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Phys. Rev. Lett. **108**, 142502 — Published 3 April 2012

DOI: 10.1103/PhysRevLett.108.142502

Abrupt Change in Radiation-Width Distribution for ^{147}Sm Neutron Resonances

P. E. Koehler,[1,*] R. Reifarth,[2] J. L. Ullmann,[3] T. A. Bredeweg,[3] J. M. O'Donnell,[3] R. S. Rundberg,[3] D. J. Vieira,[3] and J. M. Wouters[3]

[1] *Physics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA*
[2] *Goethe Universität, Frankfurt Am Main, Germany*
[3] *Los Alamos National Laboratory, Los Alamos, NM 87454, USA*

(Dated: January 18, 2012)

We obtained total radiation widths of s-wave resonances through $\mathcal{R}$-matrix analysis of $^{147}\text{Sm}(n,\gamma)$ cross-sections. Distributions of these widths differ markedly for resonances below and above $E_n =$ 300 eV, in stark contrast to long-established theory. We show that this change, as well as a similar change in the neutron-width distribution reported previously, are reflected in abrupt increases in both the average $^{147}\text{Sm}(n,\gamma)$ cross section and fluctuations about the average near 300 eV. Such effects could have important consequences for applications such as nuclear astrophysics and nuclear criticality safety.

PACS numbers: 24.30.Gd, 24.60.Dr, 24.60.Lz, 25.40.Lw

In this letter, we show that total-radiation-widths (Γ_γ) extracted from $\mathcal{R}$-matrix analysis of $^{147}\text{Sm}(n,\gamma)$ cross sections reveal an abrupt change in the shape and average value of the Γ_γ distribution near $E_n = 300$ eV. These observations are in stark contrast with theoretical expectations that both quantities should remain essentially constant across the resonance range and beyond.

The effect reported herein occurs very near the same energy as previously reported abrupt changes in the α-particle strength-function ratio [1] for the two s-wave spin states and the shape of the reduced-neutron-width (Γ_n^0) distribution [2]. Due to the difficulty of measuring the very small α widths, the former effect was of limited statistical significance. However, the effect in the Γ_n^0 data was established at about the 99% confidence level using several different tests. These two previous effects remain unexplained.

As we will show below, changes in the Γ_γ distribution shape and average are established with very high confidence. That three such deviations from theoretical expectations could occur by chance in the same nuclide at the same energy must be vanishingly small. Therefore, it is virtually certain that significant departure from standard theory has been observed and that all three effects may have a common origin. Given the relative paucity of high-quality Γ_γ data, similar effects may exist for other nuclides and, if so, could have far-reaching consequences for both basic and applied nuclear physics. For example, as we show below, these changes in the Γ_γ and Γ_n^0 distributions are reflected in abrupt increases in both the average $^{147}\text{Sm}(n,\gamma)$ cross section and fluctuations about the average that cannot be explained by the nuclear statistical model. As this theory is used to calculate cross sections for applications, similar differences in other nuclides could have important impacts in nuclear astrophysics and nuclear criticality safety.

It is expected that Γ_γ distributions for medium to heavy nuclides should be very narrow, and essentially constant across the resonance energy region. Expectation [3] that the Γ_γ distribution should be very narrow arises from i) the very complex wave functions of states at high excitation characteristic of neutron thresholds and ii) the very large number of channels for γ decay. Condition i) results in partial γ-decay widths $\Gamma_{i\gamma}$ for each channel i following a Porter-Thomas distribution (PTD) [3] (a χ^2 distribution with one degree of freedom, $\nu_{i\gamma} = 1$). Condition ii) results in the expectation that total γ-decay widths $\Gamma_\gamma = \sum_{i=1}^{n} \Gamma_{i\gamma}$ will follow a χ^2 distribution with degrees of freedom given by the number of independently-contributing channels $n \equiv \nu_\gamma = \sum_{i=1}^{n} \nu_{i\gamma}$. As $n \sim 100$, Γ_γ distributions can indeed be very narrow.

That the Γ_γ distribution should remain fairly constant arises from consideration of the physics of radiative transitions in nuclei as implemented in the nuclear statistical model, which should apply for nuclides in this mass range at these excitation energies. Partial radiation widths $\Gamma_{i\gamma}$ for transitions from resonances i to individual final levels are characterized by average values

$$\langle \Gamma_{i\gamma} \rangle = \frac{f_{XL}(E_\gamma) E_\gamma^{(2L+1)}}{\rho(E_i, J_i, \pi_i)}, \qquad (1)$$

where E_γ is the γ-ray energy, $\rho(E_i, J_i, \pi_i)$ is the density of resonances with spin J_i and parity π_i at energy E_i, and $f_{XL}(E_\gamma)$ is the photon strength function (PSF) for transitions of type X (electric or magnetic) and multipolarity L. As the resonance-energy range is rather small and all quantities are smooth functions of energy, no abrupt changes are expected.

