

A Review of the Total Radiation
Widths of the Neutron Resonances
of ^{238}U

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Abstract

An investigation has been made into the present uncertainties in the total radiation widths Γ_γ of the fertile material ^{238}U . Recent measurements of $\langle\Gamma_\gamma\rangle$ have a wider dispersion than are required in the fast breeder reactor program. In addition, the possibility of fluctuations in Γ_γ from resonance to resonance has been considered. We conclude that the quoted uncertainties in Γ_γ are probably underestimated, and that larger statistical errors would be more consistent with the various published parameter sets. Our best choice for $\langle\Gamma_\gamma\rangle$ was 23.55 meV which we obtained by weighting the recently reported data. Little evidence was found for a quasi-periodic structure of Γ_γ versus energy, with most of the variance in individual values of Γ_γ from the mean due to experimental uncertainties.

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The values of the total radiation widths of the neutron resonances of ^{238}U are among the most useful and important physical quantities in the present stage of the technological development of fission reactors. The importance of these values stems from the potential conversion of the largest fraction (99.3%) of naturally occurring uranium (^{238}U) into the fuel material ^{239}Pu by means of the radiative capture of neutrons. In present power reactors, electrical energy is produced by the fissioning of the 0.7% abundant ^{235}U isotope, with only a small fraction of the total energy coming from the fast fissioning of ^{238}U . It is possible, however, by careful engineering, to increase the production of ^{239}Pu from ^{238}U to the point where breeding occurs, and more fuel materials are produced than consumed. Breeding is accomplished by a careful choice of isotopes in the core of a nuclear reactor and their physical arrangement. In order to obtain a high breeding ratio the average energy of the neutron spectrum must be considerably higher than the "thermal" spectrum in present day reactors.

The radiation width, Γ_γ , in ^{238}U determines the probability of a capture reaction in ^{238}U and the ratio Γ_γ/Γ gives the relative probability of the capture reaction to other possible reactions. Thus, the radiation widths of the resonances, along with their average values and variances, are related to the breeding ratio of a fast reactor, and directly connected to the production and cost of nuclear fuel, potential reserves for sources of electrical power, and especially the availability (or unavailability) of cheap power. A change of a few per cent in $\langle \Gamma_\gamma \rangle$ has a large effect in terms of fuel breeding times and the eventual cost of power. The present uncertainty in $\langle \Gamma_\gamma \rangle$ of about 20% corresponds to an uncertainty of $\sim 3\%$ in the effective multiplication of a reactor factor k and 10% in the value of the breeding ratio.¹

The radiation widths of the resonances, their variance and average value are also important in safety aspects of the design of fast reactors. The Doppler effect broadens the resonance as the temperature increases. This broadening leads to increased capture of neutrons with increased temperatures because it decreases resonance escape probability in a ^{238}U rod in the reactor. The negative temperature coefficient of ^{238}U increases the safety of the fast reactor. The Doppler effect on the self-shielding of a fuel rod leads to a positive temperature coefficient which decreases the safety of a fast reactor. The temperature coefficient of a fast reactor will depend on the capture and fission widths of the fertile and fissile isotopes and the geometrical configuration of the design. Thus, the knowledge of the capture width of ^{238}U is important, for both the economic and safety aspects of fast reactor design.

Because of the importance of the radiation widths in ^{238}U , measurements of Γ_γ are frequently repeated as experimental techniques improve. With better ex-

perimental techniques the uncertainties in the Γ_γ values and other resonance parameters decrease. The more accurate values of Γ_γ allow the variation of Γ_γ with energy to be examined for the possibility of periodic structure and/or for correlations between the radiation widths and the reduced neutron widths. It is important to study the fluctuations in Γ_γ about $\langle \Gamma_\gamma \rangle$ in order to attempt to determine the reason for these fluctuations. Another recent development has been the long and short range correlations in the theory of neutron resonance spectrometry according to the Dyson and Mehta Theory. This new development provides additional information for the construction of pseudo-resonance ladders in the unresolved energy regions and improves the knowledge of $\langle \Gamma_\gamma \rangle$ in fast reactor calculations in the energy regions where this quantity cannot be measured.

