The VESUVIO Spectrometer Now and When?

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Abstract. The current layout and mechanics of the VESUVIO spectrometer are presented in light of spectroscopic measurements using electron-volt neutrons. A brief background to the theoretical framework of deep inelastic neutron scattering is presented, with focus on data collection and instrumental design. The current capabilities and research themes for VESUVIO are discussed, and possible future instrumental developments highlighted which will enhance the instrument's ability to meet scientific inquiry and expectation.

1. Introduction

The VESUVIO spectrometer at the ISIS Pulsed Neutron and Muon Source (Rutherford Appleton Laboratory, UK) is an instrument that probes a unique kinematic region in neutron scattering [1]. Despite there now being a number of spallation neutron sources across the globe, VESUVIO has remained the only spectrometer routinely serving a user community for measurements of scattering processes utilising high-energy neutrons in the epithermal region (5-150 eV), termed Deep Inelastic Neutron Scattering (DINS). In recent years the Bariloche Electron LINAC (Argentina) has witnessed the implementation of a spectrometer for DINS measurements. Whilst VESUVIO continues as both forerunner and standard for the measurement of atomic momentum distributions in condensed matter research, and remains in active development, it is hoped that further development in this field will see the advent of additional instruments.

In this article, the current layout and brief theoretical background for VESUVIO are presented, alongside the mechanics of a VESUVIO measurement as would be suitable for a prospective user of

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the instrument. In this manner, we hope to give a simple overview of the current state and operation of the instrument, highlighting both themes and capabilities in the field of electron-volt neutron scattering. Following this, there will be presented a future outlook for DINS and a discussion of how VESUVIO could be developed further to both meet the evolving-requirements of its user community, and to impact new fields of research.

2. Neutron Compton Scattering and a VESUVIO measurement

1.1. The Current Layout of VESUVIO

VESUVIO, as with the majority of spectrometers on a pulsed neutron source, is a time-of-flight (ToF) spectrometer, with ToF value, *t*, for each detected neutron given by [1,2]

$$t = \frac{L_0}{v_0} + \frac{L_1}{v_1} \tag{2.1}$$

Where L_0 and L_1 define the distance of sample from source and detector respectively, v_0, is the incident neutron velocity and v_1 the velocity of detected neutron. The neutron velocity can be related to its energy, E, through the relation $v = \sqrt{2E/m}$ where m is the neutron mass. As such, and through knowledge of t, L_0, L_1 and either E_0 or E_1, the energy transfer, $\hbar\omega$, and momentum transfer, q, can be determined [2]

$$\hbar\omega = E_0 - E_1 \tag{2.2}$$

$$q = \sqrt{2m}\left(E_0 + E_1 - 2\sqrt{E_0 E_1 \cos\theta}\right)^{1/2} \tag{2.3}$$

VESUVIO operates in inverse geometry, and as such E_1 is fixed at 4897meV.

