Response to “Comment on ‘Indications of energetic consequences of decoherence at short times for scattering from open quantum systems’” [AIP Advances 1, 049101 (2011)]

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Response to "Comment on 'Indications of energetic consequences of decoherence at short times for scattering from open quantum systems'" [AIP Advances 1, 049101 (2011)]

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The Comment by Mayers and Reiter criticizes our work on two counts. Firstly, it is claimed that the quantum decoherence effects that we report in consequence of our experimental analysis of neutron Compton scattering from H in gaseous H_2 are not, as we maintain, outside the framework of conventional neutron scattering theory. Secondly, it is claimed that we did not really observe such effects, owing to a faulty analysis of the experimental data, which are claimed to be in agreement with conventional theory. We focus in this response on the critical issue of the reliability of our experimental results and analysis. Using the same standard Vesuvio instrument programs used by Mayers *et al.*, we show that, if the experimental results for H in gaseous H_2 are in agreement with conventional theory, then those for D in gaseous D_2 obtained in the same way cannot be, and vice-versa. We expose a flaw in the calibration methodology used by Mayers *et al.* that leads to the present disagreement over the behaviour of H, namely the ad hoc adjustment of the measured H peak positions in TOF during the calibration of Vesuvio so that agreement is obtained with the expectation of conventional theory. We briefly address the question of the necessity to apply the theory of open quantum systems. [doi:10.1063/1.3660422]

I. INTRODUCTION

The main claim of the Comment by Mayers and Reiter is that the data obtained in our original paper,[1] if analysed using the standard Vesuvio instrument programs, are in "very good agreement with conventional theory."[2] Consequently, Mayers and Reiter propose that the new quantum decoherence effect that we claim to have measured[1] is "almost certainly a spurious consequence of errors in the data analysis."[2]

In section II we address the point about the reliability or otherwise of our analysis in two steps. First we demonstrate that an experimental comparison between scattering from H and D is reliable. When combined with experimental results for gaseous hydrogen and deuterium, obtained with the same instrumental setup and the same pressure cell, this suffices to prove that H and D cannot both be accounted for by conventional theory. We then show that the standard Vesuvio instrument programs produce an anomalous result for D in consequence of the "very good agreement with conventional theory" for H. Then we expand on the reasoning by which we conclude that in our data

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sets H is anomalous, while D is described by conventional theory in agreement with long-standing publications.

An additional criticism[2] concerns theoretical issues of the quantum dynamics of scattering from open quantum systems, based on the authors' belief that standard theory (Ref. 3 is advocated here) is fully sufficient in the physical context under consideration. These issues are briefly addressed in section III.

II. EXPERIMENTAL CONSIDERATIONS

The principal basis for the criticism of our work by Mayers and Reiter is that our analysis of our experimental data is faulty. The explanation offered is that "The authors of [our paper[1]] used their own analysis programs rather than the standard instrument programs [their Ref. 6], available to users of VESUVIO. They give no details of their procedure. It seems that they made no correction for multiple scattering . . . in the sample or the existing sample dependent gamma background . . . ," followed by the speculation that "The most likely explanation of the shift to positive y shown by the authors of [our paper[1]] in their Fig 2(b), is that the procedure they used to obtain $J(y)$ from the measured time of flight spectra contained an error." We now scrutinise these objections.

A. Software

We state that (i) we actually used the Vesuvio YTRANS program to transform our TOF spectra into y space; (ii) the other software that we used has been benchmarked against the Vesuvio software and produces the same results; (iii) our conclusions do not depend in any way on which software package was used.

B. Data analysis methodology

We now proceed to the crucial issue of the reliability of our data analysis. This necessarily involves an understanding of the reliability of Vesuvio itself, i.e. its calibration and stability.

