Real-Time Multi Harmonic Detection for EPR Spectroscopy using FPGA-Based Signal Processing

Masters thesis

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Electron Paramagnetic Resonance (EPR) Spectroscopy

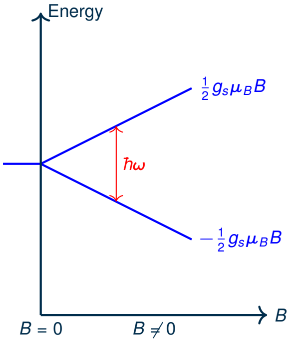
- EPR probes magnetic moments of unpaired electrons
- Example applications:
  - Medical imaging
  - Catalysis research
- Energy splitting of unpaired electrons due to Zeeman effect

$$E_{\pm} = \pm\frac{1}{2}g_s\mu_B B \tag{1}$$

- Transitions between the two states may happen for

$$\omega = \frac{g_s\mu_B B}{\hbar} \tag{2}$$

- Measure absorption
  - either vary frequency for a fixed magnetic field
  - or magnetic field for a fixed frequency



Swartz 2004; Takakusagi 2023; Attar 2024; Bonke 2021

Continuous wave EPR

- Magnetic field is modulated

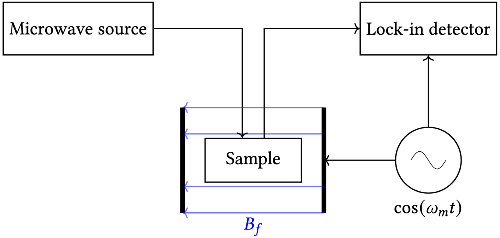
$$B_f = B + B_{\text{mod}} \cos(\omega_m t) \tag{3}$$

- Taylor expansion of the absorption $s(B_f)$

$$s(B_f) \approx s(B) + \frac{\mathrm{d}s(B)}{\mathrm{d}B} \frac{B_{\text{mod}}}{2} \cos(\omega_m t) + \dots \tag{4}$$

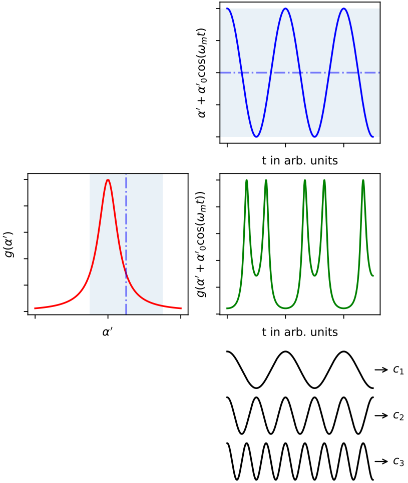
- Extract $\frac{\mathrm{d}s(B)}{\mathrm{d}B}$ with a Lock-in detector (uses $\cos(\omega_m t)$)
- Multi Harmonic Detection uses multiple higher harmonics ($\cos(\{1, 2, 3, \dots\} \times \omega_m t)$)

Multi Harmonic Detection can improve quality of spectra (SNR, broadening)



Tseitlin 2011; Yu 2015

Multi Harmonic Detection



- Determine the Fourier coefficients $c_n(\alpha')$

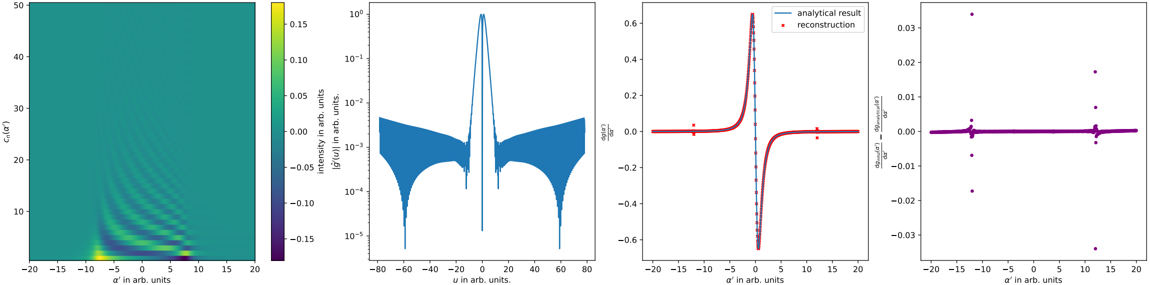
$$c_n(\alpha') = \frac{1}{T}\int_0^T \mathrm{d}t\, g(\alpha' + \alpha'_0\cos(\omega_m t))e^{-in\omega_m t} \tag{5}$$

- Recover first derivative from Fourier coefficients

$$\frac{\mathrm{d}g(\alpha')}{\mathrm{d}\alpha'} \approx \mathcal{F}_u^{-1}\left[\frac{\sum_{n=1}^{N_H} D_n^*(u)\mathcal{F}_{\alpha'}\left[c_n(\alpha')\right](u)}{\sum_{n=1}^{N_H}\left|D_n(u)\right|^2}\right](\alpha') \tag{6}$$

