

Comment on “High-energy neutron scattering from hydrogen using a direct geometry spectrometer”

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The paper by Stock *et al.* [*Phys. Rev. B* **81**, 024303 (2010)] reports measurements of neutron scattering from hydrogen in the 1–100 eV range of energy transfers, using the direct geometry MARI spectrometer at ISIS. Stock *et al.* claim that their measurements have comparable energy resolution to those on the inverse geometry VESUVIO spectrometer at ISIS. Most importantly, the main conclusions of Stock *et al.* with regard to VESUVIO are not valid unless this claim is true. Here we present overwhelming evidence that the energy resolution of the measurements in *Phys. Rev. B* **81**, 024303 (2010) is one to two orders of magnitude worse than on VESUVIO. It follows that the conclusion of Stock *et al.* that anomalous neutron cross sections measured on VESUVIO [Chatzidimitriou-Dreismann *et al.*, *Phys. Rev. Lett.* **79**, 2839 (1997)] are “the result of experimental issues using indirect geometry spectrometers” is unfounded.

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I. INTRODUCTION

There have been many previous neutron measurements on hydrogen at eV energy transfers using inverse geometry methods,¹ but the measurements reported in Ref. 2 are the first using direct geometry. Such measurements are a welcome development. Unfortunately, Ref. 2 may mislead readers who are less familiar with the technical details of neutron spectroscopy. Stock *et al.* claim that their hydrogen measurements on the MARI direct geometry spectrometer at ISIS have comparable energy resolution to measurements on the VESUVIO inverse geometry spectrometer at ISIS.

In reality, the energy resolution of VESUVIO is between one and three orders of magnitude better than that of the measurements in Ref. 2. This implies that the measurements in Ref. 2 provide no basis for the claim of Stock *et al.* that anomalous neutron cross sections measured on VESUVIO³ are “the result of experimental issues using indirect geometry spectrometers.”

II. COMPARISON OF INSTRUMENTAL RESOLUTION AT MARI AND VESUVIO

A. Heavy atoms

Stock *et al.* accept that the “intrinsic experimental resolution” of VESUVIO is better than that of MARI for scattering from heavy atoms such as lead or vanadium: they state “Such measurements find the energy widths to be narrower on Vesuvio in comparison to MARI.” It is worthwhile to investigate how superior the VESUVIO resolution for lead is as compared to MARI. Figure 1 shows lead data measured on VESUVIO. The spectra were obtained by converting time-of-flight spectra at constant angle to energy transfer, using the known final energy and standard methods.⁴ This should be compared with the lead data displayed in Fig. 7 of Ref. 2. The full width at half maximum (FWHM) of lead peaks on

VESUVIO is ~ 0.25 eV compared with between 4 and 55 eV on MARI.

Figure 2 shows Gaussians with the same FWHM as the lead peaks measured on VESUVIO and the peaks displayed in Fig. 7 of Ref. 2. After taking into account the slight broadening due to the momentum distribution of the lead atoms, the energy resolution function on VESUVIO has a FWHM of ~ 0.2 eV (Ref. 5) compared with between 4 and 55 eV on MARI. The VESUVIO energy resolution close to the elastic line is thus between 20 and 300 times better than that on MARI.

This has important implications for the stated aim of Stock *et al.* to “investigate whether neutrons can be used to study high-energy magnetic and electronic excitations at energy transfers greater than ~ 1 eV.” A significant cross section for such studies can be obtained only at low wave-vector transfer Q . This requires scattering angles $< 10^\circ$, incident energies ~ 100 eV, and close to elastic scattering.⁶ It has been shown⁷ that inverse geometry instruments give an energy resolution of ~ 0.5 eV for incident energies ~ 100 eV in the latter regime. Thus direct geometry spectrometers currently have two orders of magnitude worse resolution for such measurements.

