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# The road to a station for epithermal and thermal neutron analysis

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**Abstract.** Despite the large variety of research interests and themes motivating the current neutron research included in this collection, we have found common denominators characterising the manner in which the chosen research methodology tries to tackle the envisaged scientific questions. This article attempts to characterise those trends in current research with the aim of identifying the main mid-to-long term opportunities faced by electron-Volt neutron spectroscopy. The main realisation from this exercise is that the scientific community seems eager to combine neutron-based techniques over a broad energy range. To this end, the most natural choice seems to be to resort to neutron instruments where such capabilities are already present from the outset, with the most prominent example being the VESUVIO spectrometer at the ISIS pulsed neutron and muon source in the UK. However broad the operational basis of the existing neutron beamline infrastructure may be, progress, achievable only through further instrument upgrades, is the only way forward. The need to move forward is clearly seen within the community and is well documented by the research presented in this collection. This need for a substantial upgrade has crystallised in the form of a proposition to build a station rather than a conventional beamline, for Epithermal and Thermal Neutron Analysis station, hereafter ETNA.

## 1. Introduction

The application of electron-Volt (eV) neutron spectroscopy to scientific investigations on condensed-matter systems has witnessed, over the last few years, an impressive development. Looking at the proceedings of the VI (2014) [1] and VII (2017) international workshops on eV neutron spectroscopy [2], one can notice how the use of Deep Inelastic Neutron Scattering (DINS) to characterise Nuclear Quantum Effects (NQEs) mostly on hydrogen dynamics [3] has lead to MAss-selective Neutron SpEctroscopy (MANSE), whereby the Nuclear Momentum Distributions (NMDs) of several elements in a given sample are accessed at the same time [4, 5, 6].



Furthermore, a number of recent investigations carried out on the VESUVIO spectrometer [7], the flagship instrument for eV neutron spectroscopy at the ISIS Facility [8], successfully combined DINS with concurrent Neutron Diffraction (ND) measurements [9, 10], or employed broad-range Neutron Transmission (NT) [11, 12]. Moreover, a novel trend of concurrent characterisation of condensed-matter systems by means of DINS, ND, and Neutron Resonance Capture Analysis (NRCA) is proving to be a potent strategy and thus is gaining momentum [13].

Here, we summarise the most prominent and successful examples of the currently prevailing trends in eV neutron spectroscopy, with the aim to identify the main mid-to-long term objectives faced by the techniques employed as drivers for much-needed upgrade of the existing beamline infrastructure.

## 2. Neutron spectroscopy and modelling

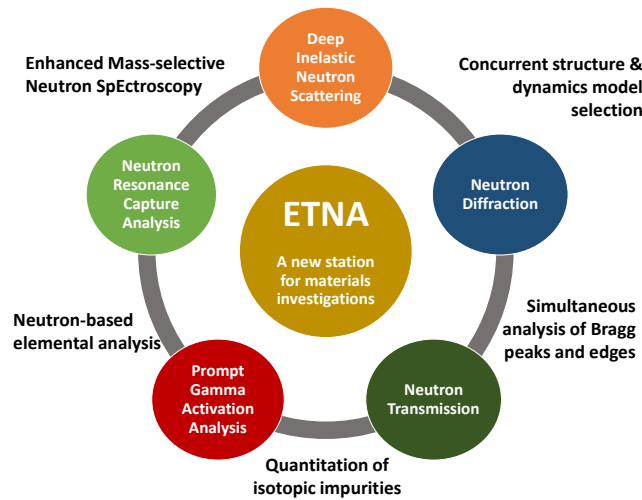
Inelastic Neutron Scattering (INS) can be considered as the neutron-based technique nearest to DINS. On the one hand, mass-resolved DINS spectra can be obtained from measured vibrational densities of states by evaluating the multi-phonon expansion well beyond the first term, and in the limit of large energy and momentum transfers [14], *i.e.*, within the celebrated Impulse Approximation (IA) [15]. On the other hand, the widths of NMDs accessed through DINS can be related via harmonic models to the frequencies of molecular vibrations [16], or to simulated atom-projected vibrational densities of states [17].

Several articles in this collection have explored the link between vibrational spectroscopy and epithermal neutrons. Parker *et al.* give an example of how thermal and epithermal neutrons at spallation neutron sources can be used to study the stretching mode in H<sub>2</sub>, with future applications to characterise the molecule's interaction with materials – a simpler task from the simulation point of view than the modelling of the 0→1 rotational transition. The article provides an instructive example on how the INS signal from light atoms, investigated with epithermal neutrons, follows the nuclear recoil line while approaching the IA. These results gain a particular interest when considered in the framework of a previous study by Senesi *et al.* [18] discussing the DINS signal obtained on direct-geometry INS spectrometers, particularly in the case of the SEQUOIA instrument at SNS, where the energy of the incident neutrons was as high as 6 eV.