The best and most recent studies on Γ_γ for ^{238}U were carried out at Harwell², Los Alamos Scientific Laboratory³, Geel⁴, Dubna⁵, and Columbia⁶. These measurements were all limited to the resonances in the energy region from thermal energy to about 2.5 keV. Considerable confusion has resulted because published values of individual Γ_γ values do not agree within their quoted uncertainties. The situation for $\langle \Gamma_\gamma \rangle$ is somewhat better, in that the various values reported tend to be in closer agreement, but in the recent measurements there is still about a 20% spread in the measured values which is much too large for this important quantity. The most controversial results have been those of Glass, et al.³ at LASL who analyzed nuclear explosion data and found a much lower value of $\langle \Gamma_\gamma \rangle$ than the other experimenters. Glass et al. also found evidences for quasi-periodic structure in the plot of Γ_γ values versus energy. Neither of these Los Alamos results have been able to be reproduced by subsequent measurements at Geel, Dubna and Columbia. We will attempt to review the present situation for ^{238}U and where possible attempt to determine the source of the uncertainties in the radiation widths for this important isotope.

Asghar, Chaffey and Moxon³ at Harwell published in 1966 their ^{238}U results for data obtained from 6ev to 823ev. They used the Harwell LINAC as a pulsed neutron source, and the time of flight technique (flight paths of 120 and 192m.) to resolve the resonances. Their data included scattering data in addition to capture data corrected for background, Doppler broadening and multiple interaction effects. Each sample thickness of the scattering and capture data yielded a different relationship between the resonance parameters (Γ_n , Γ_γ). The common intersection (in favorable cases) of all the curves in the (Γ_n , Γ_γ) plane determined the values of these parameters, and give an indication of their uncertainties. In general families of curves for different sample thicknesses for the same type of data (e.g. capture data) tend to have similar slopes, and thus the curves will intersect at small angles. Unfortunately, the intersections at small angles do not usually give a meaningful common intersection. The addition of a set of curves derived from scattering data often will be at right angles to the capture data, so that good determinations of both parameters result. Figure 1, taken from Re. 2, is an example of the analyses. The Harwell results, as well as all other time of flight measurements, are less reliable at higher energies because of the rapidly decreasing resolution as the energy increases. Data taken for capture experiments usually have another difficulty in that capture yields are usually normalized at low energy by some variation of the saturated resonance technique, in which the uncertainties increase as low energy saturated counting rates are extrapolated to higher energies. Asghar et al. determined Γ_γ for 27 resonances, with $\langle \Gamma_\gamma \rangle = 23.74 \pm 1.09$ meV. A variance analysis of their data yielded ~ 80 degrees of freedom.

Glass et al.³ at LASL published in 1968 their results on neutron capture measurements on ^{238}U using a neutron beam from an underground nuclear explosion. This source of neutrons has one great advantage over pulsed accelerator sources in that the extremely high intensity of the source has a very high signal to background ratio. As a result, nuclear explosion measurements usually "see" many more weak resonance levels than other methods. The fact that all the neutrons are a result of a single burst, however, requires substantially different experimental and recording methods than when the data is taken event by event.⁷ The neutron spectrum obtained from the nuclear explosion was also very different from that in accelerator sources. Results were found for neutron capture in individual resonances from 30 to 2000 eV. The total radiation widths were determined for 62 levels with $\langle \Gamma_\gamma \rangle = \{19.1 \pm 0.6 \text{ (stat.)} \pm 1.4 \text{ (syst)}\} \text{ meV}$. The Γ_γ values were obtained by using the obsolete values of Garg et al.⁸ for Γ_n^0 which came from 200 meter flight path transmission measurements at Columbia. In their more recent and complete results on ^{238}U (1972) the Columbia group⁶ have revised many of the older values of Γ_n^0 and found that above 1 keV the previous Γ_n^0 values were systematically lower than the recently measured values. These changes would imply a different set of Γ_γ parameters. Since the LASL results relied heavily on the Columbia reduced neutron width data, the results should be recalculated using the newer values of Γ_n^0 .

The distribution of Γ_γ for the Glass et al. data set resulted in a chi-squared distribution of $\nu=44 \pm 8$ degrees of freedom. A second approach tried was to fit the observed Γ_γ distribution with a constant (equal to 12 meV) plus a fluctuating part with 3.7 ± 0.7 degrees of freedom. Such distributions would result from the de-excitation of heavy nuclei by a small number of strong transitions to near-ground states. While there is some evidence⁹ that strong gamma transitions exist in thermal spectra for a small percentage of captures, the evidence for this "second alternative" does not seem to be very convincing. Glass et al. also found what appeared to be a significant variation in Γ_γ from level to level. This variation was quasi-periodic in nature and if real would imply possible intermediate structure in ^{238}U .

