Microwave cavity-free hole burning spectroscopy of Er$^{3+}$:Y$_2$SiO$_5$ at millikelvin temperatures

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Efficient quantum memory is of paramount importance for long-distance quantum communications, as well as for complex large-scale computing architectures. We investigate the capability of Er$^{3+}$:Y$_2$SiO$_5$ crystal to serve as a quantum memory for the travelling microwave photons by employing techniques developed for dense optical ensembles. In our efforts to do so, we have performed high-resolution microwave spectroscopy of Er$^{3+}$:Y$_2$SiO$_5$, where we identified electronic spin as well as hyperfine transitions. Furthermore, we have explored spectral hole burning technique and studied the spin relaxation process at millikelvin temperatures, determined the main relaxation mechanisms, which lay the groundwork for further studies of the topic.

I. INTRODUCTION

Microwave regime in quantum technologies is currently best represented by the superconducting circuits based on superconducting qubits$^1$. While the quantum processing units based on such superconducting qubits demonstrate outstanding results$^2$, further enhancement of calculation capacities addresses the necessity of effective microwave quantum memory$^3$. At the same time, the longest storage times are demonstrated with optical quantum memories$^4$-$^5$, which, however, rely on ultra-long coherence of the spin states of the rare earth spin ensembles.

Realization of purely microwave quantum memories has been demonstrated with electronic spins ensembles based on donors in silicon and rare earth ions$^6$. Depending on the choice of the quantization axis and magnetic field working point, storage times in such ensembles may reach six hours$^7$. Such extremely long storage time is possible due to using the zero first-order Zeeman (ZEOFOZ) transitions, which are analogous to the clock transitions and possess strongly reduced sensitivity to the fluctuations of the external decoherence sources. Coupling of the microwave fields to the spin ensembles in microwave experiments is typically performed via resonating structures$^7$-$^10$, which limits the number of transitions accessible at the same time.

Ability to effectively control the electronic spins with the propagating microwave signals$^{11}$-$^{15}$, which can couple to the same spins in broad frequency range, will pave the way to a large variety of microwave quantum memory schemes including more than two electronic levels. The atomic frequency comb (AFC) protocol is one of the most promising quantum memory protocols established in optics$^{16}$-$^{18}$, which can be realized on two electronic levels as well as extended to a third longer living state allowing for frequency domain multiplexing storage protocols$^{19}$-$^{20}$. Deterministic narrow spectral hole burning (SHB) is a necessary step in realization of AFC. While these techniques are strongly established in optics, they have not yet been widely studied in the microwave domain. The first attempt in burning microwave spectral holes has been performed on spin ensembles coupled to a resonator$^{21}$.

In this work, we explore the spectral hole burning with the propagating microwave signals in Er:Y$_2$SiO$_5$ crystal in the cavity-free regime, which allows for addressing the electronic spins in a broad frequency range. We characterize the control efficiency in our experimental scheme, identify the relaxation rates and processes, and extract the resulting spectral hole profiles. We address the potential of the SHB with the propagating microwave signals for the microwave quantum memory techniques.

II. EXPERIMENTAL DETAIL

We investigate the Erbium-doped Y$_2$SiO$_5$ (YSO) crystal, which is supplied by Scientific Materials Inc. with 0.02% of Erbium atomic concentration. Erbium is in its natural abundance with $\sim$ 23% being $^{167}$Er ($I = 7/2$), and $\sim$ 77% being 5 isotopes with zero nuclear spin. The
crystal has dimensions $3 \text{ mm} \times 4 \text{ mm} \times 5 \text{ mm}$ and is placed on the top of the co-planar transmission line, see Fig. 1(a,b). The Erbium spins are inductively coupled to the transmission line via magnetic field created by the propagating microwave signal. The orientation of the optical crystal axes is shown in Fig. 1(a). The external magnetic field vector and axes $D_{22}$ and $b$ are located in the plane of the SC chip with an angle of $45^\circ$ between axis $b$ and magnetic field vector. The magnetic field vector is parallel/anti-parallel to the wave-vector of the microwave field. Due to the low crystalline and point symmetry of YSO crystal, Erbium ions occupy two crystallographic nonequivalent sites, each showing two magnetically nonequivalent transitions. The orientation of the crystalline axes with respect to the magnetic field has been selected to fully lift the degeneracy between the magnetically nonequivalent sites.

Schematics of the measurement scheme is shown in Fig. 1(c). In the microwave spectroscopy and in hole burning measurements, the probing signal is created by Vector Network Analyzer (VNA) at the power of 0 dBm, which is additionally attenuated by $-95 \text{ dB}$ before reaching the sample. After the crystal, the transmitted signal is amplified by cryogenic low noise amplifier by 40 dB and by room-temperature low noise amplifiers by 60 dB.

