Shell Effects in Highly Excited Nuclei

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(Received August 17, 1953; revised manuscript received Februrary 1, 1954)

The boron absorption method has been used to obtain the following estimates of the energy of the lowest neutron resonance for each of the following target nuclides: Rb^{85}, 0.97 kev; Rb^{87}, 0.42 kev; Y^{89}, 5.0 kev; La^{139}, 0.08 kev; Pr^{141}, 0.38 kev; and Tl^{205}, 10 kev. The corrections were made in such a way that the figure found is a lower limit for the energy and consequently a lower limit for the order of magnitude of the spacing of *s*-wave resonances. These figures were added to data in the literature, and a plot of *s*-level spacing was made which included most target nuclides of odd mass and odd charge. This curve showed roughly symmetric maxima at the neutron magic numbers 50, 82, and 126. The cases where the compound nucleus contains more neutrons than the magic numbers are qualitatively understandable, since these nuclides are in a low state of excitation compared to normal nuclei in their neighborhood. An unusually close level spacing, which would be expected on the basis of excitation energy when there are slightly less than a magic number of neutrons, was not observed. It appears (at least near 126 neutrons and 82 protons) that the shell structure is not broken up to any great extent in the compound nucleus.

I. INTRODUCTION

HUGHES and his collaborators[1] have measured the capture cross sections of many nuclides for unmoderated fission neutrons. They have shown that capture cross sections are unusually small at and near the neutron magic numbers. They attribute these anomalously low cross sections to unusually wide level spacings. An attempt is made here to determine the anomalous spacings more directly. It seemed very likely that the resonances would lie in a region inaccessible to time-of-flight methods, and it was decided to use the methods of boron and self-absorption so that a lower limit to the spacing might be obtained by estimating the position of the lowest-energy resonance. In order to approach a quantitative comparison with normal nuclei, the spacing of *s*-neutron peaks must be determined for the nuclei of odd charge.

II. EXPERIMENTAL RESULTS

A beam of pile neutrons from the graphite pile at Oak Ridge National Laboratory was used as a source. The technique does not differ appreciably from that used by Dancoff *et al.*[2] The curves (Fig. 1) have been corrected for $1/v$ absorption and boron scattering in the same way. The measurements were extended to thicknesses at which B^{10} scattering exceeds absorption, the limit of usefulness of the method. The above workers were very successful in detecting the lowest resonance of Mn, Na, and V where their results have been checked by more direct measurements.[3] However, the method is very sensitive and sometimes detects "resonances" not otherwise verifiable. The present work probably suffers from the same difficulty which will be discussed later.

Corrected boron absorption curves are shown in Fig. 1. The Tl^{205} and Y^{89} curves indicate single resonances by their shapes. The small break at the beginning of the Y curve is considered to be due to an underestimate of the large $1/v$ component which is present in this case. The remaining curves indicate by their curvature the presence of more than one activation resonance. If these are *s*-wave resonances, the expression for the capture resonance integral shows that the ratios of the resonance activities should vary inversely as the square of the resonance energies if scattering predominates, and inversely as the three halves power if capture is predominant. [See Eq. (1) following.] With this in mind, first approximations of the steeper components of each complex curve were obtained by assuming that the sum of effects of the higher-energy resonances could be approximated by a constant term. The Rb and Pr curves, when analyzed this way, were resolved into one pure exponential and a constant term. The first resonance in Cs has been reported as 5.9 ev, and no more resonances were observed as high as 20 ev.[4] A good fit of the Cs curve was obtained using the 5.9-ev value, another component near 300 ev and a constant term. The 300-ev figure is not very reliable and should be checked by time-of-flight measurements. La, like Cs, was analyzed into three components. For reference, previous boron absorption measurements on Nb are included in

* This work was carried out under the Research Participation Program while on leave of absence from Emory University. The research was performed in partial fulfillment of the requirements for the Ph.D. degree at Duke University.

