Nuclear science for the Manhattan Project & Comparison to Today’s ENDF Data

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Abstract — Nuclear physics advances in the US and Britain, from 1939-1945, are described. The Manhattan Project’s work led to an explosion in our knowledge of nuclear science. A conference in April 1943 at Los Alamos provided a simple formula used to compute critical masses, and laid out the research program needed to determine the key nuclear constants. In short order, four university accelerators were disassembled and reassembled at Los Alamos, and methods were established to make measurements on extremely small samples owing to the initial lack of availability of enriched $^{235}$U and plutonium. I trace the program that measured fission cross sections, fission emitted neutron multiplicities and their energy spectra, and transport cross sections, comparing the measurements with our best understanding today as embodied in the Evaluated Nuclear Data File ENDF/B-VIII.0. The large nuclear data uncertainties at the beginning of the project, which often exceeded 25-50%, were reduced by 1945 often to less than 5-10%. $^{235}$U and $^{239}$Pu fission cross section assessments in the fast MeV range were reduced with more accurate measurements, and the neutron multiplicity $\nu$ increased. By a lucky coincidence of canceling errors, the initial critical mass estimates were close to the final estimated masses. Some images from historical documents from our Los Alamos archives are shown. Many of the original measurements from these early years have not previously been widely available. Through this work, these data have now been archived in the international experimental nuclear reaction data library (EXFOR) in a collaboration with the IAEA and Brookhaven National Laboratory.

I. Introduction

In 1939 the world of physics was shaken by Bohr and Wheeler’s remarkable Physical Review paper, “The Mechanisms of Nuclear Fission”, which provided physics insights and a mathematical understanding of the fission process, which had only recently been discovered. It was inspired by Meitner and Frisch’s “liquid-drop” fission model. That same paper reported on the first fast
fission uranium cross section measurements from Princeton (Ladenburg, Kanner, Barschall and van Voorhis) and from the Carnegie Institution of Washington DC (Tuve). Merle Tuve’s measurement was particularly important since it was for neutron energies below 1 MeV, which Bohr and Wheeler had predicted would be below the $^{235}\text{U}$ fission threshold and therefore could be ascribed to the $^{235}\text{U}$ isotope. The importance of these measurements was such that James Chadwick, in an interview three decades later, still remembered the page number on which the measurements were published (p. 444)!

Also, in 1939 Rudolf Peierls in Birmingham had realized that a fast unmoderated neutron chain reaction was possible in the $^{235}\text{U}$ isotope and derived a formula for its critical mass. He did not compute the actual critical mass values in his 1939 Proceeding of the Cambridge Philosophical Society paper (that came some months later, with Frisch), though he pointed out in a note added in proof how Bohr and Wheeler’s paper supported the arguments that he made.

Otto Frisch had come to Birmingham in the summer of 1939 and began to work with Peierls on whether a “super bomb” could be constructed with the $^{235}\text{U}$ isotope supporting a chain reaction. At that time, both Frisch and Peierls were classed as enemy aliens; they were allowed to work on nuclear physics as it was thought to be less sensitive compared to radar. Using Peierls’ formula, together with estimates for the cross sections and the fission neutron multiplicity, Frisch and Peierls calculated a $^{235}\text{U}$ critical mass that was surprisingly (and erroneously) small, 0.6 kg (the correct value we now know is 46 kg for a sphere of pure $^{235}\text{U}$). The mistake they made was for the $^{235}\text{U}$ fission cross section in the fast energy region, which they took as 10 b (versus the best value today of 1.2 b), influenced it seems by a measurement in Paris by which they took as 10 b (versus the best value today of 0.629 = 0.4 b).

But Chadwick also correctly guessed that Tuve’s value might be too low because it is so much smaller than the geometric cross section, which is about 1.7 b, and he remembered Bohr’s expectation that fission cross sections should not exceed the geometrical cross section (true in the fast region, not true at low energies where quantum mechanics comes in). Chadwick therefore quickly established a program of research in 1940/1941 at Liverpool’s cyclotron laboratory, with Frisch, to measure these cross sections.

Chadwick played a leading role in writing the MAUD report in 1941, a remarkable document for its clarity and prescience (originally secret of course and still hard to find - it is reproduced in Margaret Gowing’s book). It concluded in the likelihood of creating an atomic weapon assuming the successful isotope separation of $^{235}\text{U}$: “The committee said that the scheme for a uranium bomb is practicable and likely to lead to decisive results in the war.” Together with Harvard President James Conant, the US had sent two senior scientists, Urey and Pegram, to England in October 1941 to learn about the uranium work there; Chadwick told them that “if pure $^{235}\text{U}$ could be made available, there was a 99% chance of being able to produce an explosive reaction” (Gowing, p.117-119).

The parallel US National Academy of Sciences (NAS) studies, chaired by Arthur Compton came to the same conclusion, “This seems to be as sure as any untried prediction based upon theory and experiment can be”. Conant later wrote that the US government’s growing seriousness regarding the potential for an atomic bomb was influenced by both physicists in England who “had concluded that the construction of a bomb made out of uranium 235 was entirely feasible” as well as E.O. Lawrence’s 1941 proposal to use the newly-discovered plutonium. The

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aThe author, MBC, is pleased to write on MAUD, given that its chairman G.P. Thomson was MBC’s “academic grandfather” (P.E. Hodgson was Thomson’s PhD student); he knew the instigator, Peierls, at Oxford in the 1980s when Peierls held the Wykham Professor of Physics chair at New College and MBC was a lowly Scholar; and the MAUD report’s author James Chadwick shares a common ancestor with MBC (sometime after LUCA).

bThe $^{238}\text{U}$(n,f) is only 1 mb at 0.6 MeV and 14 mb at 1 MeV, but rises to 538 mb at 2 MeV.
NAS and MAUD reports helped launch what was to become the Manhattan Engineering District (MED) in August 1942 under General Groves, with the opening of the Los Alamos laboratory effort, code-named “Project Y”, in March 1943.

Robert Oppenheimer (Fig. 1) and General Leslie Groves intentionally pursued dual 235\textsubscript{U} and 239\textsubscript{Pu} pathways to develop the bomb, see Fig. 2. The uranium gun bomb approach was what we would call today the “high technology readiness level” (TRL) path, where the physics was becoming well established and had been discussed since 1940, and the principal challenge was to produce adequate quantities of enriched 235\textsubscript{U} at Oak Ridge’s MED Y-12 facility. (So, in today’s parlance, it might be called high-TRL but lower “Material Readiness Level”, MRL). In contrast, the plutonium path was much lower-TRL and MRL. Plutonium had just been discovered in 1940. Early insights from the US and Germany have been discussed by Bernstein, including by Louis Turner (Princeton) in 1940 and by von Weizsäcker in July 1940 on neptunium-239 “Eka-Rhenium” produced from a Bretscher primary source. I was able to track down such a source in Egon Bretscher’s papers held by Churchill College Cambridge's Archives. Bretscher did indeed write with great perception and intelligence. In Report II, December 19, 1940, Bretscher and Feather\textsuperscript{19} wrote (page 1) “The large cross section of element 94 to be expected is of the greatest importance in the following respects: (a) it permits (after isolation of 94) the production of a super-explosive mass. The critical radius would seem to be considerably smaller than in the case with U(235) ...”. See the end of Sec. IV. A for more on this.

Bretscher made valuable contributions at Los Alamos. He was a co-author on the important Nuclear Physics Handbook evaluations, LA-140 (1944) and LA-140A (1945), with Weisskopf, Inglis and Davis (discussed further, below) and he led work to measure the DD and DT cross sections for Teller’s Super thermonuclear studies, following earlier work at Purdue University.

Figure 3 illustrates the measurements shown in these Handbooks that will be discussed below in detail in this paper. It is evident that many of these 1940s measurements over-estimated the true value as embodied in our best understanding today (the solid curve, for ENDF/B-VIII.0); reasons for this are discussed later in this paper. The challenges of assessing “unrecognized sources of uncertainties” (USU) was the subject of a recent useful paper by Capote et al.\textsuperscript{30} It also focused on 235\textsubscript{U} fission cross sections over the years, extending the data in Fig. 3 from 1945 to the 1980s in a figure that shows differences to a 1970 evaluation by Poenitz.\textsuperscript{21}

The main purpose of the nuclear research program at Los Alamos during the Manhattan Project was to establish an understanding of the fundamental nuclear constants needed to determine critical masses, both bare and tamped, for the bomb designs, and to understand the breadth of nuclear physics to ensure there were no surprises or “show-stoppers”. A two-pronged approach was taken, one being to measure the fundamental “differential” cross sections and neutron energy and scattering angle spectra, the other being to measure integral quantities. This paper focuses on the former approach, while other papers in this issue by Hutchinson, Kimpland, Myers et al.\textsuperscript{22-24} and Ref.\textsuperscript{25} focus on the latter approach: integral critical assembly measurements. But it should be remembered that for most of the Manhattan Project, substantial amounts of enriched 235\textsubscript{U} and plutonium were simply not available in Los Alamos – they only started to arrive in late 1944 and 1945. Metal “25”\textsuperscript{c} spheres started to become available in October 1944, but the larger spheres were not available until April 1945 as described by Hutchinson\textsuperscript{22}. Plutonium “49” spheres

\textsuperscript{1}{ Los Alamos shorthand used the last number of the isotope’s Z followed by the last number of its A value, so 235\textsubscript{U} is “25”, and is also sometimes written u5; 239\textsubscript{Pu} is “49” and also p9, etc.}
were only available after February/March 1945. Thus, the integral “fast” critical assembly experiments could only begin in 1945 (some in late 1944). Most of the design work for the uranium gun bomb and for the plutonium implosion bomb was based on the fundamental cross section understandings made between 1943 and late 1944, as described in this paper.

This present paper traces the U.S. and British nuclear science developments in the period leading up to the Manhattan Project, and the subsequent advances made by the U.S., British, and Canadian scientists and engineers at Los Alamos through 1945. Some of the history of early nuclear physics measurements (at universities and at Los Alamos) has been told by the Manhattan Project’s second Physics Division Leader, Robert Wilson,\textsuperscript{26} in 1947 (LA-1009), and more recently in the 1993 excellent and comprehensive book \textit{Critical Assembly}\textsuperscript{27} by Los Alamos’ historians. The many measurements made at Los Alamos during the Manhattan Project are also documented in the “LA-” Los Alamos reports in the National Security Research Center (NSRC) archives; many of these are also available online as unclassified reports from the Los Alamos Library. A most remarkable set of documents is the Theoretical Division’s monthly progress reports, put together by Hans Bethe with his group leaders (Fig. 4). Their brilliance, and their diligent scholarship, \textsuperscript{d} helps readers reconstruct the evolution of Project Y’s work in theory, experiment and simulation. I will describe how the evolving understanding of fission cross sections and fission neutron multiplicities led to drastic swings in estimates of the critical masses for $^{235}$U and $^{239}$Pu, with corresponding mood swings of depression and elation among the Los Alamos scientists as the required material production quantities went up and down. The data that were measured are compared against our modern best understanding of the cross sections in our

\textsuperscript{d}Allan Carlson of NIST, who knew Hugh Richards at U. Wisconsin (who had been at Los Alamos), recollects Richards’ story\textsuperscript{28} of Bethe dictating a paper to his secretary (Hugh’s wife). He walked back and forth dictating in perfect English with no mistakes or changes - as if he were reading a manuscript.
Figure 3: $^{235}$U fission cross section from 10 keV to 10 MeV; the data shown are discussed in some detail in this paper and the curve is the modern ENDF evaluation. The earlier measurements had systematic errors resulting in over-estimations of the cross section. Frisch and Peierls' 1940 first guess of 10 b can't be seen on this scale.

Figure 4: Hans Bethe and Robert Bacher lab badges. They led the Theoretical Division and the Physics Division, respectively.

Evaluated Nuclear Data File ENDF/B-VIII.0 database and in the Plutonium Handbook. I will also show how uncertainties in cross sections, which were initially large (e.g. Oppenheimer suggested an uncertainty of 0.5 b on a $^{235}$U fission cross section he estimated as 2 b in a 1942 letter to Peierls), were substantially reduced to typically < 5-10% by the end of the project in 1945. Furthermore, some of the data described here have never before been openly available, and these data and related documentation are now being made available to the broader nuclear science community, via the IAEA’s Nuclear Data Section and Brookhaven National Laboratory (BNL) who have put them into the EXFOR file.

This paper does not attempt to provide a comprehensive review of all Manhattan Project nuclear science, and only covers the nuclear data used to compute fast bare critical masses. Therefore many fascinating topics are not covered here or are only covered briefly, including: tamped critical masses (see this issue, Ref.); fission delayed-neutron measurements; spontaneous fission properties; (alpha,n) reactions that meant impurities would be a concern and plutonium purification methods had to be developed (see this issue, Ref.); lower energy neutron resonance and thermal fission measurements; fission product radiochemical measurements (see this issue, Ref.), and neutron scattering measurements on non-actinides.

In April 1943, at the beginning of Los Alamos’ Project Y work, a conference was held in which the current state of understanding was summarized, see Fig. 5. Oppenheimer’s opening paper started with the fundamental nuclear data – neutron cross sections, energy spectra, and so on – that are needed to compute a fast critical mass of $^{235}$U and $^{239}$Pu. He showed a critical mass analytic formula that was in use at the time to compute critical masses.
Outline of the Organization of the Conference

1. Outline of present knowledge and main questions to be solved - OPPENHEIMER

2. Experimental results and description of available equipment - MANLEY

3. Physical constants affecting the mass of untapped gadget - BETHE

4. Energy release, role and properties of tamper - SERBER
   Information available on tube alloys - KONOPINSKI

5. Problems of detonation, detonation by shooting - OPPENHEIMER
   Autocatalysis - TELLER

6. Theory and description of slow pile - FERMI and CHRISTY
   Water-trailer - CHRISTY

7. Use of thermo-nuclear reactions - TELLER

8. Testing of gadget and expected damage - BETHE

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(1) Outline of Present Knowledge and Main Problems to be Solved
    ---Oppenheimer

Figure 5: The agenda for the first Los Alamos conference, April 1943, from LA-2.
This perspective defined the nuclear science and technology research program, worked out in detail by Bethe, Manley and Bacher (Figs. 4, 6) that was put in place for the next 30 months, see Fig. 7. In this paper, I will follow this same approach. I will first describe the critical mass formula that was used between 1943 and 1945, and show how calculations of critical masses changed as the Los Alamos scientists established increasingly-accurate experimental results.

