

Dear Patrick,

please find below the point-by-point response to both reviewer's comments. We really put a lot of effort into the revision and I hope you will find the quite interesting manuscript acceptable for publication.

Best regards,

Burkhard (on behalf of both authors)

- **RC1**

The novelty of this work is in the introduction of the Dual Bilinear Rotations which allows simultaneous spin system selective manipulations on both spins I and S as compared to the coupled spin system IS. The material is well presented and argued. Its value is in the introduction of a new concept rather than in the current presented example. I support publication of this manuscript and have only minor comments.

>>> We thank the reviewer very much for this statement.

Can authors present the 2D spectra in the SI, rather than just their projections?

>>> The 2D J-spectra are now added in the supporting information.

The authors have chosen to illustrate the technique on a fully ^{13}C labelled glucose. This introduces complications because of the presence of ^{13}C - ^{13}C coupling constants. How would these experiments behave on ^{13}C natural abundance samples? How would these experiments behave on 5-10% enriched ^{13}C natural abundance samples that are typically used to boost the sensitivity yet limiting complication of ^{13}C , ^{13}C coupling constants?

>>> The carbon spectra of natural abundance or 5-10% level isotope labeled ^{13}C samples would improve significantly due to the lack of large ^{13}C , ^{13}C -couplings. Corresponding spectra would probably be artefact free with all selected carbon signals being of very similar intensities, while unselected carbons would nearly vanish. At the same time, however, proton spectra would lose their selectivity, as the majority of protons would not be coupled to ^{13}C and a selection of OH groups would not be possible. At the same time, ^{13}C -bound protons would be very weak and artefacts originating from residual non- ^{13}C -bound protons could not be suppressed properly. These thoughts did prevent us from recording corresponding spectra. Following the comment and also the comment of the second reviewer, we recorded experiments on a sample with a mixture of different very small molecules at natural isotope abundance with different multiplicities. In this case, clearly artefacts in the ^{13}C projections are reduced to a minimum (see new figure 6). We hope the reviewer finds the new experiments representing his way of thinking.

Fig. 4c: "The blue sub spectrum is designed to mainly contain directly ^{13}C bound" Can the authors comment on why is the β -D glucose signal so pronounced?

>>> We very much thank the reviewer for this question as we were also wondering for the reason but only now found the nearly trivial answer to the question. The reason lies in the apodization of the J-resolved proton spectra: The acquisition time in the J-dimension was

250 ms. Although expected signals are in principle inphase with maximum signal intensity at the beginning of the acquisition time, apodization was chosen to be a sine function to ensure sharp, absorptive-like lineshapes for the magnitude spectra. It turns out that the H1a signal with $J(H1a,H2a) = 4$ Hz has a zero crossing at the center of the t_1 -evolution period, while its maximum sensitivity at the front and end of the t_1 -period is apodized away. The signal is therefore maximally suppressed. The H1b signal with a roughly 8 Hz coupling to H2b, instead, has its maximum intensity also at the center of the t_1 -period, where the apodization is not suppressing the signal. As a result, the anyway intensive signal appears with maximum intensity. We added now the very same projection of proton spectra using an exponential apodization function, where signal intensities are more the way one would expect them, although spectra are less resolved. While adding the spectra, we also noted that we had an issue with the Bruker Topspin parameter `nc_proc`, which caused errors in the comparison of signal intensities for proton and carbon spectra. We corrected the corresponding spectra.

Page 6, line 119: typo in “homonuclear and heteronuclear couplings evolve to the ...”

>>> The typo has been corrected.

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• **RC2:** ['Comment on mr-2026-1'](#), Anonymous Referee #2, 13 Mar 2026

General comments

The article presented by Woordes and Luy presents a new approach to application of multitude of Bilinear Rotation (BIR) elements that could be used in NO Relaxation Delay (NORD) NMR super-sequences, allowing to simultaneously acquire multiple different experiments with the same recycle delay. The family of four BIR elements (BIRD, TANGO, BANGO and BIG-BIRD) allowing to achieve different rotations on different coherences in the same time is presented. Thanks to the use of uniform rotation pulses, almost any combination of rotation angles may be achieved independently and concurrently for directly or remotely J-coupled spin systems on the chosen spectral widths. These Dual Bilinear rotations have been applied to the quadruple J-resolved experiment, in the same time revealing up to four different spectra, acquired with one relaxation delay. The manuscript is written well, and thanks to introduction to the nomenclature of the BIR elements, it should be understandable for an NMR spectroscopist, who had limited knowledge on the matter beforehand. The presented experiment has been discussed in detail with benefits and possible issues inherent to the method. However, the quality of the suppression of undesired coherences in the quadruple J-resolved experiment is slightly disappointing, and I would like to propose to test the method on a different sample, free from homonuclear ^{13}C couplings (*vide infra*). On the general side, I find that currently sufficient experimental information has been given to reproduce the experiments independently.