[1] Hughes, Garth, and Eggler, Phys. Rev. **83**, 234 (1951); Garth, Hughes, and Levin, Phys. Rev. **87**, 222 (1952); and Hughes, Garth, and Levin, Phys. Rev. **91**, 1423 (1953).

[2] Lichtenberger, Nobles, Monk, Kubitschek, and Dancoff, Phys. Rev. **72**, 164 (1947).

[3] *Neutron Cross Sections*, Atomic Energy Commission Report AECU-2040 (U. S. Government Printing Office, Washington, D. C., 1952), plus supplements 1 and 2.

[4] H. H. Landon and V. L. Sailor, Phys. Rev. **86**, 605 (1952).

Table I. The energies were determined by a two-component analysis as above.

III. DISCUSSION OF RESULTS

The energy of the lowest resonance given in Table I is valid if the resonance is an s level and the constant term subtracted is small. If the first resonance should be due to a p wave, the analysis would be invalid, but the heavy reliance placed on the initial slope would make the figure in Table I a gross underestimate of the energy of the first s resonances. These estimates for the lowest-energy s resonances are, therefore, conservative when used as lower limits to the average spacing of s resonances.

Self-absorption curves have been run on most of these materials but most of them are more difficult to interpret than the boron curves. However, self-absorption data show that in the case of Tl^{205}, if it is assumed that capture is negligible as compared to scattering, the first resonance is in the neighborhood of 10 kev or higher by a factor of 3. It is clear that Tl^{205} is not responsible for the 270-ev and the 1300-ev peaks[3] observed in natural Tl, but the Tl^{205} resonance may contribute to a 6500-ev peak[5] also seen in Tl. Attributing the 270-ev and 1300-ev peaks to Tl^{203}, one arrives at an average spacing of ~3000 ev which is in good agreement with the value ~1000 ev which may be calculated from the reduced width[6,7] of the 270-ev level.

The boron absorption method has the obvious disadvantage that, after all the corrections have been made, one is left with a very rough value of the energy of the first resonance. However, when the spacing of the resonances under study is of the order of kilovolts even qualitative information is of value since the more direct methods of time-of-flight and threshold sources have not yet been applied extensively to this region.

The method does have the advantage that separated isotopes are not necessary to study the resonances of single nuclides since radioactive properties serve to distinguish the components of an isotopic mixture. Our most interesting results were obtained on thallium and rubidium which do occur in isotopic mixtures.

A second disadvantage did us relatively little harm: the possibility that the lowest-energy resonance may be due to the p rather than s wave. In a heterogeneous neutron source varying as $1/E$, the capture resonance integral (over a single resonance)[7] is

$$R=\int\frac{\sigma(E)dE}{E}=\frac{1.3\pi g\Gamma_n\Gamma_a 10^6}{E_r^2(\Gamma_n+\Gamma_a)}, \qquad (1)$$

where E_r is the neutron energy at resonance in ev and Γ_n and Γ_a are the neutron and absorption widths,

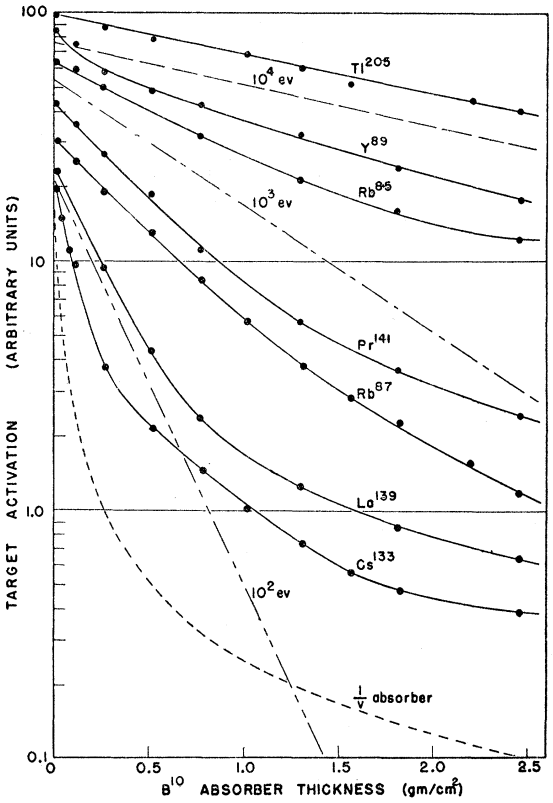

FIG. 1. Boron absorption curves corrected for scattering in the boron absorbers and for the $1/v$ activation component. The ordinates of the separate curves are not related. The broken line curves indicate the slopes associated with activation due to single resonances of various energies and pure $1/v$ activation.

