Supplementary Materials for

Enhanced quantum sensing with room-temperature solid-state masers

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Section S1: Electromagnetic simulation of microwave dielectric resonators

The dielectric resonator supporting the $|\lambda\rangle \leftrightarrow |Z\rangle$ transition among the pentacene’s triplet sublevels was designed using COMSOL Multiphysics with a 2D axisymmetric model (60). The aim of the simulation is to determine the geometry of the dielectric material, i.e. strontium titanate (STO) for generating a TE$_{01\delta}$ electromagnetic mode resonant around 1.45 GHz (i.e. close to the pentacene’s $|\lambda\rangle \leftrightarrow |Z\rangle$ transition frequency at zero field). A TE$_{01\delta}$ mode of a STO resonator has been proven, so far, to be the most suitable for pentacene-doped-$p$-terphenyl masers, which simultaneously provides a relatively high magnetic filling factor ($\sim 0.3$) (33, 35) and a high Purcell factor ($3.6 \times 10^7$) (27). The overall composition of the resonator (see Fig. S1), including a STO hollow cylinder, a support made of Rexolite, a copper tuning screw, loop antennas and an oxygen-free copper cavity with a hole drilled on the wall for optical pumping, is similar to that employed in the previous studies (21, 27, 33, 35, 48). But because the dielectric constant of STO varies with suppliers, which may arise from different impurities contained in STO, the exact dimensions of the STO hollow cylinder needs to be determined by the simulation.

Prior to the simulation, we manufactured prototypes (with arbitrarily chosen dimensions) of the STO hollow cylinder, the Rexolite support and the oxygen-free copper cavity with a cooper tuning screw. After assembly of the prototype dielectric resonator, the resonance frequency of its TE$_{01\delta}$ mode was measured with a microwave analyzer (Keysight N9917A). We then inputted the known geometries of all prototype components in the COMSOL model and adjusted the dielectric constant of STO to fit with the experimentally determined resonance frequency. By fitting, the dielectric constant of the STO raw material used in our work was determined to be 318. It is worth noting that for simplicity of the simulation, loop antennas were not included in the model. Moreover, because the Rexolite support and the pentacene-doped $p$-terphenyl crystal shown in Fig. S1 have been verified not to change the resonance
frequency significantly, only the dielectric material (i.e. STO), the copper cavity and air were constructed in the model. The height of the copper cavity was set to be a flexible parameter since it can be adjusted by the tuning screw in experiments.

Following the determination of the dielectric constant of STO, we adjusted the geometries of the prototype STO hollow cylinder and copper cavity in the model to achieve a 1.45-GHz $\text{TE}_{01\delta}$ mode (shown in Fig. S1) with a tunable range about 20 MHz (by adjusting the height of the copper cavity). The geometries of the crucial parts of the dielectric resonator were thus finalized.

![Figure. S1. A two-dimensional (2D) axisymmetric simulation of the strontium titanate (STO) microwave resonator.](image)

The heat map and red arrows represent the magnetic energy density and magnetic field vector of the $\text{TE}_{01\delta}$ mode of the STO microwave resonator. The main components used to construct the resonator, the sample position and the optical pump path through the cavity wall are labelled.

**Figure. S1.** A two-dimensional (2D) axisymmetric simulation of the strontium titanate (STO) microwave resonator. The heat map and red arrows represent the magnetic energy density and magnetic field vector of the $\text{TE}_{01\delta}$ mode of the STO microwave resonator. The main components used to construct the resonator, the sample position and the optical pump path through the cavity wall are labelled.

**Section S2: Configuration of a regenerative microwave oscillator**
The block diagram of the regenerative microwave oscillator is shown in Fig. 2(C) of the main text. The key microwave components and equipment used are summarized in Table S1.

**Table S1. List of the key microwave components/equipment used for the setup of the regenerative microwave oscillator**

| Type              | Brand        | Model           |
|-------------------|--------------|-----------------|
| Isolator          | TDK          | 11GRZ03         |
| Amplifier         | MITEQ        | 124758          |
|                   | Qorvo        | SPF5189Z        |
| Band-pass filter  | Unbranded/Generic | FBP-1420s   |
| Power splitter    | Talent Microwave | RS2DC180-S    |
| Limiter           | Mini Circuits | ZFLM-252-1WL-S+ |
| Directional coupler | Narda      | 25017          |
|                   | Narda-ATM    | C122E-10        |
| Variable attenuator | MERRIMAC | AUM-25A         |
| Phase shifter     | SAGE         | 6718-2          |
| Logarithmic detector | ADI       | AD8317         |
| Microwave analyzer | Keysight    | N9917A          |
| Oscilloscope      | Tektronix    | MSO64           |

To measure the quality of the microwave oscillator, a transmission ($S_{21}$) measurement was conducted at the coupling ports of the two directional couplers shown in Fig. 2(C). To protect the microwave analyzer (Keysight N9917A) from the oscillating signals generated in the circuit, two isolators were added at the input and output ports of the analyzers. The obtained transmission spectrum is rescaled and shown in Fig. S2. The transmission linewidth was measured to be about
8 kHz which corresponds to a boosted quality factor of ~180000. The similar ‘Q-boosting’ strategy has also been used to achieve room-temperature strong coupling of a spin ensemble with microwave photons(61) and explore gain media of room-temperature solid-state masers(36).

Figure. S2. Measured quality factor (Q) of the regenerative microwave oscillator. The linewidth was determined by the frequency difference of the two -3-dB points straddling the central frequency. The quality factor is the ratio of the central frequency to the linewidth.
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