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MUCH ADO ABOUT NOTHING: DEEP MINIMA IN ^{45}SC AND ^{56}FE TOTAL NEUTRON CROSS SECTIONS*

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The deep minima in ^{45}Sc and ^{56}Fe neutron total cross sections have been measured at the Gaerttner Linac Laboratory by using thick, ultra-pure samples in transmission experiments. The samples are used to produce quasi-monoenergetic beams at the BNL High Flux Beam Reactor. For the ^{45}Sc minimum near 2.05 keV we obtain $\sigma_{total} = 0.71 \pm 0.03$ barns, in sharp contrast to a previously reported value of ~ 0.05 barns. The ^{56}Fe measurement was carried out with a 6 kg, 68.58-cm-long sample of 99.87% isotopically pure sample of ^{56}Fe; a minimum cross section of 0.0085 ± 0.004 barns at 24.39 keV is inferred. This may be compared to a value of 0.420 barns for natural iron.

(Neutron total cross sections, ^{45}Sc measured from 0.4 to 22 keV; deduced neutron resonance parameters; ^{56}Fe measured from 0.4 to 1000 keV)

I. Introduction

Many elements exhibit deep neutron total s-wave cross section minima due to the interference between resonance and potential scattering amplitudes. These minima are of considerable interest in shielding applications and in the design of transmission filters for the production of quasi-monoenergetic neutron beams. For the latter application both steady state reactors[1,2,3] and pulsed sources[4,5] have been used. The use of such filters for dosimetry measurements has been discussed by Schwartz[6] at this meeting. Scandium and iron-aluminum filters have been also rather widely used for capture γ-ray and cross section studies. The empirical optimization of scandium and iron filters has been discussed by Greenwood and Chrien.[3] A summary of known filter facilities has recently been prepared by Tsang and Brugger.[7]

In spite of the wide applicability, the total neutron cross sections of ^{45}Sc and ^{56}Fe, in particular, are ratherly poorly known. In the installation of the HFBR scandium filter, it became obvious that the flux measured at this filter could not be reconciled with the accepted cross sections. Furthermore no accurate measurement of ^{56}Fe total cross sections has been published. For these reasons we carried out experiments to establish the cross sections at the 2.05 and 24.4 keV minima in ^{45}Sc and ^{56}Fe. Thick metallic samples of Sc (99.9% pure) and separated Fe (enriched to 99.87% in ^{56}Fe) were obtained from the HFBR Tailored Beam Facility. Neutron transmission measurements were carried out at RPI's Gaerttner Linac Laboratory, and the data subsequently were analyzed at BNL.

II. Method

A full description of the RPI Linac Facility has been reported, and only a brief description is given here.

The standard water-cooled Ta and CH_2-moderated neutron TOF target and the ^{10}B-NaI neutron detector at the 28.32-m flight path were used for these measurements. The linac was operated at a repetition rate of 500 sec^{-1}, an electron energy of $\simeq$ 70 MeV, a peak electron current of $\sim$ 1 A, and an electron pulse width of either 19, 35, or 66 ns. The counting data were recorded vs. TOF with the 31.25-ns TOF clock interfaced to the PDP-7 on-line computer. The transmission samples were cycled automatically into and out of the neutron beam by the programmed computer, and a cycle was repeated every 10 to 20 minutes to average out neutron source intensity fluctuations.

The composition of the ^{45}Sc and ^{56}Fe samples are listed in Table 1. The ^{45}Sc samples were prepared by

Table 1
Sample Properties

(A) Scandium Samples

Sample No.	Dimensions (cm)	1/N (barn/atom)
1	2.54 diameter x 10.2	2.738
2	2.54 diameter x 15.2	1.844
3	2.54 diameter x 30.5	0.922
4	2.54 diameter x ~0.2	152.5
5	2.54 diameter x 0.55	50.8
6	2.54 diameter x 2.4	11.7

Impurity	Atom Per Cent
H	0.49
O	0.18
Ta	0.037

(B) ^{56}Fe Sample

Dimensions (cm)	1/N (barn/atom)
3.22 x 3.85 x 68.6	0.175

Isotope	Wt. Per Cent
^{54}Fe	0.05
^{56}Fe	99.87
^{57}Fe	0.07
^{58}Fe	0.006

Impurity	Atom Per Cent	Impurity	Atom Per Cent
O	.617	Cr	.011
Cu	.120	W	.010
H	.059	Ni	.005
C	.020	Si	.002
N	.012	P	.002

vacuum sublimation onto a cooled Ta plate, and the sublimed metallic material was subsequently pressed into steel containers 2.54 cm in diameter. The H and O impurities were determined by the vacuum fusion method, and the heavier elements by mass spectrometry,