Tseitlin 2011

Simulated spectrum for a Lorentzian lineshape



$$c_n(\alpha')$$

$$\frac{\sum_{n=1}^{N_H} D_n^*(u)\mathcal{F}_{\alpha'}\left[c_n(\alpha')\right](u)}{\sum_{n=1}^{N_H}\left|D_n(u)\right|^2}$$

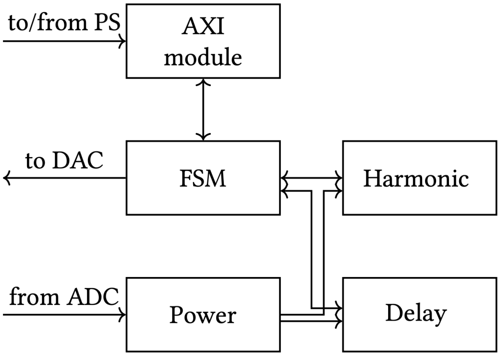
$$\frac{dg(\alpha')}{d\alpha'} \approx \mathcal{F}_u^{-1}\left[\frac{\sum_{n=1}^{N_H} D_n^*(u)\mathcal{F}_{\alpha'}\left[c_n(\alpha')\right](u)}{\sum_{n=1}^{N_H}\left|D_n(u)\right|^2}\right](\alpha')$$

Using the RFSoC for Multi Harmonic Detection

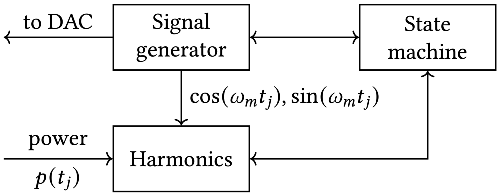
- RFSoC evaluation board
  - generates cosine FM signal
  - calculates the Fourier coefficients in parallel
  - measures delay of system
- Measurement control, Fourier coefficient processing and visualization on PC via python

FPGA implementation overview

- Complete



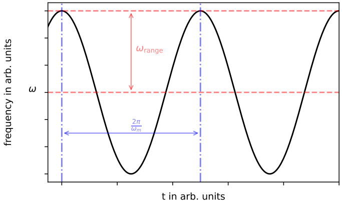
- Harmonic module

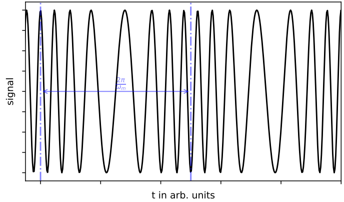


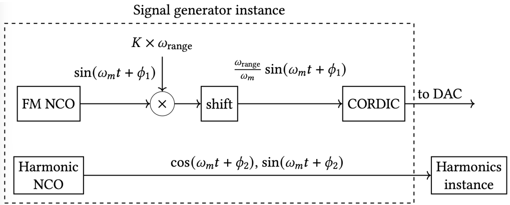
Signal generation

$$\omega_s(t) = \omega + \omega_{\text{range}} \cos(\omega_m t) \quad (7)$$

$$\cos\left(\int \mathrm{d}t\, \omega_s\right) = \cos\left(\omega t + \frac{\omega_{\text{range}}}{\omega_m} \sin(\omega_m t)\right) \quad (8)$$







Fourier coefficient calculation

- Fourier coefficient can be rewritten as

$$c_n(\alpha') = \frac{1}{T}\int_0^T \mathrm{d}t\, p(\omega + \omega_{\text{range}}\cos(\omega_m t_j - \phi_{\text{delay}}))e^{-in\omega_m t} \tag{9}$$

$$c_n = \begin{cases} \frac{a_0}{2} & \text{if } n = 0 \\ \frac{1}{2}(a_n - ib_n) & \text{if } n > 0 \\ \frac{1}{2}(a_{-n} + ib_{-n}) & \text{if } n < 0 \end{cases} \tag{10}$$

- a_n and b_n can be approximated with

$$a_n(\omega) = C\sum_{j=0}^{j_{\max}} p(\omega + \omega_{\text{range}}\cos(\omega_m t_j - \phi_{\text{delay}}))\cos(n(\omega_m t_j + \phi_{\text{adjustable}})) \tag{11a}$$

$$b_n(\omega) = C\sum_{j=0}^{j_{\max}} p(\omega + \omega_{\text{range}}\cos(\omega_m t_j - \phi_{\text{delay}}))\sin(n(\omega_m t_j + \phi_{\text{adjustable}})) \tag{11b}$$

If $\phi_{\text{delay}} = -\phi_{\text{adjustable}}$ then $b_n = 0$ due to symmetry

