Mass-selective Neutron Spectroscopy Beyond the Proton

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Abstract. We discuss ongoing methodological developments underpinning the determination of nuclear-momentum distributions from mass-resolved neutron Compton data of lightweight materials. To this end, two systems are considered in detail, namely, lithium hydride (including its deuterated counterpart) and squaric acid, an organic antiferroelectric material containing hydrogen, carbon, and oxygen. Beyond the usual case of the proton, our approach enables direct access to detailed line shape information associated with the underlying nuclear-momentum distributions of both deuterium and lithium. For oxygen and carbon, mean kinetic energies can also be obtained directly from the neutron data, as demonstrated by a detailed analysis of mass-resolved data from squaric acid. From an instrumentation point of view, this work provides a suitable platform for a detailed assessment of existing capabilities and future developments in mass-selective neutron spectroscopy on the VESUVIO spectrometer at ISIS.

1. Introduction

Neutron Compton scattering (NCS) is an experimental technique made possible by ongoing developments in accelerator-based neutron sources [1]. As described in more detail in Refs. [1, 2], the use of electron-volt (eV) neutrons to measure atomic kinetic energies is predicated upon the fact that the energy ($E$) and momentum ($Q$) transferred in the scattering process are sufficiently high such that the so-called impulse approximation (IA) provides a convenient starting point for the analysis of experimental data. In the specific case of the VESUVIO neutron spectrometer at the ISIS Pulsed Neutron and Muon Source in the United Kingdom, typical $E$ and $Q$ lie in the range 1-100 eV and 30-200 Å$^{-1}$, respectively [3, 4]. In addition to VESUVIO, we also note recent developments in NCS instrumentation by Rodríguez Palomino et al. [5, 6] at the Bariloche Electron LINAC in Argentina.

In physical terms, the IA implies that the neutron scatters from a single atomic centre, with conservation of total kinetic energy and linear momentum. Consequently, NCS probes all motions associated with a given atom as given, for example, by the relevant atom-projected vibrational density of states (VDOS) [2]. Under these conditions, it follows that the measured scattering response in the so-called Deep-Inelastic-Neutron-Scattering (DINS) regime is related in a simple way to the nuclear-momentum distribution (NMD) of a given species. In this context, NCS experiments on single-crystal specimens have allowed the reconstruction of the proton wave
function in much the same way as crystal structures may be obtained from diffraction data. Such a reconstruction is model independent and also allows a determination of the effective Born-Oppenheimer (BO) potential-energy surface of a given nuclide \[2\]. Conversely, comparison of experimental results from benchmark molecular and condensed-matter systems with \textit{ab initio} computations can also provide stringent tests for state-of-the-art first-principles calculations, including recent developments in Path Integral Molecular Dynamics (PIMD) methodologies to describe nuclear-quantum effects in condensed matter \[7\].

An important consequence of the above relates to the intrinsic mass selectivity of NCS, a condition dictated by the requisite conservation of linear momentum during the scattering process, i.e., each atomic mass follows a distinct recoil line on the \(E-Q\) plane obeying the kinematic condition \(E = \frac{\hbar^2 Q^2}{2M}\) where \(\hbar\) is Planck’s constant and \(M\) is the atomic mass. With sufficient spectral resolution (as that afforded by inverted-geometry neutron spectrometers like VESUVIO), the overall width of a given recoil line becomes sensitive to the chemical binding environment of a particular atom. Such mass and chemical selectivity (particularly for light nuclei) places NCS in a unique position relative to other scattering techniques.

To date, the primary application of NCS has focused on the study of nuclear quantum effects associated with proton motions. Systems investigated include fundamental fluids and solids \[8, 9, 10, 11, 12, 13, 14, 15, 16\], metal hydrides \[17, 18, 19, 20, 21\], glasses and amorphous materials \[22\], hydrogen-bonded systems \[11, 8, 12\], proton conductors \[23\], or hydrogen-storage nanostructured materials \[24\]. Beyond the proton, recent instrument developments offer the exciting prospects of extending the realm of applicability of NCS to MASS-selective Neutron Spectroscopic (MANSE) studies of heavier nuclei including lithium, boron, carbon, oxygen, or fluorine \[8, 9, 17, 18\].

