Neutron Capture in Iron*

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A search for resonances responsible for neutron capture in iron below 3 keV has been made with the fast choppers at the Atomic Energy Commission Laboratory, Chalk River, Canada, and at Brookhaven National Laboratory. Capture area, self-indication, and transmission techniques have been employed. The only resonance observed was that at 1200 eV. The complete set of parameters for the resonance has been derived: $E_0=1200\pm30$ eV, $\sigma_0=165\pm12$ b, $\Gamma_\gamma=0.673\pm0.074$ eV, and $\Gamma_n=0.056\pm0.06$ eV. Analysis of the capture gamma-ray spectrum shows the resonance to be due to Fe^{56} and establishes the compound state as $\frac{1}{2}^+$. The variation of total cross section with energy from 0 to 3 keV is ascribed to a neutron bound level with the following parameters: $E_0=-4390\pm1700$ eV, $\Gamma_n^0=12.5\pm8.1$ eV, $R'=3.9$ F, and $\Gamma_\gamma=1.02\pm0.46$ eV. It is demonstrated that the previously measured resonance capture integral of Fe cannot be accounted for in terms of known resonances.

INTRODUCTION

THE neutron capture cross section of Fe is of particular interest because of its extensive use for neutron shielding near reactor cores and particle accelerators and as a common structural material. There are several interesting aspects to neutron capture in iron which require investigation:

(1) No radiation width has, as yet, been measured for any resonance in iron.[1,2]

(2) No reasonable estimate for an average radiation width for resonances in iron (specifically in Fe^{56}, the dominant isotope) can account for the thermal capture cross section of[3] 2.5 b in terms of the known neutron resonances.

(3) No reasonable estimates for the average radiation width can account for the measured resonance integral of[4] 2.2 b in terms of known levels.

Furthermore, the spectrum of gamma rays following thermal capture is of some interest because of its dominance by strong ground state or high-energy gamma rays. About 35% of the transitions take place to the ground state or the first excited state. This suggests the possibility of a direct capture mechanism as outlined by Lane and Lynn.[5]

The thermal capture cross section may be explained by assuming a contribution from a negative energy level. The resonance-integral discrepancy can be explained if there is a significant contribution from "missed levels" between 1 to 10 keV of *s*- or *p*-wave character. Isakov *et al.*[6] have reported a new level near 1200 eV; however, they conclude that it is quite insufficient to account for the measured resonance integral. Furthermore, their measured capture cross section appears to exhibit a dependence on neutron velocity which is quite accurately $1/v$.

The purpose of the present investigation is an attempt to clarify these ambiguities in the neutron capture of iron by searching for levels previously missed in transmission work, to measure the parameters of such levels, particularly Γ_γ, the radiation width, and to investigate with more precision the shape of the total cross section as a function of energy up to 3 or 4 keV in an attempt to evaluate the effects of levels below the neutron binding energy.

The fast chopper facilities at Chalk River and Brookhaven were employed in this investigation. Using an array of six 3-in.×3-in. NaI crystals the following capture gamma-ray measurements were made at Chalk River: (1) gamma-ray spectra for both thermal and the 1200-eV resonance; (2) capture-area measurement of the 1200-eV level; (3) self-indication transmission measurements on the 1200-eV level obtained by cycling a sample in transmission in conjunction with a sample in the gamma-ray detection station.

Conventional transmission measurements up to 4 keV were made with the Brookhaven fast chopper. No levels other than one at 1200 ± 30 eV were discovered in these experiments. The parameters of the 1200-eV resonance, however, were determined and its spin, parity, and isotopic assignment were deduced from the gamma-ray spectrum. The parameters of the negative energy resonance were also obtained by curve fitting, on the assumption that a single such level, in combination with the known positive levels, is responsible for the shape of the σ_T curve for several keV above thermal energy. The various experiments will be described later in the following sections.

