The Hydrogen Standard-The first and the Primary Standard - Uniquely So

Allan D. Carlson
NIST Associate
Under Contract with BNL

Presented at

The IAEA Technical Meeting on Neutron Data Standards

October 9, 2023

The initial need for standards

- Early work with neutrons
  - After James Chadwick discovered the neutron in 1932 with the $^{9}Be(\alpha,n)$ reaction, numerous experiments were done with such sources. In the 1930s early measurements were made of nuclear reactions; For example Dunning at Columbia in 1935 used the $^{9}Be(\alpha,n)$ source inside spheres of differing diameters to look at transmission of neutrons. But **the focus was not on determining nuclear cross sections**, but instead on the fraction of scattered vs absorbed neutrons.
- The need for standards came largely from the Manhattan project since accurate data were required. **The first was hydrogen.**
  - One of the first hydrogen total cross section measurements for neutrons up to 6 MeV was made using accelerator produced neutrons by Bailey in 1943 at Los Alamos..
  - .Based on a paper by Barschall in 1940, Bailey knew the CMS angular distribution was nearly isotropic. Thus with the total cross section for normalization, he obtained the hydrogen angular distribution as a "standard." there was **no dependence on any cross section** in getting the hydrogen standard. It only depends on neutron counting ratios and measured quantities.
    - Barschall, at Princeton, proved that the energy distribution of recoils in an ionization chamber is proportional to the differential cross section for scattering in the CMS. He made measurements with hydrogen in the MeV energy range and found the energy distribution was flat.

Transmission experiment for determining hydrogen total cross setion - Bailey

The total cross section σ can be obtained from the following:

$$I/I_0 = e^{-n\sigma x},$$

Where I is the intensity of neutrons measured by the detector with the scatterer present, I_0 is the intensity of neutrons measured by the detector when the scatterer is absent, n is the density of the scatterer, and x is the length of the scatterer.

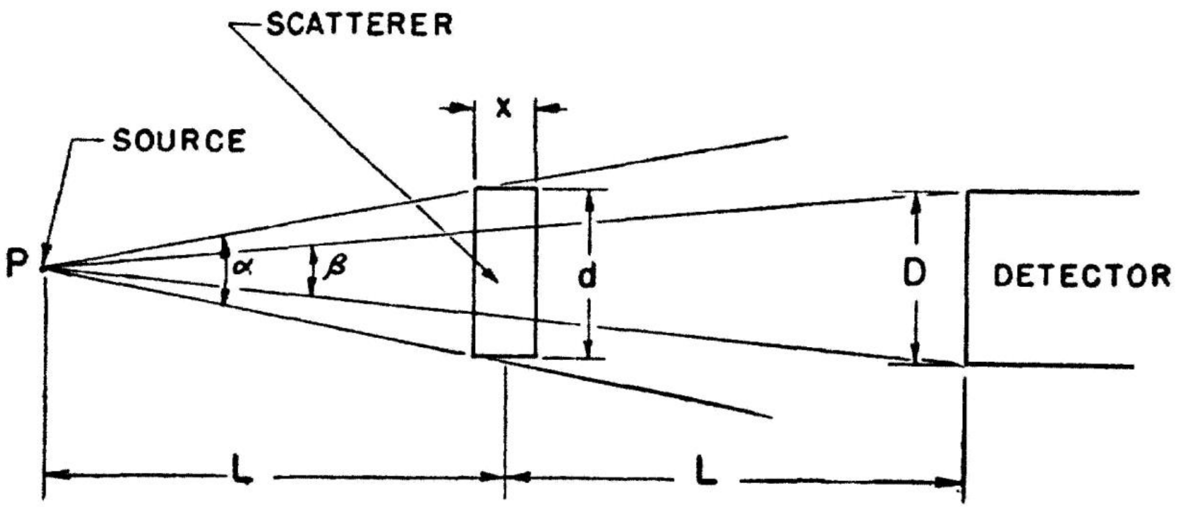


FIG. 1. Geometry of source, scatterer, and detector used in scattering experiment.

