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Remote determination of sample temperature by neutron resonance spectroscopy

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Abstract

The Doppler broadening of the lower energy neutron absorption resonances of natural hafnium, tantalum, iridium and rhenium have been studied for the purpose of measuring temperature in remote or isolated environments. Three methods for the determination of sample temperature from neutron transmission data were studied and a critical comparison of the efficacy of each method made. Fitting the observed resonance line shapes with analytical expressions incorporating instrument resolution and resonance parameters provided the most accurate measure of sample temperature with an estimated uncertainty of ± 10 °C at 1000 °C and did not require prior calibration experiments to be performed.

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1. Introduction

The preferential or 'resonant' absorption of neutrons with specific energies by target nuclei may be readily observed as dips in an energyresolved neutron transmission spectrum. The characteristics of these resonances (shape, depth, and distribution) are unique to the isotope of the

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absorbing nuclei, and may be considered as a fingerprint for the isotope. This enables discrimination of the resonances associated with a specific isotope within a multi-element system. The energy of the neutrons 'observed' by the absorbing nuclei is significantly influenced by the velocity distribution of the nuclei. As such, the profiles of resonances are sensitive to the environmental variables that affect this velocity distribution, including the thermal motion of the nuclei and the effect of vibrational modes through lattice binding. Doping of a sample with a selected isotope or element therefore offers the opportunity of investigating the atomic environment of the dopant, albeit restricted to those environmental variables to which the neutron resonances are sensitive. In practice, this is limited to the temperature to which the dopant atoms are subjected [1,2]; however, a number of successful studies have been performed investigating characteristics of the phonon spectrum of tightly bound structures [3–5].

It was realised by Bethe in the late 1930s that individual neutron absorption resonances exhibit a broadening with increasing temperature, as a result of the Doppler effect from the thermal motion of the target nuclei as viewed in the laboratory frame [6,7]. However, it was not until the 1950s that it became possible to perform studies of absorption resonances with sufficient resolution to directly measure the Doppler broadening [8]. The exploitation of the Doppler broadening of resonances as a method for remotely determining effective atomic temperatures was first proposed by Fowler and Taylor [1]. In this work, a critical assessment of the technique, its potential merits and uses were discussed: a comparison of isotopes with candidate resonances was also presented. For practical applications the technique requires the use of high flux neutron spallation sources. These sources offer access to neutrons within the requisite energy range and with the necessary resolution. Historically, restrictions on access to such facilities and the non-trivial nature of interpreting neutron resonance data, temperature determination by this neutron resonance spectroscopy (NRS) has so far received comparatively little attention and has been limited to studies where more traditional techniques such as thermocouple-based measurements have proved prohibitive. Now, however, billion-dollar projects are underway to build next-generation neutron spallation sources (SNS in the United States and JSNS in Japan). The advent of these sources should dramatically improve the range and number of experiments that may be performed at such facilities and enable experiments using such techniques to become more common. Examples of investigations which have adopted this technique so far include: the measurement of the surface temperature of an operating catalyst [9], obtaining the effective temperature of Cu atoms in a $YBa_2Cu_3O_{7-\delta}$ superconductor [10,11], characterisation of the temperature behind an explosive shock front [12], and also determining the temperature within the sample chamber in a Paris-Edinburgh cell [13]. Prompted by the requirements of nuclear engineering, considerable efforts have been directed worldwide to the acquisition and compilation of reliable nuclear reaction data including absorption resonance parameters. These data are freely available through evaluated nuclear data files (e.g. ENDF/B-VI.8, JENDL-3.3, and JEFF-3.0). For nuclear engineering applications. the Doppler broadening of absorption resonances is simply an artefact of the governing physics, that must be accounted for in the analysis and design of nuclear systems and is of limited interest in its own right. We, however, are interested in exploiting this side effect to allow measurement of samples held in enclosed or inaccessible environments, in particular those encountered in experiments under extreme temperatures and pressures [13]. In order that this phenomenon may be successfully exploited as a tool for the determination of sample temperature in such applications, it is critical that reliable procedures are established by which the temperature may be extracted from the measured resonance line shape.

In this paper, an approach based on an analytical description of the resonance line shape is suggested for temperature determination from neutron resonance data. Comparisons are also made between this methodology and with temperatures obtained using empirical methods including; individual time channel analysis, suggested by Mayers et al. [2,14] and fitting with an arbitrary function and using a calibration curve, suggested by Le Godec et al. [13].

2. Determining sample temperature from resonance broadening

It is proposed that sample temperature be determined from transmission data by a methodology which directly appeals to the equations of the governing physics rather than employing empirical methods. This offers the considerable benefit over such empirical methods that a calibration experiment is not required prior to the acquisition of the desired data. In order to accomplish this it is necessary to calculate the absoprtion cross-section as a function of neutron energy in the vicinity of the resonance including the contributions from the intrinsic line shape of the resonance and the Doppler broadening. With this result, the transmission through the sample, accounting for the thickness of the sample and instrument resolution, may be obtained. These steps are detailed in Sections 2.1 and 2.2 below.