Producing a momentum profile, $J(y)$, involves two steps. First the TOF spectrum is processed, using the universally accepted relation (Eq. (A1) in our paper[1])

$$t = \frac{L_0}{\upsilon_0} + \frac{L_1}{\upsilon_1} + t_0. \tag{1}$$

Here L_0 is the source – sample distance, L_1 the sample – detector distance for an individual detector, υ_0 is the incident velocity and υ_1 the scattered velocity of the detected neutron. Each detector is at an individual scattering angle θ and t_0 is a small time offset, caused largely by electronic delays.

As detailed in Eqs. A2–A7 in our paper,[1] the momentum transfer ($\hbar q$) and energy transfer ($\hbar\omega$) are then obtained and the TOF data are straightforwardly transformed to y space. We reiterate that we used the ISIS routine YTRANS for this transformation.

Mayers and Reiter themselves provide the answer their point about the absence of a "correction for multiple scattering in the sample or the existing sample dependent gamma background." Comparing their Fig. 2 (including these corrections) and Fig. 3 (no corrections) demonstrates that the position of $J_H(y)$ is the same with or without corrections for multiple scattering or gamma background. We deliberately did not make these corrections because (i) they are not needed in the case under consideration because the reported effect is so strong that neither correction can remove it; (ii) in our experience these corrections introduce an artificial systematic and statistical "noise" into the corrected TOF profile.

C. Reliability of comparisons between $J(y)$ for H and D

Central to the present disagreement, the position of a peak in the TOF spectrum or in y space depends entirely on the set of instrumental calibration parameters (principally L_0, L_1 and θ in

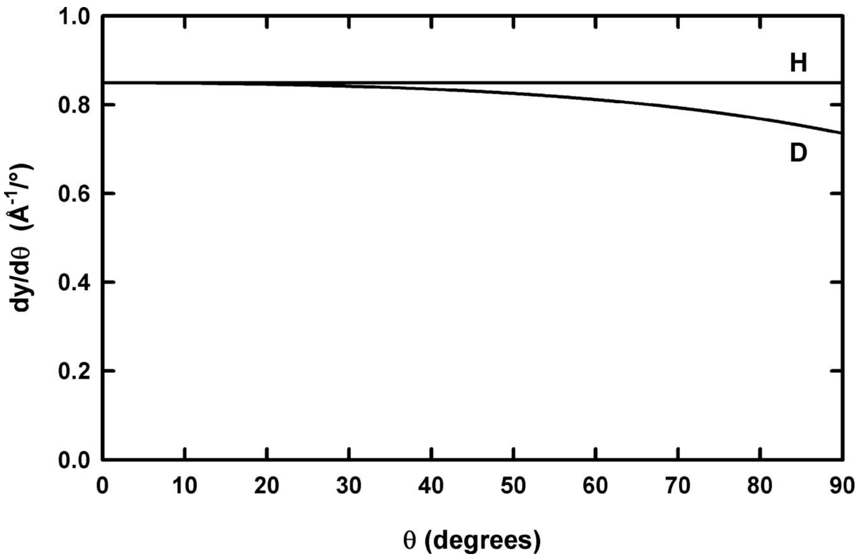

FIG. 1. Effect of changes in detector angle the positions of the H and D recoil peaks, with $\partial y/\partial\theta$ plotted on the vertical axis for $L_0 = 11.0$ m, $L_1 = 0.6$ m, $k_1 = 48.66$ Å^{-1}.