Proton recoil energy distribution measurement by Barschall for 2.53 MeV neutrons

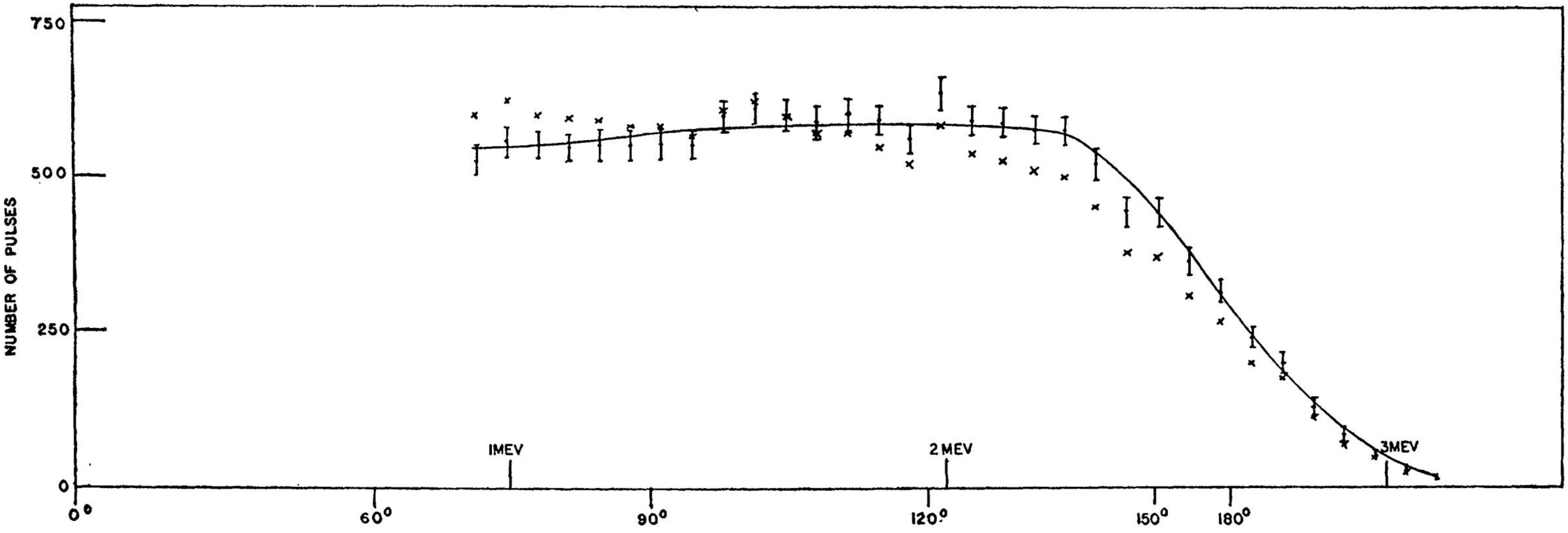
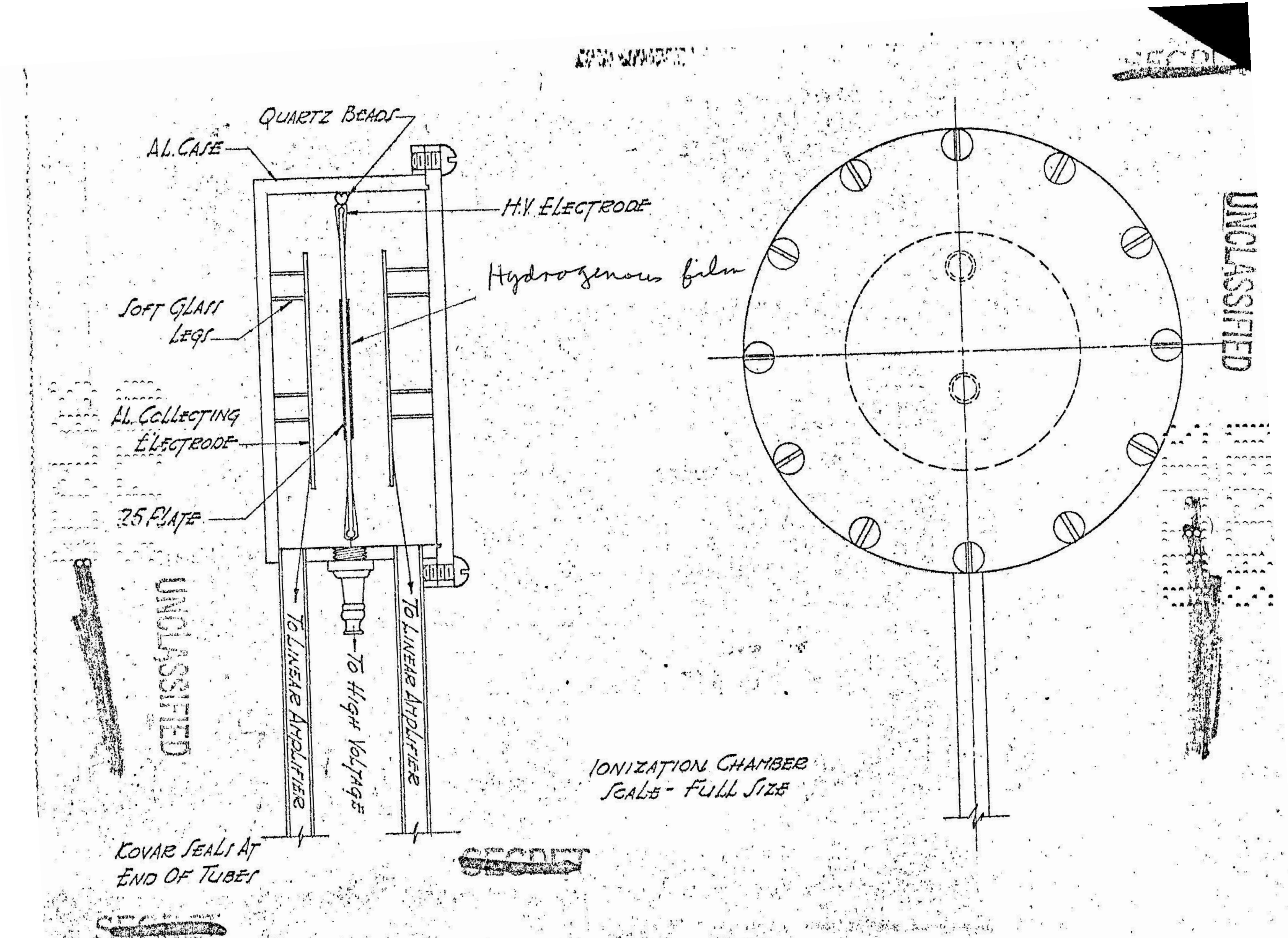