Specific comments

As in NMR spectroscopy, the rate limiting factor influencing the duration of a pulse sequence often is determined by up to a few-second long recycle delay, the presented method may bring significant time savings in the acquisition of multiple experiments following the NORD principles. I find the quadruple J-resolved experiment an elegant illustration of the capabilities of the Dual Bilinear Rotation elements, as it allows to concurrently obtain complementary information on the spin systems featured by the molecules in question.

>>> We very much thank the reviewer for this encouraging statement!

It would be intriguing to elaborate more on the potential ways of handling: (1) the long range heteronuclear couplings and (2) homonuclear couplings. Presumably, the first should be easier to handle, while the latter would lead to significant magnetization losses as it is inherent to the free from homonuclear couplings pure shift spectra. The numerical or experimental verification of such approach would probably be beyond the scope of this article, but explanation on how one can potentially deal with the two aspects could be mentioned in the main text.

>>> The effect of all kinds of coupling constants on BIRD elements as the first kind of bilinear rotation has been studied already intensively in the literature. Especially several publications by V. Rutar exist about this subject with "Effects of long-range and homonuclear couplings on bilinear rotations", JMR 71, 75-82 (1987), maybe being the publication addressing the problem most directly. Indeed, small heteronuclear as well as homonuclear couplings can be tolerated, but the element is based on the distinction via the large direct heteronuclear coupling vs. all other couplings. The sum of two ^{13}C , ^{13}C -couplings is half as large as the heteronuclear coupling and resulting spectra start to lose their full distinctive power.

Concerning the application of the Coupling Offset B_1 -compensated (COB) principle to the Dual Bilinear Rotation Elements, it would be interesting to present the method on a slightly more complex molecule but with less complexity in terms of homonuclear couplings spin system than glucose. For instance, one could try to apply the experiment to some other small molecule, which features a range of heteronuclear J-coupling values (weak long-range and strong short-range), and for which the presence of homonuclear ^{13}C - ^{13}C couplings is less problematic. For instance, one could use ^{13}C sample specifically labelled at certain distant positions, to avoid the presence of strong homonuclear ^{13}C couplings. In the same time this sample could feature a range of (short- and long-range) heteronuclear couplings for which one could design the method to either suppress or promote to give the signal, depending on the delays in the COB element(s). This addition to the article would enhance the illustration of applicability of the J-resolved method, as it is postulated in the manuscript.

>>> It would be nice to have such a molecule in hand, but we don't and we also do not find such a molecule in a catalogue. Instead, taking also the corresponding comment of reviewer 1 into account, we put together a sample with four compounds with a single carbon, but each representing a different multiplicity, i.e. Cq, CH, CH₂ and CH₃. Natural abundance spectra are recorded demonstrating the carbon multiplicity selection without heavy ^{13}C , ^{13}C -coupling compromise, but essentially useless proton spectra. We recorded spectra using the conventional dual bilinear rotations and the COB-enhanced dual bilinear rotations. Indeed, in the case of unconventional coupling constants like the 210~Hz one-bond coupling of CHCl₃, the conventional bilinear rotations fail the selection, while the COB-version of the quadruple J-resolved experiment shows almost perfect spectra without any distortions due to coupling mismatch or additional couplings. We added a figure with the carbon spectra and a describing paragraph in the main text.

Technical comments

In the line 107: I presume that "Cq" denotes quaternary carbon. Please make the assignment more explicit, as it might be not obvious for the reader.

>>> We changed the text accordingly to give a more explicit explanation.

In the figure 3, there is a plethora of spin rotations happening. One could benefit from addition of a subfigure with simplified effect of the Dual Bilinear Rotations in each sandwich (e.g. Π_x on spin I, Π_x on spin S) similarly to the colored parts of the Figure 2.

>>> We extended the Figure as suggested with several simplifications for didactics. Indeed, the sequence can be grabbed much easier this way. Thank you very much for this comment.

In the figures 4 and 5, one could increase the font size of the x-axis labels, to match the one of the titles of the spectra. The way it is right now requires to zoom-in to better see the numbers and the [ppm] marker.

>>> The x-axis labels have been increased.