Resonance Γ_γ values typically have been determined only for a relatively small subset of observed resonances, and quite often have fairly large uncertainties. On the other hand, Γ_γ data may, in principle, be a more sensitive tool for testing theory than Γ_n^0 data, for at least two reasons. First, because Γ_γ distributions are much narrower than Γ_n^0 (which are predicted to follow the PTD,

$\nu_n = 1$), it is much easier to detect a change in distribution shape in the former case with a limited amount of data. Second, systematic effects due to missed resonances should be negligible, or at least much smaller, for Γ_γ compared to Γ_n^0 data. It is a well-known fact that all experiments miss some resonances having small neutron widths, and it is well established that neglecting this effect can cause significant systematic errors in discerning the shape of the Γ_n^0 distribution from the data. Because the range of Γ_γ values is much smaller, and because they are, in general, uncorrelated with Γ_n^0 (we have verified this for the data used herein), missed resonances should have random Γ_γ values, and hence no correction for missed resonances is needed while determining the shape of the Γ_γ distribution from the data.

All resonances should have the same parity to perform a valid test of the theory. ^{147}Sm is ideal in this regard because it is at the peak of the s- and minimum of the p-wave neutron strength functions, so all observed resonances at the low energies used in our analysis should be s wave. The probability of a p-wave resonance being included in our analyses can be estimated from the average resonance parameters in Ref. [4] (p-wave neutron strength function $10^4 S_1 = 0.9$ and s-wave average resonance spacing $D_0 = 5.7$ eV), the usual assumption that the p-wave average resonance spacing $D_1 = \frac{1}{3} D_0$, and the threshold used in our analysis (see below). From these values, it is easy to show that the probability of a p-wave resonance being included in our analyses is extremely low, being less than 2×10^{-7}.

Extracting Γ_γ values from measured cross sections also requires knowing the resonance spins. For ^{147}Sm ($I^\pi = 7/2^-$), two s-wave spins are possible, $J^\pi = 3^-$ and 4^-. We overcame this potential problem by making the measurements with the Detector for Advanced Neutron Capture Experiments (DANCE) [5], with which we were able to determine firm spin assignments for all resonances used herein.

Details of the experiment have been reported elsewhere [2]. DANCE is a 4π γ-ray detector comprised of 160 BaF_2 scintillators, each coupled to its own photomultiplier tube, the outputs of which were inputted to two transient digitizers each. In this way, waveforms for each detector were recorded for each neutron beam pulse and analyzed in real time to detect peaks, whose shape and time stamp were written to disk. A 10.4-mg metallic sample, enriched to 97.93% in ^{147}Sm and mounted on a thin Al backing, was placed in the center of DANCE. A well collimated neutron beam from a water moderator at the Manuel Lujan, Jr. Neutron Scattering Center at the Los Alamos Neutron Science Center was incident on this target.

Neutron energies were determined using the time-of-flight technique during replay of the data. Cuts were applied to reduce backgrounds and restrict events to those in the range expected from ^{147}Sm(n,γ) reactions. Separate measurements were made with a blank Al backing foil. Neutron flux was redundantly measured using three different sample/detector combinations. Flux-normalized sample-out counts were subtracted from the sample-in data. Resulting neutron-capture cross sections $\sigma_\gamma(E_n)$ in the unresolved region are in agreement with the most recent high-accuracy data [6] to within the uncertainties.

As explained in Ref. [2], γ-ray multiplicity (the number of γ-rays emitted following neutron capture) information measured with DANCE makes this detector an excellent resonance "spin meter". The technique invented in Ref. [2] was further developed in Ref. [7]. We used the least-squares version in the latter reference to determine spin-separated yields as functions of neutron energy $q^J(E_n)$ for the two s-wave spins. Spins of all resonances analyzed herein could be determined from these yields by inspection. We also used these yields to calculate spin-separated neutron-capture cross sections $\sigma_\gamma^J(E_n)$, e.g., $\sigma_\gamma^3(E_n) = \sigma_\gamma(E_n) q^3(E_n)/(q^3(E_n) + q^4(E_n))$. Example cross sections are shown in Fig. 1. The spin-separated cross sections were crucial for obtaining Γ_γ values for resonances which were not fully resolved in the DANCE data.

