Neutron Transmission: A Very Powerful Technique at Small Accelerator-based Neutron Sources.

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Abstract. The determination of total cross section of a material as a function of neutron energy by means of transmission experiments is rather easy on a pulsed neutron source. The spectrometer and associated instrumentation required may be described as relatively simple, and sound statistical data are achieved even using a small, accelerator-based neutron source. Despite its simplicity, detailed scientific and technological information can be extracted from such measurements, at the cost of fairly elaborate data analysis techniques. These features make this an attractive tool for neutron-based materials research within small research groups.
Different physical properties of a material manifest over the different neutron energy ranges. At our group we have focused on the determination and analysis of the total cross section for thermal and sub-thermal neutrons, which is very sensitive to the geometric arrangement and movement of the atoms, over distances ranging from the 'first-neighbour scale' up to the microstructural level or 'grain scale'.

1. Introduction

The unique properties of thermal neutrons for the investigation of condensed matter have been recognized from the early days when neutron sources became available for research purposes. Since then, much work has been done on both experimental and theoretical aspects to fully exploit those properties, thus transforming their potentiality into a very powerful technique for the study of the structure and dynamics of matter [1].

Besides the traditional field of atomic motion studied in liquids and solids, the availability of new powerful neutron sources, advanced moderators and specialized instruments has opened new horizons for the application of neutron techniques [2]. Whether they involve the use of cold, thermal, or epithermal neutrons, large portions of the energy- and momentum-transfer space can now be explored for a range of systems and problems, from basic studies in biology to material integrity in industrial products. Yet those techniques are still limited by the intrinsic low-intensity of the sources, and in many circumstances the study of small samples cannot be performed with the required statistical accuracy.

One of the most basic of all neutron techniques is the total cross section $\sigma_T(E)$, as obtained from transmission experiments. The behavior of the total cross section of a given system as a function of the incident neutron energy provides very valuable information on its structure and dynamics. Even though fairly elaborated procedures for data analysis may be necessary to extract that information, the essentially simple and clean transmission measurements make this technique and attractive tool for materials research using even a small accelerator-based pulsed neutron source.

On the other hand, and although fewer groups are now involved in the determination of thermal neutron constants, there is still a need for improved data in this energy range, as more accurate cross section values are required in many research fields where neutron techniques are employed. That includes more precise values of the isotopes' scattering lengths, and the nuclear constants required in the development of new reactor concepts as well as in some nuclear engineering problems related to fuel management and reprocessing. Moreover, it has been demonstrated that information about some fundamental properties of the neutron itself, can be obtained from high precision total cross section data on the thermal and epithermal neutron energy regions [3,4].

In this contribution we will describe our experimental facilities, based on an electron LINAC, together with our current research topics, focused mainly around structural nuclear materials and moderators. Besides this, we will present current efforts aimed at expanding the variety of problems feasible to be studied by neutron transmission experiments. These efforts comprise experimental work for reducing experimental counting times, and maybe more importantly, theoretical and programming work aimed at producing a powerful data-analysis program for efficient materials characterization.

2. Neutron Source and Experimental Set-up

At the Neutron Physics Division of Centro Atomico Bariloche (Argentina), we have been performing transmission experiments for more than 35 years, using an Electron LINAC based neutron source, in combination with time-of-flight (TOF) techniques. Electron pulses (1.4 μs width) are accelerated up to 25 MeV, before hitting a water-cooled lead target, where bremsstrahlung radiation is produced and the induced (γ,n) reactions give rise to a fast neutron pulse. Typical repetition rates are 12.5 , 25, 50, and 100 pulses per second, with an average current corresponding to the latter frequency of 25 μA at the target. Different types of moderator systems are used depending on the specific requirements of a given experiment, optimized for production of subthermal, thermal, or epithermal neutrons, as well as in terms of the pulse-width that controls the TOF resolution. To accomplish that goal we have used throughout the years paraffin at liquid Ni temperature, light water, polyethylene and polypropylene as moderation materials, in the form of thin and thick slabs, without any decoupler or with 2-D (sandwich-type) or 3-D (grids-type) inhomogeneous poisoning.