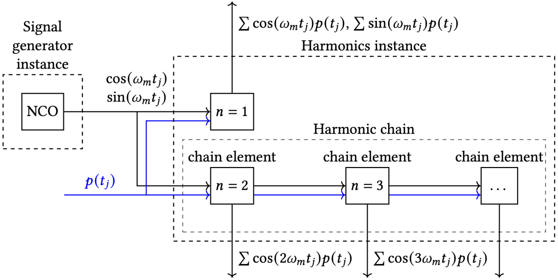
Fourier coefficient calculation

- Recursive approach, generate $\cos(\omega_m t)$, calculate all higher harmonics
- Chebyshev polynomials of first kind

$$T_n(\cos(\theta)) = \cos(n\theta) \tag{12a}$$

$$T_{n+1}(x) = 2xT_n(x) - T_{n-1}(x) \tag{12b}$$

- 12 Multipliers per coefficient, up to 356 coefficients possible
- Errors accumulate, however for $\theta \in [0, 2\pi]$ RMSE $< 6 \times 10^{-5}$ for $n = 1000$



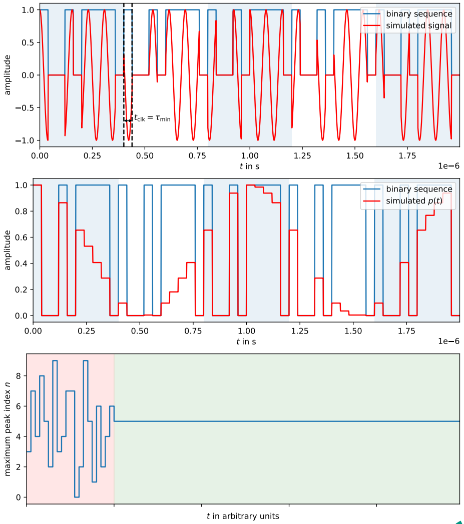
Delay measurement module

- Modulate output with semi random binary sequence
- Correlate response with time shifted sequences

$$c_\tau(t_j) = \sum_{v=0}^{j} p(t_v - \tau_{\text{delay}}) b(t_v - \tau) \qquad (13)$$

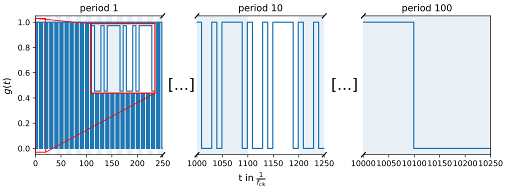
$$\tau = k\tau_{\min} \qquad (14)$$

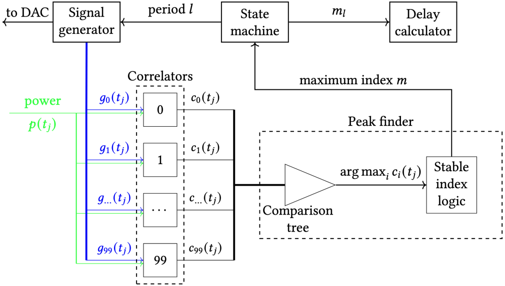
- Identify maximum sum $\arg\max_\tau c_\tau(t_j)$ by time evolution
- Sequence of length 100 selected



Delay measurement module

- To improve range use multiple periods and combine results





Phase sweep delay measurement

$$a_n(\omega) = C\sum_{j=0}^{j_{\max}} p(\omega + \omega_{\text{range}}\cos(\omega_m t_j - \phi_{\text{delay}}))\cos(n(\omega_m t_j + \phi_{\text{adjustable}})) \tag{15}$$

- First fourier coefficients a_1 and b_1 depend on phase difference

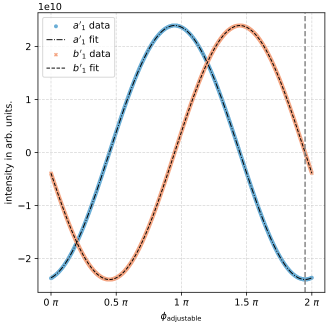
$$a_1' \propto \cos(\phi_{\text{adjustable}} + \phi_{\text{delay}}) \tag{16a}$$

$$b_1' \propto \sin(\phi_{\text{adjustable}} + \phi_{\text{delay}}) \tag{16b}$$

- Sweep $\phi_{\text{adjustable}}$ and recover ϕ_{delay} from cosine/sine fit

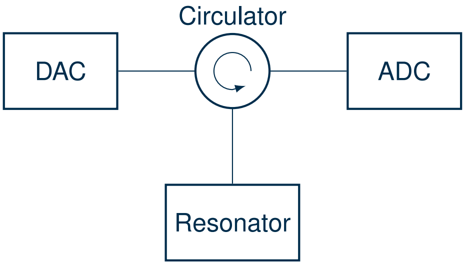
$$\tau_{\text{delay}} = \frac{\phi_{\text{delay}}}{\omega_m} \tag{17}$$