II. CRITICAL MASS CALCULATIONS

Soon after Frisch and Peierls’ secret March 1940 memorandum\(^6\) that predicted the \(^{235}\text{U}\) critical mass to be 0.6 kg, scientists in Britain and the US realized it was likely a substantial under-prediction. The subsequent fission cross section measurements at Birmingham, and then Liverpool - even though they used natural uranium targets - were able to infer the \(^{235}\text{U}\) fission cross section more accurately, as we describe later. This led the British MAUD Committee,\(^8\) in the summer of 1941, to provide an updated assessment of the critical mass in the range 9 kg (“most likely”) to 43 kg (“pessimistic”). As we will see, this range proved to be reasonable with MAUD’s pessimistic value being close to the true value of 46 kg for pure \(^{235}\text{U}\) (the critical mass would be higher still for highly-enriched uranium versus pure \(^{235}\text{U}\)).

In the US in November 1941, Compton’s National Academy of Sciences (NAS) committee did an assessment and estimated the \(^{235}\text{U}\) critical mass to be in the range 2–100 kg.\(^11,12\) Subsequently, at Compton’s request, in February 1942 Breit and Oppenheimer were asked to study the problem. They came to a result of 5 kg, a value not far from the MAUD Committee’s “most likely” value of 9 kg (Ref.,\(^27\) p.27). By the beginning of Project Y’s work at Los Alamos, in March 1943, more work had been done; other scientists such as Serber had been engaged, and at that stage the estimate of the bare \(^{235}\text{U}\) critical mass was about 60 kg (see Oppenheimer’s summary in Fig. 8, as well as Serber’s Primer\(^36\) Los Alamos report LA-1 describing how the use of more exact diffusion theory reduces the calculated critical mass estimate from 200 kg to 60 kg).

Also, Richard Tolman wrote\(^37\) a useful “Memorandum on Los Alamos Project as of March 1943”, summarizing many aspects of the current state of knowledge of nuclear and material science, where he assessed that the bare critical mass for \(^{235}\text{U}\) is 30±15 kg, while that for \(^{239}\text{Pu}\) is 10 kg (5–20 kg range), estimates that would prove to be rather good.

It was always realized that the greatest challenge for the Manhattan Project was the production of adequate amounts of fissionable nuclear materials. In comparison, the design of the bomb was somewhat easier, certainly for the uranium gun bomb (Little Boy), although once it was realized that this same gun approach for plutonium (Thin Man) would not work owing to the high spontaneous fission rate of the contaminant \(^{240}\text{Pu}\), the design of an implosion plutonium bomb (Fat Man) would prove to have substantial technical challenges, as described by Chadwick and Chadwick.\(^38\) For this reason, an accurate theoretical prediction of the critical masses was one of the highest priorities at Los Alamos. Because it was realized that substantial amounts of \(^{235}\text{U}\) and \(^{239}\text{Pu}\) would not be sent to Los Alamos for a year or two (enabling a direct integral measurements of the critical mass), Project Y focused on theoretical predictions of bare and tamped (reflected) critical masses and in determining the underlying nuclear cross section data needed for these predictions.

Beginning with Peierls’ 1939 work to derive the critical mass of an unmoderated fast critical assembly, simple analytic equations were used to calculate the fast (unmoderated) critical mass for \(^{235}\text{U}\) and \(^{239}\text{Pu}\). The historical evolution of such formulae will not be in the scope of this paper; instead, I present the formula used throughout the Manhattan Project. The rest of the paper follows the Los Alamos scientists approach and describes the research advances that they made to determine the fundamental nuclear constants that go into this formula.

The bare critical mass \(M_C\) (see Fig. 8) was taken as:

\[
M_C = \frac{4\pi^4}{35/2} \left( \frac{A}{N_A} \right)^3 \left[ \sigma_T^{-1/2} \sigma_F^{-1/2} (\bar{\sigma} - 1)^{-1/2} \right]^3 \left( \frac{1}{\rho} \right)^2 \times (1 + 0.9(\bar{\sigma} - 1) \sigma_F / \sigma_T)^{-3} (1)
\]

where \(\rho\) is the density in g/cm\(^3\), \(\sigma_F\) is the fission cross section and \(\bar{\sigma}\) the prompt fission neutron multiplicity, and

\[
\sigma_T = \frac{1}{\rho} \sum_{i} N_i \sigma_{fi}(\rho) \quad \text{and} \quad \sigma_F = \frac{1}{\rho} \sum_{i} N_i \sigma_{fi}(\rho)
\]

\[
\bar{\sigma} = \frac{1}{\rho} \sum_{i} N_i \sigma_f(\rho)
\]

where \(\sigma_{fi}(\rho)\) is the neutron fission cross section for isotope \(i\) at density \(\rho\), \(\sigma_f(\rho)\) is the total neutron cross section at density \(\rho\), and \(N_i\) is the number density of isotope \(i\).
Figure 7: Manley’s summary of the nuclear science program needed for the Manhattan Project’s success, from LA-2, March 1943.

\[ r_C = \frac{\pi}{\sqrt{3}} \sqrt{\frac{1}{(\nu - 1) \cdot n \sigma_T \cdot n \sigma_F}} \times f \]  

(2)

where \( n \) is the atom number density, \( \sigma_F \) is the fission cross section and \( \nu \) the prompt fission neutron multiplicity, and \( \sigma_T \) is the transport cross section representing the total cross section for all processes except those scatterings leading to neutrons continuing in the forward direction. \( f \) is a correction factor derived from “more accurate integral theory” (see Bethe’s summary, Fig. 9),

\[ f = \frac{1}{(1 + 0.9(\nu - 1)\sigma_F/\sigma_T)} \]  

(3)

This critical radius, when used for a solid sphere, provides the critical mass shown in Eq. (1) using \( M_C = 4/3\pi r_C^3 \rho \). Given its simplicity the formula is remarkably accurate. This can be seen by comparing the critical mass calculated with this formula with that obtained using our most sophisticated Monte Carlo neutron transport code today, MCNP6\(^c\) with ENDF/B-VIII.0. For the formula, ENDF/B-VIII.0 parameters are taken for a typical average neutron energy, 1.5 MeV.\(^f\) This comparison is shown in the last row of Table 1 where the exact MCNP6 result is 46.36 kg but the formula Eq.1 gives 55 kg. The critical mass formula over-prediction is approximately 20%, but the critical radius over-prediction is just 6%, rather impressive for so simple an equation that has just one energy group, i.e. it considers relevant nuclear constants at just one average neutron energy (and as seen in Fig. 3, the \(^{235}\)U fission cross section is fairly constant in the fast range, unlike the \(^{238}\)U cross section). By late 1944, such simple formulae were starting to be replaced by advances in neutronics simulations that were enabled by the first IBM punched card accounting machines\(^{40–42}\) to include multi-group treatments, inelastic scattering effects, and polynomial representations of angular scattering effects.

Derivations of these formulae are not provided here (see Serber’s Primer\(^{36}\) pp. 25-27 for a useful discussion); instead I will give the reader a feel for the physical basis for the critical radius Eq. 2. One might expect that the critical radius would be proportional to the mean free path before a fission collision, \( l_{FP} \) and inversely proportional to the neutron production produced in that collision, \( (\nu - 1) \), that is, proportional to \( l_{FP}^{-1} = 1/(\nu - 1) \cdot n \cdot \sigma_F \), \( n \) being the atom number density. But we might also expect the critical radius to be proportional to the mean free path for all collisions since this includes scattering the neutrons and limits their ability to stream out and escape, i.e. \( l_{FP}^{-1} = 1/(n \cdot \sigma_T) \). In fact, neutron diffusion theory

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\(^f\)Jen Alwin calculates the average energy causing fission in a critical sphere of \(^{235}\)U to be 1.48 MeV. For Jezebel, a sphere of plutonium, the average neutron energy causing fission is 1.88 MeV\(^{39}\)
The actual minimum (or critical) amount of material necessary to sustain a fast neutron chain reaction for an untamped gadget has been obtained from calculation as:

\[ M_c = \left( \frac{4\pi^2}{3} \right)^{\frac{3}{2}} \left\{ \sigma_t^{-1} \left( \frac{\nu - 1}{\nu} \right)^{\frac{1}{3}} \rho^{-\frac{1}{2}} \right\} \left\{ \frac{1}{1 + \left( \nu \sigma_f / \rho \right) \sigma_t} \right\} \]

where \( A \) is the atomic weight; \( N \) is Avogadro's number; \( \sigma_t \) the transport cross-section of the gadget material; \( \sigma_f \) the fission cross-section; \( \nu \) the number of neutrons per fission; and \( \rho \) the density of the gadget. If we omit the last factor in the formula, the remaining part is the result of a calculation based on differential diffusion theory. The last factor gives the correction due to integral theory.

Using the value for the constants

\[ \sigma_f = 4 \quad \text{(measured for 28 only)} \]
\[ \sigma_t = 1.5 \quad \text{(for 25)} \]
\[ \rho = 19 \]
\[ \nu = 2.2 \quad \text{(observed for thermal neutron fission of 25)} \]

one obtains the critical mass of 25, \( M_c = 200 \text{ Kg} \) from the differential theory and \( M_c = 60 \text{ Kg} \) from the integral theory.

If the gadget is surrounded by a neutron reflector or tamper in which no absorption or fission takes place and in which the transport mean free path is the same as in the gadget, we obtain for the critical mass the expression

\[ M_c = \frac{4\pi^2}{3} \left( \frac{\nu}{\nu - 1} \right)^{\frac{3}{2}} \left\{ \sigma_t^{-\frac{1}{2}} \left( \frac{\nu - 1}{\nu} \right)^{\frac{1}{3}} \rho^{-\frac{1}{2}} \right\} \left\{ 1 + \left( \nu \sigma_f / \rho \right) \sigma_t \right\} \]

The constants and factors have the same significance as in the previous expression. On the differential theory the critical amount is now 25 Kg of 25. On the integral theory this becomes 15 Kg; whereas for 48, on the integral theory, only about 4 Kg are needed.

Figure 8: Oppenheimer's summary of critical mass calculations from Los Alamos' first conference, April 1943, LA-2. Because the image is of poor quality, the first equation appears to have typos, with the parameters \( \sigma_t, \sigma_f, (\nu - 1) \) raised to the power \(-1/2\). A careful study though shows that they are correctly raised to the power \(-3/2\), as in Eq. (1).
REPORT OF THE THIRD MEETING HELD ON APRIL 17, 1943.

(3) The physical constants determining the critical mass and efficiency of an untamped gadget.

--- Bethe.

1. The diffusion theory critical mass and multiplication time: The rate of growth of the neutron density, $F$, is given by,

$$\frac{dF}{dt} = \frac{1}{3} J \Delta F + \frac{\nu'}{\bar{v}} F$$

where $l$ is the transport mean free path, $\nu$ is an appropriate average of the neutron velocities, $\nu'$ is the number of neutrons emitted per fission, and $t$ is the mean time between fission collisions. Aside from transient solutions, there is the persistent solution varying exponentially in time as

$$F \sim e^{-\nu' \sqrt{t}}$$

The space dependence of the solution appropriate for a spherical mass is

$$F \sim \frac{\sin \kappa r}{r}$$

where $\kappa^2 = \frac{n (\nu - \nu' - 1)}{\bar{v}}$,

where $\bar{v}$ is the (1 - cos)$^2$ average of the elastic scattering cross section plus all the reaction cross sections.

According to elementary diffusion theory the density $F$ should vanish at the boundary, which gives $\kappa R = \bar{v}$, hence

$$R = \frac{\kappa}{\bar{v}} \sqrt{\frac{\kappa R}{\bar{v}}}$$

The elementary diffusion theory is accurate only in the limit of dimensions large compared to the mean free path and for small absorption. An approach to the correct solution is obtained by making the density vanish a distance outside the actual boundary. The critical radius, i.e., that for a static solution, is given by setting $\nu' \phi$. The more accurate integral theory divides the above derived critical radius by $(\nu' (\nu' + \bar{v})/\bar{v})$. 

Figure 9: Bethe's summary of critical mass calculations from Los Alamos' first conference, April 1943, LA-2.34
finds that the critical radius \( r_C \) is proportional to both these quantities as their geometric mean (the square-root of their product), with a pre-factor of \( \pi/\sqrt{3} \), which is Eq. (2), and the result Bethe shows in Fig. 9 (noting that \( v\tau \) in Bethe’s equation, the product of the neutron velocity and the time between fission collisions, is \( v\tau=1/n\sigma F \)). As a point of reference, for a critical sphere of \( ^{235}\text{U} \) with mass 46 kg, the critical radius is 8.8 cm, whereas the fission mean free path is \( l_{F}^{\text{MFP}}=17 \text{ cm} \), and the transport mean free path is \( l_{T}^{\text{MFP}}=4 \text{ cm} \).

Using this formula, critical masses are calculated using different assumptions regarding the nuclear constants, in Table 1 for \( ^{235}\text{U} \) and Table 2 for \( ^{239}\text{Pu} \), assuming idealized spheres of the single isotope. Figure 10 shows these bare critical masses. The tables show the constants used, and the last column provides the critical mass value calculated and, in parenthesis, the value reported in the original reference. (Note that in all rows except the last, the values in the last column are from the analytic equation; in the last row for ENDF/B-VIII.0, the reported value in parenthesis comes instead from a full high-fidelity MCNP simulation.) In some cases there is very good agreement between the value I calculate from Eq. 1 and that reported by the original authors, indicating, for example, that Bethe in LA-32 or the British in MAUD (1941) faithfully computed what they intended to compute. In other cases, for example Oppenheimer’s result in Table 1 (60 kg in LA-2, see Figure 8) there is a lack of consistency, for the nuclear parameters Oppenheimer lists in Figure 8 result in 77 kg for \( ^{235}\text{U} \), not the 60 kg he quotes. Perhaps an example of Oppenheimer’s sloppiness when it came to math errors?  