Microwave hole-burning pulse is generated by a radio-frequency source (RFS) in the power range of 5 dBm to 15 dBm with a time duration of 0.5 s to 5 s. The burning pulse is attenuated on the input RF lines of the cryostat by $-55 \text{ dB}$. At the end of the burning pulse, VNA starts measuring time dependence of the transmission at a selected frequency. In order to protect the VNA input, it is cloaked from the high-power burning pulses by an RF switch, with both VNA and switch being triggered by the RFS. Both, change of absorption amplitude and phase are recorded as functions of time with intermediate frequency filter bandwidth (IFBW) of 10 Hz and sampling interval of $\sim 50 \text{ ms}$.

### III. MICROWAVE SPECTROSCOPY

Microwave spectroscopy is performed on $I_{15/2}(0) \rightarrow \frac{1}{2} \leftrightarrow I_{15/2}(0) \rightarrow \frac{3}{2}$ spin transition as a function of magnetic field at a base temperature of cryostat equal to 10 mK, see fig. 2(a). The absorption spectra are measured as $S_{21}$ parameter of VNA. In the spectrum, three strong absorption lines correspond to the crystallographic ($S^a_1$ or $S^b_2$) and magnetic ($S^a_{SD}$ or $S^b_{SD}$) nonequivalent sites of the erbium ions and are identified with the available g-tensor as marked in Figure 2(a,b). The fourth absorption line, $S^a_{1b}$, possesses the largest g-factor of $\pm 13$ and is outside the measured frequency range at applied magnetic fields. The hyperfine transitions of $^{167}$Er isotope with the nuclear spin of $\frac{7}{2}$ are observed at a much smaller absorption amplitude. The most pronounced absorption line corresponds to the $S^{1b}$ site, on which we investigate the hole-burning technique in this work.

The structure of microwave spectrum is determined by the Zeeman, hyperfine and quadrupole interactions,
1. The identified transitions are highlighted with light blue in the measured spectrum. The $S_{1b}$ transition studied with hole burning technique is highlighted with red color. Amplitude of the simulated spectra is given in arbitrary units.

2. When comparing the measured g-factors to the theoretical ones, we find an effective temperature, becomes an important parameter at millikelvin temperatures. Even small excitation result into elevated effective temperatures of spin ensembles, which can be an order of magnitude higher than room temperature. The key issue with the spin temperature is that the spin levels are strongly quantized with respect to the thermal energy level of the surrounding thermal bath. As a result, the effective spin ensemble temperature is given by $T_{eff} = \frac{\hbar \nu}{2k_B}$, where $\nu$ is the frequency of the radiation and $k_B$ is the Boltzmann constant.

3. The further absorption spectrum analysis and hole-burning spectroscopy are performed on the strongest absorption lines are marked according to the identified microwave transitions. The absorption lines are marked according to the identified microwave transitions. Hole burning spectroscopy is performed on the strongest $S_{1b}$ transition. Additional resonances at fixed frequencies are due to reflections on cable connections and bonding on the sample holder and the chip with the transmission line, see Fig. 1 for the measurement setup details.

4. Measured (black lines) and simulated (blue lines) absorption spectra at selected magnetic fields of 110 mT and 185 mT. The identified transitions are highlighted with light blue in the measured spectra. The $S_{1b}$ transition studied with hole burning technique is highlighted with red color. Amplitude of the simulated spectra is given in arbitrary units.

5. Measured absorption profiles (circles) and Lorentzian fits (solid lines) of the absorption line and extracted spectral hole at 167 mT. The inset depicts measurement sequence for the spectral hole recovery.

6. Temperature of a spin ensemble, often addressed as an effective temperature, becomes an important parameter at millikelvin temperatures. Even small excitation result into elevated effective temperatures of spin ensembles, which can be an order of magnitude higher than room temperature. The key issue with the spin temperature is that the spin levels are strongly quantized with respect to the thermal energy level of the surrounding thermal bath. As a result, the effective spin ensemble temperature

\[ T_{eff} = \frac{\hbar \nu}{2k_B} \]

where $\hbar \nu$ is the energy of the radiation and $k_B$ is the Boltzmann constant.
ture is defined as a probability of finding the electronic spin in a particular energy level, and it is calculated via Boltzmann distribution for a canonical ensemble.\textsuperscript{27,28}

To find the temperature of the spin system in the spectroscopy measurement, we fit the area under the absorption line to the Boltzmann distribution on temperature, given as $\sim \exp(\Delta E/k_B T_s)/(1 + \exp(\Delta B/k_B T_s))$, where $\Delta E$ is the energy difference between the spin states, $k_B$ is the Boltzmann constant, and $T_s = 81.9(20)\text{ mK}$ is the spin temperature. At the same time, the base temperature on the cryostat plate is measured by a sensor to be equal to $11\text{ mK}$. Thermal interface from the spin ensemble to the cryostat is mediated by spin-phonon interaction (phonon bath) and thermal boundary resistance (Kapitza resistance). The later one lead to a significant suppression of the thermal energy flow from spin ensemble to the cooling element and result into such a difference in temperature.