respectively. If $\Gamma_n \gg \Gamma_a$, R will be independent of Γ_n, and apart from the weight factor g independent of the angular momentum of the neutron wave. Even if $\Gamma_n \approx \Gamma_a$, R will depend principally on E_r^{-2}, and a low-energy p resonance may well be activated much more than a higher-energy s resonance. For instance, if a resonance is at 2.5 kev and its spacing is 10 kev, Γ_n should be roughly[8] 50 ev for an s wave and 0.5 ev for a p wave; if $\Gamma_a \leqq 0.5$ ev, R will not depend much on the angular momentum of the resonance. (This effect will not arise in measurements of the total cross section of the resonances under study. These depend on the scattering resonance integral which is always proportional to Γ_n in the energy region considered here.) Our difficulties in interpreting the self-absorption measurements probably arose from these effects. The rather low energy found for the first resonance of La may also be due to this effect, since other evidence[3] makes it seem unlikely that there is an s resonance in the neighborhood of 76 ev.

[5] W. W. Havens (private communication).
[6] Wu, Rainwater, and Havens, Phys. Rev. **71**, 174 (1947).
[7] Harris, Muehlhouse, and Thomas, Phys. Rev. **79**, 11 (1950).
[8] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1953), p. 463.

TABLE I. Neutron resonance energies from boron absorption data.

Target nuclide	N	Activated period	Cadmium ratio	$1/v$ fraction	High-energy fraction	Resonance energy (ev)	Chemical form
Rb^{85}	48	19.5 day	2.34	0.043	0.300	970	$RbNO_3$
Rb^{87}	50	17.5 min	2.02	0.033	0.074	420	$RbNO_3$
Y^{89}	50	62 hr	19.0	0.605	0	5000	Y_2O_3
Nb^{93}	52	6.6 min	···	0.153	0.210[a]	730[a]	Nb
Cs^{133}	78	3.2 hr	3.10	0.067	0.050	(5.9) 300	$CsNO_3$
La^{139}	82	40.4 hr	15.4	0.457	0.054	76	La_2O_3
Pr^{141}	82	19 hr	12.0	0.357	0.161	380	Pr_4O_7
Tl^{205}	124	4.2 min	2.98	0.063	0	10 000	Tl

[a] Calculated from Dancoff's data, reference 2.

IV. S-LEVEL SPACINGS

"Magic" spacings will now be compared with those of "normal" nuclei. This study has been restricted to the elements with odd charge and mass number in order to eliminate any possible structural effect due to other odd-even combinations of nucleons. (The data are insufficient for a study of other classes of nuclei, but in regions where considerable information is available the even-even nuclei appear to follow much the same pattern as those studied here.) In order that the close and wide spacing comparisons be valid, the spacings used here are those between resonances due to neutrons of zero angular momentum. This is necessary since close spacings are resolvable at very low energies where only s resonances are observable.