2.1. Theoretical neutron absorption resonance line shape

The reaction cross-section, $\sigma_r(E)$ for capture of a neutron, with kinetic energy, *E*, by a stationary nucleus close to the energy of an isolated resonance, E_R , may be adequately described by the single-level Breit–Wigner formula [15]

$$\sigma_{\rm r}(E) = \pi \lambda^2 g_J \frac{\Gamma_{\rm n} \Gamma_{\rm r}}{\left(E - E_R\right)^2 + \left(\frac{1}{2}\Gamma\right)^2} \tag{1}$$

where $\tilde{\lambda}$ is the de Broglie wavelength of the incident neutron, Γ_n is the neutron width, Γ_r is the partial width for the reaction and g_J is a statistical spin factor for the formation of a compound state with angular momentum, J. For narrow resonances, a reasonable approximation to the resonance width, Γ , is $\Gamma = \Gamma_n + \Sigma_r \Gamma_r$ [16]. However, practical investigations seldom encounter stationary nuclei and as such it is necessary to accommodate the velocity distribution of the target nuclei to obtain an effective cross-section, σ_{eff} . The typical approach is to define an energy transfer [17], E', to be the difference between the kinetic energies of the nuclei and incident neutron, E. The effective cross-section can then be obtained from the convolution of the Breit–Wigner formula (Eq. (1)), with an energy transfer function, S(E'), accounting for the velocity distribution of the target nuclei [18].

$$\sigma_{\rm eff}(E) = \int dE' S(E') \sigma_{\rm r}(E - E').$$
⁽²⁾

In the simplest case, for a monatomic free gas or classical solid, by taking a Maxwellian distribution of velocities for the target nuclei corresponding to a temperature, T, the energy transfer function becomes [6,7]

$$S(E') = \frac{1}{\Delta\sqrt{\pi}} \exp(-\left(E' - E\right)^2 / \Delta^2)$$
(3)

where Δ is the so-called Doppler width and is given by

$$\Delta = \sqrt{\frac{4mME_Rk_{\rm b}T}{(M+m)^2}} \tag{4}$$

where m and M are the neutron and nuclei masses respectively and k_b is Boltzmann's constant.

Thus the observed resonance line shape may be obtained from [6]

$$\sigma_{\rm eff}(E) = \sigma_0 \frac{1}{2\sqrt{\pi}} \frac{\Gamma}{\Delta} \int_{-\infty}^{\infty} dy \frac{\exp\left(-\frac{1}{4}\frac{\Gamma}{\Delta}(x-y)^2\right)}{1+y^2}$$
(5)

where, $x = (E - E_{\rm R})/\frac{1}{2}\Gamma$, $y = (E' - E_{\rm R})/\frac{1}{2}\Gamma$ and $\sigma_0 = 4\pi\lambda^2 g_J \Gamma_{\rm n}\Gamma_{\rm r}/\Gamma^2$.

However, this result is only applicable for a monatomic free gas or classical solid. In a crystalline solid, the thermal motion of the target nuclei is governed by the phonon spectrum of the crystal lattice. Lamb derived the general energy transfer function for the (n, γ) reaction in a crystalline solid accounting for the creation or annihilation of phonons [19]. Further, by treating the crystal as a monatomic Debye solid, Lamb also showed that at temperatures in excess of the Debye temperature of the crystal, T_D , with weak lattice binding, (where $\Delta + \Gamma \gg k_b T_D$), the resonance line shape has the same form as that of a free gas with the exception that the mean average energy per degree of freedom, $k_{\rm b}T$, in the Doppler width is replaced by the average energy per vibrational degree of freedom of the lattice given an effective temperature, $T_{\rm eff}$ such that,

$$k_{\rm b}T_{\rm eff} = \frac{1}{2} \int \mathrm{d}\nu \, \coth\left(\frac{h\nu}{2k_{\rm b}T}\right) g(\nu) \tag{6}$$

where g(v) is the phonon density of states of frequency v. This model is referred to as the effective free gas model (EFGM). For pure elements above room temperature a simple Einstein model for g(v) is sufficient and yields [16,17]

$$k_{\rm b}T_{\rm eff} \approx \frac{1}{2}hv_{\rm E}\,\coth\left(\frac{hv_{\rm E}}{2k_{\rm b}T}\right)$$
(7)

where the Einstein frequency, v_E can be obtained from the Debye temperature with $hv_E = \frac{3}{4}k_bT_D$. For strong lattice binding and lower crystal temperatures the line formed is more complex and it is necessary to adopt a full harmonic crystal model (HCM) to properly describe the resultant resonance line shape [20].

It is therefore suggested that pure metallic samples should be used, as the mean frequency of lattice vibrations are lower than that for compound materials. Indeed, it is also appropriate to select elements which possess low Debye temperatures to further ensure that the EFGM provides a reasonable description of the resonance line shape.