In light of the unexpected results of Glass et al., the Geel group of Rohr, Weigmann and Winter redid the ^{238}U measurements and presented their results at the 1970 Helsinki conference.⁴ Rohr et al. measured the radiation widths while another group at Geel¹⁰ (Carraro and Kolar) determined the reduced neutron widths. The experiment was done at the 60 MeV electron LINAC using a 60 meter flight path. A combined area analysis of the capture data with the transmission data yielded the radiation parameters for 28 resonances in the range 50 to 1000 eV, with a $\langle \Gamma_\gamma \rangle = (24.64 \pm 0.85) \text{ meV}$. This was in general agreement with the Harwell value of the mean radiation width, but very different and much higher than that of Glass. Rohr et al. pointed out that a value of Γ_γ as low as that of Glass et al. (19.1 meV) would give a resonance integral computed with this mean width more than 10 barns smaller than the experimental value of Kusters et al.¹¹ at Karlsruhe. Moreover, the Geel group could not reproduce the quasi-periodic structure in the Γ_γ values versus energy in the energy range from 50 to 1000 eV.

Recent measurements (1972) were performed at Dubna by Malecki, Pikeiner, Salamatin and Sharapov⁵. They obtained transmission and capture data using the

pulsed fast IBR reactor with a linear accelerator injector as a neutron source. A 500 meter flight path was used and the resolution was ~ 6 nsec/m. Radiation widths for 31 resonances between 66 and 1197 eV were obtained, with $\langle \Gamma_Y \rangle \sim 24$ meV. All the radiation widths fell within a very narrow range, with the smallest and largest reported values being 22 and 27 meV. Their distribution of Γ_Y had a very small variance and relatively high number of degrees of freedom. The variance for their individual Γ_Y values from $\langle \Gamma_Y \rangle$ does not exceed their quoted uncertainty in Γ_Y values. The very small range of Γ_Y values is totally inconsistent with the periodic fluctuations of Glass et al. The Dubna results for $\langle \Gamma_Y \rangle$ are also in much better agreement with those of Asghar and Rohr than the Glass value.

The latest published data (1972) on the radiation widths of ^{238}U resulted from a series of capture, self-indication and transmission measurements performed at Columbia University by Kahn et al.⁶ These measurements utilized the Nevis cyclotron and flight paths of 33 m, 40 m, and 200 m. The analysis used the area technique for the capture yields, self-indication ratios and transmission areas to obtain the (Γ_Y, Γ_n^0) parameters. Values of the radiation widths were obtained for 71 levels up to 2400 eV. The $\langle \Gamma_Y \rangle$ was $(22.9 \pm 0.5 \text{ (stat)} \pm 0.9 \text{ (syst)})$ meV with few values of Γ_Y more than 20% away from the mean. These results are consistent with a large number of possible γ transitions between the initial and final states. A chi-squared test yielded $\nu = 70$ degrees of freedom, with most of the spread in the individual Γ_Y values from the mean probably coming from experimental errors in the measured values, so that the true ν is probably very much larger. There was no evidence of significant periodic fluctuation in the Γ_Y values versus energy. The $\langle \Gamma_Y \rangle$ was in much better agreement with the Harwell, Geel and Dubna values than with that of LASL.

The reasons for the differences in the quoted values for the individual Γ_Y values and the mean $\langle \Gamma_Y \rangle$ are difficult to determine. Ideally, all the experiments are equivalent and therefore should yield parameters which agree to within the stated uncertainties. In fact, the experimental scatter in values often is quite large. There are several factors which contribute to the uncertainty in the results. Some of the more important are treated below.

There appears to be real differences in the data from the LASL nuclear explosion experiment and the pulsed accelerator results. Fig. 2, part (a) shows a portion of the experimental capture data from the LASL nuclear explosion results. This figure was taken from Ref. 3. Of particular interest are the two resonances at 937 and 958 eV. These two resonances are well separated from other resonances and have Γ_n values which are approximately the same. Experimental values of Γ_n for these two resonances range between 145 and 214 meV. Glass reports that the ratio of the capture yields of the two resonances are quite different, with $\Gamma_Y(937) / \Gamma_Y(958) = 1.7 \pm 0.1$. He found $\Gamma_Y(937) = 20.9 \pm 1.6$ meV. Inspection of his data would seem to confirm the large variation of Γ_Y in these two levels. However, in part (b) of Fig. 2 capture yields in the same energy region for the Dubna data show that the ratios of capture yields of the same resonances to be nearly equal and this is reflected in the ratio of the capture widths $\Gamma_Y(937)/\Gamma_Y(958) \sim 1.1$ with $\Gamma_Y(937) = 24 \pm 3$ meV and $\Gamma_Y(958) = 22 \pm 3$ meV. The Harwell and Columbia

data sets give similar results. Asghar at Harwell did not quote Γ_γ values for these resonances but Rahn et al. at Columbia found $\Gamma_\gamma(937) = 25 \pm 2$ meV and $\Gamma_\gamma(958) = 21 \pm 2$ meV confirming the Dubna values. Therefore the fluctuations reported by Glass seem to appear in his data, while not in the data of the other groups. The fundamental questions are whether or not the nuclear explosion is valid (i.e. correctly obtained) data and if so why are the data sets from the other laboratories, which seem more consistent, different? Satisfactory answers have not been found. Figure 3 taken from the paper by H. Malecki et al.⁷ show Γ_γ versus energy for the LASL and Dubna results. The differences in the fluctuations of the two are quite marked.

