Pulsed and continuous-wave magnetic resonance spectroscopy using a low-cost software-defined radio

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ABSTRACT
Software-defined radios (SDRs) constitute a modern and highly adaptive realization of a radio-frequency transceiver system. This work demonstrates how a particular radio transceiver, namely, the LimeSDR board, can be configured for pulsed and continuous-wave magnetic resonance spectroscopy. As a first step, the board needed to be extended by a bit pattern generator, so as to facilitate synchronization of other spectrometer equipment. The upgraded board was incorporated into two different spectrometers, namely, into a pulsed spectrometer operating at either 30 MHz for nuclear spins or 150 MHz for electron spins and into a continuous-wave spectrometer at 150 MHz for electron spins. Pulse sequencing capabilities were approved by relaxometry and Rabi oscillations of electron spins at a time resolution of 33 ns. Upon exhaustive averaging of acquired transients, unwanted oscillations that are characteristic for fast analog-to-digital converters emerged out of the noise floor. Methods for cancellations of these oscillations are presented, in particular a new acquisition scheme with cyclic incrementation of the acquisition position. The continuous-wave spectrometer provided derivative spectra of absorption and dispersion by phase-synchronous sideband demodulation. Furthermore, this spectrometer featured a software-defined automatic frequency control to account for probe drift, which has been incorporated into the open-source spectrometer control software.

I. INTRODUCTION

Electronics built for communication systems and information technology achieve ever more complex functionality at high performance and low cost. Accordingly, a number of chip modules are nowadays available that interface directly to radio-frequency (RF) and microwave domains. As an example, RF transceivers with highly programmable analog characteristics became available in order to have one single chip that complies with multiple communication standards. Owing to this flexibility, such chips are termed field-programmable radio-frequency (FPRF) chips. Clearly, this nomenclature is inherited from its established digital counterpart, namely, the field-programmable gate array (FPGA). Such FPRF chips are often found in software-defined radios (SDRs), which constitute a modern realization of radios with signal processing largely relying on computational resources. As a result, the entire communication system ranging from the software interface to the RF front-end can be adjusted to the intended purpose.

Thanks to the coverage of RF and microwave frequency domains, communication electronics proved of course also very useful in applications other than communication systems. For magnetic resonance spectroscopy that is of relevance in this study, such electronic devices found use in a number of home-built spectrometers. In nuclear magnetic resonance (NMR), for instance, a number of low-cost spectrometers based on FPGAs or chip modules, as well as spectrometers based on SDRs, have been proposed. The motivation to reduce the footprint of NMR spectrometers even triggered the design of application-specific integrated circuits (ASICs). The situation is similar in electron paramagnetic resonance (EPR), where operation frequencies can range up to several gigahertz and the involved time scales of transient spin precession are generally shorter than in NMR. In particular,
spectrometers that rely on FPGAs or chip modules\textsuperscript{26–29} or on ASICs\textsuperscript{30–31} were introduced.

In general, the design of such home-built spectrometers is motivated by (i) enhanced functionality for development of new spectroscopic methods, (ii) reduced system size for portability, or (iii) reduced costs as compared to a commercial spectrometer. However, an obstacle for the construction of such spectrometers is imposed by the knowledge required by the targeted electronic component and its interface. As an example, high-performance analog-to-digital converter (ADC) chips can be bought at rather low cost, but a paramount effort needs to be taken to transform such a chip into an actual digitizer ready for spectroscopy. Accordingly, only few specialist groups can benefit directly from the availability of state-of-the-art integrated circuits. Indirectly, of course, the innovation in electronic components manifests in improved performance of test and measurement instruments, such as digitizers, arbitrary waveform generators, synthesizers, or lock-in amplifiers. These instruments are certainly easier to assemble into a spectrometer system since these are delivered with a documented software interface.

A paradigm shift is expected from open-source hardware platforms. These devices unify state-of-the-art hardware with a ready-to-use open-source interface at low cost. Due to the open interface, application-oriented adaptations to the hardware platform become feasible without building an entire system from scratch. This study focuses on such an open-source hardware platform, namely, the LimeSDR board. This low-cost SDR board is equipped with an FPRF that is interfaced by an FPGA and a universal serial bus (USB) 3.0 controller. Already in a previous study,\textsuperscript{32} the open-source firmware of the LimeSDR’s FPGA could be adapted for pulsed NMR spectroscopy. In this work, a different approach for the integration of the LimeSDR into a spectrometer is presented. Moreover, this approach is tested with pulsed NMR and EPR, as well as with continuous-wave (CW) EPR.

This paper is organized as follows. First, a description of the relevant specifications of the LimeSDR and of the spectrometers for pulsed and CW experiments is given in Sec. II A. Since the software of the spectrometers is available for further use, a separate description of the software follows in Sec. II B. The experimental results for pulsed NMR and EPR are presented and discussed in Sec. III. In addition to spectroscopic results, a dedicated aspect of signal acquisition is treated separately in Sec. III C, namely, the cancellation of unwanted oscillations introduced by the ADC. Subsequently, the experimental results for CW EPR are presented in Sec. IV, and the study is concluded in Sec. V.

II. SPECTROMETER DESCRIPTION

A. Hardware

1. LimeSDR board

The LimeSDR-USB is equipped with two full-duplex RF channels. Each channel has therefore a transmission (TX) output and a reception (RX) input that can be accessed simultaneously. The actual transition from digital data to analog RF signals takes place within the FPRF chip on the LimeSDR board. A simplified schematic of the FPRF is shown in Fig. 1(a). This chip interfaces to the digital domain by 12 bit digital-to-analog and analog-to-digital converters (DAC/ADC). The maximum rate at which the output of these converters can be changed is determined by the maximum bandwidth of the USB interface (61.44 MSa/s). In principle, faster conversion rates are possible by means of interpolation and decimation that is available within the FPRF’s built-in digital processing unit. In this work, however, this was not used. Among other reasons (see below), interpolation would flatten the pulse flanks of excitation pulses.

On the analog side of the converters, the corresponding baseband (BB) signals are translated to RF by mixing with a local oscillator (LO). The frequency translation is realized by single-sideband modulation, therefore using a pair of mixers (and a pair of BB signals) in phase quadrature. For each pair of mixers, gain and phase balances as well as DC offsets can be adjusted so that the mixing can be optimized within the targeted frequency range. The available RF range is determined by the LO range (30 MHz–3.5 GHz) and the maximum feasible BB modulation bandwidth (61.44 MHz). Within the FPRF, different RF paths can be selected to optimize the performance for a certain frequency range. Moreover, each of these paths has its own RF matching network on the LimeSDR board. As the frequency range in this study was below 200 MHz, an inductor (with label MN18) needed to be removed in order to improve the RX performance.\textsuperscript{32}

In order to use the LimeSDR as a spectrometer, several conditions need to be fulfilled. First of all, the RX and TX channels need to be phase coherent, which is achieved by selecting the same LO source for the RX and TX mixers. Second, a time synchronization between RX and TX is required, which is established within the FPGA by means of hardware timestamps. In fact, the primary task of the FPGA is to relay digital data between the FPRF and the
USB interface. Digitized RX samples from the FPRF are grouped into packets (1360 samples) and complemented with a timestamp, which is the total number of received samples since the stream has been started. TX sample packets (1020 samples) from the host computer can be scheduled at a specific timestamp, which needs to be a multiple of 4080. Note that the packet size for TX and RX samples is different because a 16 bit representation is intentionally selected for TX samples, whereas the RX samples have a direct correspondence to the 12 bit ADC. TX samples have thus an unused overhead of 4 bits.