The contribution by Kolensikov *et al.* is an exquisite example of how the understanding of a given system, in particular ultra-confined water, can be enhanced by comparing INS and DINS data. In this case, INS peaks interpreted as tunnelling modes affecting the confined water molecule were directly related to the delocalisation of water's hydrogen accessed through the analysis of its NMD [19]. Moreover, the comparison of INS and DINS data at several temperatures allows for a deeper discussion on the origin of hydrogen bonding and its evolution with the thermodynamic phase.

The attempt to reproduce both INS and DINS spectra by first-principles simulations can be regarded as a stringent selection protocol for a given theoretical model, especially for those systems particularly affected by NQEs or anharmonic local potentials, as in the case of formic acid [10]. State-of-the-art computer simulations including NQEs, based on the path integral formalism [20], have found in DINS experiments a much-needed benchmark, such as in [21, 9]. However, the time-consuming nature of first-principles simulations has pushed the community to look for new strategies [22]. In this regard, Druzinski *et al.* show how to successfully convert the results from a *per se* classical computer simulation based on molecular dynamics so as to account for NQEs by using a coloured-noise thermostat previously suggested by Ceriotti [23].

The relation between NMDs and the ordering degree of the local potential affecting carbon in several allotropes was tackled by Armstrong *et al.* The authors replaced the concept of bond strength with the Hausdorff-Besicovitch fractal dimension, a concept borrowed from mathematical topology, deriving a relation between the fractal dimension of a given system and the carbon nuclear kinetic energy, supporting the experimental results with simulated atom-



**Figure 1.** Experimental possibilities available on ETNA.

projected vibrational densities of states.

### 3. Concurrent investigation of structure and dynamics

Building upon the synergy between INS and DINS, and taking advantage of the possibility to concurrently measure DINS spectra and ND data, recent investigations were able to model and optimise the structure and dynamics of complex systems over a uniquely large time- and space-window [24, 25]. Following this strategy, Krzystyniak *et al.* tackle the challenging problem of glassy-versus-polycrystalline structures in Zr–Be binary mixtures (see also Ref. [26]).

Another contribution by Krzystyniak *et al.*, applies concurrent ND and DINS to nitrogen-doped superconducting radio-frequency niobium cavities. The conditioning procedure was found to lead to systematically larger Nb lattice parameters, also providing an estimate of nitrogen concentration. Moreover, DINS was found particularly sensitive to the broadening of the niobium NMD in the doped sample as compared to a standard one, that was related to an increased degree of ordering and binding in the niobium local potential.

### 4. Neutron-induced photon spectroscopies

Epithermal neutrons are particularly suitable for applications to elemental analysis of bulk materials, in scattering experiments based on DINS and MANSE, as well as in absorption experiments based on NRCA, neutron resonance transmission analysis, and Prompt-Gamma Activation Analysis (PGAA). The effort of the community towards this end is well represented in the collection of articles presented here, as well as in Ref. [13]. For example, Nardini *et al.* present an interesting combination of NRCA and other chemometric tools, in particular principal-component analysis, applied to archaeological artefacts so as to identify trace elements, and thereby their origin and history.

The contribution by Romanelli *et al.* continues along the lines of previous studies by Mayers *et al.* [27] and Postma and Schillebeeckx [28] to use NRCA to analyse the resonance line shape to obtain information about the local quantum temperature affecting an atom, and the associated NQEs. This approach is suggested as a high-throughput technique whereby relatively fast measurements can be applied to parametric studies, for example as a function of temperature.

The technique is able to tackle the dynamics of heavy-weight elements for which the DINS resolution would otherwise be a limiting factor.

Recent studies made use of a high-purity Ge detector to acquire high-resolution photon-energy and time-of-flight spectra, whereby the photon peaks can be correlated to the arrival time of neutrons in the sample area. The contribution by Onorati *et al.* (see also Ref. [29]) discusses the origin of the  $\gamma$ -background levels on the VESUVIO spectrometer, as well as other beam-lines at ISIS, including the effects due to a recent change to the water moderator serving the instrument [30, 31]. Details on the absolute calibration of the high-purity Ge detector are given in the contribution by Parmentier *et al.* The characterisation and indexing of the  $\gamma$ -ray spectra is the first step to design low-background measurements and increase the detection and quantitation thresholds for elemental analysis.

## 5. Detection and instrument modelling

As already mentioned in the conclusions of the proceedings of the VI workshop [32], the long acquisition times to collect DINS data with good statistics is one of the limiting factors preventing parametric studies on the VESUVIO spectrometer. The preliminary work by Ulpiani *et al.* shows how it is possible to increase the count rate for the front-scattering YAP detectors by fine-tuning the electronics parameters underpinning the detection procedure. In addition to shorter measurements, such an upgrade could improve the detection limits of hydrogen in materials, paving the way for a new application of the technique in industrial problems.