MANSE studies have been reported recently on bulk and nanoencapsulated NaH \[25\], \(^7\)LiF \[26\], lithium hydride (LiH) and its deuterated counterpart (LiD) \[17, 18\], and heavy water and ice \[27\]. In spite of the relatively low neutron-scattering cross sections of elements like Li, these studies have demonstrated sufficient sensitivity to the underlying NMD for this technologically relevant nuclide via a judicious choice of experimental conditions. Building upon these recent efforts, the present work seeks to provide the foundations of a general methodology for future MANSE studies. To this end, we assess the information content of NCS data measured on VESUVIO and extend existing data-analysis protocols for the treatment of MANSE data, always with a view to a detailed comparison with the predictions of first-principles calculations. Two systems are considered in detail, namely: LiH and its deuterated cousin LiD; and squaric acid (SA), an organic antiferroelectric material containing hydrogen, carbon, and oxygen.

2. Methodology

2.1. Theoretical Considerations

We recall the basic features of NCS, and the reader is referred to Refs. \[1, 2\] for further details. Within the IA introduced earlier, the dynamic structure factor \(S(Q, E)\) for a given atom of mass \(M\) reduces to a single peak centered at the recoil energy \(E_r = \frac{\hbar^2 Q^2}{2M}\) \[28, 29, 30, 31, 32, 33, 34, 2\]. Consequently, \(S(Q, E) = \left(\frac{M}{\hbar^2 Q}\right) J(y)\), where \(y\) is the initial radial momentum \(p\) of the nucleus projected onto the scattering vector \(Q\). \(J(y)\) is the so-called NCS profile \[29, 28\] and formally corresponds to the longitudinal nuclear momentum distribution (NMD) of the target nucleus along \(y\). Mathematically \[28, 35\],

\[
y = p \cdot \hat{Q} = \frac{M}{\hbar^2 Q} (E - E_r) = \frac{M}{\hbar^2 Q} \left( E - \frac{\hbar^2 Q^2}{2M} \right)
\]

(1)

where \(\hat{Q}\) is a unit vector along \(Q\). Hereinafter, \(Q\) will be given in units of \(\text{Å}^{-1}\), \(E\) in meV, and \(M\) in \(\text{amu}\). With this choice of units, Planck’s constant is given by \(\hbar = 2.0446\) (meV \(\text{amu}\)^{1/2} \(\text{Å}\).
In what follows, systems where \( J(y) \) depends only on the magnitude of \( y \) but not on \( \hat{Q} \) will be considered in the IA limit. However, in this class of systems we have to consider two cases: (i) systems where departures from the approximation of a harmonically bound isotropic nucleus are observed, either through anharmonicity or anisotropy of the local effective BO potential, and (ii) harmonically bound isotropic systems. In the second case, a spherically averaged three-dimensional longitudinal NMD, \( J_{IA}(y) \), can be conveniently expressed via recourse to a Gram-Charlier expansion of the form \([2, 36]\)

\[
J_{IA}(y) = \frac{\exp\left(\frac{-y^2}{2\sigma^2}\right)}{(\sigma\sqrt{2\pi})} \sum_n c_{2n} \frac{1}{2^{2n}n!} H_{2n}(\frac{y}{\sigma\sqrt{2}}).
\]

where \( H_{2n} \) denotes a Hermite polynomial and the standard deviation (STD) of a given NMD is denoted by \( \sigma \). In Eq. (2), the term with \( n = 0 \) is set to unity due to the requisite normalization of the NMD. Moreover, with \( c_2 \) set to zero, the kinetic energy \( E_k \) of a nucleus of mass \( M \) is related to the STD of its NMD, \( \sigma \), via \( E_k = 3h^2/2M \). This relationship is independent of the precise values of the remaining coefficients \( c_{2n} \). Therefore, the fitting of \( J_{IA}(y) \) to the Gram-Charlier expansion amounts to varying both the width of the momentum distribution \( \sigma \) and the coefficients \( c_{2n} \), with \( n \geq 2 \).