For brevity this paper describes just bare critical mass comparisons, but the focus at Los Alamos in 1943 quickly became tamped (reflected) critical masses (as described in Hutchinson’s paper in this issue). Tamped critical assemblies typically require a little less than half the amount of \( ^{235}\text{U} \) or \( ^{239}\text{Pu} \) compared to the bare assemblies, and therefore they use the available precious special nuclear material more efficiently. Indeed, this is the reason that Tables 1, 2 (far-right column) do not show a reported critical mass from Serber and Rarika’s 1945 paper, LA–235 – the focus then was entirely on tamped assemblies. A for-
Table 1: $^{235}\text{U}$ bare critical mass calculated with the listed nuclear data “constants” using Eq.(1). In the last column, the critical mass in parenthesis is the one reported in the original reference. The last row shows our best values today (ENDF/B-VIII.0) at 1.5 MeV neutron energy, the critical mass in parenthesis here coming from an MCNP6 calculation.

| Author, Date | $\sigma_F$ | $\sigma_T$ | $\rho$ | Crit. Mass $^{235}\text{U}$ (kg) |
|--------------|-----------|-----------|-------|-------------------------------|
| Frisch, ’40  | 10. 2.3   | 10. 15.   |       | 0.44(0.6)                     |
| Peierls’6    |           |           |       |                               |
| MAUD’41      | 2. 3. 5.  | 19.6      |       | 8.5(9.)                       |
| likely’8     |           |           |       |                               |
| MAUD’41      | 1.5 2.5  | 3.5 19.6  |       | 44.5(43.)                     |
| pessimistic’8|           |           |       |                               |
| Compton case | 3. 3. 9.  | 18.6      |       | 2.6(3.4)                      |
| ’41, NAS’11,12 |           |           |       |                               |
| Oppenh. ’43, LA-234 | 1.5 2.2  | 4. 19.   |       | 77.(60.)                      |
| Bethe ’43, LA-32’43 | 1.6 2.  | 5. 19.   |       | 85.(83.)                      |
| Serber, ’45 | 1.34 2.4 | 4.7 18.7 |       | 66. (−)                       |
| LA-235      |           |           |       |                               |
| ENDF’29, 2018 | 1.24 2.57| 5.0 18.9 |       | 55.(46.)                      |

Table 2: $^{239}\text{Pu}$ bare alpha-phase critical mass calculated with the listed nuclear data “constants” using Eq.(1). In the last column, the critical mass in parenthesis is the one reported in the original reference. The last row shows our best values today (ENDF/B-VIII.0) at 1.5 MeV neutron energy, the critical mass in parenthesis here coming from an MCNP6 calculation.

| Author, Date | $\sigma_F$ | $\sigma_T$ | $\rho$ | Crit. Mass $^{235}\text{U}$ (kg) |
|--------------|-----------|-----------|-------|-------------------------------|
| Oppenh. ’43, LA-2 ’43 | 3. 2.2 | 4. 19. |       | 13.(15.)                    |
| Bethe’43 | 2.6 2.  | 5. 19.   |       | 29.(31.)                     |
| LA-32, ’43 |           |           |       |                               |
| Serber’44 | 1.85 2.8 | 4.8 19.4 |       | 14.(−)                       |
| LA-235, ’45 |           |           |       |                               |
| ENDF’29, 2018 | 1.93 3.09| 5.29 19.6|       | 8.6(10.2)                    |
mula for the tamped mass is also shown in Oppenheimer’s paper in Fig. 8. In that Figure, we see that in April 1943 Oppenheimer estimated a tamped critical mass for $^{239}$Pu of 4 kg; today with our best simulation tools, MCNP6 with ENDF/B-VIII.0 data, we find the same result, 4.0 kg of $\alpha$-phase plutonium, for an infinite $^{238}$U tamper. As was the case for $^{235}$U, we will show that again, this agreement was a lucky outcome resulting from canceling errors between fission $\sigma_F$ and $\nu$ assessments.

The rest of this paper is devoted to discussing the developing understanding of the nuclear cross sections used in Tables 1, 2, based on the research programs in Britain and the USA through 1945.

### III. EVALUATIONS OF CROSS SECTION DATA

The concept of periodically documenting our best understanding of nuclear cross sections began at Los Alamos with Bethe, setting the stage for a series of documents with the name “Los Alamos Handbook of Nuclear Physics”. After the war, this scholarly approach was continued at Los Alamos (e.g. Hansen and Roach), and was adopted by other laboratories, e.g. Goldsmith at Brookhaven Laboratory, leading to Brookhaven’s well-known BNL-325 documents on neutron cross sections. It was later extended by the international nuclear science communities who created the Evaluated Nuclear Data Files (ENDF) databases in the US, JEFF in Europe, BROND in Russia, JENDL in Japan, and CENDL in China. The ENDF databases have been documented in detail in a series of publications in Elsevier’s Nuclear Data Sheets.

The term “evaluated data” describes the process by which subject matter experts have reviewed available information from measurements, together with physics insights from theory and modeling, to determine “best values” that are recommended. Theory and modeling often play a role in extending the measured data to regions of energy or scattering angle that have not been measured. Such evaluated data can then be used in neutron transport codes for applied nuclear technology calculations.

Table 3 lists the main review documents written during the project, especially the three versions of the Los Alamos Handbook of Nuclear Physics. The earlier MAUD reference is included because it documented what was known in 1941, much of which proved to be remarkably accurate and played an important role in influencing subsequent efforts. Beyond these evaluated data compilations, there were many calculational papers written during the Manhattan Project, by Bethe, Christy, Peierls, Serber, and others, that summarized the data they were using to calculate critical masses and yields. Table 3 highlights just one of those, the LA-235 article by Serber, which provides a useful summary of what was being used in the Theoretical Division towards the end of the project.

Robert Wilson’s comprehensive summing-up of what was learned at Los Alamos is the last entry, being written in 1947. This “Nuclear Physics” LA-1009 review is quite remarkable in the breadth of its scope, and in describing what had been accomplished during the project under intense time deadlines.

The present work is making a number of historically-important measurements available to the broader nuclear science and technology community, via the internationally-maintained EXFOR database, through collaboration with the IAEA’s Nuclear Data Section and BNL. These publications are listed in Table 4.

Below, we summarize the early work in Britain and the US at Berkeley, Carnegie (Washington DC), Wisconsin, Chicago, and Rice before the Manhattan Project began, and what was subsequently accomplished at Los Alamos by 1945. We compare the advances made with our best understanding of these data today, as embodied in our latest ENDF/B-VIII.0 evaluated cross section database.

### IV. FISSION CROSS SECTIONS

In the 1930s and early 1940s, fast cross sections had been measured in three neutron energy ranges, requiring different sources. Photoneutrons from 0.1 – 1 MeV were used from radioactive gamma sources (e.g. $\gamma$-Be with radioactive $^{88}$Y made in a cyclotron from the (p,n) reaction on Sr; and a Ra-Be photoneutron source) allowing

| Author       | Date   | Reference          |
|--------------|--------|--------------------|
| Chadwick,    | 1941   | MAUD, UK           |
| Thomson      |        |                    |
| Oppenheimer, | 1943   | LA-2               |
| Bethe        |        |                    |
| Christy      | 1943   | LA-11 Handbook     |
| Bretsch, Davis, | 9/1944 | LA-140 Handbook    |
| Inglis, Weisskopf | 3/1945 | LA-140A Handbook   |
| Serber       | 3/1945 | LA-235             |
| Wilson       | 1947   | LA-1009 Nuclear Physics |

$^h$Pure plutonium is in the alpha phase at room temperature; this is distinguished from its delta-phase when alloyed with gallium, which has a lower density and larger critical mass, e.g. as used in the Jezebel critical assembly.
1940, led to urgent efforts to measure the fission cross section. This was restricted to obtaining 235\textsuperscript{U} fission data for neutrons comprising energies from 0.2 MeV to nearly 1 MeV. The value obtained was 2.3E-24cm\textsuperscript{2} (this was the MAUD report, which stated: “The Liverpool measurements give values for the fission cross section of 235\textsuperscript{U} varying from 2.1E-24 cm\textsuperscript{2} for neutrons of about 0.35 MeV to 1.5 E-24 cm\textsuperscript{2} for neutrons of about 0.8 MeV.” (versus our best values today of 1.2 b and 1.1 b, respectively). Then MAUD says “Another measurement of this quantity has been obtained by Dr. Frisch using a mixed beam of neutrons comprising energies from 0.2 MeV to nearly 1 MeV. The value obtained was 2.3E-24cm\textsuperscript{2}” (this was the aforementioned 1940 Birmingham measurement).

A uranium sample enriched in 235\textsuperscript{U} to 15% was sent from Lawrence’s Berkeley laboratory to Liverpool in December 1942, and enabled Bretscher to determine the 235\textsuperscript{U} fission cross section above the 238\textsuperscript{U} fission threshold (see below). Brown says (Ref.,\textsuperscript{18} p. 24) of Lawrence, “it says much about his opinion of Chadwick and the Liverpool department that he was prepared to part with the precious sample”. From the British perspective, the sample was sent just in time, for Roosevelt was convinced that a new policy be put in place to limit information flow between countries if that information could not be used to win the war. Conant’s January 1942 memorandum on areas that would be impacted included fast neutron reactions (Ref.,\textsuperscript{18} p. 235). There were also discussions on sending a plutonium sample to Britain, but the risk of its loss in transit from enemy attack was considered to be too high.

235\textsuperscript{U} fission to be measured without interference from 238\textsuperscript{U}; between 2 – 3 MeV, D(d, n) neutrons were made from low-voltage accelerators; and around 4 MeV, broad-energy source neutrons were made from Ra-Be sources. For the higher (>2 MeV) source neutron energies, fission on natural uranium targets was dominated by 239\textsuperscript{Pu} fission. These methods, and the subsequent development of monoenergetic sources (e.g. via Li(p, n) reactions with accurately-controlled accelerator voltages) were reviewed by Goldsmith\textsuperscript{45} in 1947.

### IV.A. Early Work in Britain

The MAUD committee’s deliberations, including their study of the Frisch-Peierls memorandum from March 1940, led to urgent efforts to measure the fission cross section. Before moving to Liverpool in August, Frisch and Titterton set up fission measurements on natural uranium using a Ra–Be photoneutron source at Birmingham. This was restricted to obtaining 235\textsuperscript{U} fission data for neutron energies 0.2-1 MeV that were below the 238\textsuperscript{U} fission threshold. They found a 235\textsuperscript{U} fission cross section of 2.3 b that showed that the previous 10 b estimate was too high (Ref.\textsuperscript{5} p.131).\textsuperscript{67} Afterwards in 1945 Chadwick wrote that it was “The first reasonable measurement, but still very rough” in handwritten marginal notes in Peierls’ 1945 summary which is being made available by Moore in this Issue\textsuperscript{67}. From Bretscher’s papers at Churchill College Cambridge, it is evident that in late 1940 he also made a measurement using the high tension “H.T. set” (high voltage) accelerator at Cambridge,\textsuperscript{68} again with a natural uranium source, using neutrons from the d+C reaction. By combining his data with the aforementioned Princeton measurements by Ladenburg \textit{et al.}, he was able to conclude that for neutrons produced near 1.8 MeV energy, the fission cross section of 235\textsuperscript{U} had to be less than 2.8 b. (We now know this is indeed the case; its value is about half of this). Bretscher concluded by saying “The only way to get a better value for this cross section seems now to enrich U in U(235).”\textsuperscript{68}

In the Autumn of 1940, Chadwick, Frisch and collaborators (Ref.,\textsuperscript{18} p. 203) used the Liverpool cyclotron with unenriched natural uranium targets with a Li(p,n) source reaction to measure the cross section below 1 MeV (to a factor of 2 or better). Peierls stated that they showed “Frisch’s previous figure as somewhat high and also that the cross section decreased with increasing energy”.\textsuperscript{67} These Liverpool and Birmingham values were reproduced in the MAUD report, which stated: “The Liverpool measurements give values for the fission cross section of 235\textsuperscript{U} varying from 2.1E-24 cm\textsuperscript{2} for neutrons of about 0.35 MeV to 1.5 E-24 cm\textsuperscript{2} for neutrons of about 0.8 MeV.” (versus our best values today of 1.2 b and 1.1 b, respectively). Then MAUD says “Another measurement of this quantity has been obtained by Dr. Frisch using a mixed beam of neutrons comprising energies from 0.2 MeV to nearly 1 MeV. The value obtained was 2.3E-24cm\textsuperscript{2}” (this was the aforementioned 1940 Birmingham measurement).

### Table 4: Measurements newly added to EXFOR through this work, via the IAEA Nuclear Data Section and BNL. In the references, “CF” reports stands for Central File Number; these reports are now held at ORNL/OSTI.

| Author          | Lab       | Reaction        |
|-----------------|-----------|-----------------|
| Tuve            | Carnegie  | 235\textsuperscript{U}(n,f) |
| Chadwick        | Liverpool | 235\textsuperscript{U}(n,f) |
| Frisch          | Birmingham| 235\textsuperscript{U}(n,f) |
| Chamberlain     | Berkeley  | 235\textsuperscript{U}(n,f) |
| Kennedy, Segrè  | Berkeley  | 235\textsuperscript{U}(n,f) |
| Heydenburg      | Carnegie  | 235\textsuperscript{U}(n,f) |
| Hanson          | Wisconsin | 235\textsuperscript{U}(n,f) |
| Benedict        | Wisconsin | 235\textsuperscript{U}(n,f) |
| Williams        | Wisconsin | 1H(n,p)         |
| Bloch           | Stanford  | 235\textsuperscript{U}(n,f) PFNS |
| Chadwick        | Cambridge | 235\textsuperscript{U}(n,f) |
| Kinsey          | Cambridge | 235\textsuperscript{U}(n,f) |
| Wiegland, Segrè | Los Alamos| 239\textsuperscript{Pu}/235\textsuperscript{U}(n,f) |
| Koonitz, Hall   | Los Alamos| 235\textsuperscript{U}(n,f) |
| Williams        | Los Alamos| 239\textsuperscript{Pu}/235\textsuperscript{U}(n,f) |
| Snyder, Williams| Los Alamos| 239\textsuperscript{Pu}/235\textsuperscript{U}(n,f) |
| Wilson          | Los Alamos| 239\textsuperscript{Pu}/235\textsuperscript{U}(n,f) |

| Issue summary which is being made available by Moore in this Issue\textsuperscript{67}. From Bretscher’s papers at Churchill College Cambridge, it is evident that in late 1940 he also made a measurement using the high tension “H.T. set” (high voltage) accelerator at Cambridge,\textsuperscript{68} again with a natural uranium source, using neutrons from the d+C reaction. By combining his data with the aforementioned Princeton measurements by Ladenburg \textit{et al.}, he was able to conclude that for neutrons produced near 1.8 MeV energy, the fission cross section of 235\textsuperscript{U} had to be less than 2.8 b. (We now know this is indeed the case; its value is about half of this). Bretscher concluded by saying “The only way to get a better value for this cross section seems now to enrich U in U(235).”\textsuperscript{68}

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Later in 1943, following the August 19, 1943 Quebec agreement between “The U.K. and the U.S.A. in the Matter of Tube Alloys”, the British learned for the first time about the Los Alamos site in New Mexico where there were already hundreds of scientists working under Oppenheimer. Approximately 25 British scientists...
would move to Los Alamos beginning in December 1943, and consequently the British nuclear science effort was then curtailed. Chadwick led the British Mission in Los Alamos, although in practice spent most of his time in Washington, helping ensure the smooth-running of the collaboration’s administrative aspects. Peierls deputized for Chadwick and had a highly impactful role at Los Alamos as Bethe’s Theoretical Division’s T-1 group leader for implosion physics (at Los Alamos he was known as much for his hydrodynamics expertise as for his nuclear physics). Frisch became the leader of the Gadget Division’s criticality group, G-1, and made numerous nuclear physics contributions at Los Alamos (for example, the Dragon experiment described in this issue), and Peierls tells the nice story that Frisch was equally renowned for his talents on the piano, and at one musical evening in Los Alamos the remark was overheard: “This guy is wasting his time doing physics!” (Ref. p.133).