2.2. Prediction of transmitted signal

If material of thickness w is placed in a neutron beam, then the transmitted signal, $\Phi(E)$, may be obtained from the observed cross-section (Eq. (5)) by

$$\Phi(E) = \exp(-nw\sigma_{\rm eff}(E)) \tag{8}$$

where n is the number density of absorbing nuclei in the material per unit area.

In interpreting neutron resonance data, it is also critical to consider the effect of the instrument resolution on the measured line shape, as the resolution broadening may be comparable to the width of the resonance. In time-of-flight spectroscopy, broadening and asymmetry of the measured resonance profiles arise from a number of sources associated with the production, moderation and measurement of neutrons. These sources include [21] the finite width of the pulse produced by the accelerator; the finite size of the spallation target and moderator: the finite width of the time bins of the time of flight detector system; uncertainty in the time of the pulse start; electronic noise; and the recovery time of the scintillator. A number of instrument resolution functions have been proposed that are appropriate for time-of-flight neutron sources. Of these, the most popular are those based around the convolution of a Gaussian or pseudo-Voigt function with leading and trailing exponential components [22–25] or a χ^2 function [4,5]. The exponential components of these functions account for the effect of moderation of the neutrons between the spallation source and the moderator; they display a strong energy dependence. With resolution broadening the detected signal, N(E), becomes

$$N(E) = \int dE' P(E') R(E') \Phi(E - E')$$
(9)

where R(E) is the instrument resolution function and P(E) is the pulse source function. Provided that the pulse source does not vary significantly over the width of the resonance and resolution function, it is legitimate to approximate the effect of the pulse source function to a product, i.e.

$$N(E) = P(E) \int dE' R(E') \Phi(E - E').$$
(10)

This equation may then be fitted to the measured transmission data using an iterative, least-squares, fitting routine. It is suggested that the convolutions required in calculating Eq. (10) may be most readily accomplished with a numerical approach. The fitting variables will include the sample temperature, number density of absorbing nuclei, and any variables required to describe the open beam signal and instrument resolution (unless the parameters are already known).

3. Experimental description

Experiments to quantify the temperature dependence of the Doppler broadening of neutron absorption resonances for natural hafnium, tantalum, rhenium, and iridium were conducted on the PEARL beam line (viewing a 100 K CH₄ moderator) located at the ISIS neutron spallation source at the Rutherford Appleton Laboratories, UK. Thin foils $(50 \,\mu\text{m} \times 25 \,\text{mm} \times 25 \,\text{mm})$ of each element were mounted on a small purpose-built stainless-steel frame and secured into position with thin tungsten wires. The frame was suspended within the centre of a vanadium element vacuum furnace (RAL-1) in which the foil temperatures were independently monitored by two K-type thermocouples; one located in contact with the top of the small frame supporting the foil; the second located in the centre of the furnace beneath the foil. The temperatures recorded by these two thermocouples therefore provided an indication of the temperature gradient experienced by the foils and was typically less than $\pm 1^{\circ}$ C. Feedback control from the thermocouple in contact with the frame enabled the temperature recorded by this thermocouple to be controlled to within ± 0.1 °C of the selected set point during each measurement.

Short exposure ($\sim 60 \,\mu A \,h$) 'shap-shots' of the transmission spectra of each element were initially taken with the foils mounted individually on the frame without evacuating the chamber. For subsequent measurements conducted at elevated temperature the foils were mounted together in pairs (iridium and rhenium, hafnium and tantalum). These combinations were selected to ensure that no overlap of the resonances of interest occurred. To prevent oxidation of the foils at elevated temperatures, the furnace was evacuated to $\sim 2 \times 10^{-8}$ bar. For each pair of foils, measurements were made in steps from 100 °C to 1000 °C in 100 °C increments. After each increase in temperature the foils were permitted to equilibrate prior to starting the data acquisition. This period varied from approximately 40 min at 100 °C to 10 min at 1000 °C and was chosen so that the temperature recorded by the two thermocouples was seen to be the same and invariant $(\pm 1^{\circ}C)$ during a 2-min period.

Transmission data were gathered with a custommade detector consisting of a ⁶Li-enriched glass scintillator block which presented a face of 7 mm



Fig. 1. Photograph of the detector system developed for the measurement of neutron transmission spectra.

 \times 7 mm to the incident beam and had a length of 25 mm along the direction of the beam. Scintillation detection was accomplished using a 10-stage linear photomultiplier tube. A photograph of the detector system is presented in Fig. 1. The flight path to the detector was determined to be 13.81 m by calibration against the resonance spectrum from a ²³⁸U foil measured in the same configuration as the foils under investigation. These data were also used for the subsequent calibration of the instrument resolution (see Section 5). Transmission data were gathered through both the ISIS data acquisition electronics system (limited to 250 ns resolution) and through a FAST ComTec P7886 multiscaler PCI data acquisition card (with 2 ns time bins for higher accuracy data). The 2 ns time bins corresponded to energy resolution of 0.36 eV and 3.7×10^{-7} eV at 2000 eV and 0.2055 eV, respectively. With this system, transmission data were gathered for neutron energies between 0.2055 eV and 2000 eV. The duration of data acquisition at each temperature was approximately 40 min (approximately 120 µA hours).

