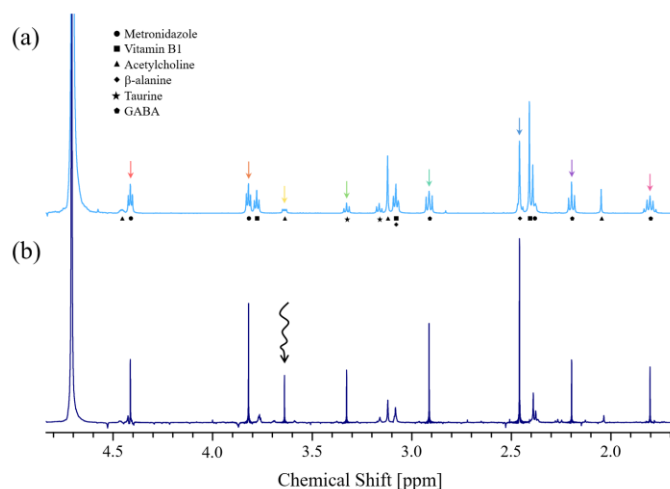


Editor comments are shown in black, our reply in green and the revised text in blue.

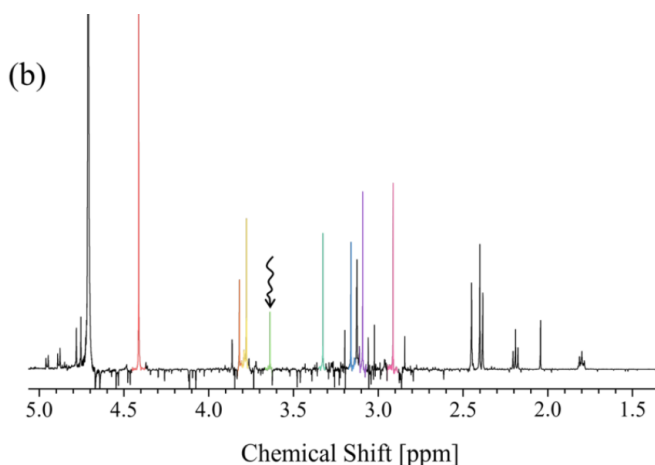
The authors have addressed most of the concerns and questions raised at the discussion, where appropriate indicating that some of the points need further research, which is a positive outcome of a good paper. Both referees have indicated that the paper describes an exciting development in NMR methodology and should be published. There are a couple of minor points raised by the referees that in my opinion would benefit from further clarification.

1) Both referees point out the lack of mentioning the Bloch-Siegert (BS) effects in polychromatic selective spin-lock (SSL) experiments. While the authors brush aside this argument suggesting that the BS effects were negligible, they certainly are not in Figure 9 where two of the simultaneously irradiated multiplets are only 25 Hz apart (response to referee 1 in point 4), or even 6.5 Hz as mentioned by the referee 1, which is comparable with (or even less than) the SSL field strength (typically 25 Hz as indicated in the manuscript).

- 1) We thank the editor for pointing out this detail. In fact, we realized that while selecting the irradiation frequencies of SSL pulses in the experiment illustrated in originally submitted version of Fig. 9, we made a poor choice of applying two irradiations very close to each other. We believe that this might explain the presence of multiple artifacts in the resulting spectrum. To test this hypothesis, we repeated the experiment with the same sample, but this time chose 8 SSL frequencies such that they are at least 25 Hz apart from each other. The resulting revised Fig. 9 is shown below:



The number of artifacts has dropped significantly with respect to the original submission:



Note that one can easily identify the Bloch-Siegert (BS) shifts (Bloch and Siegert, 1940) in these spectra; for instance, the set of peaks at ca. 2.38 ppm is right-shifted by 8 Hz, 25 Hz away from the irradiation frequency (blue arrow at 2.48 ppm), which is a clear signature of a BS shift. Another set of BS-shifted peaks at 3.76 ppm are right-shifted by 7 Hz compared the orange arrow. The peak at 2.05 ppm is also right-shifted by 7 Hz compared the purple arrow.

The multiplet at 3.08 ppm is slightly left-shifted by *ca.* 1 Hz because it is exposed to two RF fields represented by two green arrows that are almost symmetrically disposed with respect to the multiplet.

Another observation from Fig. 9 is that the peaks that are BS shifted have strongly attenuated intensities. This can be explained by noting that the initial magnetization vector in the transverse plane can be decomposed into two components, one that is parallel to the B_1 field and therefore spin-locked, while the component orthogonal to the effective field rotates in a plane perpendicular to this field. Therefore, these signals are both frequency-shifted *and* attenuated.

Quantitative comparisons between conventional and SSL spectra also require consideration of the steady state established during repeated scans. In a conventional 1D experiment, the interval available for longitudinal recovery is the recovery delay plus acquisition period, because recovery towards equilibrium continues during acquisition. In an SSL experiment, the repeated RF pulses applied during the acquisition can perturb longitudinal magnetisation as it recovers, so that the effective unperturbed recovery interval for the irradiated spins may be closer to only recovery delay. Consequently, when it is not sufficiently long compared with T_1 , the conventional and SSL experiments may establish different steady states, leading to attenuation of the SSL integrals. This effect is relevant only to multi-scan acquisitions.

Another important consideration is that during SSL acquisition, the receiver is blanked while the RF pulses and associated dead-time intervals are applied, and each complex FID point is derived from the signal acquired during the remaining observation window. This interrupted acquisition reduces the effective observation time per dwell and may decrease the signal-to-noise ratio or introduce implementation-dependent amplitude scaling and modulation artefacts. To first order, such a receiver-window effect should act as a common factor on all resonances and therefore cannot by itself explain selective attenuation of individual signals. A systematic separation of these effects using calibrated windowed reference experiments is left for future work.

Bloch, F. and Siegert, A.: Magnetic resonance for nonrotating fields, *Phys. Rev.*, 57, 522–527, <https://doi.org/10.1103/PhysRev.57.522>, 1940.

As pointed out by the referee 2 if the polychromatic SSL waveforms were created using the Wavemaker (Bruker) the BS shifts were compensated for automatically which would explain the lack of the interference effects as observed by the authors.

We did not use any Wavemaker, only phase incrementation calculated on-the-fly with the pulse sequence running.

Otherwise the statements claiming that the BS effects were negligible in responses to points 4 and 6 of referee 2 would remain unresolved.

We now have responded in great details.

- 2) Surely some of the artifacts in Fig. 9 can be identified as decoupling sidebands (both of the same phase) or modulation sidebands (opposite phases as observed in homonuclear decoupling experiments) due to RF modulation in polychromatic SSL.
Other than that the manuscript is ready for publication.

The artifacts in the revised version have essentially disappeared.