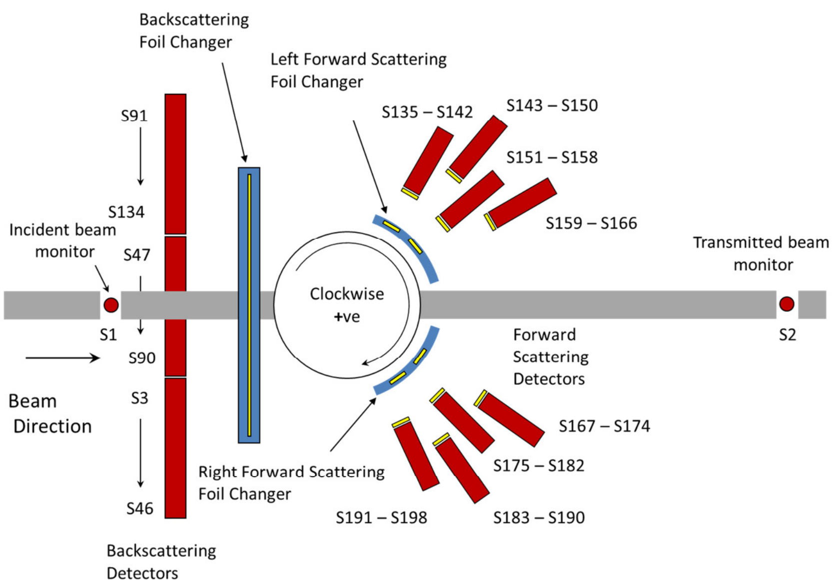

Figure 1. Schematic layout of the VESUVIO spectrometer

The layout of VESUVIO is given in figure 1. The instrument comprises 196 detectors in addition to an incident (S1) and transmitted (S2) beam monitor. It can be seen that two distinct geometries for detector banks exist, those in forward scattering (S135-S198) and backscattering (S3-S134). In backscattering the detectors are laid out in three banks perpendicular to the beam direction. These Li-doped glass scintillator detectors cover an angular range of 130°-163°, with each detector of approximate area 20 cm^2.

E_1 is defined and fixed on VESUVIO through the incorporation of gold analyser foils. In backscattering this is done by taking measurements with and without foils between detector and sample [3]. Gold exhibits a strong nuclear resonance, absorbing neutrons at an energy value calibrated as 4897meV. Thus a difference measurement will yield ToF spectra corresponding to scattering processes resulting in this fixed E_1. By taking measurements of different gold foil thicknesses, the energy resolution of VESUVIO is improved. This 'Double Difference' (DD) foil cycling technique is detailed elsewhere [1,3-4]. It should be noted that running VESUVIO in 'foil-out' mode, *i.e.* without analyser foils present, allows for the instrument to be utilised as a diffractometer. Foil-cycling the 132 ^{6}Li-doped scintillator detectors in backscattering geometry on VESUVIO thus allows for simultaneous diffraction measurements in the thermal neutron region during a standard DINS measurement.

The detector banks at forward scattering on VESUVIO were upgraded in 2008, now consisting of γ-detecting Yttrium Aluminium Perovskite (YAP) detectors [5]. The gold resonance at 4897 meV is still used to define E_1, but in this case it is the ejected γ-ray that is detected. The DD technique is instigated through the use of the secondary gold foils detailed in figure 1. The cycling of these secondary analyser foils produces a sample-independent γ-background of ~5% in forward scattering [6]. This unwanted contribution to the signal is subtracted from spectra as part of the data analysis process.

1.2. Atomic Momentum Distributions and the Impulse Approximation

The aim of any measurement on VESUVIO is to probe the atomic dynamics of a system [7], through determination of atomic momentum distributions or kinetic energies. The reduction and analysis of raw data is viewed by the user in time of flight, *C(t),* or in a given, mass-specific response function in *y*-space $J_M(y)$ (rather than the scattering law $S(\boldsymbol{q},\omega)$ more common for other neutron spectrometers), and so in this section we shall briefly outline the mechanics of VESUVIO data handling within the framework of DINS.

At the high energy transfers available on VESUVIO, the scattering event is described within the Impulse Approximation (IA), whereby conservation of momentum and kinetic energy occurs between the incident neutron and single target nucleus. As such, the scattering law may be written as [7-9]

$$S(q,\omega) = \int n_M(\boldsymbol{p})\delta\left(\omega + \frac{p^2}{2M} - \frac{(\boldsymbol{p}+\boldsymbol{q})^2}{2M}\right)d\boldsymbol{p} = \frac{M}{q}J_M(y_M,\hat{\boldsymbol{q}}) \tag{2.4}$$

where

$$J_M(y_M,\hat{\boldsymbol{q}}) = \int n_M(\boldsymbol{p})\,\delta(y_M - \boldsymbol{p}\cdot\hat{\boldsymbol{q}})d\boldsymbol{p} \tag{2.5}$$

and

$$y_M = \frac{M}{q}\left(\omega - \frac{q^2}{2M}\right) \tag{2.6}$$

where $n_M(\boldsymbol{p})$ is the radial nuclear momentum distribution for species of atomic mass M, and $J(y,\boldsymbol{q})$ thus being the distribution of momentum states along the direction $\hat{\boldsymbol{q}}$ $(= \boldsymbol{q}/q)$, the Compton profile. It is immediately apparent that within the IA there is a mass dependence, with the transform to y-space centring the contribution from each atomic species at y=0, dependent on the recoil energy for a given mass M, $q^2/2M$. This is expressed in the count rate for VESUVIO spectra, given by

$$C(t) = \frac{E_0 I(E_0)}{q} \sum_M A_M M J_M(y_M) \otimes R_M \tag{2.7}$$

where $I(E_0)$ is the incident flux of neutrons with energy E_0. The mass-dependent amplitude factor, A, is proportional to the number density (N_M) and scattering cross-section (σ^T) of the atomic species, and R is the mass-dependent instrument resolution function.