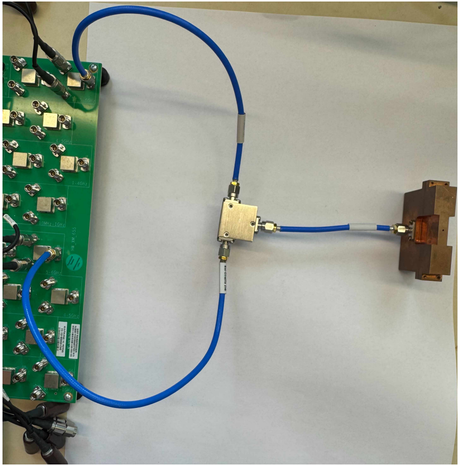
- Only requires adjustable phase of first harmonic generation



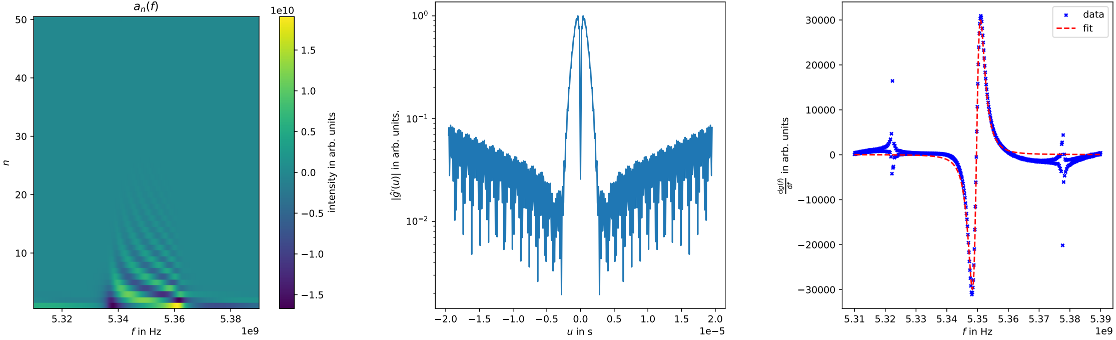
Resonator measurement setup

- 5.35 GHz copper cavity resonator

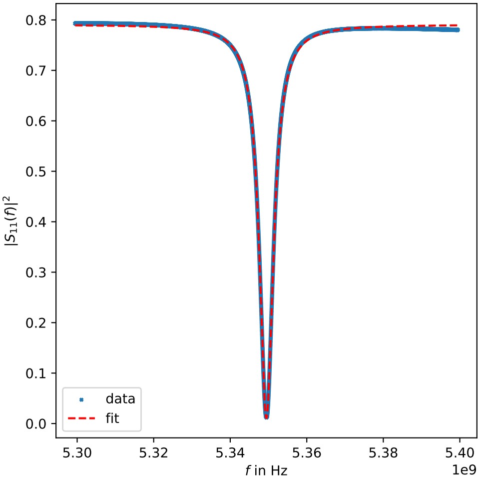




Resonator measurement



Comparison to VNA measurement



- Multi harmonic result

$$f_0 = 5.349\,637(15)\,\text{GHz} \tag{18a}$$
$$\gamma = 2.276(26)\,\text{MHz} \tag{18b}$$

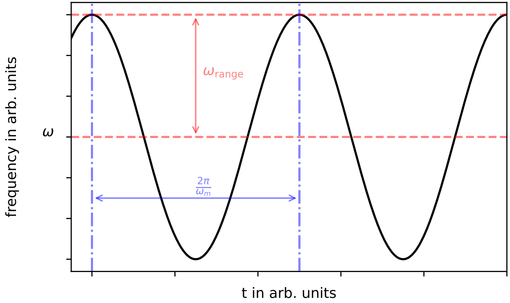
- VNA result

$$f_0 = 5.349\,444(8)\,\text{GHz} \tag{19a}$$
$$\gamma = 2.182(1)\,\text{MHz} \tag{19b}$$