This overhead is important for the last condition to use the LimeSDR as a spectrometer, namely, for synchronization with external hardware. In its native configuration, those 4 overhead bits are left disregarded in the FPGA. By modification of the FPGA firmware, these 4 bits have been routed to the general purpose input-output (GPIO) pins of the FPGA. In this way, a four channel bit pattern generator is added to the LimeSDR. Importantly, this firmware modification is fully compatible with the existing application programming interface (API). One only needs to combine the 12 bit DAC samples with the 4 GPIO bits into a single 16 bit value and respect the corresponding sample rates. In fact, the four GPIO bits have twice the sampling rate of the DACs. The reason is that the DACs are operated synchronously as a pair for quadrature modulation, thus requiring twice the sampling rate from the FPGA. Furthermore, it is worth mentioning that a total of 8 GPIO pins are available on the FPGA. One could therefore also envisage buffering the 4 overhead bits to all 8 pins, which would then run at the same sampling rate than the DACs. Note that since the GPIO is in low voltage transistor-transistor logic (LV-TTL), a logic level translator and line buffer were added to provide 5 V TTL signals. In pulsed experiments, these TTL signals can, for instance, be used for gating and blanking. As will be discussed below, TTL capability was also important for CW experiments. Accordingly, the LimeSDR with the modified firmware provided the RF and TTL lines that are necessary for basic experiments.

In a previous account on a pulsed NMR spectrometer with the LimeSDR, a different synchronization strategy has been pursued. In particular, an external bit pattern generator has been used as the acquisition trigger for the LimeSDR via a GPIO pin. In addition, the synchronization of multiple LimeSDR devices has been considered. Also in this case, a modification of the firmware was necessary since the LimeSDR did not provide these capabilities at the time of writing. In brief, the trigger on the GPIO pin modified the timestamp within the FPGA so that the host computer could identify RX packets during the acquisition trigger. Moreover, the timestamp of the RX trigger served as basis to schedule subsequent TX data.

The study point at a subtle issue, namely, the clocking of the converters within the FPRF. The clocks of DAC and ADC are derived from the same master clock that runs four times faster. However, different clock dividers are used to generate the TX and TX clocks. Since these dividers are initialized independently of each other, there are multiple possibilities for the relative phase of the DAC and ADC clocks. As an example at an ADC/DAC sampling rate of 30.72 MHz, the master clock is at 122.88 MHz. Depending on clock initialization, the converter clocks can thus have a relative offset of N⋅8.1 ns, where N ranges from 0 to 3. Using another firmware modification, this relative offset has been monitored and set to zero by intentional reinitialization of the clocks. In this way, TX pulses scheduled upon a RX trigger had a well-defined timing, even from two different LimeSDR boards. In this study, such initialization-dependent offset between ADC and DAC clocks was not considered. Importantly, this clock offset has no effect on the timing between the TX output and the GPIO trigger outputs. In particular, the GPIO trigger outputs are clocked at a rate that is phase-locked to the DAC sampling rate. Note, however, that this is only true as long as DAC samples are not interpolated to a faster sampling rate within the FPRF and as long as one single channel is used. Otherwise, the DAC sampling clock is susceptible to be relayed to the FPGA upon clock division, which introduces initialization-dependent offsets.

2. Setup for pulsed experiments

In order to test pulsed operation, a basic setup using a tuned coil as a probe was deployed. The setup is shown in Fig. 1(b). Pulse sequences from the LimeSDR TX output were sent to a high power amplifier (HPA; NMR: TOMCO BT04000-AlphaS, EPR: Kalmus LP1000). The amplifier was blanked with a TTL trigger from the LimeSDR with calibrated timing. In order to maximize power transmission to the coil and counteract saturation of the receiver, crossed diodes and a quarter-wavelength transformer were used. The receiver consisted of a low noise amplifier (LNA; NMR: 46 dB gain with NF Corporation SA-220ES and a 50 Ohm resistor at its input, EPR: 20 dB gain with Avantek UAA916B) and another set of crossed diodes to protect the LimeSDR input. For NMR at 30 MHz, the probe was a saddle coil with a diameter of 14.5 mm and the sample had a diameter of 3.5 mm and a height of 8 mm. The magnetic field \(B_0\) of 0.7 T was provided by an electromagnet (Drusch et Cie). For EPR at 150 MHz, the probe was a 7.5 mm long, 6 turn solenoid coil wound around a 5 mm tube. The sample was contained in a quartz tube with 2.4 mm inner diameter. The magnetic field \(B_0\) of 5 mT was provided by a magnet available from a previous study. In order to reduce the dead time for pulsed EPR, the quality factor of the coil was reduced to 10 by a 10 Ω resistor connected in series to the coil. Note that this introduces a loss in sensitivity and more elaborate probe designs in this frequency range make use of active circuits to absorb the high-power excitation pulses. In addition, a pronounced video leakage signal at few megahertz upon blanking of the HPA needed to be separated using a third-order frequency diplexer with cutoff at 54 MHz. The video leakage could therefore be absorbed into a 50 Ω resistor.

3. Setup for CW experiments

CW EPR at 150 MHz was based on the same sample and coil setup as for pulsed EPR. A schematic of the setup is shown in Fig. 1(c). In the absence of the series resistor, the coil had a quality factor around 90 and 25 dB return loss. The receiver LNA was identical as in pulsed EPR; however, no driving amplifier was used for transmission. An isolation of 40 dB between transmission and reception was achieved by a directional coupler (Mini Circuits ADC-15-4), at the expense of 15 dB attenuation for transmission. The excitation tone at 3 dBm was therefore attenuated to ~12 dBm before reaching the coil. Based on the geometry of the coil and its resistance of 1.1 Ω, a driving field \(B_1\) on the order of 77 mG was estimated.
Modulation of the main field $B_0$ was achieved by an additional saddle-shaped coil that was tuned to 105 kHz and provided a field of 0.68 G/V. Two different devices were used to drive this coil. The first consisted of an ordinary function generator (Agilent 33522A), whose 10 MHz reference was used as a reference for the LimeSDR. The second device was a custom circuit that generated a sine-wave from a TTL square wave at 105 kHz [gray box in Fig. 1(c)]. Using this circuit, the field could be modulated using a phase-coherent TTL trigger from the LimeSDR. The circuit consisted of four parts: (i) a unity-gain 105 kHz bandpass filter with the Sallen-Key topology ($R = 1k$, $C = 2 \text{nF}$), (ii) a fifth-order Butterworth filter with cutoff at 130 kHz ($R = 5k$, $L = 10 \text{mH}$, $C = 2 \times 150 \text{pF}$, and $1 \times 470 \text{pF}$), (iii) a variable gain stage that can be trimmed with a potentiometer, and (iv) an output driver (National Semiconductor LM 6321). The resulting sine wave had its higher harmonics reduced to below -40 dB of the principal oscillation, whose maximum amplitude was 3 V_{PP}.