It is appropriate to point out here that certain advantages may materialize when one uses a capture gamma-ray detector to investigate the properties of neutron resonances. In the present experiment we are interested in identifying weak levels which, conse-

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[1] An estimate of $\gtrsim0.8$ eV has been made for the radiation width of the resonance near 1200 eV by Isakov *et al.* (Ref. 6).

[2] H. Goldstein and M. H. Kalos, NDA Report 2-62, Nuclear Development Corporation of America, 1957 (unpublished).

[3] *Neutron Cross Sections*, compiled by D. J. Hughes and R. B. Schwartz (U. S. Government Printing Office, Washington D. C., 1958), 2nd ed.

[4] A. E. McArthy, P. J. Persiani, B. I. Spinrad, and L. J. Templin, ANL Reactor Physics Constants Center Newsletter No. 1 (1961).

[5] A. M. Lane and J. E. Lynn, Nucl. Phys. **17**, 563, 586 (1960).

[6] A. I. Isakov, Yu. P. Popov, and F. L. Shapiro, Zh. Eksperim. i Teor. Fiz. **38**, 989 (1960) [translation: Soviet Phys.—JETP **11**, 712 (1960)].

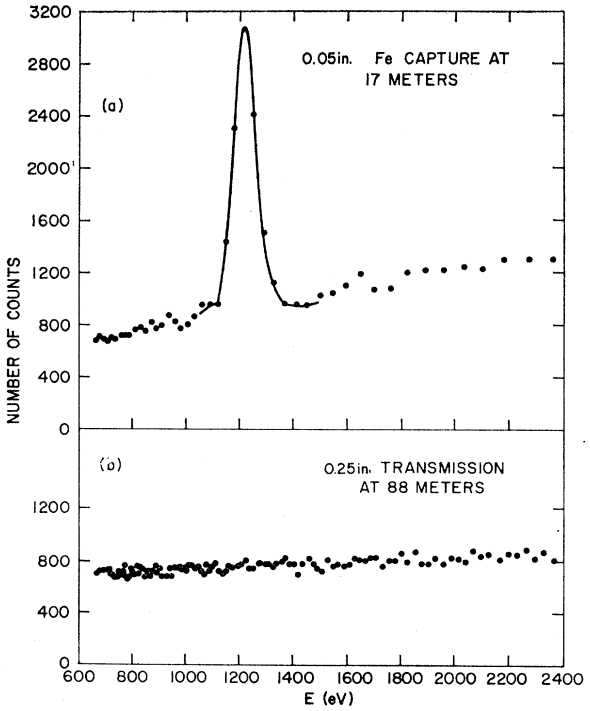

FIG. 1(a) The 1200-eV resonance in Fe^{56} as detected by an array of NaI crystals. A target foil of 0.015 in thickness was placed at 17 m from the chopper. The nominal time resolution is 75 nsec/m. (b) A counting rate curve for neutrons transmitted through a 0.25 in slab of iron and detected by a BF_3 counter array at 88 m from the chopper. The nominal time resolution is 15 nsec/m.

quently, may have eluded observation in transmission. Figure 1(a) shows a plot of capture gamma rays above 2 MeV observed from a 15-mil Fe sample by the NaI-crystal array stationed at 17 m from the Chalk River fast chopper. The corresponding neutron energy resolution is 75 nsec/m. Figure 1(b) shows the corresponding transmission of the incident beam through a $\frac{1}{4}$-in. Fe sample taken at the Chalk River 88-m station; a neutron energy resolution of 15 nsec/m. Despite the use of inferior neutron energy resolution and a relatively thin sample, the 1200-eV level is very clearly seen by the observation of capture gamma rays, but is almost unobservable in transmission. Evidently, the use of a capture gamma-ray detector makes possible the determination of level parameters for narrow resonances that cannot even be seen in a conventional transmission measurement. There are two principal reasons why this is so:

(1) The background is inherently lower for a capture gamma-ray measurement where a peak is observed rather than a dip as in the transmission case. Put in another way, one looks at the difference of two small numbers instead of a comparable difference in two very large numbers.