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for both samples, at the Ames Laboratory Analytical Center. In addition the Ta content of the Sc was checked by emission spectrography, by resonance x-ray fluorence, by neutron activation, and by neutron transmission. This wide variety of methods for Ta analysis was necessitated by the discovery of a sharply inhomogeneous distribution of Ta impurity throughout the bulk of the Sc filter. Both activation and transmission methods were able to sum over large sections of the filter and thus produce a more reasonable average impurity content than was the case for the other techniques, which were applied to small samples of the filter material.

The ^{56}Fe sample was prepared at Oak Ridge National Laboratory from electromagetically enriched iron in the form of iron oxide. The metallic sample was obtained by reducing the oxide in a hydrogen atomosphere. The isotopes and impurities listed in Table 1 were determined from measurements of the iron oxide and metal respectively.

In the final analysis, these cross section measurements are only as reliable as the impurity content determination. At the Sc minimum, the impurity correction is about 112 mb out of a measured 822 mb: while in the ^{56}Fe, the correction is about 51.5 mb out of a measured $\sim$ 60 mb. In each case ENDF/B IV evaluated cross sections were used in the correction.

Two sets of ^{45}Sc transmission measurements were carried out. In one measurement the 10.2-cm-long scandium sample No. 1 was placed in the neutron beam to produce a TOF-filtered spectrum of neutrons which is peaked near the interference minimum in scandium. This removes most of the neutrons from the beam and results in a very low background of neutrons with energies far from the minimum. Then samples No. 2 and 3 were cycled into the filtered beam to obtain an accurate measurement of the cross section near the interference minimum. The other measurement was a conventional transmission measurement. The 10.2-cm-long scandium filter was removed from the beam and samples No. 4, 5 and 6 were cycled into the beam. This latter measurement enabled the cross section to be determined near the peaks of the resonance as well as near the minima.

For the ^{56}Fe measurements a 20.3-cm-long filter of pure Armco iron was placed in the neutron beam to produce a TOF-filtered spectrum peaked near the iron minimum. The 68.6-cm-long ^{56}Fe sample was then cycled into and out of the filtered neutron beam. The filtering effect in iron is illustrated in Fig. 1 near the 24 keV minimum. Here the ^{10}B-NaI detector relative counting rate is shown for Armco iron filters varying in thickness from 5.1 cm(2") to 50.8 cm(20"). For this experiment the 20.3 cm(8") thickness was used, and from Fig. 1 we see that the peak transmission through the filter is 48% and that most of the neutrons at energies several keV away from the peak are removed from the beam.