**B. Control software**

The LimeSDR was controlled from the host computer (Lenovo Thinkpad X1 Yoga, 2016) via USB 3.0. The control software had two components [see Fig. 1(a)]: (i) a low-level C++ routine that interacts with the LimeSDR through the provided API and (ii) a Python class to ease scripting of the spectrometer. In Secs. II B 1 and II B 2, the most relevant details on these programs are given. For further insight, the entire code is available for noncommercial use.\(^{9,11}\) Note that the gateware was modified from version 2.17 from June 2018, which was used with API version 18.04.19. At the time of preparing this manuscript, the gateware evolved to version 2.21 with corresponding API updates, indicating active development and rectification of pending issues.

1. Low-level C++ routine

In brief, the C++ routine contains data buffers for the TX and RX data that are continuously streamed to/from the LimeSDR. On the TX side, all the waveforms that define the requested experiment are calculated and scheduled. On the RX side, the received data packets are inserted into the output buffer, averaged, and stored. If the experiment is finished, the RX data and all the input parameters are stored using the hierarchical data format (HDF 5) standard. Apart from this general structure, different low-level routines were used for pulsed and CW experiments.

For pulsed experiments, the routine accepts a one-dimensional pulse sequence specification, with the only variable parameter being the pulse phase. The routine therefore realizes a phase-cycled pulse sequence with constant timing and pulse flip angles. The RX data for the different pulse phases are stored in different output buffers so that the necessary receiver phase cycle can be applied during postprocessing. Note that the smallest possible buffer size is 4080 samples, which is due to the requirement on scheduling TX packets at a certain RX timestamp. At the used sampling rate of 30.72 MSa/s, this corresponds to 133 $\mu$s. All the pulse experiments even used a minimum buffer size of 12 240 Sa, thus 398 $\mu$s. For pulsed EPR, this time scale is much longer than the typical duration of echo transients. Accordingly, only a fraction of the output buffer contains the relevant data. Moreover, a number of spin systems restore equilibrium faster than 398 $\mu$s so that several sequence repetitions are possible within the utilized buffer size. In the EPR results, up to 50 repetitions within 398 $\mu$s were used, so as to achieve a repetition rate of 126.4 kHz. For pulsed NMR, the minimum buffer size is generally short as compared to the duration of recorded transients. The resulting output data can thus attain impractical sizes and decimation to a lower sample rate is advisable.

For CW experiments, the pulse routine has been adapted. The three most important changes were (i) inclusion of field control for field-swept spectra, (ii) calculation of Fourier components of the RX output buffer for demodulation, and (iii) implementation of an automatic frequency control (AFC) scheme.\(^{12,13}\) The CW routine thus realizes a two-dimensional CW experiment, with an optional phase cycle in the first dimension and a field sweep in the second dimension. Due to the demodulation, only data at the requested frequencies was stored in the HDF file. Otherwise, the data buffer used in CW experiments would result in very large files. In particular, CW experiments were performed at a conversion rate of 15.36 MSa/s and a buffer of 1.3 Msa, thus representing a t_{buff} = 87 ms long time window. Note that except for the LO frequency, all frequencies in the experiment were an integer multiple of the inverse buffer period 1/t_{buff}. The concerned frequencies were the principal CW tone at BB synthesized by the DAC, eventual amplitude-modulation sidebands of this principal tone for AFC, and the field modulation frequency.

Concerning the amplitude-modulation sidebands for AFC, one can choose up to four AFC frequencies. Each modulation frequency results in two sidebands, whose Fourier coefficients are stored in the output file. For prototyping purposes, the frequency deviation between the CW drive and the cavity was calculated based on the sideband amplitudes, assuming a linear response between the amplitude deviation and the frequency shift. The speed of the AFC feedback depended on several parameters. The control software required three buffer periods to update all involved frequencies: one buffer period was required to prepare a complex oscillator $\hat{Y}_{\text{H}}$ at the shift frequency in a separate buffer, based on a lookup table. During the subsequent buffer period, the software needed to wait until updated TX data packets could be scheduled. As soon as this took place, the requested TX data packets were shifted by multiplication with the shift oscillator $\hat{Y}_{\text{H}}$ and relayed to the TX stream. In the third buffer period, the CW frequency was therefore updated. Concurrently, the oscillators to calculate Fourier components were shifted in frequency during reception of data packets with the updated frequency. When using two AFC frequencies and when averaging AFC readings over three buffer periods, the total feedback time was 435 ms and the calculation load of this software-defined AFC could be sustained without interrupting the continuous data stream at 15.36 MSa/s. As it turned out, the actual frequency shifts took place on a much slower time scale. In order to avoid additional noise introduced by the AFC feedback, much longer averaging times to calculate the frequency shift have been used in the experiments (see Sec. IV C).

2. Python class for scripting

A Python class called limr serves as the main interface for spectrometer control. In essence, the limr class contains all the parameters that are required for the low-level C++ interface. Furthermore, an additional arbitrary one-dimensional parameter sweep can be added. All the pulse sequences that are presented in Sec. III B were
realized by adding such an additional parameter sweep specification. For CW experiments, an additional sweep was not used since the low-level routine already provided the capability for recording field-swept CW spectra. Nevertheless, such an additional sweep could be of interest for CW, as for instance for power saturation studies.

In order to give an example, Fig. 2 illustrates the script for an echo sequence with swept interpulse delay. All the variables that are relevant for the experiment are identified with a three-letter label, as for instance lof, which stands for LO frequency. Based on the code comments, the designations can be identified within Fig. 2. As the sequence has two pulses, any pulse parameter is provided as a list with two entries. The phase cycle is specified using two variables that determine the number of phases, pcn, and the stacking level of multiple cycles, pcl. In this example, the first pulse has two phases and is at the lowest stacking level. The second pulse has four phases and is stacked on top, resulting in an exorcycle that selects exclusively pathways due to echo refocusing. Moreover, the parameter pba prioritizes phase cycling before averaging. The phase is thus varied at the repetition rate of the experiment. This has the advantage that not only static receiver offsets and imbalances can be canceled, but also fluctuations with time scales longer than the repetition time.