(2) For many resonances strong high-energy gamma rays are emitted. This enhances the favorable background situation mentioned above. It should be noted here that this condition may be present in the case of resonances due to p-wave capture, where favorable spin and parity assignments permit transitions not allowed for the s-wave case.

The favorable circumstances present in capture gamma-ray measurements may be further utilized in the self-indication method,[7] which will be described in detail in a following section. Here we note merely that it makes possible resonance parameter analysis where the neutron resolution is rather poor, as is the case with choppers in the keV region.

II. 1200-eV RESONANCE PARAMETERS

The determination of a complete set of Breit-Wigner parameters for a resonance requires, in general, a measurement of both total and partial cross sections, e.g., thick and thin sample transmission areas plus scattering or capture area. However, in the present case, the use of conventional transmission measurements is extremely difficult because of the resolution and background problems previously discussed. The weak strength of the resonance at 1200 eV indicates that $\Gamma_\gamma \gg \Gamma_n$. Consequently, a combination of a capture area ($\sigma_0\Gamma_\gamma$) measurement with a self-indication (σ_0) measurement is optimum for determination of the parameters $\Gamma\gamma$ and $g\Gamma_n$. It is shown below that the resonance capture gamma-ray spectrum is consistent only with s-wave capture in Fe^{56}, which implies that $g=1$.

A. Resonance Capture Spectrum

The capture gamma rays from the 1200-eV resonance were observed by the array of six 3-in.$\times$3 in. NaI detectors and recorded in an electrostatic memory two-parameter analyzer.[8] In addition, the Fe-capture spectrum due to thermal neutrons has also been measured. Using line shapes measured for 4.43-MeV and 7.73-MeV gamma rays, the spectra have been unfolded into the constituent lines by means of a least squares program written by Ferguson.[9] The results of thermal capture have been compared to those of Groshev,[10] and to the measurements on the 1200-eV level. The relative intensities for the thermal capture gamma-ray transitions are in satisfactory agreement with those obtained by Groshev (Table I). Figure 2 shows the comparison between thermal and resonance capture. Note the similarities between the two spectra; the relative intensities are summarized in Table I. The presence of the strong 7.6-MeV gamma rays as well as the weaker lines shows that the resonance is formed by s-wave capture in

[7] J. L. Rosen, J. S. Desjardins, J. Rainwater, and W. W. Havens, Jr., Phys. Rev. **118**, 687 (1960).
[8] R. E. Chrien, H. H. Bolotin, and H. Palevsky, Phys. Rev. **127**, 1680 (1962).
[9] A. J. Ferguson, Atomic Energy of Canada, Ltd. Report 1398, (1962) (unpublished).
[10] L. V. Groshev, A. M. Demivov, V. N. Lutsenko, and V. I. Pelekov, *γ-Ray Spectra from Radiative Capture of Slow Neutrons* (Pergamon Press, Inc., New York, 1959).

TABLE I. Relative intensity of capture gamma rays from Fe^{56}.

E_γ	Thermal capture[a]	Thermal capture[b]	Thermal capture[c]	1.2-keV neutron capture[d]
7.639	100	100	100	100
7.273	9.7	17	23	25
6.932	...	...	...	35
6.440	...	2.2	...	2.6
6.373	1.1	...	−1.6	...
6.009	15.5 } 30	25 } 53	} 50	} 49
5.911	14.5 }	28 }		

[a] B. B. Kinsey and A. Bartholomew, Phys. Rev. **89**, 375 (1953).
[b] L. V. Groshev *et al.*, J. Nucl. Energy **3**, 258 (1956). See Ref. 10.
[c] Present work.
[d] Present work.

Fe^{56}. Further corroboration of this assignment was provided in the self-indication measurement described below.

The appearance of a remarkably strong ground-state transition in resonance capture somewhat weakens the argument that the presence of a strong ground state transition in thermal capture is evidence for a direct capture effect in Fe. Here we see the same effect occurring through compound nucleus formation.

The strong similarities in the thermal and the 1200-eV resonance spectra made practical the measurement of the resonance capture area $\sigma_0\Gamma_\gamma$ by using the known Fe thermal capture cross section as a normalization.