In part the uncertainties in the capture width Γ_γ are due to uncertainties in the neutron width Γ_n^0 . An example of this is found in the resonance parameters for the 347-eV level reported by the various groups. The 347 eV resonance is strong, well isolated and at low energy so that resolution and other problems associated with the data at higher energies are not significant. However, the dispersion of Γ_γ parameters for this level is quite large. Table 1 shows that experimental Γ_γ values are found from a low of 17.9 meV to a high of 28 meV. Figure 4 shows the Columbia results for a large number of different sample thicknesses and types of experiments for this resonance. Superimposed on this figure are the $(\Gamma_n, \Gamma_\gamma)$ values from Table 1. A close inspection of the figure shows that the pairs of values from Columbia and Harwell lie close to the set of curves by the Columbia group for their capture samples. This indicates that the capture experiments for the two groups yielded identical information, but that the wildly different Γ_γ values are due to the values of Γ_n^0 which were chosen. The Harwell value for Γ_n is, in retrospect, too low. They have a very pronounced dip in their beam spectrum at 340 eV in ^{238}U . Figure 4 also shows that the capture results from Dubna and Geel are about the same. The difference in Γ_γ reported is due to the differences in the value of Γ_n^0 chosen. The LASL is quite inconsistent with the results from the other laboratories (note that Γ_n^0 was used from an older Columbia determination). Differences between the Columbia and Harwell; Dubna and Geel; and LASL experimental Γ_γ parameters are not due to the uncertainty in Γ_n but inherent in the Γ_γ measurement itself. The uncertainties in the capture yields are caused mainly by background fluctuations due to nearby levels or statistics and the normalization of the capture data. The normalization problem leads to systematic differences and will be discussed below.

Fig. 4 shows that some fairly large discrepancies can occur for data taken during the same experiment. The results for the 40m and 200m transmission results for this level are greater than would be expected. For this reason it is desirable to have a large number of sample thicknesses in any series of measurements, and to measure the capture, transmission (or scattering) and self-indication of a sample at the same time. A large number of different measurements will yield a large body of information that will provide various cross checks and insure internal consistency of a given data set. The uncertainties quoted in experimental measurements of Γ_n and Γ_γ parameters which involve only a few different sample thicknesses are probably underestimated. Fig. 1, for instance, shows the 65.95eV level analysis taken from the Harwell results. The error bars indicate the estimated uncertainties in the capture and scattering areas, and the "most likely" values of Γ_n and Γ_γ obtained by a chi-squared technique. The error bars indicate a band of uncertainty on either side of the curves, in which the parameters should lie. For capture

data, errors due to counting are usually quite small but the data are subject to numerous other possible errors which are partly systematic in nature. The sources of these other errors include the absolute normalization of the data, corrections for multiple interaction effects, flux variations with energy, detector calibration and loss of gamma ray counts due to dead time and self-shielding. A number of these errors are usually treated as being partly statistical.¹² Treating these effects as entirely systematic gives too low an estimate on the statistical error on Γ_γ and too much weighting of the capture results over the scattering (or transmission) data. On the other hand the assumption that the errors are entirely statistical clearly gives a too high uncertainty in the individual Γ_γ parameters. Each experimental group has the problem of dividing the errors into a statistical and systematic part. We are inclined to choose somewhat larger statistical errors than most groups. The variations in results taken at Columbia under slightly different experimental conditions such as different flight paths or detectors indicate these larger statistical errors. Therefore, our choice of Γ_γ given the results in Fig. 1 would be approximately 26 ± 2.5 meV, giving small weighting to capture result (b) because of its large uncertainty and noting that only results from two curves, (a) and (c) determine our result.