FIG. 5. Distribution in energy of recoil protons. Crosses indicate measured points, bars points corrected for wall effect.

Establishment of the $^{235}U(n,f)$ cross section as a "standard"

- Bailey made some of the first $^{235}U(n,f)$ measurements. He used a double ionization chamber with a well defined hydrogenous foil on one side and a ^{235}U deposit of known mass was on the other side. With neutrons incident on the chamber fission fragments are detected going at back angles and proton recoils were detected going in the forward direction.
  - Thus neutron fluence determination with the hydrogenous foil allowed the $^{235}U(n,f)$ cross section to be determined. But it was relative to the hydrogen cross section.
  - Using the determination of the neutron fluence with the hydrogenous chamber, they calibrated a long counter they had designed. It was then used for some neutron fluence determinations.
- Due to the simplicity of making ratio measurements to the $^{235}U(n,f)$ cross section, a number of ratio measurements to the $^{235}U(n,f)$ cross section were made during the Manhattan project. It was treated as a standard at that time.
- Note the ^{6}Li, ^{10}B, Au and ^{238}U standards (in addition to ^{235}U) do not have a convenient normalization as H does.
- Carbon scattering does have convenient normalization but it has a limited energy range and experimental problems..
- There are some special situations, e.g. associated particle measurements where absolute cross sections are obtained but they are limited.
  - Comment: The "barn" originates from WWII when Manhattan scientists considered that hitting such large atoms would be as easy as "hitting the side of a barn."

Actual drawing of the dual-ionization chamber used by Bailey



INTER-OFFICE MEMORANDUM

DATE 28 December 1945

TO: N. E. Bradbury

FROM: H. Bethe

SUBJECT: Thesis by Carl L. Bailey

B 2/10/82

I have read the thesis written by Carl L. Bailey.

It is my opinion that the thesis is highly satisfactory, and I would have no hesitation to accept it for a Ph.D. degree.