B. Capture Area Measurements

The counting rate, integrated through the 1200-eV resonance, as measured by the NaI gamma-ray detector,

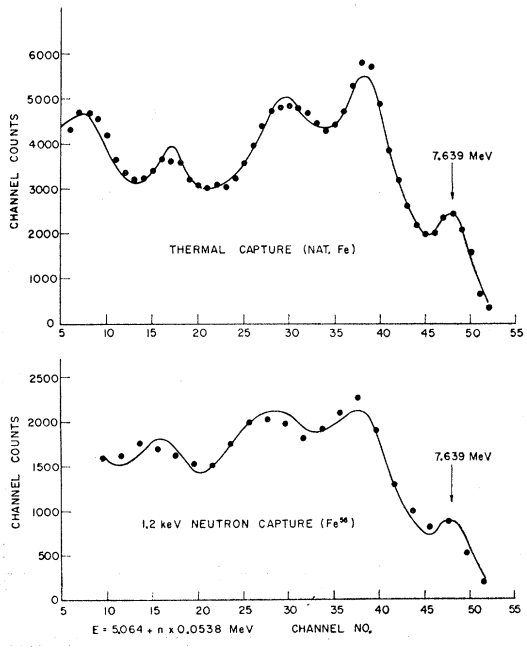

FIG. 2. A comparison between the spectra of gamma rays following resonance and thermal neutron capture in iron. The gammas are detected in 3 in.×3 in. NaI crystals.

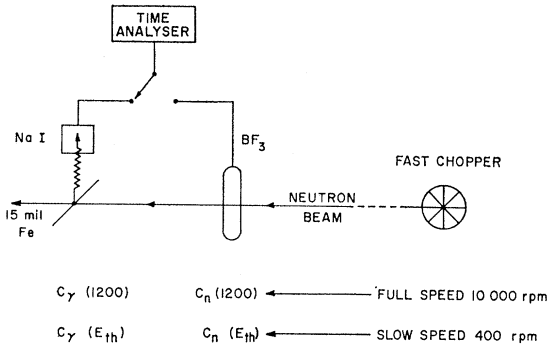

FIG. 3. A schematic representation of the experimental arrangement for the capture area measurements.

is

$$C_\gamma(1200\text{ eV})=\left[\int_{\text{res}}(1-e^{-n\sigma(E)})dE\right]N(1200)\frac{\Gamma_\gamma}{\Gamma}\Sigma_\gamma, \quad (1)$$

where Σ_γ is the efficiency for the detection of a neutron-capture event. Since the resonance capture spectra is very similar to that from thermal capture, the efficiency Σ_γ was measured in terms of the known thermal neutron capture cross section of Fe;

$$\Sigma_\gamma=\frac{C_\gamma(E_{\text{th}})}{n\sigma_{\text{cap}}(E_{\text{th}})N(E_{\text{th}})}. \quad (2)$$

From (1) and (2), the capture area A_γ is

$$A_\gamma=\left[\int_{\text{res}}(1-e^{-n\sigma(E)})dE\right]\frac{\Gamma_\gamma}{\Gamma}$$
$$=\frac{C_\gamma(1200)}{C_\gamma(E_{\text{th}})}\frac{N(E_{\text{th}})}{N(1200)}n\sigma_{\text{cap}}(E_{\text{th}}). \quad (3)$$

The ratio of the incident neutron flux near thermal and resonance was measured by a BF_3 counter approximately 1 m ahead of the gamma-ray detector. Since this counter is known to be $1/v$ throughout this range, the capture area, expressed in measured quantities, is

$$A_\gamma=\frac{C_\gamma(1200)}{C_\gamma(E_{\text{th}})}\frac{C_n(E_{\text{th}})}{C_n(1200)}\left(\frac{0.0253}{1200}\right)^{1/2}n\sigma_{\text{cap}}(0.0253). \quad (4)$$

Figure 3 shows, schematically, the experimental arrangement. Four separate time-of-flight spectra were measured to determine C_γ, $C_n(1200\text{ eV})$ and C_γ, $C_n(E_{\text{th}})$. The total counting rate in the BF_3 counter was used to monitor the incident beam in each case.