For a harmonically bound isotropic system where \( J(y) \) depends only on the magnitude of \( y \), the NCS profile in the IA limit can be written as a normalized Gaussian \([34, 33, 2, 37, 30]\) which can be expressed as a special case of the Gram-Charlier expansion with coefficients \( c_{2n} = 0 \) for \( n > 0 \) in Eq. (2).

For finite values of \( Q \), corrections to the IA are known as ‘Final-State Effects’ (FSEs) \([34, 2]\). To account for FSEs, the method of Sears \([28]\) is routinely incorporated into standard NCS data treatments \([34]\) by expressing the measured NCS profile \( J(y) \) as a series of the form

\[
J(y) = J_{IA}(y) + J_{FSE}(y) = J_{IA}(y) - \frac{M(\nabla^2 V)}{36\hbar^2q} \frac{d^3}{dy^3} J_{IA}(y) + ... \quad (3)
\]

where \( J_{IA}(y) \) is the IA result given by Eq. (2). \( \langle \nabla^2 V \rangle \) is the mean value of the Laplacian of the potential energy of the nucleus expressed in meV Å\(^{-2}\) (cf. Refs. \([34, 38]\)).

The STD of a given NMD, \( \sigma \), as well as its associated Laplacian, \( \langle \nabla^2 V \rangle \), may be calculated from the underlying VDOS under the assumption that the NMD for a given nucleus in a crystal along the \( \hat{Q} \) direction assumes a purely multivariate Gaussian functional form. This procedure is commonly referred to as the ‘Gaussian’ approximation (GA) \([39, 2, 8, 17]\). Similarly, the spherically averaged value of \( \sigma \) for nucleus \( n \), \( \sigma_n \), can be obtained using the appropriate atom-projected VDOS (PVDOS), \( G_n(\omega) \), from \([8]\)

\[
\sigma_n^2 = \frac{M_n}{\hbar^2} \int G_n(\omega) \frac{\omega}{2} \coth \left( \frac{\omega}{2k_BT} \right) d\omega. \quad (4)
\]

\( G_n(\omega) \) for a harmonic solid can also be used to estimate the magnitude of FSEs \([17]\) since \([40]\)

\[
\langle \nabla^2 V_n \rangle = \frac{3M}{\hbar^2} \int \omega^2 G_n(\omega) d\omega. \quad (5)
\]

For the case of an isotropic three-dimensional harmonic oscillator (3D-HO) we can write

\[
\langle \nabla^2 V \rangle = \frac{12\hbar^2\sigma^4}{M}. \quad (6)
\]

Rewriting Eq. (3) using a new variable, \( x = \frac{y}{\sqrt{2\sigma^2}} \), and using the definition of the third-order Hermite polynomial, \( H_3(x) = 8x^3 - 12x \), we arrive at
Then, an explicit connection between the parameter \( k \) and \( \sigma \) can be established for the case of a harmonically bound nucleus

\[
k = \sigma \frac{\sqrt{2}}{12}
\]

Both \( \sigma \) and \( k \) or, more generally, the mean Laplacian of the potential energy of a given atomic nucleus, can be used as fitting parameters in the analysis of NCS data and then compared with a prediction based on an \textit{ab initio} calculation. Since the second moment of a given NMD as well as the magnitude of FSEs can be calculated directly from the underlying VDOS, these two parameters can (in principle) be disentangled by devising analytical criteria constraining NCS observables.

2.2. Experimental Protocol

All NCS measurements were performed on the latest incarnation of the VESUVIO spectrometer at the ISIS Pulsed Neutron and Muon Source, United Kingdom [3, 4, 41]. As an inverted-geometry neutron spectrometer, the sample is exposed to a polychromatic neutron beam characterized by an incident-energy spectrum \( I(E_0) \). Incident neutrons having initial energy \( E_0 \) travel a distance \( L_0 \) from the pulsed source to the sample. After scattering at an angle \( \theta \), neutrons of final energy \( E_1 \) travel a distance \( L_1 \) to the detector as schematically shown in Fig. 1. To achieve energy selection, a thin foil is placed in front of the detectors. This foil absorbs neutrons over a narrow range of energies, thereby fixing the final energy of the detected neutrons. All NCS measurements described in this work used a gold (Au) foil with a resonance energy of \( E_1 = 4.9 \) eV. With this experimental arrangement, data collection necessarily involves foil cycling in both forward and backscattering geometries in order to obtain TOF data with and without energy selection, followed by a subtraction of these two data sets to obtain NCS spectra. In forward scattering, the energy-resolution function is adequately described by the convolution of a Lorentzian of half-width-at-half-maximum (HWHM) 24 meV and a Gaussian with STD 73 meV [42]. In backscattering, the resolution function is nearly Gaussian with extended (small) wings, and can be accurately described as the convolution of a Gaussian (STD=74 meV) and a Lorentzian (HWHM=24 meV) [43]. For further details of the VESUVIO spectrometer, see the contribution by Seel \textit{et al.} in the same issue.

