Epithermal neutron instrumentation at ISIS

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Abstract. The advent of pulsed neutron sources makes available high epithermal neutron fluxes (in the energy range between 500 meV and 100 eV). New dedicated instrumentation, such as Resonance Detectors, was developed at ISIS spallation neutron source in the last years to apply the specific properties of this kind of neutron beam to the study of condensed matter. New detection strategies like *Filter Difference* method and *Foil Cycling Technique* were also developed in parallel to the detector improvement at the VESUVIO beamline. Recently, epithermal neutron beams were also used at the INES beamline to study elemental and isotopic composition of materials, with special application to cultural heritage studies. In this paper we review a series of epithermal neutron instrumentation developed at ISIS, their evolution over time and main results obtained.

Detectors for spectroscopy

The advent of pulsed neutron sources, such as the ISIS spallation source, has made available relatively large fluxes of neutrons in the *epithermal* energy range between 500 meV to 100 eV. Such energy range is usually not available in reactor-based neutron sources and even in pulsed facilities most of the beamlines are subdued to neutron moderators made to provide a beam of thermal (about 25 meV) neutrons. Only in recent times the possibilities given by epithermal neutrons have generated increased interest in the scientific community, due to the fact that neutrons in the lower part of the epithermal range provide information about atomic and molecular phenomena with characteristic energies of some eV, previously of difficult access through traditional probes. Consequently, new research paths for the investigation of certain aspects concerning condensed matter were opened, as for instance the short time single particle dynamics in quantum and molecular systems and high-energy excitations in materials. Since 1985 the ISIS spallation neutron source has been providing opportunities for research and development in experimental instrumentation and in new detection strategies to extend the kinematical region accessed by neutron scattering experiments to exchanged momenta $\hbar q \leq 10$ Å^{-1} and $\hbar q > 200$ Å^{-1} coupled with exchanged energies $\hbar\omega \geq 1$eV.
The VESUVIO spectrometer [1, 2] at ISIS was designed and built by British and Italian scientists for inelastic neutron scattering (INS) at such energy and momentum transfers ranges; it has been operational at the ISIS pulsed neutron source since September 2001. This instrument was an upgrade of the previous eVS spectrometer, that operate in the same energy range since 1985, pioneering inelastic neutron scattering at the eV energies in the last decades. The experimental technique

implemented at VESUVIO is known as *Deep Inelastic Neutron Scattering* (DINS). The VESUVIO Spectrometer used the high intensity of neutrons in the eV energy range (about 10^6 n/s cm^2 eV) and the pulsed nature of the ISIS source to measure atomic momentum distributions in a variety of condensed matter systems [3]. When the momentum transfer in the scattering process is sufficiently large (at least 10 times higher than the mean atomic momentum), the scattering can be interpreted within the *Impulse Approximation* (IA) framework. In IA scattering is essentially a single-atom "billiard ball" scattering, entirely determined by conservation of momentum and *kinetic* energy in the collision (molecular effects in the sample are irrelevant). By measuring the energy and momentum changes of the neutron, the momentum component of the target atom along the direction of the scattering vector *q* can be determined. Energy transfers in the 1-30 eV region and wave-vector transfers between 30 A^{-1} and 200 A^{-1} are achieved using a *filter difference* (FD) technique. Being VESUVIO an inverted geometry spectrometer [4], the energy of the scattered neutron is fixed by a nuclear resonance absorption foil; the incident energy, and hence the energy and momentum transfers, are determined using standard time of flight (TOF) techniques. The nuclear resonance absorption foil is a basic element for eV neutron spectroscopy in an inverted geometry instrument, in that it selects the final energy of the scattered neutrons. The experimental method used at VESUVIO consists of cycling the analyzer foil in and out of the scattered neutron beam. In this way, two measurements are taken for every sample, one with the foil between sample and detector and one with the foil removed, as shown in figure 1_a.