A DINS measurement on VESUVIO (for a given detector geometry) thus consists of a set of peaks in ToF, centred at values relating to each atomic species' recoil energy, itself dependent on the atomic mass. In practise a majority of experiments on VESUVIO are on isotropic samples, whereby the $\hat{\boldsymbol{q}}$ dependence of $J(y)$ is lost. The most common model for $J(y)$ is a Gaussian form in y, exact for an isotropic, harmonic potential. In this case the atomic kinetic energy $\langle E_K \rangle$ relates to the standard deviation of the Gaussian, σ, through

$$J_M(y_M) = \frac{1}{\sqrt{2\pi\sigma_M^2}} \exp(-\, y_M^2/2\sigma_M^2) \tag{2.8}$$

$$\langle E_K \rangle = \frac{3\hbar\sigma_M^2}{2M} \tag{2.9}$$

This simple expression represents the elegance of DINS. For a given system, each mass is expressed as an individual peak in ToF, the widths of which (in actuality the standard deviation of Gaussian peak in the y-space for each mass) are a direct measurement of their mean kinetic energy and thus confining potential. Further models for $J(y)$ that move beyond the isotropic harmonic-oscillator model include expansion of equation (2.8) to include final-state effects [7,9], higher-order Hermite polynomials [7,10] (the so-called Gram-Charlier expansion), and the use of a multivariate Gaussian distribution [11].

3. Current Scientific Directions and Future Developments for VESUVIO

1.3. The Problem of Mass

It has been stated that in a DINS measurement each mass is exhibited as a peak centred at its recoil energy (in ToF this is dependent on instrument geometry), with intensities related directly to number density and scattering cross-section. As mass is increased, the separation between neighbouring elements becomes progressively shorter, as can be seen from equations (2.4-2.7). It is therefore a simple matter to separate and compare the lightest masses, such as hydrogen, deuterium (D) or lithium (Li), but for masses such as carbon (C) and nitrogen (N) there is swift convergence. Figure 2 demonstrates this convergence, and highlights the difference in convergence rates in forward and backscattering geometries.

As such, for measurements whereby masses >5 amu are of interest, and particularly for systems containing multiple masses which are desired to examine simultaneously, it is the backscattering banks that are of interest. By verity of being (nearly) isobaric with the neutron, hydrogen will not backscatter

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in the epithermal region, and thus a peak arising from neutron scattering (strictly we confine this to a single scattering event) from a proton will only appear in forward scattering spectra. We can thus divide discussion of the scientific capabilities (or directions) of VESUVIO into two thematic areas, those involving the proton and deuteron, and those of simultaneous measurement of heavier masses.

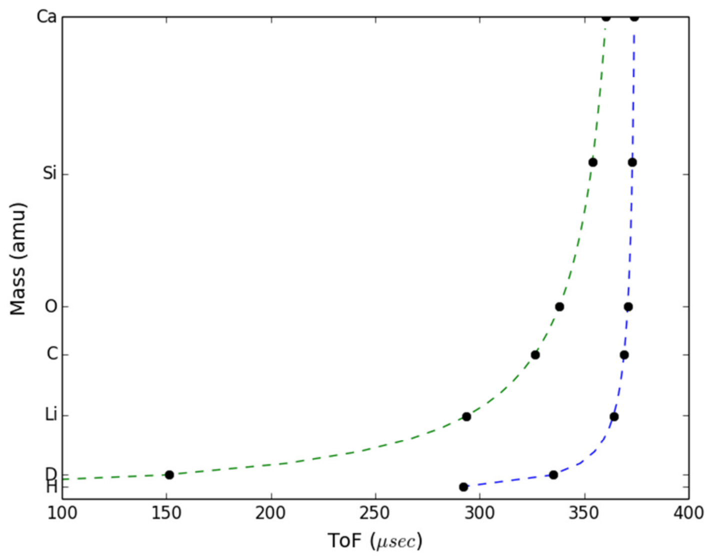

Figure 2. Recoil peak positions for atomic masses 1.0079-40.0 amu. Detector geometries simulated in forward (blue) and backscattering (green) for θ=40.0° and 140.0° respectively, with L_1=0.52m. These are close to the values for detectors S69 and S156.