The $\mathcal{R}$-matrix code SAMMY [8] was used to fit the

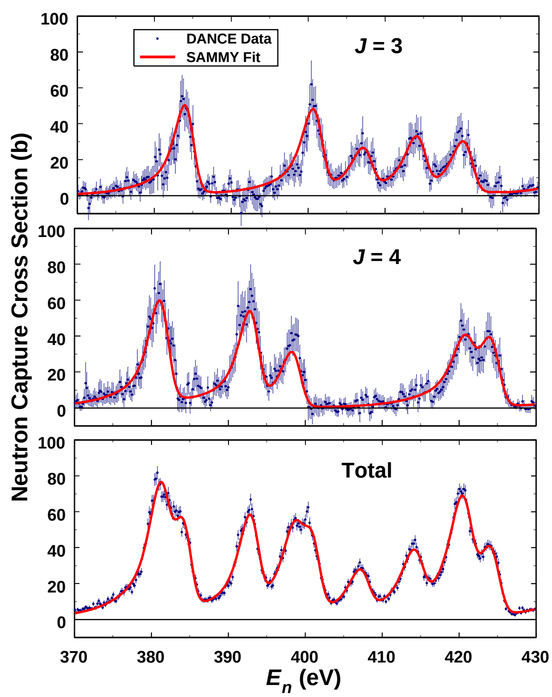

FIG. 1: (Color online) A small part of our ^{147}Sm(n,γ) cross sections versus neutron energy. Top, middle, and bottom panels show $J = 3$, 4, and total capture cross sections, respectively. See text for details.

^{147}Sm(n,γ) cross-section data and obtain resonance parameters. Because natural widths $(\Gamma_n + \Gamma_\gamma)$ of the resonances were almost always smaller than the sum of the experiment resolution plus Doppler broadening, fitting the capture data alone yields capture kernels,

$$K_\gamma = g_J \Gamma_n \Gamma_\gamma / (\Gamma_n + \Gamma_\gamma), \tag{2}$$

(where $g_J = (2J+1)/[(2I+1)(2j+1)]$ is the statistical factor for resonance, target, and neutron spins J, I, and j, respectively) and not the individual widths. However, neutron widths are available from resonance analysis of total cross section data. Therefore, Γ_γ values were determined by fitting our data using the $g_J\Gamma_n$ values from Ref. [9].

There are two components to the total uncertainty $\Delta\Gamma_\gamma$ for each Γ_γ value. First, there is the contribution due to fitting the data, which was calculated by SAMMY. Second, there is a contribution due to the uncertainty in the neutron width, which was derived in the standard manner using Eq. 2 and the $\Delta\Gamma_n$ values from Ref. [9]. These two components were added in quadrature to obtain total uncertainties. As can be seen from Eq. 2, if Γ_n is significantly larger than Γ_γ, then the capture kernel is relatively insensitive to Γ_n and the resulting $\Delta\Gamma_\gamma$ is essentially that calculated by SAMMY. However, as Γ_n decreases, $\Delta\Gamma_\gamma$ increases. In the limit $\Gamma_n \ll \Gamma_\gamma$, the capture kernel is essentially equal to $g_J\Gamma_n$, and hence Γ_γ cannot be determined. Therefore, some limit has to be imposed on the subsequent analysis. Experience has shown $\Gamma_n \geq \Gamma_\gamma/2$ to be reasonable, and hence we limited the subsequent analyses to such resonances. We have repeated the analyses described below using other reasonable limits (e.g., $\Delta\Gamma_\gamma/\Gamma_\gamma < 10\%$, 5%) and obtained essentially the same results.

The Γ_γ values for the 62 (out of 112 observed) resonances below 700 eV meeting the above criteria are shown as a function of resonance energy in Fig. 2. As can be seen in this figure, there is no discernible difference in Γ_γ values for the two s-wave spins, and the Γ_γ distribution becomes noticably broader for $E_n > 300$ eV. Therefore, we combined data for the two spins in subsequent analyses, and divided the data into two groups; $E_n < 300$ eV and $300 < E_n < 700$ eV.

Cumulative Γ_γ distributions for the two energy regions are shown in Fig. 3. We performed several tests [10] to discern the statistical significance of the change in distribution shape which is evident in this figure.

The median (variance) test indicates the null hypothesis that medians (variances) of the two distributions are the same can be rejected at the 99.8% (99.9%) confidence level. Similarly, the Smirnov and Cramer-von Mises two-sample tests reveal the null hypothesis that data in the two energy regions were sampled from the same population can be rejected with > 99% and > 99.9% confidence, respectively. In essence, all these statistical tests indicate that the change in the Γ_γ distribution evident in Fig. 3 is highly statistically significant.

