purcell enhanced emission rate $\Gamma_p = \frac{4g_0^2}{\kappa} \approx 1 \text{ Hz}$.

These results indicate the coupling regime is limited by the spin linewidth $\gamma_s > g_{\text{ens}}$, or equivalently that the Nd spin ensemble is decohering at a faster rate than excitations are being exchanged between it and the resonator. Steps toward improving this coupling strength could include using a more strongly doped rare-earth sample to increase $g_{\text{ens}}$ though this could also increase $\gamma_s$, or exploiting the coherence-enhancing ZEFOZ transitions in REIs [28] to decrease $\gamma_s$. The significant $B_1$ inhomogeneity in this resonator design also poses challenges for performing coherent operations on the entire spin ensemble with pulsed ESR. This could be mitigated by coupling to implanted layers of REIs [23, 29], or by using resonators designed to generate a more homogeneous $B_1$ field.

IV. CONCLUSION

We investigated the use of $\text{Y}_2\text{SiO}_5$ as an alternative to sapphire or silicon substrates for the fabrication of superconducting devices. The fabricated NbN lumped-element resonator had a quality factor $Q \approx 400 000$ and a loss-tangent $\tan \delta = 4 \times 10^{-6}$ comparable to sapphire [15], and yielded a high-cooperativity coupling $C = 25$ between the resonator and a $^{145}\text{Nd}$ spin ensemble. These results demonstrate $\text{Y}_2\text{SiO}_5$ is well-suited for superconducting devices, while also enabling an interaction with optically-accessible rare earth spins.

Further studies of this device include performing pulsed ESR measurements in the high-cooperativity regime and measuring coherence properties of the spin ensemble. The methods shown here are also applicable to other REIs in $\text{Y}_2\text{SiO}_5$, for example Er with its 1540 nm telecom band optical transition [6] and Yb which exhibits a large oscillator strength [30] and coherence-enhancing ZEFOZ and near-ZEFOZ transitions [28]. It may also prove interesting to study the suitability as a substrate of other crystalline hosts for REIs, such as yttrium aluminium garnet (YAG), yttrium lithium fluoride (YLF), yttrium orthovanadate (YVO$_4$), and calcium tungstate (CaWO$_4$).

Fabricating superconducting devices on crystals doped with REIs as demonstrated here shows promise for integrating these optical elements alongside the fast information processing available from superconducting qubits. This is important step toward making a microwave–optical transducer capable of connecting such quantum processors within a quantum network. By exploiting long coherence times available from REIs this is also a route to integrating fast quantum electronics with a spin ensemble acting as a quantum memory.

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