EVIDENCE FOR A 3% “DOPPLER-ERROR” IN THE OXYGEN-16 NEUTRON CROSS SECTION DATABASE

Cecil Lubitz, Paul Romano, Timothy Trumbull, Devin Barry

Knolls Atomic Power Laboratory

Bechtel Marine Propulsion Corporation

Box 1072, Schenectady, New York, 12301, U.S.A.

NEMEA7 WORKSHOP

NOVEMBER 5-8. 2013

Background - I

- Scattering cross sections of light elements are typically flat at zero Kelvin but acquire a "Doppler-1/v-tail" at T>0. At room temperature and 2200 m/s the increase factor is (1 + 1/2A), ($A=M/m_n$) For O16 it is 1.0315. [LU2008]
- Neutron optics measurements of the coherent scattering length give a 0K value irrespective of the temperature of the sample or of its physical composition or structure.
- The thermal scattering cross sections in the Atlas, mostly coherent cross sections, are zero kelvin values inconsistent with the 2200 m/s reaction cross sections, which are explicitly room temperature.

Background - II

- As a consequence, someone using the Atlas O16 thermal scattering cross section as a room temperature value might unknowingly incur a 3.15% error.
- The tabulated cross sections in the Standards are 0K values, but are mostly reaction cross sections exhibiting 1/v invariance to Doppler broadening. The thermal values for H1 and C12(n,n) are Standards and are 0K values.
- Doppler-broadening codes used to routinely neglect the “small term” in the theoretical formula because it was negligible at high energy. That loses the low-energy Doppler-tail so that someone looking at the room-temperature thermal range for a light isotope could be misled into believing it was flat down to E=0.

The 3% Hypothesis - I

- At PHYSOR 2012 Kozier, et al, showed widespread occurrence of 3% high thermal scattering cross sections in evaluated data libraries. [KO2012]
- The simplest explanation is that the room-temperature value unknowingly got into the 0K evaluated files. Hale said that was unlikely because the ENDF/B-VI.0 value, 3.8883 b, was driven by high-energy total cross section measurements, not thermal values.
- Examination of various high-energy experiments found some, Cierjacks 1968, Ohkubo 1985, and Johnson 1974 which were apparently several percent high, clustering around 3%.

The 3% Hypothesis - II

- Understanding the reasons was difficult because of insufficient documentation.
- Cierjacks 1968 is unpublished and no information is available other than EXFOR.
- Ohkubo's Santa Fe paper is on cerium oxide and says they subtracted 2 times 3.85 barns for the oxygen, later changed to 3.85 – 0.002E (keV). They suspected 3.85 was high and ascribed it to an unknown amount of water contaminant.
- Johnson 1974 is unpublished and the data below 0.3 MeV were withdrawn. "Unknown problems" and "unknown impurities" are suspected.

The 3% Hypothesis - III

- These experiments and their atypical documentation suggest a hypothetical scenario: To check their transmission results they extrapolated to thermal and compared to the “well-known” room-temperature 2200 m/s cross section they believed to be flat but which incorporated a 3.15% Doppler increase. They would have interpreted the 3% of ~4 barns as a 120 mb background error. At higher energy where the cross section is more like 2 barns it would have looked like 6%. Not knowing how they could have missed a background of that magnitude, they decided to not publish the results.

The 3% Hypothesis - III

- In retrospect, they are accurate measurements, simply normalized (for whatever reason) about 3% high. When consistently normalized they agree very well.

Five High-Precision Data Points - I

- The following experiments are characterized by either using the words "high-precision" or "precision" to describe their work or by attaching small uncertainties to their results.
- Their other characteristic is that they are immune to the Doppler-rise error and can therefore serve to identify files which were incorrectly normalized.
- Schneider [SC1976] subtracted the scattering length of silicon from quartz (SiO_2) to get Bcoh(O)=5.830(2)f. The 0K elastic cross section is 3.7938(0.0026) and the .0253 eV total is 3.7940. ***This needs updating to whatever Bcoh of silicon is now.***

Five High-Precision Data Points - II

- Dilg [DI1971] subtracted silicon from quartz transmission using a cobalt-resonance to scatter the beam at 130 eV. ***Measured quartz and silicon in different experiments*****.** Their value, 3.761±0.007(0.19%) is quoted in the Atlas of Neutron Resonances. ***ENDF/B-VII silicon is 2% below their value so we quote them as 3.7831.***
- ***This reduces the discrepancy with Schneider from 13 sigma to 4 and removes "transmission support" for the Atlas "optics" value 3.7614.***
- ***Recommendation: Arrange for an expert examination of the optics measurements which cluster around Bcoh=5.08 f to see why they are so low.***

Five High-Precision Data Points - III

- Koester [KO1990] double-scattered from Cu63 and Se80 then transmitted through silicon and quartz at 1970 eV. 3.77±0.03(0.8%). Measured silicon in the same experiment. No need for an update, since the silicon and backgrounds cancel.

Five High-Precision Data Points - IV

- Block [BL1975] subtracted iron-filtered transmission at 23.5 keV through Al_2O_3, SiO_2, Al, and Si. No update needed. ***3.736±0.007b Is sometimes quoted as corroboration of Dilg's 3.761 but is actually lower because the cross section has fallen between .0253 eV and 23.5 keV.***
- [Window] This is a representative value for the bottom of the window at 2.35 MeV. It received a lot of attention and the Cierjacks 1980 value is 100 mb. Although small, it is clearly distinguishable from measurements reporting 103 or higher.

Cierjacks 1980 High-Precision Transmission

- Cierjacks [CI1980] used a liquid oxygen target to eliminate silicon problems. Often used as an energy standard.
- One problem: They say “Typical background corrections were of the order of 1-3% ... Due to possible long term effects in the oxygen run, where the transmission and the background spectra were measured at different times, *the total cross section curve was normalized at 3.5 MeV to our previous thin sample measurement.”*

  No numbers given.

Peak Height Analysis of Cierjacks 1968 – I

- There is a resonance in O16 at 1 MeV. In evaluated files its height is limited by unitarity *but that does not apply to normalized experimental data.*
- The observed height is a combination of the unitary peak riding on the backgounds from the other non-interfering channels. The unitary height is uniquely determined by the resonance energy $(J+1/2)4\pi/k^2$, but the other channels may differ a little from evaluation to evaluation.
- In SAYER 2000 the height is ***8.147***
- In JENDL40 the height is ***8.201.*** The SAYER-JENDL average is ***8.174.***
- In CI68 it is ***8.430***. ***8.430/8.174 = 1.0313***