IV. DYNAMICS OF SPECTRAL HOLES

Spectral hole burning is performed in a saturation recovery experiment, when a microwave burning pulse of a fixed frequency is sent to the crystal for a fixed time. The population of the ground state is then transferred to the excited state, as indicated with the red arrow in fig. 1(c). As a result, the absorption amplitude is reduced. The recovery of the saturation amplitude over time is detected with a week probing signal of VNA and is governed by the dynamics of relaxation processes.

Relaxation processes. Below $1\text{ K}$, spin relaxation is limited to two main processes: flip-flop, $R_{FF}$, and direct process, $R_d = \frac{1}{\tau_d}$. These processes are schematically shown in fig. 1(c). Third process depicted in fig. 1 is spectral diffusion, $\Gamma_{SD}$, which contributes into broadening of the spectral hole.

The spin-spin relaxation rate given by flip-flop process is defined as $\text{29}$

$$R_{FF} = w_{FF} \tanh^2 \left( \frac{g \mu_B B}{2k_B T_s} \right), \quad (2)$$

where $w_{FF}$ is the flip-flop coefficient, which, following the approach from Car et al.\textsuperscript{30}, we estimate to be equal $50\text{ mHz}$. Constants $\mu_B$ and $k_B$ are Bohr magneton and Boltzmann constant, respectively, and $T_s$ is the effective temperature of the spin system.

Rate of the direct process over the magnetic field and temperature in absence of the phonon bottleneck is given by $\text{31}$

$$R_d = \frac{1}{\tau_d} = w_d g^5 B^5 \coth \left( \frac{g \mu_B B}{2k_B T_s} \right), \quad (3)$$

where the spin-phonon relaxation rate coefficient $w_d$ is estimated to be equal to $23\text{ Hz/T}^{-5,7,31,32}$.

Thus, the final equation including flip-flop and direct process relaxation rates reads as $R = R_{FF} + R_d$, which is then fit independently to the magnetic field, fig. 3(a), and temperature, fig. 3(b), dependent data. Derived coefficients are listed in table II and show very good agreement with analytically derived values.

Effective temperature. The effective temperature of the spin ensemble $T_s$, which is derived from relaxation processes, is in a good agreement with the spin temperature derived from the microwave spectrum, $\sim 70\text{ mK}$, see table II for detail. It also means that below magnetic fields of $B_{pol} \approx 2 T_s$, $T_s \approx 140\text{ mT}$: the spin ensemble is not polarized to the ground state. This results into stronger flip-flop rate as well as into smaller hole amplitudes leading to smaller signal-to-noise ratio in the data. Below the magnetic field of $70\text{ mT}$, splitting of the spin states is smaller than the thermal energy level, $k_B T_s \geq g \mu_B B$, and saturation recovery is not measurable.

Apart from extracting temperature of the spin ensemble, we directly measure the temperature of the cryostat itself which reaches down to $11\text{ mK}$. Temperature of the cryostat remains unchanged during the sweep of the magnetic field and is controllably changed during the temperature-dependent measurement. For fitting the relaxation processes to experimental data, we introduce the correction of the actual temperature of the spin ensemble with respect to that of the cryostat $\text{32,33}$:

$$T_s = T_{min}(1 + (T/T_{min})^2)^{1/2}, \quad (4)$$

where $T$ is the temperature of the cryostat and $T_{min}$ is minimal temperature attainable by the spin system. At the limit of $T < T_{min}$, $T_s = T_{min}$, while for $T > T_{min}$, $T_s = T$. Relaxation rates extracted thus from the temperature dependent data are in good agreement with the magnetic field dependence. Largest declination of the data points from the fit-curves are observed for the lowest and higher fields, see fig. 3(b). However, such a straggle of the data points is similar to that observed for the multiple measurements in the magnetic field dependence, fig. 3(a).

