Review for mr-2025-11: "Optimized shaped pulses for 2D-SIFTER" by Trenkler et al.

In this study, Trenkler et al. discuss and show the use of shaped pulses (chirps in this case) for SIFTER. The broadband excitation by these pulses allows the "direct" detection of the whole EPR spectrum for each time point in the SIFTER trace, via a Fourier transform of the echo. This correlates the dipolar coupling in the indirect dimension with the EPR spectrum in the direct dimension. Since the EPR spectrum of nitroxides at X-band essentially encodes orientational information, the 2D correlation in these experiments allows to determine not only the distance between two labels, but also their orientation.

The use of shaped pulses in SIFTER and the dipolar/EPR correlation is not new. Chirped SIFTER was shown in (Schöps et al., 2015), albeit without a direct FT dimension. The dipolar/EPR correlation was demonstrated by (Doll and Jeschke, 2016) in Q-band, and by (Bowen et al., 2018).

Nevertheless, the present study discusses many theoretical and practical intricacies in a clear and nice way, and the application to rigid labels in RNA plus the comparison with orientation-selective DEER/PELDOR is certainly interesting to many researchers in the field. I also believe that the influence of Q_{crit} on ϕ_0 was not discussed so clearly before (at least I never thought about it in detail). This doesn't matter in many sequences, but in SIFTER it does. This should also have implications for other sequences, such as 5p-ESEEM.

Given these points, I think the paper is a valuable contribution, and certainly well suited for *Magnetic Resonance*.

I have a few points which should be considered for a revised version, and then I have some questions about the hardware. The latter is not the focus of this paper, but I ask anyway because I am interested.

1) The chirp-SIFTER papers above are already cited in the manuscript, but given the title, I think it would be appropriate to mention them in the introduction already.
2) Line 16: I think "orientation selective SIFTER" is a bit unfortunate in this context. I know where the author's come from, but the particular experiment is as orientation NON-selective as it gets. I would use "EPR-correlated", or "Orientation-correlated", although the latter is a bit less accurate.
3) Line 23: I think the nuclear spin with the largest magnetic moment is (^{3}H) ;)
4) Line 46: "However, for shaped pulses it is essential to keep all amplifiers in their linear regime, to preserve the designed pulse shape at all power levels."
   I disagree. At X- and Q-band where there are true amplifiers and not amplifier-multiplier chains, the power amplifier can be driven into saturation. Especially for chirps, the amplitude non-linearity is not very detrimental, unless the input

signal is not clean. If LO leakage is significant, the non-linearity will introduce harmonics at $\omega_{LO} + n\omega_{awg}$, which can be very bad for low AWG frequencies.
I do agree however, that the non-linearity must be taken into account, especially for amplitude modulation. In commercial NMR spectrometers, this is routinely done, in a process called "linearization".

5) In the definition of the WURST pulse as given in the paper, the phase at t=0 (middle of the pulse) is not 0, but $\phi(t) = \int_{t_p/2}^{t} \Delta\omega(\mathrm{t}')\mathrm{dt}' = \pi SW\left(\frac{t^2}{tp} - \frac{tp}{4}\right)$, so $\phi(0) = -\frac{1}{4}\pi\ SW\ t_p$. This is the point where it crosses the spins at 0 offset and contains the TBP. I would just like the authors to confirm that the particular definition has no influence on their discussion regarding Q_{crit}, ϕ_0 and TBP.
6) Line 267: " It also needs to be verified that the output of the AWG (arbitrary waveform generator) is linear with respect to the input amplitudes for fast amplitude changes."
I would just like to highlight that the Nyquist criterion alone is not sufficient to talk about the bandwidth of an AWG or a digitizer. While the authors use an AWG with 0.625 ns sampling step, the info I found online also mentioned an analog bandwidth of 400 MHz, which is exactly what the authors see in Figure A 3. In fact, the analog bandwidth can be smaller (oversampling) or higher (undersampling) than the Nyquist frequency.
7) Line 685: "All shaped pulses used in this study were corrected with the transfer function obtained from the resonator profile by the procedure described by Doll et al. (2013)." Is unclear to me. Did you get the transfer function from the resonator profile as A. Doll did, or did you correct the pulses like Doll, or both? The "correction" could mean a) deconvolution with the transfer function, or b) adjusting the sweep rate of the chirp according to the resonator profile.
8) Line 695: "This profile was in most cases not at all flat and there still seem to be considerable imperfections in either the excitation of the nitroxide or in the broadband detection". It is expected that the detection is influenced by the resonator profile. It has to be, and this cannot be compensated in the excitation.
9) Line 277: "In both spectrometers, there was no clock synchronization of the spectrometer pulses and transient recorder to the AWG. In the case of our home-built spectrometer, since the oscilloscope has a much higher time resolution (0.1 ns compared to 0.625 ns of AWG), this was not a concern."
I find this puzzling. I don't see how the higher time resolution of the oscilloscope solves the syncing problem, unless you digitally phase-correct each transient. I can see that for synchronized triggers and low frequency (IF) signals, this might not matter so much. But I would expect that for the higher frequencies in the nitroxide spectrum, averaging echoes with varying phase should lead to some attenuation. Did you verify that echoes average coherently over many shots and over longer time periods?

10) Line 359: “Importantly, 360 the great agreement between the SIFTER echo FT and the EDFS was achieved only after identifying, through initial testing, a spectrometer carrier frequency at which standing waves did not significantly distort the microwave pulse shape or the detection.”
    I suspect not only standing waves, but also mixing artifacts/spurious frequencies, due to the IQ mixing with the LO in the middle of the resonator and spectrum.
11) Line 414: The echoes in orientation-selective DEER/PELDOR could also be Fourier-transformed.