- Slight broadening also observed in other works

Tseitlin 2011; Yu 2015

Stimulation parameter influence

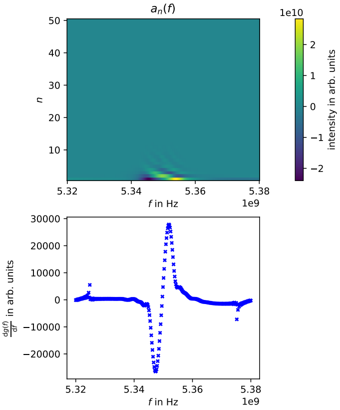


- for previous measurement

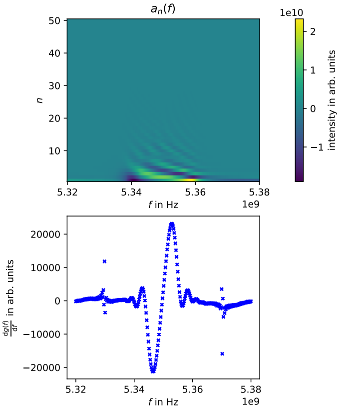
$$f_m = 50\,\mathrm{kHz}$$

$$f_{\mathrm{range}} = 12.5\,\mathrm{MHz}$$

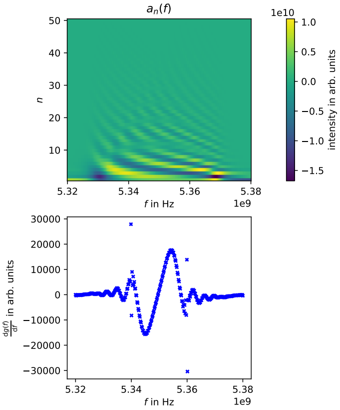
Constant $f_m = 500\,\text{kHz}$



$f_{range} = 5\,\text{MHz}$

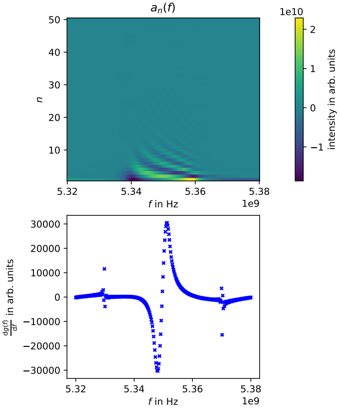


$f_{range} = 10\,\text{MHz}$

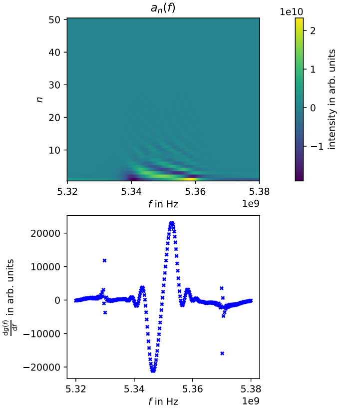


$f_{range} = 20\,\text{MHz}$

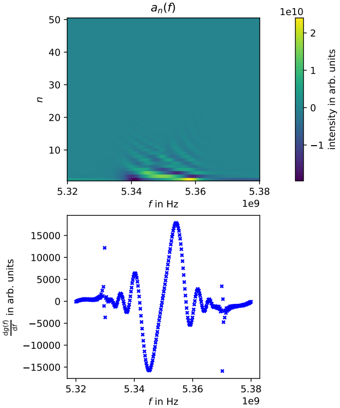
Constant f_{range} = 10 MHz



f_m = 100 kHz

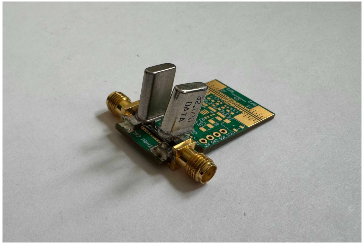


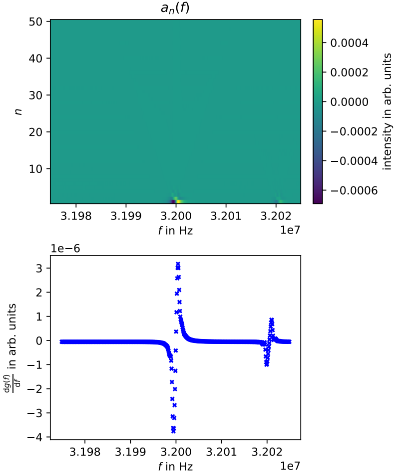
f_m = 500 kHz



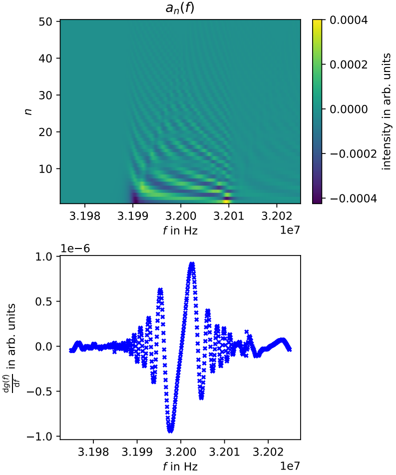
f_m = 1 MHz

Similar behaviour for 32 MHz quartz resonator



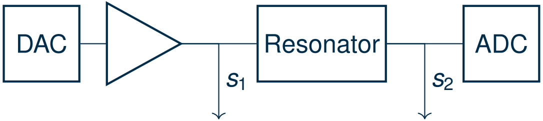


f_m = 0.5 kHz



f_m = 10 kHz

Quartz resonator transmission



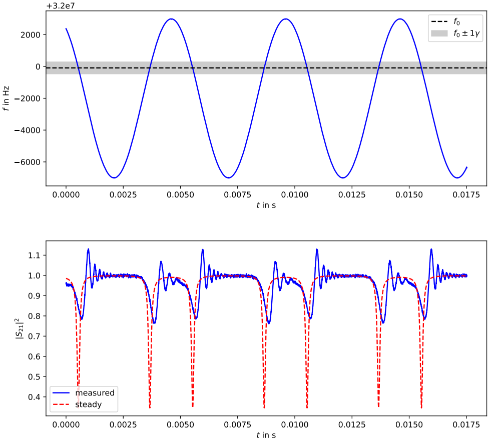
- from voltages of signals $s_{1,2}$ calculate