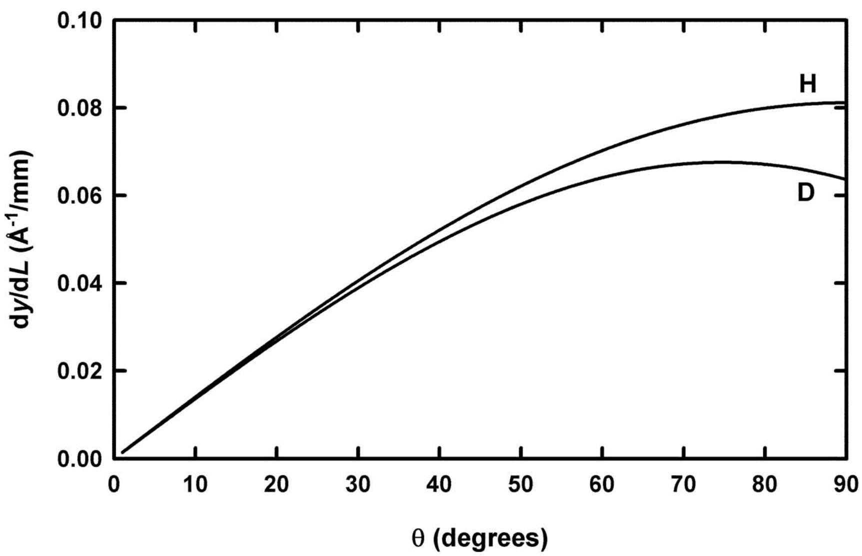

FIG. 2. Effect of changes in sample position, L, on the positions of the H and D recoil peaks, with dy/dL plotted on the vertical axis for $L_0 = 11.0$ m, $L_1 = 0.6$ m, $k_1 = 48.66$ Å^{-1}.

the present context) that is tabulated for each detector in the Instrument Parameter (IP) file. The methodology used to construct the Vesuvio IP file is detailed (albeit incompletely) in Ref. 4 The standard Vesuvio instrument programs use this file. The IP file changes from time to time as Vesuvio is recalibrated or its calibration is adjusted (see subsection II D), so it is important to use the file associated with the period of each experiment.

It is reasonable to expect that data sets obtained during the same instrument epoch (meaning that the physical instrument is unchanged and the same basic IP file applies) may be reliably compared, even if they are regarded as unreliable on an absolute basis. This is especially true when, as in the present case, the data sets were also obtained with the same ancillary apparatus and methodology, so that only differences between the samples, or differing effects caused by the samples, can contribute to the observed differences in scattering. Aside from the physics of the

