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# VESUVIO+: The Current Testbed for a Next-generation Epithermal Neutron Spectrometer

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**Abstract.** We present an overview of ongoing developments in epithermal neutron spectroscopy using the VESUVIO+ beam line at the ISIS Facility. In its current incarnation, VESUVIO+ provides a suitable platform for further and much-needed progress in the judicious exploitation of epithermal neutrons at a pulsed spallation source, as well as constitutes a necessary milestone towards a next-generation station for Epithermal and Thermal Neutron Analysis, hereafter ETNA. In particular, we discuss recent improvements in capability relative to its predecessor VESUVIO. These include the concurrent use of mass-resolved neutron spectroscopy, transmission, and diffraction to explore the properties of complex functional materials, as well as the implementation of techniques unique to pulsed neutron sources such as $\gamma$-ray dopplerimetry and energy-resolved prompt-$\gamma$ activation analysis.

## 1. Introduction

Over the past decade, the VESUVIO spectrometer at the ISIS Pulsed Neutron & Muon Source [1–3] has supported an international community across many areas of contemporary science including chemistry, physics and cultural heritage [2, 4, 5]. Deep Inelastic Neutron Scattering (DINS) has been the primary technique for the measurement of Nuclear Momentum Distributions (NMDs). The past few years have witnessed a progressive shift of DINS beyond fundamental condensed-matter systems [3, 5–7]. This has been the primary driver behind ongoing improvements to the instrument, hereafter denoted as VESUVIO+. Figure 1 provides a visual summary of the recent transition from VESUVIO to VESUVIO+. In particular, a series of concurrent techniques are now available, including transmission, diffraction, (n,$\gamma$)-resonance dopplerimetry, and Prompt-Gamma Activation Analysis (PGAA) [8]. Recent changes to the water moderator in the ISIS Target Station 1 have lead to a substantial increase in the thermal neutron flux without compromising the resolution of the instrument [8]. Methodological advances in neutron transmission have allowed the determination of total cross sections over a broad energy range, as well as the treatment of total-scattering data, especially for hydrogenous systems [9]. Moreover, (n,$\gamma$) resonance dopplerimetry for incident neutron energies between 1 and 100 eV creates the opportunity to explore nuclear quantum effects in heavy atomic species with a much-higher count-rate and spectral resolution than DINS [7]. PGAA has the potential of providing enhanced capabilities in the quantitative determination of chemical composition [10]. VESUVIO+ offers the possibility of expanding the remit of PGAA beyond its implementation in reactor-based neutron sources use by exploiting the inherent time resolution of pulsed spallation neutrons over a wide range of incident energies. These improvements in instrumentation have also been accompanied by advances in data analysis and computational materials modelling. The first task has now been largely achieved with a new data-analysis package developed within the MANTID framework [7, 11], including the use of Bayesian inference methods [12]. Ongoing efforts also aim to develop *ab initio* techniques to predict neutron-scattering observables across the epithermal and thermal regimes, as recently illustrated in Ref. [14]. Overall, these developments provide a suitable testbed for the subsequent development of a fully fledged ETNA, schematically shown in the right panel of Fig. 1 and explained in more detail in Ref. [2]. The order-of-magnitude increase in count rates and underlying information content possible with ETNA will enable the full exploitation of epithermal neutrons in materials research, particularly in studies requiring parametric scans as a function of an external variable or across a set of specimens within a given family of materials.