$$|S_{21}^{measured}(t)| = \left|\frac{V_2(t)}{V_1(t)}\right| \tag{20}$$

- compare to steady state S_{21} from VNA measurement using measured frequency

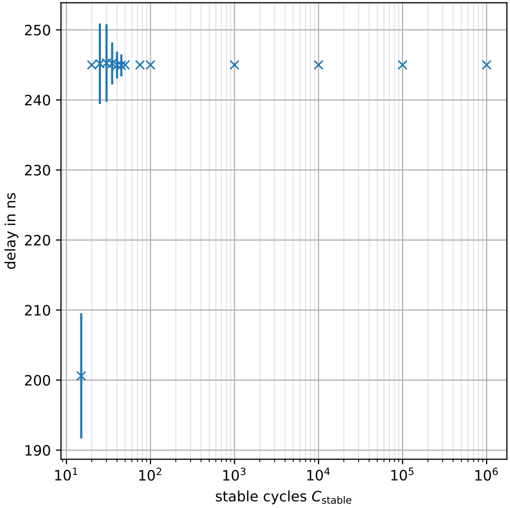
$$|S_{21}^{\text{steady}}(f_{\text{measured}}(t))| \tag{21}$$

Frequency sweep may become too fast, similar behaviour seen for fast linear sweeps

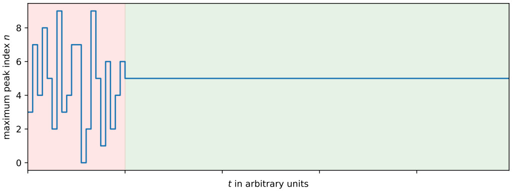


Schmitt 1966

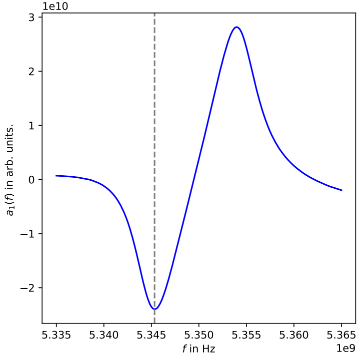
Delay measurement module sensitivity

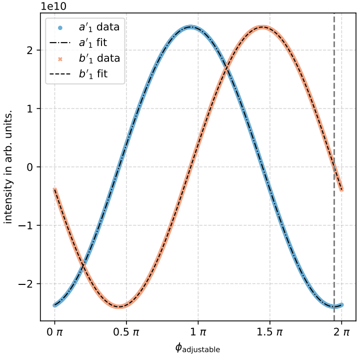


- Copper resonator circuit
- Measure at 5.3 GHz
- C_{stable} delay peak detection criteria (constant indize for C_{stable} cycles)