The treatment of experimental NCS data is routinely performed in the time-of-flight (TOF) domain for a set of detectors placed at a series of scattering angles \( \theta \). For a given mass \( M \), instrument-calibration procedures establish a unique relation between the values of \( Q \), \( E \), and \( y \) on each TOF channel \( t \) and scattering angle \( \theta \) [43, 34]. With these considerations in mind, the total number of neutrons detected in an NCS spectrometer for a given mass \( M \) in a TOF channel \( t \) is proportional to the Compton profile, \( J[y_M(t)] \), convolved in \( y_M \) space with a (mass-dependent) instrumental resolution function \( R[y_M(t)] \). Thus, for a total of \( N \) different masses present in the sample, the total count-rate at a fixed scattering angle \( \theta \), \( C_\theta(t) \), can be written as the following sum [cf. [34], Eq. (2.24)]:

\[
C_\theta(t) = A'[\frac{E_0 I(E_0)}{Q}]_t \times \\
\sum_{n=1}^{N} I_n M_n J_n[y_n(t)] \otimes R_n[y_n(t)]
\]
Figure 1. Schematic diagram of the VESUVIO spectrometer. For further details, see the main text and Seel et al. in the same issue.

where $A'$ is a mass-independent experimental constant and the mass-independent factor $\frac{\langle E_0 I(E_0) \rangle}{q}$, depends on the incident neutron spectrum, $I(E_0(t))$, the initial neutron energy, $E_0(t)$, and the momentum transfer $Q(t)$, all explicit functions of $t$. In Eq. (9), the NMD for mass $M$, $J_M(y_M(t))$, is given by Eq. (3) with the expression for the NMD in the IA limit, $J_{IA}(y)$, given by Eq. (2). Integrated peak intensities $I_M$ for a given scattering mass $M$ are proportional to the scattering density $I_M = AN_M \sigma_M$, where $\sigma_M = 4\pi b^2_M$ is the total (bound) neutron-scattering cross section [44, 34].

For the NCS measurements on LiH and LiD, the specimens were enclosed in 6x6 cm$^2$ flat aluminum (Al) cells resulting in a total transmission of 85 and 90 % for LiH and LiD, respectively. In the case of SA, the sample was enclosed in a flat niobium (Nb) cell with 50-μm-thick windows leading to a total transmission of ca. 80 %. Use of thin Nb foils served to minimise contributions of the empty cell to the overall NCS response as well as spectral overlap with oxygen and carbon. All measurements reported herein were carried out over the range 4-300 K using a dedicated helium closed-cycle refrigerator.
3. Results and Discussion

