Shaped pulses for transient compensation in quantum-limited electron spin resonance spectroscopy

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**A B S T R A C T**

In high sensitivity inductive electron spin resonance spectroscopy, superconducting microwave resonators with large quality factors are employed. While they enhance the sensitivity, they also distort considerably the shape of the applied rectangular microwave control pulses, which limits the degree of control over the spin ensemble. Here, we employ shaped microwave pulses compensating the signal distortion to drive the spins faster than the resonator bandwidth. This translates into a shorter echo, with enhanced signal-to-noise ratio. The shaped pulses are also useful to minimize the dead-time of our spectrometer, which allows to reduce the wait time between successive drive pulses.

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1. Introduction

Electron spin resonance (ESR) spectroscopy allows to analyze the composition and structure of paramagnetic samples [1]. In the inductive detection method, the sample is coupled to a resonant microwave cavity of frequency \(\omega_0\). When the spins are tuned to \(\omega_0\) by application of a magnetic field \(B_0\), they interact with the magnetic component of the intra-cavity microwave field \(B_1(t)\). After being driven by appropriate microwave pulse sequences, their Larmor precession dynamics induces the subsequent emission of weak microwave spin-echo signals, which carry information on the properties of the paramagnetic species present in the sample.

The cavity has several roles in this process. It amplifies the \(B_1\) driving field by a factor proportional to \(Q/\sqrt{\omega_0} \) (\(Q\) being the resonator quality factor), which lowers the incident microwave power requirements. It also amplifies the emitted spin-echo signals by the same factor, which conversely enhances detection sensitivity. On the other hand, the cavity bandwidth \(\Delta \omega = \omega_0/Q\), which sets both the spin excitation and spin detection bandwidths, which can be a severe issue since typical electron spin linewidths may exceed \(\Delta \omega\) by far. The cavity also causes transients in the drive fields which limit the degree of control achieved on spin dynamics, and whose characteristic time scale is given by the cavity field amplitude damping time \(T_1\). As a result, the cavity quality factor needs to be carefully optimized for the purpose of a given experiment.

A number of these issues can be mitigated by the use of drive pulses with a more complex time-dependence than a simple rectangular pulse (so-called shaped pulses) [2,3], as schematically shown in Fig. 1. Thanks to modern arbitrary waveform generators (AWG) [4], microwave pulses with arbitrary time-dependent amplitude and phase can be generated. If the transfer function between the AWG and the intra-cavity field is known, it is possible to compute the drive pulse shape needed to obtain arbitrary intra-cavity field temporal profiles. This strategy in principle eliminates all the drive pulse distortions caused by the cavity; in particular, spins can be driven with a bandwidth much larger than \(\Delta \omega\) using shaped pulses [5]. Cavity filtering of the signal emitted by the spins, however, remains unavoidable [5].

The use of shaped pulses [6] has been proposed and demonstrated first in nuclear magnetic resonance spectroscopy [2,3,7–9]. They have then been applied in ESR spectroscopy [10] for cavity ringdown suppression [11,4,12], wide-band Fourier-transform...
The signal is finally demodulated using an IQ mixer, and its two rapid microwave switch makes sure that the sample is protected from the noise at the amplifier except when the pulses are on. The input line is strongly attenuated at the microwave switch, whereas the total damping rate \( \kappa = \kappa_r + \kappa_i \) also includes the internal cavity loss rate \( \kappa_i \).