B. Hydrogen

Despite the much better resolution of VESUVIO for measurements of heavy atoms, Stock *et al.* argue that, specifically for scattering from hydrogen, the energy resolution of VESUVIO is either worse or at best, comparable to that of MARI. At a number of points in their paper they stress that the widths of hydrogen peaks at constant angle on VESUVIO are broader than on MARI and infer that the inelastic energy resolution of MARI at eV energies is therefore “better or comparable” to that of VESUVIO. In fact the instrument resolution has virtually no effect on hydrogen peak widths measured on VESUVIO. These are almost entirely determined by the way the scan in Q (wave-vector transfer) and E (energy transfer) crosses the dynamic structure factor

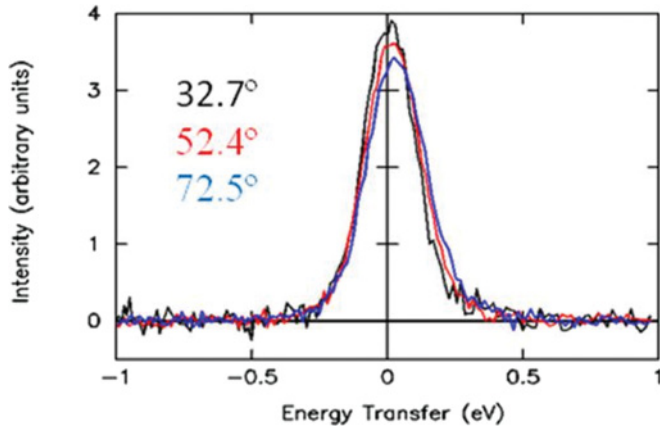


FIG. 1. (Color online) Data from 2 mm of lead on VESUVIO. The slight shift of the peak with angle is due to the increasing recoil of the lead atoms as the scattering angle increases.

$S(Q, E)$. Contrary to the inferences in Ref. 2, measurements on polythene also demonstrate quite clearly that VESUVIO has much better resolution than MARI.

The most basic method of analyzing hydrogen data at eV energy transfers is to assume that the dynamic structure factor $S(Q, E)$ has the impulse approximation (IA) form

$$S(Q, E) = \frac{M}{\hbar^2 Q \sigma} \frac{1}{\sqrt{2\pi}} \exp\left[-\frac{y^2}{2\sigma^2}\right], \quad (1)$$

where

$$y = \frac{M}{\hbar^2 Q} \left(E - \frac{\hbar^2 Q^2}{2M} \right) \quad (2)$$

and M is the mass of the target atom. Equations (1) and (2) are exactly correct in the limit $Q \rightarrow \infty$ and when the binding is by isotropic harmonic forces.⁸ Equations (1) and (2) are equivalent to Eq. (6) of Ref. 2, which Stock *et al.* fitted directly to their data. The full widths at half maximum values 2Γ discussed in Ref. 2 are therefore related to σ via

$$2\Gamma = 2\sqrt{2 \ln 2} \frac{\hbar^2 \sigma Q}{M} = 0.00977 Q \sigma, \quad (3)$$

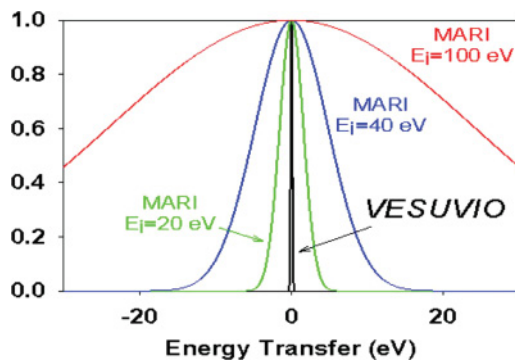


FIG. 2. (Color online) Gaussians with the same FWHM in energy transfer as the lead peaks measured on VESUVIO and MARI. The widths (FWHM) of the MARI data were taken from Fig. 7 of Ref. 2. To a good approximation these curves show the energy resolution functions of the two instruments close to zero energy transfer.

where the second equality applies if 2Γ is in eV and Q and σ are in \AA^{-1} .