The Adjustment Procedure - I

- The value 8.430 for the CI68 peak height was obtained by smoothing the experimental data in the vicinity of the peak with the shape from SAYER 2000. It used the same procedure we used for adjusting all the cross sections.
- The effects we are trying to unravel are multiplicative normalization factors and additive constant backgrounds, so we use as the form of the adjusted data set $\sigma^{T\prime} = A\sigma^{T} + B$, where T stands for "to-be-adjusted". The high-precision set to which we adjust is σ^{H}. In order to not alter σ^{H} we interpolate σ^{T} to the H-energies and write

$$\chi^2 = \sum_i \left(A\sigma_i^T + B - \sigma_i^H\right)^2$$

The Adjustment Procedure - II

- The solution to the two equations

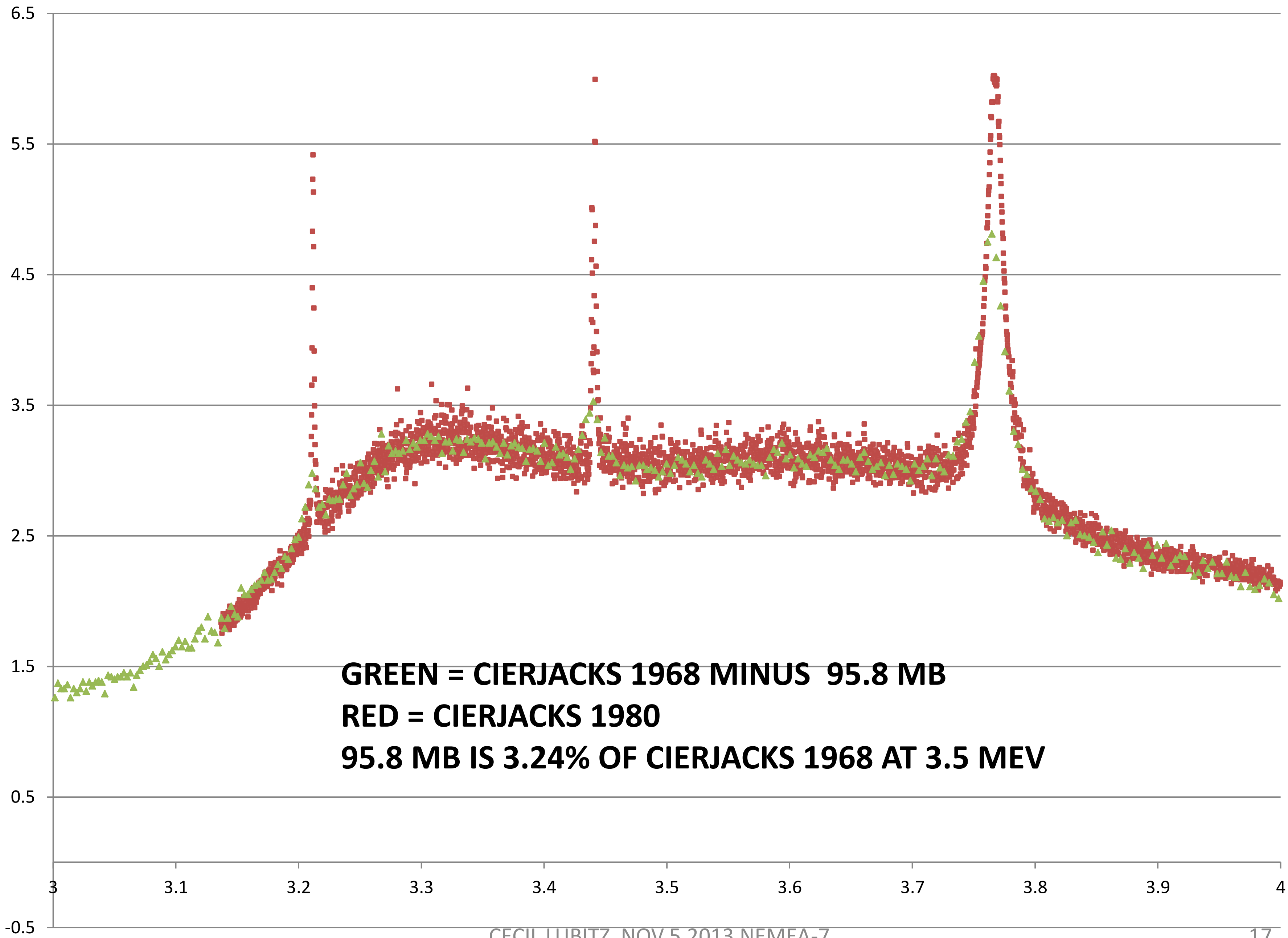
$$\frac{\partial \chi^2}{\partial A} = 0; \quad \frac{\partial \chi^2}{\partial B} = 0$$

gives the desired values of A and B. The code also prints out the RMS deviation between the adjusted file $\sigma^{T'}$ and the "high-precision" file σ^{H}.

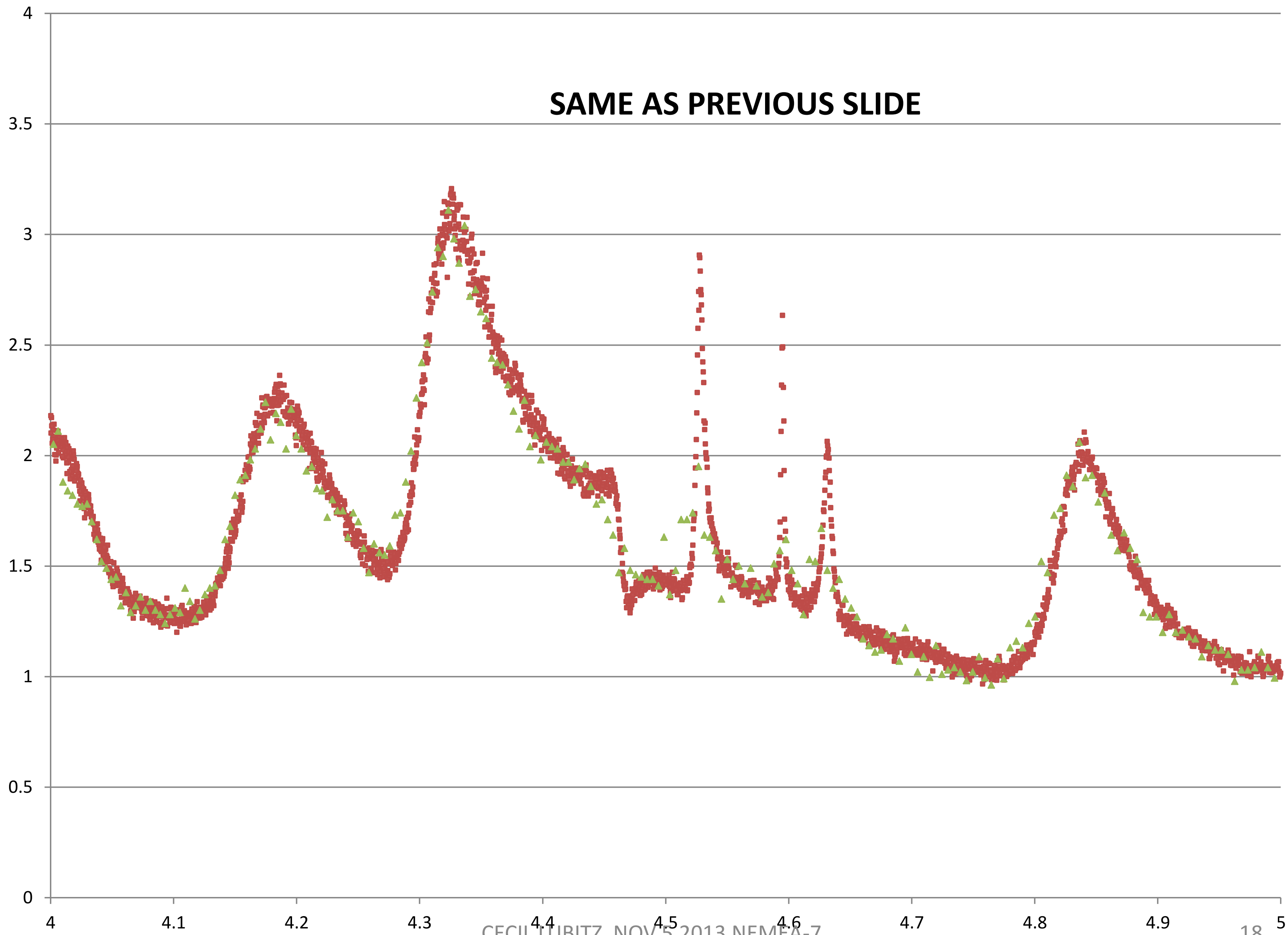
- In the case just discussed, the jittery CI68 points in the vicinity of the peak were the "high-precision" data and the SAYER 2000 points defining its peak were the T file. T' is then a smoothed shape of the experimental data.
- An energy shift would be useful but is much more complicated.