The cavity is coupled to an ensemble of \( N \) spins 1/2 with spin operators \( S_{ij}^{(I)} \). The equations that govern the evolution of spin \( j \) are

\[
\begin{align*}
\dot{S}_l^{(l)} &= -\Delta S_l^{(l)} + g_j Y_l^{(l)} \\
\dot{S}_l^{(II)} &= \Delta S_l^{(II)} - g_j X_l^{(II)} \\
\dot{S}_l^{(I)} &= g_j X_l^{(I)} - g_j Y_l^{(I)},
\end{align*}
\]

where \( g_j \) is the spin-resonator coupling constant [21] which is proportional to the amplitude of \( B_1 \) at the spin location, and \( \Delta = \omega_L - \omega_B \) is the detuning from the resonator frequency. These are identical to the usual Bloch equations, with the Rabi frequency given by the product of the coupling constant \( g_j \) and the corresponding field quadrature amplitude [25]. The cavity field is coupled both to the input drive field quadratures \( \beta_{X,Y}(t) \) and to the spins via

\[
\begin{align*}
\dot{X}(t) &= \sqrt{\kappa_r} \beta_X(t) - \frac{1}{2} X(t) - \sum_{j=1}^N 2g_j Y_j^{(II)} \\
\dot{Y}(t) &= \sqrt{\kappa_r} \beta_Y(t) - \frac{1}{2} Y(t) + \sum_{j=1}^N 2g_j Y_j^{(I)},
\end{align*}
\]

Eqs. (1) and (2) enable to compute the intra-cavity field and spin dynamics for a given drive field \( \beta_{X,Y}(t) \), provided the distributions of spin Lambor frequency \( \rho_\delta(\Delta) \) and coupling constant \( \rho_g(g) \) (caused by the spatial inhomogeneity of the \( B_1 \) field) are known. Here we are interested in finding the drive fields \( \beta_{X,Y}(t) \) to achieve a given intra-cavity field profile specified by the quadratures \( X_p(t), Y_p(t) \). This problem is drastically simplified if the field generated by the spins (third term on the right of Eqs. (2)) is negligible compared to the intra-cavity field produced by the drive field. That is the case when the so-called cooperativity parameter \( C = \sum g_j^2 / (\kappa \Delta) \) verifies \( C \ll 1 \) [25] (\( \Delta \) being the characteristic width of the \( \rho_\delta(\Delta) \) distribution), which is the usual situation in both NMR and ESR. In the following we will assume to be in this situation. A targeted dynamics \( X_p(t), Y_p(t) \) is then obtained with the drive fields

\[
\begin{align*}
\beta_X(t) &= [X_p(t) + \frac{1}{2} X_p(t)] / \sqrt{\kappa_r} \\
\beta_Y(t) &= [Y_p(t) + \frac{1}{2} Y_p(t)] / \sqrt{\kappa_r},
\end{align*}
\]

which is identical to the formula derived in [2]. Our target pulse shape in this work is the so-called “bump pulse”, defined as

\[
X_p(t) = X_0 e^{-t^2/(2t_p^2)}, \quad Y_p(t) = 0,
\]

for \( |t| < t_p/2 \), and \( X_p(t) = Y_p(t) = 0 \) for \( |t| > t_p/2 \). \( X_0 \) denotes the maximum amplitude and \( t_p \) the pulse length. This pulse shape goes smoothly to zero at \( \pm t_p/2 \) and does not need to be truncated, contrary to Gaussian pulses for instance [25]. From Eq. (3), the time-dependent drive pulse needed to obtain a bump-shaped intra-cavity field is readily obtained.

In the experiment, we detect the field leaking out of the cavity. The latter contains both the reflected drive pulses, as well as the spin free-induction-decay and echo signals. The output field quadratures are given by the input-output relation

\[
\begin{align*}
\dot{X}_{out}(t) &= \sqrt{\kappa_r} X(t) - \beta_X(t) \\
\dot{Y}_{out}(t) &= \sqrt{\kappa_r} Y(t) - \beta_Y(t).
\end{align*}
\]

In the following, we resort to numerical simulations to compare our model to the experimental results. For that, we first compute the distributions \( \rho_g(g) \) and \( \rho_\delta(\Delta) \). We then discretize these distrib-