3.1. Lithium Hydride and Deuteride

In recent works, Krzystyniak et al. [17, 18] have examined in detail the NCS response of both LiH and LiD, including a comparison of experimental NCS data with first-principles calculations and an assessment of the harmonic and impulse approximations over the kinematic range currently accessible on the VESUVIO spectrometer. As part of these efforts, a number of physical models for the underlying NMDs were compared with available experimental data, leading to the conclusion that for this particular system a purely Gaussian NMD and FSEs compliant with the 3D-HO model ($k = \frac{\sigma^2}{12}$) provide a satisfactory description of the NCS response for all nuclei in these materials. For LiH, it was found that $\sigma_H = 3.5 \pm 0.1 \text{ Å}^{-1}$ and $\sigma_{Li} = 7.0 \pm 0.4 \text{ Å}^{-1}$ in forward scattering, in excellent agreement with previous \textit{ab initio} calculations ($\sigma_H = 3.57 \text{ Å}^{-1}$ and $\sigma_{Li} = 7.14 \text{ Å}^{-1}$) [17]. For LiD, the same model gives $\sigma_D = 4.4 \pm 0.1 \text{ Å}^{-1}$ and $\sigma_{Li} = 7.2 \pm 0.9 \text{ Å}^{-1}$ in forward scattering. These values are within one STD from \textit{ab initio} predictions, namely, $\sigma_D = 4.43 \text{ Å}^{-1}$ and $\sigma_{Li} = 7.15 \text{ Å}^{-1}$. Although considerably less accurate, the widths of the Li momentum distributions in backscattering are also consistent with the above results, with values for $\sigma_{Li}$ of $9.0 \pm 2.2 \text{ Å}^{-1}$ and $7.0 \pm 0.8 \text{ Å}^{-1}$ for LiH and LiD, respectively. Ref. [18] also explored the possibility of increasing the information content of NCS data by examining Cumulative Angle-Averaged Data (hereafter CAAD) in both forward and backscattering geometries. In this approach, this sum was directly compared with the corresponding sum of individual fits for a given detector at a given scattering angle $\theta$ using unit-area-normalised expressions for $C(\theta, t)$ given by Eq. (9), i.e., $\text{CAAD} = \sum_{i=1}^{N} \frac{C(\theta, t)}{\int C(\theta, t)\,dt}$. The summation range for this quantity corresponds to a selection of detectors after exclusion of outliers and faulty detectors. In practice, the selection process involves the removal of individual TOF spectra recorded by detectors which, when fitted with the expression given by Eq. (9), lead to reduced $\chi^2$ values lower than 0.8 and higher than 1.2 [18]. This data-analysis procedure leads to a more transparent assessment of the performance of a given NMD model as well provides a convenient starting point for the development of ‘detector-focusing’ schemes aimed at increasing the effective count rate of TOF data relative to current data-analysis protocols.

As a natural extension of the above strategy, we consider the analysis of ‘focused’ NCS data directly obtained from a CAAD following the same criteria for the rejection of noisy and/or faulty detectors and outliers described in our previous work [18]. Figures 2 and 3 show LiH and LiD summed TOF data in forward and backscattering along with their respective fits. TOF data and associated error bars are shown in light grey, highlighting an order-of-magnitude improvement in signal-to-noise ratios (SNRs) relative to single-detector NCS data. These TOF data were pre-processed using the protocol described in detail in Ref. [18]. This procedure included the subtraction of multiple-scattering contributions, self-shielding and attenuation, and absorption. Contributions from $\gamma$ backgrounds affecting the performance of the forward-scattering detector bank were removed using the standard VESUVIO data-reduction routines [45]. The data fits shown in these figures demonstrate effective mass separation of all nuclei in both forward and backscattering geometries. As visual aid, individual mass contributions have been colour-coded: blue for H and D in LiH and LiD, respectively; green for Li; and red for Al. The thick black lines in both figures describe the total TOF count rates, including sample container. The experimental data with associated error bars are shown as grey points.