Normalization of Cierjacks 1980 to 1968 – I

- To estimate the smoothed values of CI68 and CI80 at 3.5 MeV we used a least-squares quadratic fit, The presence of little blips in the experimental data threw off an attempt to use the SAYER 2000 “shape” as we did for the peak.
- The CI68 quadratic is $y = 12178.01532\,x^2 - 85792.42052\,x + 154141.8751$ mb. The value at 3.5 MeV is ***3049.1*** mb.
- The CI80 quadratic is $y = 282971.6335\,x^2 - 1977339.946\,x + 3457240.563$ mb. The value at 3.5 MeV is ***2953.3*** mb.
- ***The ratio is 1.0324.***
- ***The difference is 95.8 mb.***
- Since Cierjacks in 1980 blamed background, ***they probably added this amount as a constant to their data***.



6.5
5.5
4.5
3.5
2.5
1.5
0.5
-0.5
3
3.1
3.2
3.3
3.4
3.5
3.6
3.7
3.8
3.9
4
GREEN = CIERJACKS 1968 MINUS 95.8 MB
RED = CIERJACKS 1980
95.8 MB IS 3.24% OF CIERJACKS 1968 AT 3.5 MEV



SAME AS PREVIOUS SLIDE
4
3.5
3
2.5
2
1.5
1
0.5
0
4
4.1
4.2
4.3
4.4
4.5
4.6
4.7
4.8
4.9
5

Normalization of Cierjacks 1980 to 1968 – II

- Apparently they sent their *unadjusted data* to EXFOR. The good agreement says that the experiments are consistent.... 1980 reproduced the 68 data very well. The 68 data are multiplicatively high but in 1980 they probably interpreted the 3.5 MeV difference as a background of unknown origin. A multiplicative adjustment would probably look similar.

Relating Cierjacks 1980 to the Five High-Precision Lower-Energy Measurements – I

- It’s important to know how well CI80 agrees with the five HP points at lower energy. This can be done by adjusting SAYER 2000 (or any other good evaluation) to the five points and to CI80 ***independently*** and seeing how closely the results agree.
- Adjusting SAYER 2000 to the five points gives A=***0.9691*** B=***1.7*** mb RMS=***14.8*** mb ***1/A=1.0319.***
- Adjusting SAYER 2000 to CI80 gives A=***0.9684*** B=***3.7*** mb RMS=***67*** mb ***1/A=1.0326.***

Relating Cierjacks 1980 to Thermal – II

- This table compares the five HP points plus 3.5 MeV as they are given by ***completely independent*** adjustments of SAYER 2000 to the five points and to CI80.

POINT	SAYER 2000 5POINTSAB	SAYER 2000 CI80AB	DIFF MB	DIFF MB AFTER CI80 + 0.9 MB
SCHNEIDER 0253 EV 3.7940	3.7853	3.7844	-0.9	0
DILG 130 EV 3.7831	3.7848	3.7839	-0.9	0
KOESTER 1970 EV 3.77	3.7805	3.7796	-0.9	0
BLOCK 23500 EV 3.736	3.7310	3.7302	-0.8	-.1
WINDOW 2.35 MEV 0.100	0.0996	0.1015	+1.9	2.8
3500000 2.9533	2.9907	2.9904	-0.3	-.6

- This says that CI80 is ***completely consistent*** with five ***totally independent*** high-precision measurements and that adjusting SAYER 2000 to both of them simultaneously will give an accurate total cross section.

A Comment on SAYER 2000-HPP Adjusted

- We just showed that multiplying SAYER 2000 by 0.9691 and adding 1.7 mb will fit the five points with an RMS deviation of 14.8 mb.
- Since the hypothesized error is an inadvertent multiplication at 2200 m/s by 1.0315, we can ask "If you start with the adjusted cross section $\sigma^{T\prime}$, what multiplicative constant will give you back the unadjusted SAYER 2000 data set?" The answer is $\sigma^{T}/\sigma^{T'} = \frac{1}{A}\left(1 - B/\sigma^{T'}\right)$. At 2200 m/s $\sigma^{T'} =$ 3.7853 and the factor is ***1.0314*** .
- ***The SAYER 2000 experimental database, after normalizing CI80 up by 3.5%, was a high-quality collection, uniformly high by 3%.***

Analysis of Johnson 1974 - I

- A plot of JO74 against CI80 shows it to be high. Adjusting it to CI80 gives the very interesting result

  A = 0.93972, B = 59.7 mb, RMS = 17.4 mb

- Since 0.93972 = 1.0641, the code is saying that you will get a very accurate match of the cross sections, a 17.4 mb RMS difference, if you reduce Johnson's data by 6.4% and then add back 59.7 mb.
- It's possible that 6.4% is coincidentally twice 3.2% and that 60 mb is coincidentally 3% of 2 barns, a reasonable average O16 cross section, but the hypothesis says:

Analysis of Johnson 1974 - II

- "Johnson in 1974 believed Cierjacks 1968 without knowing it was 3% too high ***multiplicatively***. In his subsequent efforts to normalize his own data, he repeated the error and got a file that was 6% too high ***again multiplicatively***. That was obviously wrong and not knowing that he had erred ***multiplicatively*** he brought it back down ***additively***, assuming that he had missed some background. But he suppressed the low-energy points where the "higher" cross sections, with their higher percentage errors, didn't look so good."

Looking Ahead - I

- If one accepts the above discussion, we should stop fitting any data that is inconsistent with either the five high-precision points or the CI80 total cross section.
- Below 3 MeV we should adjust all experimental data so they are consistent with the five high-precision points. Adjusted SAYER 2000 shows that is feasible. The job will be to reproduce the adjusted cross sections with new R-matrix parameters.
- Above 3 MeV the CI80 total and published resonance parameters should be basic data with our attention put on splitting that total into scattering and (n, α).
- We have not considered the region above 6.2 MeV.

Looking Ahead - II

- Since unitarity constrains the resonance peak heights, in order to tie down the (n, α) reaction while maintaining the CI80 total we will need to adjust the non-resonant potential cross sections via the channel radii since the resonance tails are probably well-known
- It’s not customary, but there is no reason the channel radii cannot vary from channel to channel, since that is consistent with R-matrix theory, $a_c, c = \{\alpha JLS\}$. We should pin the scattering radius to the channel radius, barring some theoretical objection.
- It might be possible to fix the bound states at “known values” and not include them as fitting variables.

Looking Ahead - III

- Benchmark calculations should determine the precise value of the (n, α) cross section. The three goals are
    1. No trend with oxygen absorption.
    2. No trend with leakage.
    3. An average eigenvalue close to unity.
- This will involve adjusting the angular distributions, which can be tricky. KAPL's work in that area, currently in the low-energy end of the ENDF/B-VII.1 evaluation, was done in the context of a 3% high scattering cross section and 32% high absorption, so will surely need modification. SAYER 2000 "matches" that data so the same comment applies to it.