The 1200-eV resonance measurements were made at the 17-m station using the maximum chopper speed of 10 000 rpm, i.e., a neutron-energy resolution of 75 nsec/m. Gamma rays were observed from a 3-in.×3-in., 15 mil foil of SAE 1070–1090 steel (98.3% Fe) at 45° to the beam. The detector discriminator bias corresponded to 3-MeV gamma rays. Discriminator settings

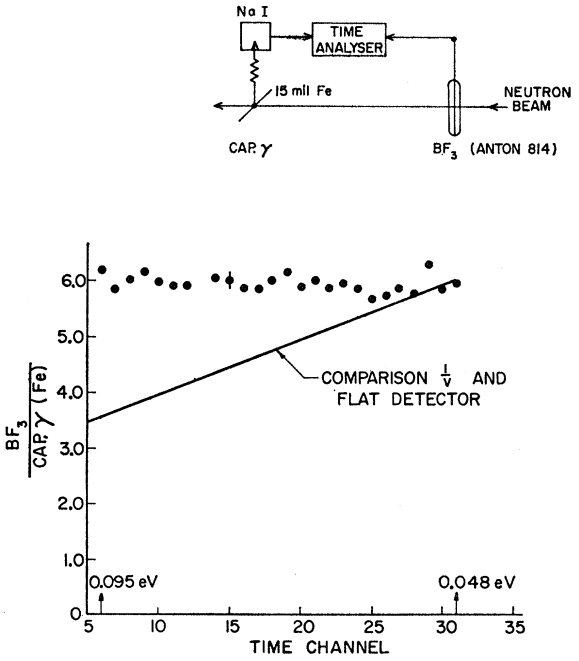

FIG. 4. The ratio of counting rates from a BF_3 detector and from the Fe foil viewed by NaI detectors from 0.05 to 0.10 eV. The solid line is drawn to indicate the ratio which would be obtained if the $1/v$ BF_3 detector were compared to a detector whose efficiency does not vary with energy. The constant value of the ratio with energy indicates that the iron foil is a $1/v$ absorber in this region.

of both the gamma ray and BF_3 detector were checked daily. Drifts of less than 1% were observed and tolerated. The background for the capture detector was determined from the "wings" of the 1200-eV resonance.

The procedure was repeated with the chopper at 450 rpm to cover the energy range of 0.05 to 0.10 eV. Time-of-flight spectra from both detectors were again recorded. The background radiation in the thermal capture measurement is determined from a run with a carbon scatterer of equivalent neutron scattering power replacing the iron target. Figure 4 shows the ratio of neutron to gamma ray counts is independent of energy in this energy region. This result is expected since the thin Fe foil and BF_3 counter act as $1/v$ absorbers.

Using a value of the Fe thermal-capture cross section of 2.02 ± 0.06 barns, A_γ was measured to be 0.677 ± 0.030 eV.

Two possible corrections to the above value of A_γ have been considered. The first concerns the effect of elastic neutron scattering and probability of subsequent capture. Since the foil is thin to both thermal and resonance neutrons, the correction has been estimated by considering only single scattering. An elementary calculation shows that the ratio of secondary captures (those captures occurring after a single scattering) to primary captures is just the product $n_{av}\sigma_s$, where n_{av} is the average path length for scattered neutrons in atoms/b, and σ_s is the scattering cross section. At near thermal energies, the scattering cross section is about 11 b and the total correction to the observed capture rate is -3.5%, where the foil self-shielding is included in addition to the scattering effect. At the resonance, neutrons which scatter suffer an energy loss, $\Delta E \gg \Gamma$, on the average, and so escape from the resonance. However, neutrons of energies higher than the resonance energy may scatter with such an energy loss as to result in resonance capture. The instrumental neutron energy resolution is poor enough so that such captures will be included in the measured resonance capture area. The number of neutrons scattered into the energy region of the resonance via potential scattering is equal to the number scattered out of that energy region by potential scattering. Application of the thin sample approximation used in the thermal case results in a net correction of -3.1%, including the foil self-shielding factor. Consequently, to an accuracy of $\approx 0.4\%$, the effects of neutron scattering in the determination of A_γ are self-cancelling.