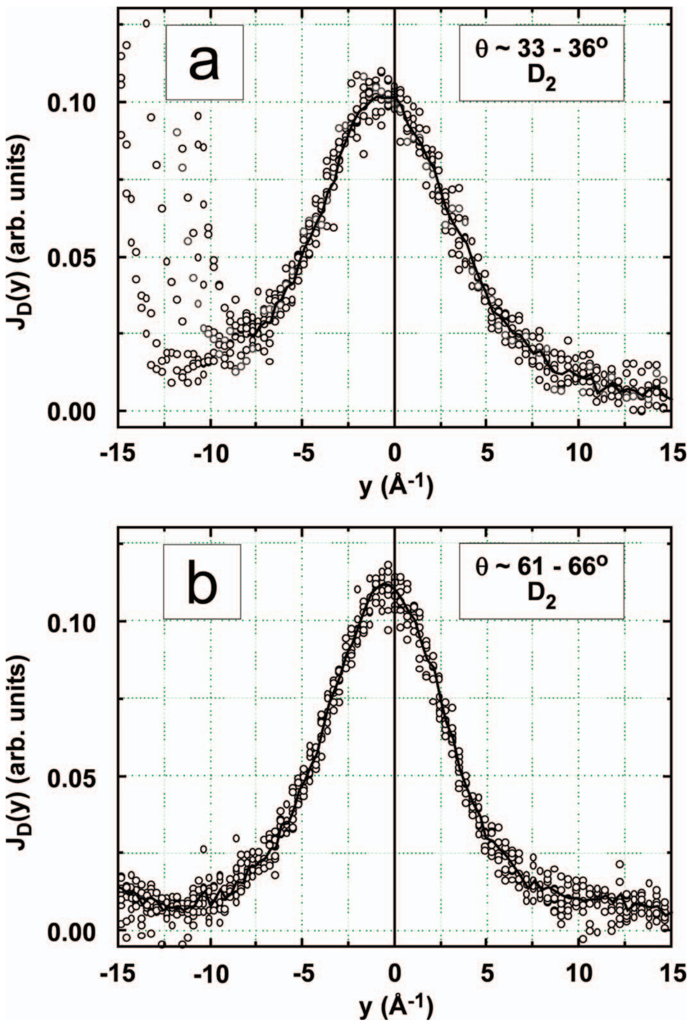

FIG. 3. The experimental (not normalised) Compton profile $J_D(y)$ of D in D_2 gas,[1] obtained from untreated (i.e. as measured, with no corrections) time-of-flight data using the "standard" IP file. $J_D(y)$ measured (a) with one block of 8 detectors, at the low scattering angles $\theta \sim 33$–$36°$, and (b) with one block of 8 detectors at the high scattering angles $\theta \sim 61$–$66°$. In (b) the D peaks exhibit an artificial shift to $y < 0$, which contradicts numerous literature reports; see the text.

samples themselves, the potential contributors to an observed difference between samples are the gamma background, multiple scattering, and inconsistencies in the calibration that affect the TOF peaks for the two sample masses differently, including differing location of the two samples with respect to the ideal position. Having discounted corrections for the gamma background and multiple scattering as inconsequential at the absolute level (see subsection II B), more so at the differential level, we turn to calibration effects.

The dominant parameter affecting the absolute TOF peak position on Vesuvio is the detector angle.[4,5] The sensitivity of the position of the recoil peak to detector angle θ is given by[5]

$$\left.\frac{\partial y}{\partial \theta}\right|_{y=0} = \frac{k_1^2 \kappa s}{q}$$

where

$$\frac{1}{\kappa} = \frac{\upsilon_1}{\upsilon_0} = \frac{k_1}{k_0} = \frac{\mathrm{c} + \sqrt{\mu^2 - \mathrm{s}^2}}{1 + \mu}$$

and

$$q^2 = k_1^2(\kappa^2 - 2\kappa c + 1)$$

with $c = \cos\theta$, $s = \sin\theta$, k_0 the initial neutron wavevector and k_1 the final neutron wavevector. This result is equivalent to that obtained by a completely different analytic route by Andreani *et al.*[6] Figure 1 shows this sensitivity with parameter values appropriate to Vesuvio ($L_0 = 11.0$ m, $L_1 = 0.6$ m, $k_1 = 48.66$ Å^{-1}). H and D are affected in very nearly the same way, with only 0.03 Å^{-1} per degree difference in sensitivity to an error in θ at $\theta = 60°$. That is, an error of even several degrees in the measured detector angle would not significantly affect an H–D comparison. Therefore a comparison performed between data from H and D with all other experimental parameters held constant must be valid with high reliability on a y scale conservatively set at 0.1 Å^{-1},[5] which is very small compared to the scale of effects at issue here.

A potentially more significant effect is that of the independent error in the location of the H and D samples between two experiments. This can be calculated from the sensitivities to L_0, L_1 and θ.[5] Figure 2 shows this sensitivity (with parameter values appropriate to Vesuvio) to be 0.06–0.07 Å^{-1} per mm of sample displacement along the beam axis at $\theta = 60°$, for H or D. Because the gaseous samples were contained in a pressure vessel firmly and reproducibly located within the cryostat, which is itself reproducibly centred on the ideal sample position, we estimate that the maximum likely error in the sample location is 1.5 mm, leading to a maximum likely difference between H and D owing to sample mis-location of 0.2 Å^{-1}, again insignificant on the scale of effect at issue.

We therefore conclude that the comparison between H and D displayed in Figs. 1 and 2 of our paper[1] is reliable on a y scale that is small compared to the shifts greater than 1 Å^{-1} in the $J(y)$ peak that we claim to have measured.

The inescapable corollary to this conclusion is that if, as Mayers and Reiter claim, the results for H are in "very good agreement with conventional theory",[2] then our $J(y)$ profiles for D (Fig. 1 in Ref. 1) need to be shifted significantly in the negative y direction. In consequence, the (presumed) final state effects[7,9] (FSE) will become much unreasonably large and will not tend towards zero at large q, as required by the Impuse Approximation,[7–9] seriously contravening conventional theory.