4. Resonance data

Normalised transmission spectra obtained with foils of natural tantalum, hafnium, iridium, and

606

rhenium mounted individually (the 'shap-shot' data) at room temperature and ambient pressure, binned with 250 ns time bins are given in Fig. 2. In each of these spectra the stronger resonances in the range 0–100 eV may be readily identified. With 2 ns time bins the number of time bins across each resonance varied from approximately 50000 for resonances with energies of approximately 1 eV to 500 for resonances at approximately 100 eV. However, with this binning the counting statistics associated with each time bin was poor. To minimise the uncertainty associated with each data point for the subsequent analyses the data were rebinned to a level considered appropriate for each resonance investigated (from 256 to 1024 ns time bins) such that a minimum of 12 data points were obtained across the full-width half-maximum of each resonance peak. Due to the limitations of the instrument used and acquisition time available the counting statistics for data gathered from resonances above approximately 10 eV were inadequate for a detailed investigation of the Doppler broadening of these resonances. Subsequent analysis was therefore limited to the stronger low energy resonances of the four materials investigated—the ¹⁷⁷Hf 1.098 eV, ¹⁷⁷Hf 2.388 eV, ¹⁸¹Ta 4.28 eV, ¹⁸¹Ta 10.34 eV, ¹⁸⁵Re 2.156 eV, ¹⁹¹Ir 0.6528 eV, ¹⁹¹Ir 1.298 eV and ¹⁹¹Ir 5.36 eV resonances. These are identified in Fig. 2. Notably, the strong resonances seen in Fig. 2 at approximately 8 eV in the Hf spectrum and 9 eV in the Ir spectrum were not analysed due to the difficulty in separating the contributions from overlapping resonances associated with different isotopes of these elements.

5. Results

The determination of sample temperatures from neutron transmission data by the approach outlined in Section 2 requires reliable values of the



Fig. 2. Normalised transmission spectra obtained from $50\,\mu\text{m}$ thick foils of natural tantalum, hafnium, iridium and rhenium at room temperature with the ISIS data acquisition electronics in 250 ns time bins. Arrows indicate the eight resonance peaks used to extract sample temperatures.

resonance parameters associated with each resonance to be obtained. Much published data exists for the resonance parameters of all naturally occurring isotopes through evaluated nuclear data libraries such as ENDF/B-VI, JENDL-3.3, and JEFF-3.0. However, whilst the resonances of certain elements, such as those with direct nuclear engineering applications, are reliably known, the data available for other materials is often less well determined. For example, the EXFOR database, which provides a comprehensive compilation of experimentally determined nuclear reaction data, lists some 28 references providing data for the resonance parameters of the principal naturally occurring isotope of Tantalum, (181Ta), in the energy range 1–10 eV. Of these references, only four provide data on the widths of the 4.28 eV resonance and even these show significant variations.

The published resonance parameters of each of the eight strong resonances investigated in this study are given in Table 1. It can be seen that significant scatter exists in these data. Mercifully, the Doppler broadening is not sensitive to the resonance parameters (see Eq. (4)); nor do the resonance parameters vary with temperature. Thus, if a resonance is well-described by use of an appropriate resolution function and resonance parameters at a given temperature, it should be possible to determine the temperature of the sample, measured at a different temperature, using the same resolution function and resonance parameters with some degree of accuracy. A key requirement of the analysis of the resonance line shape is therefore the determination of the instrument resolution function.

To obtain a suitable resolution function for the data acquired, the rebinned resonance profiles measured were fitted with Eq. (10). Debye temperatures for each of the elements investigated were obtained from Kittel [26]. Comparison between the fits produced with a selection of resolution functions indicated that a normalised Gaussian offered the most effective description of the instrument resolution in this experiment, i.e.

$$R(E) = \frac{1}{\delta\sqrt{2\pi}} \exp\left(-\frac{(E-E_R)}{2\delta^2}\right)$$
(11)

where δ is the resolution width.

The absence of an exponential decay term that is typically included in resolution functions for neutron spallation sources is consistent with the under-moderation of the epithermal neutrons studied here.

Similarly, testing of a number of candidate background functions indicated that a simple power function provided the best description of the open beam signal, i.e.

$$P(E) = P_0 + P_1 E^{\alpha} \tag{12}$$

where P_0 , P_1 , and α are arbitrary constants.

The resolution widths required to obtain a good fit to the data at 100 °C were determined using the resonance parameters obtained from Mughabghab [27] (see Table 1). A plot of the variation of the resolution widths against the nominal energy of the resonance is given in Fig. 3. It may be expected that if the resonance line shape is well characterised by the functions and parameters used, then this plot would show a smooth variation with the nominal energy of the resonance. However, the plot exhibits significant scatter in the data. This scatter was not seen to improve with higher sample temperatures as may be expected if arising as a result of failure of the EFGM to adequately describe the resonance line shape at temperatures close to $T_{\rm D}$. It was therefore concluded that the scatter occurred as a result of uncertainties in the published resonance parameters used.