In our transmission facility we have two detector stations, located at 8m and 17.9m from the neutron source, where the detection bank, consisting of seven ^{3}He proportional counters (10 atm. filling pressure, 6” active length, 1” diameter) can be placed. The sample changers intersect the evacuated flight path at 4m and 8m from the neutron source, respectively. The sample-in/sample-out method is normally employed to minimize the effect of beam power fluctuations, and the raw data are corrected for dead time, mean emission time of the neutron source, and background. In FIG. 1 we show the lay-out of our facility.

3. A Few Scientific and Technological Applications

This section describes a few examples where total cross section measurements were performed in order to obtain fundamental information on some basic and applied studies. In most cases they correspond to experiments done using our facility, but we will also mention others performed elsewhere to illustrate the range of possibilities that the technique is able to support.

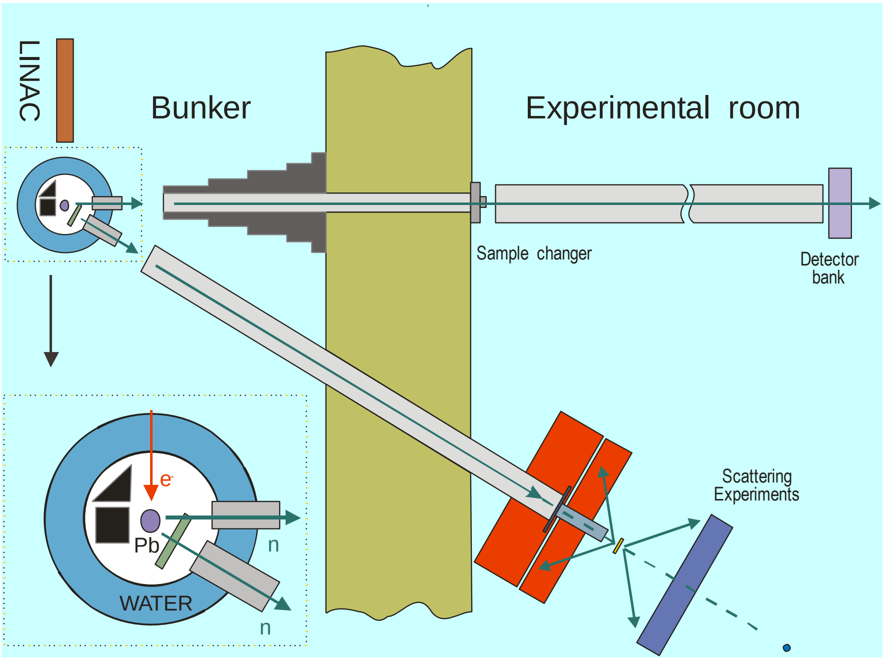

FIG. 1. Lay-out of the Bariloche pulsed neutron facility.

3.1. Total Cross Sections of Molecular Systems

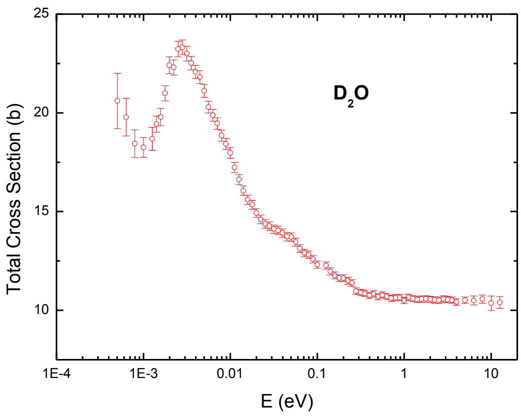

FIG.2. Total cross section of D_2O at room temperature over the thermal energy range.

Thirty five years ago the total cross section of D_2O was not known with the required accuracy, and that problem was tackled in our lab as one of the first applications of the then recently built facility. A moderator consisting of a paraffin block cooled down to liquid nitrogen temperature was used, in combination with a low repetition rate, to achieve statistically significant data below one millielectronvolt. Mixtures of light and heavy water were also measured, in order to obtain the total cross section of the molecular species HDO, to subtract its contribution to the real heavy water transmission and eventually determine the total cross section of D_2O at room temperature. The result is shown in FIG. 2 [5].