Another specification that requires further clarification is the TTL trigger at the GPIO output pin 1, which is programmed by the variable t1d. The timing of this trigger is always relative to the start and end flanks of the RF pulses. The timing specification used in the example means the following: the TTL trigger starts 150 samples before an RF pulse (pad0) and ends exactly with the RF pulse (pad1). In addition, a global delay of 50 samples (shift) is applied to align the TTL trigger to the RF pulse.

The arbitrary parameter sweep is implemented at the very end of the script (l.parsweep), just before the experiment is executed (l.run). Using this sweep command, any variable can be swept. In the example here, the variable pof, i.e., the pulse delay, is modified individually for each of the two pulses. The parameter sweep specification involves an initial and a final data point, followed by the total number of sweep points and by an optional pulse index. In principle, it would be sufficient to sweep only the delay between the first and the second pulses (pof with pulse index 1). However, this would lead to a shift of the echo position in the RX output buffer. For a constant echo position throughout the experiment, the position of the first pulse is also shifted (pof with pulse index 0).

Overall, this scripted sequence specification employs an entirely parameterized representation of the experiment. Thanks to this parameterization, parameter sweeps that define a specific pulse sequence can be implemented following a rather general strategy. Moreover, the fact of defining the sequence directly within Python gives access to a number of mathematical functions to support sequence scripting. For comparison, one should emphasize that for a number of pulsed spectrometers, the experiment is defined as a text file with a strict input syntax so that it can be read by a pulse interpreter. With such a scheme, the time flow of the experiment might be more evident than with a parameterized specification. However, the degrees of freedom for sequence specification are certainly enhanced with the integration into a scripting environment. This is especially important when incorporating excitation pulses with complex excitation patterns. At least for a previous spectrometer that relied heavily on such shaped pulses, this concept has proven to be vital.

III. PULSED MAGNETIC RESONANCE

A. Pulsed NMR at 30 MHz

Pulsed NMR was tested at an intermediate field strength of 0.7 T. The principal aim of this experiment was to affirm that the same information can be obtained with the LimeSDR as compared
to an ordinary spectrometer. In particular, a locally built spectrometer available from previous studies\textsuperscript{45,46} served as a reference. This spectrometer was connected in the same way to the HPA, coil, and LNA as illustrated for the LimeSDR in Fig. 1(b). In addition, the principal layout of the reference spectrometer was the same as for the FPRF of the LimeSDR [see Fig. 1(a)]. The received signal was therefore frequency-translated by mixing with two RF LOs in phase quadrature. Subsequently, the down-converted BB signals were low-pass-filtered with cutoff at 12 kHz and digitized at 100 kSa/s. The benchmark experiment was a FID upon a 20 μs long excitation pulse. The sample consisted of water that was doped with NiCl\textsubscript{2} (3 g/l), which allowed for a repetition time of 400 ms.

Figure 3(a) shows the resulting FID for the LimeSDR (black) and the reference spectrometer (gray). As is readily seen, the principal decay signal is of comparable shape and amplitude. However, the trace from the LimeSDR appears more noisy. As clarified in the following, this is solely due to the difference in the receiver bandwidth. While the reference spectrometer had its bandwidth fixed at 12 kHz, the LimeSDR had a much larger analog filter bandwidth of 5 MHz. The NMR signal therefore occupied only a very small fraction of the available BB bandwidth. Thanks to this large bandwidth, the LimeSDR has been configured such that the (complex) BB signal was oscillating at −1 MHz. During data evaluation, the relevant frequency range was selected by digital frequency translation (by +1 MHz) and low-pass-filtering with a 100 kHz cutoff. Moreover, the sampling rate was decimated by a factor of 10 from 7.68 MHz to 0.768 MHz. During data evaluation, the relevant frequency range was selected by digital frequency translation (by +1 MHz) and low-pass-filtering with a 100 kHz cutoff. Moreover, the sampling rate was decimated by a factor of 10 from 7.68 MHz to 0.768 MHz in order to reduce the number of data points. As a consequence, the FID obtained with the LimeSDR had a bandwidth that was about 8 times the bandwidth of the reference spectrometer, which explains the enhanced noise contribution in time domain.

Most importantly, the spectral noise density was comparable for both spectrometers. This is illustrated in Fig. 3(b), where the magnitude spectrum around the NMR peak is shown on a logarithmic scale. As is clearly seen, the spectral noise level is comparable. Since the dispersive component of the FID introduced a quite broad background onto the magnitude spectrum, the noise level has also been compared in the absence of any input signal. The resulting noise spectra in Fig. 3(c) corroborate that the spectral noise floor is the same within the bandwidth of the reference spectrometer. Note that it is the spectral noise contribution that is of relevance for NMR spectroscopy. Keeping in mind that pulsed experiments are rarely single shot measurements as presented here, it has also been verified that the noise floor follows a \( \sqrt{N} \) dependence in repeated experiments. In particular, the LimeSDR noise floor showed the theoretically predicted reduction by 13 dB when combining 20 averages.

In summary, the NMR benchmark experiment affirms that the LimeSDR provides the required functionality and performance for pulsed experiments. As also mentioned previously,\textsuperscript{7} the small fractional bandwidth required for NMR can be advantageous since the positioning of the relevant frequency range within the BB bandwidth can be optimized. In general, one wants to avoid that nonidealities of the frequency translation as well as spurious contributions from the ADC are overlapping with the relevant frequency range. For the electron spin systems studied in Sec. III B, the fractional bandwidth is no longer small so that different strategies are required to avoid such nonidealities.

B. Pulsed EPR at 150 MHz

Pulsed EPR was performed in order to outline the attainable time resolution with the LimeSDR. In fact, at a sampling rate of 30.72 MSa/s, the time resolution is 33 ns. In principle, even a faster sampling rate of 61.44 MSa/s would be supported. However, with the host computer used in this work, the data stream was not stable at this faster rate. Even at the chosen rate of 30.72 MSa/s, there were actually occasional interruptions of the stream. In order to avoid missing data points in pulse sequences, acquisition was resumed upon such interruptions.

For the chosen field strength of 5 mT, a 500 μM solution of the so-called “Finland trityl” radical\textsuperscript{44} dissolved in a 20 mM HEPES buffer at pH of 7.5 was used. This radical is known for its very narrow single-line spectrum as well as its sensitivity to local oxygen concentration.\textsuperscript{49,50} Accordingly, there are a number of applications of this radical, as for instance EPR-based oxygen sensing and imaging\textsuperscript{51–53} and NMR signal enhancement by polarization transfer from electron spins to nuclear spins.

