ELECTRON-VOLT NEUTRON SPECTROSCOPY, WITHER GOEST THOU?

S. Fletcher,[1] A. G. Seel,[1] R. Senesi,[2] F. Fernandez-Alonso[1,3]

[1] ISIS Pulsed Neutron and Muon Source, Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire OX11 0QX, United Kingdom

[2] Dipartimento di Fisica e Centro NAST, Università degli Studi di Roma Tor Vergata, Via della Ricerca Scientifica 1, 00133 Roma, Italy; Consiglio Nazionale delle Ricerche, CNR-IPCF, Sezione di Messina, Messina, Italy

[3] Department of Physics and Astronomy, University College London, Gower Street, London, WC1E 6BT, United Kingdom

In January 2014, scientists from across the world came together in Abingdon (United Kingdom) for a two-day meeting on the latest developments in electron-Volt neutron spectroscopy (eVs) [1]. This international workshop was jointly organised by the ISIS Pulsed Neutron and Muon Source, the Consiglio Nazionale delle Ricerche, the Università degli Studi di Roma Tor Vergata, and the Università degli Studi di Milano Bicocca. Interest in the use of epithermal neutrons for condensed matter dates back to the advent of proton-driven spallation neutron sources in the 1970s and 1980s, resulting in a first major scientific meeting at Los Alamos (USA) in 1984 [2]. Following these initial efforts, the first and second editions of the present eVs workshop series were held in Abingdon (United Kingdom) in May 1995 and October 1998, the third edition was held in Santa Fe (New Mexico, USA) in April 2005 [3], the fourth at Oak Ridge National Laboratory (Tennessee, USA) in 2006, and the fifth in Rome (Italy) in 2010 [4]. This sixth edition [1] provided a comprehensive and up-to-date overview of eVs, with particular emphasis on the current science and instrumentation program on the VESUVIO spectrometer at ISIS [5]. The event also represented an opportunity to bring together experimental and theoretical communities interested in nuclear quantum dynamics in condensed matter and the application of eVs techniques in materials science. In addition to talks and Q&A sessions, the meeting was wrapped up by a two-hour discussion to delineate the way forward in the short, medium, and long terms.

Figure 1. Workshop participants during a brief lapse of (well-deserved!) orderly respite

The use of epithermal neutrons for high-resolution spectroscopic studies remains a unique feature of short-pulse, accelerator-driven spallation neutron sources. Much of the progress in the field is owed to joint and sustained research & development efforts at ISIS by the Italian and British communities over the past couple of decades, including novel detection methods,

data-analysis tools and protocols, not to forget emerging applications beyond fundamental condensed-matter research. In this context, VESUVIO remains the only neutron spectrometer of its kind in the world and has seen recent and significant improvements of its capabilities, leading to substantial gains in accuracy and reproducibility. This meeting provided a timely platform to discuss these developments in the context of current scientific trends.

The opening session focused on the quantum behaviour of hydrogen and deuterium as seen via nuclear-momentum-distribution measurements, an area of research that continues to be at the heart of the science programme on VESUVIO. Recent collaborative studies between experimentalists and theoreticians were presented, primarily aimed at a quantification of nuclear quantum effects in aqueous and hydrogen-bonded systems, including cases of extreme spatial confinement. These presentations provided a firm starting point for subsequent discussions on the underlying nature of eVs observables, including the analysis and interpretation of Compton profiles in both ordered and disordered media. On a closely related front, these experimental studies also offer a direct link to the output of path-integral molecular dynamics simulations, yet it was also acknowledged that much remains to be done in order to facilitate a direct comparison between experimental data and first-principles predictions. The second session detailing ongoing studies on heavier nuclides followed naturally from the above, and included lithium- and oxygen-containing binary and ternary systems, as well as complex materials such as biocements. Falling under the umbrella of MAss-selective Neutron SpEctroscopy (MANSE), these more recent studies served to examine current capabilities and limitations on VESUVIO, as well as to explore future opportunities for MANSE in the study of heavier atoms, a new frontier for eVs with applications to the wider materials and applied sciences.