Assuming that the IA is valid, if the instrument resolution was perfect, the value of σ obtained by fitting Eqs. (1) and (2) to data would be W_H , where $\hbar W_H$ is the rms momentum of protons in the sample. In reality the fitted value of σ is always greater than W_H due to instrument resolution broadening. To a good approximation the value of σ obtained from fitting is increased to

$$\sigma = \sqrt{W_H^2 + W_R^2}. \quad (4)$$

Equation (4) defines W_R , the instrument resolution width in the hydrogen momentum (y) space. The VESUVIO resolution width in E at constant Q is approximately proportional to W_R .

Figure 3 shows a typical fit of Eqs. (1) and (2) to the carbon and hydrogen peaks in VESUVIO polythene data. The data, shown as the red points with error bars due to counting statistics, is uncorrected for multiple scattering and background. Note that contrary to the claim of Stock *et al.* that “it is impossible to subtract the short time background on inverse geometry machines” (Sec. VI, Ref. 2), Fig. 3 demonstrates that the background at short times on VESUVIO is very low. The background subtraction is automatically performed by the foil cycling method, used to define the final neutron energy.⁹

The values of σ obtained at different angles by fitting Eqs. (1) and (2) directly to the VESUVIO polythene data, with no correction for the instrument resolution, are shown as the red circles in Fig. 4. The mean value over all detectors of these points is 4.84 \AA^{-1} with a standard error in the mean of 0.01. After correction for the VESUVIO resolution using the methods described in Refs. 10–12, values of W_H shown as the black points were obtained. In the latter case the mean over all detectors is $W_H = 4.77 \text{ \AA}^{-1}$, again with a standard error of 0.01. It follows from these values and Eq. (4) that the average resolution width over all detectors is $W_R \sim 0.7 \text{ \AA}^{-1}$. This is $\sim 15\%$ of the width W_H .

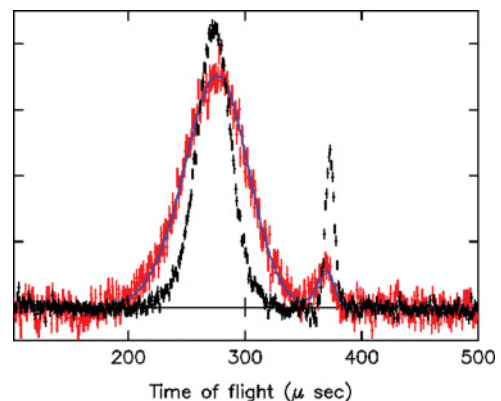


FIG. 3. (Color online) Uncorrected time-of-flight data from CH_2 and liquid H_2 on VESUVIO at a scattering angle of 45.9° . The red points are data from CH_2 . The blue line is the fit of Eqs. (1) and (2). The black points were taken from a sample of liquid hydrogen contained in an aluminum can. The intensity scale of the y axis is determined by the normalization region chosen in the incident beam monitor and is thus arbitrary. However, it is the same for the H_2 and CH_2 data.