First, it is demonstrated that a FID (and a Hahn echo) can be acquired with a sample at reduced oxygen concentration. For this purpose, nitrogen was gently bubbled into the sample tube for several minutes through a thin pipette. Just afterward, the sample tube was sealed. The 130 μl sample was then inserted into the spectrometer for the measurements. The resulting time traces are shown in Fig. 4, where the origin of the time axis is defined by the starting flank of the first pulse. For the FID upon the shortest excitation pulse (top), a few oscillations can be observed after a dead time of 1.2 μs.
For the Hahn echo (bottom), a signal free of dead-time artifacts was already observed 800 ns after the last pulse. The fact that the FID has decayed at times where the echo is nonzero ($t > 3.5 \mu s$) indicates the presence of inhomogeneous line broadening, which originates from hyperfine couplings to protons of the molecule’s CH$_3$ groups. Also note that with the signals emitted from the coil on the order of 100 nV, the traces were averaged $10^5$ times. For the FID, raw data were superimposed by ADC spurs, which have been removed, as described in Sec. III C.

To demonstrate the pulse sequencing capabilities of the spectrometer, the same sample tube was used. However, the oxygen concentration was augmented so that the relaxation times were faster as compared to Fig. 4. Under these conditions, the echo appeared as depicted by the black curve in Fig. 5(a) upon removal of ADC spurs (see Sec. III C). The gray curve corresponds to an exponential decay with time constant $T_2^* = 1.1 \mu s$. In the pulse sequences shown in the following, this exponential window function was used to weight the raw time-domain echo for optimum balance between signal and noise in the spectral domain. The echo area $I_{echo}$ was then obtained from the peak amplitude of the phased spectrum.

In order to verify proper excitation of the spins, Rabi oscillations have been driven at three different pulse amplitudes. The pulse sequence consisted of a driving pulse at an amplitude of 30%, 60%, or 90%, followed by a spin echo detection sequence that probed the longitudinal component upon the first pulse [see the inset in Fig. 5(b)]. From basic theory, one would expect that the flip angle $\phi_{flip}$ follows $\phi_{flip} = \omega_1 \cdot t_{nut}$, where $\omega_1$ is the driving pulse amplitude and $t_{nut}$ is its duration. Using the minimum time increment of 33 ns to prolong the duration $t_{nut}$ of the first pulse, transient nutation under the first pulse could be followed clearly. The resulting $I_{echo}(t_{nut})$ curves are shown in Fig. 5(b), with data for descending pulse amplitudes vertically arranged from top to bottom. The dashed gray lines indicate critical positions. For instance, at the first dashed line around $t_{nut} = 200$ ns, the weakest pulse realized a $\pi$ rotation, whereas the stronger pulses rotated the spins by $2\pi$ and $3\pi$. Note that the reduction in the oscillation amplitude is ascribed to inhomogeneity in the driving field and to relaxation effects since both $T_1$ and $T_2$ are on a comparable time scale. Corresponding rotation angles are found at the second dashed gray line around $t_{nut} = 400$ ns, namely, $2\pi$, $4\pi$, and $6\pi$. Accordingly, the flip angles are in good agreement with the pulse amplitudes, which confirms that spin excitation is working as intended.

Having verified proper spin excitation, basic pulse sequences to determine spin relaxation have been implemented. In Fig. 5(c), the decay of the Hahn echo as a function of the interpulse delay is shown in black. An exponential decay with a time constant $T_2 = 1.34 \mu s$ was obtained upon least-square curve fitting (gray). Note that the sequence script for this experiment is shown in Fig. 2. The decay of longitudinal magnetization was monitored with the inversion recovery sequence [inset in Fig. 5(d)]. The resultant inversion decay (black) in Fig. 5(d) was fitted in least-squares sense by an exponential decay (gray) with $T_1 = 1.54 \mu s$.

Overall, the experiments in this section confirm that the pulse sequencing capabilities demonstrated for NMR in Sec. III A as well as in an independent study can be extrapolated to EPR as long as the time scale of 33 ns is sufficient for the sample under study. Nevertheless, a critical aspect when working in this regime is the presence of spurious frequencies from the ADC, which needed to be removed in a postprocessing step for a number of time traces presented in this section. Owing to its importance, Sec. III C treats this aspect in further detail. Other than these ADC spurs, imbalances in the quadrature frequency translation scheme might also be relevant for both RX and TX paths. Currently, however, such imbalances are considered to be less problematic since these can be corrected for by...
fine-adjustments of the mixers, at least within a certain bandwidth and parameter range.

C. Cancellation of digitizer spurs

As mentioned in Sec. III B, spurious oscillations due to the ADC have been observed in data with exhaustive averaging. To further elaborate on these imperfections, a 240 µl trityl solution that was filled into the tube under ambient conditions was used. The raw echo trace obtained after 6.5 x 10^7 averages is shown in black in Fig. 6(a), together with the corresponding magnitude spectrum in panel b. While the time domain data appear rather noisy, the spectrum reveals that this noise corresponds to spurious oscillations at f_s/2 and f_s/4. Such spurious oscillations are rather common for fast ADCs, especially when several converters are interleaved for a faster sampling rate. Without knowing the detailed internal architecture of the ADC within the FPRF, it is a valid assumption that the ADC is constructed by interleaving four ADCs that run at a fourth of the sampling frequency and are time shifted with respect to each other. If the time shift between the four ADCs is exact and the analog-to-digital conversion is identical for each ADC, the interleaved ADCs are equivalent to one single ADC at four times the sampling rate. However, in the case of ADC mismatch and timing errors, interleaved ADCs are prone to imperfections and a number of dynamic correction techniques have been proposed.\textsuperscript{12}

For the raw echo trace in Fig. 6(a), a decomposition of the signal into four interleaved signals is shown in the inset. Each color corresponds to a different subtrace. As is readily seen, the subtraces have a different DC offset. On device level, these DC offsets might correspond to mismatch in either conversion gain or offset among the interleaved ADCs. By individual subtraction of the DC offset of each subtrace and recombination of the data, the traces illustrated in blue in panels a and b are obtained. As is clearly seen in both the time- and frequency-domain representation, the spurious oscillations were suppressed using this rather simple static correction. Likewise, the FID trace in Fig. 4 and the echo trace in Fig. 5(a) have been corrected using this technique.

Other than a correction during postprocessing, one certainly prefers to reduce these spurious oscillations at the first place. The most obvious strategy is to ensure that the analog signal is well scaled to the input range of the ADC. Here, the input noise was scaled to roughly 20% of the full ADC range. Accordingly, there was not much headroom left to further enhance the gain before the ADC. With the echo buried in the noise floor, this gain selection was actually already rather high and chosen due to the presence of ADC spurs. Note that such a high gain setting comes at an expense of dead time due to the reduced dynamic range. Another possibility that is incorporated into the LimeSDR FPRF would be the use of oversampling and decimation. One could, for instance, operate the ADC at a four times faster sampling rate and decimate to f_s. This was not done in this study since oversampling in the RX path would also imply oversampling in the TX path. As mentioned in Sec. II, interpolation and oversampling in the TX path flatten the pulse flanks and are prone to introduce uncertainty in the timing between the added TTL triggers and the TX DACs.

