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PULSED NEUTRON SCATTERING ON VANADIUM IN THE THERMAL ENERGY RANGE

J.R. GRANADA, F. KROPFF and R.E. MAYER
*Centro Atómico Bariloche *, 8400 Bariloche, Argentina*

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A scattering experiment was performed on a polycrystalline vanadium sample using a pulsed neutron source. The result shows that the differential cross section of vanadium remains constant for neutron wavelengths up to $\lambda \simeq 1.7$ Å, but a rapid increase in the cross section is observed for longer wavelengths in qualitative agreement with a previous evaluation.

1. Introduction

Absolute normalization has always been a difficult stage in neutron diffraction data analysis, whether a steady state or a pulsed neutron source is being used. Besides the attainment of an absolute differential cross section, the latter case requires the knowledge of the neutron spectrum shape incident on the sample. This information can be obtained from a direct measurement, that is by placing a detector at 0°, but strangely this seemingly safe procedure has rarely been employed [1–3]. Instead, scattering from polycrystalline vanadium has been extensively used in time-of-flight diffraction experiments in order to obtain the shape and intensity of the incident neutron spectrum, due to its property of being an almost totally incoherent, isotropic scatterer. However, elasticity in the scattering process from vanadium, a quality commonly accepted, has been the subject of a recent evaluation [4] which indicated a non negligible energy dependence of the scattering cross section. If this is the case, a vanadium scattering spectrum should no longer be considered as a direct measurement of the energy distribution in the incident neutron beam of a time of flight experiment, although vanadium can still be redeemed as a standard if its scattering cross section is precisely known over the energy range of interest.

In this paper we report an experimental study intended to investigate the thermal neutron scattering properties of vanadium.

* Comisión Nacional de Energía Atómica.

2. The experiment

2.1. Experimental set-up

A detailed description of the Bariloche spectrometer was given previously [3] and therefore only a brief account of its main features is given here.

A 25 MeV electron Linac with a lead target was used as a pulsed neutron source, operating at a repetition rate of 100 pps.

The moderator was a disk of polypropilene, 152 mm in diameter and 25 mm thick. We preferred to use this geometry instead of a sandwich-type moderator [5] in order to enhance the slow neutron current, and because for this experiment there are no special resolution requirements.

Eight ^{3}He detectors (10 atm pressure, 1" diameter, 6" active length) were placed in pairs forming a square, each pair lying on a $tQ_{el} = constant$ surface [3].

The source–sample distance was 490 cm, the sample-detectors distance of approximately 40 cm and the mean angle of the detectors was 161°. The incident flight tube was air evacuated and contained several collimators which defined a beam size of 1" in diameter at the sample position. Both the sample and detection bank were cased into a cube with 10 cm thick borated paraffin walls, lined inside with 1 mm thick cadmium.

The data acquisition line involved conventional NIM electronics, a Laben TV 60 time encoder connected on-line to an HP-2100 computer and this in turn interfaced with an IBM 360/44 computer.

2.2. Measurements

The sample was a coin-shaped piece of polycrystalline vanadium (99.9% purity), with a diameter of 29.4 mm; thickness of 6.71 mm and number density of

0.0718×10^{24} atoms/cm^3. It was mounted on a two-position sample changer with its axis parallel to the incident beam direction.

Twenty seven scattering measurements were performed, each consisting of four sample-in sample-out cycles of a preset duration. The data were sorted by the TOF encoder into 2048 channels of 4 μs width and the sample and background spectra were recorded together with the monitor counts. The sample temperature was kept at ~ 15°C throughout the measurements.

The incident spectrum was measured in a separate run with a ^{3}He detector, similar to those of the detection bank, placed in the direct beam at a distance from the source equal to the total flight distance in scattering measurements. This direct beam detector was located inside a paraffin cylinder, of such dimensions as to provide a 13 cm shield in any direction. The neutrons reached the detector through a hole of rectangular section (2.54×0.3 cm^2) lined with 1 mm thick cadmium. Except for that part of the ^{3}He tube facing the entrance slit, the rest of the detector was also covered by the same thickness of cadmium.

Scattering measurements were also performed with a Cu sample in order to calibrate the time-of-flight scale. Ancillary measurements were carried out: (1) with a Cd sample to search for contributions to the scattering background from neutrons going through the sample position and (2) with the direct beam detector to investigate the beam profile at the sample position.