1.4. Nuclear Quantum Effects

The last decade has seen a dramatic change in the science conducted on VESUVIO. An examination of the number of days requested on the instrument over recent cycles (figure 3) shows that the proton is the focus of a majority of research on VESUVIO. Increasingly more complex systems are being measured, whereby not only proton dynamics are important, but also nuclear quantum effects associated with the exchange of the proton (H) by the deuteron (D). Importantly, there is increasing interest to measure systems involving both H and D.

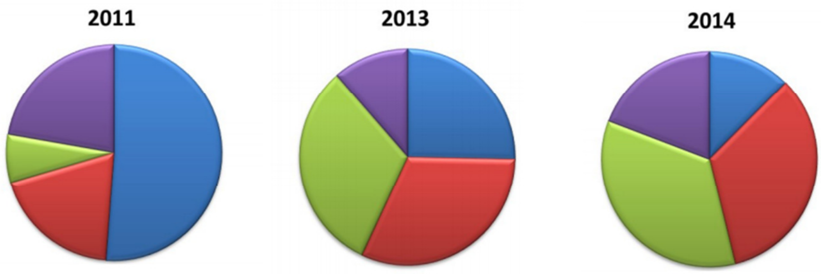

Figure 3. Recent requested time on VESUVIO divided into research themes. *Red*: H_2O, *Blue*: investigations into proton dynamics excluding H_2O; *Purple*: deuterium dynamics; *Green*: M>5 amu.

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The large total scattering cross section of the proton combined with its light mass results in the proton peak being of particular high intensity and isolated from others in DINS spectra. As such VESUVIO has proven particularly adept in examining proton kinetic energies and momentum distributions in a variety of condensed matter systems [7], including diatomic H_2 [12,13], energy materials [14-16] and H_2O both in bulk states and confined within host materials [17-19].

Recent investigations into nuclear quantum effects, specifically those involving H/D comparison have all involved the measurement of H/D species separately [20,21]. Whilst not an outlandish approach by any means, this is primarily limited to systems with single proton environments. If VESUVIO and by extension DINS are to contribute to systems of partial deuteration, a greater capability in the measurement of both H and D simultaneously must be achieved. In principle, this is something relatively simple on VESUVIO, with the mass-specific nature of the scattering response yielding signals from H and D separately in ToF. However, the sparseness of VESUVIO data (by verity of the low detected flux on the instrument) combined with the often far larger H>D signal can make reliable measurement of D kinetic energy difficult. This is exemplified in the simulated spectrum for monodeuterated/monohydrogenated phenol, shown in figure 4.