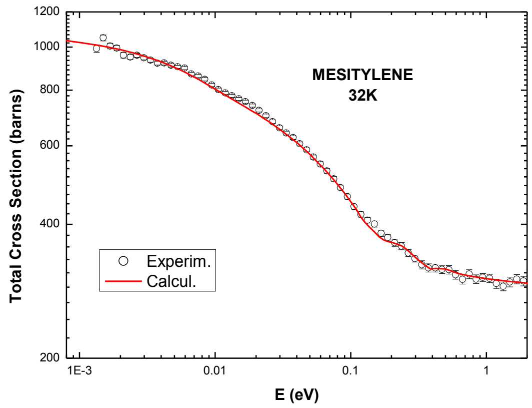

FIG.3. Total cross section of Mesitylene at 32K, comparing experimental data with calculation.

After the development of a Synthetic Scattering Model to describe the interaction of slow neutrons with molecular systems [6], we used total scattering cross section measurements to refine some of its parameters before generating the scattering kernel for a material, as it is shown in FIG. 3 for Mesitylene at 32K [7]. Conversely, that kind of measurements represent the first validation test for any calculation.

3.2. Determination of Scattering and Absorption Cross Sections

In the mid 70's there was a lack of information on Mo, an element considered of importance as structural material in reactor design. In addition, the values of the nuclear constants σ_c , σ_i , σ_{bound} , quoted in the literature showed some inconsistencies between them. We performed an experiment in order to get a set of consistent parameters for scattering and absorption through a measurement of total cross section. The experimental curve for polycrystalline molybdenum is shown in FIG. 4, and was fitted theoretically in the whole energy range using our code CRIPO [8], from which all the looked after nuclear constants were obtained. This procedure was used for several elements and isotopes.

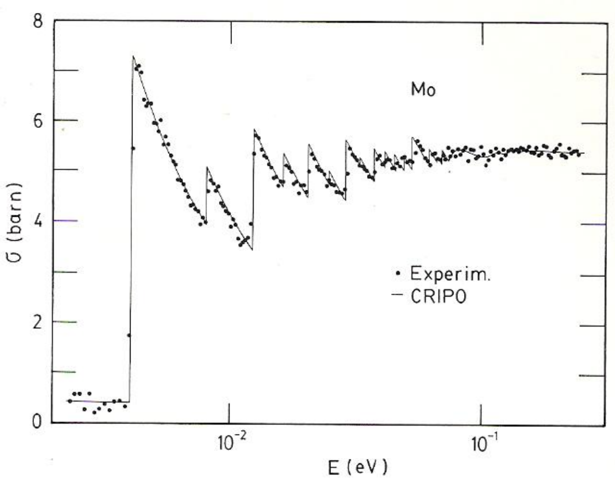

FIG. 4. The total cross section (absorption subtracted) of Mo in the thermal neutron range.

3.3. Condensed Matter Studies

Once the absorption cross section is subtracted from $\sigma_T(E)$, the remaining total scattering cross section $\sigma_s(E)$ contains information relative to structural and dynamical properties of the target system. In the following paragraphs we mention just a few examples where that information was obtained.

a) Structure of Condensed Systems

As it was already shown in FIG. 4, the structure of a polycrystalline material is clearly reflected through the Bragg edges, and from that pattern the lattice structure and unit cell parameters are obtained. Although the latter will in general be not as accurate as those obtained from a conventional diffraction measurements, there are many circumstances in which either the sample size, or the extreme conditions required, or the need to follow some rapid evolution through the phase diagram, may demand the use of more intense incident and outgoing neutron fluxes. Also, it was demonstrated many years ago that transmission measurements can provide very useful structural information in the case of amorphous systems [9], where the radial distribution function for silica obtained from that technique was compared well with the one resulting from traditional diffraction experiments.

b) Main Dynamical Features

Even though the total scattering cross section is an integral magnitude, related to the Van Hove scattering function S(**Q**,ω) [1] by

$$\sigma(E) = \frac{\sigma_b}{4\pi} \int \frac{k'}{k} S(\mathbf{Q},\omega)\, d\Omega\, dE' \ ,$$

where $\hbar\mathbf{Q}$ and $\hbar\omega$ represent the momentum and energy transfer, respectively, some broad dynamical features can be extracted from the sample's transmission. A simple example is the determination of the Debye temperature θ_D of a polycrystalline material from the variation of the Bragg jumps caused by the Debye-Waller factor, under the hypothesis of a Debye model to describe the lattice dynamics. An usually more efficient and precise method to obtain θ_D , not limited to polycrystals, is related to the behavior of the inelastic contribution to $\sigma_s(E)$ which is dominated at low neutron energies by one-phonon processes [10].