RELATIVE NEUTRON INTENSITY FOR 2", 8", 14", AND 20" IRON FILTERS

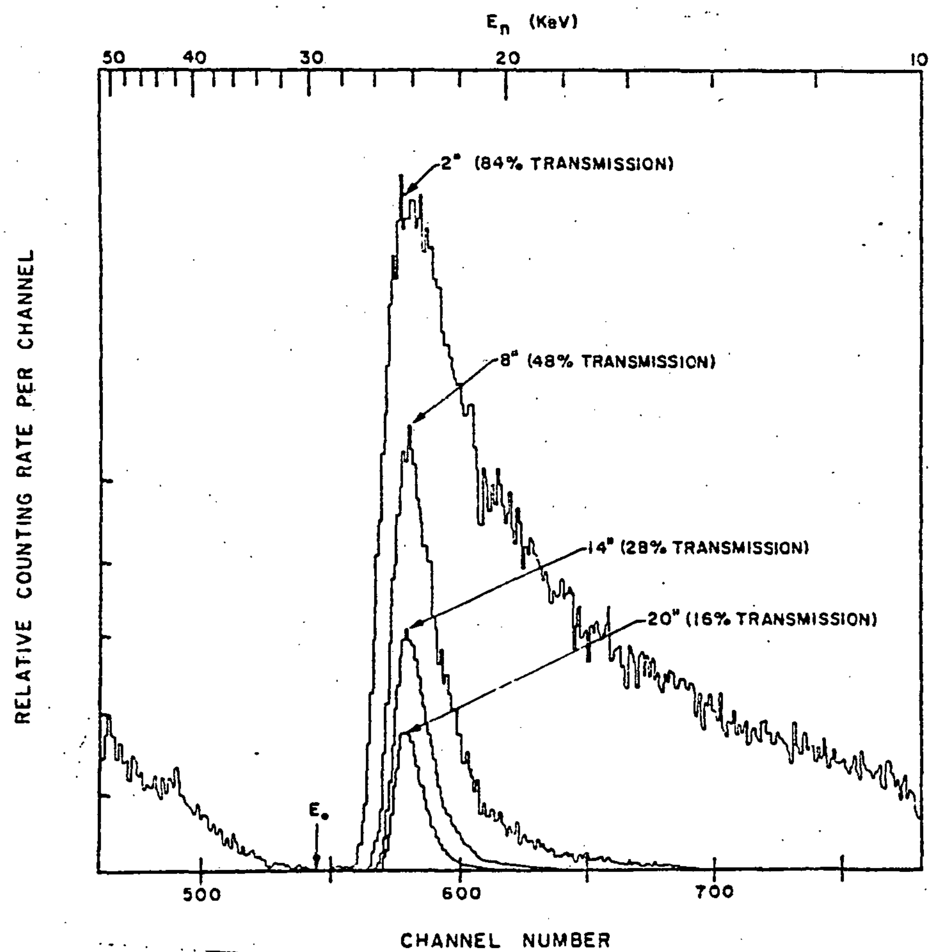

Fig. 1: The ^{10}B-NaI detector counting rate vs. TOF with Armco iron filters 5.1-cm(2"), 20.3-cm(8"), 35.6-cm(14") and 50.8-cm(20") thick. The peak transmission through each filter is shown in parentheses.

The TOF counting data were corrected for dead-time losses in the electronics and for background, and the neutron transmission was then determined. The total cross section σ_t was obtained from the neutron transmission with the following equation

$$\sigma_t = -(1/N) \ln T - (N_{air}\sigma_{air})/N \qquad (1)$$

where N is the thickness of the scandium or ^{56}Fe sample, T is the neutron transmission, N_{air} is the thickness of air displaced by the sample, and σ_{air} is the neutron total cross section of the air. The cross section of air was obtained from the oxygen and nitrogen cross sections plotted in BNL-325.[10]

III. Results

A. ^{45}Sc

The ^{45}Sc total cross section obtained from equation (1) is plotted in Fig. 2 over the neutron energy range from $\simeq$ 0.4 to 22 keV. This plot is a "blend" of all the ^{45}Sc data, where the data near the peaks in the cross section are from the thinnest sample and the data near the deep minima are from the thickest sample. The data have been corrected for the presence of the contaminants listed in Table 1.

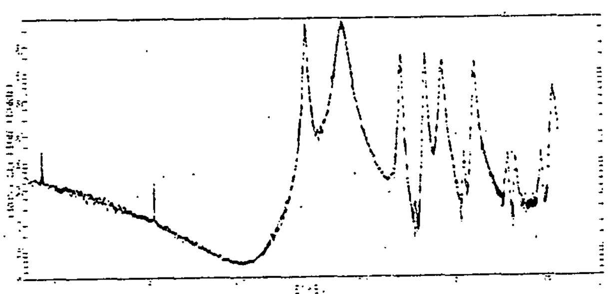

Fig. 2: The neutron total cross section of ^{45}Sc. The experimental data are shown as solid points with error bars (standard deviations determined from the counting statistics). The solid curve is a resolution-broadened "R-matrix" fit to the data using the resonance parameters listed in Table 2.

The measured minimum cross section of ^{45}Sc near 2 keV is (0.71 ± 0.3)b, where the error is derived from a combination of the counting statistics and the uncertainties in the H and O corrections. The minimum occurs at an energy of (2.05 ± 0.02) keV. This minimum cross section of (0.71 ± 0.03)b is in serious disagreement with earlier measurements reported by Wilson[11] of ~ 0.05 b, and it is also in disagreement with the evaluated minimum cross section[12] of ~ 0.085 b which was based on these older measurements. Our result of 0.71 b has serious implications for the effective use of expensive scandium for filtered beams. The 0.71 b cross section at 2.05 keV is comparable to, or larger than, the cross sections in the higher energy ^{45}Sc minima, and thus the length of the ^{45}Sc filter should be limited to enhance the transmitted neutron flux near 2 keV relative to that transmitted at higher energies.