To obtain resonance parameters directly from the data acquired, a second-order polynomial was fitted to the variation in the resolution widths with neutron energy (see Fig. 3). The rebinned resonance profiles were fitted again with the width of the resolution function constrained to this curve, whilst the resonance parameters were permitted to vary. The resonance parameters obtained in this fashion were seen to be insensitive to sample temperature; consistent with the observations of Gressier et al. [28] in a study of the resonances of NpO₂. The parameters obtained were averaged over the values obtained at all temperatures for each resonance and are given in bold in Table 1. The values obtained are typically consistent with those determined by other studies. However, some parameters are seen to deviate significantly from the values accepted in the evaluated nuclear data

 Table 1

 Resonance parameters of the eight resonances investigated

Isotope	E_0 (eV)	$2g\Gamma_n$ (meV)	Γ_{γ} (meV)	Reference
¹⁷⁷ Hf	1.098	2.171	66.2	ENDF/B-VI, JENDL-3.3, JEFF-3.3
	1.098	2.219	62.83	JEF-2.2
	1.098 ± 0.02	1.90 ± 0.03	66.2 ± 0.7	Mughabghab [27]
	1.100 ± 0.005	2.10 ± 0.05	67 ± 2	Igo and Landon [30]
	1.095 ± 0.005	1.77 ± 0.002	66 ± 1	Joki et al. [31]
	1.0964 ± 0.0015	2.232 ± 0.013	65.64 ± 2.9	Moxon et al. [32]
	1.099 ± 0.002	1.92 ± 0.04	66.4 ± 1	Fuketa and Harvey [33]
	$\textbf{1.099} \pm \textbf{0.001}$	1.88 ± 0.01	62.6 ± 0.5	This study
¹⁷⁷ Hf	2.388	8.0	60.8	ENDF/B-VI. JENDL-3.3. JEFF-3.3
	2 3837	8.0	61.0	IFF-2 2
	2.3637 2 388 ± 0.002	9.0 ± 0.1	60.8 ± 0.7	Mughabahah [27]
	2.300 ± 0.002 2.39 ± 0.01	9.0 ± 0.1 9.3 ± 0.2	60 ± 1	Igo and Landon [30]
	2.39 ± 0.01	3.1 ± 0.2	$\frac{00 \pm 1}{70 \pm 7}$	Levin and Hughes [34]
	2.380 2 3837 \pm 0 0002	3.4 ± 0.3 8 0628 \pm 0 068	70 ± 7 61 74 ± 0 74	Movon et al. [32]
	2.3837 ± 0.0002	8.0028 ± 0.008	61.74 ± 0.74	Fuketa and Harvey [22]
	2.384 ± 0.002 2.386 ± 0.001	8.9 ± 0.2 9.9 ± 0.1	60.7 ± 0.6	This study
1817-	4.29	2.0	52	ENDED VILLEE 2.2
Ta	4.28	3.9	53	ENDF/B-VI, JEF-2.2
	4.28	3.2	53	JENDL-3.3, JEFF-3.3
	4.28 ± 0.01	3.60 ± 0.16	53 ± 4	Mughabghab [27]
	4.282 ± 0.008	3.9 ± 0.6	49 ± 6	Wood [35]
	4.28	3.6 ± 0.1	62.1 ± 2.6	Evans et al. [36]
	4.28 ± 0.01	3.62 ± 0.12		Belanova et al. [37]
	4.279 ± 0.001	3.5 ± 0.1	63 .1 ± 1.4	This study
¹⁸¹ Ta	10.34	4.66	55	ENDF/B-VI, JEF-2.2
	10.36	4	65	JENDL-3.3, JEFF-3.3
	10.36 ± 0.05	3.50 ± 0.18	65 ± 6	Mughabghab [27]
	10.38 ± 0.01	4.5 ± 0.5	50 ± 10^{1}	Harvey et al. [38]
	10.37 ± 0.03	3.36 ± 0.14	76.5 ± 3.5	Belanova et al. [37]
	10.40	3.6 ± 0.1	65 ± 6	Evans et al. [36]
	10.33 ± 0.01	3.4 ± 0.1	69 ± 4	This study
¹⁸⁵ Re	2 156	2 8286	54.9	ENDE/B-VI JEFE-3 3
	2 160	2.83	54.0	IFF-2 2
	2.156 ± 0.004	33 ± 03	54.9 ± 0.9	Mughabahah [27]
	2.160 ± 0.001	33 ± 0.3	54 + 5	Friesenhahn et al [39]
	2.158 ± 0.001	3.2 ± 0.1	58.8 ± 0.6	This study
191	0.6528	0.4276	72.2	ENDE/D VI
11	0.0528 ± 0.005	0.4370 0.547 \pm 0.006	72.2 + 0.5	Mughababab [27]
	0.0320 ± 0.003	$0.34/\pm 0.000$	12.2 ± 0.3	Mughaoghao [27]
	0.034 ± 0.002	0.25 ± 0.01	$/4 \pm 1$	Landon $[40]$
	0.055 ± 0.004	0.334 ± 0.003	72.0 ± 0.5	Masyanov et al. [41]
	$0.05/\pm0.001$	0.314 ± 0.001	$/4.1 \pm 0.3$	vertebnyj et al. [42]
	0.6551 ± 0.0014	0.668 ± 0.007	70.8 ± 0.9	Brunner and Widder [43]
	0.6528 ± 0.0005	$0.6/9 \pm 0.011$	12.1 ± 0.5	Fischer et al. [44]
	0.6536 ± 0.0005	0.44 ± 0.05	72.2 ± 0.4	This study