3. Data reduction and results

3.1. Raw data handling

Scattering data from the Cu sample were used to calibrate the spectrometer, following a peak analysis procedure recently given [5]. According to this formulation the position, shape and integrated intensity of a Bragg reflection in a TOF experiment, is described by:

$$I(t) = A(2\alpha)^{-1} \exp\left\{ -\frac{\sigma}{\alpha}\left[\tfrac{1}{2}\frac{\sigma}{\alpha} + \sqrt{2}\mu\right]\right\} \times [1 + \mathrm{Erf}(\mu)],$$

where

$$\mu \equiv 2^{-1/2}[(t - t_0)/\sigma - \sigma/\alpha]$$

and

$$\mathrm{Erf}(\mu) \equiv 2\pi^{-1/2}\int_0^{\mu} \mathrm{e}^{-y^2}\,\mathrm{d}y.$$

The parameters σ and α characterize the symmetric and asymmetric contributions to the TOF uncertainty respectively, and in practice they are controlled by the spectrometer's geometry and the mean emission time of the neutron source. The scale factor A gives directly the peak integrated intensity, whereas t_0 represents the peak position which is the quantity of interest for the time scale calibration.

We obtained for the elastic scattering vector:

$$Q_e[\text{Å}^{-1}] = 16771.3/(t - 19.2 + \alpha), \qquad t \text{ in } \mu\text{s},$$

which, combined with an independent determination of the total flight distance, gave:

$$\lambda[\text{Å}] = 7.408 \times 10^{-4}\,(t - 19.2 + \alpha), \qquad t \text{ in } \mu\text{s}.$$

The mean emission time $\alpha(\lambda)$ showed the conventional behaviour [5], the value at the long wavelength plateau being ~ 39 μs for the thick neutron source used in this experiment.

From the initial twenty seven scattering measurements on the vanadium sample, two were discarded due to spectrum distortion caused by noise in the acquisition line. The twenty five remaining data sets were then added together, and sample and background spectra were normalized by their corresponding monitor counts after a small dead time correction was performed. The two normalized spectra are shown in fig. 1, as a function of the neutron time of flight.

The data from the direct beam detector were treated in the same manner and the monitor-normalized spectra of incident neutrons and its corresponding background are shown in fig. 2. An important feature is the almost complete elimination of the thermal neutron background, a convenient situation for a reliable determination of the incident spectrum shape.

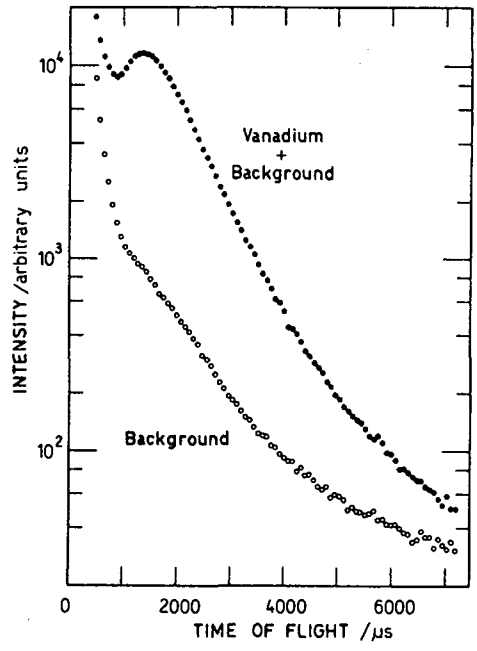

Fig. 1. The recorded time-of-flight spectra for the backscattering detector.

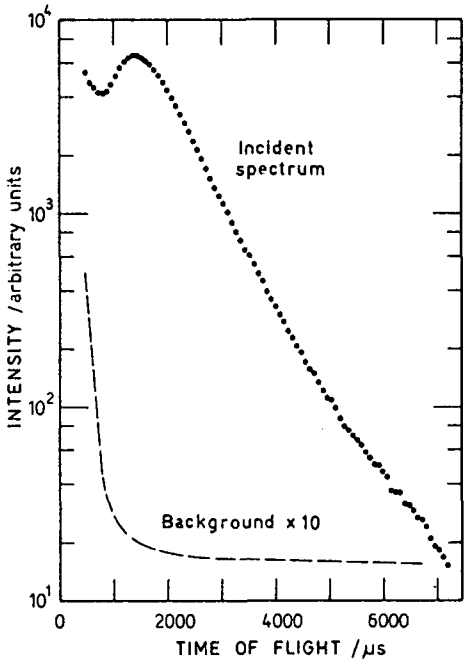

Fig. 2. The recorded time-of-flight spectra for the direct beam detector.