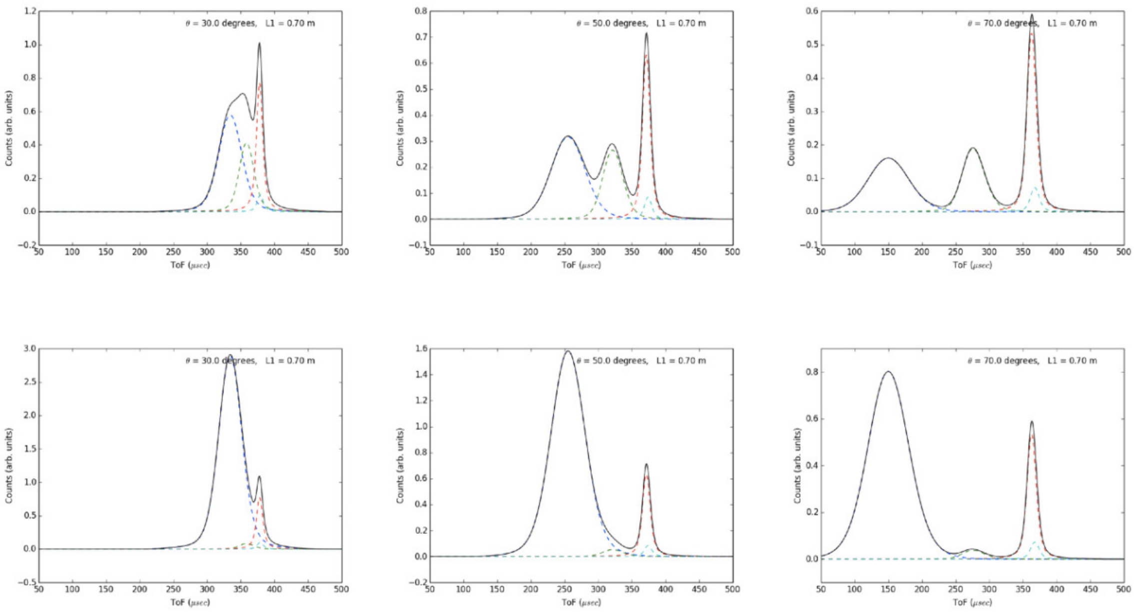

Figure 4. Simulated DINS spectra for isotopically labelled phenol for L_1=0.7 m and θ=30° (*left*), 50° (*centre*) and 70° (*right*). *Top row*: C_6D_5OH. *Bottom row*: C_6H_5OD.

VESUVIO is currently flux-limited, particularly for the traditional, detector-by-detector approach to data reduction and analysis. An average measurement of the proton momentum distribution of a single sample at one temperature takes of the order of 18 hours. After this time the signal-to-noise levels for single-detector spectra is suitable for analysis of peak width and shape. Examining the simulated (and noiseless) spectra for C_6H_5OD it is clear that simultaneous fitting of the D peak is only possible for higher angles, $\theta \gtrsim 50°$.

In principle, measurement of D-profiles is possible in backscattering geometry, where the proton recoil signal is excluded from the kinematics of proton-neutron collisions using epithermal neutrons. It thus seems natural that for samples dominated by proton scattering the backscattering banks could be utilized in the isolation of D from H. Unfortunately, the current geometry of the VESUVIO banks yields a D-signal at ToF values <150μs, a region suffering from low signal-to-noise for the ^{6}Li-doped scintillator detectors in backscattering. Were the D-peak brought up to higher ToF values by shifting to lower angles in backscattering, routine measurements of D on these banks would be feasible. As such, we can conclude that VESUVIO is not currently exploiting two important angular ranges for simultaneous measurement of H/D – higher angles in forward scattering and lower angles in backscattering. These regions are shown in figure 5. An alternative to this would be to extend L_0 by physically moving the instrument further from the neutron source. This is currently not feasible at the operational levels of ISIS due to the concomitant loss of flux this would entail.

1.5. Heavier Masses and MANSE

Perhaps the most striking change in the scientific direction of VESUVIO shown in figure 3 is that ‘heavy-mass’ measurement, by which we refer to masses >5 amu, those of boron and above. Focus has been brought to the theoretical framework of DINS as a measure of atomic dynamics as separable by mass, now termed MAss-selective Neutron SpEctroscopy, or MANSE in the literature [22,23]. The advent of MANSE has been recent enough that the capabilities of VESUVIO in this field are yet to be explored to its full extent, with fresh approaches not only to sample chemistry/stoichiometry, but also data analysis and sample environment. Here we shall briefly comment on these, focusing on the manner in which VESUVIO could be developed in the future to accommodate MANSE in its scientific repertoire.