Alternatively, if D does tend to obey the Impulse Approximation at high momentum transfer as we concluded[1] in agreement with previously published work by Andreani *et al.*,[8] then H does not obey conventional scattering theory.

These considerations suffice to discount the claim by Mayers and Reiter that our observation of effects outside the framework of conventional theory is wrong. We further point out that Mayers and Reiter address only our analysis for H and do not consider the robustness and significance of the comparison with D.

D. Absolute reliability of *J*(*y*) for H and D

We now proceed to examine the origin of the disagreement over the analysis of our experimental data for H and D.

Using the same "standard" IP file (meaning that available at ISIS for use by the standard Vesuvio instrument programs) applying to the period in which the experiment was performed, we obtain exactly the same result for H that is shown in Fig. 1 of Ref. 2, implying (apparent) agreement with conventional theory.

Figure 3 shows our data for D from Fig 1 of our paper[1] reprocessed using again the standard Vesuvio instrument programs and the standard IP file applying to the period in which this experiment was performed. As expected from the discussion in the previous subsection, the D peak has been artificially shifted towards negative y for large scattering angles. Furthermore, the shift is independent of scattering angle within experimental error, so that the (presumed) FSE does not diminish as expected from conventional theory.[7–9]

As detailed in our paper,[1] we used calibration parameters measured directly with steel rules and a protractor,[4] by the ISIS Metrology Group, for our data analysis. In contrast, the standardly available Vesuvio IP files are initially obtained by a sequential procedure involving inelastic and diffraction measurements.[4] Crucial to the present disagreement, however, the calibration parameters may then be adjusted to ensure that the position of the H peak, with the standard correction for FSE incorporated in the peak shape, conforms to conventional theory.[10] In our view this adjustment renders Vesuvio invalid for investigations of hydrogen in which the location of the H recoil peak is of importance for further analysis, such as correcting for FSE and calculating the momentum distribution. As an example, an important consequence of this adjustment is that violation of y-scaling[7] (i.e. the non-constancy of FSE-corrected $J(y)$ with q), which is clearly visible in Fig. 2 of our original paper,[1] is obscured by summing Compton profiles over all detectors in the "standard Vesuvio instrument programs."

Our results for D, shown in Fig. 1 of our original paper,[1] agree with well-established and repeatedly confirmed results documented in the scientific literature.[8,9,11] This does not suffice to prove that D does obey conventional theory, but the following examination of published experiments and theory strongly indicates that it is so. Comparing the y positions of the experimental D Compton profiles measured with the eVS ancestor of Vesuvio, shown in Fig. 26(a,c) of the review article[9] with our D profiles shown in Fig. 1(a,b) of our original paper[1] demonstrates that they are equivalent in the common angular range of the two experiments. Given that D causes backscattering (in contrast to H), one might argue that the Impulse Approximation may not be obeyed at these forward scattering angles, but only at backscattering angles where the momentum transfer to D becomes similar to that to H at $\theta \sim 65^\circ$. This point is resolved by Fig. 26 of Ref. 9, which also shows the theoretical Comtpon profile of D (in ortho-D_2) calculated in the frame of conventional theory. The Impulse Approximation is theoretically predicted to be obeyed at high forward scattering angles (i.e. $\sim 60^\circ$–70°), so there is a genuine agreement between experiment and conventional theory for D.[8,9,11]

Because we obtained excellent agreement with previously (and independently) analysed and published data on D,[8] using calibration parameters that were not adjusted according to expectations for H, we conclude that (i) it is in fact D that obeys conventional theory within the (ω,q) domain of the data reported in our paper[1] and (ii) H does not obey conventional scattering theory.

III. THEORETICAL CONSIDERATIONS

Mayers and Reiter[2] agree with us[1] that, in the frame of every conventional theory (including that put forward in Ref. 3), the Compton profile can only be centred at momentum $\hbar y \leq 0$. Demonstrating that the recoil peak in y space can move to positive y is therefore tantamount to requiring a different theory, which we believe has to be the quantum dynamics of open systems.