3.2. Beam attenuation and multiple scattering corrections

Theoretically, the differential scattering cross section is given by the observed count rate in a given direction due to single scattering processes divided by the incident neutron flux. In an actual experiment however, the neutron beam is attenuated as it travels inside the sample, the result being that different volume elements do not see the same incident beam falling upon them nor are they able to contribute equally to the observed scattered intensity. Furthermore, neutrons which were scattered more than once inside the sample are also recorded and their contribution must be subtracted from the scattering spectrum.

These two effects are accounted for through the application of beam attenuation and multiple scattering corrections to the raw data [6]. The observed scattered intensity (normalized by the incident neutron spectrum) at any given time channel can be written as:

$$I_D = I_1 + I_M, \tag{1}$$

where I_1 represents the contribution of single scattering processes and I_M is that of multiple scattered neutrons:

$$I_M = \sum_{k=2} I_k. \tag{2}$$

It is customary to express I_M in terms of I_1:

$$I_M = I_1(f_M - 1), \tag{3}$$

and this equation defines the multiple scattering correction factor f_M.

The once-scattered neutron intensity I_1 must be corrected by the attenuation factor f_A [7] in order to obtain the 'ideal' intensity I_i:

$$I_1 = I_i f_A, \tag{4}$$

or, in terms of the experimentally observed intensity I_D:

$$I_i \equiv K\left(\frac{d\sigma}{d\Omega}\right) = I_D/(f_A \cdot f_M). \tag{5}$$

We employed two methods to evaluate the correction factors. The first one was a semi-analytic approach in the spirit of the calculation made by Soper and Egelstaff [8]. The coin-shaped geometry of the sample was divided in the following way:

1) the total thickness L in m parts, each of thickness h;
2) the circular face in n angular sectors, each subtending an angle φ and
3) the total radius R in p parts of non-equal lengths ΔR_k $(k = 1,\ldots,p)$.

The radius partition is obtained from the requirement that all volume elements have the same volume, i.e.:

$$\Delta V_{ijk} = (\varphi/2)(R_k \Delta R_k)h = \pi R^2 L/(npm), \tag{6}$$

with

$$R_k = \sum_{\mu=1}^{k-1} \Delta R_\mu + \tfrac{1}{2}\Delta R_k,$$

from which follows the condition $\Delta R_k = R^2/(2pR_k)$ and the recursive relationships to calculate R_k:

$$R_{k-1} = \tfrac{1}{2}\left[A_k + \sqrt{A_k^2 - R^2/p}\right]$$

and (7)

$$A_k = R_k - R^2/(4pR_k); \qquad A_p = R.$$

In this way, the sample is represented by a tridimensional arrangement of points, defined (in catersian coordinates) by:

$$\boldsymbol{R}_{ijk} = R_k \cos\left[\frac{\pi}{n}(2j-1)\right]\boldsymbol{x} + R_k \sin\left[\frac{\pi}{n}(2j-1)\right]\boldsymbol{y} + \frac{L}{2m}(2i-1)\boldsymbol{z}, \tag{8}$$

$i = 1,\ldots,m; \quad j = 1,\ldots,n; \quad k = 1,\ldots,p,$

with a volume ΔV_{ijk} associated with each of them.

The two following quantities were then numerically calculated over the neutron energy range of interest:

$$S_1 = \sum_i \exp\left\{-N\sigma_t \frac{L}{m}(m - i + 1/2)(1 + 1/\cos\Psi)\right\},$$

and

$$S_2 = \sum_{i,k} \exp\left\{-N\sigma_t \frac{L}{m}(m - i + 1/2)\right\} \times \sum_{i',j',k'} \exp\left\{-N\sigma_t \frac{L}{m}(m - i' + 1/2)/\cos\Psi\right\} \times \exp\{-N\sigma_t |\boldsymbol{R}_{i'j'k'} - \boldsymbol{R}_{i1k}|\}/|\boldsymbol{R}_{i'j'k'} - \boldsymbol{R}_{i1k}|^2,$$

where N is the sample number density and Ψ the angle between the sample normal and the sample–detectors direction. We used the formula [9]:

$$\sigma_t(E) = 4.75 + 5.17\sqrt{0.0253/E}, \qquad E \text{ in eV},$$

to describe the vanadium total cross section at thermal energies.