Looking Ahead - IV

- KAPL used a non-standard R-Matrix code, which incorporated a potential-well phase shift to augment the hard-sphere. SAYER 2000 was apparently able to achieve the same result with SAMMY and we should go that route.
- Using the benchmarks as an integral part of the process makes it computationally intensive, but that will be a CIELO theme. We now have good eigenvalues with the wrong cross sections and we should be able to keep them at least as good.

References - I

[LU2008] C. R. Lubitz, “The Temperature of Coherent Scattering Cross Sections”, Cross Section Evaluation Working Group, 2008.

[KO2012] PHYSOR 2012

[SC1976] Schneider, Acta Cryst. (1976), A32, 375.

[DI1971]W. Dilg, L. Koester, W. Nistler, Phys Letters V36B #3 (1971) 208.

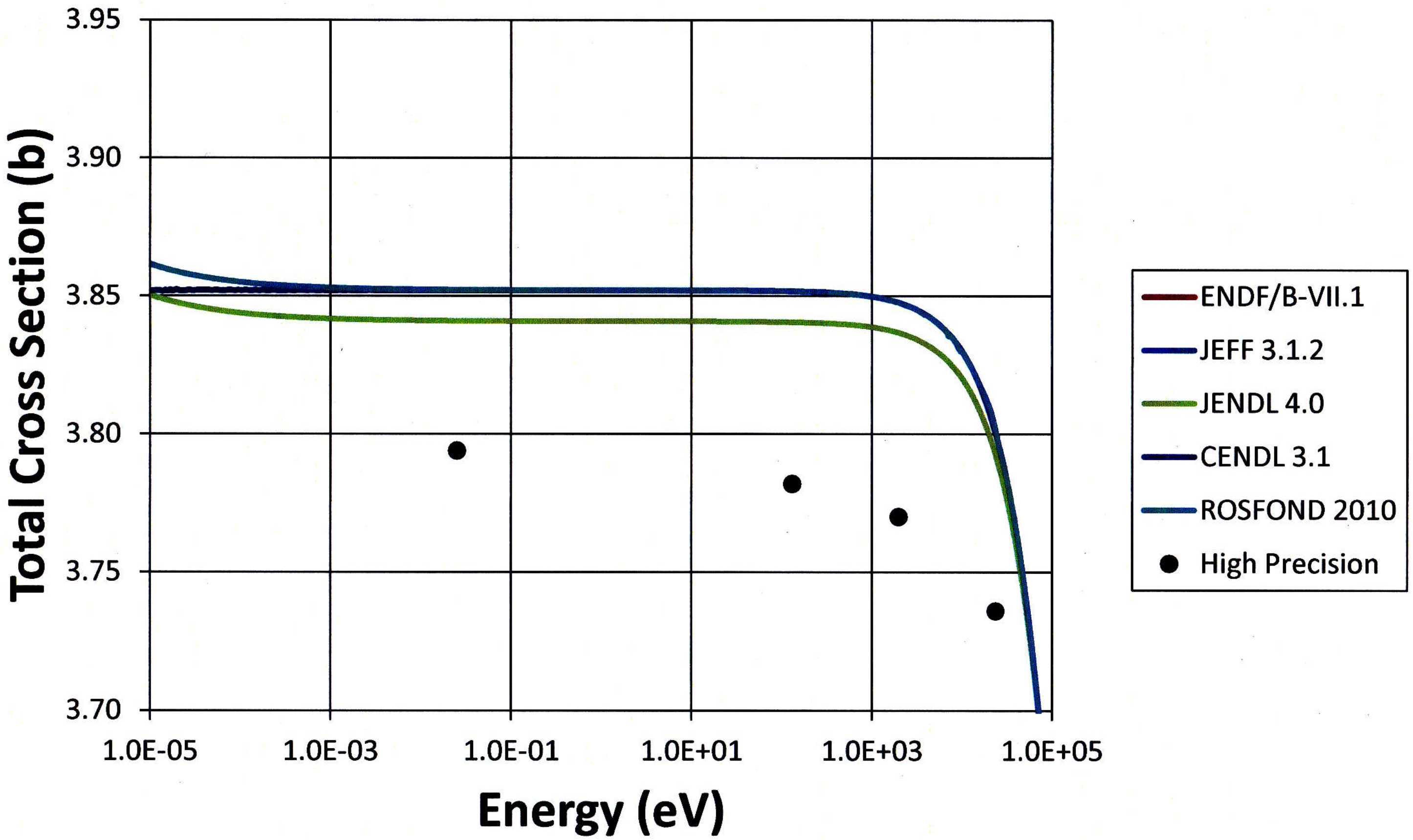
[KO1990] L. Koester, W. Waschkowski, J. Meier, Z. für Physik A Atomic Nuclei 1990, V337, Issue 3, pp 341-348.

[BL1975] R.C. BLOCK , Y. FUJITA , K. KOBAYASHI & T. OOSAKI (1975), “Precision Neutron Total Cross Section Measurements Near 24 keV”, J. of Nuclear Science and Technology, 12:1, 1-11.