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**Fig. 1.** (a) Basic concept. A short microwave pulse with a rectangular envelope (left green line) is sent onto an ESR cavity and is subsequently distorted by the cavity’s transfer function. The resulting intracavity field (center green line) then drives the spins for much longer than intended resulting in spin-echo signals which are stretched in time (right green line). In contrast, if a tailored shaped pulse is employed (left blue line), the cavity distortion is compensated. A short intracavity field (center blue line) now drives the spins resulting in wide-band spin manipulation and shorter echo duration. (b) Schematics and optical images of the LC resonator used in the experiment. Dark areas represent the Si substrate while light areas are made of Al. Bi spins are implanted everywhere below the Si surface, in a box-like profile at a depth ~100 nm. The resonator active area is a 0.2, pl. volume around the 100 μm-long, 500 nm-wide inductance. (c) Technical implementation. Shaped drive pulses are generated by modulating the output of a continuous-wave microwave generator (LO) by two analog outputs of an AWG via an IQ mixer. A rapid microwave switch makes sure that the sample is protected from the noise at the amplifier except when the pulses are on. The input line is strongly attenuated at low-temperatures in order to thermalize the microwave field to millikelvin temperature. The pulses are routed to the sample via a circulator, and the reflected signal is routed to the output line. It then undergoes amplification at several stages, starting from a JPA at 12 mK, followed by a HEMT at 4 K and then at room-temperature. The signal is finally demodulated using an IQ mixer, and its two quadratures digitized. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
butions into $N_x \times N_z$ bins of spins with identical coupling and frequency. We integrate $N_x \times N_z$ times Eq. (1) and finally compute the cavity and output fields using Eqs. (2) and (5).

All that precedes is the standard treatment of inductively-detected magnetic resonance, be it nuclear or electronic. One specific aspect of our experiments is that due to the small mode volume, high quality factor of the superconducting micro-resonator, and to the low temperature of the experiments, spins relax towards thermal equilibrium dominantly by spontaneous emission of a microwave photon into the cavity [26] - the so-called Purcell effect. The Purcell relaxation rate for spin $j$ is given by

$$\Gamma_{ij} = \kappa - \frac{g_j^2}{\Delta_j^2 + \kappa^2/4}. \quad (6)$$

In our experiments, Purcell relaxation plays no role during a spin-echo sequence because it occurs on a much longer timescale, which is why it was not included in Eqs. (1). It however plays a role in the steady-state polarization profile of the spin-ensemble, because of the dependence of the relaxation rate on the detuning and coupling constant. We take this phenomenon into account by initializing the density matrix of spin $j$ at $t = 0$ as $\rho_j = -1 + e^{-t/\Delta_j} |S_j; t\rangle \langle S_j; t|$, being the repetition time of the experimental sequence.

3. Materials and methods

The spectrometer is built around a superconducting micro-resonator that was described in [23]. It consists of an Aluminum thin-film patterned as a lumped-element LC circuit with a 100 μm-long, 500 nm-wide inductance, on top of a silicon substrate [see Fig. 1(b)]. This inductance defines the active area of the resonator around which $B_1$ is sizeable and spins are therefore detected; the corresponding mode volume is ~0.2 pl. Finite-element modelling yields the spatial distribution of the $B_1$ field generated around the inductance, which in turn allows us to compute the coupling constant density function $\rho_s(g)$ needed in the simulations. As explained in [23], the average coupling constant over the detection volume is found to be $g/2\pi \approx 450 \text{ Hz}$.

The resonator chip is mounted in a sample holder which is cooled down at 12 mK in a dilution refrigerator. The 50 Ω coaxial line used for delivering the control pulses and collecting the signal is connected to the sample holder with a SMA bulkhead to which an antenna is soldered [21]. By tuning the antenna length, one can adjust the capacitive coupling of the resonator to the line and thus its quality factor ($Q \approx 4 \times 10^4$ in this work). Spectroscopic measurements allow us to extract the resonance frequency $\omega_0/2\pi = 7.274 \text{ GHz}$, the coupling rate $\kappa = 7.1 \cdot 10^3 \text{ s}^{-1}$, the internal energy loss rate $K_i = 2.8 \cdot 10^3 \text{ s}^{-1}$ (measured at the single photon limit), and the total energy loss rate $\kappa = 10^5 \text{ s}^{-1}$. The corresponding field decay time is $\tau = 2\kappa/\kappa = 2\mu s$.