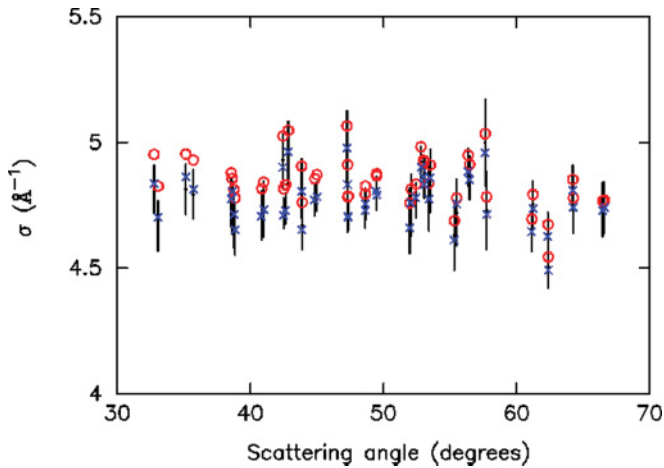


FIG. 4. (Color online) Values of σ obtained by fitting VESUVIO data directly to Eqs. (1) and (2) with no correction for the instrument resolution are shown as the red circles. The values of the rms proton momentum W_H obtained from the standard instrument program are shown as the black points. The instrument programs incorporate the VESUVIO resolution and a peak shape correction for deviations from the IA due to the finite Q . The blue crosses were obtained with a resolution correction, but no correction for deviations from the IA. The Q ranges from $\sim 31 \text{ \AA}^{-1}$ at the lowest scattering angle of 33° , to $\sim 115 \text{ \AA}^{-1}$ at the highest scattering angle of 67° . The error bars due to counting statistics obtained from the instrument programs are shown as vertical black lines. The error bars for the black, red, and blue points are almost identical.

The data displayed in Figs. 3 and 4 demonstrates that the impulse approximation is very well satisfied at the (Q, E) values attained on VESUVIO. Very good fits are obtained and values of σ and W_H obtained from the fitting are independent of angle to within the errors of $\sim 4\%$ due to counting statistics (see Fig. 4). This must be so if the IA is valid and the resolution function does not significantly broaden the hydrogen peak width, since σ is then physically the rms momentum of hydrogen atoms in the sample. The VESUVIO instrument programs also incorporate a correction for the small deviations from the IA ($\sim 1\% - 5\%$) due to the finite Q of measurement, using the formalism developed by Sears.¹³ However, this makes very little difference to the values of W_H obtained from the fitting as can be seen by comparing the blue crosses (no correction) and black points (corrected) in Fig. 4. This is again strong evidence the IA is well satisfied for hydrogen measurements on VESUVIO.

On both inverse and direct geometry time-of-flight neutron instruments, the only really accurate way to obtain a constant Q scan is to create a map in (Q, E) space using time-of-flight data taken from detectors at different angles. This is not necessary for the measurement of proton momentum distributions. It is only possible in practice on VESUVIO over a very limited region of (Q, E) space, due to the limited angular coverage of the detectors on the instrument. However, a calculation is possible and Fig. 5 shows the hydrogen peaks from polythene at constant Q which would be obtained on VESUVIO, with the current instrument resolution and an unbroken range of angular coverage from 30° to 70° at steps of 0.1° .

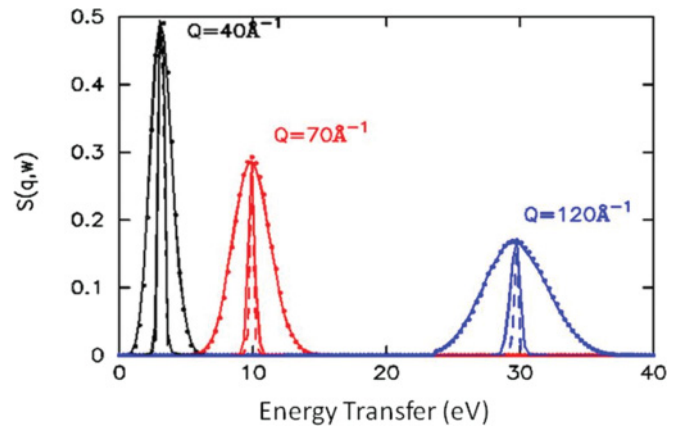


FIG. 5. (Color online) Shows a calculation of data at constant Q on VESUVIO generated as described in the text. The broad peaks were calculated assuming $W_H = 4.77 \text{ \AA}^{-1}$, the mean value obtained from fitting VESUVIO data. The solid lines are simulations with the current VESUVIO resolution included. The points are the peaks that would be obtained with perfect resolution. The slight asymmetry of the peaks is due to the incorporation of the Sears correction (Ref. 12) for deviations from the IA at finite Q in the calculation. The narrow peaks were calculated for $W_H = 0$ and define the instrument resolution at constant Q . In the latter case the solid lines were calculated with all resolution components included and the dashed lines with only the uncertainty in final energy included.