As a matter of fact, the formalism of conventional neutron scattering theory describes transitions between stationary states, which are the eigenstates of the complete Hamiltonian H_c of a closed system.[12] Open quantum systems[13] are not considered in conventional theory.[3,9,12] Consequently, quantum phase-destruction phenomena, like decoherence and the continuous quantum Zeno and anti-Zeno effects,[14] are nonexistent in conventional theory. In more formal terms, time-dependent processes of conventional theory exhibit a unitary time evolution, whereas the quantum dynamics of open quantum systems, which in general includes decoherence and dissipation and thus breaks time-inversion invariance, is non-unitary.[13,14] This difference is fundamental and therefore it can have serious consequences.

The theoretical paper[3] to which Mayers and Reiter refer as able to account for decoherence effects offers only a time-independent perturbative treatment of the scattering process including the electronic degrees of freedom. This treatment is only valid for closed systems and cannot account for decoherence. Moreover, it should be stressed that scattering necessarily introduces an entanglement of the neutron with the scattering particle(s) of H_2,[1] which however plays no role in Ref. 3 or in any other conventional neutron scattering theory.

We further remark that the "strong coupling limit" advocated in Ref. 3 leads to an intensity deficit[15] of the main H recoil peak owing to the breakdown of the Born-Oppenheimer approximation,[3] producing a broadly distributed spectral density which the gamma and multiple-scattering corrections applied via the standard Vesuvio instrument programs[2] do not correct for. Therefore, this should add an artificial contribution to the wings of the Compton profile at large $|y|$, which then could be misinterpreted as a feature of the physics of the sample. We are unaware of any application of this theory[3] to experiments to test its validity.

IV. SUMMARY AND CONCLUSIONS

Mayers and Reiter claim[2] that our observation[1] of a shift to positive *y* values of the H recoil peak is wrong owing to faulty data analysis. We have refuted this claim by establishing the following points.

1. The software that we used for data analysis is not the origin of the disagreement between us over the position of the H recoil peak in *y* space.
2. The effects that we observe are not removed by correcting for multiple scattering or gamma background.
3. By analysing the sensitivity of Vesuvio to changes or errors in the flight-path calibration parameters,[5] we showed that a comparison between the positions of the H and D recoil peaks is reliable to 0.2–0.3 $Å^{-1}$ in our experiments, to both of which the same basic instrument calibration applies. This is small compared to the observed deviations[1] from the conventionally expected peak positions of more than 1 $Å^{-1}$.
4. Consequent on Point 3, if the position of the H recoil peak in *y* space is in agreement with conventional theory as Mayers and Reiter claim it is, then D is anomalous, thus contradicting numerous literature reports[8,9,11] that it obeys conventional theory well as to the position of its recoil peak and thereby refuting those reports.
5. To the contrary, we found using independently measured instrument flight-path parameters that the position of the D recoil peak does agree with conventional theory and existing literature reports[8,9,11] in the parameter space spanned by our reported experiment.[1]
6. The agreement between the D results and conventional theory necessarily means that H is anomalous: the H recoil peak is shifted to positive *y* values at high momentum transfer,[1] which is agreed by Mayers and Reiter to be impossible within any conventional theory.
7. Consequent on Point 6, a different theoretical frame is required to account for this reality. We maintain that the theory of open quantum systems[13,14] is applicable.[1]
8. The origin of the disagreement over the position in *y* space of the H and D recoil peaks is the ad-hoc adjustment of the Vesuvio calibration parameters to ensure that the position of the H recoil peak agrees with conventional theory.[10]

Given that the standard calibration procedure applied on Vesuvio assumes that the H recoil peak is in the expected place, Vesuvio cannot credibly be used for research that depends in detail on recoil peak positions and peak shapes unless an independent calibration is performed, which we regard as urgently necessary.

This outcome also raises the question of how many of the numerous publications based on measurements on H and D performed with Vesuvio or its eVS ancestor rely on wrong absolute H and/or D peak positions.

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