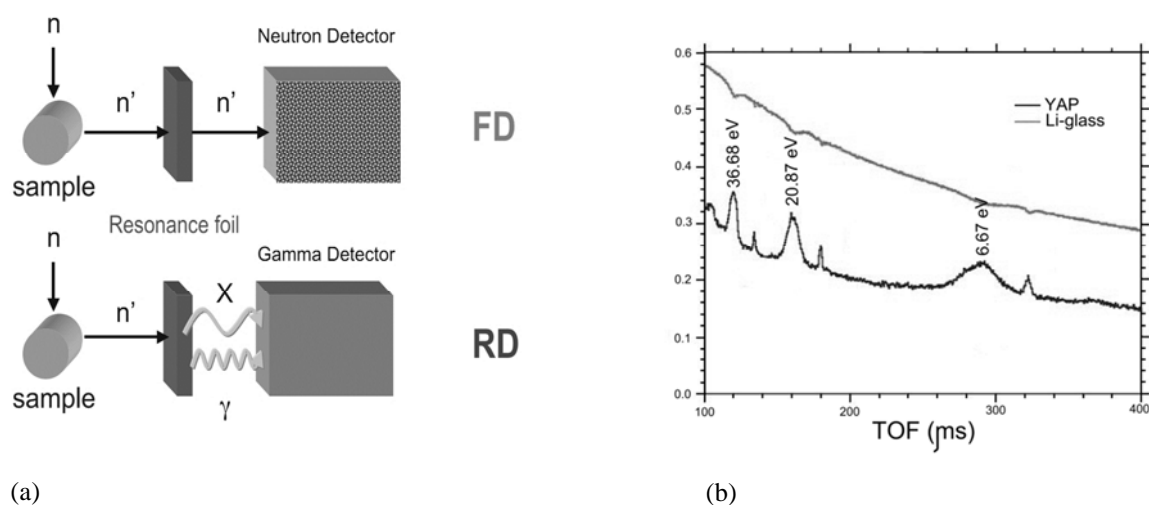(a)   (b)

Figure 1. (a) Principles of Filter Difference and Resonance Detector techniques. (b) Time of flight spectra for Filter Difference (Li-glass detector based) and Resonance Detector (YAP detector based) techniques.

The difference between these two data sets is indeed the experimental signal, i.e. a measurement of the intensity of neutrons scattered from the sample with final energy E_1. The upgraded VESUVIO spectrometer employs routinely ^{197}Au or ^{238}U foils as filter analyzers. The resonances of uranium are quite narrow compared to the resonance energy ($\Delta E_r/E_r \sim 0.5\%$), however the broadening due to the thermal motion of the lattice in which the absorbing nuclei are embedded, is an important contribution to the spectrometer resolution function; it was experimentally shown that the latter can be effectively reduced by cooling the filter foils.

   Resonance Detectors (RD) were developed for VESUVIO and replaced the Lithium-glass detectors previously used on the eVS spectrometer [5, 6]. In the case of Resonance Detectors, the detection of the prompt γ-ray radiation emitted by the absorber foil after resonant neutron capture was used (figure 1_b). Neutron resonance radiative capture is a physical process which occurs in two steps: i)

absorption of a neutron in a nucleus at a certain energy, mostly determined by the internal structure of the absorbing system and ii) prompt emission of a γ-rays cascade due to primary (from the absorbing nuclear energy level to the ground state) and multi-step radiative transitions [7]. The new RD set-up in VESUVIO was optimized and the performance of a new kind of photon detectors was tested including solid-state cadmium-zinc-telluride (CZT), a silicon detector based system, a Li-glass scintillator and yttrium aluminum perovskite (YAP) scintillators coupled with photomultipliers. The best configuration in terms of efficiency and signal to background ratio enhancement resulted to be YAP inorganic crystal coupled with a photomultiplier [8, 9, 10, 11]. YAP, indeed, is a fast, mechanically and chemically resistant scintillator material. Main characteristics of this material are a good light yield (about 18000 photons/MeV) and a short decay time of 27 ns for λ = 350 nm (wavelength of maximum emission). It is insensitive to neutrons and has a 100% efficiency at 100 KeV and 10% at 1 MeV. In figure 2 the TOF spectrum from an ice sample in an Aluminum cell in the time range 50-500 μs recorded through the YAP-based detector is shown as an example of the performances of the new system: details of the analysis of such spectrum are presented in ref. 12.
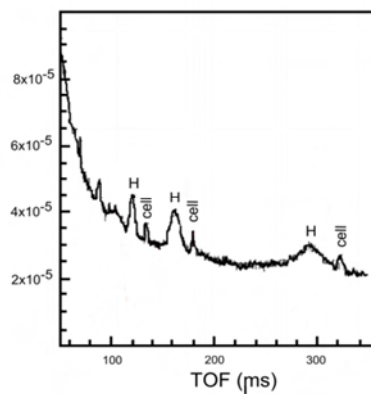

Figure 2. Time of flight spectra in the time range 50-500 μs recorded through the YAP based detector (sample: ice in an Aluminum cell). Four Hydrogen resonances are present and could be analyzed.