Inspection of Figs. (2) and (3) demonstrates the adequacy of a physical model based on a Gaussian NMD with FSEs given by the 3D–HO model for both LiH and LiD in both forward and backscattering. The ratios of integrated signals associated with Li and Al, namely, $N_{Li}/\sigma_{Li}/N_{Al}/\sigma_{Al}$, read 0.8 and 2.8 for LiH and LiD, respectively. These ratios remain equal for forward and backscattering, further confirming the robustness of our data-analysis protocols. We note that, at present, CAAD fitting curves are generated assuming no TOF-dependent...
Figure 2. LiH CAADs and associated fits for (a) forward and (b) backscattering geometries. For further details, see the main text.
these limitations, only the width of the Li-recoil peak in LiD backscattering data was considered for comparison with previous studies. Overall, the NMD parameters obtained using the CAAD compare very well with those previously reported using other approaches [17, 18]. For LiH, we find $\sigma_H=3.45\pm0.03$ Å$^{-1}$ and $\sigma_{Li}=7.09\pm0.95$ Å$^{-1}$ in forward scattering, and $\sigma_{Li}=7.02\pm0.25$ Å$^{-1}$ in backscattering. For LiD, $\sigma_D=4.09\pm0.10$ Å$^{-1}$ and $\sigma_{Li}=8.4\pm0.5$ Å$^{-1}$ in forward scattering, and $\sigma_{Li}=6.4\pm0.4$ Å$^{-1}$ in backscattering. On the whole, the analysis of NCS data using a CAAD provides improved estimates of NMD parameters for Li in both forward and backscattering geometries relative to data-analysis protocols relying on far more sparse data associated with individual detectors. Forward and backscattering data for D in LiD still pose a number of challenges in either case owing to the presence of TOF-dependent experimental backgrounds. Work is in progress to improve the CAAD method to account for the presence of backgrounds in TOF data. Notwithstanding these limitations, forward-scattering data can still be used to provide reliable estimates for the underlying NMD and these results can still be used to check the consistency of the NCS data across detector banks. The essentially harmonic and highly isotropic character of the underlying VDOS in both LiH and LiD is ascertained by the analysis of NCS data presented in this Section. Extension of this work to highly anisotropic or anharmonic systems represents a natural extension of the above work in order to assess the loss of sensitivity to higher-order terms describing the underlying NMD, as NCS data are progressively averaged over scattering angle to generate a CAAD.
3.2. Hydrogen, Carbon, and Oxygen in Squaric Acid

Encouraged by the results presented above on LiH and LiD, we have applied the same methodology to solid SA, a carbon-based antiferroelectric material containing H, C, and O (molecular formula C$_4$O$_4$H$_2$). In the solid, individual SA molecules are arranged in layers perpendicular to the $b$-axis, with strong hydrogen bonds within the $a-c$ plane. Below the onset of paraelectricity at 373 K, the four-membered carbon ring associated with a single molecular unit is distinctly asymmetric and hydroxyl (OH) groups across adjacent layers are arranged in an antiparallel fashion, giving rise to the observed antiferroelectricity. In parallel with the NCS measurements reported herein, we have performed detailed single-crystal neutron diffraction measurements on SA as a function of temperature using the SXD instrument at ISIS [46]. The temperature dependence of lattice constants and hydrogen-bond parameters shows an increasing decoupling of the crystal layers with temperature (expansion of the $b$-axis), and a concomitant weakening of the hydrogen bonds across individual SA units on the $a-c$ plane, a trend which starts well below room temperature. Given the above observations, SA constitutes a suitable molecular system to explore the sensitivity of NCS data to the anisotropic character of strong hydrogen bonds, as well as possible (and hitherto unexplored) changes in the nuclear dynamics of the heavier C and O species as a function of temperature.

The total NCS response of SA has been described as a angular sum of unit-area-normalised spectra accounting for the total count rate in TOF, i.e., $\text{CAAD} = \sum_{i=1}^{N} \int C(\theta, t)\,d\theta$. Underlying NMDs for all nuclei were assumed to be Gaussian and the detector sum to run over all forward and backscattering detectors. FSEs were constrained within the 3D-HO model as shown by Eq. (8). In principle, the quality of the data allows for a determination of NMD parameters for the Nb cell used as sample container, although we note that these values are heavily dominated by the present resolution function of the instrument. Thus, Nb NCS widths at 18 K and at room temperature or above were constrained using tabulated Debye values corresponding to the low- and high-temperature limits [47]. These two limits give NMD STDs for Nb of 14 and 24.4 Å$^{-1}$ at 18 K and 300 K, respectively. Additionally, CAAD fits in forward and backscattering were constrained to obey the expected 1:1 stoichiometry of C and O in SA. In order to check the feasibility of our MANSE approach in forward scattering for O- and C-containing systems, no constraints were imposed on the NMD STDs of these two nuclei, i.e., their numerical values were left free in the fitting sessions of both back- and forward-scattering data. Data reduction protocols including the removal of noisy and/or faulty detectors and outliers were identical to those employed for LiH and LiD in the previous Section.