The VESUVIO resolution function and the Sears¹³ corrections to the IA were incorporated in the calculation of the time-of-flight data $C(t)$ as described in Ref. 11. The resolution was calculated from the measured instrument uncertainties derived from the calibrations described in Ref. 5. Given the calculated $C(t)$, Q and E were determined for each time-of-flight bin t . The corresponding value of $S(Q, E)$ was obtained from the standard expressions for $C(t)$ on an inverse geometry instrument⁴ and binned in a (Q, E) matrix.

Figure 5 shows sections of this matrix at different Q values. The broad peaks were calculated with the mean value $W_H = 4.77 \text{ \AA}^{-1}$ obtained from the VESUVIO measurements shown in Fig. 4. The full lines show the calculation with the VESUVIO resolution included. The points were calculated assuming perfect resolution. The narrow peaks were calculated for $W_H \rightarrow 0$, when Eqs. (1) and (2) imply that $S(Q, E)$ is a delta function in E at constant Q . It is conventional to take the instrument resolution as the measured peak width for a delta function $S(Q, E)$. Following this convention the narrow peaks show the VESUVIO resolution function for scattering from hydrogen at constant Q . The full line was calculated with all instrument resolution components included, the dashed line with only the uncertainty in the final energy taken into account. At higher Q values, the resolution for hydrogen scattering on VESUVIO is in fact dominated by the angular resolution.⁵

The values of 2Γ obtained by inserting the mean fitted VESUVIO value of $\sigma = 4.84 \text{ \AA}^{-1}$ into Eq. (3) are shown as a function of Q as the red line in Fig. 6. The FWHM's of the simulated polythene data in Fig. 5 are indistinguishable from this line on the scale of Fig. 6. The values of 2Γ given in Fig. 13 of Ref. 2 are also shown. It is obvious that in almost

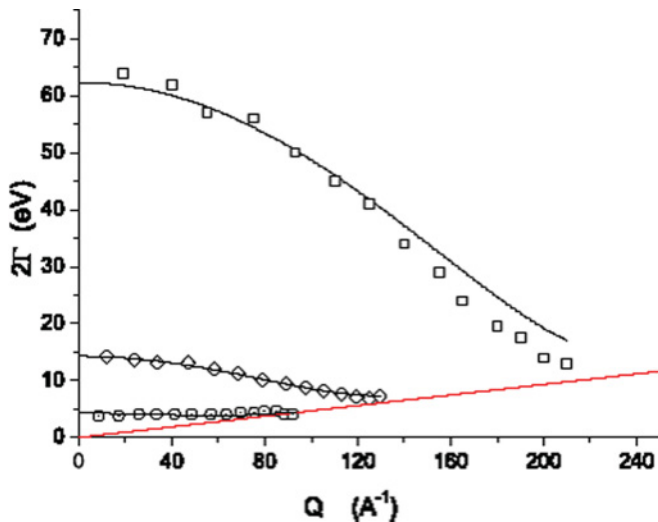


FIG. 6. (Color online) The red line shows the FWHM 2Γ of $S(Q, E)$ calculated from Eq. (3) and the mean value $\sigma = 4.84 \text{ \AA}^{-1}$ obtained from the VESUVIO data shown in Fig. 4. Values of 2Γ obtained from Eq. (3) and individual VESUVIO data points in Fig. 4 are indistinguishable from the red line on the scale shown. The circles, diamonds, and squares show the data displayed in Fig. 13 of Ref. 2 at incident energies of 20, 40, and 100 eV, respectively. The solid black lines are the best fit to Eqs. (4) and (5) of the single parameter ΔE_0 .

all Q, E space the FWHM 2Γ of the H peak from polythene is much larger on MARI than on VESUVIO. This can only be due to the much coarser resolution of MARI. The ratio W_R/W_H is approximately equal to the ratio of the instrument energy resolution width and the intrinsic hydrogen peak width in E at constant Q . For most of the MARI points shown $W_R \gg W_H$, and it follows from Eqs. (3) and (4) that 2Γ is determined almost entirely by the MARI energy resolution. On VESUVIO it is always the case that $W_R \ll W_H$ and 2Γ is determined almost entirely by the sample response.