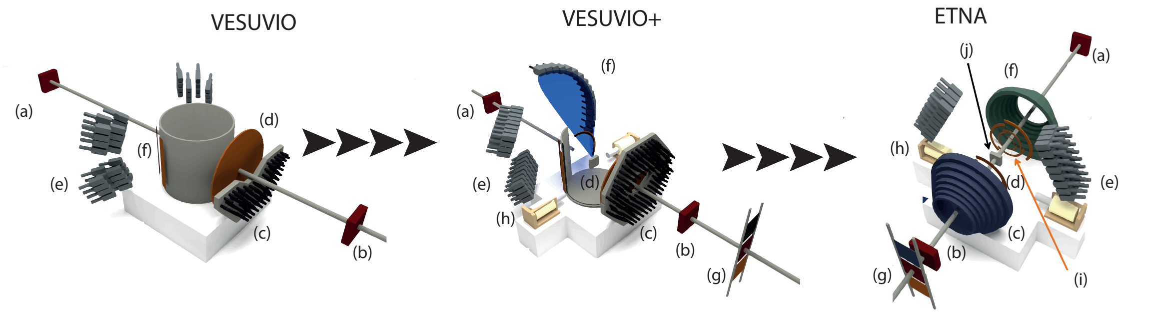


**Figure 1.** From VESUVIO to ETNA. Left: VESUVIO. Middle: VESUVIO+, currently under development. Right: ETNA. Beamline components: (a) transmitted and (b) incident beam monitors; (c) backscattering Li-glass neutron detectors; (d) backscattering Final Neutron Energy Analyser Mechanism (FNEAM); (e) forward-scattering YAP detectors; (f) forward scattering FNEAM; (g) incident foil-changing mechanism; (h) Ge detectors for PGAA; (i) FNEAM in Debye-Scherrer geometry; (j) sample position and concurrent *in-situ* techniques such as Raman spectroscopy [13]. See text for details.

## 2. A Glimpse at the Future

To illustrate the enhanced capabilities of VESUVIO+, we present a case study on $Mo_{4,5}Nd_{12,5}B_{20,8}O_{62,2}$ (MoNdBO), a rare-earth molybdate phase characterised by a complex stoichiometry involving both light and heavy nuclides [15]. Figure 2 shows the neutronic response of MoNdBO at Room Temperature (RT) measured in a simultaneous fashion using:
(a) MAss-selective Neutron SpEctroscopy (MANSE) in backscattering geometry;
(b) (n, $\gamma$) resonance dopplerimetry for Mo and Nd, using the forward-scattering YAP detectors;
(c) high-resolution powder diffraction.

The main panel in Fig. 2 demonstrates the use of MANSE to isolate the spectral response of heavy nuclei in a complex material well beyond previous attempts restricted to atomic masses below 25 amu [16–21]. As a first step, a high-precision instrument calibration was performed, extending previous protocols [22]. This procedure involved a Le-Bail fit to Pb-powder diffraction data [Fig. 2 (c) – inset], followed by a full Rietveld refinement, both using JANA2006 [23]. The result of the Le-Bail refinement is shown in Fig. 2 (c). A monoclinic unit cell was found (space group P21/c), with lattice parameters a $= 10.0425 \pm 0.0011$, b $= 4.1630 \pm 0.0003$, c $= 11.9077 \pm 0.0009$ Å, $\alpha = \beta = 90$ degrees, and $\gamma = 116.462 \pm 0.005^o$.

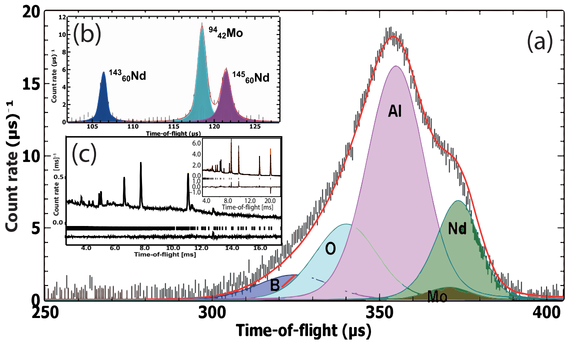


**Figure 2.** VESUVIO+ data for MoNdBO at RT, all measured simultaneously: (a) MANSE data; (b) (n,$\gamma$) resonances from Mo and Nd; and (c) powder-diffraction patterns. See text for details.