H. A. Bethe

H. Bethe

INTER-OFFICE MEMORANDUM

DATE December 20, 1945

TO: N. E. Bradbury

FROM E. Fermi

SUBJECT: Thesis by Mr. C. L. Bailey

B 2/10/81

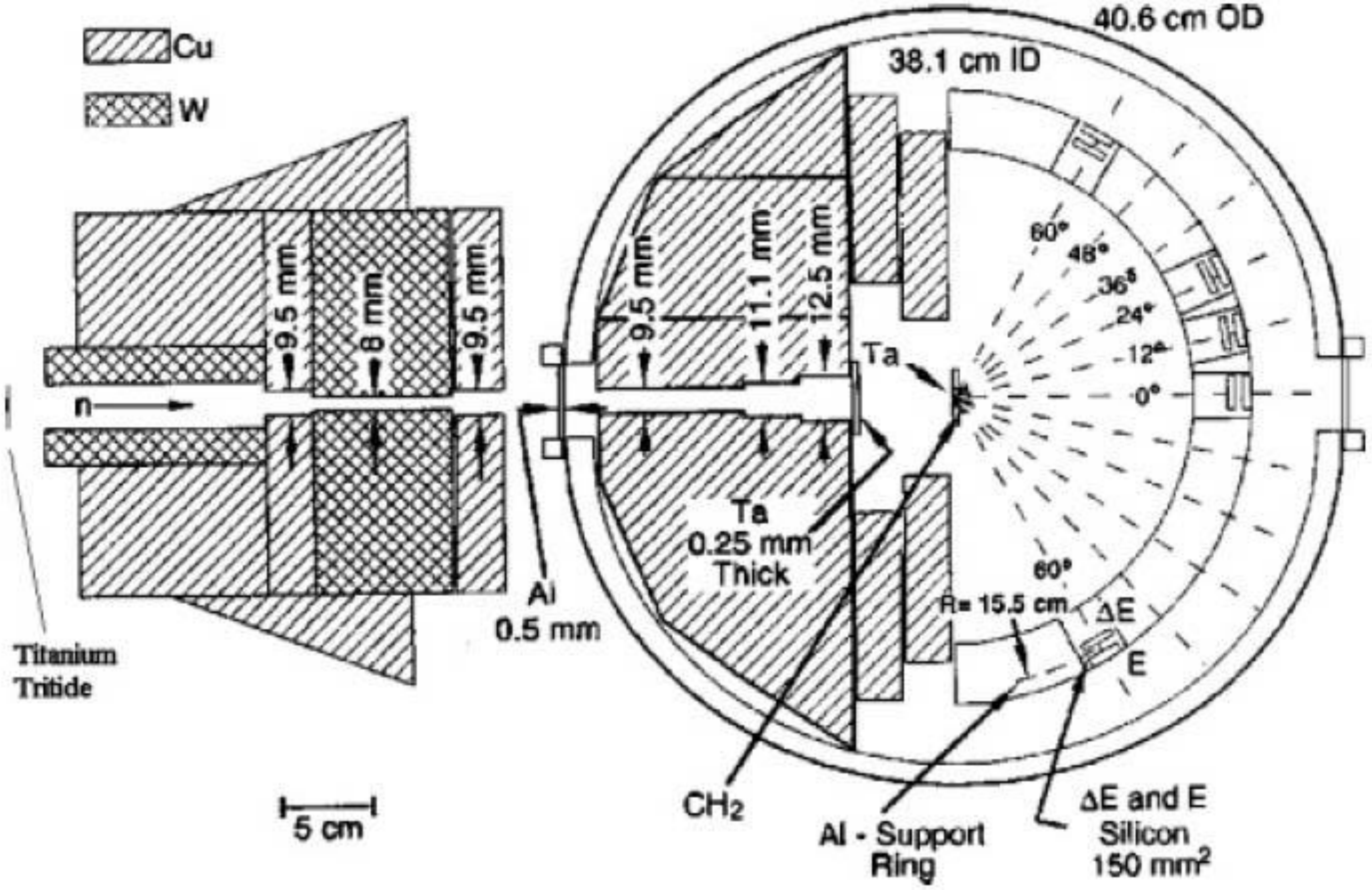
According to your request, I have examined the paper that Mr. Bailey proposes to submit to the University of Minnesota as a thesis for the Ph.D. Degree, and I have discussed the work with Mr. Bailey. In my opinion the work has been conducted with good experimental technique and could properly be accepted as a thesis for the Ph.D.

CC: Williams
Wilson
Bethe
Kennedy

E. Fermi

Establishment of the hydrogen scattering angular distribution at high energies

- The method for measuring angular distributions by Barschall is limited in enery range
  - The problem is the range of proton recoils is too large for high energy neutrons. Also edge effects and end effects are a problem.
- Use of scattering chambers leads to good measurements of angular distributions.
- Data at all angles are taken simultaneously. Thus they only depend on ratios of count rates and solid angles