An overview of both the low-temperature part of the setup and the room-temperature electronics is schematically shown in Fig. 1 (c). The input line is heavily attenuated at low temperatures to thermalize the microwave field close to the cryostat base temperature. A circulator routes drive pulses from the input waveguide towards the sample, and the reflected signal (together with the spin signal) towards the output waveguide. It is amplified first at 12 mK by a quantum-limited Josephson Parametric Amplifier (JPA) [27], then at 4 K by a High-Electron-Mobility-Transistor (HEMT), and at room-temperature for the final amplification stage. After demodulation by mixing with the local oscillator, the down-converted signal is sampled by a digitizer yielding the field quadratures $I(t), Q(t)$ (in the following only the signal-carrying quadrature is shown). Note that due to the unusually small resonator mode volume, low microwave powers (of order picoWatt) are needed to drive the spins; as a result, we can measure with the exact same setup both the drive pulses after their reflection at the resonator input and the spin signal, which is useful for the shaped pulses characterization. More details on the resonator, JPA, and spectrometer design, can be found in [21,23]. In [23] the spin detection sensitivity was estimated to be 65 spin$/\sqrt{\text{Hz}}$ for the same setup but with a quality factor of $8 \cdot 10^4$. Assuming that the sensitivity scales like $k$ (since the echo amplitude scales like $1/\sqrt{k}$ and the repetition time like $k$), we estimate it to be in the present case ~150 spin$/\sqrt{\text{Hz}}$.

One issue with superconducting micro-resonators is their low power handling capability. Indeed, in a superconducting strip such as used for the inductance, non-linear effects appear when the current density approaches the critical current density of the film. For Aluminum at low temperatures it is of order $10^{11} \text{ A/m}^2$ [28], which implies that in our geometry non-linear effects are expected for ~1 mA ac current in the wire. Because of the large spin-resonator constant however, spins can be driven efficiently well below that threshold as will be shown in the following.

In order to generate microwave drive pulses with arbitrarily-controlled amplitude and phase, we modulate the continuous signal delivered by a microwave synthesizer (the local oscillator LO) using two analog outputs of an AWG (model Tektronix 5014), using an IQ mixer. The AWG sampling rate is 1 Gs/s, and its analog bandwidth is 300 MHz. After calibration of the IQ mixer, LO leakage was ~60 dB. At the mixer output, the pulse goes through a voltage-controlled attenuator and is then amplified to obtain sufficient driving strength. Right before the low-temperature part of the setup, a fast microwave switch controlled by a digital output of the AWG with 80 dB dynamics transmits microwave signals into the cryostat only during the application of the drive pulses. This protects the spins from the amplifier output noise, which would otherwise reduce their equilibrium polarization [29]. Phase cycling is performed using the IQ modulation by the AWG.

To test the spectrometer operation with shaped pulses, we use the electron-spin resonance of bismuth donors in silicon as a model system. Bismuth atoms were implanted into the silicon sample on which the resonator is patterned, at a typical depth of 100 nm. At low temperatures, bismuth atoms can bind an electron; details on the spin Hamiltonian of such bismuth donors can be found in [30,31]. A field $B_0 = 3.74 \text{ mT}$ is applied in order to tune the lowest-frequency transition of the bismuth donors in resonance with $\omega_0$. As explained in [21,32,23] the spin resonance of the donors in our experiment is broadened by strain in the substrate caused by thermal contraction of the thin-film resonator upon cooling, which affects inhomogeneously the hyperfine-coupling constant between the bismuth nuclear and electron spins. The resulting spin linewidth was measured to be of order ~20–30 MHz [23], considerably larger than the 200 kHz resonator linewidth. This situation is frequently encountered in ESR spectroscopy. In the simulations we will therefore model the spin density $\rho_s(\Delta)$ as being a constant.