Figure 2 shows a plot of spacings against neutron number. Aside from a few omissions noted below, all stable odd-Z nuclei and the radioactive nuclei Rb^{87} and Re^{187} are included. The nuclide corresponding to each point may be identified from the Z scale and the numeral in the circle which is the last digit of the atomic number. Where both isotopes of the same element are plotted, the displacement in the N scale is sufficient to distinguish the lighter from the heavier. The open circles and double circles were determined by inspection of curves of cross section against energy.[3,9] The energy range over which the resolving power appeared to be reasonably adequate was estimated by inspection of the curves. It was assumed, also, that below 125 kev, all peaks shown in the published curves were s resonances. The peaks used in computing these spacings, then, were those appearing in the region below 125 kev for all the lighter elements. Other upper limits of neutron energy adopted were: for copper, 25 kev; gallium, 600 ev; bromine, 200 ev; arsenic, 150 ev; silver, 150 ev; indium, 30 ev; antimony, 27 ev; iodine, 200 ev; tantalum, 45 ev; rhenium, 30 ev; iridium, 10 ev; gold, 250 ev; thallium[203], 3 kev; and bismuth, 80 kev. One subzero resonance was counted in europium for which the energy range was taken from -1 to 13 ev. Where two or more peaks were observed on a cross-section curve, the average spacing was determined and multiplied by two to correct for the fact that two independent reactions take place depending on the relative orientation of the neutron and nuclear spins; the result was then multiplied by the number of isotopes. The reader may check these points from the curves shown in the neutron cross-section compilation, AECU 2040.[3]

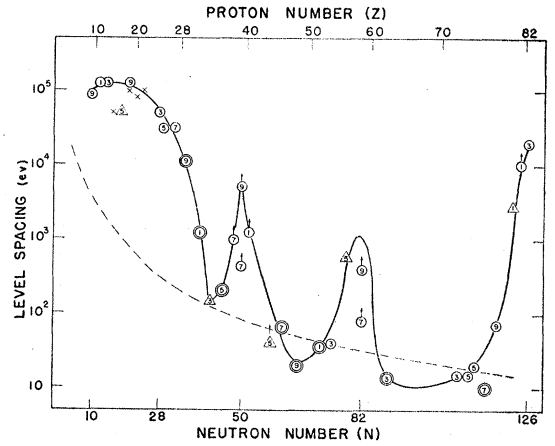

FIG. 2. The average spacing of s-wave neutron resonances of the odd elements. Circles with arrows indicate that a lower limit is plotted. (See Table I and the discussion of the interpretation of the boron absorption data.) Other circles indicate spacing estimated from resonance peaks given in reference 3; double circles signify that the spacing is the average for two isotopes of the element in question. Points plotted as crosses and triangles are explained in Table II. Numerals inside the symbols give the second digit of the atomic number, of the element. The dashed curve represents spacing calculated from the liquid drop model (reference 11).

The data from Table I appear as open circles with arrows pointing upward to indicate that the boron absorption method leads to an approximate lower limit to the level spacing. Where more direct observations are unavailable, the "nominal spacings" $D^* \simeq \Gamma_n E^{-\frac{1}{2}} \times 10^4$ ev were used.[8] D^* may be estimated with considerable uncertainty from cadmium ratios.[2,7,10] The best available estimates for a few of the nuclides were so determined, and are shown as crosses in Fig. 1. (Many of the anomalous spacings found by boron absorption were predicted from previous measurements of cadmium ratios.[10]) Table II specifies the nuclides for which the spacings have been so estimated,

[9] R. K. Adair, Revs. Modern Phys. **22**, 249 (1950).

[10] H. W. Newson (unpublished). Some of the data were taken in collaboration with R. H. Rohrer and E. Rogers.

and also includes four nuclides for which the estimated spacings were not obtained directly from Table I or from AECU 2040.

The final curve is qualitatively about what one would expect from the work of Hughes *et al.*[1]; however, the direct method appears more sensitive. The largest peak-to-valley ratio of spacings is 10^3 against a capture cross-section ratio of 100. The values of the average spacings obtained by counting resonance peaks in the existing cross-section curves may be expected to be high, in general. Very few of all the resonances so far observed have been measured with sufficient resolving power to be reasonably sure that two or more levels are not combined to form a single peak in the cross-section curve. It is even more likely that apparently flat portions of a cross-section curve will be found to include resonances after being remeasured with greater resolving power. One might, therefore, expect the true values at the minima to be somewhat lower than shown in Fig. 2. These difficulties are particularly troublesome in the region between 20 and 40 neutrons. In addition, among the still lighter elements (below 20 neutrons), it is often difficult to distinguish resonances of different angular momenta. Any discussion of these lighter nuclei ($N<40$) must be postponed until more and better data are available.

