#### DETERMINATION OF THE PARAMETERS OF THE 1.15 keV RESONANCE IN 56Fe BY PROMPT GAMMA-RAY DETECTION

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<u>Abstract</u>: Time-of-flight measurements of the capture cross-section of iron have been made on the Harwell electron linac HELIOS with a gamma-ray detector using the pulse amplitude weighting technique. The work is in support of the Task Force which was set up by the NEANDC to resolve the large discrepancies between capture and transmission measurements of the 1.15 keV resonance parameters.

Two series of measurements were made. In the first, relative capture yields from a 2 mm thick natural iron sample were measured. These data allow the 1.15 keV parameters to be determined relative to the parameters of other resonances in iron. In the second series a thin laminated sample of iron and gold was used, allowing absolute resonance parameters to be obtained by normalizing the capture yield at the "saturated resonance" in gold at 4.9 eV. Results are presented based on recent calculations of the weighting function using a sophisticated electron transport code.

(56Fe, 1.15 keV resonance parameters, capture detectors, weighting functions)

#### Introduction

The 1.15 keV resonance in <sup>56</sup>Fe is important in reactor technology particularly because of its large contribution to the Doppler coefficient of reactivity for fast reactors. It is unfortunate, therefore, that reported measurements of the neutron width of this resonance obtained with "total energy capture detectors" are as much as 30% greater than the value derived from accurate neutron transmission data. The seriousness of the situation has been recognised by the NEANDC in setting up a Task Force¹ whose aim it is to resolve this discrepancy. Matters related to the work of the Task Force are summarized elsewhere in these Proceedings.

A total energy detector is defined as one in which the probability of detecting a capture event is proportional to the total excitation energy. Provided that no more than one gamma-ray is observed from each capture event, any dependence of the efficiency of detecting the event on the shape of the capture gamma-ray spectrum is then removed. In one such type of detector the required efficiency is achieved by applying a weighting function to the observed pulse amplitude spectrum. The weighting function is derived from a series of pulse amplitude spectra for different gamma-ray energies2. In the absence of convenient monoenergetic gamma-ray sources above 2.6 MeV, it is usual to calculate the spectra with a Monte Carlo code. The validity of the weighting function therefore depends on the ability of the code to model a capture event in the detector. Much of the present discrepancy between capture and transmission data for the 1.15 keV resonance stems from capture measurements using the amplitude weighting technique. This is probably because, until recently, calculated gamma-ray responses only poorly represented the observed responses for higher energy gamma-rays. The capture gamma-ray spectrum from the 1.15 keV resonance is dominated by particularly strong

high energy components and a weighting function generated from calculated responses is likely to produce incorrect results. As part of the Harwell contribution to the Task Force a total energy detector using the weighting technique is being studied in some detail.

# Experimental

## Time-of-flight measurements

The detector consists of two 0.4% cells of deuterated benzene (C6D6) mounted on photomultipliers with quartz windows and placed on either side of the capture sample. The sample itself is an 80 mm disc placed normal to the incident neutron beam and contained in an evacuated flight tube made of thin-walled carbon fibre. The detector was designed for the investigation of the structural materials whose s-wave resonances have neutron widths which can be a factor of a thousand or more times greater than their radiation widths. The materials of the detector were accordingly chosen for their low neutron capture cross-sections to achieve a very low sensitivity to background events caused by neutrons scattering from the sample. The scintillator cells consist of thin-walled beryllium cylinders with quartz windows.

Capture cross-section measurements are made with the detector arrangement placed 42 m from the Fast Neutron Target of HELIOS at the end of an evacuated flight tube. The present investigations were made with electron pulse lengths in the range 10-40 ns.

# Detector response to gamma-rays

The computer code which is used to derive the weighting function is tested by comparing calculated and observed responses of the capture detector to monoenergetic gamma-rays. At energies below 2.6 MeV this can be done with standard radioactive sources, but at higher

energies it is necessary to use a nuclear reaction which leads to the emission of monoenergetic gamma-rays. The  $^{19}F(p,\alpha\gamma)^{16}O$  reaction is used in the present investigation. In this process, the second, third and fourth excited states of  $^{16}O$  de-excite almost exclusively to the ground state with the emission of gamma-rays with energies of 6.13, 6.92 and 7.12 MeV respectively. When the incident proton energy is below 500 keV the contribution of the two higher energy transitions is small and the reaction provides an effectively monoenergetic source of 6.13 MeV gamma-rays.