4. Experimental results

4.1. Shaped pulse characterization

In NMR and ESR experiments using shaped pulses, the intracavity field is often characterized by inserting a pickup coil in the resonator [3,5,17], or by using the Rabi nutation of the spins [5]. Here we directly measure the reflected drive pulse, and use it to extract the intra-cavity field. This is facilitated by the low microwave power (picoWatts at the sample level) of the drive pulses used in our experiments, which is itself a consequence of the large spin-resonator coupling.
As can be seen from Eq. (5), the intra-cavity field can be obtained by subtracting the reflected signal from the incident drive pulse, both measured with the very same setup. To do so, we measure the reflected pulse at \( \omega_0 \), whereas the incident pulse is measured by applying it 10 MHz off-resonance from \( \omega_0 \), so that it is fully reflected at its input without deformation. Because of the small frequency difference, the transfer function of the whole setup can be considered identical in both cases, so that the signals can be simply subtracted, yielding the intra-cavity field \( X(t), Y(t) \).

Examples are shown in Fig. 2 (only the quadrature carrying the signal is displayed). For a 1 \( \mu \)s-long square-shaped pulse, the intra-cavity field shows a linear rise during the pulse, followed by an exponential decay with a time constant of \( 2 \beta / k \) as expected. We then generate a “bump” pulse with \( t_p = 1 \mu \)s (Eq. (4)), using Eqs. (3) to compute the AWG drives [2]. The measured intra-cavity field of the bump pulse is in excellent agreement with the expected pulse shape as given by Eq. (4), with the maximum amplitude as the only fitting parameter. Transients are efficiently suppressed, by at least an order of magnitude; and the intra-cavity field rise- and decay time is \( 250 \) ns, an order of magnitude shorter than the cavity field decay time \( 2 \beta / k \). This successful transient suppression also confirms that the resonator remains linear upon application of the drive pulses (square as well as bump).

4.2. Echoes with shaped pulses

We proceed with the measurement of spin-echoes, using a \( \pi / 2 - \tau - \pi - \tau \) Hahn-echo sequence with \( \chi / \chi \) phase-cycling on the first pulse. To optimize the pulse amplitude, Rabi nutation data was taken by measuring the integrated echo amplitude \( A_e \) as a function of the refocusing pulse amplitude for a square pulse shape (see Fig. 3). Damping of the oscillations is caused by the spread in Rabi frequency \( \gamma(g) \) due to \( B_1 \) spatial inhomogeneity, and is qualitatively reproduced by the simulations. Fig. 3 also shows the reflected signal quadrature of a full echo trace, including the echo, both for 1 \( \mu \)s-long square-shaped and bump-shaped pulses. The reflected control pulses again demonstrate transient suppression in the bump-pulse case. The bump-pulse echo is both shorter and higher-amplitude than the square-pulse. These features are well captured by simulations.

We study the shaped pulse echo in more details in Fig. 4. In order to measure the contribution from spins outside of the cavity bandwidth, a low repetition rate is necessary, since those spins relax more slowly than spins at resonance because of the Purcell effect (see Eq. (6)). Fig. 4 shows a time trace of a Hahn echo, measured using bump pulses with \( t_p = 1 \mu \)s, and a repetition time
The echo shows a pronounced asymmetric shape, with a sharp rise in less than a microsecond, and a slower decay. The fact that the echo signal rises faster than $2/\kappa$ demonstrates that the echo originates from the rephasing of spins lying in a broader frequency range than the cavity bandwidth. After the echo reaches its peak amplitude, it decays in $\sim 2/\kappa$. Indeed, despite the use of shaped pulses to drive the spins over arbitrarily wide bandwidth, their emission remains unfiltered by the cavity [5]. To visualize more quantitatively the dynamics of the spin-ensemble magnetization, we have numerically deconvoluted the cavity response (see Fig. 4a inset), resulting in a Gaussian-shaped time dependence with a FWHM width of 1.3 $\mu$s. The Fourier transform of the magnetization is also shown in Fig. 4, and shows a linewidth 4 times greater than $\kappa$. Numerical simulations were performed using the computed distributions $\rho_x$ and $\rho_y$, based on the equations of Section II, and taking into account the Purcell relaxation. The echo shape and duration are quantitatively reproduced, as well as the spin magnetization dynamics.