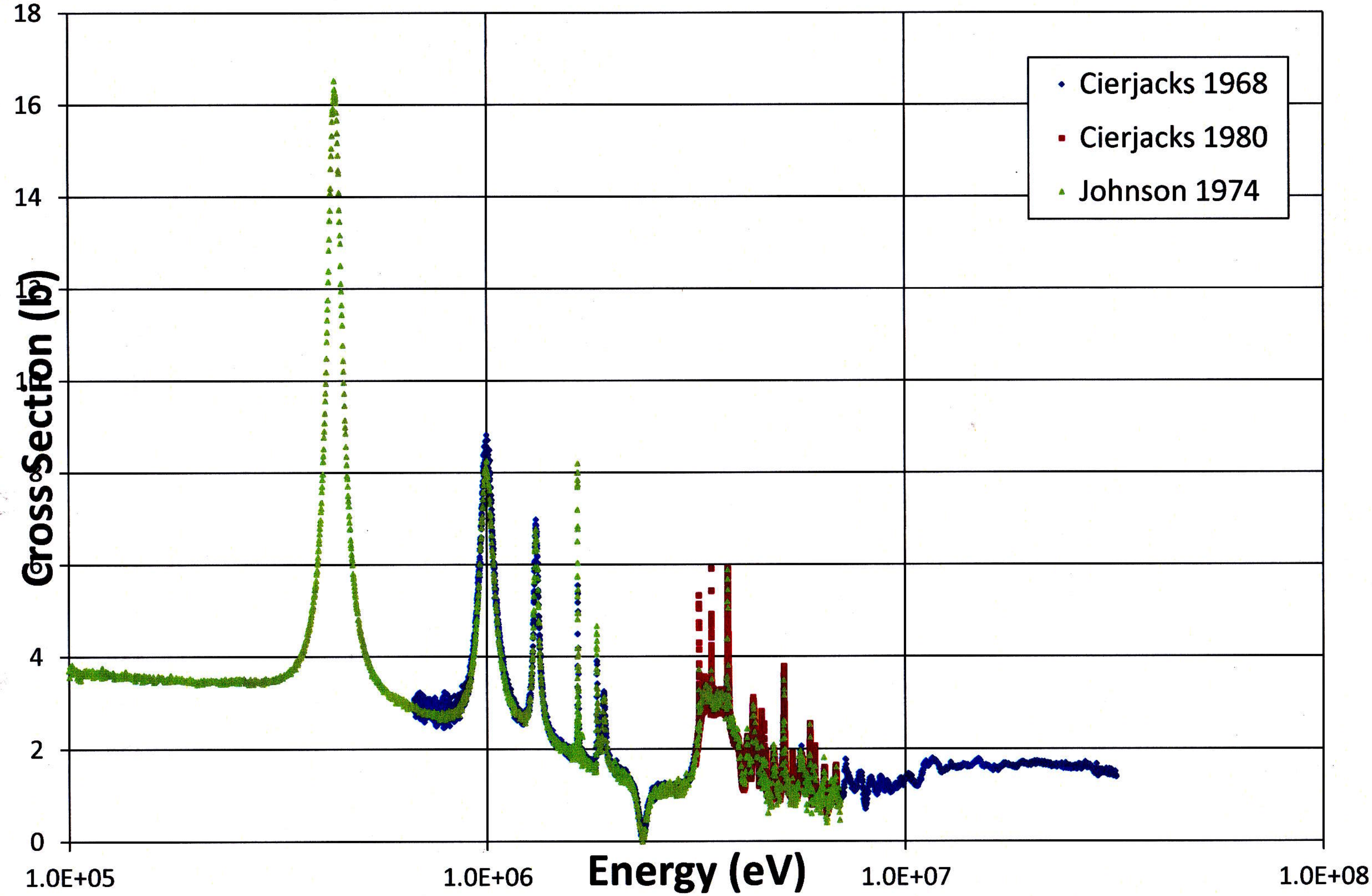
References - II

CI1980] S. CIERJACKS, F. HINTERBERGER, G. SCHMALZ, D. ERBE, P.v. ROSSEN, B. LEUGERS “HIGH PRECISION TIME-OF-FLIGHT MEASUREMENTS OF NEUTRON RESONANCE ENERGIES IN CARBON AND OXYGEN BETWEEN 3 AND 30 MeV“, Nucl. Inst. & Methods 169 (1980) 185-198.

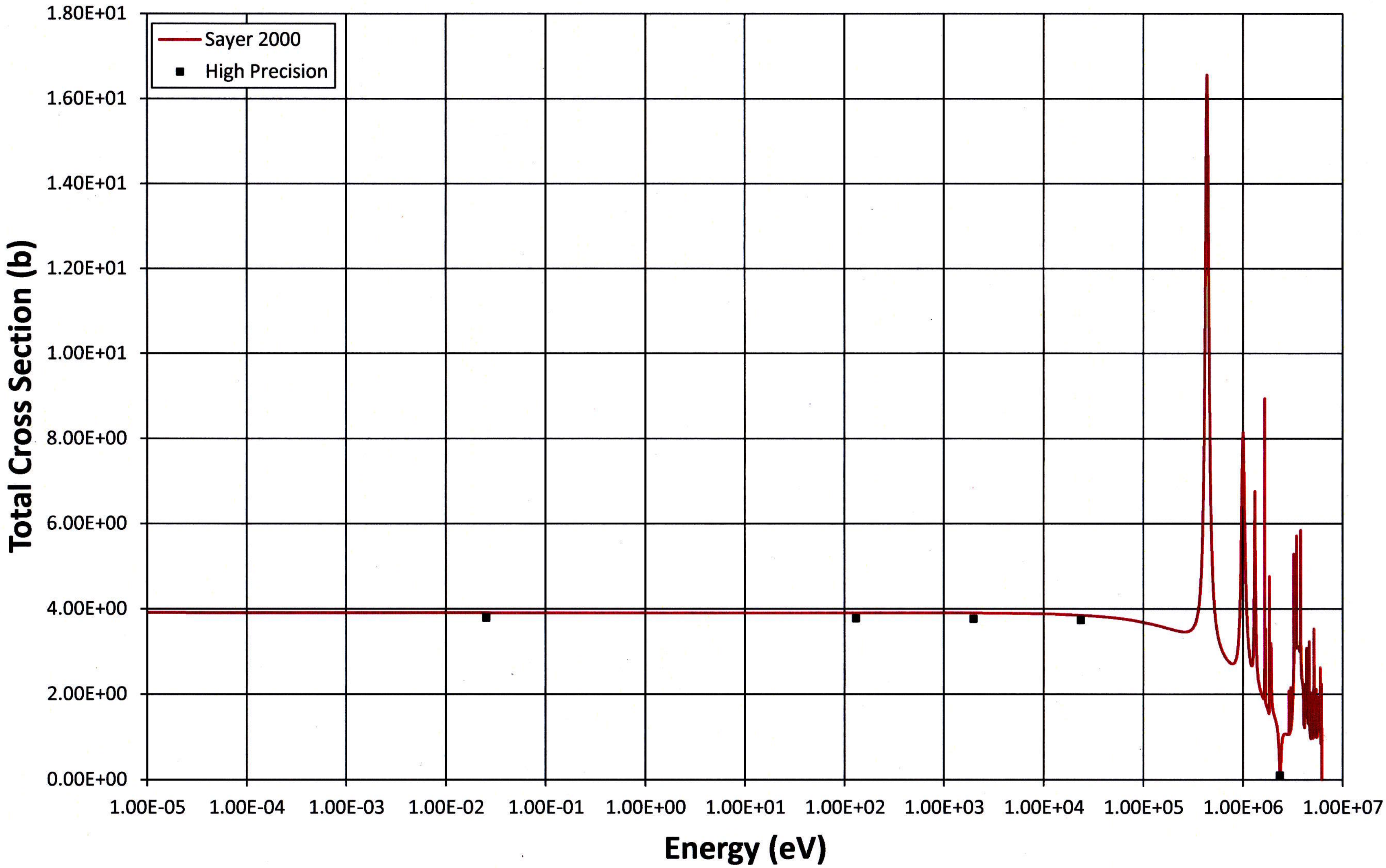
FIVE TYPICAL EVALUATIONS ARE A FEW PERCENT ABOVE FOUR HIGH-PRECISION MEASUREMENTS