The IBIS accelerator was used to measure the response of one C6D6 detector to gamma-rays produced in the  $^{19}F(p,\alpha\gamma)^{16}O$  reaction by 500 keV protons incident on a target of CaF2 evaporated on to a tantalum backing. In Fig. 1 the circles represent this response and the histogram shows the response calculated with the EGS4 code<sup>3</sup> (F. G. Perey, private communication), the two responses being normalised at the double escape peak at 5.1 MeV. An experimentally determined contribution of 7% from gamma-rays at 6.92 and 7.12 MeV is included in the calculation. code EGS4 originates from the Stanford Linear Accelerator Centre and treats in a comprehensive way the Monte Carlo simulation of electromagnetic cascade showers. Arbitrary geometry can be handled using the combinatorial geometry

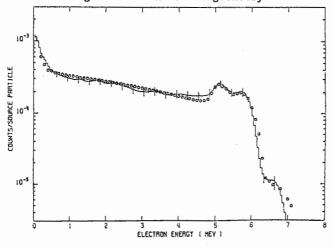


Fig. 1 Detector response to 6.13 MeV gamma-rays.

sub-programs of the MORSE-CG code\*. Perey et al show in another contribution to these Proceedings that when such a comprehensive code is applied to the problem, and when all the materials surrounding the source and the detector are properly accounted for, reasonable agreement with experiment is obtained. The present comparison shown in Fig. 1 is an example of this, the shapes of the two spectra agreeing to within about 15% over most of the energy range. This agreement contrasts markedly with earlier comparisons where calculated spectra under-estimated by a factor of two or more the number of events below 4 MeV when normalized to experiment as in the figure. The failure of these original calculations to reproduce the experimental responses was most noticeable for higher energy incident gamma-rays, and arose mostly from inadequate treatment of electron tracking in materials local to the source and detector. Clearly, weighting functions generated with such codes will give unreliable results, particularly if applied to

capture gamma-ray spectra with strong transitions to the ground state as in the 1.15 keV resonance in 56Fe.

The measurement shown in Fig. 1 consists only of the shape of the gamma-ray response. Further measurements have since been made on the Harwell 5 MV Van de Graaff in which the absolute response of the detector to 6.13 gamma-rays was determined by comparison with a calibrated Ge(Li) diode. As in the IBIS measurements a CaF2 target on a tantalum backing was used. The results are not yet fully evaluated, but the indications are that the absolute efficiency calculated with EGS4 agrees with the observed efficiency to within 15%. In analysing these measurements, EGS4 calculations show that a significant source of secondary electrons is generated in the % mm thick tantalum target backing. These produce an enhancement of the gamma-ray response towards low energies. Experimentally, it is found that a significant reduction in this component can be achieved by placing a 12 mm thick layer of aluminium on the front face of the  $C_6D_6$  detector. This thickness is sufficient to prevent electrons of energy up to ~6 MeV from reaching the scintillator while producing minimal attenuation of the 6.13 MeV gamma-rays. This results in a pulse amplitude spectrum which is almost flat below ~4 MeV. It is apparent that in the capture cross-section measurement itself a significant fraction of the observed amplitude spectrum will be due to secondary electrons produced in the sample.

Tests of the sensitivity of resonance parameters derived by the weighting technique to the actual shape of the weighting function have been made. These indicate that, at the above level of agreement between observed and calculated gamma-ray responses, the uncertainty introduced in the parameters from the application of a calculated weighting function should not be greater than about 3%. A weighting function for energies up to 12 MeV has been generated with the EGS4 code for each of the capture sample geometries used in the present measurements and the results obtained with these functions are now considered.