Figures 1–6 provide conclusive evidence that at eV energies, the energy resolution of VESUVIO is greatly superior to that of MARI for both hydrogen and lead. Nevertheless, Stock *et al.* still claim that their measurements of hydrogen in Ref. 2 have better or at worst, comparable energy resolution to those on VESUVIO. Stock *et al.* have misinterpreted in an elementary way the VESUVIO data shown in their Fig. 1. They correctly state, “the broad widths of the hydrogen recoil lines on VESUVIO are the result of the detector trajectories intersecting the recoil line more tangentially in the indirect geometry setup on VESUVIO than on direct geometry machines such as MARI.” It is hard to understand why they do not draw the unavoidable conclusion: hydrogen peak widths at fixed angle on MARI and on VESUVIO do not convey any direct information about the relative resolutions of the two instruments.

There seems to be a basic misunderstanding in Ref. 2. Stock *et al.* have assumed that the hydrogen peak width is determined entirely by the instrument resolution on both MARI and VESUVIO; that is, that $\sigma = W_R$. This is a reasonable approximation for all the MARI data in Ref. 2 except that at the highest Q values. It is completely wrong on VESUVIO, where $\sigma = W_H$ to within $\sim 2\%$ at all Q . Figure 3

also shows uncorrected time-of-flight data at constant angle on VESUVIO from liquid H_2 (black) in addition to the CH_2 data (red). The same detectors in identical positions were used for both measurements. The different widths (and line shape) of the hydrogen peaks centered at $\sim 280 \mu\text{sec}$ can only be due to the different sample responses. As is immediately obvious from the data, the momentum distribution of the protons in CH_2 has a FWHM about twice that in liquid H_2 . On MARI the energy resolution is so poor that virtually no difference in the hydrogen peak width from these samples would be observed.

III. FITTING THE HYDROGEN PEAK WIDTH AND MARI RESOLUTION

In our view the analysis of the MARI resolution in Appendix B of Ref. 2 is incorrect. Stock *et al.* argue that on MARI there are two components in the time pulse width; the width τ_0 produced by the Fermi chopper and a second component $\tilde{\tau}$. The physical content of τ_0 and $\tilde{\tau}$ is given unambiguously by Eqs. (B1) and (B3) of Ref. 2. According to these equations τ_0 is determined by the width in energy, ΔE_0 , of the incident pulse and $\tilde{\tau}$ by the width in energy, ΔE_1 , of the scattered pulse. It is evident that ΔE_1 cannot be statistically independent of ΔE_0 . Hence these two uncertainties cannot be added in quadrature as Stock *et al.* assume. For example, if Stock *et al.* were correct and $\tau_0, \tilde{\tau}$ were independent, then their analysis implies that installation of a perfect chopper on MARI ($\tau_0 = 0$), would still give very poor energy resolution.

The variation of 2Γ with Q in the measurements of Ref. 2 is almost certainly a straightforward consequence of the fact that the incident energy is very coarsely defined by the MARI Fermi chopper, due to its transparency at eV energies. Stock *et al.* state that “the neutrons are emitted in a short pulse about $0.5 \mu\text{sec}$ long that will be treated by our simplified model as instantaneous.” This assumption (with which we agree) implies that ΔE_0 is determined almost entirely by the characteristics of the MARI Fermi chopper. Under these circumstances the energy resolution of a direct geometry spectrometer is⁴

$$W_R = \left(1 + \frac{L_0 E_1^{3/2}}{L_1 E_0^{3/2}} \right) \Delta E_0, \quad (5)$$

where E_0 is the incident energy, E_1 is the final energy, L_0 is the incident flight path, L_1 is the final flight path, and ΔE_0 is the spread of neutron energies incident on the sample.