The normalization process of the capture data, even when carefully done, gives rise to systematic uncertainties in the Γ_γ values. Various methods are used to normalize the measured capture yield. The LASL nuclear explosion results were determined relative to the ${}^6\text{Li}(n,t){}^4\text{He}$ cross section which is quite straight below 100 keV. The referenced Li cross section¹³ appears to be ~4% higher between 20 and 80 keV than a more recent determination.¹⁴ This would imply that the Glass capture data would systematically be at least 4% too low. The Geel normalization method used the ${}^{10}\text{B}(n,t){}^7\text{Li}$ reaction, and assumed the ${}^{10}\text{B}$ cross section varied as $(1/v)$ in the region of interest. The absolute calibration of the product of detector efficiency times neutron flux was done with the "black resonance" technique for one resonance at 6.67eV in ${}^{238}\text{U}$. In this method care must be taken because the ${}^{10}\text{B}$ becomes progressively more transparent to neutrons. Thus the spatial distribution of γ rays produced in the ${}^{10}\text{B}$ changes, and counting losses and γ ray self-shielding may be energy dependent. These effects may cause an apparent distortion in the $(1/v)$ dependency of cross section of the reference material, which could lead to systematically increasing Γ_γ values.

The Columbia group determines the product of the saturated counting rate times detector efficiency for a number of resonances of ${}^{238}\text{U}$. Also, Au capture samples are run separately, and together with ${}^{238}\text{U}$ in the beam to determine the absolute normalization and its energy dependence at a series of energy points. The estimated systematic uncertainty in the Columbia data was $\pm 5\%$ when uncertainties due to counting losses, gamma ray self-shielding, multiple interactions and energy dependence were added to this absolute normalization error.

At Dubna the data normalization was also accomplished by determining the product of neutron flux times detector efficiency. Resonances in ${}^{238}\text{U}$ with $\Gamma_n \ll \Gamma_\gamma$ were used. A thin boron film placed in the neutron beam was employed to obtain the energy dependence of the flux. This is similar to

Harwell's approach to the normalization problem. The absolute value of the capture cross section and Γ_γ parameters are due to a normalization of low energy resonances, and a determination of the shape of the neutron flux as a function of energy. It is estimated that the normalization was accurate to $\pm 3\%$ for the Harwell data below 100eV and rising to $\pm 7.5\%$ at 50keV. For all groups, the uncertainties in the normalization are at a minimum at low energy.

Pinning down the source of the uncertainty in the individual Γ_γ values and $\langle \Gamma_\gamma \rangle$ is a complex task. Differences in the way different experimental groups collect and process their data, as well as the complexity of the measurement are difficult to assess. It is clear that the differences in the data sets has not been completely resolved. We are inclined to regard the LASL value for $\langle \Gamma_\gamma \rangle = 19.1$ meV as being too low and well outside the range of error for other, more recent measurements. The remaining published values of $\langle \Gamma_\gamma \rangle$ after removing the LASL value lie between 22.9 and 24.6 meV. Our best choice for $\langle \Gamma_\gamma \rangle = 23.55 \pm 0.16$ meV. We arrived at this value by weighting the Harwell, Geel, Dubna and Columbia data, taking into consideration the relative completeness of the data, errors quoted by the various groups and the time the measurement was performed. The LASL data was neglected. The relative weights we assigned were somewhat arbitrary. Our choice of 23.55 meV for $\langle \Gamma_\gamma \rangle$ is within the quoted errors of all the groups with the exception of the LASL results. The discrepancy in the nuclear explosion results of LASL remains unexplained. All the other data sets show little evidence for periodic structure in Γ_γ . Most individual values of Γ_γ observed lie close to the mean. Chi-squared analyses yield a quite high number of degrees of freedom, with most of the variance due to uncertainties in the measurements themselves. We therefore conclude that there is no significant variation of Γ_γ from level to level.

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TABLE 1
Resonance Parameter Results for the 347 eV Level
in ^{238}U

Γ_{γ} (meV)	Γ_n (meV)		
26 $\pm$ 3	80 $\pm$ 6	Rahn et al.	Columbia(1972)
23.5 $\pm$ 1.4	81.7 $\pm$ 1.6	Rohr et al.	Geel
22 $\pm$ 2	78 $\pm$ 10	Melecki et al	Dubna
20.4 $\pm$ 1.9	57.4 $\pm$ 3.3	Asghar et al.	Harwell
17.9 $\pm$ 1.5	82 $\pm$ 7*	Glass et al.	LASL
		*Garg et al.	Columbia(1964)