Research about Resonance Detectors improved the performance of the VESUVIO spectrometer by providing a high-resolution *Forward Scattering Detector* (FSD) bank and a *Very Low Angle Detector* (VLAD) bank (figure 3_a and 3_b). VLAD was designed to perform *High energy Inelastic Neutron Scattering* (HINS) measurements in the region of the kinematical space *(q, ℏω)* characterized by $1.5 \text{ Å}^{-1} \leq q \leq 10 \text{ Å}^{-1}$ and $0.3 \text{ eV} \leq \hbar\omega \leq 20 \text{ eV}$, thus complementing the capability of the instrument beyond DINS measurement. The VLAD configuration equipment has proven to be effective up to final neutron energies above 100 eV [13, 14, 15]. On VLAD the YAP crystals are given a trapezoidal shape in order to cover a large area (about 1075 mm^2) in the same angular range (figure 3_c).
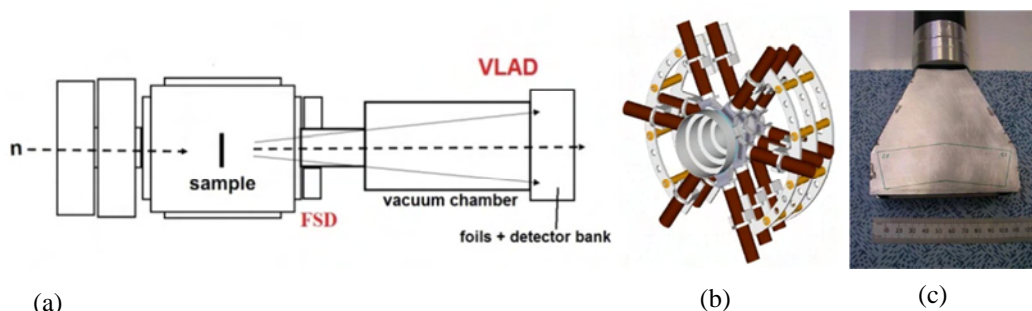

(a)   (b)   (c)

Figure 3. (a) VLAD and Forward Scattering Detector (FSD) set up on VESUVIO beamline; (b) scheme of VLAD detector bank; (c) VLAD detector: the YAP crystal is shaped in a trapezoidal and connection with a photomultiplier tube, through optical coupling by a light guide system.

The particular crystal shape does not allow direct connection with a photomultiplier tube, so that the optical coupling of the active medium with the PMT is provided by a light guide system therefore ensuring adequate light collection. The FSD bank was also developed and installed on VESUVIO (figure_4).

Figure 4. Photograph of Forward Scattering Detector bank at VESUVIO beamline.

A preliminary set-up was made of two detector banks with 32 scintillators (eight rows of four scintillators each) placed on either side of the beam pipe. With such configuration the bank presented a noticeable cross-talk between detector elements and the isotropic emission of the gamma rays was a source of corruption of the angular information. To solve the issue an optimized set up was developed, increasing the distance between the scintillators and sixteen new modules with four scintillators each were mounted vertically (see figure_4). The FSD cover a range of angles from 33^0 to 67^0.

A further improvement to the beamline design was the application of a *Foil Cycling Technique* (FCT). The FCT uses two foils of the same neutron-absorbing materials; the first foil is placed in front of the gamma detector and is used as the energy *analyzer* for RD configuration; the second is the cycling foil that is placed between the sample and the analyzer. During measurements, the foils are cycled in and out the scattered neutron beam and two measurements are taken (*foil-in* and *foil-out*) combining the RD and the FD techniques. Final data are obtained as the difference between the two measurements.