### Results

## 2 mm iron sample

Time-of-flight measurements have been made of the capture yield from a 2 mm thick sample of natural iron in the energy range 190 eV to 120 keV. The shape of the incident neutron spectrum was determined by replacing the iron sample with a 10B sample. The yield obtained is not absolute, but the parameters of the 1.15 keV resonance are determined relative to those of the 22.8 keV resonance in 56Fe which has a much "softer" capture gamma-ray spectrum than that from the 1.15 keV resonance. This difference in spectra led Weston and Todd<sup>5</sup> to compare measured capture areas  $(g\Gamma_n\Gamma_\gamma/\Gamma)$  of the two resonances as a means of testing the validity of their weighting funtion. The ratio of the areas obtained using the area analysis code CAREA are compared below with the values reported by Weston and Todd.

Table 1 Ratio of capture area 22.8 keV/1.15 keV

Measurement	Ratio
ORNL <sup>5</sup> (transmission)	2.83±0.16
Weston and Todd <sup>5</sup> (capture)	2.91±0.17
Present (capture)	2.55±0.16

Although the present ratio is 10% lower than the transmission value, the difference is not unreasonable in view of the uncertainties. Analysis with the original Harwell weighting function<sup>2</sup> gives a ratio of 2.39±0.16. It is apparent that the use of the weighting function based on EGS4 calculations which properly model the experimental geometry gives somewhat better agreement with the transmission value.

### Laminated sample of iron and gold

Time-of-flight measurements have been made on the 42 m flight path in the energy range 2.4 eV to 110 keV using a laminated sample of iron and gold sheet in the form, 0.3 mm Fe - 25  $\mu$ m Au - 0.3 mm Fe. This arrangement allows absolute iron capture cross-sections to be obtained by normalizing the observed capture yield by the "saturated resonance" method at the 4.9 eV resonance in gold. This method relies on the fact that in a resonance with a large peak cross-section, and in which the dominant interaction is neutron capture, the capture yield (capture events per incident neutron) will approach unity near the peak, even when the sample is relatively thin. Calculation of the exact yield for the purpose of normalization is thus insensitive to the precise value of the resonance parameters. Figure 2 shows a calculation of the capture yield for the present sample obtained with the shape analysis code REFIT' in comparison with the experimental data after normalization. The abscissa is the time-of-flight channel number.

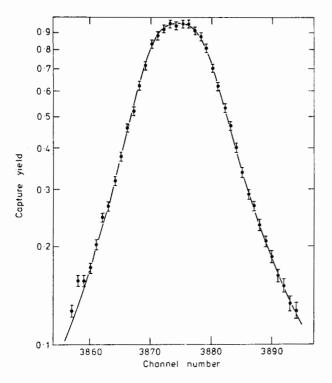


Fig. 2 Normalization of capture yield at 4.9 eV

The shape of the incident neutron spectrum was determined by replacing the iron/gold sample with a 10B sample. The capture area of the 56Fe 1.15 keV resonance derived from the measured capture yield with the code CAREA6 is shown below.

Table 2 Capture area of 1.15 keV resonance

Measurement	$g\Gamma_n\Gamma_\gamma/\Gamma$ (meV)
ORNL (transmission)¹ Harwell (transmission)® Present (capture)	55.7±0.7 57.8±0.7 59.5±3.0

Analysis of the present measurements with the original Harwell weighting function  $^2$  gives an area of 65.0 $\pm$ 3.0 meV.

#### Discussion

In this work we have applied to our measurements a weighting function which was derived from a comprehensive Monte Carlo code which models accurately the experimental geometry. Values have been obtained for the ratio of the 22.8 keV to the 1.15 keV resonance areas and for the absolute area of the 1.15 keV resonance. These are in better agreement with the values obtained from accurate transmission data than those derived from our previous analysis based on a less satisfactory weighting function. These results go someway to resolving the longstanding discrepancy between capture and transmission measurements of the parameters of the 1.15 keV 56Fe resonance.

Analysis of the measurements will continue and attention will be given to improving the calculated gamma-ray response of the  $C_6D_6$  detector. The resonance analysis will also be improved by using the comprehensive R-matrix code REFIT?

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