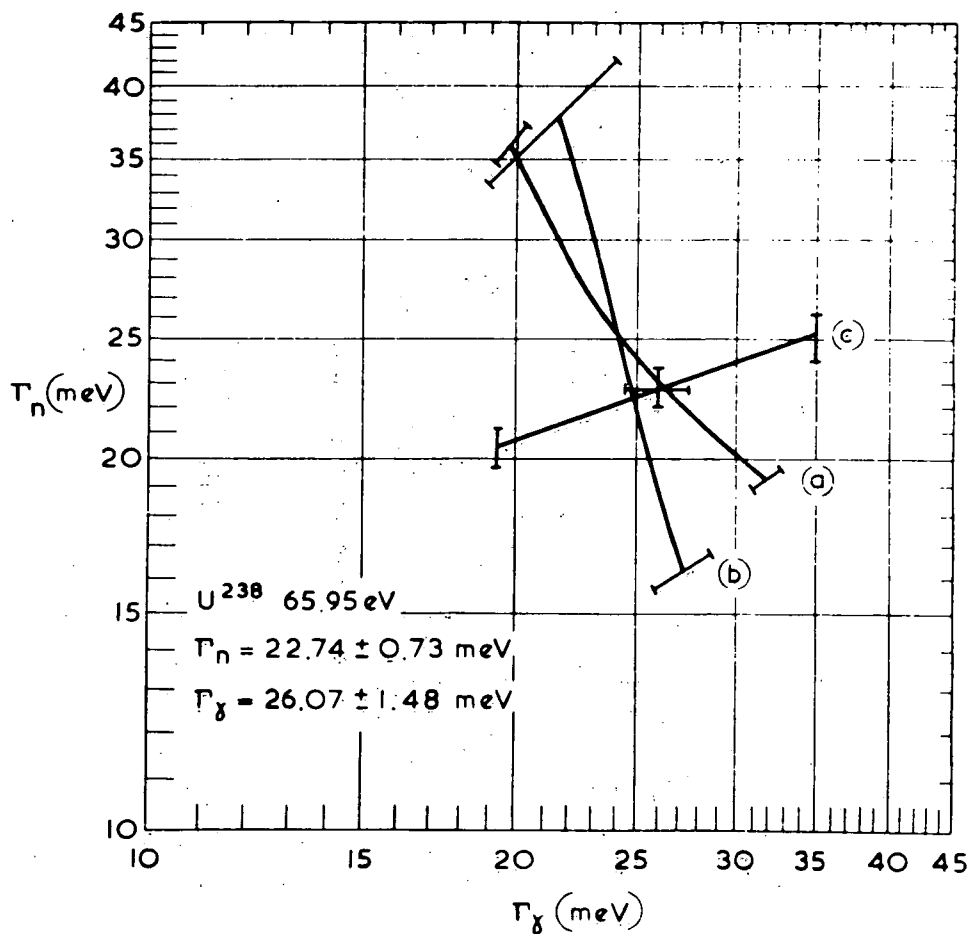


FIGURE 1

An example of the area analysis used in finding the neutron resonance parameters. This figure shows the results for the 69 eV resonance in ^{238}U as obtained at Harwell. Curves (a) and (b) are from capture samples with thicknesses of 1.17×10^{-4} and 2.56×10^{-3} atoms/barn and curve (c) is from scattering data with a sample thickness of 1.21×10^{-4} atoms/barn. In favorable cases the common intersection of the curves determines Γ_γ and Γ_n .

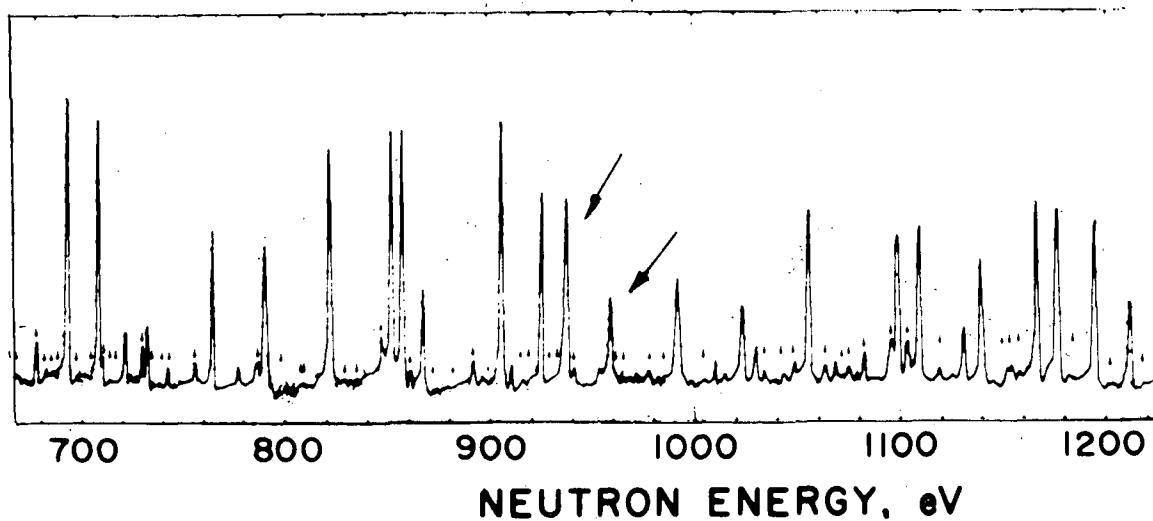


FIGURE 2(a)