Given the above constraints, an alternative cancellation strategy dedicated to repeated acquisitions was elaborated. In particular, the spurious oscillations only add up in phase if the repetition time is an integer multiple of the oscillation period. For the data presented so far, this was always the case. By incorporation of intentional shifts to the repetition time, the spurious oscillations can be averaged out. The point of departure is the chosen repetition time that corresponds to M_rep samples. On the LimeSDR architecture (and certainly also on numerous other spectrometers with synchronization between RX and TX), M_rep is automatically bound to numbers that are a multiple of 4. Accordingly, f_s/2 and f_s/4 spurs are a multiple of the repetition time and will add up in phase. To counteract this addition, the data are averaged consecutively in four separate buffers, where the repetition time M_rep is incremented by one sample from buffer to buffer. The experiment is therefore effectively repeated with a consecutive repetition time cycle of [M_rep + 1, M_rep + 1, M_rep + 1, M_rep + 3]. In other words, the position of the echo transient within each buffer is incremented by one sample. When combining the data from the four buffers with the corresponding shift in the echo position, the f_s/2 and f_s/4 spurs average out.

Data acquired in this way are shown in orange in Fig. 6. Both time-domain and frequency-domain representations approve the absence of the spurs. Note that the apparent improvement of the signal-to-noise ratio of the orange trace as compared to the blue trace is solely due to the number of scans. In fact, the blue trace...
and black trace represent data from the first out of the four averaging buffers. As a technical remark, it is noted that there was actually no need to allocate new RX data buffers for this experiment as the minimum buffer size of the LimeSDR already accommodated 50 sequence repetitions at the chosen repetition time of 7.9 μs. For longer sequence repetition times, separate data buffers are obviously required. In general, it is advisable to cycle between the different repetition times as fast as possible, as opposed to averaging each buffer individually one after another. The reason is that the spurious contribution might drift while averaging each buffer individually. Ideally, one would thus combine the phase cycle and the repetition time cycle together and cycle both parameters at the nominal repetition rate. In the experiment here, only the repetition time was cycled at the nominal repetition rate. The phase was cycled at a 50 times slower rate of 2.5 kHz.

It is expected that this scheme can be helpful in a number of other situations. First of all, it lies at hand that also higher order spurious frequencies up to $f_{s/n}$ can be averaged by a corresponding repetition time cycle of length $n > 4$. Second, the presence of such spurs is certainly not constrained to the converters in the LimeSDR. As an example, such spurs have also been observed on a state-of-the-art EPR spectrometer with high-end data converters operating in the gigahertz range.

### IV. CONTINUOUS-WAVE ELECTRON SPIN RESONANCE

While pulse experiments capture transient precession dynamics, CW experiments record steady-state spin precession under RF excitation. The field modulation at frequency $\omega_{mod}$ required for separation of the precession signal creates a number of sidebands around the RF excitation frequency $\omega_{RF}$. In the limit where the field modulation is weak compared to the linewidth of the sample, only sidebands at the fundamental frequency are generated. The received signal can be written as

$$y_{CW}(t, B_0) = \alpha \cdot S(B_0) \cdot e^{i\omega_{RF}t + \phi_{RF}} \cdot \cos(\omega_{mod}t + \phi_{mod})$$

$$+ \beta \cdot e^{i\omega_{RF}t + \phi_{RF} + \phi_{lock}},$$

(1)

where $S(B_0)$ is the complex-valued derivative CW spectrum at $\omega_{RF}$ with absorption and dispersion components, $\phi_{RF}$ and $\phi_{mod}$ are the phases of the corresponding frequencies, and $\alpha$ and $\beta$ are the complex-valued parameters that relate to experiment, instrument, and sample conditions. The first line thus corresponds to the precession signal at the lower and upper sideband, whereas the second line denotes the excitation signal reflected from the cavity. Note that an additional phase $\phi_{lock}$ was added explicitly to the reflected signal, so as to include a leakage path from the transmitter to the receiver. For the low-cost CW spectrometer used here [Fig. 1(c)], such leakage took place within the directional coupler used to isolate reception from transmission. In particular, leakage through the coupler was on the order of 6 dB below the signal reflected from the cavity. As a consequence, the signal detected at $\omega_{RF}$ cannot be used to retrieve the phase $\phi_{RF}$ that is required to extract the spectrum $S(B_0)$. A different approach to obtain $\phi_{RF}$ was therefore required, and two different options are explained below. In order to avoid leakage at the first place, better isolation might be achieved with a circulator or even by leakage cancellation. With a circulator, also the transmitter power could be used more efficiently, at least up to the point where the receiver becomes saturated. In fact, receiver saturation is due to the reflected signal from the cavity and critically depends on the isolation between reception and transmission. This reflected signal might be reduced by active cancellation— or by an improved probe design.

Other than knowledge of the RF phase $\phi_{RF}$, also the modulation phase $\phi_{mod}$ needs to be known in order to properly extract the spectrum $S(B_0)$. Ultimately, this requires phase coherence between the phases $\phi_{RF}$ and $\phi_{mod}$ throughout the entire experiment, as clarified in Sec. IV A. As a technical side remark, it is noted that all CW experiments have been performed with a two-step phase cycle in the modulation phase $\phi_{mod}$. The corresponding intentional phase shifts of $\pi$ have been reduced during data analysis. When using the TTL outputs to generate the modulation signal at 105 kHz, such a phase cycle was specified within the experiment script. When using an external signal generator to generate this signal, the modulation frequency needed to be incremented by half of the inverse buffer period $t_{buff}$. Indeed, independent of whether a phase cycle was used or not, matching of the external modulation frequency and the buffer period required an appropriate choice of the buffer period $t_{buff}$. The choice needed to be such that the resulting $f_{mod}$ could be realized with the external signal generator’s frequency resolution. Since no evident advantage of this phase cycling scheme over the standard experiment without such cycling has been observed, it will not be commented further throughout this study.

#### A. Modulation using an external source

In order to illustrate the need for a phase-synchronous field modulation in CW EPR with a SDR, results obtained with an external benchtop signal generator are first presented. The sample used for this experiment was a commercially obtained powder of dilithium phthalocyanine (Alfa Aesar #39334: lithium phthalocyanine). In principle, this sample represents a precursor for the production of the widely used lithium phthalocyanine (LiPc) radical that contains only a single lithium atom and has a specific crystalline stacking. However, the compound used as received contained an intense EPR signal with X-band peak-to-peak line width $\Delta_B^{pp}$ of 0.86 G and no apparent sensitivity to oxygen. Accordingly, it was assumed that a significant fraction of the sample already oxidized to stable LiPc radicals during synthesis. It was further assumed that the crystalline properties inhibited efficient oxygen diffusion.