One motivation for short control pulse duration is their higher robustness to spin detuning. Indeed, the nutation frequency of spin $j$ is given by $\sqrt{\omega_0^2 + \Delta^2}$, with $\omega_0 = g \sqrt{\Delta^2 + \gamma^2}$. Due to the transient decay, square-shaped pulses have an effective minimum duration of order of $2/\kappa$, implying that the maximum value of $\omega_0$ for a $\pi$ pulse is $\sim \kappa$. Spins on the edges of the resonator bandwidth (those with $\Delta \sim \kappa/2$) therefore unavoidably undergo a Rabi nutation with an angle and axis that are significantly different from those at resonance with a square-shaped pulse. Bump-shaped refocusing pulses on the other hand have a shorter duration, leading to higher $\omega_0$ for a $\pi$-pulse and thus to better performance for spins with $\Delta \sim \kappa/2$. To test this prediction, we compare the signal-to-noise ratio of an echo obtained with the same square-shaped $\pi/2$-pulse, but with either a bump- or square-shaped refocusing $\pi$ pulse. The signal and noise are given by $A_x = \int I(t)u(t)dt$ and $\sigma = \sqrt{\int [I(t)]^2 dt}$, respectively, where $u(t)$ represents the mode shape of the echo normalized such that $\int [u(t)]^2 dt = 1$ [21]. This mode shape is obtained by fitting a skewed Gaussian function to the echo signal

$$u(t) = \frac{1}{\mathcal{N}} \exp \left[ -\frac{t - t_0}{2\sigma^2} \right] \text{erfc} \left[ \frac{t - t_0}{\sigma \sqrt{2}} \right],$$

where $\mathcal{N}$ is the normalization, $\sigma$ the scale and $\alpha$ the shape factor. We find a SNR improvement of 16% in the bump refocusing pulse case compared to square-pulse refocusing, which confirms the better performance of bump-shaped refocusing pulses.

Suppression of cavity ringdown enables to minimize the delay between control pulses, which is particularly useful for dynamical decoupling pulse sequences where refocusing pulses are applied shortly one after the other. As a proof-of-principle, we run a Carr-Purcell-Meiboom-Gill pulse sequence using bump pulses. Even with a delay of 10 $\mu$s, the echoes are still clearly separated as seen in Fig. 5; whereas with square pulses the minimal separation was found to be closer to 50 $\mu$s. Because of the short delay and long coherence time of donors in silicon (particularly with silicon isotopically enriched in nuclear-spin-free $^{29}$Si as in our experiment), $\sim$ 1000 echoes can be measured in this way (see Fig. 5). Such a long echo train may be used in particular for further increasing the single-shot signal-to-noise [33,23]. The shorter inter-pulse delay within the CPMG sequence reduces the sensitivity to higher frequency noise contributions, which enables more echoes to be generated and thus further SNR enhancement. Here, assuming a white-noise background, the CPMG averaging would translate into a SNR improvement as high as a factor 20. Recent experiments however have shown that this figure is overestimated in the presence of low-frequency or correlated noise, which may occur in experiments with SC resonators due to resonator phase noise [34,23] or long-term spin fluctuations.

5. Conclusions

We have demonstrated cavity ringdown suppression and wide-band spin excitation using shaped control pulses in a high-sensitivity, quantum-limited ESR spectrometer at millikelvin temperatures. So far only bump pulses were used, which were shown to robustly suppress cavity transients and increase the SNR of Hahn-echoes by 16%. Implementing optimal control techniques for special-purpose pulse sequences [17,11,18,35] would be a natural next step to further improve the level of spin control achieved. Also, the ringdown suppression demonstrated here would enable to measure spins with short coherence times, bringing quantum-limited ESR spectroscopy one step closer to real-world applications.

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