The ^{45}Sc total cross section has been fit by an approximate R-matrix formalism,[13] and the solid curve through the experimental points in Fig. 2 is a "best fit" to the data. The curve has been resolution broadened; the resolution FWHM is approximately 3 channels near the 2 keV minimum. In Table 2 are listed the resonance parameters derived from this fit.

Table 2

^{45}Sc Resonance Parameters

s-wave level parameters: Γ_γ = 0.4 eV

E_o (eV)	Γ_n (eV)	J	E_o (eV)	Γ_n (eV)	J
-500	4.0 (Γ_n^o)	3	11575	290	4
-220	0.67 (Γ_n^o)	4	14525	20	3
3295	75	3	14740	26	4
4330	340	4	15560	28	4
6684	130	3	15850	5	3
8023	145	4	18580	32	3
9092	300	3	18870	62	4
10625	10	3	20500	80	4
10735	6	4	20780	710	3

p-wave level parameters

E_o (eV)	$g\Gamma_n$ (eV)
460.6	0.0022
1060.4	0.0050
7377.0	0.4
7458.0	0.4
7548.0	0.25

Resonance parameters for ^{45}Sc derived from shape fits to the total cross section.

This fit was determined by the following procedure:

(a) The spins of the positive energy resonances were obtained by fitting the peak cross sections and the shape of the interference between resonances.

(b) Negative energy levels were introduced to fit the cross sections at thermal and in the low energy region.

(c) The neutron widths were obtained for all the resonances such that (i) the calculated R-matrix cross section curve produced an acceptable overall fit to the data, and (ii) the R-matrix minimum total cross section near the 2 keV minimum equaled the observed value of 0.71 b.

(d) A single radiation width was then determined for all the resonances such that the thermal capture cross section resulting from the sum of contributions from all the resonances equaled the evaluated value[12] of (26.5 ± 1.0) b.

The best fits to the data are obtained when the J=3 channel spin contributes significantly to thermal capture. The "best fit" parameters listed in Table 2 produce a thermal capture cross section which has approximately equal contributions from J=3 and J=4 channel spins.

Thermal neutron capture γ-ray spectral measurements by Bolotin[14] favor a significant J=3 channel spin contribution. He observed a primary gamma-ray transition to the 1^- state in ^{46}Sc at an excitation energy of 142 keV. Thermal capture in scandium consists of a mixture of capture into 3^- or 4^- states and Bolotin's observed transition strength of 1.3 gammas per 1000 captures indicates that this gamma ray is an E2 transition from a 3^- to a 1^- state. The partial radiation width for this E2 transition can be calculated from the observed transition strength and the resonance parameters deduced from the R-matrix fit to the total cross section. The E2 width calculated from the "best fit" parameters in Table 2 is about 6 times larger than the typical E2 width observed in this mass and energy range, and this is reasonable considering the fluctuations of the observed E2 widths. However, when the E2 width is determined from the R-matrix parameters which produce predominantly J=4 channel spin thermal capture, the E2 width is about 500 times larger than the typical E2 width.

Such a 500 times larger E2 width is very unlikely, and thus Bolotin's measurements favor thermal capture which has a significant J=3 channel spin contribution. We have independently confirmed Bolotin's observation in a separate experiment to be reported elsewhere.

A polarized neutron experiment by Roubeau et al.[15] claims to have measured the difference in scattering lengths at thermal between the (I + 1/2) J=4 and (I - 1/2) J=3 spin states. They report a value of $(a_+ - a_-) = + 1.2 \times 10^{-12}$ cm. The implication of their result is that a J=4 state dominates thermal scattering. This result is inconsistent with our present result, since it would mandate a deep minimum near 2 keV, as a result of interference between the dominant J=4 resonance at 4.330 keV and a strong J=4 bound state. An attempt to fit our data subject to this constraint results not only in a poor fit, but requires absurd R-matrix parameters, (e.g., $R^{\prime}_{J-\frac{1}{2}}/R^{\prime}_{J+\frac{1}{2}} \approx 8$). We conclude that the experiment of Roubeau et al. is in error.

B. ^{56}Fe

The ^{10}B-NaI detector counts per TOF channel are plotted in Fig. 3 for the 20.3-cm (8") Armco iron filter in the neutron beam, and in Fig. 4 for the 20.3-cm Armco iron filter plus the 68.6-cm (27") ^{56}Fe sample. The 68.6-cm ^{56}Fe sample produces a quasi-monoenergetic peak of neutrons which peaks near an energy of 24 keV.