A portion of the experimental data from the LASL atomic explosion measurement. These results are from the capture of neutrons in the first sample in the beam with a thickness of 5×10^{-3} atoms/barn. The two large arrows indicate the levels at 937 and 958 eV discussed in the text.

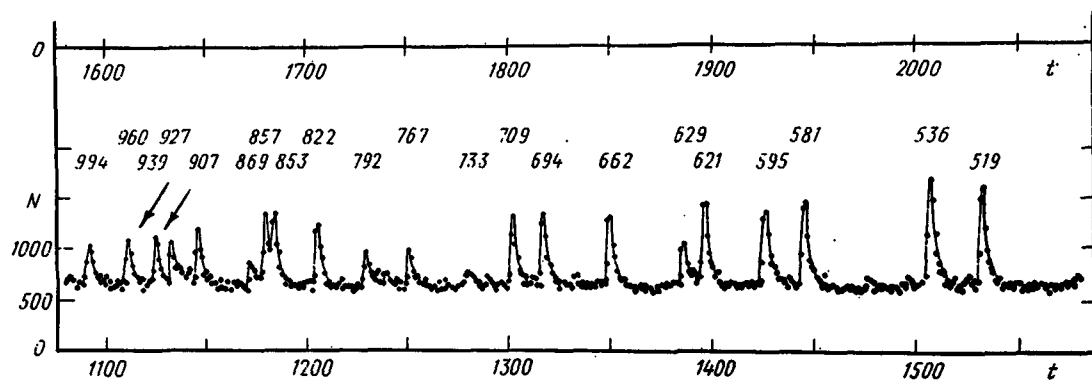


FIGURE 2(b)

A portion of the experimental data from the IBR pulsed reactor at Dubna. Radiative capture was measured in a 1 mm thick (4.8×10^{-5} atoms/barn) sample. The arrows indicate the same pair of levels as in part (a) of this figure.