Other notable measurements made in England during the war are two fast \(^{235}\text{U}\) fission measurements made in 1944, by Bretscher, and by Chadwick and Kinsey, reported in Koontz’s August 1944 Los Alamos report LA-128 and Robert Wilson’s 1947 Nuclear Physics review LA-1009. These are shown below in Fig. 11 and Table 5 and are seen to be fairly accurate. They were made using the enriched \(^{235}\text{U}\) target sent from the USA, before Bretscher and Chadwick came to Los Alamos in 1944. This same LA-128 (p. 19) mentions Chadwick-Kinsey data used a three-counter method to measure the flux.

In this paper’s Introduction, Bretscher, Feather, and Rotblat’s early 1940 insights into the high potential of \(^{239}\text{Pu}\) were noted. Here I transcribe Bretscher’s progress report from December 1940:

“Report on Work Carried out in Cambridge, September - December, 1940”, from Churchill College Cambridge’s Archive Center who holds papers of The Royal Commission on Historical Manuscripts, Egon Bretscher, CBE, Tube Alloys, D.37 (1940): Two distinct lines of research have been followed, (i) by Halban’s and Kowarski’s team on the conditions necessary for the realization of a divergent chain of fissions produced by slow neutrons, and (ii) by Bretscher and his collaborators, in the Cavendish H.T. Laboratory, on problems bearing upon the realization of a divergent chain with fast (or medium fast) neutrons. It was assumed at the outset that only (ii) but immediate relevance to the problem of producing super-explosives, but, as the following Reports show, it now appears likely that the success which has already been achieved in (i) one method of producing a divergent chain has been shown to the practicable – see Report 1) will enable us to approach (ii) with the greatest chance of success, also. Briefly, this comes from the following considerations. Early work was based on the expectation that separation of the rare isotope \(^{235}\text{U}\) was the necessarily preliminary to the practical realization of the divergent-chain super-explosive. It was considered likely that the fission properties of \(^{235}\text{U}\) would be satisfactory in this respect – but a difficult practical problem was encountered in showing conclusively that this was the case. (Part of Report 2 deals with an attempt in this direction). On the other hand, general considerations indicate that the body \(^{239}\text{X}_{94}\) would probably be even more satisfactory as regards fission properties than \(^{235}\text{U}\) and the realisation of the slow neutron chain process makes possible the production of this body, even more readily than \(^{235}\text{U}\) may be produced by isotope separation. For preliminary work on the fission properties of \(^{239}\text{X}_{94}\), it seems that sufficient material may be obtained by irradiation of large quantities of uranium on the Cavendish H.T. set. \(^{239}\text{X}_{94}\) is left after two successive \(\beta\) disintegrations of \(^{239}\text{U}\), formed from the abundant uranium isotope \(^{239}\text{U}\) by slow neutron capture. The advantage of depending upon a reaction of the abundant isotope (99.3%) does not require elaboration.

Note that Report II is by Bretscher (page 1), Report I by Halban and Kowarski (page 9), both in document D.37. James Chadwick pointed out that it was discussed still earlier, in his laboratory at Liverpool. After Peierls’ wrote in the late summer of 1940, when publications from Berkeley on the discovery of plutonium were received, the suggestion was made that this could be produced in quantity by means of a slow-neutron reaction, and that it was likely to be suitable for a military weapon. This suggestion was presented to the M.A.U.D. committee by the group at Cambridge where meanwhile Bretscher had begun some experiments on fast neutrons at Chadwick’s request, but I have no first-hand information on how exactly it originated. Chadwick adds as a marginal hand-written note (transcribed as a footnote in Ref.: First suggestion that I know of was made by Rotblat about June–Early July 1940. Common topic in my laboratory before Cambridge was brought in.”.

**IV.B. Early Work in the USA: \(^{235}\text{U}\)**

In a 1985 colloquium at Los Alamos Harold Agnew, Los Alamos’ third Director, gave a talk that described the beginnings of US nuclear science and his own role in the Manhattan Project. He noted that the earliest advances were occurring under Compton in Chicago, Fermi and Dunning at Columbia, and Lawrence at Berkeley. Early in 1940, Columbia physicist John R. Dunning studied the fissioning of a sample of \(^{235}\text{U}\) that had been separated in a mass spectrometer by Alfred O. Nier at the University of Minnesota. This work showed that \(^{235}\text{U}\) is the isotope responsible for slow neutron fission in uranium,
as predicted by Bohr and Wheeler. The thermal fission cross section that was measured, 400-500 b, is not far from the evaluated value today, 587.3 ±1.4 b. The 1940 state of understanding of nuclear fission was summarized in an extensive Review of Modern Physics paper by Louis Turner (Princeton), though that paper had only minimal references to neutron cross section measurements.

In 1942, as the USA expanded its efforts to assess the feasibility of developing an atomic bomb, the growth of nuclear research across the country’s universities was staggering. John Manley, at Chicago’s Met Lab, served as a manager for directing and coordinating the work at nine separate institutions: Berkeley (Seaborg, Segre), the Carnegie Institution in Washington (Heydenburg), Cornell (Rossi, Hollowell, Bacher), MIT, Minnesota (Williams), Purdue, Rice (Richards), Stanford (Bloch), and Wisconsin (McKibben). Los Alamos’s NSRC contains Manley’s extensive correspondence with these researchers.

As the preparations began for a Manhattan Project, Oppenheimer wrote on Nov. 1, 1942 to Rudolf Peierls in Britain to summarize his present understanding and raise some differences of opinion on the underlying science and technology. He summarized the recent 235U fission cross section experiments made at the Carnegie Institution and at Berkeley: Radioactive yttrium photons on Be making 220 keV neutrons gave 2.9 b and radioactive Na+D gave 1.8 b (these Berkeley values, described below, were reported as 2.8 b and 1.9 b respectively) and 12C+D making 500-900 keV neutrons gave 2 b (note this Carnegie value soon after was reported as 1.1 b, which we now know to be more accurate). The more accurate data from Wisconsin’s Van de Graaff were not yet available.

The unit of barn, 10−28m², has its origin during those times. In Los Alamos Manuscript Series LAMS-523, “Note on the origin of the term ‘barn’”, Baker and Holloway describe how they were inspired by their rural origins to choose this term, and that it was first concluded that for nuclear processes “a cross section of order 10−24cm² was really as big as a barn”. This was coined in December 1942 when they were at Purdue, working under John Manley’s direction. Its first written use was in December 1942 in their Los Alamos LAMS-2 report. They came to this choice, having ruled out (for various reasons) other possible names including an “Oppy”, a “Bethe”, a “Manley”, and a “John”.

At the Carnegie Institution of Washington’s Department of Terrestrial Magnetism (DTM), Heydenburg in late 1942 used a Van de Graaff neutron source from the reaction D+D and 12C + D→13N + n on targets of natural U and those enriched in 235U. The neutron energies available were limited by the maximum incident energy of the Carnegie electrostatic generator. The measurements suffered from high energy (1.8 and 5.6 MeV) neutrons from the presence of 13C which caused fission in 238U, which was present in the uranium sample. The value σF(25) =1.1 b was measured for 640 keV neutrons, from the C+D reaction. But the data from these C+D and D+D neutron source experiments was viewed as problematic at the time for the aforementioned reasons, even though their 1.1 b measured value is now known to agree exactly with our modern ENDF/B-VIII.0 assessment. It was concluded that Li(p,n) source reactions were preferred, as used at Wisconsin and Liverpool (see further below).

At Berkeley, Segrè and collaborators used radioactive sources to make neutrons. In 1942, Chamberlain, Kennedy and Segrè used quasi-monoenergetic photoneutrons from the photodisintegration of Be by yttrium radioactive-decay γ rays, and of deuterium by 24Na gamma rays. The powerful radioactive yttrium and sodium sources were made in the Berkeley cyclotron. Segrè introduced the idea, following Fermi, of a manganese bath to measure the neutron source strength: the neutrons were thermalized and the resulting activation from the capture reaction was measured. These experiments concluded that σF(25) = 2.8 b at 160 keV and 1.7 b for 430 keV neutrons, with uncertainties estimated as 15%. A natural uranium sample was used so that measured values could only be inferred for incident neutron energies below the 1 MeV 238U fission threshold.

A major breakthrough was made by the University of Wisconsin nuclear physics group group in 1942, through the development of quasi-monoenergetic source neutrons from the Li(p,n)Be reaction on enriched and natural uranium samples. The Van de Graaff electrostatic generator was capable of accelerating single charged species up to approximately 4 MeV, producing monoenergetic neutrons up to 2 MeV. Two different approaches were developed to determine the neutron flux, one of which measured recoil protons, comparing against the n-p collision cross section as a primary standard, the other used the manganese bath technique. In April 1943 this led to 235U fission results of 1.66 b by Hanson and 1.22 b by Benedict and Hanson at a neutron energy of 0.53 MeV, the latter result being impressively close to our best value today, 1.13 b. The Hanson measurement was the first ever that I am aware of that made a fission cross section measurement in ratio to the n-p scattering cross section, a standard approach used today. The neutron energy dependence was also investigated for the first time, from 0.2 to 1.8 MeV, indicating a constant value above 600 keV, with an increasing cross section from 600 keV to the lower energies. They also showed that 238U has a threshold of fission of 1.0 MeV, as predicted by Bohr and Wheeler, and rises to 0.45 barns at 1.8 MeV.
IV.C. Work at Los Alamos: $^{235}\text{U}$

At the beginning of the Manhattan Project, the state of understanding was discussed in the March 1943 conference and summarized in the LA-2 report, with relevant presentations by Oppenheimer, Bethe, and Manley. For the fast neutron energy region, the $^{235}\text{U}$ fission cross section was thought to be about 1.6 b, and the $^{239}\text{Pu}$ cross section was thought to be about twice as large. These assessments were influenced by the recent 1942/1943 uranium measurements from Wisconsin using the Li(p,n) source reaction, and plutonium measurements from the Carnegie Institution. The fission neutron average multiplicity $\bar{\nu}$ for $^{235}\text{U}$ was taken as 2.2 based on a recent measurement at Chicago by Fermi (Fermi, report CP-257,p.3), and the same value was assumed for $^{239}\text{Pu}$, which had not yet been measured. It was guessed, correctly, that the $\bar{\nu}$ for fast neutrons would be just a little larger than for thermal neutrons (which could be more easily measured). It was also recognized that the existing British and US measurements of the fission neutron energy spectrum suffered from serious systematic errors. The summary remarks by Bethe, Manley, and Bacher emphasized the large uncertainties in all these data, and the urgency to more accurately measure the key nuclear constants needed for the project.

Existing accelerator equipment was brought to Los Alamos from various universities across the country. Wisconsin provided two Van de Graaff electrostatic voltage generators (2.4 and 4 million volts), allowing the creation of quasimonoenergetic neutrons from a few tenths of an MeV up to 1.8 MeV using a Li(p,n) reaction and up to 6 MeV with D+D reactions. The “short-tank” accelerator had been built mainly by Joseph McKibben, and most of the group there came to Los Alamos. Dick Taschek got his PhD under Breit at UW in 1941 and both Donald Benedict and Alfred Hanson finished their PhDs in 1943 under Ray Herb, the architect of the best Van de Graaffs. These Van de Graaff machines, referred to as the “Short Tank” and “Long Tank” proved to be the most useful at Los Alamos for precision fission experiments, in William’s group. Illinois provided a 600 keV Cockroft Walton generator, creating 2.5-3 MeV neutrons with D+D reactions. Manley had worked with this machine at Illinois, and proved to be Oppenheimer’s “right hand man” in assembling all the accelerator equipment; having moved to Chicago, he put Harold Agnew in charge of moving the Illinois machine to Los Alamos and putting it in the basement of the Z building, for his group. For Robert Wilson’s group, a cyclotron was provided by Harvard that could produce protons up to 7 MeV and deuterons up to 11 MeV. Thermal neutrons were produced following a D+Be reaction with subsequent moderation in graphite, as well as neutrons with energies in the 0.001-100 eV range using time-of-flight methods. Manley describes the process by which “I was the one in charge of getting all those damned machines up to Los Alamos”, and the impressive feat of getting them all operational by July 1943.

Los Alamos established the practice of measuring cross sections relative to a H(n,p) “standard”, measuring the recoil protons from a thin film of hydrogenous material in the neutron beam. This was enabled by William’s and Bailey’s seminal measurement of the H(n,p) cross section. The better understanding of the neutron fluence in experiments allowed $^{235}\text{U}$ fission cross sections to be measured more accurately, versus earlier methods where as we discussed above discrepancies of 30% or more were common. Once fission cross sections were accurately determined at Los Alamos, they in turn were used as standards to determine other cross sections in relative measurements.

This method was used by Hall, Koonz and Rossi in 1944 to more accurately measure the fission cross section in a double chamber that measured fission recoils compared to proton recoils. Hall et al. used the Li(p,n) reaction with the Van de Graaff high voltage generator in building W for neutron energies up to 1.6 MeV, and a D-D source of neutrons at 2.5 MeV using the Cockroft-Walton in building Z. They established at 1 MeV a $^{235}\text{U}$ fission cross section of $1.33\pm0.05$ b, which served as a standard cross section at Los Alamos during the Manhattan project. Indeed, this value was reasonably accurate, differing by 10% with the best “standard” value today in ENDF/B-VIII.0 at 1 MeV, 1.203b±1.3%.

Later in October 1944, Williams reported on similar measurements, from 5 keV to 2 MeV, for a range of activities that include $^{235,8}\text{U}$, $^{239}\text{Pu}$, $^{237}\text{Np}$, . . . Compared to the work of Hall et al., Williams developed “long counters” that improved the accuracy below 400 keV and above 3 MeV, with help from Hanson who had come from Wisconsin. Williams used the Van de Graaff with a Li(p,n) source, obtaining data from 5 keV to 2 MeV. Thus, by the Fall of 1944 Williams was able to conclude that “the general form of $\sigma(25)$ as a function of neutron energy is fairly well established . . . and is known to between 5 and 10 percent from 200 keV to 2 MeV”. This assessment was actually pretty optimistic; we now know that at the important higher energy of 1.5 MeV Williams’ LA-150 $^{235}\text{U}(n,f)$ data of 1.32 b was fortunately only 6% too high, but below 1 MeV his data remained substantially high (10% overestimate at 1 MeV, 25% at 500 keV, 61% at 100 keV, 56% at 30 keV). Late in 1944, the Los Alamos Handbook of Nuclear Physics (LA-140), 2nd Ed., by

\footnote{Yes, the spy. See A. Carr, “The Project Y Spies”, Los Alamos report LA-UR-28986 (2014).}
Bretscher, Davis, Inglis, Weisskopf summarized all these data for use by Los Alamos’ researchers.

After the war ended, in Nov. 1945 Bailey, Wilson et al. published\textsuperscript{77} updates to the LA-150 data of Williams in the region 25 – 500 keV. This work correctly argued that the previous LA-150 cross sections were too high for the lower neutron energies below 500 keV. These new results brought the fission cross section down, but the Bailey values were still high compared to our best estimates today. Likewise, David Frisch’s proportional counter results in 1946 also provided lower fission cross section measurements that were lower and more accurate,\textsuperscript{78} see Fig. 3.