The total cross section of ^{56}Fe near the 24 keV minimum is plotted in Fig. 5. For iron both impurity cross section corrections and resolution corrections are very important, which was not the case for scandium. The total correction for impurities amounted to 51 mb in the region of the minimum; and we infer a minimum observed cross section of ~ 15 mb at an energy of 24.39 $\pm$ 0.04. This cross section is, however, seriously affected by our resolution function, which has a FWHM of about 200eV at the minimum (~ 2.1 channels). Using the parameters of Pandey and Garg[16] we fitted a series of resolution broadened cross section curves to the minimum, using the radiation width of the major s-wave resonance near 27 keV as a parameter. We find the best fit for a radiation width of about 2.2 eV, which provides an independent measurement of that important parameter. We also include a p-wave potential scattering contribution in this calculation. From the best fit, we infer a minimum cross section of 8.5 $\pm$ 4 mb, of which 1.3 mb is due to ℓ=1 scattering, and the balance is due to the (n,γ) reaction.

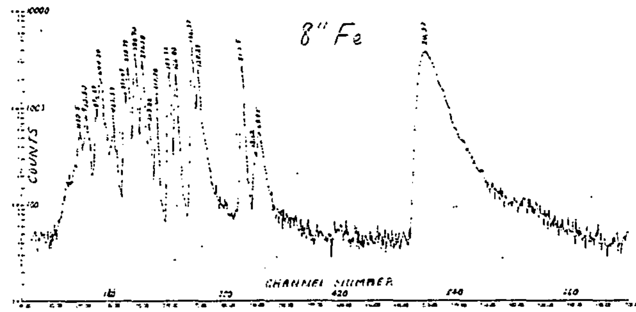

Fig. 3: The ^{10}B-NaI detector counts vs. TOF channel number for a 20.3-cm(8") Fe filter placed in the neutron beam. The TOF channel width is 31.25 ns.

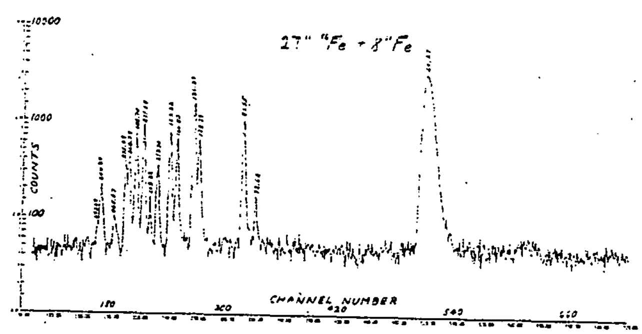

Fig. 4: The ^{10}B-NaI detector counts vs. TOF channel number for a 20.3-cm(8") Fe filter plus a 68.8-cm(27") ^{56}Fe sample placed in the neutron beam. The TOF channel width is 31.25 ns.