1. Detectors for other applications: Neutron Resonance Transmission and Neutron Resonance Capture Analysis imaging techniques

Epithermal neutron beams can also be used to study materials in terms of elemental and isotopic composition identification. *Neutron Resonance Capture Analysis* (NRCA) uses the unique resonance absorption properties of epithermal neutrons. Many elements have neutron absorption resonances in the energy range below 500 eV. Their absorption cross-sections are well known and vary from one element to another and actually also between isotopes of the same element, thus a resonance is a sort of fingerprint of a chemical element and of its isotopes [16]. Neutron absorption is often followed by the prompt emission of a gamma-ray cascade, with typical total cascade energies up to about 8 MeV. Being a non-destructive, non-invasive technique, thus especially suited to samples that cannot be damaged, NRCA has been used for analysis of archaeological artifacts to determine elements' composition [16, 17-25].

In the framework of the *ANCIENT CHARM* (*Analysis by Neutron resonant Capture Imaging and other Emerging Neutron Techniques: new Cultural Heritage and Archaeological Research Methods*) project, funded by the European Community "New and Emerging Science and Technology" program, new imaging techniques based on improvements of the NRCA approach were developed at ISIS. A

Neutron Resonant Capture Imaging combined with Neutron Resonance Transmission (NRCI/NRT) set-up was installed on the INES beamline at ISIS (figure_5).
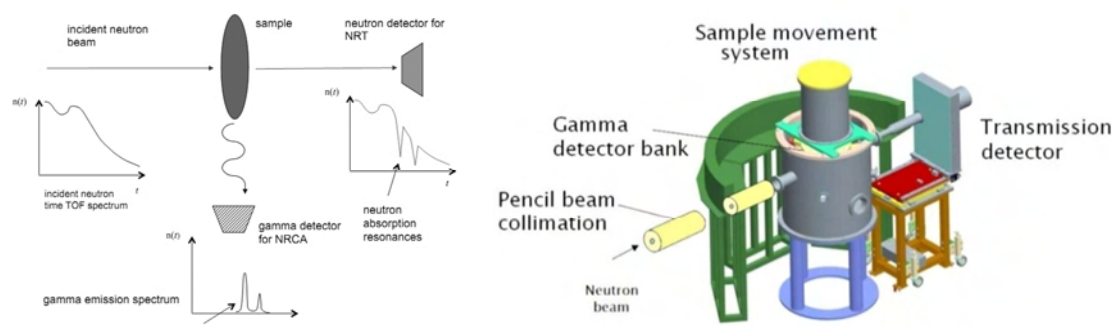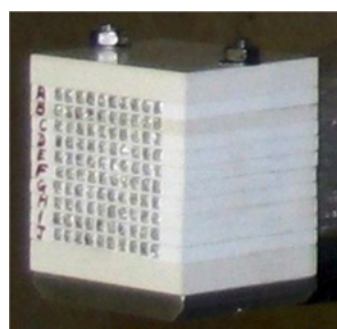

Figure 5. Neutron Resonant Capture Imaging combined with Neutron Resonance Transmission (NRCI/NRT) set-up installed at INES beamline, ISIS.

NRCI/NRT uses both gamma emission and neutron transmission measurements of resonant epithermal neutrons to determine the 3D elemental composition of the sample. The time of flight technique allows to recognize the resonance energies through the positions of the absorption peaks in the time spectrum. While the time position provides qualitative information (i.e. the nature of the element), the peak area provides quantitative information on the amount of the specific recognized element within the sample. Typical detection limits for NRCA are down to concentrations of 10^{-2}–10^{-4}%. In the case of NRT detection limits are worse around 10^{-1}–10^{-2}%.

The set-up for NRCI uses a 5 mm pencil beam of epithermal neutrons and an array of 28 YAP detectors mounted in a bank made of 3 support rings of cast lithium carbonate, surrounding the sample position. Each detector has a circular crystal front face with a 50 mm diameter and 25 mm depth (figure 6_a).

(a)

(b)

Figure 6. (a) Neutron Resonant Capture Imaging detector array; (b) Neutron Resonance Transmission detector.

The whole gamma detector bank is installed inside the INES aluminum tank. The pencil beam collimation was obtained through two sections (400 mm long and 150 mm long), each made of 50% Li_2CO_3 and 50% epoxy resin and installed inside the INES shutter, approximately 3 m upstream from the sample position.