Finally, we obtained the attenuation factor from:

$$f_A = \frac{L}{m} S_1,$$

and δ, the ratio of secondary to primary scattering from:

$$\delta = N \frac{\sigma_s}{4\pi} \frac{\Delta V}{p} \frac{S_2}{S_1}.$$

The multiple scattering correction factor f_M was then calculated according to Sears's prescription [10]:

$$f_M = [\exp(2\delta) - 1]/(2\delta).$$

The entire calculation was performed for a number of equally spaced points (twenty) in the TOF scale, using two partitions of 720 and 900 volume elements and a subsequent linear extrapolation to an infinite partition to obtain the final values of f_A and δ.

We also performed a Monte Carlo calculation for a few points of the TOF scale, which gave results in excellent agreement with the semianalytic method, thus adding credibility to this crucial stage of data processing.

3.3. Results

The vanadium scattering spectrum was normalized *in shape* by the incident neutron spectrum as measured by the direct beam detector. It must be emphasized that we did not attempt to perform an absolute measurement of the vanadium differential scattering cross section, but to investigate whether or not it remains constant as a function of neutron energy.

The experimental ratio I_D is shown in fig. 3 (closed circles), together with the calculated attenuation and multiple scattering corrections. The total correction f_T is also shown in the same figure and from the comparison with the experimental points it is evident that the "ideal" intensity I_i [in eq. (5)] does not have a constant behaviour along the TOF scale. This is further shown in fig. 4, where the experimental data are compared with our calculated differential scattering cross section [4]. The multiplicative constant K in eq. (5) is obtained by forcing the experimental point at 0.107 eV to coincide with the evaluation at that energy.

Although no error bars have been drawn, the accuracy of the experimental result can be assessed from the correlation of data points. The observed differential scattering cross section of vanadium for the backscattering detector can be represented by

$$F_1, \qquad \text{if } 3\times10^{-3} \leq E < 2\times10^{-2} \text{ eV} \quad (5.2 \geq \lambda > 2.0\,\text{Å}),$$

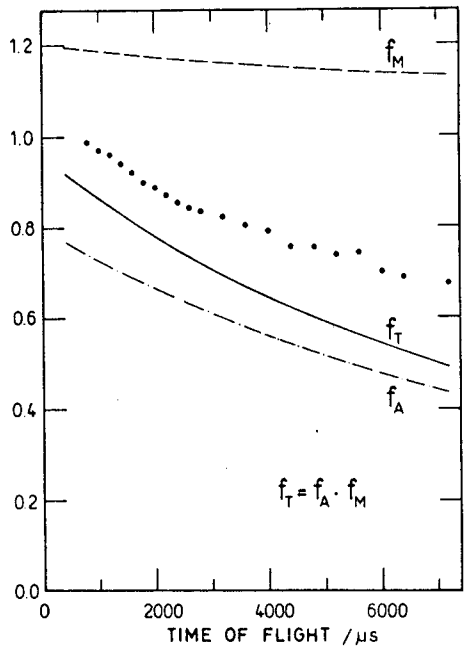

Fig. 3. Correction factors for attenuation (f_A), multiple scattering (f_M) and the combination of both effects (f_T), calculated for vanadium under the conditions of the present experiment. The closed circles indicate the experimental ratio I_D (see text for details).