Hydrogen scattering angular distribution predictions

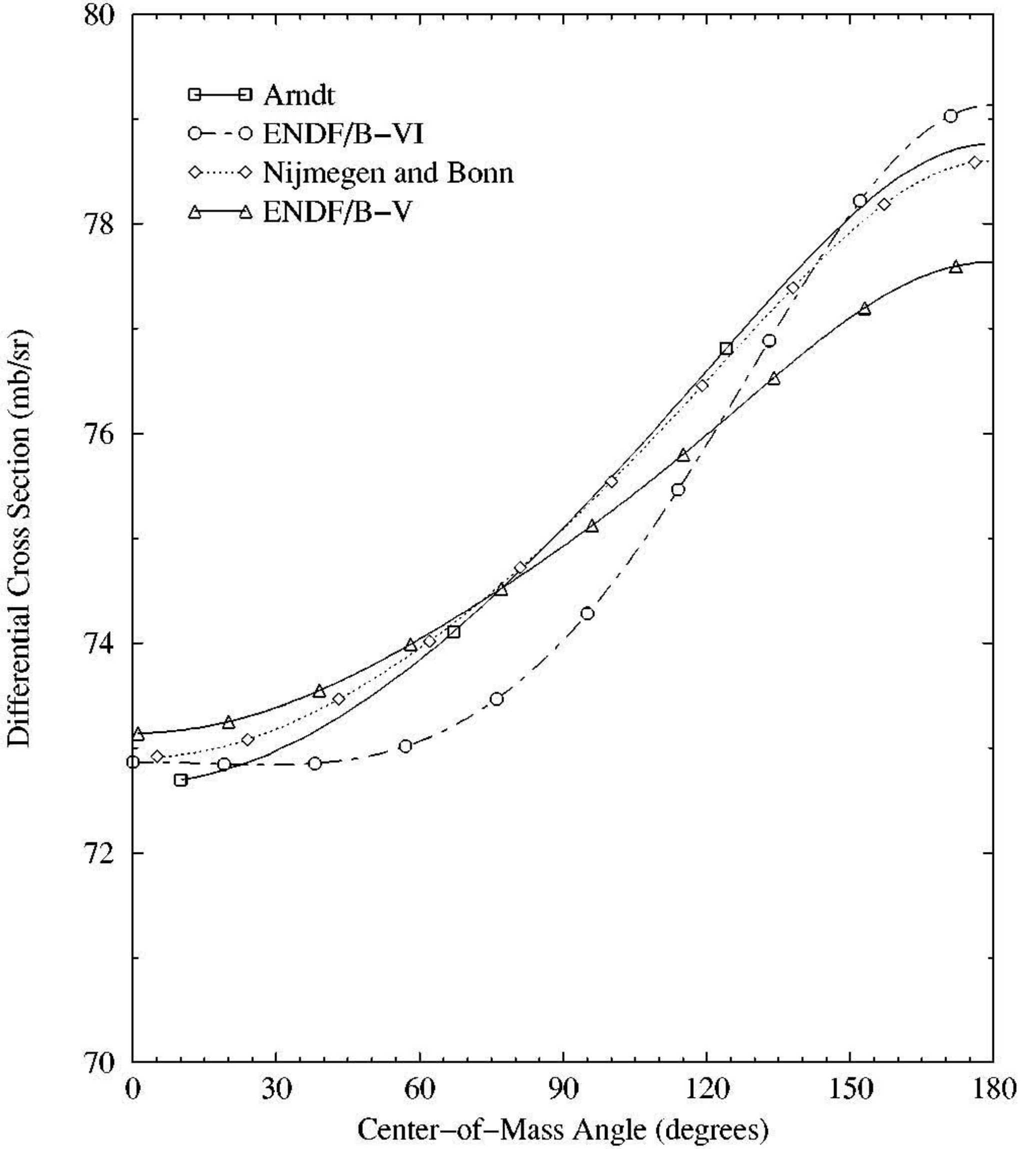
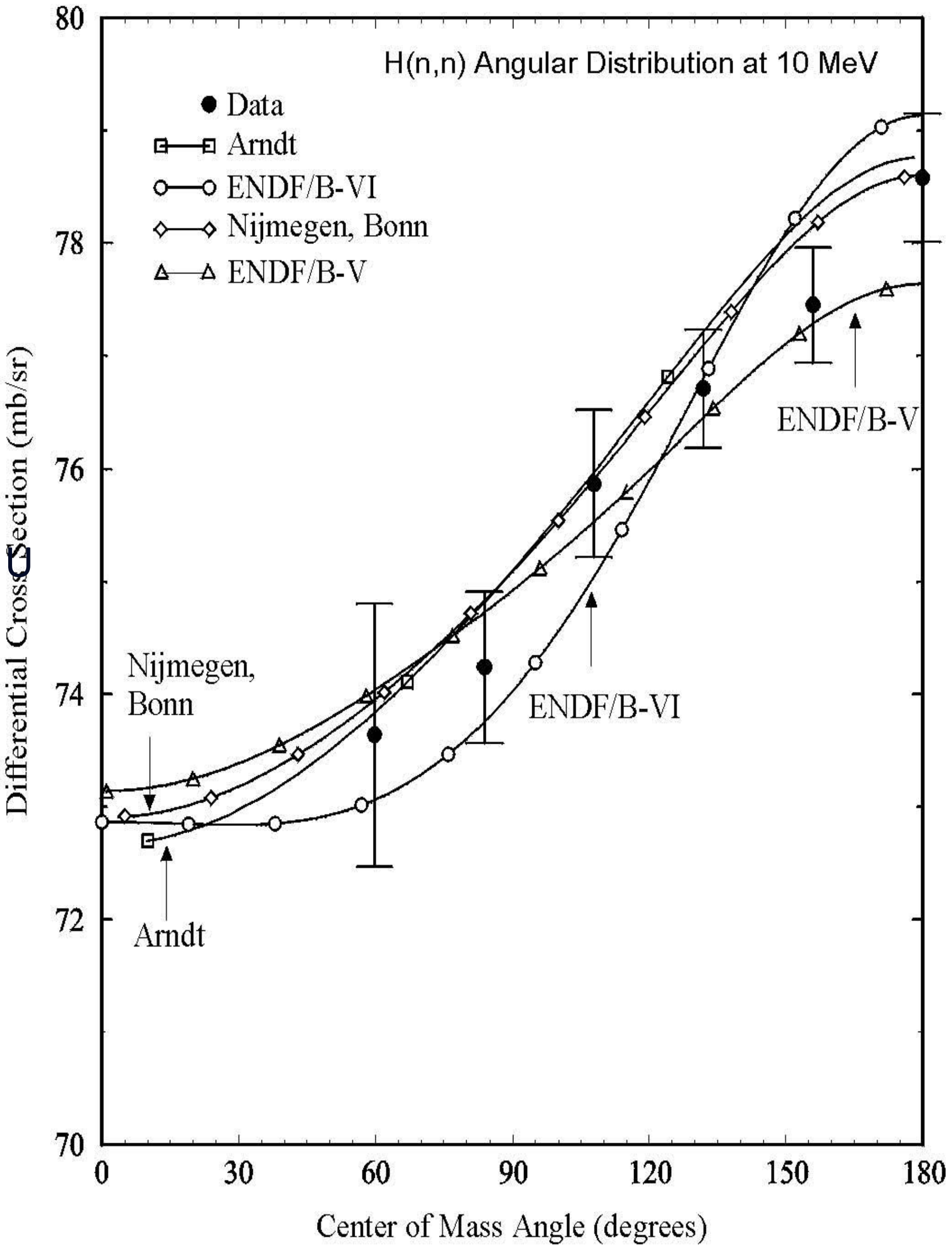


