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A single chip electron spin resonance detector based on a single high electron mobility transistor

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ABSTRACT

Single-chip microwave oscillators are promising devices for inductive electron spin resonance spectroscopy (ESR) experiments on nanoliter and subnanoliter samples. Two major problems of the previously reported designs were the large minimum microwave magnetic field (0.1–0.7 mT) and large power consumption (0.5–200 mW), severely limiting their use for the investigation of samples having long relaxation times and for operation at low temperatures. Here we report on the design and characterization of a single-chip ESR detector operating with a microwave magnetic field and a power consumption orders of magnitude lower compared with previous designs. These significant improvements are mainly due to the use of a high electron mobility transistor (HEMT) technology instead of a complementary metal-oxide semiconductor (CMOS) technology. The realized single-chip ESR detector, which operates at 11.2 GHz, consists of an LC Colpitts oscillator realized with a single high-electron mobility transistor and a co-integrated single turn planar coil having a diameter of 440 μm. The realized detector operates from 300 K down to 1.4 K, at least. Its minimum microwave magnetic field is 0.4 μT at 300 K and 0.06 μT at 1.4 K, whereas its power consumption is 90 μW at 300 K and 4 μW at 1.4 K, respectively. The experimental spin sensitivity on a sensitive volume of about 30 nL, as measured with a single crystal of α,γ-bisdiphenylene-β-phenylallyl (BDPA)/benzene complex, is of 8×10^{10} spins/Hz$^{1/2}$ at 300 K and 2×10^{9} spins/Hz$^{1/2}$ at 10 K, respectively. In a volume of about 100 pL, located in proximity to the coil wire, the spin sensitivity improves by two orders of magnitude.

1. Introduction

Methods based on the electron spin resonance (ESR) phenomenon are powerful spectroscopic tools, used in physics, chemistry, material science, biology and medicine, to investigate the structure, dynamics, and spatial distribution of paramagnetic species in a wide range of frequencies (typically from 100 MHz to 1 THz) and temperatures (typically from 10 mK to 1000 K) [1–3]. At frequencies above 1 GHz, the measurements are usually performed using either relatively large cavities (mL to μL sensitive volumes) or miniaturized conducting [4–18] and superconducting [19–27] resonators (μL to sub-pL sensitive volumes). Miniaturized resonators are used in order to maximize the signal-to-noise ratio in experiments with small samples. Inductive methods based on microresonators have sensitivities at the level of 10^7–10^{11} spins/Hz$^{1/2}$ at room temperature [8,10,11,13,17] and of 10^2–10^6 spins/Hz$^{1/2}$ at low temperature [10,16,21,22,25–27], mainly depending on the sample relaxation times, the sensitive volume of the microresonator, the operating frequency, and the adopted normalization criteria (see Appendix C). Non-inductive techniques can achieve single electron spin sensitivity [28–30] but are generally considered as less versatile than those based on the more conventional inductive approach [16,22,31].

In Refs. [8–12] we presented single-chip integrated inductive ESR detectors, fabricated using complementary metal oxide semiconductor (CMOS) technologies, operating between 8 GHz and 146 GHz in the temperature range from 300 K down to 4 K. The ESR phenomenon was detected as a variation of the frequency of an integrated LC-oscillator due to an effective variation of its coil impedance caused by the resonant complex susceptibility of the sample (see Appendix A). Two major problems of the previously reported designs were the large minimum microwave magnetic field (0.1–0.7 mT) and the large power consumption (0.5–200 mW), severely limiting their use for the investigation of samples having long relaxation times and for operation at low temperatures. Here we report on the design and characterization of single-chip ESR detectors operating with microwave magnetic fields down to 0.4 μT (at 300 K) and 0.06 μT (at 1.4 K), and power consumptions down to 90 μW (at 300 K) and 4 μW (at 1.4 K), i.e., orders of

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magnitude lower compared with previous designs [8–12]. These significant improvements are mainly due to (1) the use of an InGaAs high electron mobility transistor (HEMT) technology instead of a Si CMOS technology [32], and (2) the drastic simplification of the integrated electronics to a single transistor with a few passive components instead of a more complex circuitry. Reducing the microwave field amplitude and power consumption allows to extend the use of the single-chip ESR detector approach to samples having much longer relaxation times (up to T_1T_2 products as large as 10^{-10} s^2 at 300 K and 10^{-8} s^2 below 30 K) and to lower temperatures (down to 1.4 K with ordinary ^{4}He variable temperature inserts and, probably, also at lower temperatures with high cooling power ^{3}He probes and ^{3}He-^{4}He dilution refrigerators).

2. Description of the single-chip ESR detector

The single-chip ESR detector described in this work is manufactured using a high electron mobility transistor (HEMT) technology (D007IH mHEMT, OMMIC, France). The transistor channel consists of a two dimensional electron gas (2DEG) in $In_{0.7}Ga_{0.3}As$. The gate length is 70 nm. The key properties of transistors and low noise amplifiers operating up to 220 GHz realized with the technology used in this work have been investigated in Refs. [33,34], in the temperature range from 300 K down to 20 K. In this work we report on the design and characterization of a single-chip integrated ESR detector based on an 11 GHz oscillator operating in the temperature range from 300 K down to 1.4 K, at least.