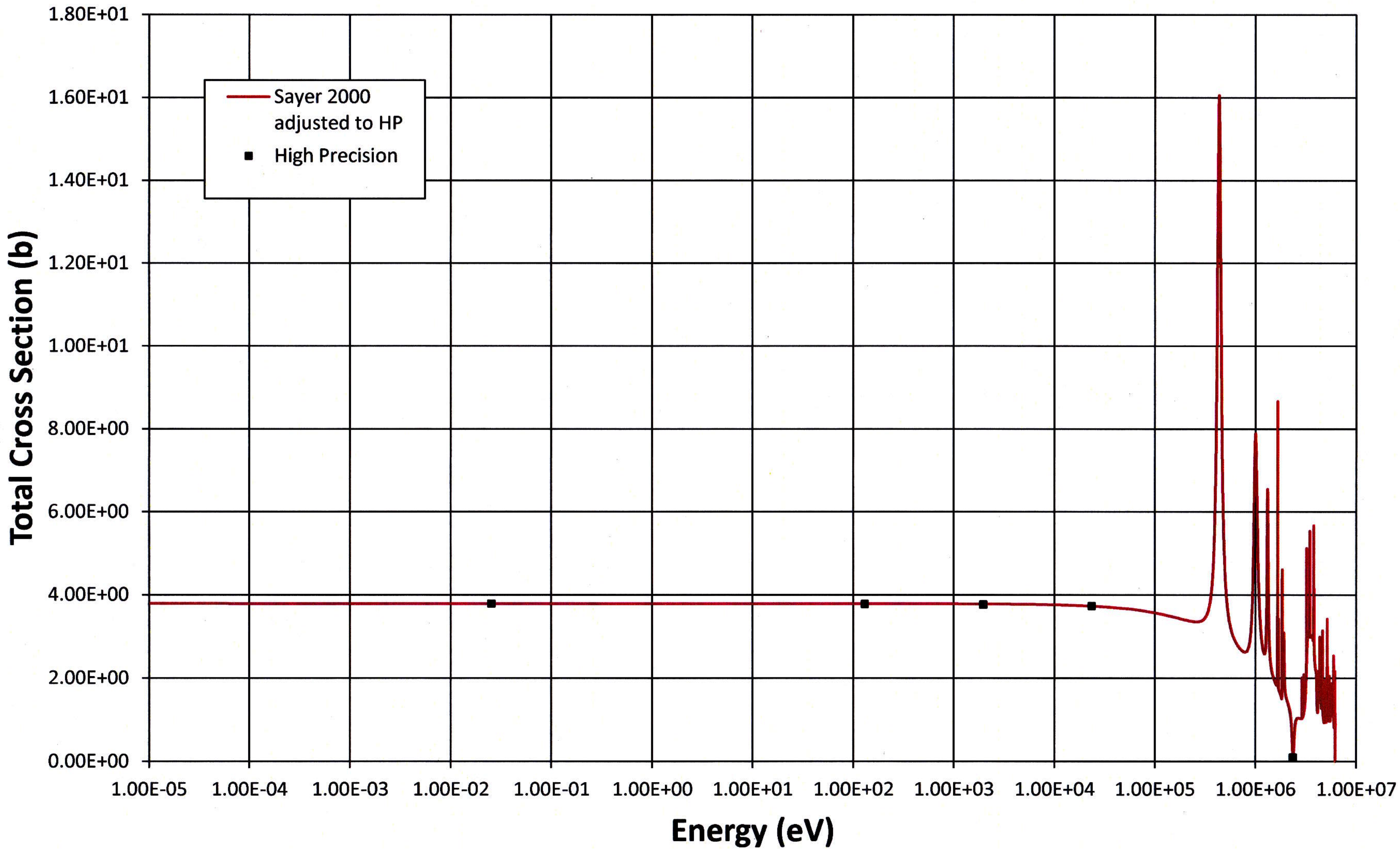
THREE EXPERIMENTAL DATA SETS



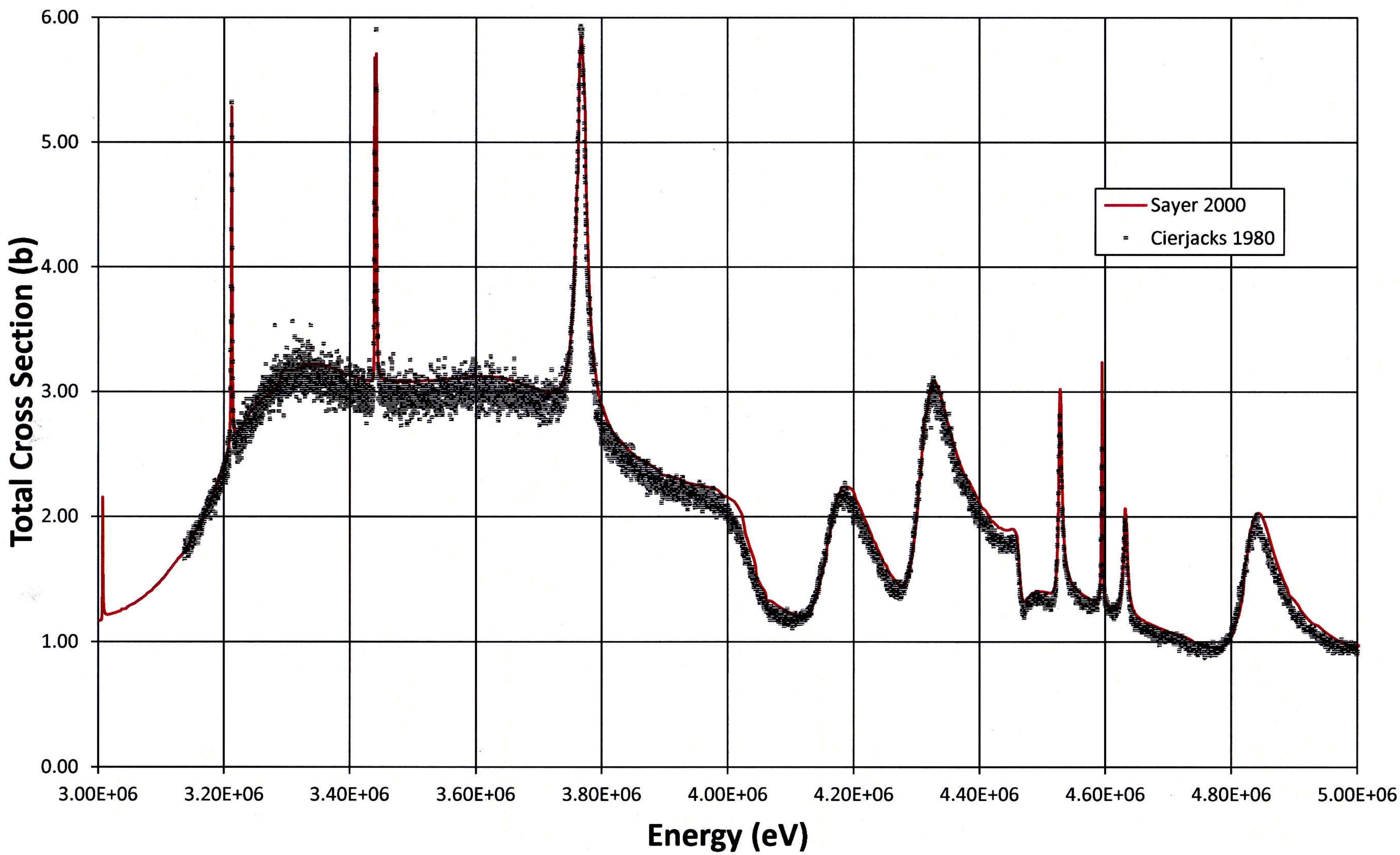
RAW SAYER 2000 3% ABOVE HPP'S



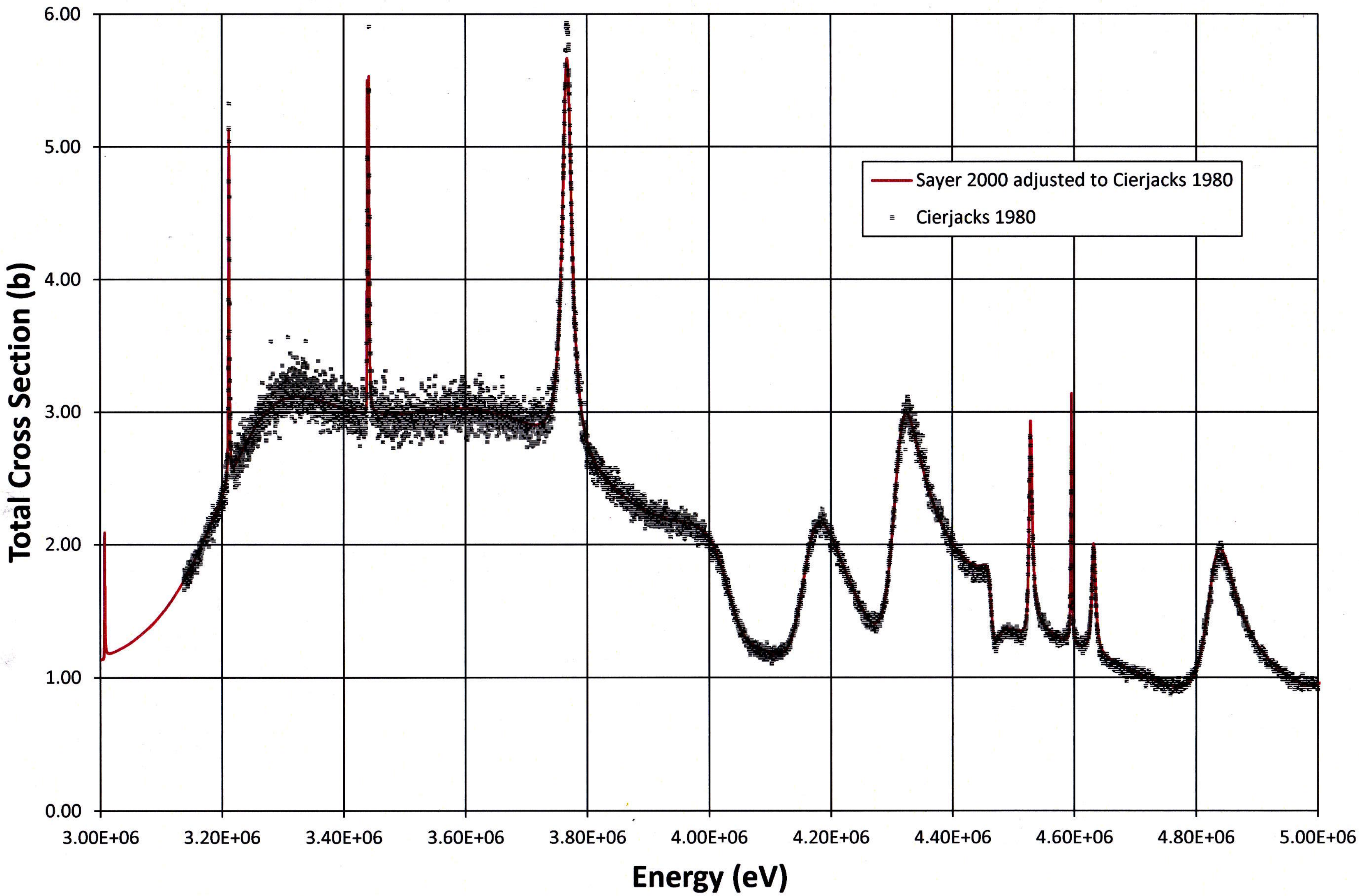
SAYER 2000 ADJUSTED TO THE FIVE HIGH-PRECISION POINTS