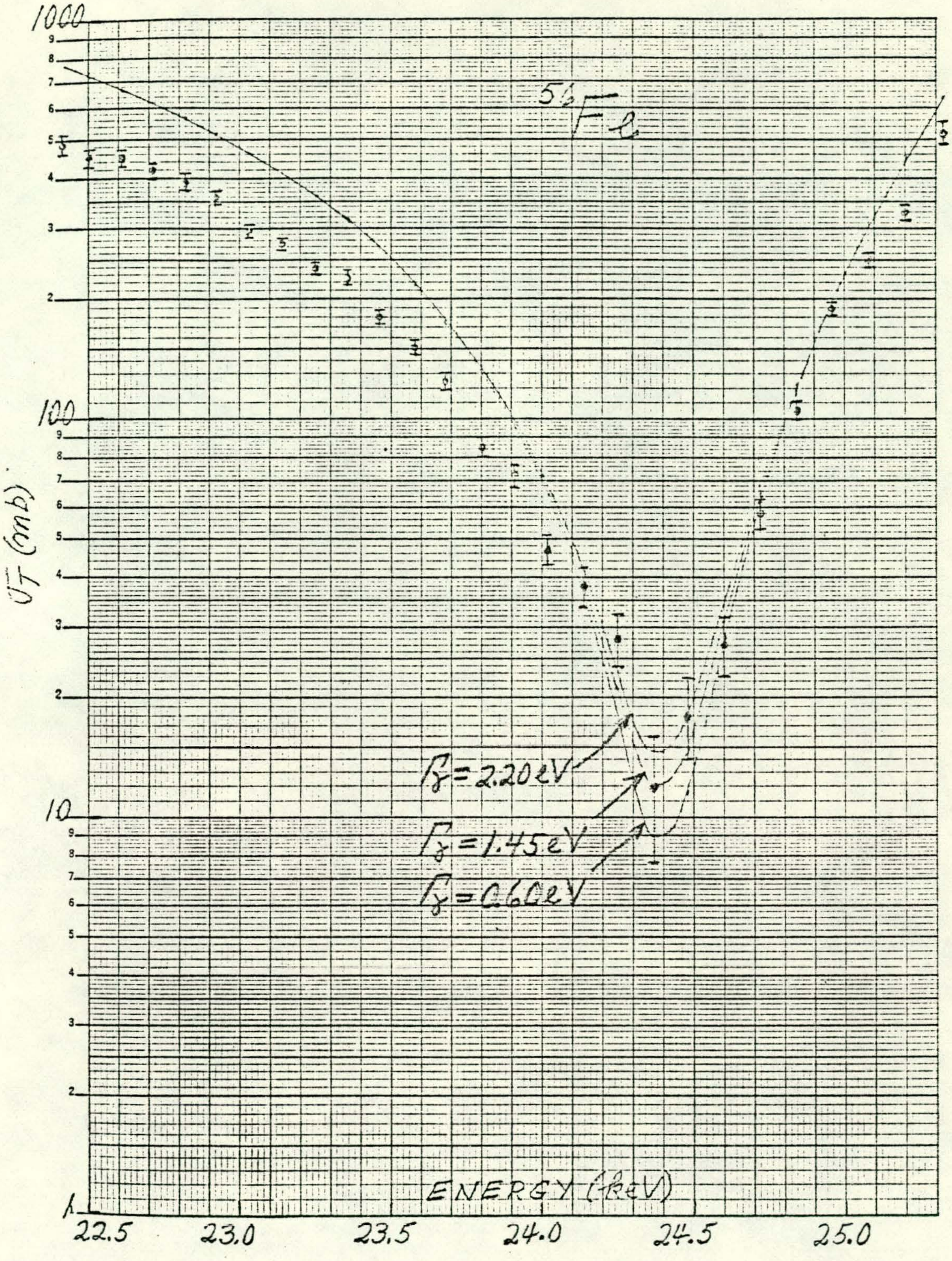

Fig. 5: The neutron total cross section of ^{56}Fe near the 24.37 keV minimum. The solid curves are resolution-broadened "R-matrix" calculations using the resonance parameters shown in the figure.

The minimum cross section measured for this ^{56}Fe sample is almost two orders of magnitude smaller than the $\simeq$ 420 mb cross section measured for elemental Fe.[17,18] This results in a much more intense beam of quasi-monoenergetic $\simeq$ 24 keV neutrons from this filter than from a filter of Fe of the same length. For example, for the 68.6-cm long filter with $1/N = 0.175$ b/a, the transmission through the ^{56}Fe filter at 24.4 keV is 72% (allowing for impurities), whereas the transmission through the same thickness of Fe is only 9%. Thus a quasi-monoenergetic beam of $\simeq$ 24 keV neutrons can be obtained with a ^{56}Fe filter which has excellent transmission through the 24.4 keV minimum. The filter can be used in very thick configurations to reduce unwanted fast neutrons and gamma rays.

Summary

The neutron total cross section has been measured for ^{45}Sc and ^{56}Fe with particular emphasis upon measuring the cross section in the minima. The ^{45}Sc cross section has a prominent minimum at (2.05 $\pm$ 0.02) keV which is (0.71 $\pm$ 0.03) b. This cross section is an order of magnitude larger than estimated from earlier measurements, and this has serious implications in the design of a ^{45}Sc filter for reactors. Although the design of a ^{45}Sc filter system depends upon the application of the system (e.g., for neutron capture spectra, dosimetry, etc.), this higher cross section of 0.71 b should lead to the selection of a thinner ^{45}Sc filter than one based on the former $\sim$ 0.05 b value.

The ^{56}Fe cross section has a prominent minimum at 24.4 keV which is (8.5 $\pm$ 4.0) mb. This is considerably smaller than the $\simeq$ 420 mb minimum in elemental iron, and thus thick filters of ^{56}Fe can provide intense quasi-monoenergetic beams of $\sim$ 24 keV neutrons with a very small contamination of gamma rays and fast neutrons.

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