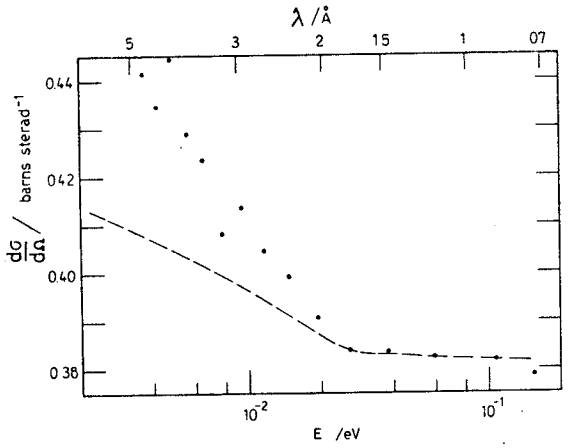

Fig. 4. The observed (closed cirlces) and calculated (dashed line) differential scattering cross section of vanadium for the backscattering (~161°) detector. See text for details on normalization.

$$\left.\frac{d\sigma}{d\Omega}\right|^{obs} = \begin{cases} F_1 + F_2, & \text{if } 2\times10^{-2} \leqslant E < 3\times10^{-2} \text{ eV} \\ & (2.0 \geqslant \lambda > 1.7\,\text{Å}), \\ F_1 + F_3, & \text{if } 3\times10^{-2} \leqslant E < 1 \text{ eV} \\ & (1.7 \geqslant \lambda > 0.3\,\text{Å}), \end{cases}$$

where:

$F_1 = 0.2743 - 0.0296 \ln E$,

$F_2 = 0.03\,(\ln E + 3.912)^2$,

$F_3 = 0.1073 + 0.0292 \ln E$.

4. Discussion

It has been proposed [4] that departures from a flat behaviour in the differential scattering cross section of vanadium exist in the thermal neutron energy region. However, it is expected that such deviation should amount to only a few percent of the free atom value and this imposed demanding conditions on the whole experiment.

The background contribution in both scattering and incident beam measurements was brought down to low levels and this was particularly noticeable in the direct beam case.

Even though the sample-in sample-out method was employed in the scattering measurements, we further investigated the effect of a possible error in monitor normalization on the background subtracted scattering spectrum. Taking a 1% error – a figure which is representative of the standard deviation of the mean value in the twenty five accepted monitor ratios – only the low energy tail of the scattering spectrum is affected (less than 1% at ~0.005 eV).

In order to avoid any distortion of the measured spectra, no smoothing process was applied at any stage of the data reduction.

The attenuation and multiple scattering corrections are fairly large due to the size of the sample used in this experiment. This is not a major obstacle however, as both corrections can be calculated with a high degree of accuracy if the relevant total cross section σ_T is precisely known over the required energy range. Incidentally, we verified that a simple attenuation factor which includes only the $1/v$ absorption component of σ_T is an excellent representation (less than 1% discrepancy) of the total correction factor f_T, the small difference arising due to the path dependence of the multiple scattered neutrons; this result, valid for isotropic scatterers, was already anticipated by Page [11].

The experimentally observed scattering cross section of vanadium for the backscattering detector is shown in fig. 4 as a function of neutron energy and wavelength. There is no quantitative agreement with our calculated curve [4] at low energies and a few comments have to be made at this point:

a) The scattering cross section as calculated by our CRIPO computer programme can not be expected to have an accuracy better than ~2%, as it is based on a Debye frequency spectrum and performs an interpolation between tabulated points. Its predictions however, are well supported by an independent one-phonon calculation at low neutron energies.

b) On the experimental side, it is this energy region which presents more difficulties, due to the rapid fall-off in the recorded spectra for long times of flight. Furthermore, we had to measure the incident neutron spectrum with an entrance slit of a smaller section than that presented by the sample to avoid saturation problems in the detector; this procedure was preferred to that of diminishing the Linac electron current in order to ensure the same conditions at the neutron source. Because the backscattering detectors are arranged in a square configuration (see sect. 3.1) the scattered neutrons are recorded while falling on the detectors with incident angles ranging from 0° to ~ 10°; this is not the case for the direct beam measurements but a calculation showed that no difference in the detector efficiency can be expected across the above mentioned angular range and for the neutron energies under consideration.

Finally, both experimental and calculated curves show a gratifying agreement on the point at which the departure from a flat behaviour is produced; it is very unlikely that a combination of experimental errors would be able to produce such a striking coincidence.

Our data clearly indicate that vanadium does behave as an almost completely elastic scatterer for neutrons of wavelengths up to ~ 1.7 Å, but beyond this limit a rapid increase in the differential cross section is observed. This result is relevant to time of flight diffraction work when the vanadium reference method is employed and we hope that it will prompt neutron users to investigate this matter further.

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