The dotted curve is taken from Bethe's[11] review article; it was calculated from the liquid drop model. There is fair agreement for most of the heavy elements, but serious deviations appear for light elements and at the neutron magic numbers. The peak at 50 neutrons appears to spread at least two neutrons on either side of the magic number. The minimum at about 44 neutrons has been well established by Havens *et al.*[3,12] who found four definite resonances in bromine (2 isotopes) between 0 and 200 ev, and by Harris and Bollinger[3,13] who found resonance peaks in $_{33}As^{75}$ at 45 ev, 92 ev, and other resonances above 100 ev. The peak at 82 neutrons is somewhat less prominent than might be expected from the work of Hughes *et al.* The most interesting feature here is the fact that Cs^{133} (78 neutrons) may have a spacing as wide as 600 ev.

The most interesting peak is that at $N=126$ and $Z=82$. $_{81}Bi^{209}$ has been investigated by Gibbons and Newson[3,14] who find an average spacing of about 10 kev for both possible J values of compound nuclei formed by an s neutron. The same authors have also verified to some extent the spacings assigned to the two isotopes of Tl; the spectrum contains several small peaks at energies below 10 kev. A large peak above 10 kev may be the one found in Tl^{205} by boron absorption. Even as far away from the magic number as $_{79}Au^{197}$ the spacing is definitely anomalous (about 60 ev).[3,15]

The even elements show a similar behavior just below 126 neutrons. $_{82}Pb^{208}$ appears to have only one s resonance between 0 and 800 kev[16]; $_{82}Pb^{206}$ has many s resonances with a spacing of the order of 20 kev,[16] while an examination of the cross-section curves of even elements[3] between $_{80}Hg$ and $_{74}W$ reveals a qualitative increase of spacing for the heavier elements.

TABLE II. Level spacings plotted in Fig. 2 as triangles and crosses.

Target nuclide	N	Symbol in Fig. 2	Estimated spacing	Basis for estimate
$_{45}Rh^{103}$	58	Triangle with arrow	>40 ev	Resonances were not found between 1.4 and 1000 ev; they are very probably not present below 22 ev.[a]
$_{55}Cs^{133}$	78	Triangle	600 ev	It is assumed that the first resonance is at 5.9 ev[a] and the second at 300 ev (Table I).
$_{81}Tl^{203}$	122	Triangle	3000 ev	Resonances[a] at 270 and 1300 ev are assigned to Tl^{203}. There are apparently no additional resonances up to 3 kev in unpublished curves taken at Duke University.
$_{17}Cl^{35}$	18	Triangle	50 kev	Four resonances found at Duke University below 100 kev in natural Cl are assumed to be due to Cl^{35}. (Unpublished)
$_{15}P^{31}$	16	Cross	50 kev	Cadmium ratio[b]
$_{17}Cl^{37}$	20	Cross	100 kev	Cadmium ratio[c,d]
$_{19}K^{41}$	22	Cross	80 kev	Cadmium ratio[d]
$_{21}Sc^{45}$	24	Cross	100 kev	Cadmium ratio[b]
$_{33}As^{75}$	42	Triangle	150 ev	There are resonances at 45 and 92 ev.[e] No others are apparent up to at least 150 ev.

[a] See reference 3.
[b] See reference 7.
[c] See reference 2.
[d] See reference 10.
[e] See reference 13.

[11] H. A. Bethe, Revs. Modern Phys. **9**, 69 (1937).
[12] W. W. Havens (private communication).
[13] S. P. Harris and L. M. Bollinger, Phys. Rev. **87**, 222 (1952); also private communication.
[14] J. H. Gibbons and H. W. Newson, Phys. Rev. **91**, 209 (1953).
[15] J. Tittman and C. Sheer, Phys. Rev. **83**, 746 (1951).
[16] Barschall, Bockelman, Peterson, and Adair, Phys. Rev. **76**, 1146 (1949); Peterson, Adair, and Barschall, Phys. Rev. **79**, 935 (1950).