Over time the better cross section measurements led to smaller cross sections in the fast region, as can be seen in Table 5 and Fig. 11 for the fission cross section at 1.5 MeV. This is because the cross section increases substantially as one goes to energies below 0.5 MeV; earlier measurements were more subject to systematic error contamination processes in which the source neutrons were down-scattered by surrounding material. Dr. Fredrik Tovesson of ANL stated to me that “I have seen this artificially increase the measured fission cross section before, both for fissile targets and non-fissile targets with fissile contaminants. Additionally, many early measurements in the fast region used quasi-monoenergetic neutron sources, while newer measurements have often been performed at time-of-flight (TOF) facilities such as LANSCE/WNR.\textsuperscript{80} When using neutron TOF you are less susceptible to low-energy neutron background, which supports the hypothesis”. In Table 5 a high-accuracy measurement is shown for comparison, by Dr. Allan Carlson,\textsuperscript{79} made at the National Bureau of Standards’ linac. This measurement is absolute, with the neutron fluence determined from a well-characterized black detector and the fission rate determined with a well-understood fission chamber.

Carlson, who worked with some of the early Los Alamos researchers, adds a useful perspective: “Carl Bailey who also worked on cross section measurements with Williams said the work was very primitive by today’s standards. The cross sections measured were too high because backgrounds were very high and difficult to properly mea-
Table 5: $^{235}$U fission cross sections near 1.5 MeV “fast” neutron energy, from measurements and evaluations (italicized). The exact energy or ranges of energies are given in parentheses. The “LA-” references denote work done at Los Alamos.

| Authors                        | Date   | $\sigma_F$   |
|--------------------------------|--------|--------------|
| Tuve, Carnegie Inst. DC       | 1939   | 0.4 b (0.6 MeV) |
| Frisch & Peierls             | 1940   | 10 b (a bad guess) |
| MAUD, Chadwick, Frisch, Liverpool | 1940   | 2.1 b (0.35 MeV) |
| MAUD, Frisch, Birmingham    | 1940   | 2.3 b (0.2-1 MeV) |
| Betscher, Cambridge         | 1940   | < 2.8 b (1.8 MeV) |
| MAUD "most likely"          | 1941   | 2 b |
| MAUD "pessimistic"          | 1941   | 1.5 b |
| Compton, NAS                | 1941   | 3 b |
| Chamberlain, Kennedy, Segré, Berkeley | 1942   | 2.8 b (220 keV) |
| Heydenburg, Meyer           | 1942   | 1.1 b (0.64 MeV and lower) |
| Hanson, Wisconsin           | 3/43   | 1.66 b (0.5-1.8 MeV) |
| Benedict, Hanson            | 3/43   | 1.22 b (0.53 MeV) |
| Oppenheimer, LA-2           | 3/43   | 1.6 b |
| Bethe, LA-32                | 10/43  | 1.6 b |
| Bretscher, Cambridge        | 1/44   | 1.31 b (2.05 MeV) |
| Chadwick, Kinsey            | 05/44  | 1.45 b (1.5-2 MeV) |
| Williams, LA-150 (1944)     | 1944   | 1.32 b |
| Koontz, Hall, LA-128        | 1944   | 1.31 b ±5% |
| D. Frisch, LA-554, LA-1009  | 1946   | 2.1 b (35 keV) |
| Wilson, LA-1009             | 1947   | 1.33 b±5% (1 MeV) |
| Carlson, NBS                | 1985   | 1.24 b±2% |
| ENDF/B-VIII.0               | 2018   | 1.24 b±1.3% |
sure. He noted to me that increased background means more fission detector counts thus a higher cross section. They also were not sure the neutron sources they used were truly monoenergetic. They lucked out on that.\textsuperscript{1,0}

IV.D. Early Work in the USA: $^{239}$Pu

The first measurements of plutonium fission were made for thermalized neutrons at Berkeley in 1941 (Ref.,\textsuperscript{13} pp. 355-6.), and reported in a Berkeley report A-33 by Seaborg, Segre, Kennedy and Lawrence. At thermal energies, Chamberlain \textit{et al.}\textsuperscript{81} found that the fission cross section of $^{239}$Pu was 1.87 times greater than $^{235}$U, an amount that was found later by DeWire at Los Alamos to be an overestimate owing to the neutrons not being completely thermalized (see below, and Table 8 – the correct value is 1.28). A January 1943 letter from Chamberlain, Kennedy, Segre and Wahl to Manley (NSRC A84-019-49-9) reported slow neutron values for this ratio of 1.238 and 1.294, much improved. Later, Lawrence would comment that the fast fission plutonium cross sections is ten times that of (238) uranium (Ref.,\textsuperscript{13} p. 368.), while a measurement by Seaborg and Segre in 1941 found a factor of 3.4 (Ref.,\textsuperscript{27} p. 23.) While these might seem to be contradictory claims, both could be correct depending upon the exact neutron energy, owing to the fast-changing $^{238}$U fission cross section as it rises from its threshold; today we assess this ratio to be 10 at 1.4 MeV, but 3.7 at 2 MeV.

By 1942, Oppenheimer and Manley urged the measurement of fast neutron fission of plutonium. In October, Seaborg wrote to the Carnegie Institution of Washington to say he was sending them a 10 \textmu g sample of plutonium. Heydenburg and Meyer wrote\textsuperscript{82} to Manley in Los Alamos, on April 9 1943, on “Comparative Fission Cross Section for Element 49 and Normal Uranium”, communicating their late 1942/1943 measurements of the $^{239}$Pu to $^{235}$U fission cross section ratio. They obtained ratios 1.64 at thermal energies, 1.58 at 650 keV, and 1.68 at 3.95 MeV, and reported an absolute $^{239}$Pu(n,f) cross section of 2.18 b at 650 keV. As seen in Tables 6 and 7 these measurements are impressively accurate when compared to modern ENDF/B-VIII.0 values. Later, Taschek\textsuperscript{83} in LA-28 quotes a Heydenburg\textsuperscript{84} (CF-626) Pu9/u5 fission ratio of 1.76 at 650 keV, just slightly different. In the first Los Alamos conference documented in LA-2 Manley\textsuperscript{34} discussed the data measured at two fast energies with results, “about two times 25” at 0.4 and 6 MeV. This appears to be a reference to the aforementioned Heydenburg measurements, though the energies and the ratios quoted aren’t exactly the same and the “two times” seems strangely optimistic.

IV.E. Work at Los Alamos: $^{239}$Pu

As we have seen, at the start of Project Y’s work the assessment was that the $^{239}$Pu fission cross section would be “about two times 25” in the fast neutron energy range, leading Oppenheimer, Bethe, Manley and so on to use 3 b in their initial critical mass calculations.\textsuperscript{34,35} These same values can be seen in Serber’s Primer book\textsuperscript{36} p. 15, Fig. 1 where the plutonium (“49”) curve shows 3 b for higher neutron energies. Williams, chairman of one of the conference sessions, talked of the importance of “absolute measurements of the fission cross sections of 25, 49, 01 [n], 11 [p]… Someone should gather these materials”,\textsuperscript{35} and getting good data on plutonium became the priority; these were the first measurements made at Los Alamos.

Measurements of plutonium fission posed a challenge for the experimentalists. At first, no samples were available, and then by the summer of 1943, plutonium started arriving but only in tiny \textmu g quantities from Chicago’s Met Lab. Also, measuring the ionization effects of a fission fragment above the alpha-particle pile-up from alpha decay is challenging. Nevertheless, once measurements began at Los Alamos by Segrè and collaborators from the summer of 1943 on, the fission ratio data obtained for “49/25”, \textit{i.e.} $^{239}$Pu(n,f) in ratio to $^{235}$U(n,f), were remarkably accurate. But when multiplied by the $^{235}$U fission cross sections, which at that time were measured to be too high (see previous section), the resulting plutonium fission cross sections were also too high. The experience during the Manhattan project was, therefore, one in which the plutonium fission cross section measurements decreased

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\textsuperscript{4}Carlson also notes: “Bailey also has a $^{235}$U fission cross section that he made with Williams before the work of Williams. Both are high. Bailey was doing his PhD thesis work at LA and he mentioned to me that when he defended his thesis the panel was composed of Williams, Wilson, Fermi, Bethe and Kennedy. That could be rather intimidating!”
during the project, from the initial assessments of 3 b to just under 2 b after the Fall of 1944, for fast neutrons around 1.5 MeV, see Fig. 12.

The first measurement of plutonium fission cross sections at Los Alamos came from Segrè and Wiegand, August 31, 1943 (LA-21). Accelerator neutron-sources were not yet used; at that stage they relied on neutron sources from radioactive targets. They had supplied a 17 µg sample of plutonium together with an enriched 235U sample containing 35 µg of 235, allowing fission ratio measurements. They found that the fission cross sections of 49 and 25 for 220 keV neutrons have a ratio of 1.14 (using an yttrium-plus-beryllium source). For higher energy radium-plus-beryllium neutrons around 3.6 MeV the fission cross sections of 28, 24 and 49 are respectively 0.32, 0.7 and 1.57 times that of 25. These ratio results are shown in Table 6 and are seen to be extremely accurate compared to σ values are: 

\[ \sigma_{\text{yttrium-plus-beryllium source}}. \]

For higher energy radium-plus-beryllium neutrons around 3.6 MeV the fission cross sections of 28, 24 and 49 are respectively 0.32, 0.7 and 1.57 times that of 25. These ratio results are shown in Table 6 and are seen to be extremely accurate compared to ENDF/B-VIII.0 data today. The earlier 49/25 ratio results by Heydenburg et al. were reasonably accurate too.

Two months later, in October 1943 Taschek and Williams measured\(^4\) the 49/25 fission ratio, but this time with the Van de Graaff, and a Li(p,n) source reaction with the 17 µg Pu target. This allowed them to map out the incident energy dependence of the cross section ratio, from 0.1 MeV to 1.5 MeV. Their result at 1.5 MeV incident energy was about 8% high, and when combined with the uranium 235U fission value in use at the time (1.6 b from Wisconsin, also high) they obtained a plutonium fission cross section at 1.5 MeV of 2.7 b, still very much too high.

Bethe’s Theoretical Division progress reports\(^4\) show how the new data were quickly adopted. By October 1943, although the 235U fission cross section was still taken to be 1.6 b, the value at the beginning of the project that had come from Wisconsin, Bethe had reduced the 239Pu estimated fast fission cross section to 2.6 b based on the new Segrè and the new Taschek fission ratio data, and used these updated values in his critical mass calculations. This plutonium fission cross section of 2.6 b was considerably too high, but the problem was not in the recent Los Alamos 239Pu/235U fission ratio data but rather in the too-high 235U fission of 1.6 b from Wisconsin.

As the year progressed in 1944, the accurate 239Pu/235U fission cross section measurements combined with the more accurate 235U fission results led to decreasing estimates of the plutonium fission cross section. Bacher had to report\(^8\) to Oppenheimer and Los Alamos’ Governing Board that it “appeared certain that the Pu cross section must be revised downwards by at least 15%, because of incorrect lifetimes assumed”. He said “The best present values are: \(\sigma_f\approx 25\) at 1 MeV = 1.4 b; \(\sigma_f\approx 49\) at 1 MeV = 2.2 b; \(\bar{\nu}_f\approx 25 = 2.4, \bar{\nu}_f\approx 49=2.8\).” In fact, as we will see, the plutonium fission value of 2.2 b would still prove to be an overestimate, being about 13% higher than best values later found of 1.94 b, see Table 7.

Spring-summer of 1944 was the time when Segrè’s data on reactor-made plutonium (with a higher 239Pu content) was showing spontaneous fission rates five times higher than they expected, a shocking discovery that is well-told in the introductory chapter of Critical Assembly.\(^5\) This forced the gun Pu bomb design (“Thin Man”) to be abandoned owing to risks of pre-initiation and led to the lab pivoting to focus on plutonium implosion. Bacher moved from Physics Division Leader to being put in charge of the new Gadget Division to build the plutonium implosion gadget, and Robert Wilson took over from Bacher as leader of the Research Physics Division. The fundamental nuclear physics data for plutonium fission was all-important, and the intense pressure was captured well in Critical Assembly, which quoted (p. 196) Wilson: “As Wilson remembers, P-Division realized “there were mistakes in the measurements,” some of which were “very large.” The spontaneous fission measurements were the principal worry, but Wilson was also upset by the status of fission cross-section and \(\bar{\nu}\) measurements, which were, in his opinion,”a disgrace.” Wilson reacted with a tremendous furor. So many important measurements seemed to be “based on shifting sand .... I mean, how could we call ourselves physicists?”

Yet in the late summer of 1944, as for 235U, Williams’s

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| Authors                | Date | \(\sigma_f\) | Reference |
|------------------------|------|-------------|-----------|
| Heydenburg, Meyer      | 1943 | 2.18 b      | Carnegie Inst. |
| Oppenheimer, Wiegland | 3/43 | 3 b         | LA-21     |
| Wiegland, Segrè        | 8/43 | (2.5 b)     | LA-21     |
| Taschek, Williams      | 10/43| (2.7 b)     | LA-28     |
| Bethe                  | 10/44| 2.6 b       | LA-32     |
| Bacher                 | 8/44 | (2.2 b)     |           |
| Williams               | 10/44| 1.95 b      | LA-150    |
| Handbook               | 1944 | 1.95 b      | LA-140    |
| Wilson                 | 1947 | 1.94 b      | LA-1009   |
| Toveson                | 2010 | 1.92 b±0.7% | ENDF/B-VIII.0 |

**Table 7: Fast 239Pu fission cross sections near 1.5 MeV neutron energy, from measurements and evaluations (italicized).** The exact energy or ranges of energies are given in parentheses. The “LA-” references denote work done at Los Alamos. Values in parenthesis indicate that they were ratio measurements to 235U fission (that were quite accurate, see Table 6) but were multiplied by the by the 235U fission cross section of that era (e.g. 1.6 b in 1943, which we now know was too high).
group made substantial advances\textsuperscript{63} that allowed them to make excellent plutonium fission cross section measurements on the Van de Graaff, using a quasi monoenergetic Li(p,n) source. Advances in electroplating target production of larger plutonium samples, faster electronics, and use of “comparison chambers” to measure fission cross section ratios between $^{239}\text{Pu}$ and $^{235}\text{U}$ led to the improvements. Their uncertainty estimates were 3\% statistical and 3\% systematic, which was dominated by the mass of the foil. The result they obtained at fast neutron energies, for example 1.95 b at 1.5 MeV, are in excellent agreement (to 1\%) with our best assessment today, see Table 7 and Fig. 12.