Theoretical interpretation of this change may be aided by estimation of distribution parameters for the two regions. To this end, we used the maximum likelihood (ML) method. As noted above, Γ_γ data are expected to follow a χ^2 distribution with many degrees of freedom, $\nu_\gamma \sim 100$. For such large values of ν_γ, a χ^2 distribution is very close to Gaussian in shape. One advantage of using a Gaussian rather than χ^2 distribution for the analysis is that uncertainties $\Delta\Gamma_\gamma$ can easily be included [11].

Therefore, we used the technique described in Ref. [11] to estimate most likely values for the means $\langle\Gamma_\gamma\rangle$ and standard deviations σ_N of the Γ_γ distributions in the two energy regions. Resulting ML estimates are $\sigma_N = 4.67 \pm 0.81$, $\langle\Gamma_\gamma\rangle = 52.0 \pm 1.1$, and $\sigma_N = 11.7 \pm 1.5$, $\langle\Gamma_\gamma\rangle = 59.6 \pm 2.0$, for the lower- and upper-energy regions, respectively. Hence, the ML results also indicate that Γ_γ distributions in the two energy regions are significantly different. Translated to χ^2 distributions, these ML results lead to $\nu_\gamma = 241$ and 51 for the Γ_γ distributions in the lower and upper energy regions, respectively.

Because our dividing energy is slightly different than that used in Ref. [2], our new Γ_n^0 values for smaller resonances are slightly different, and the ML technique of Ref. [12] is better than that used in Ref. [2], we reanalyzed the Γ_n^0 data using the technique of Ref. [12] to obtain ν_n values for the two energy regions. As explained in the Ref. [12], the ML analysis technique employs an energy dependent threshold to properly account for the effect of missed small resonances. The results given in Table I were obtained with a threshold $g_J\Gamma_n^0 \geq 3.3 \times 10^{-4} E_n$, where $g_J\Gamma_n^0$ is given in meV for E_n in eV. As explained

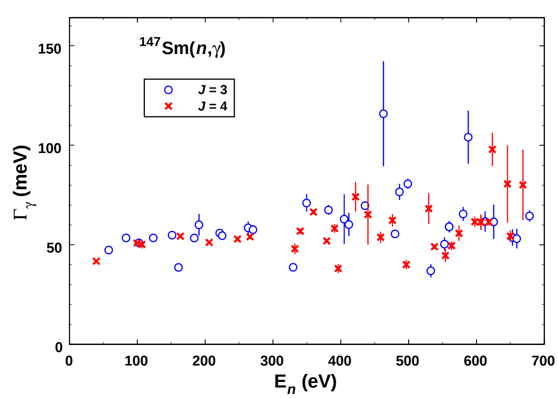

FIG. 2: (Color online) Resonance Γ_γ values versus energy from the SAMMY fits to our data for the 62 resonances meeting the criteria discussed in the text. Values for 3^- and 4^- resonances are shown as blue cirlces and red X's, respectively.

in the Ref. [12], this same threshold excludes p-wave resonances from the analysis with equal effectiveness at all energies. We then applied the technique of Ref. [13] (using the same threshold, and modified to work for any ν_n) to obtain average s-wave resonance spacings D_0 and neutron strength functions S_0, corrected for missed small resonances. These values, along with parameters for the Γ_γ distributions, are given in Table I. Our resulting ν_n values are consistent with those of Ref. [2] and so confirm that the Γ_n^0 distribution also changes shape near 300 eV.

That these changes in the Γ_γ and and Γ_n^0 distributions are mirrored in the $^{147}\mathrm{Sm}(n,\gamma)$ cross section is shown in Fig. 4, in which our DANCE data averaged over 80-eV-wide bins are shown. The bin width must be wide enough to contain several resonances so that the large fluctuations in resonance sizes are damped, but not so large that the change near 300 eV is averaged out. As the average resonance spacing is about 6 eV, the chosen bins should contain about 13 resonances on average, which should be a good compromise. For comparison, the typical rule of thumb for statistical model calculations is that the energy interval contain at least 10 resonances.