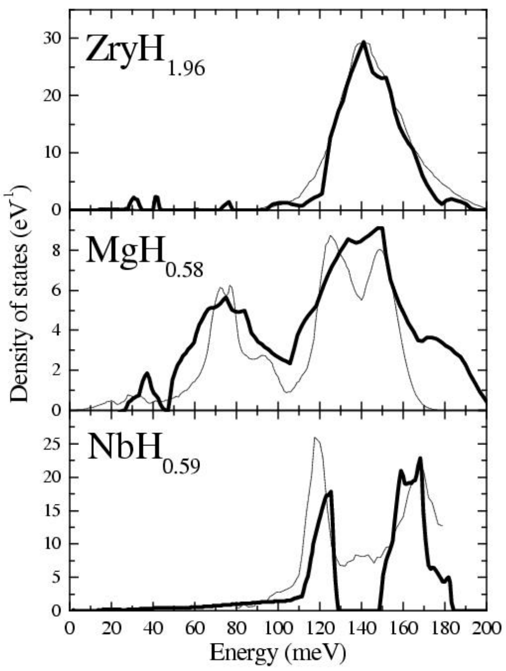

FIG. 5. Calculated densities of states from the total cross-section measurements (thick line). The dotted line are the results of inelastic scattering experiments.

In another class of systems, we showed that dynamic information can be obtained from total cross-section measurements, and a method was introduced to obtain the density of states (DOS) from the incoherent part of the inelastic total cross section [11]. For the application of this method, the elastic and multiphonon contributions must be known beforehand, to subtract them from the experimental data. This can be achieved by the approximation of the density of states through some simple model, such as a combination of a Debye spectrum to describe the acoustic modes plus Gaussian functions to account for the optic ones. We have applied this method in the case of three metal hydrides: zircalloy, magnesium and niobium, whose total cross sections were measured at a low-flux neutron source. Those DOS results, shown in FIG. 5, are in agreement with the inelastic scattering experiments performed at high-flux sources over the range from 40 to 250 meV.

c) Quantum Effects at Low Temperatures

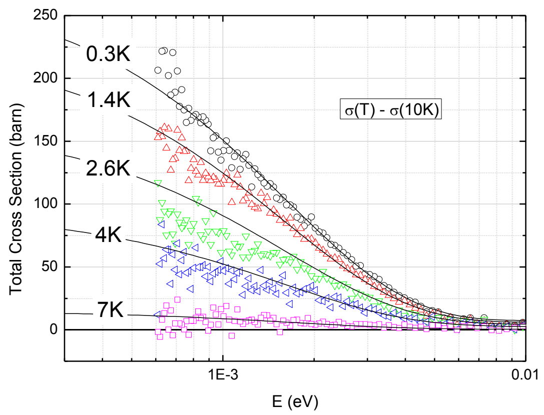

FIG. 6. Total cross section differences for Methane in phase II, between different temperatures and the values at 10K, as resulting from calculations [12] and experiment [13].

We have recently introduced a new model to describe the interaction of slow-neutrons with solid methane in phase II, that can be used to describe any molecular system under situations when spin correlations between its identical atoms are apparent [12]. In particular, we have indicated the strong dependence of the spin correlations effects with temperature, as displayed by the total cross section at low neutron energies. We show in FIG. 6 the difference between total cross sections at different temperatures with that at 10K, as obtained from calculated [12] and experimental [13] results, suggesting the use of that effect as a thermometer in some special circumstances.

3.4. Bragg Edge Measurements

The position and height of the Bragg edges appearing in the total cross sections of polycrystalline materials can be measured to high accuracy with the TOF technique [JRS1]. As the position and height of the edges depend on the same laws as the position and intensity of conventional diffraction peaks, information about the stress state [JRS2], texture [JRS3] or the phases present in the sample [JRS4] is readily available from an analysis of the transmission pattern. For multiphase materials, the total cross section displays Bragg edges from all crystallographic phases. Hence, the volume fractions of the constituent phases can be determined from an analysis of the height of the edges. Under certain circumstances, these measurements provides some advantages over conventional neutron diffraction: for relatively thick samples transmission patterns can be collected faster, as the transmitted count rate is high compared to the intensity diffracted into a certain solid angle. For high-intensity sources such as Isis, this allows studying kinetics of structural phase transitions in solids. FIGURE 7 shows an example of such studies performed on the ENGIN instrument at ISIS [JRS5]. An EN24 steel sample was austenised at 830°C furnace, and subsequently cooled to 380°C, and the total cross section was measured in-situ. The disappearance of the γ-(111) and γ-(200) edges and the appearance of the α-(110) are clearly seen in the figure. The inset shows the time evolution of the α and γ phases, obtained after a Rietveld analysis of the transmitted spectra using the code BetMan developed by Vogel [JRS6]. We are currently expanding this computer code to deal with textured materials.