The NRT detector was installed in transmission. The transmission geometry allows, in principle, to use a position-sensitive neutron detector and a rotating/translating sample holder to achieve a full 3D scan of the object under measurement. The detector developed for Ancient Charm project (figure 6_b)

consists of a 10x10 pixels array; each pixel is made of neutron-sensitive GS20 scintillator glass (6.6% total lithium of which 95% ^{6}Li) with an active area of 1.8×1.8 mm^2 and it is embedded in a 16 mm thick support made of boron nitride and separated by Al foils from neighboring pixels. The individual pixels are coupled via a 0.5 mm thick glass disperser to optical fibers which transport the light to photomultiplier tubes. Reference NRT measurements were performed on different samples to test the element-sensitivity of the imaging technique and some example images are shown in figure 7.
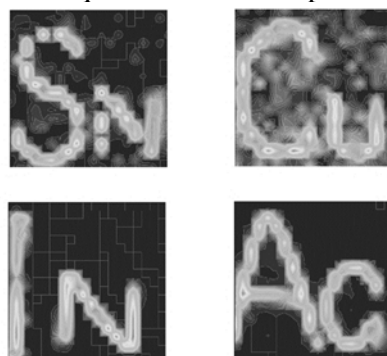

Figure 7. Spatial resolution in Neutron Resonance Transmission element sensitive imaging. Each square image has a side of 40 mm and the wires of different materials (tin-Sn, copper-Cu, indium-In, silver-Ag) are 1mm thick.

These are images of different 1 mm thick metal wires (tin-Sn, copper-Cu, indium-In, silver-Ag), superimposed and enveloped in a 1 mm thick Al foil. Wire shapes are clearly visible in the NRT images.

The NRCI/NRT set-up at INES was used on selected archaeological findings. In figure 8_a and 8_b a typical NRCA and NRT spectra are shown as an example of application to a realistic cultural heritage samples.

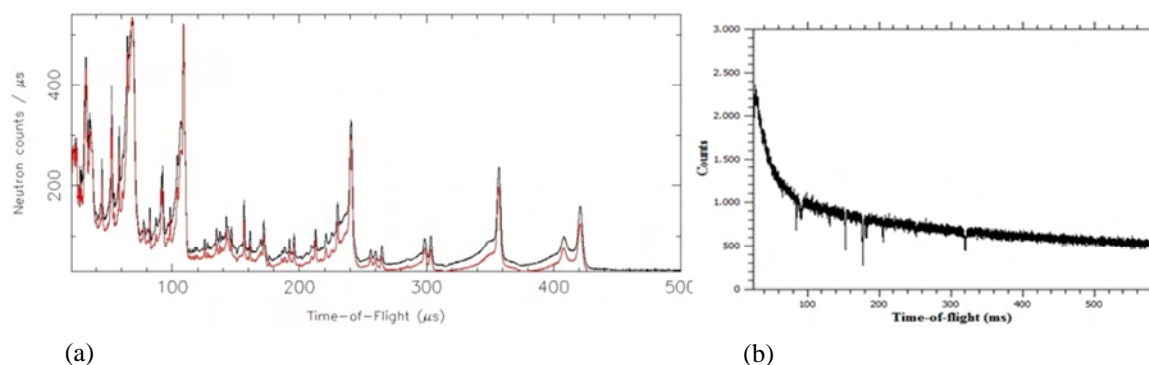(a)   (b)

Figure 8. (a) Spectrum from NRCA gamma detector of an archaeological interest sample; (b) Spectrum of NRT detector of an archaeological interest sample.

Shown spectra are related to the study of a gilded bronze head relief by the famous *maestro* Lorenzo Ghiberti. The sample comes from the East doors of the Florence Baptistery, also known as *'The Gates of Paradise'* for which information about the gilding state was obtained [26].

Conclusions and future prospective

An overview on how the availability of epithermal neutron fluxes has improved the development of dedicated neutron instrumentation at ISIS spallation neutron source has been provided. Epithermal neutron beams were used to study kinematical properties of matter and composition of materials in

terms of both elemental and isotopic identification. In this framework, with the recent development of new instrumentation, new prospective is opened in different fields, for instance cultural heritage. Combined use of techniques like NRCI/NRT and the widely known Prompt Gamma-rays Activation Analysis [27] could increase - the possibilities of identifying "exotic" or unexpected elements and isotopes in Cultural Heritage samples. The detection of particular rare elements such as lanthanides or rare earths through mentioned imaging techniques could open a new generation of traceability strategies.

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