Isotope	E_0 (eV)	$2g\Gamma_n$ (meV)	Γ_{γ} (meV)	Reference
¹⁹¹ Ir	5.36	4.8	82.0	ENDF/B-VI
	5.36 ± 0.01	6.0 ± 0.1	82 ± 2	Mughabghab [27]
	5.36 ± 0.04	3.6 ± 0.2	67 ± 5	Landon [40]
	5.36	1.29 ± 0.03	101.6 ± 9.3	Bolotin and Chrien [45]
	5.36 ± 0.009	6.81 ± 0.31	84 ± 5	Masyanov et al. [41]
	5.362 ± 0.005	1.4 ± 0.02	80 ± 3	Vertebnyj et al. [42]
	5.357 ± 0.001	3.4 ± 0.1	$\textbf{84.4} \pm \textbf{1.2}$	This study
¹⁹³ Ir	1.298	0.7304	83.2	ENDF/B-VI
	1.298 ± 0.001	0.913 ± 0.017	83.2 ± 8.0	Mughabghab [27]
	1.303 ± 0.005	0.46 ± 0.01	86.5 ± 1	Landon [40]
	1.301 ± 0.003	0.773 ± 0.013	82.2 ± 1.9	Brunner and Widder [43]
	1.298 ± 0.001	0.80 ± 0.01	83.2 ± 0.8	Fischer et al. [44]
	1.307 ± 0.0025	0.385 ± 0.013	87 ± 1.5	Vertebnyj et al. [42]
	1.304 ± 0.004	0.749 ± 0.024	96 ± 8	Masyanov et al. [41]
	1.303 ± 0.001	1.3 ± 0.2	86.6 ± 0.8	This study

Values in bold are those obtained in this study by fitting the observed neutron transmission data with analytical expressions.

Fig. 3. Width of resolution function required to fit the resonances profiles obtained at $100 \,^{\circ}$ C using the resonance parameters from Mughabghab [27].

libraries by more than the experimental uncertainty.

With a resolution function and resonance parameters providing an adequate description of the resonance lineshape, sample temperatures could then be obtained by fitting the resonance profiles constraining the width of the Gaussian resolution function and resonance parameters (obtained as described above). Examples of the fits obtained with the ¹⁸⁵Re 2.156 eV resonance at 100 °C and 1000 °C are given in Fig. 4.

The quality of the fits produced are generally seen to be excellent with no fine structure evident in the residual greater than the error associated with counting statistics. Notable exceptions are those fits obtained for the ¹⁹¹Ir 0.6528 eV and ¹⁷⁷Hf 1.098 eV resonances which displayed a pronounced asymmetry in their residuals. It is believed that this asymmetry arose as a result of the failure of the function chosen to adequately describe the background in the vicinity of these resonances and some improvement was observed with the use of a quadratic polynomial background instead. Again, it may be expected that if the EFGM did not provide an adequate description of the resonance line shape then it is likely that the quality of the fits produced would be worst for those data acquired at low temperatures (close to the Debye temperature of the material) and for those materials with the highest Debye temperatures, i.e. Iridium $(T_{\rm D} = 420 \,\mathrm{K})$ [26] and Rhenium $(T_D = 430 \text{ K})$ [26] and would be

Fig. 4. Examples of the fits obtained with the data gathered with the FAST ComTec P7886 multiscaler PCI data acquisition card for the ¹⁸⁵Re 2.156 eV resonance examined by analysis incorporating neutron resonance parameters at 100 °C and 1000 °C.

expected to improve with increasing sample temperature. However, no evidence to support this was observed in the fits obtained for any resonance and as such the EFGM was assumed adequate for this analysis.

The temperatures show reasonable agreement with those measured by the thermocouples over the range of temperatures investigated. The standard deviation of the temperatures obtained from those obtained with the thermocouples are given in Table 2 and are typically of the order of $\pm 10-\pm 15$ °C.