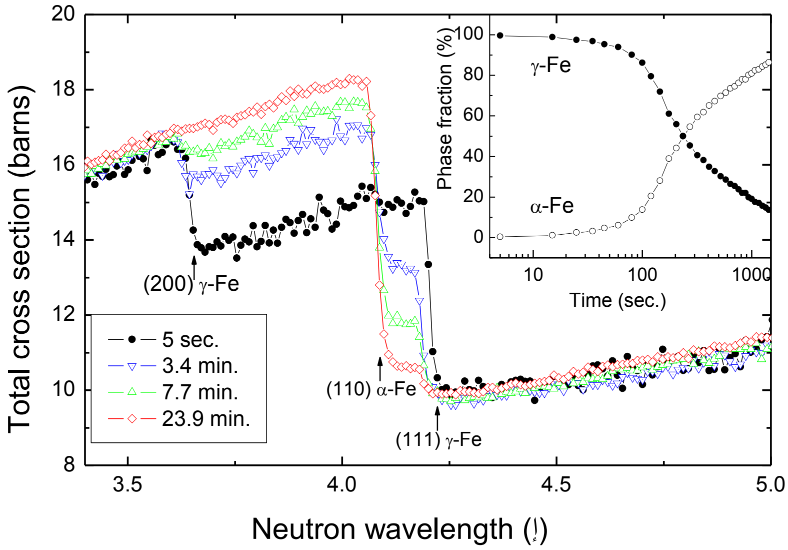

FIG. 7: Detail of the total cross section of EN24 steel at 380°C after austenisation at 830°C [18]. The times are measured from the time of insertion in the furnace. The inset shows the evolution of the phases, obtained after Rietveld analysis of 32 spectra.

3.5. Neutron Absorption Resonances

Transmission measurements to determine or refine neutron absorption resonances' parameters in nuclear fuel or structural materials are of interest by their own, on account of the importance of such type of information for Reactor Physics calculations. Besides, the development of more accurate yet simple algorithms to describe Doppler broadening effects on the natural line-width, able to cover the whole range of energies and temperatures [20], can be applied to the field of thermometry, improving and extending to the low temperature range the predictions that can be made with the use of the traditional effective temperature gas model.

3.6. Fundamental Neutron Interactions

The measurement of neutron total cross section for heavy elements over an energy range from zero up to several KeV, permits a determination of the neutron-electron scattering length and the neutron electrical polarizability [3,4]. We have used our facility and the KURRI Linac (Kyoto University, Japan) based pulsed source to perform transmission experiment on Pb and Bi, over epithermal neutron energies, together with a full calculation of the different electromagnetic interactions and their corresponding contributions to the scattering length [21]. From the high precision total cross section values (uncertainties varying from 0.2% to 0.3%), we were able to extract a value for the neutron's electric polarizability quite consistent with the best values obtained at that time.

4. Conclusions

In this paper we have reviewed some scientific and technological applications of neutron transmission experiments, including part of our research activities related to Nuclear, Neutron, and Condensed Matter Physics. A cold neutron source based on solid mesitylene is under construction in our facility, to tackle a number of interesting problems using longer wavelength neutrons. The main purpose of this contribution is to emphasize the range of situations in which the knowledge of the total cross section provides very useful information on a given system. Although the complexity associated to the integral character of this magnitude demands a careful accountancy of its different components, the experimental setup and data processing are essentially simple in nature, and that renders confidence on the measured values. In spite of its lack of details as compared with traditional elastic or inelastic experiments, both the structure and dynamics of matter can be revealed by the total cross section under appropriate conditions, and could be a unique tool to explore rapidly varying phenomena.

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