Phase sweep delay measurement





- First minimize a_1' over frequency, then sweep phase
- Fit results

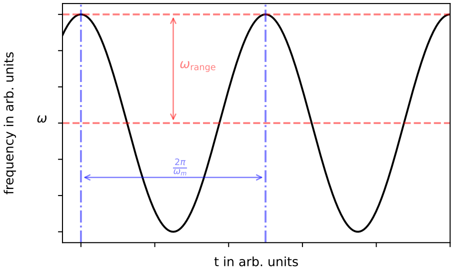
$$\tau_{a_1'} = 259.06(14)\,\text{ns}$$

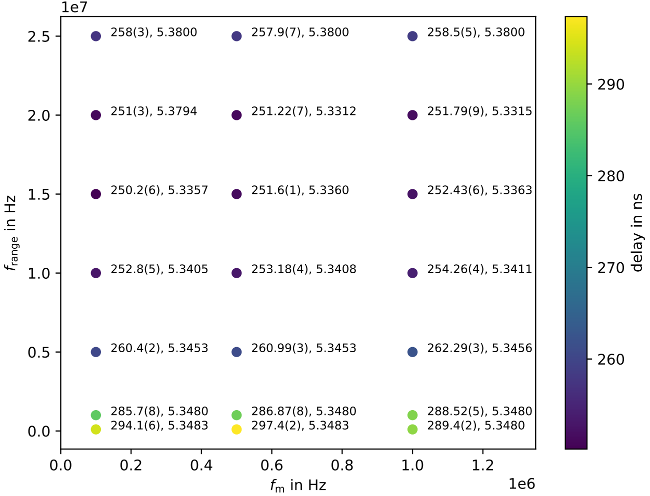
$$\tau_{b_1'} = 260.57(3)\,\text{ns}$$

- Average

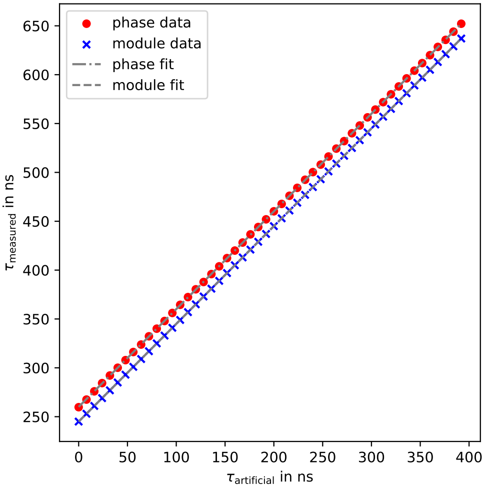
$$\tau = 259.81\,\text{ns}$$

Delay parameter sweep





Artificial digital delay



- Linear fit

$$\tau_{\text{measured}}(\tau_{\text{artificial}}) = m\tau_{\text{artificial}} + \tau_{\text{const}}$$

- Delay module result

$$m = 1.0000 \tag{22a}$$

$$\tau_{\text{const}} = 245\,\text{ns} \tag{22b}$$

- Phase method result

$$m = 1.0001(3) \tag{23a}$$

$$\tau_{\text{const}} = 260.07(6)\,\text{ns} \tag{23b}$$

Conclusion

- Implemented Multi Harmonic Detection on RFSoC evaluation board
  - Recursive higher harmonics calculation
  - FM signal generation + measurement
- Verified using microwave resonators
  - FM signal parameters limited by resonator response time
- Implemented two delay measurements methods
  - Dedicated module gives stable results
  - Phase sweep unknown influence of FM signal parameters

Outlook

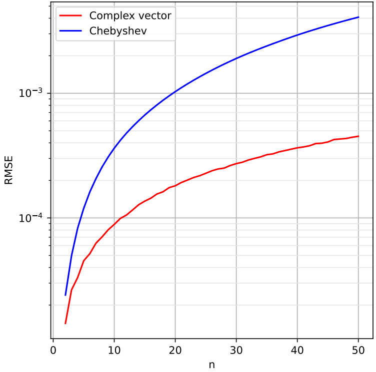
- Further investigate phase sweep delay (*B*-field modulation)
- Adjust system for real EPR setups

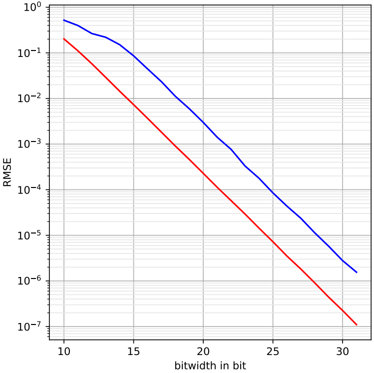
Resource usage

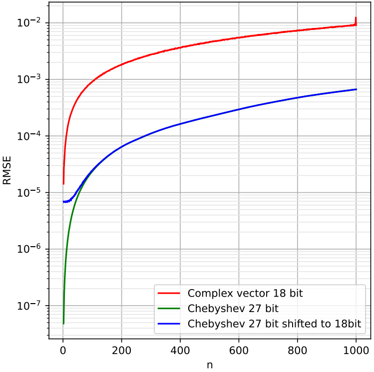
Table: All values are in percent of the Zynq UltraScale+ RFSoC Gen 3 ZU49DR . 50 harmonics are generated.

	CLB LUTs	CLB Registers	BRAM	DSPs
Overall	22.61	17.71	0.09	14.42
Harmonic measurement module	14.97	10.63	0.0	14.23
Delay measurement module	8.47	6.99	0.09	0.0
AXI interface	0.13	0.04	0.0	0.0
Power calculator	0.03	0.03	0.0	0.28