The (n, $\gamma$) resonances of $^{143}_{60}$Nd, $^{94}_{42}$Mo, and $^{145}_{60}$Nd were fitted with Voigt line shapes [22]. In fitting, the peak positions and Lorentzian line widths were fixed at $55.3 \pm 0.4$, $44.7 \pm 0.4$, and $42.5 \pm 0.3$ eV, and 112, 150, and 411 meV, respectively [24], whereas Gaussian Doppler widths were fitted and yielded NMD widths of $47 \pm 4$, $27 \pm 2$, and $55 \pm 5$ Å$^{-1}$. The Mo NMD width was then fixed to 27 Å$^{-1}$ in fitting the MANSE data of largely overlapping Mo and Nd recoil peaks. MANSE data fitting yielded NMD widths of $8.6 \pm 0.5$, $10.1 \pm 0.2$, $14.0 \pm 0.1$, and $47.5 \pm 1.1$ Å$^{-1}$ for B, O, Al, and Nd, respectively. The Nd NMD widths obtained from fitting (n,$\gamma$) resonances and MANSE data agree, thus validating the methodology adopted to model these complex data.

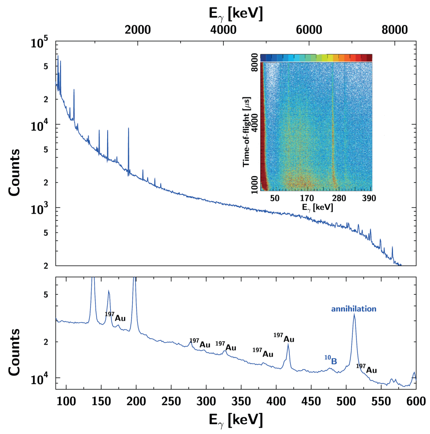


**Figure 3.** PGAA and multi-dimensional TPGAA spectra on an ancient golden coin. Upper panel: PGAA data over the $\gamma$-energy range 100 keV $\leq E_\gamma \leq$ 9 MeV. Lower panel: 100 keV $\leq E_\gamma \leq$ 600 keV, with labels indicating specific features associated with $^{197}Au$, $^{10}$B, and electron-positron annihilation. Inset: TPGAA spectrum in the time-of-flight range between 1000 and 8000 $\mu$s.

As a second example, we present PGAA and Time-of-flight-resolved multidimensional PGAA (TPGAA) [25] capabilities for simultaneous chemical and isotopic analysis, currently under development. In particular, the epithermal and thermal fluxes available on VESUVIO+ offer the potential for the further development of TPGAA, thus providing innovative ways to enhance the sensitivity and isotopic selectivity to intermediate- and heavy-mass isotopes using resonance scattering in the epithermal region. As an example, Fig. 3 reports PGAA and TPGAA spectra on an ancient golden coin. Spectra were taken with a high purity germanium detector using a coaxial close-ended N-type crystal of 8 cm diameter and 16 cm height. Detector resolution was tested using $^{60}$Co source and assessed to be 3% in energy corresponding to 3 keV at the

$^{60}$Co peak in the 1.33 MeV $\gamma$ energy spectrum. The TPGAA acquisition electronics chain was composed of a digitizer CAEN DT5724. Each channel of the digitizer was provided with a SRAM memory organized in a programmable number of circular buffers. When all buffers were filled, the board was considered full, that is, no trigger was accepted and the acquisition stopped. When the acquisition restarted, no trigger was accepted until at least the entire buffer was written. This means that the dead time was extended for a certain time (depending on the size of the acquisition window) after the board had exited the full condition. The dead time was estimated to fluctuate between the 10% and the 30%. There was a blind zone for the first 200 $\mu$s of each time-spectrum due to the high count rate and the difficulties for the digitizer to be as fast as needed to record all the signals. The digitizer could register 100 MS/s.

## 3. Outlook and Perspectives

VESUVIO+, an important stepping stone to develop ETNA, has been described. It already offers enhanced capabilities for the analysis of concurrent MANSE and diffraction data. In addition, the use of (n,$\gamma$) resonances and TPGAA enables a detailed characterisation of complex materials using both thermal and epithermal neutrons. The two examples presented in this work demonstrate the largely untapped synergies across a variety of experimental techniques, with a view to increasing the information content of a given experiment relative to what is currently possible using more conventional neutron-based methods.

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