No points are included for Li^7, B^{11}, and N^{15}. In this region, s resonances are very rare and significant estimates of spacing cannot be made[3]; the spacings must be very large (10^5-10^6 ev). No useful cross-section curves[3] are available for the rare earths, $_{65}$Tb, $_{67}$Ho, $_{69}$Tm, and $_{71}$Lu, but their very high thermal cross sections ($\sim$100 barns)[3] make it likely that the spacings are of the order of 10 ev. This is about the same spacing as $_{63}$Eu and $_{73}$Ta, so that this wide gap is probably at a relatively flat portion of the curve.

The most interesting feature of the curve shown in Fig. 2 is the apparent symmetry of the peaks about the magic numbers. Hughes *et al.*[1] were able to account qualitatively for anomalously wide spacings when the compound nucleus has more than a magic number of neutrons by the fact that these nuclei are excited by about 2 Mev less than normal (i.e., the neutron binding energy is about 2 Mev less). This explanation cannot be used when the compound nucleus has slightly less than a magic number of neutrons. In this case the excitation is greater than normal, and an entirely different explanation is necessary.

After examining the curves of Harvey[17] which show the change of neutron binding energy near the magic numbers, we estimate that it requires all of the excitation energy of the compound nucleus to excite a few neutrons when there is just a magic number of neutrons in the nucleus, and the situation will be about the same if there are a few less neutrons. Presumably, the proton binding energies will behave similarly near the magic numbers.

The wide peak at $Z=82$ and $N=126$ is then easily understood in terms of the shell model. The extra binding energy at a closed or nearly closed shell tends to hold the nucleons in their ground states since the stable orbits, which must be postulated to account for the shells, cannot in general exist for excited states of individual nucleons.[18] Thus, few nucleons in the compound nucleus are in excited states, the life of the compound nucleus will be short, and the neutron width and level spacing, unusually large. It is not immediately clear that the same should be true when $N=50$ or 82; the above argument holds for the neutrons but the proton numbers are not at all magic, and extensive excitation of the protons might be expected. In fact, except for $_{37}Rb^{85}$ and the doubtful point $_{45}Cs^{133}$, the peaks near $N=50$ and 82 may be attributed to low neutron binding energy, and more data must be obtained before the possible symmetry of the peaks around 50 and 82 is established. We note that there is no peak around 50 protons; so that, if the symmetry really exists at all three neutron magic numbers, the protons and neutrons must play a very different role in the compound nucleus.

[17] J. A. Harvey, Phys. Rev. **81**, 353 (1951).
[18] V. F. Weisskopf, Helv. Phys. Acta **23**, 187 (1950).

IV. CONCLUSION

Merzbacher and Newson[19] have proposed a "semiconductor" model to account for the anomalies just below the magic number 126. This model assumes that there is a gap of about 2 Mev between the ground state of the highest-energy neutron in the nucleus and the first excited state for an individual neutron. This is obviously the approximate energy necessary to excite a neutron from a closed shell. While the model as developed, thus far, is by no means adequate to explain the details of Fig. 2, it does predict that the ratio of the spacing of Tl^{205} to Ta^{181}, a normal heavy nucleus,[3] should be about a thousand. This agrees with the data used in Fig. 2. The wide spacing of the resonances of $_{83}Bi^{209}$, due in part to excitation energy effects, is too complicated for the theory in its present state to explain.

We wish to express our indebtedness to the management and staff of Oak Ridge National Laboratory for making the facilities of the Laboratory available to us. We also wish to thank W. W. Havens, A. C. Langsdorf, and D. J. Hughes for unpublished information. The part of the work done at Duke University was supported by the U. S. Atomic Energy Commission.

[19] H. W. Newson and E. Merzbacher, Phys. Rev. **91**, 241 (1953).