Later measurements of Williams in LAMS-135 (Ref.\textsuperscript{27} p. 342), 1 September 1944, confirmed these results. As for $^{235}\text{U}$, over time the better plutonium cross section measurements led to smaller cross sections in the fast region, as can be seen in Table 7 and Fig. 12 for the fission cross section at 1.5 MeV. There, a “modern” 2010 measurement is also shown for comparison, by Tovesson, made at Los Alamos’ LANSCE facility. This measurement was in ratio to $^{235}\text{U}(n,f)$ and also normalized to thermal. Other accurate plutonium measurements have been made by Lisowski and by Shcherbakov.

IV.E.1. Thermal fission

This paper will not describe thermal neutron measurements in detail. But we note that the earliest thermal fission measurements from Berkeley (Chamberlain, Kennedy, Segrè, Wahl), Carnegie (Heydenburg), and then Los Alamos (Wiegland and Segrè), gave results that were compromised because the neutrons were not completely thermalized. This was appreciated by J. DeWire, R. Wilson and W. Woodward\textsuperscript{87} in June 1944 who discussed the different energy-dependence of $^{235}\text{U}$ versus $^{239}\text{Pu}$ fission cross sections in the thermal energy region. DeWire more completely thermalized the neutrons to obtain a fission cross section ratio for $^{239}\text{Pu}(n,f)/^{235}\text{U}(n,f)$ of 1.28 at 0.025 eV (2200 m/s), a value that is in perfect agreement with our modern assessments, see Table 8. When combined with their estimated $^{235}\text{U}$ thermal cross section they obtained a thermal $^{239}\text{Pu}$ fission cross section of 705 b,\textsuperscript{59} a value that is only 6\% below our best value today, 752 b. The thermal $^{235}\text{U}$ cross section was evaluated to be 542 b in the 1944 Los Alamos Nuclear Physics Handbook,\textsuperscript{39} a value 8\% below our best value today (587 b).

V. FISSION $\bar{\nu}$ (NEUTRON MULTIPLICITY)

Before Project Y began in Los Alamos, a number of measurements on the average multiplicity of neutrons emitted in thermal $^{235}\text{U}$ fission had already been made, beginning with Halban’s seminal 1939 measurement in Paris showing that a self-sustaining chain reaction could be produced. The early 1939-1940 values measured by Halban ($\bar{\nu}$=3.5±0.7), by Anderson and Fermi ($\bar{\nu}$=2.2), by Zinn and Szilard ($\bar{\nu}$=2.3), and by Turner ($\bar{\nu}$=3.05), influenced the values adopted in the early calculations of $^{235}\text{U}$ critical mass by Frisch and Peierls (who assumed $\bar{\nu}$=2.3), and by the MAUD Committee ($\bar{\nu}$=2.5-3.0) and Compton’s NAS committee’s ($\bar{\nu}$=3.0).\textsuperscript{12}

At Chicago, Fermi’s team had measured the neutrons produced from $^{235}\text{U}$ per thermal neutron absorbed. This was combined with measurements of the ratio of fission to

\begin{table}[h]
\centering
\caption{Thermal plutonium fission ratio $^{239}\text{Pu}/^{235}\text{U}$ measurements in the neutron energy region, compared with ENDF/B-VIII.0. The “LA-” references denote work done at Los Alamos.}
\begin{tabular}{llll}
\hline
Authors, Energy & Date & Ratio & Today \textsuperscript{ENDF} \\
\hline
Heydenburg,\textsuperscript{62} \textsuperscript{Thermal} & 4/43 & 1.64 & 1.28 \\
Chamberlain, Segrè\textsuperscript{61} \textsuperscript{CN-469, thermal} & 1942 & 1.87±0.14 & 1.28 \\
Chamberlain, Segrè \textsuperscript{letter to Manley} & 1/43 & 1.24,1.29 & 1.28 \\
Wiegland, Segrè\textsuperscript{01} \textsuperscript{LA-21, thermal} & 8/43 & 1.74±0.2 & 1.28 \\
DeWire\textsuperscript{67} \textsuperscript{LA-103, thermal} & 6/44 & 1.28 & 1.28 \\
\hline
\end{tabular}
\end{table}

Figure 12: The $^{239}\text{Pu}$ fission cross section measurements changing with time, for a neutron energy near 1.5 MeV. Over time the measured values decreased; the horizontal line shows the best value today. Measurements are shown as solid symbols whereas evaluations or estimates are crosses. The numerical values are listed in Table 7.

\begin{itemize}
\item \textbf{Table 8: Thermal} plutonium fission ratio $^{239}\text{Pu}/^{235}\text{U}$ measurements in the neutron energy region, compared with ENDF/B-VIII.0. The “LA-” references denote work done at Los Alamos.
\end{itemize}
capture in $^{235}\text{U}$ (which was not known reliably at thermal) to infer $\bar{\nu}^{27}$ (Par. 1.60); (Fermi, report CP-257).

The Manhattan Project physicists made some assumptions about $\bar{\nu}$, based on general physics principles, that turned out to be correct. They assumed that the dependence with incident neutron energy ought to be weak, implying that measurements at thermal (where the fission cross section is higher, giving better counting statistics in experiments) provide a good approximation to the $\bar{\nu}$ values needed for fast neutrons. They also expected $^{239}\text{Pu}$’s $\bar{\nu}$ to be at least as large as that of $^{235}\text{U}$.

At the first conference at Los Alamos, Oppenheimer reported a value of $\bar{\nu}=2.2\pm0.2$ in $^{235}\text{U}$ for fast neutrons, from Fermi (CP 257, p.3), see Fig. 8. Nothing was known experimentally for $^{239}\text{Pu}$; as mentioned above, on general physics principles it was assumed that it would be similar to $^{235}\text{U}$’s value, and one of the project’s highest priorities was to obtain a measured value for plutonium. Indeed, the very first nuclear physics measurement at Los Alamos was for plutonium $\bar{\nu}$. Critical Assembly (p.78) describes Robert Wilson’s Physics Division view at the time that “if you’re going to spend a billion dollars” to build an atomic bomb, you have to be sure that $\bar{\nu}$ is large enough to sustain a chain reaction, and that the fraction of fission neutrons that are delayed, versus prompt, needs to be small.

Bacher, the first Physics Division Leader (Fig. 4), crafted a research program that pursued complementary and parallel studies, to both mitigate risks and provide a variety of insights. This involved supporting continued thermal measurements at Chicago by Fermi’s team, and new measurements at Los Alamos focused on both thermal and fast neutron energies. Bacher wanted to be able to tie together Fermi’s measurements at thermal with Los Alamos measurements also for thermalized neutrons, and additionally to determine the neutron energy dependence of $\bar{\nu}$ from thermal up to fast neutron energies (Ref. 27 p.184). Diven, Manley and Taschek describe how hard the fast neutron measurements were, given the small fission cross section at fast versus thermal energies. This is because most neutrons in the experiment go through the target without interacting and provide a large “noise” background to the measured signal.

In the summer of 1943 at Los Alamos, Williams used the the Van de Graaff with Li(p,n) source reactions slowed down by paraffin and by water to make the first plutonium $\bar{\nu}$ measurement. Not only did the slow neutron source reaction result in a higher fission cross section and better statistics, but it also ensured that the source neutrons were separable from the higher-energy fission neutrons of interest. A tiny $142\ \mu$g plutonium sample from Chicago’s Met Lab was prepared into a foil by Art Wahl, and the ratio of prompt $\bar{\nu}$ for plutonium, versus $^{235}\text{U}$, was measured to be $1.20\pm0.09$ and plutonium $\bar{\nu}$ itself determined to be $2.64\pm0.2$ (Ref. 27 p.79). The fact that plutonium’s $\bar{\nu}$ was larger than uranium’s was very good news, confirming their expectation. Richard’s memoirs relate this very first experiment at Los Alamos, and stated that the few-$\mu$g speck of plutonium came from the Washington University (St. Louis) cyclotron (perhaps a precursor to the aforementioned Met Lab sample?), and the experiment’s success earned the group a camping trip into the Pecos Wilderness. This result was the last good news on $\bar{\nu}$ for quite a while, as a series of contradictory results for the $^{235}\text{U}$ $\bar{\nu}$ were subsequently found.

The first of the disappointing results was in the Fall of 1943 when Fermi reported a new measurement that reduced $^{235}\text{U}$ $\bar{\nu}$ to 2.0 from his previous value of 2.2. Later, this was found to be a change in the wrong direction (the best prompt value today is 2.41 at thermal and 2.57 at the fast neutron energy of 1.5 MeV). Fermi’s reputation was such that his $\bar{\nu}=2.0$ value was immediately adopted for $^{235}\text{U}$ and $^{239}\text{Pu}$, as seen for example in the updated criticality calculations shown in Bethe’s Theoretical Division monthly report for October 1943, LA-32, see Table 1. Oppenheimer wrote to Groves stating that “even this small change means an increase by 40% in the amount of material required” (Ref. 31 p.191).

Robert Wilson captured the crisis of confidence with his statement in the introduction to Nuclear Physics, LA-1009, “We were constantly plagued by worry about some unpredicted or overlooked mechanism of nuclear physics which might make our program unsound”. Oppenheimer explained to Groves “even the most careful experiments in the field may have unexpected sources of error”. He pointed out the value of pursuing independent approaches so that the $\bar{\nu}$ values obtained could be validated by all promising means.

At Oppenheimer’s request, Segrè traveled to Chicago to make thermal measurements, taking advantage of the reactor’s high neutron fluence. In early 1944, he found a result for $^{235}\text{U}$ thermal $\bar{\nu}$ of 2.15, while Fermi obtained a new value of 2.18 (Ref. 27 p.195). These were changes in the right direction, but more accurate values had to await improved experimental methods being developed back at Los Alamos, together with larger amounts of “Clinton” plutonium that would come from Oak Ridge’s Clinton Engineer Work’s reactor.

In June 1944, accurate measurements of thermal prompt $\bar{\nu}$ were made by Snyder and Williams using thermalized neutrons from the cyclotron, with an ion chamber for counting fissions. Their result for the $\bar{\nu}$ ratio of $^{239}\text{Pu}/^{235}\text{U}$, was $1.17\pm0.02$, in excellent agreement with our best value today from the ENDF/B-VIII.0 standards,
1.19. Compared to the first Segrè measurement in 1943 that had been done with only 142 µg, this experiment used 562 mg of Pu. Results were also obtained for an “integral” broad energy Ra-Be neutron source, with a known source strength, resulting in absolute measurements\textsuperscript{76} of thermal $\bar{\nu}$-25=2.44, and thermal $\bar{\nu}$-49=2.86, with estimated uncertainties of 0.05, i.e. about 2% (LA-140). These agree to 1% with our best ENDF/B-VIII.0 values today, of 2.41 and 2.87 respectively at thermal. The increasingly-accurate measurements of the $\bar{\nu}$ ratio for $^{235}$Pu/$^{239}$U during the Manhattan Project is also usefully described by Hutchinson in this issue.\textsuperscript{22}

Robert Wilson\textsuperscript{65} reported similar excellent results on July 4, 1944 (no time off for celebrations\textsuperscript{7}). They found a similar result for the prompt $\bar{\nu}$ $^{239}$Pu/$^{235}$U ratio of 1.18±0.01 (compared with 1.19 today). They were also able to show that the prompt neutrons did not include any “slightly delayed” neutrons that were delayed by >5.E-9 seconds; an important result for consideration of a fast chain reacting system.

By late in 1944, the (small) energy dependence of $\bar{\nu}$, from thermal to fast energies, had also been established (Ref.\textsuperscript{27} p.342). William’s group found 0.98±0.04 for $^{235}$U, and 1.01±0.04 for $^{239}$Pu, for the ratio $\bar{\nu}$(250 keV)/$\bar{\nu}$(thermal).\textsuperscript{91} These experiments, and similar ones in Manley’s and Wilson’s groups\textsuperscript{92} confirmed the initial expectation that $\bar{\nu}$ would be similar for fast and thermal energies, and agree with our modern assessments: ENDF/B-VIII.0 today shows a ratio of 1.02 and 1.01 for $^{235}$U and $^{239}$Pu, respectively, for 250 keV v. thermal energies$^1$.

Combining Snyder and Williams’ $\bar{\nu}$ data with Wilson’s data, the September 1944 LA-140 Nuclear Physics Handbook assessed\textsuperscript{76} thermal evaluated values of $\bar{\nu}$-25=2.47 and $\bar{\nu}$-49=2.91, which agree with our modern values to 2% and 1% respectively. By 1945, the LA-140A Nuclear Physics Handbook had updated the fast $^{235}$U $\bar{\nu}$ to 2.50. The values of $\bar{\nu}$ data determined throughout the 1940s are shown in Fig. 13.

### VI. PROMPT FISSION NEUTRON SPECTRA, PFNS

The topic of the energy dependence of prompt neutrons emitted in fission has the distinction of being documented in the first manuscript LAMS-1, an early 1943 paper by W.E. Bennett and H.T. Richards.\textsuperscript{93} The prompt fission neutron spectrum (PFNS) was needed during the Manhattan Project for a variety of reasons. It set a substantial part of the neutron energy distribution in fast metal regions of critical systems, defining the average neutron energy for
which cross sections and \( \bar{\nu} \) needed to be known (around 1–2 MeV) and influencing calculated criticality. For example, Chadwick\(^{94} \) quantified the calculational uncertainty on the criticality k-eff owing to the uncertainty of the average energy of the PFNS; for Godiva, a fast highly enriched uranium \(^{235}\)U assembly, an uncertainty of 300 keV in the average PFNS energy (roughly the accuracy of understanding in 1943) corresponds to about 1% relative uncertainty in calculated k-eff, which is about 2 kg out of 46 kg in the critical mass of an idealized \(^{235}\)U sphere. Compared to the other nuclear data uncertainties in Table 1 circa 1944-1945 this is a relatively small effect. But there is another phenomenon in dynamical systems where the PFNS is important: it determines the neutron velocities and therefore the time between subsequent chain reaction fissions, see Lestone’s paper\(^{95} \) in this issue.

Today, neutronics simulations access ENDF data on the PFNS spectra that are also dependent upon the incident neutron energy; the “Chi matrix”. The PFNS are different for \(^{235}\)U, \(^{238}\)U, and \(^{239}\)Pu, with plutonium being “hotter” than uranium. However, in the early days of nuclear science it was correctly appreciated that the PFNS dependence on incident energy, and actinide target type, would be small. This proved to be the case, and so in this section (as was also the case at Los Alamos 1943-1945) I will mostly not concern myself with incident-energy differences, and will show PFNS plots that combine data from various experiments, sometimes with different incident energies. It was common to intentionally thermalize accelerator-source neutrons to benefit from higher statistics and lower backgrounds owing to the large fission cross section at low energies.