5.1. Comparison with empirical methods for temperature determination

5.1.1. Individual time-channel analysis using transmission ratio data and a calibration matrix

In the work of Mayers et al. [2,14] it was suggested that the transmission of neutrons of a specific energy through the sample in the vicinity of absorption resonances could be used to provide an individual measure of the sample temperature. With time-of-flight data, each time bin, associated with neutrons of a specific energy, can therefore be used to provide an independent estimate of the sample temperature. By constructing a calibration matrix from a well-determined experiment at known temperatures, the temperature of the sample may be uniquely determined from each time/energy channel. Further, by performing a

Table 2

Standard deviations of temperatures determined from each of the resonances investigated in this study from those obtained with thermocouples for temperatures between $100 \,^{\circ}$ C and $1000 \,^{\circ}$ C using the analytical method suggested and two empirical approaches of Mayers et al. [2,14] and Le Godec et al. [13]

Isotope	Energy (eV)	Analytical method $\pm^{\circ}C$	Individual channel analysis $[2,14] \pm^{\circ}C$	Empirical fit and calibration [13] \pm° C
¹⁷⁷ Hf	1.099	15	19	39
¹⁷⁷ Hf	2.386	11	18	13
¹⁷⁷ Ta	4.279	16	25	13
¹⁷⁷ Ta	10.33	23	40	29
¹⁷⁷ Re	2.158	5	9	10
¹⁷⁷ Ir	0.6536	12	30	28
¹⁷⁷ Ir	1.303	12	10	19
¹⁷⁷ Ir	5.357	13	24	15

weighted sum over all neutron energies available (or time bins) a more accurate measure of the sample temperature may finally be obtained. In addition to circumventing the need to characterise the resonance line shape in terms of the governing physics, this methodology also eliminates the need to describe and quantify the instrument resolution.

To compare the efficacy of this technique with that described above, a calibration matrix was constructed according to the methodology detailed by Mayers et al. [2] with the variation of the neutron transmission as a function of temperature for each time channel being fitted with a secondorder polynomial which could subsequently be used for temperature determination. Examples of the transmission data normalised against room temperature data is given in Fig. 5. In this figure, four time channels (A, B, C, and D) are identified. The calibration curves obtained from these time channels are given in Fig. 6 as examples. The uncertainty associated with the temperatures determined for each time channel were obtained by considering the uncertainties in the coefficients of the polynomial calibration curves. Samples temperatures were subsequently extracted from the transmission data following the suggestions offered by the authors. However, in applying this procedure it was observed that a significant proportion of the time channels failed to produce a sensible estimate of the sample temperature. This behaviour may be anticipated for those time bins away from the thermally broadened resonances where statistical uncertainties will dominate the variations in transmission seen between successive temperatures. However, it was also observed for many time bins associated with the resonances for which significant variations in the neutron transmission with temperature would be expected. In constructing the calibration matrix it was observed that the uncertainty associated with the data often resulted in significant scatter about the polynomial calibration curve for each time channel. This was also seen to lead to nonsensical polynomial fits for some time channels. As identified by Mayers et al. [2], the time required to acquire the calibration curve data must be significantly longer than that of the subsequent temperature measurements. If this is not the case, the errors associated with the

Fig. 5. Neutron transmission data gathered with a FAST ComTec P7886 multiscaler PCI data acquisition card in the vicinity of the ¹⁸⁵Re 2.156 eV resonance at 300 °C and 1000 °C normalised against that obtained at 100 °C. Four time channels are identified (A, B, C, and D) for which the associated calibration curves are given in Fig. 6.

Fig. 6. Calibration curves obtained for the four time channels identified in Fig. 5 in the vicinity of the 185 Re 2.156 eV resonance.

polynomial calibration curve coefficients must also be taken into consideration. A consequence of higher statistical uncertainty in the calibration is the propagation of significant errors in to the temperature determined for each time channel. This effect may be ameliorated by rebinning the data into larger time bins. However, whilst rebinning of the spectra with larger time bins enabled more reasonable temperatures to be obtained in each individual channel, the net reduction in the number of independent measures of the sample temperature resulted in no improvement in the accuracy of the temperature obtained from the weighted sum over all time channels across each resonance. As a consequence, no improvement was seen in the final temperatures obtained for rebinning beyond 1024 ns.

The standard deviations of temperatures obtained by this method from those measured with the thermocouples are given in Table 2. The deviations obtained are greater than those obtained with the analytical approach described above. Larger deviations are also observed for higher energy resonances as a result of fewer time channels across each resonance contributing to the final temperature determined. The accuracy of the temperatures determined by this method are in contrast to those reported by Mayers et al. [2] in their study of ± 1 °C. The higher errors obtained in this study are attributed to the lower counting statistics of the data gathered compared to that of Mayers et al. This is a particular problem due to the sensitivity of this method to the propagation of errors through the data processing steps to obtain the final temperature.

5.2. Analysis using an empirical fit and calibration curve

In the study of Le Godec et al. [13], temperatures were effectively determined from measured transmission data of the ¹⁸¹Ta 10.34 eV resonance with an accuracy of ± 20 °C by fitting the resonance line shape with a Gaussian function and comparing the fitted width to a calibration curve. However, this method is effective only for those resonances whose line shape may be well described by a Gaussian function. For this to be the case, the intrinsic (Breit–Wigner) line width must be small compared to the broadening arising as a result of the Doppler effect, i.e. those resonances for which $\Delta/\Gamma > 1$. This is the case for the ¹⁸¹Ta 10.34 eV resonance used by Le Godec et al. for the determination of sample temperature in a Paris–Edinburgh cell. However, for the majority of resonances, the contribution of the Breit–Wigner line width to the observed line shape is significant. As a consequence, problems will be encountered in treating the resonance line shape with a simple Gaussian.