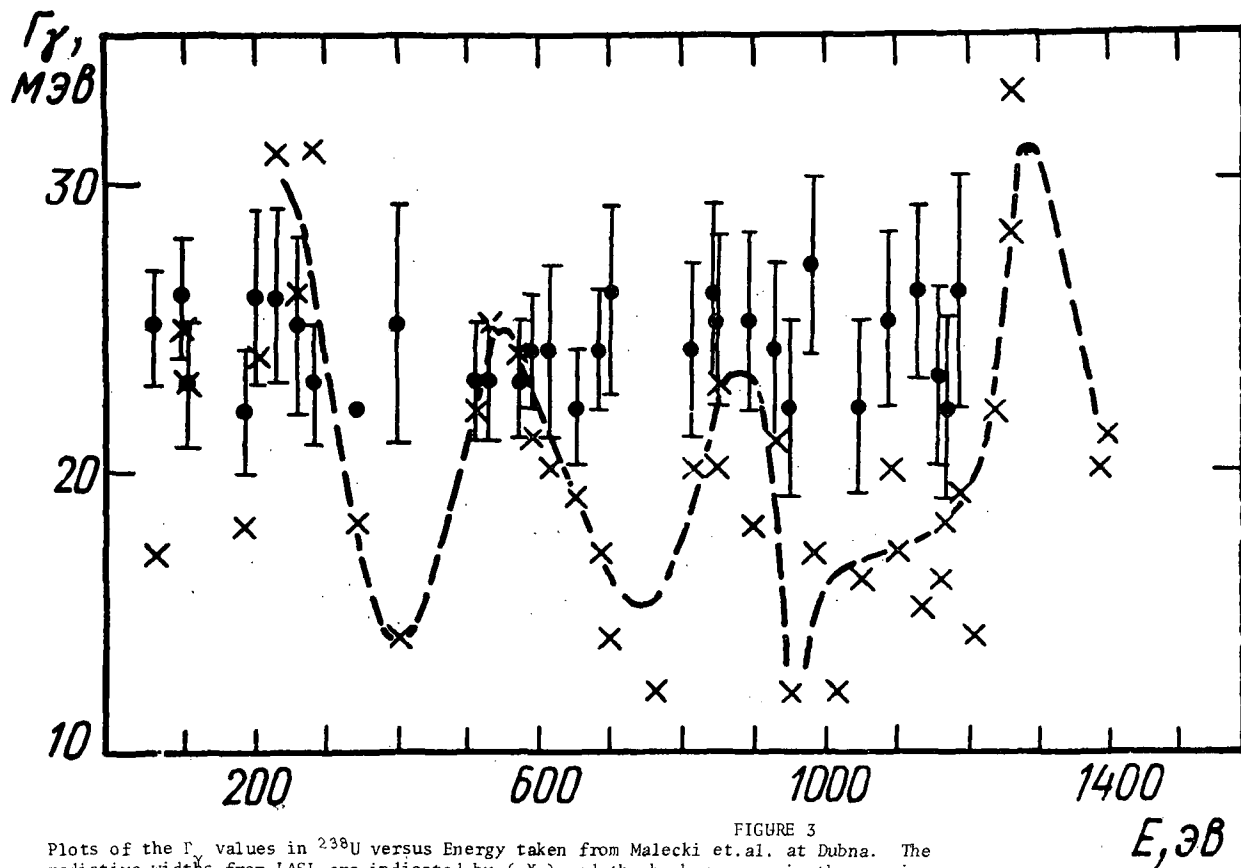


FIGURE 3

Plots of the Γ values in ^{238}U versus Energy taken from Malecki et.al. at Dubna. The radiative widths from LASL are indicated by (X) and the broken curve is the quasi-periodic structure suggested by Glass et. al. The other data are the Dubna values, all of which are quite close to the mean. The differences in the variances of the two data sets are quite striking.

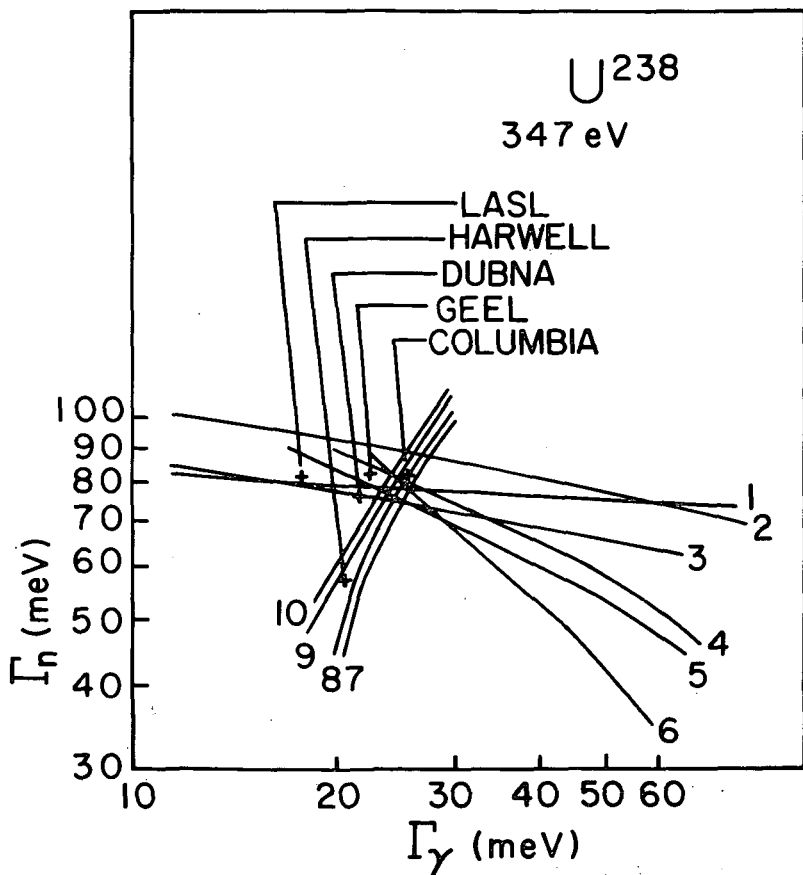


FIGURE 4

The analysis of the resonance parameters for the 347 eV level in ^{238}U obtained at Columbia. The curves numbered 1 through 6 are from 200 and 40 meter transmission samples of inverse thicknesses of $(l/n) = 8.5$ to 478 barns/atom; curves 7 through 10 are from capture samples of 119 and 478 barns/atom from two different detectors. The dispersion in the curves is an indication of the uncertainties in the resonance parameters. The (+) indicate the values of Γ_γ and Γ_n obtained by the various laboratories. The resonance at 347 eV is strong and well isolated.