The spectrum as obtained by demodulation of the two modulation sidebands is shown in Fig. 7(a), showing the real (blue) and the imaginary (orange) component. The traces appear rather noisy since only $\phi_{RF}$ has been corrected, but not $\phi_{mod}$. Accordingly, these traces represent the spectrum $S(B_0)$ multiplied by $\cos(\phi_{mod})$. Because the modulation phase $\phi_{mod}$ was different for each point in the field-swept spectrum, the traces appear distorted. Based on magnitude spectra, $\phi_{mod}$ could be reconstructed for each point. The resulting spectrum $S(B_0)$ is shown in Fig. 7(b), and its components represent the dispersion (blue) and the absorption (orange) component. This spectrum was fitted in least-squares sense by a homogeneous line with $\Delta_B = 2.95$ G (black and gray). Note that the RF phase $\phi_{RF}$ was also retrieved based on the magnitude spectrum, namely, by enforcing symmetry of the absorption component.
While phase retrieval for $\phi_{\text{mod}}$ and $\phi_{\text{RF}}$ is an option at a high signal-to-noise ratio as it is the case here, it is certainly not a general approach. The phase should therefore be the same for each data point. Here, the phase was different since for each point in the fieldsweep, the SDR was stopped in-between field adjustments. In principle, one could keep the SDR running during field adjustments and thus establish that field adjustment will be executed within a time that is an exact multiple of the buffer period $t_{\text{buffer}}$. In this way, the modulation phase $\phi_{\text{mod}}$ would be coherent throughout the experiment. However, it would still vary from spectrum to spectrum since the phase of the external source is random at the time the experiment starts. Accordingly, a phase synchronization between the SDR and the modulation drive is required, which can be accomplished using a triggering scheme.

Similar to pulsed operation, there is the option to synchronize the SDR with an external trigger, as demonstrated previously, or to use an internal trigger from the SDR, as pursued in this study. Benchtop signal generators do, however, rarely accept a trigger to reset the phase. Accordingly, a solution based on an integrated oscillator chip module with a synchronous phase reset functionality could be an option. As an example, such functionality is provided by a few analog and digital synthesizers as well as by clock dividers. The phase of the external oscillator module could thus be reset by a TTL trigger from the SDR. Yet another option is to directly generate a digital oscillator via the FPGA GPIO output and translate this digital oscillator to a sine wave for field modulation, as implemented in this study and presented in the following.

**B. Phase-synchronous modulation using the FPGA GPIO**

As discussed in Sec. IV A, the phase synchronization problem has been solved by deriving the field modulation from the FPGA GPIO output. The modulation phase $\phi_{\text{mod}}$ was therefore constant and determined once by calibration. An additional issue was leakage through the directional coupler, which inhibited a straightforward determination of the RF phase $\phi_{\text{RF}}$ from the reflected signal. As a workaround, the RF phase $\phi_{\text{RF}}$ has been extracted by means of two auxiliary RF sidebands around the carrier. To this end, the CW excitation tone was extended by two amplitude-modulation sidebands at 80 kHz offset and with amplitude at 0.5% of full scale. Due to the frequency offset to the probe, these sidebands experienced a more pronounced reflection from the probe such that the unwanted phase shift from leakage through the coupler was negligible. Since the phase response of the matched coil is (i) linear around its center and (ii) symmetric with respect to its center, the RF phase was determined by the mean phase of the two sidebands. An additional phase shift of $\pi/2$ to $\phi_{\text{RF}}$ is obtained in this way for the absorption component to the imaginary part of the spectrum $S(B_0)$. While calculating the mean phase of the two sidebands, one should account for phase wrapping. Here, with $\phi_1$ being the phase of the lower sideband and $\phi_2$ being the phase of the upper sideband, $\phi_2$ has been augmented by $2\pi$ if $\phi_2 < \phi_1$. Without such a correction, the phase $\phi_{\text{RF}}$ would experience an artificial phase shift of $\pi$ in the case of phase wrapping. Indeed, the amplitude-modulation sidebands for determination of $\phi_{\text{RF}}$ are part of the AFC functionality demonstrated in Sec. IV C. Frequency correction is, however, not a necessity for the extraction of the RF phase based on the excitation sidebands.

In Fig. 8(a), the resulting absorption (orange) and dispersion (blue) components of the LiPc sample are illustrated. In analogy to Sec. IV A, the shape of the components was fitted in least-squares sense using a homogeneous line with a comparable $\Delta g$ of 2.93 G (black and gray). This approves the calibration of the modulation phase $\phi_{\text{mod}}$ and the reconstruction of $\phi_{\text{RF}}$ from the excitation sidebands. However, there is an appreciable DC offset in both curves, which is particularly pronounced for the absorption component. Notably, this offset was also present without a sample inserted, but it vanished almost completely when disconnecting the modulation coil. Such a DC offset could originate from vibrational coupling between the modulation coil and the EPR probe. However, the employed probe did not show such an offset with an external signal generator [see Fig. 7(b)]. Accordingly, this offset is attributed to an electrical decoupling of the drive circuit, such as residual modulation of the RF signal by the modulation drive circuit. It is therefore anticipated that further electrical decoupling of the drive circuit can reduce this DC offset.
Thanks to the synchronous phase, also CW spectra of trityl solutions were acquired. To this end, the same sample tube as used for Figs. 4 and 5 was used but at different oxygen concentrations. The absorption spectrum of the sample tube prepared under ambient conditions is shown in blue. After bubbling nitrogen for a few minutes and subsequent sealing, the spectrum shown in orange was obtained. Both curves were fitted in least-square sense by homogeneous lines (dashed) with $\Delta_{pp}$ of 197 mG and 93 mG, respectively. Note that both traces are normalized to the same $y$ scale. The apparent difference in the DC offset of the two spectra is due to the different modulation amplitudes used for the two samples. In contrast, modulation of the degassed sample with 75 mVpp modulation, as used for the native sample, resulted in an overmodulated spectrum (data not shown). Furthermore, based on the estimated $B_1$ driving field of 77 mG (see Sec. II A 3), it is assumed that the 93 mG wide line was broadened considerably by the drive (see also Sec. IV C).

Overall, the results in this section approve that the complex-valued spectrum $S(B_0)$ is accessible using the LimeSDR with upgraded GPIO functionality. Apart from this encouraging proof-of-principle demonstration, it should be noted that the proposed setup can benefit from a number of improvements. First, the CW setup has been designed for reduced cost, which brought certain performance losses discussed above, namely, isolation and leakage. Unless for teaching purposes, one certainly wants to improve the performance by implementing established approaches, such as probes with higher isolation. As mentioned above, the DC offset due to coupling between $\omega_{mod}$ and $\omega_{RF}$ is another important aspect related to the hardware design.