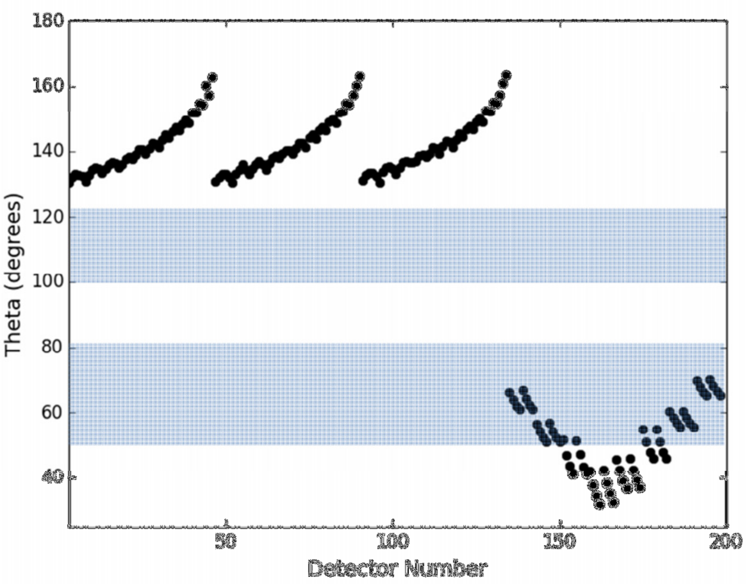

Figure 5. Angular coverage of detectors on VESUVIO in backscattering (3-134) and forward scattering (135-198). Shaded regions represent the angular coverage that could be exploited to further optimise VESUVIO for deuterium studies.

Examining again figure 2 we can see that for elements in the first two periods of the periodic table, H-Ne, their recoil positions are not yet converging in backscattering to an extent that is comparable to forward scattering. Of course this simplistic approach does not yet incorporate peak intensities or widths in ToF, but it is a suitable first estimation for the capability of VESUVIO to accurately and simultaneously measure the atomic kinetic energies of these elements in a given system. Within this range exist elements particularly important for modern materials and chemical research, for example lithium (battery and energy storage materials), boron (superhard materials), carbon and nitrogen (organic molecules and layered materials), oxygen (oxide devices) and fluorine (superacids and ion conductors). These serve only as examples, but highlight the potential breadth of application for DINS and VESUVIO.

As with forward scattering, the problem of flux and signal-to-noise is encountered in MANSE. Noiseless spectra for elements from Li to F are simulated in figure 6, with arbitrary values for profile width, σ_M. Now it can be seen that despite peak centroid separation in ToF, appreciable overlap in signal will result in ambiguity in fit parameters if data is sparse, as is the case for the low detected flux on VESUVIO. In addition to this, the sample environment itself contributes strongly to measurements of heavier masses. It is common to run samples within Al sample containers for neutron spectroscopic measurements, due to its low total scattering cross-section, 1.5 barn. This is equivalent to many elements of interest in DINS, and is indeed higher than some (σ^T_{Li}=1.37, σ^T_O=4.232 barn), and thus for smaller amounts of sample this contribution can dominate. This is demonstrated in figure 6, with measured spectra for a 1-mm-thick Li_2O sample run in a standard VESUVIO Al sample container compared to that simulated for a Sn container. Clearly the contribution from sample container can be reduced not only in absolute scattering power (by moving to thinner sample containers) but also by utilizing materials heavier than Al. For this reason, candidates include Sn-foils for standard and low-temperature measurements, and Nb containers for higher temperatures.