Early 1939 data on the PFNS from thermal fission neutrons had been published by W.H. Zinn and L. Szilard (PR 56, 619 (1939)) and H. von Halban, F. Joliot, and L. Kowarski (Nature 143, 939 (1939)). These very early measurements were suggestive but could not characterize the spectrum in any detail. The earliest theoretical treatment of the fission process, beyond brief references to the phenomena in Bohr and Wheeler and Zinn and Szilard, was by Norman Feather in Cambridge, February 1942.\(^{96} \) Feather correctly understood the emission mechanism to be the isotropic evaporation of neutrons from excited fission fragments in their center-of-mass frame, followed by the kinematical boosting of these neutrons based on the fragment’s motion, and he developed a mathematical model to describe them, see Fig. 14.

In the early 1940s, programs to measure the PFNS were established in Britain and the USA. In Britain, the PFNS was measured at Liverpool by Rotblat, Pickavance, Rowlands, Hall and Chadwick (B report 86), with a photographic emulsion plate method. At Rice, W.E Bennett and H.T. Richards (CF Report 325; H.T. Richards PR59, 796 (1941)) made PFNS measurements by observing recoils in the cloud chamber. Subsequently, Bennett and Richards would move to Minnesota with Williams to measure the PFNS with photographic plates using the Van de Graaff; they thought that this emulsion technique would allow neutrons to be measured over a wide range of energies, keeping scattering material to a minimum.\(^{97} \) At Los Alamos, many measurements would be made to determine the neutron spectrum through proton recoil studies in ionization chambers, for thermal and fast incident neutrons, using approaches that followed Bloch and Staub’s work\(^{98} \) (report CF-525) at the Stanford cyclotron (which proved to be very accurate, see below). At Chicago, measurements were made with the pile thermal neutrons, with the cyclotron source, and with neutrons from spontaneous decay sources.\(^{27} \)

As was often the case, it was Bethe who first quantified the likely average energy of the neutrons, and understood their basic spectrum shape from theoretical insights. In LA-2 he assessed that they should have an average energy of about 1.7 MeV, see Fig. 15, influenced by the 1943 Stanford measurements (see below). This is close to the best value today of 2.0 MeV for thermal neutrons on \(^{235}\)U (2.04 MeV for fast 1.5 MeV incident neutrons). Bethe correctly appreciated that nuclear theory insights could provide useful quantitative guidance, perhaps as good or better than many experiments of that time period owing to their substantial background scattering problems (see below). This is no longer the case, and nowadays our best theoretical treatments tend to be calibrated to high-precision measured data. In the last decade, flagship experiments\(^{97–99} \) at Los Alamos, by LANL, LLNL and CEA scientists have focused on precise PFNS measurements.

It is interesting to try to follow Bethe’s train of thought in the text in Fig. 15, given that this represents the first quantitative theoretical perspective on the PFNS’s average neutron energy, \( E_{av} \). First he considers the moving fragments’ velocity and notes that this corresponds to a moving velocity of 0.8 MeV/nucleon (which will be used to do a kinematical boost into the lab frame). Our assessments today are that the average kinetic energy for the fragments is 169.1 MeV at thermal, 169.4 MeV at fast energies, and so when divided by the number of nucleons (236 for \(^{235}\)U+n) we obtain 0.72 MeV/nucleon, close to Bethe’s 0.8 MeV/nucleon. Next he states that the average lab-frame energy of the PFNS neutrons will be this 0.8 MeV plus the mean (center of mass, CM) energy for

\(^{96} \) Richards moved to Los Alamos for Project Y and subsequently was on the faculty at the University of Wisconsin until 1988. He had 49 graduate students, several continuing in the field of neutron-nuclear physics.
formed with a high energy of excitation. It has been assumed that
the general features of the emission are capable of representation
on the "evaporation" hypothesis of Bohr (c.f. Weisskopf(1)) but,
besides a brief reference to the effect in papers of Bohr and
Wheeler(2) and Ziegele(3), no treatment of the influence of the motion of the
fission fragments on the energy spectrum of the emitted neutrons
appears to have been undertaken. Since these fragments are brought

Figure 14: An image extract from Norman Feather's report BM-148, Emission of Neutrons from Moving Fission Fragments, February 1942.
evaporation, which is \( E_{av}=3/2kT_{CM} \), \( T_{CM} \) being the temperature for evaporation (see Eq. (3) in Ref. [10]). Simply adding the two quantities is correct for isotropic neutron emission in each fragment’s center-of-mass system. Next we must consider whether Bethe’s text implies that he is (a) using a \( T_{CM}=0.6 \) MeV from some consideration, see below, to derive an average CM fission fragment kinetic energy of 0.9 MeV, to derive \( E_{av}=1.7 \) MeV PFNS average energy; or the opposite, (b) assuming 1.7 MeV from “present measurements” of PFNS to work backwards and infer \( T_{CM}=0.6 \) MeV and imply it is reasonable. I think it is the latter since Bethe was aware of Bloch’s summary in LA-4 that showed Stanford’s recent PFNS measurement (reported on March 11, 1943 letter to Manley, NSRC-A84-19-Box49-7) with an average energy of 1.7 MeV (although there was much uncertainty at the time, and early PFNS measurements from Liverpool and Rice reported PFNS average energies that were erroneously high). We can still ask, what might Bethe thought of the value \( T_{CM}=0.6 \) MeV from his nuclear physics insights? Where might that have come from?

A Maxwellian evaporation spectrum has a functional form \( P(\epsilon) = \sqrt{\epsilon} \exp(-\epsilon/kT_{CM}) \), \( \epsilon \) being the neutron CM-frame energy, and has an average CM energy \( \epsilon_{av} = 3/2kT_{CM} \). A simple estimate of temperature can be obtained from the average post-neutron emission excitation energy \( U \) of the decaying fragment and the level density parameter \( \alpha \), \( U = \alpha T_{CM}^2 \) (Bethe, 1937, p.81) with the pre-neutron excitation energy \( U' = U + S \), \( S \) being the separation energy. If one disregards the complexities of the double-humped fission fragment distribution, with different spectra from the heavy and light peaks, one could estimate an average fission fragment level density level parameter \( \alpha = A/8 = 236/2/8 = 14.75 \) \( \text{MeV}^{-1} \). At the time Bethe thought \( \pi \) was about 2 (1 per fragment on average); one might guess a neutron separation energy of \( S = 5 \) \( \text{MeV} \) for neutron-rich fragments; after neutron emissions occurred and residual systems were bound to further emission, their remaining excitation energies would be somewhere between 0 and 5 MeV and heavily weighted towards the upper end owing to the shape of the evaporation spectrum, about 4.7 MeV on average; so one would estimate an average post-neutron emission excitation energy per fragment of \( U = \epsilon_{av} + 4.7 \) MeV, which is then equal to \( \alpha T_{CM}^2 \), so \( 4.7 + 3/2 T_{CM} = 14.75 T_{CM}^2 \), giving \( T_{CM} = 0.62 \) MeV, \( \epsilon_{av}=0.93 \) MeV, close to Bethe’s \( T_{CM} = 0.6 \) MeV, \( \epsilon_{av}=0.9 \) MeV in LA-2, Fig. 15.

So Bethe’s approach had a PFNS average energy of \( E_{av} = 0.8 + 3/2 \times 0.6 = 1.7 \) MeV, whereas my “recreation” of what he might have done gives 0.72 + 3/2 \times 0.62 = 1.65 MeV. For comparison, a modern estimate of these fission fragment values by Kawano et al.,[101] based on a statistical model informed by empirical data, gives temperatures in the 0.75-0.85 \( \text{MeV} \) range – somewhat higher than the 0.62 \( \text{MeV} \) obtained from the above simple argument. Such modern modeling results would be expected to give higher temperatures, since Bethe’s \( E_{av}=1.7 \) MeV is a little small compared to our best understanding today, 2.0 \( \text{MeV} \) for thermal incident neutrons.

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{Figure15}
\caption{An image extract from Bethe’s report in LA-2, April 1943, predicting the average energy of PFNS neutrons to be 1.7 MeV (versus the best evaluations today of 2-2.1 MeV).}
\end{figure}

In 1942, Christy and Manley[103] made a clever and essential integral measurement of the average energy of the PFNS neutrons, at Chicago, by measuring the energy of the neutrons absorbed in water following thermal \( ^{235}\text{U} \) fission, an approach that was used at Liverpool too.[93] This was particularly important because of the experimental challenges that were being faced by the many groups that were trying to measure the spectrum directly. Their result, \( 2.2 \pm 0.2 \) MeV, is very close to the correct result 2.0 MeV in ENDF/B-VIII.0, and furthermore they correctly assessed that if anything it would be an over-estimate of the true value. It led Serber to state[36] the PFNS has an average energy of 2 \( \text{MeV} \) (Primer, p.17; p.70), probably a rough average of Christy’s measurement (2.2 \( \text{MeV} \) and the Stanford measurement (1.7 MeV) quoted by Bethe and Manley, and pointed experimentalists to appreciate possible systematic errors in the PFNS measurements that they were making.

At the March 1943 conference in Los Alamos, Bethe in LA-2 described the motivation for understanding the PFNS, and the experimentalists present agreed on the unsatisfactory nature of the spectrum data taken so far at Rice and at Livermore, which suffered from large systematic errors from multiple scattering processes. Bethe said that the measurements were “unsatisfactory largely because too much fission material has been used. If enriched material were available smaller masses could be used and inelastic scattering of the fission neutrons avoided”.[49]. However,

\[ \text{Further comparisons with Kawano’s simulations: He obtains average pre-neutron emission excitation values of } U = 13.7 \text{ MeV (light)} \text{ \ and } 8.8 \text{ (heavy), so } 11.3 \text{ MeV on average, versus my } U' = 10.7 \text{ MeV; average CM temperature of } 0.85 \text{ MeV (light)} \text{ \ and } 0.78 \text{ MeV (heavy) so } 0.815 \text{ MeV on average, versus my } 0.62 \text{ MeV, and average CM neutron evaporation energy of } 1.22 \text{ MeV versus my } 0.93 \text{ MeV} \]
Figure 16: The $^{235}$U prompt fission neutron spectrum for thermal neutrons at the beginning of Project Y, as presented in LAMS-1. The black solid line is our modern value of the thermal spectrum from ENDF/B-VIII.0, which comes from an IAEA evaluation, and shows the accuracy of the 1943 Stanford measurement.

Figure 17: The $^{235}$U prompt fission neutron spectrum for neutrons, showing measurements made during Project Y. The incident neutron energy varies for these different experiments and while in principle the spectrum changes with energy, the changes are much smaller than the differences seen between these data sets. The black solid line is our modern value of the spectrum from ENDF/B-VIII.0, and the red points are recent measurements made at Los Alamos for an average incident neutron energy of 1.5 MeV.

Bethe did provide optimism in a theoretical understanding of the shape of the spectrum, which “can be predicted theoretically.” A few weeks later, in LA-4, at the April 27 workshop Bloch (p.3) described the Stanford PFNS measured for thermal neutrons on $^{235}$U, from a cyclotron source, which had an average emission energy of 1.7 ±20% MeV, and peaked at 1.1 ±20% MeV (F. Bloch and H. Staub, CF-525, and see NSRC A84-019-49-7). He noted that these energies were considerably lower than those from Rice and Liverpool, and was concerned that they suffered from inelastic scatterings that distorted the spectrum to lower energies, although we now know the Stanford measurements to be remarkably accurate (ENDF/B-VIII.0 has a PFNS average energy of 2.0 MeV for thermal neutrons on $^{235}$U), see Fig. 16. Manley was more positive about the Stanford results, saying “Data of ion chamber pulse size distributions from Stanford, which looks reasonable theoretically, show neutrons tailing off from 1 MeV” (in LA-2); subsequent experimental methods at Los Alamos would also use ion-chamber pulse distributions and obtain relatively accurate results, as discussed below.

Bennett and Richards (1943) in LAMS-1 noted a likely problem with the Liverpool Rotblat-Chadwick measurement owing to multiple scattering in the experiment; LAMS-1 attempts to correct for this to give the data in their figure II. The curve drawn has a mean energy of 2.9 MeV – “50% too large if the measurements of mean energy by absorption in a water tank are reliable.” This refers to the aforementioned Chicago integral measurement of 2.2 MeV by Christy and Manley, that was correctly trusted. Figure 16 shows the PFNS data described in LAMS-1 together with their “best estimate” (green line) of the time - an average of two of the data sets (but unfortunately not the Stanford data, which at the time they thought could be suffering from systematic inelastic scattering errors). Today’s ENDF/B-VIII.0 spectrum is shown for comparison, and is seen to be substantially softer with an average neutron energy of 2.0 MeV. The Bloch and Staub Stanford 1943 PFNS data are also shown and seen to be remarkably accurate.

During the course of the Manhattan project, the spectrum would be measured many times at Los Alamos, at Stanford, and at Minnesota. Richards’ Minnesota data were documented in LA-60 where an average energy of 1.85 MeV was found – fairly accurate compared to our best result today of 2.0 MeV, see Fig. 17. But after that, with an improved enriched $^{235}$U source, instead of finding more accurate PFNS results, the Minnesota data seem to become less accurate over time for reasons I don’t understand – Fig. 17 shows LA-84 (May, 1944) data for thermal fission on $^{235}$U and LA-200 (Jan 15, 1945) data for fast 300-650 keV neutrons on $^{235}$U, and it is evident that these orange and green data points were not very accurate. Richards’ measured change in average energy from thermal to fast incident energies (2.3 to 2.6 MeV) is much greater than our best assessments today (2.00 to 2.01 MeV in ENDF/B-VIII.0). Also, plutonium data in LA-84 differed substantially from uranium, a result not seen in subsequent measurements at Los Alamos by Staub.
and Nicodemus. Richards’ LA-200 paper provides a cautionary example of how one can face “one step forward, two steps back” in science. On the (erroneously) hot 2.6 MeV-average PFNS he obtained for $^{235}$U, he suggested that it was in fact correct by showing comparisons of an integral of the $^{238}$U(n,f)/$^{235}$U(n,f) ratio obtained with his spectrum, compared to a new direct integral measurement of this fission by Wilson. The good agreement he obtained was presumably from canceling errors.

The most accurate $^{235}$U and $^{239}$Pu PFNS data were obtained by Nicodemus and Staub at Los Alamos in 1944 using moderated neutrons from the Li(p,n) reactions at the Van de Graaff generator, and were published after the war. These $^{235}$U PFNS data are shown in Fig 17 and are seen to agree well with the earlier Stanford data and our best ENDF/B-VIII.0 evaluation today (black curve, which is based on Capote et al.’s IAEA evaluation).

For comparison, some recent 2020 high-accuracy $^{235}$U “Chi-Nu” data from Los Alamos’ LANSCE facility are shown as red points. In April 1944 William’s group also saw small differences between the $^{235}$U and $^{239}$Pu PFNS (Ref., p.196) between 1 and 2.5 MeV outgoing neutron energy. They were aware of substantial differences between their ionization chamber data and those from Minnesota’s photographic plate method. The Los Alamos results would prove to be the most accurate.