Precision recursive calculation





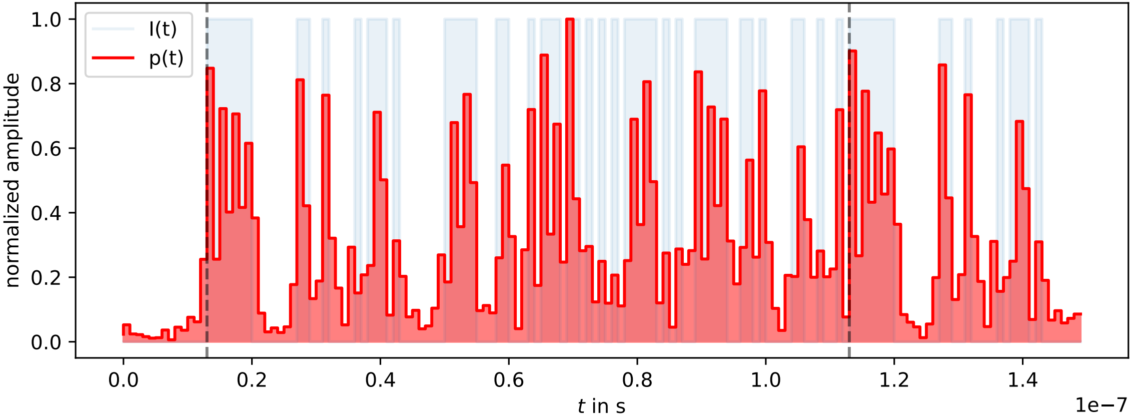


Complex vector using Eulers formula

$$\cos(n\phi) = \Re\left[\exp\left(in\phi\right)\right]$$

$$\exp\left(in\phi\right) = \exp\left(i(n-1)\phi\right)\exp\left(i\phi\right)$$

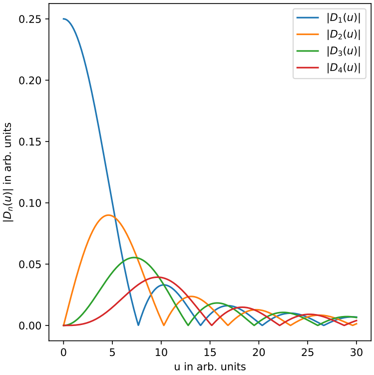
Example delay measurement

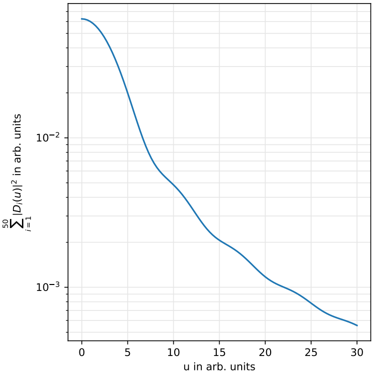


Filter function

$$D_n(u) = \left(\frac{\alpha_0'}{2n}\right) i^{n-1} \left(J_{n-1}\left(\alpha_0' u\right) + J_{n+1}\left(\alpha_0' u\right)\right) \tag{24}$$

$J_n(x)$ Bessel function first kind





Fourier coefficient delay

$$p(\cos(\omega_m t)) = \sum_{n=-\infty}^{\infty} c_n e^{in\omega_m t} \tag{25}$$

time shift $t_0 = \frac{\phi_{\text{delay}}}{\omega_m}$

$$p(\cos(\omega_m t - \phi_{\text{delay}})) = \sum_{n=-\infty}^{\infty} c_n e^{in(\omega_m t - \phi_{\text{delay}})} = \sum_{n=-\infty}^{\infty} c_n e^{-in\phi_{\text{delay}}} e^{in\omega_m t} \tag{26}$$

coefficient

$$c_1' = \frac{1}{T}\int_0^T \mathrm{d}t p(\cos(\omega_m t - \phi_{\text{delay}}))e^{-i(\omega_m t + \phi_{\text{adjustable}})} \tag{27}$$

$$c_1' = \frac{1}{T}\int_0^T \mathrm{d}t \sum_{n=-\infty}^{\infty} c_n e^{-i\phi_{\text{delay}}} e^{in\omega_m t} e^{-i(\omega_m t + \phi_{\text{adjustable}})} \tag{28a}$$

$$c_1' = e^{-i(\phi_{\text{adjustable}} + \phi_{\text{delay}})} \sum_{n=-\infty}^{\infty} c_n \frac{1}{T}\int_0^T \mathrm{d}t e^{in\omega_m t} e^{-i\omega_m t} \tag{28b}$$

$$\tag{28c}$$

Due to the orthogonality of the integral the first Fourier coefficient c_1' with adjustable phase is related to the non time shifted coefficient with

$$c_1' = e^{-i(\phi_{\text{adjustable}} + \phi_{\text{delay}})} c_1 \tag{29}$$

Delay correlation calculation

$$R_{x,y}(\tau) = \int \mathrm{d}t\, x(t) y(t-\tau) \tag{30}$$

response

$$s_2(t) = \alpha s_1(t-\tau_{\text{delay}}) + \sigma(t) \tag{31}$$

$$s_1 = b(t)\cos(\omega_c t) \tag{32}$$

$$p(t) = s_2^2 = \alpha^2 b^2(t-\tau_{\text{delay}})\cos^2(\omega_c(t-\tau_{\text{delay}})) + n(t) \tag{33}$$

$$\cos(x)^2 = \frac{1}{2}(1+\cos(2x)) \tag{34}$$

$b^2(t) = b(t)$ if $b(t) \in \{0, 1\}$, cross correlation $R_{p,b}(\tau) = c_\tau$

$$c_\tau = \frac{\alpha^2}{2}(R_{b,b}(\tau-\tau_{\text{delay}}) + R_{b\,\cos,b}(\tau-\tau_{\text{delay}})) + R_{n,b}(\tau-\tau_{\text{delay}}) \tag{35}$$