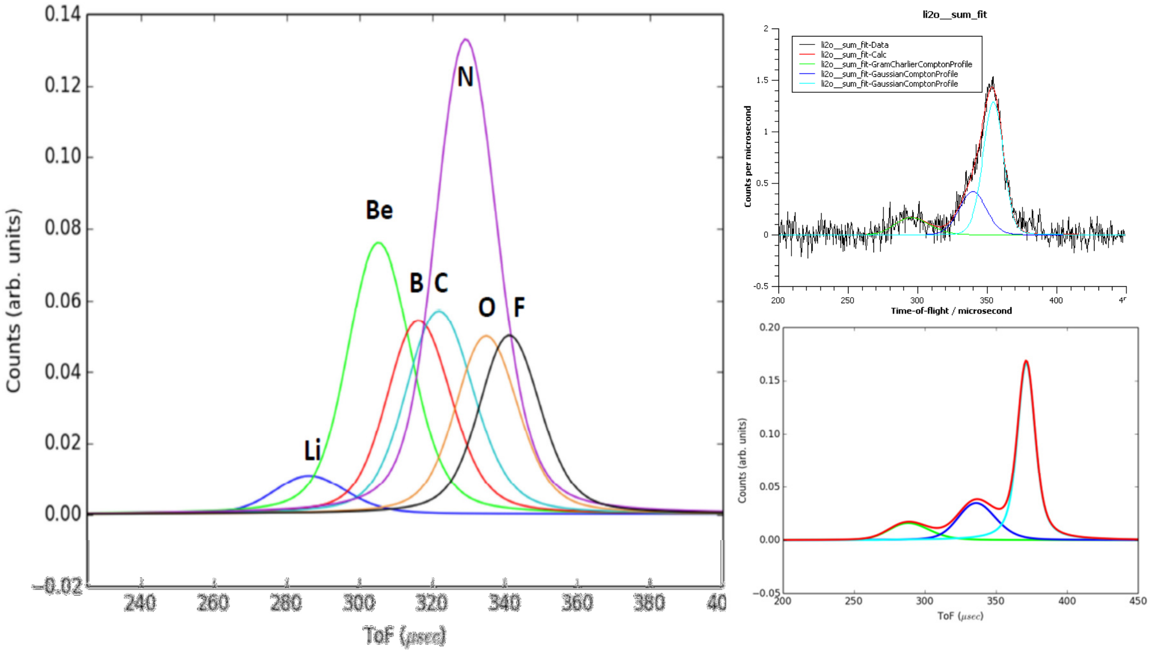

Figure 6. *Left:* Simulated DINS spectra for elements Li to F, L_1=0.55 m θ=140°. *Right Top*: Measured Li_2O spectra in standard Al sample container. *Right Bottom*: Simulated Li_2O in Sn container of equal scattering power.

Traditional analysis of DINS spectra on VESUVIO have been performed using a detector-by-detector methodology [1,24]. In this manner, the $J(y)$ for each mass is constructed in their individual y-spaces, with the composite function fitted to the ToF spectra for each detector, utilising the individual geometries for each. In this manner it follows that from each detector an independent σ_M is fitted. This historical perspective arises from the first measurements of σ_M for light elements, testing the impulse approximation and invariance of σ with detector angle through conversion to y-space.

But is this necessary? For the sparse data on VESUVIO, any means that can increase the statistics of the ToF signal for the purpose of spectral fitting, without degrading the signal, would be beneficial. An immediate solution would be to sum over detectors within a narrow $d\theta dL_1$ range. In this manner, if the difference in the ToF expression of *J(y)* (which we can simplify as a difference in centroid positions and peak widths (dC,$d\Gamma$) is smaller than the error in fitted σ_M (in ToF) for individual detectors, a fit to the summation of these detectors will not yield spurious σ_M values. There is also the possibility of a great saving in measurement time, whereas current runs for a single temperature take upwards of 18 hours. A visual representation for this is given in figure 7, where it can clearly be seen that spectra condense at lower and higher angles (128°-132°, 158°-162°). In practise it would therefore be feasible to define artificial detector banks within the backscattering detector modules, whereby ToF spectra for detectors of similar θ and L_1 values are summed and fitted to a single C(t) of averaged detector geometry. As discussed by Krzystyniak *et al.* in the same issue, a way to increase the information content of sparse data on VESUVIO involves an examination of so-called Cumulative Angle-Averaged Data (hereafter CAAD) in either forward or backscattering geometry. In this approach, scattering data measured at a given detector is first integrated in the ToF domain using numerical integration, then normalized by its TOF integral, and finally summed over a given range of detectors with the CAAD fitting function following the same fate as the CAAD signal. This data-analysis procedure leads to a more transparent assessment of the performance of a given nuclear-momentum-distribution model, as well provides a convenient starting point for the development of the 'detector-focusing' schemes described above.