More generally, there is a need to optimise the geometry of the VESUVIO spectrometer so as to increase the beam flux without sensibly worsening the instrument's resolution. The modelling of the instrument using McStas, presented by Di Giulio *et al.*, goes in this direction. Codes for the simulation of neutron transport are not optimised for epithermal neutrons, therefore the development of new components, such as those simulating energy filters based on nuclear resonances, are much needed and will speed-up any foreseeable instrument-development project.

## 6. Strategies for data analysis

Following a previous update [33], Romanelli *et al.* have presented a new script to run the analysis of DINS spectra in MANTID. The script makes use of the algorithms included in the software to load and visualise the data, perform preliminary fits on the raw data, and run corrections for multiple scattering and  $\gamma$  backgrounds. Moreover, a number of consistent analysis strategies are presented, including sequential or global fits of hydrogen NMDs from different detectors in the West-scaling variable. A detailed and independent approach to the analysis of DINS data is presented by Dawidowski *et al.* as well. The combination of several procedures to reduce and fit DINS data is the strongest proof of consistency in the data analysis, and the implementation of these strategies is being implemented progressively in the new MANTID releases.

The novelty in the handling of DINS data in MANTID includes the implementation of Bayesian tools for the selection of the fitting model, also discussed in Ref. [34], here stressing non-negativity and other physical constraints to fitted NMDs. Krzystyniak *et al.* also show how to extract value of the mean kinetic energy from the final-state corrections to the IA, a procedure that can provide an estimation of the width of experimental NMDs at an early stage of data reduction, to be used as a starting parameter for subsequent data analysis.

## 7. Outlook and ways forward

Looking at these proceedings and at a recent review [4], the scientific community using eV neutron spectroscopy on the VESUVIO spectrometer has broadened the area of investigated subjects towards, for example, energy materials [35] and real-life systems [36]. Such an evolution in the scientific programme has been catalysed, amongst other reasons, by an increased attention to the several techniques simultaneously available during a DINS experiment. Although the

instrument was designed originally for applications in neutron spectroscopy, it is now considered a good-resolution diffractometer [30], as well as a station where neutron cross sections can be accurately measured over a broad energy range, from a fraction of meV up to tens of keV. Additional attention is now paid to the NRCA capabilities of the photon-sensitive front-scattering detectors, and PGAA experiments are becoming more regular.

Driven by the needs the community has expressed, there are a number of upgrades that could pave the way to even more challenging investigations. The fine tuning of the detection electronic chain has already showed promising results [37], with saturation problems arising from the increase in count rates being mitigated by the recent installation of last-generation data-acquisition electronics, DAE3, now in its test period on VESUVIO. The optimisation of the detection procedures should be reinforced by an increase of the angular coverage of scattered neutrons, a limiting factor at present for acquisition times, as well as a rearrangement of the existing detectors onto Debye-Scherrer cones at constant scattering angle, a way to increase the signal-to-noise ratio at the data-analysis stage.

Cleaner measurements of NRCA and PGAA require a suppression of the background within the VESUVIO blockhouse, a task addressed by additional Pb shielding towards the beam stop and the moderator, under installation at present, and the possibility to cycle in the primary path a Cd foil to attenuate the neutron flux at thermal energies. The foil changer, in its design stage at present, will allow additional tests of direct-geometry DINS spectra by cycling a resonant foil in the incident beam, with the possibility to improve the MANSE capabilities of the instrument. Also, the instrument is at present collecting data with a fine binning of  $0.1\ \mu\text{s}$ , instead of the traditional  $0.5\ \mu\text{s}$  binning, so as to improve the resolution both for NRCA and DINS, at low time-of-flight values.

Furthermore, the capabilities available with neutron transmission can be increased by including a monitor with both spatial- and energy-resolution, therefore making neutron imaging available, with possible applications to the characterisation of catalytic processes as in the case of [12]. Such a detector would serve as a monitor for a more robust correction of DINS data, and as a diagnostic tool.

The evolution of eV neutron spectroscopy has proved to be successful and exciting. Driven by this well-documented and highly exciting upsurge in research activity, a clearly emerging community-driven need for a substantial upgrade of the VESUVIO instrument will pave the way to tackle challenging investigations yet to come. The main result is the awareness of the need for a station for Epithermal and Thermal Neutron Analysis, a future instrument already referred to as ETNA.

As organisers, we have enjoyed the VII international workshop on electron-Volt neutron spectroscopy, and the related proceedings collected in this volume. We look forward to the next workshop, eager to understand, from this prolific scientific community, what are the best ways forward. As always, the sky is the limit.

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