FIG. 1. Comparison of the predicted angular distribution for the n-p elastic scattering at 10 MeV (Refs. [12–15,17]).

Hydrogen scattering angular distributions

the^{235} U



High energies for the hydrogen angular distribution

- Unfortunately, measurements of sufficient quality of angular distributions are not available for all energies and angles. Thus one must use evaluated data.

- One of the first sources used for hydrogen data was the Gammel phenomenological model. It did well up to about 30 MeV for the total cross section. Angular distributions were not as well reproduced.

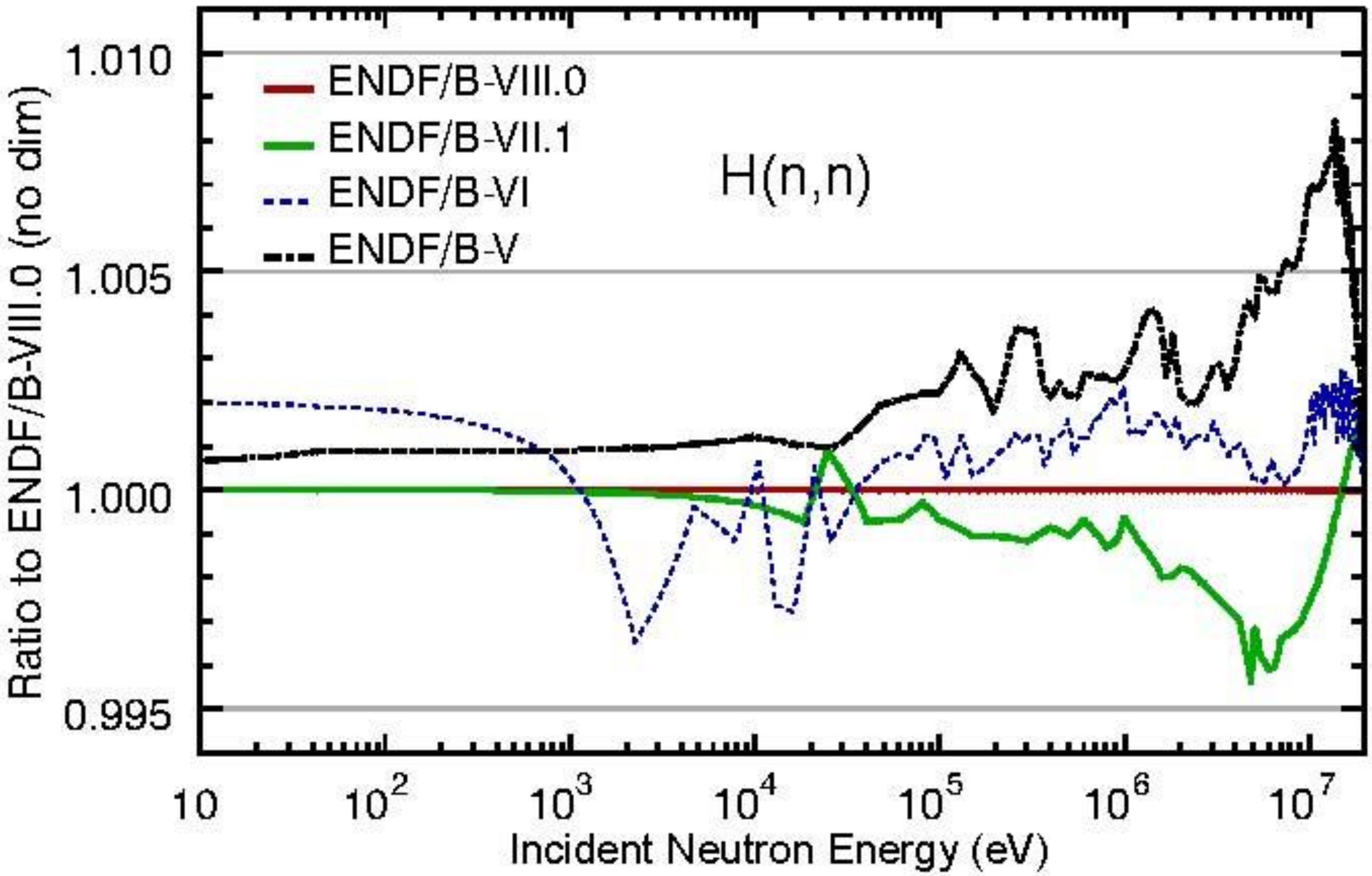
- Improvements came with various phase shift calculation by several groups
  - The Hopkins-Breit (Yale) phase shift analysis was accepted for the hydrogen standard for ENDF/B-III, ENDF/B-IV and ENDF/B-V
  - Phase shift analyses were done and continue to be done by the former Arndt group

- The most recent work on the hydrogen standard has been done at LANL using the EDA R-matrix program. The ENDF/B-VI, ENDF/B-VII and ENDF/B-VIII hydrogen standards were obtained with it.

- Though the hydrogen measurements do not depend on any cross section, It should be noted that after the use of phase shift and R-matrix analyses for obtaining evaluations of the hydrogen cross section, there is some dependence of those evaluations on other nuclear data.

- Changes do occur with new hydrogen evaluations though they are rather small for the total cross section.