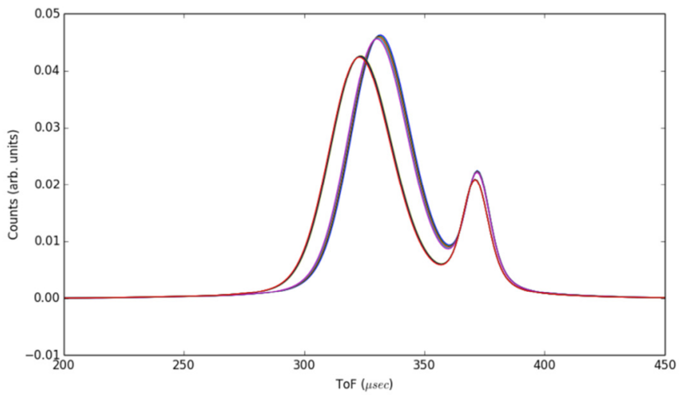

Figure 7. Simulated DINS spectra for C in Sn-foil. The overlaid spectra are for θ=128.0,129.0,130.0, 131.0 and 158.0,159.0,160.0,161.0,162.0.

4. Conclusions: VESUVIO When?

The VESUVIO spectrometer remains the only instrument providing a user programme for DINS research. Since its last major upgrade, a great advancement and diversity in the science performed by the instrument has been witnessed, as demonstrated by the research themes underpinning beam-time requests. We have proposed a number of instrument upgrades and extensions to VESUVIO that would enhance the current experimental capabilities of the instrument.

The most important limitation of VESUVIO is not one of design, but of flux. Were an upgrade to the ISIS target and moderators to be undertaken that would result in a higher incident flux (and thus count-rate) on VESUVIO, the effects on scientific capability cannot be overstated. Experimental durations would be brought down from the current 18 hours for routine measurements to those of inelastic or quasielastic neutron scattering. Increased flux would also enable an increase in L_0, which could vastly improve mass discrimination for H/D and MANSE experiments. This flux would currently come at a cost for the existing backscattering detectors, due to saturation effects. The ^{6}Li-doped glass scintillator detectors in backscattering are limited in terms of their electronics, in that they will saturate at approximately 45% scattering under the current flux on the instrument. Pronounced Bragg scattering from either sample or sample container can thus affect backscattering performance. An increase in flux on VESUVIO would either need to be attenuated in backscattering to avoid detector saturation or be compensated by an increase of L_1 (thus negating any possible gains on the instrument). Alternatively, replacing these detectors with the γ-detection technology already existing in forward scattering, where saturation is not a problem, would overcome this problem all together. The installation of a specific diffraction bank (scintillator detectors in backscattering at larger L_1) would still enable crystallographic identification in conjunction in this case.

There is a great need to extend the angular-coverage and number of detector banks on VESUVIO. By concentrating on higher forward-scattering and lower backscattering regions (a stark gap in the current instrument design) isolation and measurement of the kinetic energy and momentum distribution of D could be undertaken *simultaneously* to H. This extends DINS to a range of partially-deuterated samples that are currently difficult to measure on VESUVIO, but that are potentially important for the isolation and examination of nuclear quantum effects.

The manner in which MANSE samples are both measured and analysed is being revisited. Sample environment, particularly sample containers) plays a far greater role in these measurements than for those of the proton. We have demonstrated that by moving to foils of heavy-elements (Sn, Nb) rather than the Al containers commonly used, environment-intensity is shifted away from masses of interest in MANSE studies. The detector-by-detector methodology for DINS is also not optimised for MANSE, due to sparse, overlapping-intensity from masses >5 amu. One alternative proposed here is the approximation of average detector fitting. We propose that the error inherent in fitting to an averaged $C(t)$ is negligible for heavier masses, and far out-weighed by the improved statistics of summation.

Finally, one point should be mentioned that has been highlighted previously for future upgrades of VESUVIO [25,26], and that is the redesign of vertical detector- geometry in forward scattering. The resolution of VESUVIO for the measurement of both H and D is unparalleled, approximately 25% and 20% of the intrinsic width in *y*-space respectively. Of this, angular resolution dominates for H and is significant for D. By moving to a conical detector array around a Debye-Scherrer cone relative to the transmitted-beam axis, not only would the angular resolution for these detectors be improved by a factor of approximately 2, but the γ-background from the secondary analyser foils would be averaged to zero. This would be particularly important for increases in flux whereby mitigation of this sample independent γ-background would become relevant. If these detectors were also moved to a longer L_1 (again increasing the angular resolution for the detectors), the capabilities of VESUVIO for H/D momentum profiles would be drastically improved. This is, again, dependent on an increased flux.

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