Using the standard IA result that $E_f/E_i = \cos^2 2\theta$ at the hydrogen peak center [see Eq. (1) of Ref. 2 and inserting the MARI values $L_i = 11.79 \text{ m}$ and $L_f = 4.02 \text{ m}$, Eqs. (3)–(5) can be fitted to the MARI data with the single fitting parameter ΔE_0 . The mean value of $W_H = 4.77 \text{ \AA}^{-1}$ obtained from the VESUVIO data in Fig. 4 was used to calculate σ from Eq. (4). The fits are shown as black lines in Fig. 6. The fitted values of ΔE_0 are listed in Table I for the three incident energies displayed in Fig. 6. Also listed are values of ΔE_0 obtained from the FWHM of the lead peaks shown in Fig. 7 of Ref. 2.

It can be seen from Fig. 6 that the 20 and 40 eV data is very well fitted by the single parameter ΔE_0 . For $E_0 = 100 \text{ eV}$ the fit is not so good. However, this is not surprising given the data analysis procedures in Ref. 2. Stock *et al.* write that the values

TABLE I. The second column gives $t \Delta E_0$, the range of incident neutron energies obtained by fitting hydrogen data as described in the text. The third column gives the FWHM's of the lead peaks shown in Fig. 7 of Ref. 2.

Incident energy (eV)	ΔE_0 (eV) from H	ΔE_0 (eV) from Pb
100	15.9 ± 0.4	14
40	3.63 ± 0.03	2.8
20	1.08 ± 0.04	0.9

of 2Γ in Fig. 13 of Ref. 2 were obtained by “fitting the constant 2θ scans to the sum of two Gaussians to represent the recoil lines from hydrogen and carbon.” It is clear from Fig. 11 of Ref. 2 that the hydrogen peak shape in constant 2θ scans becomes progressively less Gaussian as E_0 increases. It seems very different from a Gaussian for $E_0 = 100$ eV. The same is true for the lead peak shape in Fig. 7 of Ref. 2. This change in peak shape is probably due to the increasing transparency of the MARI Fermi chopper as the incident energy is increased and implies that their analysis cannot give accurate values for 2Γ at $E_0 = 100$ eV.

It can also be seen from Table I that the values of ΔE_0 obtained from the lead and hydrogen MARI data are in quite good agreement. In fact, the Q dependence of 2Γ in the MARI measurements can be explained rather well by Eqs. (3)–(5), with ΔE_0 taken from the MARI lead data. This textbook calculation gives a better overall description of the MARI hydrogen data than the model of Ref. 2, Appendix B, which includes two free fitting parameters, but cannot explain the 20 eV MARI data at all. A more accurate evaluation of the CH_2 data in Ref. 2 could be made by fitting the lead data to determine the shape of the MARI energy resolution function in a similar way to that described in Ref. 5.

IV. SUMMARY AND DISCUSSION

The scientific motivation of Ref. 2 was the ongoing debate about the cross-section anomalies observed on VESUVIO^{3,11} and by two separate groups using electron scattering.^{14,15} The results in Ref. 2 are consistent with the measurements of Moreh *et al.*,^{16,17} that with very coarse energy resolution no anomalies are observed. However, as previously pointed out,¹⁸ existing theories of anomalous cross sections predict^{19–21} that there will be no anomalies with very coarse resolution. Hence the measurements of Ref. 2 provide no basis for the conclusion of Stock *et al.* that anomalous neutron cross sections measured on VESUVIO^{3,11} are “the result of experimental issues using indirect geometry spectrometers.”

The measurements of Stock *et al.* do, however, demonstrate that at eV energies, direct geometry chopper spectrometers are currently a long way from being competitive with inverse geometry spectrometers, based on resonance foil methods. Chopper spectrometers have between one and two orders of magnitude worse resolution for the study of momentum distributions and two orders of magnitude worse resolution for the study of magnetic and electronic excitations at eV energies.

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