Comparison of hydrogen total scattering evaluations



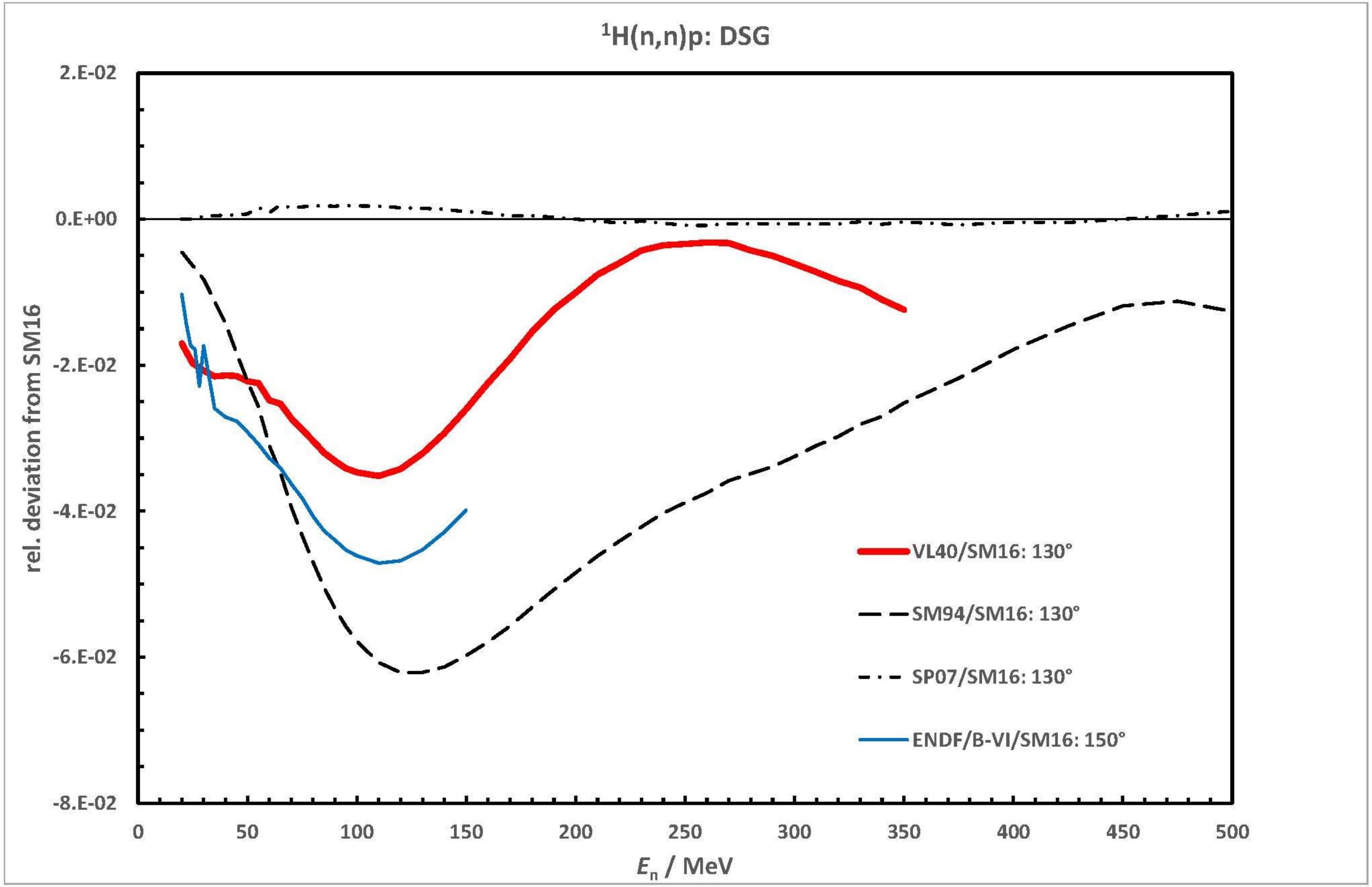
Upper MeV energies for the hydrogen angular distribution

- An important problem now is getting hydrogen scattering values in the upper MeV energy region
  - For the ENDF/B evaluations, 20 MeV is the highest energy.
  - However, for the ENDF/B-VI evaluation, in addition to the "official" evaluation there was another evaluation with an extension up to 150 MeV. Gerry Hale provided an EDA R-Matrix analysis up to 30 MeV that was "matched" to a portion of an Arndt phase shift analysis from 30 MeV to 150 MeV.
    - This "unofficial" evaluation was done (requested) so LANL fission data relative to the hydrogen cross section could be converted to fission cross sections. Gerry had concerns about this extension. I know there were problems with Legendre coefficients being continuous.
  - Work is underway at LANL by Paris and Hale using EDA. The analysis is now up to about 100 MeV with the objective of going up to 350 MeV. However data above 20 MeV are not going to be available until the next standards evaluation is completed.
  - So for measurements being made, what should they use for the hydrogen standard

So for measurements being made, what should experimenters use for the hydrogen standard

- For the n_TOF fission cross section work they were considering one of the Arndt evaluations.
  - Both the Pirovano data from 20 MeV to about 150 MeV and the Manna data up to about 450 MeV are affected.
    - Note all the higher energy evaluations considered were from Arndt (phase-shift work) who at one time evaluations 4 times a year; note SM stands for summer and SP stands for spring.
    - It is a concern that the Arndt SP07 evaluation differs from our ENDF/B R-matrix evaluations by about a percent. It only differs from the Hopkins-Breit (phase-shift) result by 0.3%
    - Unfortunately there are differences for the various Arndt evaluations and the "unofficial" ENDF/B-VI evaluation that extended to 150 MeV

Several hydrogen angular distribution evaluations



So how should an experimenter show the data obtained

- Present the data in its primary form. For the fission data it would be absolute fission data divided by absolute proton rates.

- If comparisons to other data are required, clearly state what hydrogen evaluation was used to convert the ratio.

- Until the determination of accurate hydrogen data has been done this is all you can do. When those data are available one can easily convert with the new hydrogen standard dataset.