MANSE spectra of SA along with their fits are shown in Figs. (4) and (5) for base (18 K) and room temperature (300 K), respectively. The experimental data with associated error bars are shown as grey points. The total fitted NCS response is shown in black, and the coloured lines represent contributions from individual nuclei: (i) blue for H; (ii) green for C; (iii) red for O; and violet for the Nb cell. At both temperatures, the fits provide an excellent description of the experimental NCS data, clearly showing the effective discrimination of all masses when both forward and backscattering data are analysed synchronously. Forward-scattering data can effectively discriminate between the NCS of the proton relative to other masses. In backscattering, the NCS response from C, O, and Nb can be separated into three distinct contributions.

To the best of our knowledge, these data represent the first instance where MANSE has been used with success to unambiguously separate up four different nuclei in a condensed matter system. Moreover, we note that a judicious choice of sample-environment conditions leads to a minor contribution from the container to the overall NCS response, a situation to be contrasted with the LiH and LiD data presented earlier. On the basis of these results, we anticipate that further technical developments on this front will be necessary for the development of MANSE into a fully fledged technique for the study of complex materials using NCS.
From the fits of CAAD data in forward-scattering shown in Figs. (4) and (5), we obtain $\sigma_H = 4.6 \pm 0.2 \text{ Å}^{-1}$ and $4.7 \pm 0.2 \text{ Å}^{-1}$ at $T = 18 \text{ K}$ and $T = 300 \text{ K}$, respectively. In comparison with LiH, these values represent a ca. 30 % increase in NCS widths, illustrating the sensitivity of NCS to an increase in ‘chemical temperature’ (binding strength) of the proton in SA relative to LiH in spite of its predominantly two-dimensional nature. Moreover, our results on SA demonstrate that the CAAD analysis followed in this work provides a convenient platform for MANSE studies of masses $M > 10 \text{ amu}$. NMD STDs for C and O in SA at $T = 18 \text{ K}$ amount to $\sigma_C = 12 \pm 2.7(12 \pm 0.3) \text{ Å}^{-1}$ and $\sigma_O = 12 \pm 2(11.4 \pm 0.3) \text{ Å}^{-1}$ in forward(backscattering). At $T = 300 \text{ K}$, $\sigma_C = 14 \pm 2(14 \pm 0.3) \text{ Å}^{-1}$ and $\sigma_O = 14 \pm 2(14 \pm 0.3) \text{ Å}^{-1}$ in forward(backscattering). Work is in progress to provide a basis for comparison of the above experimental values with first principles DFT results. As expected on the basis of previous considerations, the higher resolution of the backscattering detector banks leads to improved estimates of NMD STDs. Thus, a sensible data analysis strategy for MANSE data given current experimental capabilities on VESUVIO involves the use of forward-scattering CAADs for the analysis of proton-recoil data, and backscattering data for heavier nuclei, particularly those with masses greater than 10 amu. The contribution of Seel et al. in the same issue discusses a number of ways to circumvent these limitations in the future.

4. Outlook
The results presented herein provide a suitable platform for ongoing developments in MANSE using NCS techniques. We have shown that our data-analysis strategy enables the analysis of
Figure 5. CAADs and associated fits for SA in (a) forward and (b) backscattering geometries at a temperature of $T=300$ K. For further details, see the main text.

MANSE data from composite materials containing H, D, Li, C, and O. Unlike neutron-scattering techniques using thermal and cold neutrons, neutron absorption by naturally occurring Li (or B) is substantially reduced in the epithermal (eV) region. Further work is needed to establish absolute sensitivity levels for these technologically relevant elements using MANSE, as well as a detailed assessment as to whether there is a real need for (typically costly) isotope enrichment protocols.

In the above context, we have also reported for the first time the analysis of MANSE data involving four distinct nuclei over the atomic-mass range 1-100 amu. MANSE data are also amenable to direct comparison with first-principles predictions and, as such, we anticipate that this much-needed synergy between NCS experiments and computational materials modelling will become an increasingly important area of activity in the foreseeable future. Extension of our approach to heavier masses and more complex materials requires both advances in instrumentation, to increase count rates and mass resolution, as well as an increasingly sophisticated and robust data-analysis toolkit to improve model-selection criteria.

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