Fig. 1a shows the detailed schematics of the single-chip ESR detector. The integrated detector is a Colpitts LC oscillator [35–37], consisting of a single HEMT transistor, a single turn planar coil inductor, a resistor, and two capacitors. The resonator is connected to a common drain amplifier in order to achieve low output impedance without loading the LC resonator. The DC bias of the transistor is obtained connecting the source to ground through an integrated 500 Ω resistor (acting as current generator) and connecting the gate to ground (the coil inductor has a DC impedance of about 0.6 Ω and no DC current flows through it). The positive feedback from the transistor source through the two resonator capacitors creates an equivalent negative resistance which compensates for the losses in the resonator, and then leads to stable oscillations. In the HEMT technology used in this work, the availability of on-chip connections to ground using metalized holes from the front-side to the back-side of the chip (commonly called vias) allows to avoid the use of wire-bonding for the connection to ground, which makes it feasible to integrate simple non-differential topologies with relatively low parasitic capacitances, inductances, and resistances. The oscillator frequency varies from 11.2 GHz to 11.4 GHz, depending on the temperature and the DC bias voltage V_{DD}. The overall size of the integrated Colpitts LC-oscillator is 0.8×0.5 mm^2 (see Fig. 1c). The chip is glued with an conductive epoxy (Epo-Tek, H20E-FC) on a standard FR4 printed circuit board and electrically connected by wedge-wedge Au wire bonding (wire diameter 20 μm).

In Fig. 1b we show the block diagram of the complete set-up used to perform the ESR experiments reported in this work. Since the integrated common drain amplifier is not saturated, it is possible to measure both the oscillator amplitude and the oscillator frequency at the same time. For this purpose, the output signal from the integrated oscillator is amplified and, subsequently, processed by two separated electronic chains. The frequency detection branch consists of a frequency divider, a mixer, and a delay line discriminator used as a frequency-to-voltage converter. The amplitude detection branch consists of a microwave diode detector. Field modulation with lock-in synchronous demodulation is used, as in conventional continuous-wave ESR spectroscopy, for both the oscillator frequency and the oscillator amplitude detection.

In Fig. 1d we report the key features of the realized single-chip ESR detectors, including the geometrical and the electrical details of the integrated coil, the minimum power consumption (and the corresponding minimum microwave magnetic field B_1) required for stable oscillation, and the spin sensitivity. B_1 and N_{min} values are given for samples placed in the geometric center of the coil as well as in close proximity to the internal side of the coil wire. The value of B_1 is estimated from the value of the microwave current in the coil obtained from electronic circuit simulations performed using ADS (Advanced Design System, Keysight). Taking into consideration that the simulations accurately predict the oscillation frequency, the oscillation amplitude, and the DC power consumption (both current and voltage), we estimate that the computed value of B_1 is accurate within a factor of two. The spin sensitivity (given in spins/Hz$^{1/2}$) is computed from the experimental results obtained with a sample of BDPA as $N_{min} = (3N_S/SNR)$, where N_S is the number of spins in the sample and SNR is the signal-to-noise ratio. The signal expressed as its peak-to-peak value is given in Hz and the noise is expressed as its spectral density in Hz/Hz$^{1/2}$ (see Appendix C for more details).

In Fig. 1e we report the experimentally measured frequency noise at 300 K, 100 K, and 1.4 K. The measured noise floor (i.e., the noise at frequencies above the 1/f noise corner frequency) corresponds, within a factor of two, to the thermal frequency noise originating from the coil resistance given by $(kTR)^{1/2}(f_{LC}/V_{LC,0})$, where $V_{LC,0}$ is the oscillation amplitude and R is the coil resistance at the operating frequency [38] (see Fig. 1a). The experimentally measured thermal noise and 1/f noise have an opposite dependence on the oscillator amplitude, in agreement with the model reported in Ref. [39]. The thermal noise is minimized by maximizing the oscillation amplitude whereas the lowest 1/f noise is obtained close to the minimum oscillation amplitude (i.e., close to the start-up condition for the oscillator). Since at the largest field modulation frequency possible with our set-up (about 150 kHz) the 1/f noise is still dominant, the noise is minimized at the minimum bias voltage. A detailed discussion on the achieved spin sensitivity is reported in Section 3 and in Appendix C.

3. Electron spin resonance experiments

Fig. 2 shows the results of measurements performed with the single chip ESR detector at temperatures from 300 K down to 1.4 K. We performed experiments on three samples having different characteristics, an exchange narrowed standard system (a single crystal of BDPA) and two hyperfine split systems (a single crystal of 5% Cu^{2+} in $Ni(mnt)_2$ and a powder of 0.2% Mn^{2+} in MgO). Details of each sample are given below. All samples are placed on the single-turn planar coil using a toothpick sharpened with a metallic cutter. The samples adhere to the chip surface by van der Walls forces without any adhesive or with the help of a small drop (<100 pL) of high vacuum grease (Dow Corning). The dimensions of the samples are measured by means of an optical microscope, with an error in the estimation of the volume of less than a factor of two. The reported ESR signals are obtained by measuring the oscillation frequency variation in presence of field modulation. Hence, the signal shape is the first derivative of a dispersion signal (see Appendix A).

In Fig. 2a we report the results obtained with a single crystal of BDPA (1:1 α,γ-bisdiphenylene-β-phenylallyl/benzene complex, Sigma Aldrich 152560) having a volume of about $30 \times 30 \times 5$ μm^3. Thanks to the low B_1 value (0.4 μT at 300 K and 0.06 μT at temperatures below 30 K), the signal is neither saturated nor broadened (the linewidth is 0.1 mT). The signal amplitude grows approximately as $1/T$ down to 30 K (i.e., it follows the Curie law). Below 10 K the signal amplitude decreases, probably due to a decrease in the susceptibility caused by the proximity with the