Consideration has also been given to the validity of the assumption that the capture gamma-ray detector is equally efficient for resonance and thermal capture. Figure 5 shows the observed ratio $C_\gamma(1200)/C_\gamma(E_{th})$ as a function of discriminator bias; this has been computed from the measured spectra shown in Fig. 2. The departure from a constant ratio is a consequence of the spectral differences; the effect is not large but it is difficult to evaluate. Therefore, we have assigned to the measured value of $C_\gamma(1200)/C_\gamma(E_{th})$ an error of 11% equal to the mean deviation from constancy. Therefore, $A_\gamma = 0.677 \pm 0.073$ eV. This result when corrected for self-protection effects involving the σ_0, as measured by the self-indication method, leads to the following:

$$\sigma_0\Gamma_\gamma = 110 \pm 12 \text{ eV b.}$$

This result is somewhat larger than the previously reported Russian value of 74 ± 7 eV b.[5] The value of the

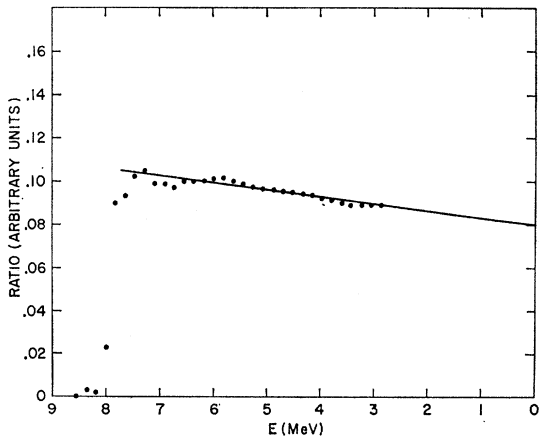

FIG. 5. The ratio of the cumulative gamma-ray spectra, as observed in 3 in.×3 in. NaI, from resonance and thermal capture. A straight line is fitted to the data between 3 and 7 MeV, and used to extrapolate the ratio to zero energy.

radiation width deduced using the σ_0 measured below is

$$\Gamma_\gamma = 0.673 \pm 0.074 \text{ eV}.$$

C. Self-Indication Experiment

The self-indication method is a transmission measurement where the detector is a foil of the same material as the transmission foil. Figure 6 is a simple schematic illustration of the experimental arrangement. Neglecting multiple-scattering effects (which are small for this case where $\Gamma_\gamma \gg \Gamma_n$), the number of interactions in the resonance which result in gamma ray emission is simply

$$N_\gamma = \left[\int N(E)(1-e^{-n_D\sigma(E)})dE\right](\Gamma_\gamma/\Gamma)$$

in the absence of a transmission foil. With a transmission foil placed near the chopper $N(E)$ is modified and we have

$$N'(\gamma) = \left[\int N(E)(e^{-n_T\sigma(E)})(1-e^{-n_D\sigma(E)})dE\right](\Gamma_\gamma/\Gamma).$$

The measurement is of $N(\gamma)$ and $N'(\gamma)$ and we form the ratio $R = N'(\gamma)/N(\gamma)$.