The average energy of these PFNS measurements on $^{235}$U is shown in Fig. 18 (upper panel). After the Manhattan project, in 1952 Watt at Los Alamos published his seminal paper on how to represent the PFNS with a straightforward semi-empirical analytic expression $N(E) = c \exp(-E/a) \sinh(\sqrt{(bE)})$, where $c$ is a normalization constant and $a, b$ are fit parameters. The average energy of this spectrum is $3a/2 + a^2 b/4 = 3.90$ MeV. His analysis included the use of 1952 data from Bonner (Los Alamos) and from Hill’s (Argonne) reactor experiment, obtaining $a = 1, b = 2$ with an average energy of 2.00 MeV for the $^{235}$U thermal PFNS. This is exactly the same as our best ENDF/B-VIII.0 estimate today, see Fig. 18, which comes from an IAEA standards committee evaluation, although between 1952 and 2018, the evaluated value drifted down to 1.95 MeV on the low side and up to 2.03 MeV on the high side (e.g. ENDF/B-V to VII.1) (Fig. 18 lower panel).

Plutonium spectrum data are shown in Fig 19. Both Staub and Nicodemus’ data, and those from Richards (LA-84) are reasonably accurate when compared to the best evaluated data today, ENDF-B-VIII.0 (black line). For comparison, also shown is a recent Los Alamos measurement from the LANSCE/Chi-Nu experiment (red symbols), and Lestone and Shore’s NUEX experiment from test data (orange points), which is further

Figure 18: The $^{235}$U PFNS fission spectrum’s average neutron energy measurements changing with time. (a) Upper panel: Data from 1942 – 1952. Although not strictly correct, for simplicity the figure includes values measured at different incident energies, from thermal to fast energies. Measurements are shown as solid symbols whereas evaluations or estimates are crosses; (b) Lower panel: Data evaluations from 1952 – present, for thermal incident neutrons and for 1 MeV incident energy. H-R denotes the 1961 Hansen and Roach evaluation, values from various ENDF database versions are shown from 1970-present (Credit: N. Gibson). The horizontal lines show the best thermal value today.
Figure 19: The $^{239}\text{Pu}$ prompt fission neutron spectrum for neutrons, showing measurements made during Project Y. The incident neutron energy varies for these different experiments and while in principle the spectrum changes with energy, the changes are much smaller than the differences seen between these data sets. The black solid line is our modern value of the spectrum from ENDF/B-VIII.0, and the red points are recent measurements made at Los Alamos for an average incident neutron energy of 1.5 MeV.

Figure 20: An image from the MAUD report on the transport cross section, which they called the “collision cross section”.

As discussed in Section II, the transport cross section $\sigma_T$ is a key quantity needed for computing the critical mass, Eq. 1. The 1941 MAUD report included numerous insightful discussions on the magnitude of nuclear cross sections; one of them is shown in Fig. 20 for the transport cross section, referred to there as the “collision cross section”. The “most likely” value they assessed was 5 b for fast neutrons on uranium, with a possible range 3.5-5 b, agreeing extremely well with our ENDF/B-VIII.0 value today of 4.85 b at 1.5 MeV for $^{235}\text{U}$ in ENDF/B-VIII.0.

At the beginning of the Los Alamos Project Y, Oppenheimer was using 4 b, but as we see below, Los Alamos soon moved to use values from 4.7-5 b (See Tables 1, 2).

At Los Alamos, Manley’s group, with Heinz Barschall (Fig. 21) and other strong experimentalists was set up to measure these cross sections. After the war Barschall moved to the University of Wisconsin and maintained close collaborations with neutron physics researchers at Los Alamos and Livermore laboratories. With his students, he played an important role in the development of the optical model of neutron scattering. His contributions also included advances in nuclear medicine and long-term service as editor of the Physical Review C.

The transport cross section has various definitions in the literature. In the 1940s, they understood it to be a quantity that included all reactions, that is, the neutron total cross section, but subtracted from this should be neutrons that are scattered into the forward direction (where the collision didn’t essentially change the transport process). This
led to assessments that used a transport cross section:

$$\sigma_T = \sigma_{\text{non}} + 2\pi \int \sigma_{\text{el}}(\theta) (1 - \cos(\theta)) \sin(\theta) d\theta$$

$$= \sigma_{\text{non}} + (1 - <\mu>) \sigma_{\text{el}}, \quad (4)$$

where $\sigma_{\text{non}}$ is the total nonelastic cross section (which includes the fission cross section), $\sigma_{\text{el}}$ is the total elastic cross section, $\sigma_{\text{el}}(\theta)$ is the angle-differential elastic cross section and $\mu$ is the average of the cosine scattering angle. With ENDF/B-VIII.0 today, at 1.5 MeV we find for $^{235}\text{U}$, $\sigma_{\text{non}}=3.248 \text{ b}$, $\sigma_{\text{el}}=3.557 \text{ b}$, $\mu=0.549$, so that $\sigma_T=4.852 \text{ b}$.

A cross check using the PARTISN multi-group transport code is useful, since this explicitly computes a transport cross section. Tom Saller obtains 4.83 b in a 618-group around 1.5 MeV using PARTISN, which is consistent with the point value from ENDF/B-VIII.0.

Because the transport cross section required a determination of the angular dependence of scattering, $\sigma_{\text{el}}(\theta)$, Manley’s group established a program to systematically measure such differential elastic and inelastic scattering angular distributions for a wide range of materials: actinides and potential tamper and structural elements. As well as directly measuring the scattered neutron, Barschall made a breakthrough in which he determined elastic scattering probabilities at different angles by measuring the recoiling kinetic energy of the struck target nucleus; simple kinematics allowed him to infer what neutron angle would have produced such a recoil energy.26

Figure 22 shows the evolution of assessments of the uranium transport cross section over time. Unlike the fission data, this was a case where the earliest assessments from 1941 on proved to be remarkably accurate.

VIII. CRITICALITY UNCERTAINTY REDUCTION

I close by tracing the calculated criticality uncertainty reduction that took place through Project Y’s campaign to measure nuclear constants. In the 1940s it was not a routine practice to provide uncertainty assessments on measured nuclear cross sections. In the measurements described in this paper, specific uncertainty estimates were often missing. The LA reports did, however, frequently discuss experimental problems that would lead to systematic errors, and the Project Y scientists worked hard to develop improved methods to mitigate these challenges. In none of the LA documents quoted were numerical uncertainties on calculated critical masses given, except for the aforementioned article by Tolman who, in March 1943, assessed $\approx 50$% critical mass uncertainties [$^{235}\text{U}$: $30\pm15 \text{ kg}; ^{239}\text{Pu}: 10 \text{ kg} (5–20 \text{ kg})$].

At the beginning of the project, uncertainties in the fission cross section exceeded 25–50% and by 1945 these were reduced to 5–10% (today they are 1–2%); uncertainties in the fission neutron multiplicity were initially 15% or more and were reduced to a few percent (today they are a fraction of a %); the neutron spectrum PFNS was measured fairly accurately by the end of the project with an average energy measured to about 10% (today it is known to less than 2%), and the transport cross section, initially known to 20%, was determined to 5% (and today this is known to a few %).

Based on these uncertainties in the fundamental data, the bare critical mass uncertainty can be determined, either using Eq. (1) or using our modern computational transport codes such as MCNP6. Fig. 23 shows some calculated uncertainties for a bare $^{235}\text{U}$ critical sphere using Eq. (1), with the last value shown (from 2018) instead based on MCNP6. The very large error bars shown at the beginning of the project in 1943 were reduced to about 20% by the spring of 1945 (66 kg $\pm 14$ kg). As was noted, it was fortuitous that Oppenheimer’s initial 1943 estimate of 60 kg for the mass was very similar to what Serber was calculating in the spring of 1945, using the much more accurate nuclear data measured at Los Alamos. At the beginning of Project Y, the fission cross section assessment was overestimated, whereas $\bar{\nu}$ was underestimated, and these errors canceled.
Critical mass $^{235}$U (bare) from calculation

Figure 23: The calculated $^{235}$U bare critical mass with uncertainties - based on differential cross section data - at the beginning of Project Y, $60(+210, -33)$ kg and at the end of Project Y, $66(\pm 14)$ kg as well as from the first integral experiment measurements at Los Alamos in 1945 as described by Hutchinson et al.,$^{22}$ $50.8\pm 2.5$ kg (solid circle point). The horizontal line represents our best understanding today from MCNP6 simulations using ENDF/B-VIII.0, $46.4\pm 1.7$ kg, which were in practice calibrated to the Godiva bare HEU critical mass experiment in the 1950s.
The best value today from a ENDF/B-VIII.0 and MCNP6 Monte Carlo transport calculation, 46.4±1.7 kg, is shown as a horizontal line in Figure 23. The figure also shows an accurately-calculated value of 45 kg obtained in 1952 by Bengt Carlson using the Serber–Wilson neutron diffusion method\textsuperscript{113} with three neutron energy groups (an approach that was developed by the US and the British during the Manhattan Project, 1944-1945). Note that Carlson and Wilson both had connections with the Montreal/Chalk River Manhattan Project laboratories as described in this issue by Andrews, Andrews and Mason;\textsuperscript{114} Carlson came to Los Alamos in 1945, where he spent the rest of his career. Carlson would go on to invent the SN neutronics method that has been used so widely at Los Alamos and across the world.\textsuperscript{115}

The final 1945 calculated uncertainty of 20\% in the critical mass is still quite high, and the calculated value using Eq. (1), 66 kg ±14 kg, was actually 43\% above the value we know today, 46.4 kg. That is why Los Alamos performed integral critical assembly experiments of criticality, which became possible in 1945 once substantial kg-quantities of HEU began to arrive, see Hutchinson et al.'s paper in this Issue.\textsuperscript{22} Hutchinson et al. shows how extrapolations from integral subcritical experiments on “25” in 1945 gave estimates of the 235U bare critical mass of 50.8±2.5 kg, a value only 9\% above our best understanding today, see the solid circle data point in Figure 23. Such integral measurements, which were mostly focused on tamped fast assemblies, allowed the critical masses to be determined more accurately. Likewise, today the MCNP6 calculation-based uncertainty of ±1.7 kg remains substantial (corresponding to 1\% uncertainty in calculated k-eff that comes from our ENDF/B-VIII.0 nuclear data uncertainties\textsuperscript{94,97}); our true knowledge of this critical mass is actually about an order of magnitude more accurate because the integral Godiva bare HEU assembly was measured to high accuracy (0.1\% in k-eff), a few years after the war (1951).

IX. CONCLUSIONS

Research at Los Alamos 1943-1945 dramatically advanced our understanding of nuclear science. Even though many experimental measurement techniques had been established earlier, they were vastly improved and extended at Los Alamos. Methods were developed that allowed fission cross sections to be measured to unprecedented accuracy, over a wide neutron energy range. Compared to the period after the war, the electronics that they had at the time was primitive, but many of Project Y’s best experimental physicists had become experts in electronics.\textsuperscript{96}

After the war, many of the scientists returned to universities across the USA, and some moved to laboratories at Berkeley and Oak Ridge, Argonne (1946), Brookhaven (1947), Idaho (1949), and Livermore (1952). Their subsequent nuclear physics research at these universities and laboratories was built upon the methods in experiment and theory developed during Project Y.

The initial large systematic errors on the fission cross section were reduced to below 5-10\% by the end of Project Y – many of the figures in this paper have show the measured data trending with time towards our best evaluated values today. But rather remarkably, in the case of the prompt fission neutron spectrum’s (PFNS) average energy, Serber’s 1943 beginning first guess of 2.0 MeV (with an uncertainty I estimated as 300 keV) is exactly where we have ended, 2.00(1) MeV. “What we call the beginning is often the end, and to make an end is to make a beginning.”\textsuperscript{94} The large uncertainty reduction comes from much-improved experimental and analysis methods forming the basis for the recent 2018 IAEA/ENDF/B-VIII.0 evaluation\textsuperscript{90,104-106} for thermal neutrons on 235U.

This paper showed how calculated critical masses from Project Y, using improved cross sections, became much more accurate with uncertainties reduced to about 20\% by 1945 (66 kg ±14 kg for a bare 235U sphere). Nevertheless, they were not as accurate as those obtained from the integral criticality experiments which became possible later in 1945 once larger amounts of enriched uranium and plutonium became available (giving 50.8±2.5 kg for 235U, a value only 9\% above our best understanding today). One might ask whether the differential cross section work was ultimately valuable, given the more accurate integral measurements? It was, because these cross sections then became available for use use in accurate simulation codes.\textsuperscript{113} Their use extended well beyond addressing the original critical mass question. The rapidly-improving neutronics simulation capability opened up the ability to rapidly explore a wealth of physics questions, to advance the technologies in both defense and civilian nuclear power arenas.

The Manhattan Project benefited from bringing together a remarkable cohort of first-rate scientists, who brought their best graduate students with them. This had an enduring impact on the scientific culture at the Laboratory, creating a self-confidence\textsuperscript{7} in what can be accomplished.

\textsuperscript{9}Ursula von der Leyen also used this T.S. Eliot line on the occasion of the final Brexit trade deal.

\textsuperscript{7}Allan Carlson also relates an occasion on which he heard Dick Taschek (a Project Y nuclear physicist who later became Los Alamos’ P-Division leader) giving the first talk at the Neutron Standards and Flux Normalization meeting at Argonne in 1970; “He subtly referred to the values of the 235U fission cross section by a newcomer who probably did not understand the problems involved with making very
Los Alamos National Laboratory has sustained a long-standing reputation for “all things nuclear” (to this day it is also especially known for material science and computing). An example relates to an innovative determination of the prompt fission neutron spectrum from plutonium by Lestone et al., that was shown in Fig 19. This analysis used experimental data from the nuclear test diagnostic, NUXE, which benefitted from having certain advantages compared to lab experiments (notably, higher neutron fluences with better signal to noise), but had some other complementary systematic uncertainties (e.g. for determining the spectra below 1 MeV outgoing neutron energy). When Los Alamos published these data in 2011 and 2014, their claimed accuracy was immediately accepted by the international community, even though the Laboratory was unable to openly publish all the details that went into the analysis. The NUXE result was quite important in a subsequent 2016 international assessment led by Capote with a collaboration organized through the IAEA. This was a consequence of the international regard for Los Alamos’ reputation in nuclear science, and it was only five years later that these data could be independently corroborated by high-precision lab experiments by the CEA, Los Alamos, and Livermore (The LANL-LLNL “Chi-Nu” data, also shown in Fig 19 – compare the red and orange points).

But any pride in our Laboratories’ technical accomplishments must, in the end, play second fiddle to the importance of sustaining a scientific questioning culture as articulated by our first Director, J. Robert Oppenheimer: “There must be no barriers to freedom of inquiry. There is no place for dogma in science. The scientist is free and must be free to ask any question, to doubt any assertion, to seek for any evidence, to correct any errors.”

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