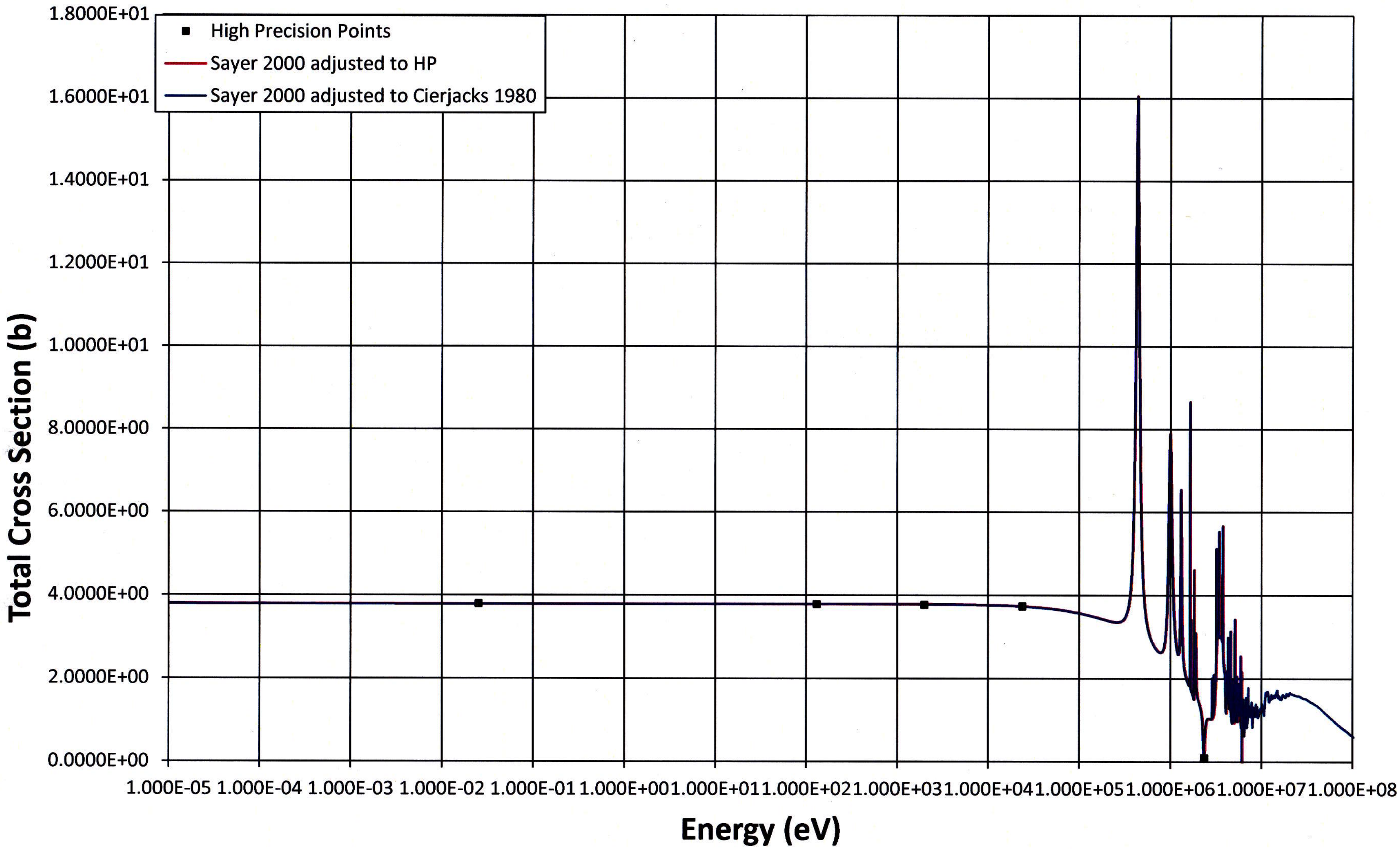
RAW SAYER 2000 AND CIERJACKS 1980



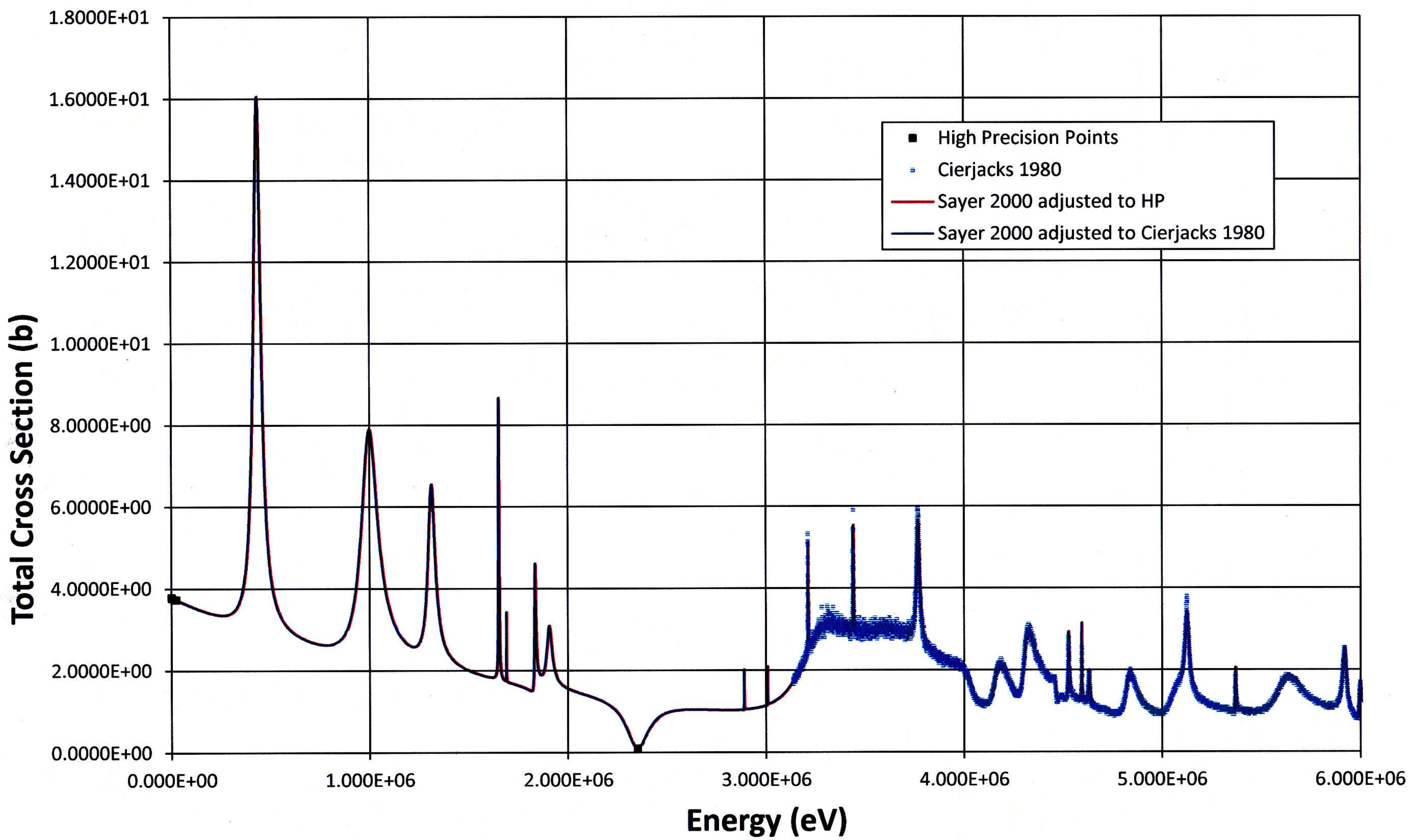
ADJUSTED SAYER 2000 AND CI80



OVERLAY PLOT OF SAYER 2000 ADJUSTED TO THE 5 HPP'S AND ***INDEPENDENTLY*** ADJUSTED TO CI80 (I)

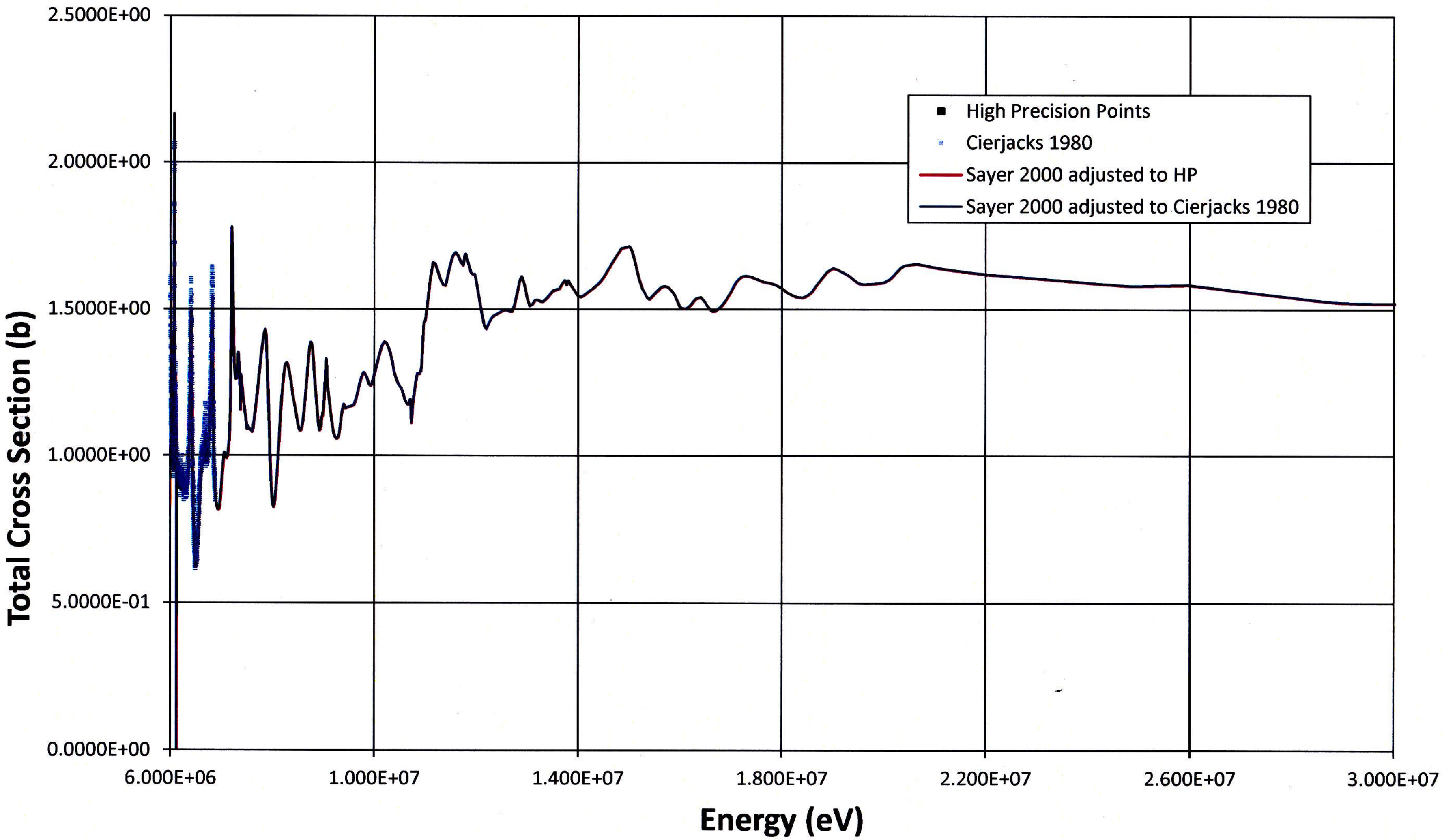


OVERLAY PLOT OF SAYER 2000 ADJUSTED TO THE 5 HPP'S AND ***INDEPENDENTLY*** ADJUSTED TO CI80 (II)



9

OVERLAY PLOT OF SAYER 2000 ADJUSTED TO THE 5 HPP'S AND ***INDEPENDENTLY*** ADJUSTED TO CI80 (III)



CONCLUSION

- *The O16 experimental database is extraordinarily well-measured and well-evaluated. Below 6.2 MeV there is no need to measure anything else.*
- *All we have to do is identify the spurious 3% in those data which were distorted by the Doppler rise in the thermal scattering cross section.*