To provide a better comparison with the analytical approach described for the resonances investigated in this study it is volunteered that it may be possible to extend the applicability of this approach to include such resonances by fitting the observed line shape with functions possessing a Lorentzian component such as Voigt or pseudo-Voigt functions. With such a function, the Doppler broadening of any resonance would be expected to manifest as an increase in the width of the Gaussian component. In contrast, the intrinsic line shape, being invariant with increasing sample temperature, should result in the width of the Lorentzian component showing little no variation with temperature. Whilst both Voigt and pseudo-Voigt functions were tested for this purpose, marginally greater success was achieved using a pseudo-Voigt function as it provided a better fit to the complex line shape measured. Examples of the fits produced with a pseudo-Voigt function, to the observed line shape of the ¹⁸⁵Re 2.156 eV resonance at 100 °C and 1000 °C, are given in Fig. 7. In general, it is observed that some structure appears in the residual that is not encountered when fitting with the analytical expressions; this is evident in comparison of Figs. 4 and 7. In spite of this limitation, the widths of the Gaussian and Lorentzian components do, in general, display the anticipated behaviour (see Fig. 8). However, for many resonances the Lorentzian component does exhibit a pronounced increase with temperature which may be attributed to the failings of this function to accurately describe the observed line shape.

The temperatures obtained by this method showed greater departure from those measured

Fig. 7. Examples of the fits obtained with a pseudo-Voigt function to the data gathered with the FAST ComTec P7886 multiscaler PCI data acquisition card for the ¹⁸⁵Re 2.156 eV resonance at 100 and 1000 °C.

with the thermocouples than with the other two data processing techniques. The standard deviation of the temperatures obtained by this method are given in Table 2. It is believed that these discrepancies arise as a result of the limitations of this method to adequately describe the observed line shape. Whilst the temperatures determined from the ¹⁸¹Ta 10.34 eV resonance did not reproduce the level of accuracy reported by Le Godec et al. [29] of ± 20 °C, it was observed that the fitting residual displayed less fine structure than the other resonances investigated. It is therefore concluded that the larger errors associated with the temperatures obtained for this resonance may be attributed to higher counting statistics errors than those encountered in their work.

Fig. 8. Widths of the Gaussian and Lorentzian components of a pseudo-Voigt function fitted to the 191 Ir 0.6528 eV resonance as a function of the sample temperature.

6. Discussion

All of the three methods for the determination of sample temperature from Doppler broadening of neutron transmission data examined in this study provide a reasonable measure of the sample temperature. With all three methods temperatures were most successfully determined with the lowest energy resonances. From reference to Eq. (4) it would be expected that high-energy resonances from heavy nuclei would exhibit the greatest Doppler broadening and hence also be the most desirable for temperature determination by NRS. However, with narrow resonances, particularly those at higher energies, the resonance spans fewer time channels so greater data acquisition times are required to adequately determine their line shape and hence also the sample temperature. It is therefore concluded that it may be more appropriate to perform measurements for temperature determination with the lowest energy resonances of the materials available.

Sample temperatures were most effectively obtained by fitting the measured transmission data with the analytical expressions suggested incorporating the theoretical line shape including resolution broadening. This method also offered a considerable advantage over the others that it does not require a calibration to be performed in advance of the experiment. This is particularly useful as, for many of the applications for which temperature measurement by NRS is considered, it is difficult to obtain conditions for a suitable calibration. Additionally, as access to spallation neutron sources is necessarily limited, circumventing the requirement of performing calibration measurements enables all of the beam time to be dedicated towards making the desired measurements. It was noted, however, that this method required considerable care to obtain a resolution function and resonance parameters such that the line shape for a given temperature was welldescribed. Once this was established, however, the temperatures of measurements made at subsequent temperatures were readily determined. If such data is not available it is suggested that the instrument resolution be permitted to vary such that the line shape measured at a reference temperature (typically room temperature) is confidently established before subsequent application to other temperatures. This circumvents the additional care required to accurately determine the resonance parameters which are not a requisite for temperature determination as the Doppler broadening of the resonance is not dependent upon them.

7. Conclusions

An analytical method has been used to obtain sample temperatures by measuring the thermal broadening of neutron absorption resonances. The temperatures were most readily determined from low-energy resonances ($<10 \,\text{eV}$) where the accurate characterisation of the resonance line shape was most rapidly accomplished. Accuracies of ± 10 °C were achieved by fitting the measured transmission data with analytical expressions, although, it is noted that higher accuracies may be possible with extended data acquisition times than those used in this study. As only small quantities of material $(1 \text{ mol } m^{-2})$ were required to provide reasonable resonance absorption, only small amounts of resonant material added to the sample would be required for temperature determination to be performed. For these reasons we have demonstrated that neutron resonance spectroscopy offers a practicable method for the determination of sample temperatures in remote or inaccessible environments.

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