Rabi frequencies and acting microwave power. For efficient control of the a spin ensemble over the transmission line, we need to know the effective acting power of the magnetic field component of the microwave signal and correct Rabi frequency. To identify the Rabi frequency, we vary the length of the burning pulse and measure the maximally achieved amplitude of the spectral hole. From the observed oscillations, we derive the Rabi frequency to be equal to $3.9\text{ Hz}$. Taking into account the size of the propagating microwave mode, see fig. 1(d), we derive the

| Fit of magnetic field dependence | 23(1) \text{ Hz} \quad 50(1) \text{ mHz} \quad 69(8) \text{ mK} | 89 |
| Fit of temperature dependence | 22(2) \text{ Hz} \quad 30(1) \text{ mHz} \quad 73(21) \text{ mK} | 85 |
| Analytically derived spectroscopy | 23 \text{ Hz} \quad 51 | 81.9 \quad 50 |

\text{TABLE II. Coefficients of relaxation processes and effective temperature values}
RMS amplitude of the magnetic component of the microwave field $B_{AC} \simeq 0.35$ nT, or total acting microwave power $P_{act-R} \simeq 45$ fW ($-105$ dBm).

From the spectroscopy measurement, we also derive additional losses on the sample and SC to be in the order of $-40$ dB. Simulating the propagation of the microwave mode as shown in fig. 1(d), we estimate the mean loss of amplitude in the main acting volume of the microwave mode to be in the order of $-20$ dB. Applying the total loss from the spectroscopy measurement, comprising of $\sim -115$ dB, see fig. 1, to the microwave power of $15$ dBm generated by the RFS, we obtain expected acting power of $P_{act-a} = -100$ dBm in the microwave mode. We thus see that the acting powers from Rabi frequency, $P_{act-R}$, and from analytical calculations, $P_{act-a}$, are in good agreement. The further hole burning experiment is performed with $5$ dBm output power of RFS, with Rabi frequency of $\simeq 1$ Hz, at which we have not observed any pronounced oscillations when varying the pulse duration.

V. SPECTRAL HOLE PROFILES

By keeping the burning frequency fixed and changing the probing frequency, we identify the profiles of the spectral holes. Such a three-dimensional spectrum of a spectral hole is shown in fig. 4(a), where the pump frequency was kept fixed at $3.651$ GHz and probing frequency was scanned around the pump with total span of $20$ MHz. When increasing the burning time, the amplitude of the spectral hole is increased, while the width of the spectral hole and relaxation time are independent of the burning time, which has been confirmed at several magnitudes of magnetic field. Therefore, the burning pulse length of $3$ s has been selected for the hole burning experiment as an optimal one. Independence of the spectral hole width of the burning pulse length is attributed to the rate of spectral diffusion, which is faster ($\sim 1$ kHz) than possible shortest burning pulse ($0.1$ s-1 s).

The profiles of spectral holes have been probed for a set of magnetic field values. By fitting the profiles to the Lorentzian shape, we extract amplitude and width of the created spectral holes as functions of time, fig. 4, and of magnetic field. In the range of magnetic fields below $170$ mT, area and amplitude of the holes are nearly constant. At increase of magnetic field above $170$ mT, the width of spectral hole increases while amplitude decreases. The relative amplitude of spectral holes is at the level of $\sim 7.5$ % of the spectral line amplitude, and similarly the relative width slightly varies around $\sim 65$ % of the spectral line width.

Measuring the relaxation of the spectral hole over its profile, we observed it to be faster when measured outside of width at half maximum for magnetic field of $185$ mT, see fig. 4(b). Inside of the spectral hole width region, the relaxation time can be considered constant. Such behavior of spectral holes appears then as narrowing of the spectral hole with time as shown in fig. 4(c). Interestingly, the spectral hole burned at $167$ mT, which profile is demonstrated in fig. 2(c), has shown nearly no change of the width over time. Moreover, there is a narrowing of the spectral hole after $20$ s of relaxation, after which the width returns to it’s initial value of $\sim 37$ MHz. The wider spectral hole observed at $185$ mT converges to a width value of $\sim 40$ MHz, which is similar to that at $167$ mT resulting approximately $60$ % of the absorption line width. At other magnetic fields, width of spectral holes is also $\sim 60$ %, revealing no change over the time. We suggest that the large initial broadening on spectral hole at $185$ mT and it’s subsequent narrowing is related to interference of one of the on-chip parasitic resonance at a close frequency ($\sim 3.75$ GHz) to the transition in-study.
Earth spin ensembles.

This publication pave the way towards the realization of microwave power in burning pulse. Results presented in first-order Zeeman transitions and increase of the acting resonator. Further enhancement of the control of the spin systems in cavity-free regime require exploiting the zero first-order Zeeman transitions and increase of the acting microwave power in burning results. Results presented in this publication pave the way towards the realization of cavity-free microwave quantum memory based on rare-earth spin ensembles.

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