Other than the hardware design, there is one important observation that is related to the operation of the LimeSDR itself. In particular, certain points within a CW field sweep were corrupted. A clear spike around $B_0 = 45$ G in Fig. 8(a) is visible in both curves. In addition, a pronounced spike is visible in the blue curve in Fig. 8(b) around 53.1 G. The origin of these spikes has not been investigated further, and it is presumed that these are related to problems with data streaming and buffering with the specific combination of LimeSDR firmware, host computer, and API. In principle, one could detect at least some of these spikes during a scan and reacquire the corrupted data points. As an example, the spike in Fig. 8(b) can be identified as fluctuation in all demodulated signals.

C. Automatic frequency control

Automatic frequency control (AFC) is an important technique in CW EPR. The majority of AFC schemes rely on a voltage-controlled microwave oscillator, whose frequency is locked to the cavity by means of a feedback onto the oscillator’s control voltage. For digital CW spectrometers, such an analog feedback loop is not possible and a digital feedback is required. Accordingly, a way to adjust the frequency of the CW oscillation is required. Particularly, such a feedback loop has been implemented directly within the FPGA of a fully digital spectrometer. In this way, a very fast digital feedback is achieved, as is, for instance, required for in vivo imaging.

In this work, a software-defined digital AFC was implemented. The feedback thus needed to pass via the host computer, which required more time to adjust the frequency. As described in Sec. II B 1, the feedback mechanism was based on excitation sidebands and it took 435 ms to adjust the output frequency. A special feature of the software implementation is the support for more than one single sideband modulation frequency, which allows for more precise frequency estimation protocols than with a single modulation frequency. Here, the frequency shift was the mean frequency shift determined independently from two modulation frequencies at 80 kHz and 82.5 kHz. When operating the feedback loop at the fastest possible rate, an enhanced noise floor was observed, indicating that a longer observation window was required in order to estimate the frequency with adequate precision. Accordingly, the AFC time window was set such that there was one frequency correction per data point in a field sweep.

An example with the same sample as used in Fig. 5 is shown in Fig. 9. Panel a shows the absorption (orange) and dispersion (blue) component averaged for 9 out of 10 repeated scans and a fit in least-square sense (dashed). Panel b shows the shift of the CW frequency $\Delta f_{CW}$ during the acquisition of these 10 scans, which corresponds to a time interval of 4.25 h. As can be inferred from the frequency shift, the AFC followed drift in the probe frequency. The slow time constant of this drift justifies the long feedback time for the AFC of 23 s. In the spectrum, the noise floor is visibly enhanced for $B_0 > 54.4$ G. This was due to corrupted data points in the 9 averaged spectra, as observed in Sec. IV B. One single spectrum had actually spikes that were so large that the trace was excluded from averaging. Other than these spikes, the very first data point is offset since each scan was started with the same CW frequency. In panel b, this starting frequency corresponds to $\Delta f_{CW} = 20$ kHz. Accordingly, this first point indicates how the field-swept spectrum is influenced by a frequency jump on the order of 30 kHz.

The fit to the spectrum reveals a line width $\Delta_{pp} = 103$ mG, which corresponds to a transverse relaxation time of 638 ms. This relaxation time is by a factor of 1.7 faster than the corresponding $T_1^*$ relaxation time determined with pulsed EPR [see Fig. 5(a)]. The reason for the larger line width for CW is the amplitude of the driving field $B_1$. In fact, by assuming the estimated $B_1$ field of 77 mG of the coil alone, line broadening by a factor of 2.0 is predicted. With a slightly lower field around 61 mG, the same factor of 1.7 is obtained. Within the precision of the estimated $B_1$ field, the CW EPR results are therefore in agreement with the pulse EPR data from Fig. 5 using exactly the same sample.
V. CONCLUSIONS AND OUTLOOK

Overall, the experimental results approve basic pulsed and CW operation of the SDR-based low-cost spectrometer. An important feature is that only a few modifications are required in order to upgrade the off-the-shelf LimeSDR board for spectrometer development. It is therefore anticipated that the main electronic interface of the upgraded SDR, namely, the single-channel RF transceiver and the four-channel bit pattern generator, is readily portable to other environments. With the RF frequency range of the transceiver reaching up to 3.5 GHz, a number of scenarios that benefit from such a hardware setup are envisaged.

For pulsed NMR, the SDR-based solution can probe $^1$H resonances at magnetic fields of 0.7 T and higher. In situations where a suitable magnet is available, basic NMR functionality can thus be added with a rather low effort. This is, for instance, the case for the development of alternative NMR detection methods, for dynamic nuclear polarization (DNP) experiments, for NMR magnetometry purposes, or to replace outdated spectrometer consoles that are no longer supported despite a functional magnet. For the case that a RF power amplifier for pulsed excitation is not available, even CW approaches that have here been demonstrated for EPR could be envisaged. In this respect, it should be noted that a configurable low-cost solution that combines CW and pulsed approaches might prove useful for educational purposes. For magnetic fields below 1 T, which can be realized with compact permanent magnets, 0.2 T appears as the acceptable lower limit for the RF frequency range of the LimeSDR. A certain operation window for combination with such compact and portable magnet systems is therefore also provided.

For EPR spectroscopy, the LimeSDR’s frequency range is rather well suited, given that frequencies beyond the transceiver’s cutoff at 3.5 GHz could be reached by frequency translation. However, a limitation for pulsed EPR is the time resolution, which was at 33 ns in this study. With current pulsed EPR spectrometers operating on nanosecond time scales and the trend toward sub-nanoseconds, a versatile pulsed EPR spectrometer is out of reach. Nevertheless, there is a multitude of experiments that can be performed at this resolution, as exemplified by the number of EPR spectrometers with operation frequency below 8 GHz with comparable time resolution. With an effort in reconfiguring the dual-channel digital interface for maximum single-channel bandwidth, an improved time resolution of 8 ns should actually be feasible. For ordinary CW experiments, the time resolution is largely sufficient. By further elaborating on the electronics for phase-coherent field modulation, even spectroscopy in the rapid scan regime is within reach.

A potential limitation of this entire approach lies within the correction of open issues and performance limitations of the LimeSDR board. The most notable issue in this study was occasional interruptions of the data stream. These are, however, expected to be less prominent on a different host computer. Another independent study with the LimeSDR pointed at intrinsic difficulties in synchronizing multiple boards. On the one hand, the open-source hardware concept already proved useful in correcting for certain limitations, such as the absence of trigger functionalities. On the other hand, it still remains an open question to what extent adaptations to the firmware are maintainable besides official firmware releases, which are updated quite regularly.

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