The ratio R is a function of $n_T\sigma_0$ and Δ/Γ, where n_T is the transmission foil thickness and Δ is the resonance Doppler width. For the sample thicknesses used it can be demonstrated that this ratio is almost independent of n_D, the detector foil thickness, and only weakly dependent on Δ/Γ. A computer program has been written to relate this ratio R to the sample parameter $n_T\sigma_0$. The result is combined with the $\sigma_0\Gamma_\gamma$ as measured above and the analysis reiterated to account for the dependence of R on Δ/Γ. The iterative analysis is rapidly convergent in this case. A transmission foil of 60 mils of SAE 1010 steel was used and three separate detector foils of 15, 30, and 50 mils of steel were run. Table II gives the individual results. The average value is

$$n_T\sigma_0 = 1.92 \pm 0.14.$$

For the transmission foil $n_T = 0.01164$ atoms/b for Fe^{56}; therefore,

$$\sigma_0 = 165 \pm 12 \text{ b}.$$

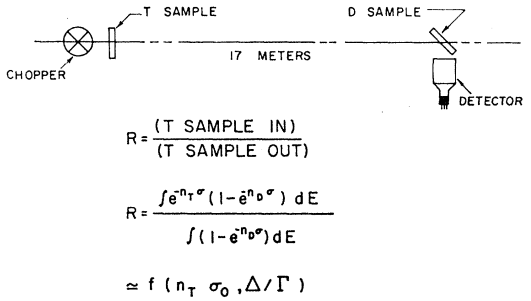

FIG. 6. A schematic diagram of the experimental arrangement for the self-indication measurements.

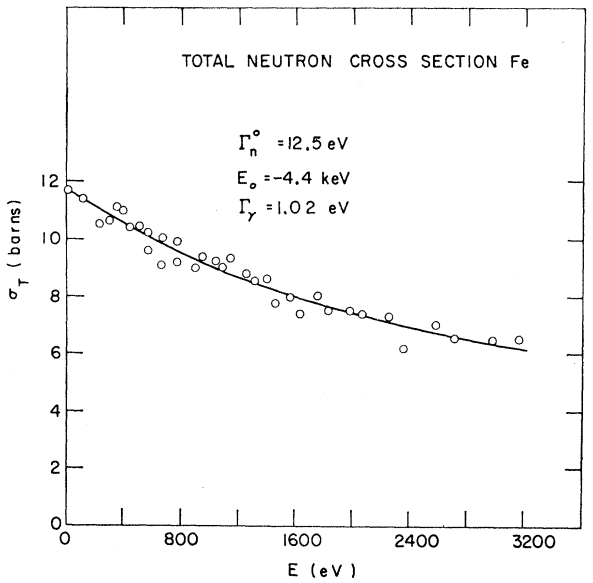

FIG. 7. The total cross section of iron up to 3 keV as measured with the BNL chopper at a resolution of 70 nsec/m. The parameters shown are for a neutron-bound level which will best fit the observed curve.

The measured value of $n_T\sigma_0$ taken with the known isotopic abundances of Fe^{54}, Fe^{57}, and Fe^{58} is enough to rule out the possibility that the resonance is in one of those isotopes, and this confirms the isotopic assignment made on the basis of the capture gamma-ray spectrum mentioned above. For example, the peak cross section, assuming the resonance to be in F^{58}, is 2600 ± 180 b whereas the maximum possible σ_0 is 2160 b for an s-wave resonance at this energy.

III. Fe TOTAL CROSS SECTION

In parallel with the capture area and self-indication measurements carried out at the joint BNL-AECL facility at Chalk River, an investigation of the total neutron cross section of iron was carried out with the fast chopper at BNL from 1 eV up to an energy of 5 keV. It has been suggested that the cross section in this region varies with energy in such a way as to suggest the effective presence of a level below the neutron binding energy. Not only does the shape of the total

TABLE II. Results of self-indication measurements.

Foil thickness	Self-indication ratio[a]	σ_0 (b)
0.060 transmission 0.015 capture	0.684±0.034	153±15
0.060 transmission 0.030 capture	0.688±0.023	149±10
0.060 transmission 0.050 capture	0.625±0.027	193±20

[a] The ratio of areas derived from gamma-ray counts from the capture sample with transmission sample in and out of the beam, corrected for potential scattering in the transmission sample.

cross-section curve suggest such a level, but the size of the thermal capture cross section supports this suggestion. The experiment was carried out with a piece of mild steel (99.5% Fe) of nominal thickness ½-in. and an actual thickness of 0.1066 Fe atoms/b.