Also shown in Fig. 4 are two statistical model calculations based on the average resonance parameters given in Table I for the two energy regions. As all statistical model codes of which we are aware assume the PTD for Γ_n^0, and essentially a single, constant $\langle\Gamma_\gamma\rangle$, we wrote our own simple code which randomly samples over χ^2 distributions with parameters given in Table I. As can be seen in Fig. 4, there is a substantial, fairly abrupt, change in the measured cross section near 300 eV, and calculations based on the parameters for each region are in good agreement with the data in that region, but inconsistent with data in the other region. It also is evident that fluctuations in the data about the theoretical value are larger in the upper-energy region, which is consistent with the interpretation [1, 2] of a non-statistical effect in this region. The cross section in the upper-energy region is approximately 30% larger than calculated using parameters for the lower-energy region. Our calculations indicate that about one third of this increase is due to the changes in the Γ_γ distribution, and the remaining two thirds is mainly due to width-fluctuation effects in the neutron channel.

To our knowledge, there is no model which can explain the two previously reported or the current effects in $^{147}\mathrm{Sm}+n$ widths near 300 eV. The above Γ_γ results could be interpreted as a decrease in the number of effectively independent channels by 190, or a decrease in the degrees of freedom for each channel by approximately a factor of 1/5, between the two energy regions. Given the extremely limited Γ_α data available, the paucity of high-quality Γ_γ data, and the near universal practice of assuming Γ_n^0 follow the PTD, similar effects could exist in other nuclides. In addition to interest in understanding the underlying theory, such effects may be important to, for example, nuclear astrophysics and nuclear criticality safety, in which models often are used to calculate important quantities beyond the reach of measurement. Because our results do not agree with predictions and assumptions of these models, it is prudent to assume that quantities predicted by these models may be more uncertain than previously thought. Similar quality data on other nuclides likely will be needed before the origin and extent of the effects presented herein can be understood.

The authors would like to thank R. R. Winters for useful discussions. This work was supported by the Office of Nuclear Physics of the U.S. Department of Energy under

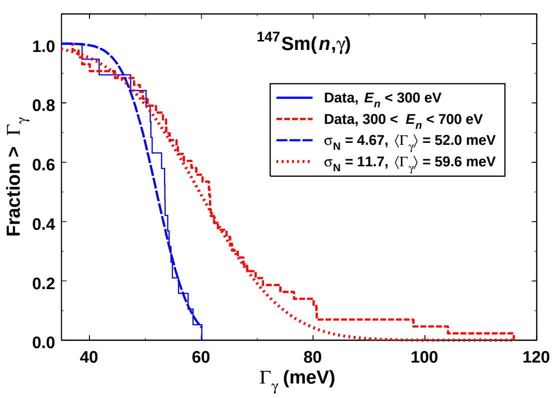

FIG. 3: (Color online) Cumulative Γ_γ distributions. Shown are the fraction of resonances with Γ_γ larger than a given value versus the value. Staircase plots depict the measured data whereas smooth curves show Gaussian distributions from the ML analyses.

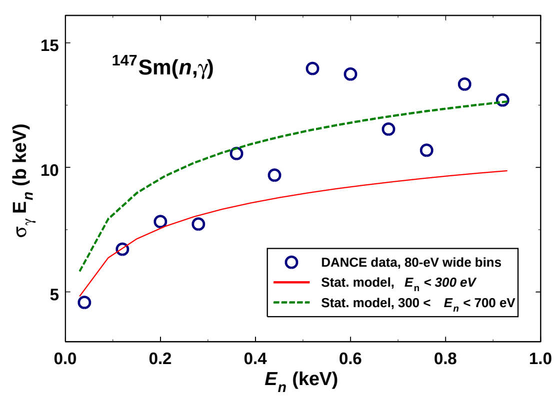

FIG. 4: (Color online) Open blue circles depict our DANCE $^{147}\mathrm{Sm}(n,\gamma)$ cross sections averaged over 80-eV-wide bins. Error bars corresponding to one-standard-deviation statistical uncertainties are smaller than the symbols. The solid red and dashed green curves show results of statisical model calculations based on the average resonance parameters for the lower- and upper-energy regions, respectively. See text for details.

TABLE I: Average parameters for ^{147}Sm+n resonances.

Region (eV)	$\langle\Gamma_\gamma\rangle$ (meV)	ν_γ	$10^4 S_0$	D_0 (eV)	ν_n
0 - 300	52.0 ± 1.1	241 ± 42	4.56 ± 0.94	5.78 ± 0.44	$1.04^{+0.33}_{-0.31}$
300 - 700	59.6 ± 2.0	51.0 ± 6.5	4.53 ± 0.81	6.18 ± 0.40	$2.67^{+0.60}_{-0.57}$

Contract No. DE-AC05-00OR22725 with UT-Battelle, LLC. This work has benefited from the use of the LANSCE facility at Los Alamos National Laboratory which was funded by the U.S. Department of Energy and operated by the University of California under Contract W-7405-ENG-36.

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