Figure 2. Dan Major (Bar-Ilan University, Israel) explains the importance of nuclear-quantum effects in enzyme catalysis

The last session of the first day was necessarily devoted to developments in eVs instrumentation, taking as starting point the uniqueness of VESUVIO on the global stage. The detection of epithermal neutrons remains a key area of research & development efforts at ISIS, as well as a superb illustration of fruitful and continued collaborative efforts between ISIS and its international partners, particularly the Italian community. Moreover, this session emphasized the yet-to-be-tapped potential offered by further technical developments on VESUVIO, so as to provide a step-change in experimental capabilities for detailed parametric and time-resolved studies, as well as much-needed enhancements in mass discrimination to expand the realm of applicability of the MANSE technique. Alternative and complementary epithermal neutron spectrometers such as MAPS at ISIS or SEQUOIA at the SNS were also brought to the fore and discussed in the context of current and future scientific needs and challenges. Beyond neutron instrumentation per se, new capabilities to treat eVs data within the MANTID framework [6] were also presented, leading to a subsequent discussion on the needs of the community in the provision of data-reduction and analysis tools so as to facilitate the interpretation of experimental data.

The second day focused on theoretical approaches, associated state-of-the-art computational tools, and their application to a variety of phenomena including collective proton tunnelling and ferroelectric behaviour in hydrogen-bonded media or enzyme catalysis. As illustrated by a number of international speakers, atomic momentum distributions are of great interest and relevance to theorists developing new methodologies for molecular and condensed-matter dynamics, a high-profile area in contemporary chemistry and physics. The ultimate aim of this line of research is to predict and quantify non-trivial effects associated with the quantum behaviour of the lighter elements in condensed matter, from hydrogen and its isotopic cousin deuterium to lithium, boron, carbon, nitrogen, or oxygen. Whereas this work is fundamental in nature, it is also transformational as it questions the very essence of our current understanding of ubiquitous phenomena such as proton transfer in aqueous and biological media or charge and mass transport in materials for energy applications. Likewise, these conceptual and methodological developments in first-principles simulations can also provide fresh insights into the properties of increasingly complex materials amenable to investigation with eVs.

The second day ended with a lively discussion on perspectives and challenges for eVs techniques. The discussion emphasised the central role that VESUVIO continues to play in joint experimental and theoretical efforts to explore and unravel the essence of nuclear quantum dynamics in condensed matter. This discussion session, as well as the questions posed to all speakers throughout the workshop, were documented and will form a set of conference proceedings. The attendees amply demonstrated that the science undertaken on VESUVIO

could be conveniently subdivided into an unabated interest in fundamental nuclear-quantum effects associated with particle delocalisation and interference, and emerging applications of MANSE in more practical scenarios. These two complementary areas were discussed in terms of future prospects, highlighting both systems to which each could contribute, as well as the role and importance of parallel efforts in computational modelling. The room may have been filled with experimentalists and theoreticians, yet it was abundantly clear that participants were certainly not confining themselves to their own communities!

More details of the conference programme including copies of the presentations can be found in Ref. [1] below. Conference proceedings will be published in a thematic volume in the open-access journal *Journal of Physics: Conference Series*. Based on the extensive input from all participants, these proceedings will follow a 'Faraday Discussion' style, including a detailed record of all discussions held, associated conclusions, and future prospects.

.....

References

[1] www.isis.stfc.ac.uk/news-and-events/events/2014/
[2] www.osti.gov/scitech/biblio/5858602 and www.osti.gov/scitech/biblio/5860709
[3] www.fisica.uniroma2.it/~vesuvio/dins/html/eV_workshop240405.htm
[4] web129.its.me.cnr.it/workshop/
[5] www.isis.stfc.ac.uk/instruments/vesuvio/
[6] www.mantidproject.org/