The experimental data up to 3 keV are shown in Fig. 7. Above that energy resonances in Fe^{54} appear and the data are inferior to those of the Van de Graaff groups.[11] Note that there is no indication of the narrow 1200-eV resonance in these data.

The experimental data were fitted by the method of least squares to the following function:

$$\begin{aligned}\sigma(E)=&0.651[\Gamma_n^0(29\text{ keV}]^2/[E-(29\times10^3)]^2\\&+0.5722(R)[\Gamma_n^0(29\text{ keV})]/[E-(29\times10^3)]\\&+0.651(\Gamma_n^0)^2(E-E_0)^{-2}\\&+0.5722R'\Gamma_n^0(E-E_0)^{-1}\\&+4\pi R'^2/100,\end{aligned}$$

where Γ_n^0 (MeV), E_0(eV) are the reduced width and resonance energy for the neutron bound level.

R'(F) is the effective radius parameter and Γ_n^0(29 keV) is the reduced width of the Fe^{56} resonance at 29 keV. The other resonances in iron do not effectively contribute in this energy region. The terms above represent resonance scattering, potential scattering and resonance-potential interference scattering. The level-level interference and resonance absorption terms have been evaluated and are negligible. The least-squares fitted values for the negative energy resonance are as follows

$$\begin{aligned}E_0&=-4390\pm1700\text{ eV},\\\Gamma_n^0&=12.5\pm8.1\text{ eV},\\R'&=3.9\pm1.9\text{ F}.\end{aligned}$$

With the knowledge of the thermal-capture cross section of Fe^{56} (2.7 b), the Γ_γ for this level may be deduced from

$$\Gamma_\gamma=\sigma_{\text{th}}(E_0)^2/\Gamma_n^0(4.093),$$

where the units are as given before. The result is $\Gamma_\gamma=1.02\pm0.46$ eV. The result is, unfortunately, subjected to the large errors inherent in curve fitting to the "tails" of distant resonances; however, within this error it is the same as the radiation width for the 1200-eV resonance. The value of R' necessary to fit these data is surprisingly low compared to the value of 6.7 F obtained by Bilpuch *et al.*[12] It was not possible to obtain a satisfactory fit using the latter value.

V. DISCUSSION

The radiation width herein reported, 0.670 eV, is near the values generally quoted for neighboring nuclides. For Mn^{55} Levin and Hughes[13] calculate a radiation width of ~0.500 eV from the assumption that the 337-eV level accounts for all of the thermal-capture cross section. The 132-eV resonance of Co^{59} has a measured radiation width of 0.400±0.04 eV.[14]

The resonance absorption integral has been measured and the currently accepted mean of those measurements is 2.2±0.2 b.[4] The non-$(1/v)$ portion of the resonance integral is obtained by subtracting one half of the thermal cross section and is, therefore, 1.0±0.2 b. We may compute this reduced resonance integral from the known resonances in Fe below 30 keV. It is easy to show that resonances above this energy have a negligible contribution to the resonance integral for any reasonable assumption for the radiation widths.

The resonance integral may be written as

$$\int_\gamma=\sum_i f_i\frac{4090}{E_i^2}\frac{g\Gamma_{ni}\Gamma_\gamma}{\Gamma},$$

where Γ_γ is given in meV, and f_i is the isotopic abundance. The contribution for the 1200-eV level, calculated from the parameters measured in the present experiment is

$$0.134\pm0.010\text{ b}.$$

The balance of the known resonances contributes very little to the resonance integral (about 0.01 b).

Thus, it is clear that the 1200-eV resonance does not explain the discrepancy between the calculated and observed resonance integral. Since any capture cross section goes as $1/v$ (including level-level interference terms) near zero energy, it is difficult to account for the non-$(1/v)$ capture in iron.

ACKNOWLEDGMENT

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[11] *Neutron Cross Sections*, compiled by D. J. Hughes, B. A. Magurno, and M. K. Brussel (U. S. Government Printing Office, Washington D. C., 1960), 2nd ed.

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