# Progress on Deuterium

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# Outline

- Background
- New nuclear-theory results
- Ongoing ZED-2 validation testing
- GELINA measurements
- Summary

# Background

- R. Mosteller, LANL reported (CSEWG 2004) decrease in *k* of **~10 mk** (1000 pcm) for HEU-fluoride/$D_2O$ solution critical experiments, using ENDF/B-VI.5 (& later) $^2H$ relative to ENDF/B-VI.4
  - Mainly due to changes made to the *energy-angle distributions for elastic scattering* (MF 4, MT 2) <3.2 MeV
- Initial tests for low-leakage, natural-uranium ZED-2 critical lattices at CRL showed low *k* impact (<1 mk), but noticeable effect (**~0.6 mk**) on *Coolant Void Reactivity* (CVR) simulation bias
  - M. Williams, ORNL TSUNAMI ACR-700 analysis also demonstrated high sensitivity of CVR to $^2H$ *elastic scattering cross section* (but can't address angular distributions)
- AECL contracted L. Townsend, U. of Tennessee to review the experimental $^2H(n,n)^2H$ energy-angle database
  - Conclusion: old, sparse, inconsistent, tended to support increased backscatter relative to ENDF/B-VII.0; new measurements recommended
  - NEA's High Priority Request List submission (2006)
- AECL commissioned new three-body nuclear-theory calculations
- *Deuterium Data Workshop* held in Deep River on May 14, 2009

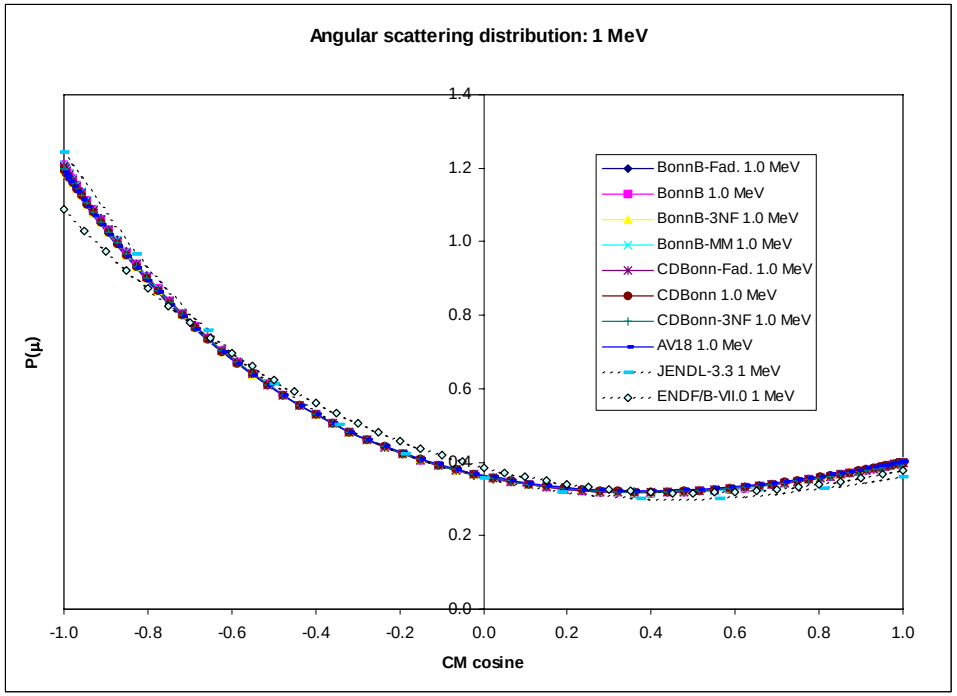
# New nuclear-theory results

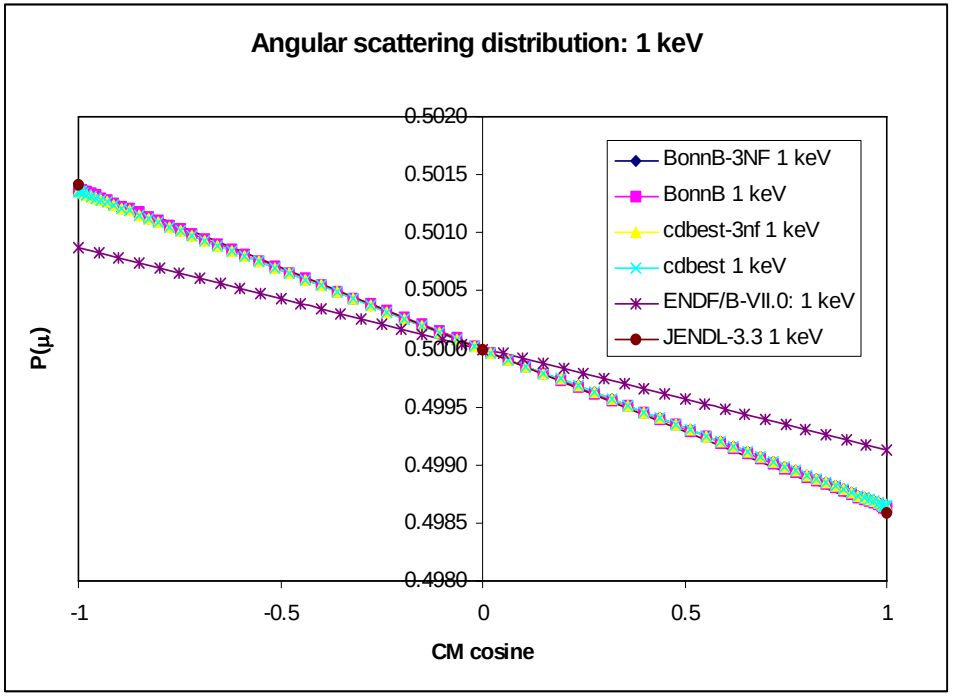
*(L. Canton - Universita di Padova; J. Svenne - University of Manitoba)*

- 8 new data sets [$\sigma_{elastic}$ for 74 CM cosines at each energy]
  - **Bonn-B** nucleon-nucleon potential using Alt-Grassberger-Sandhas (AGS) equations (73 energies; 1 keV to 29 MeV); may be the best potential
  - **Bonn-B** with 3-nucleon forces (3NF) (50 energies; 1 keV to 29 MeV)
  - **CD-Bonn** potential using AGS (73 energies; 1 keV to 29 MeV)
  - **CD-Bonn** with 3NF (51 energies; 1 keV to 29 MeV)
  - **Bonn-B** with magnetic moment (MM) interaction [no 3NF] (69 energies; 50 keV to 29 MeV)
  - **AV18** potential using Faddeev equations (32 energies; 20 keV to 3 MeV)
  - **Bonn-B** using Faddeev (32 energies; 20 keV to 3 MeV)
  - **CD-Bonn** using Faddeev (32 energies; 20 keV to 3 MeV)

# New angular scattering probability distributions P(E,μ)

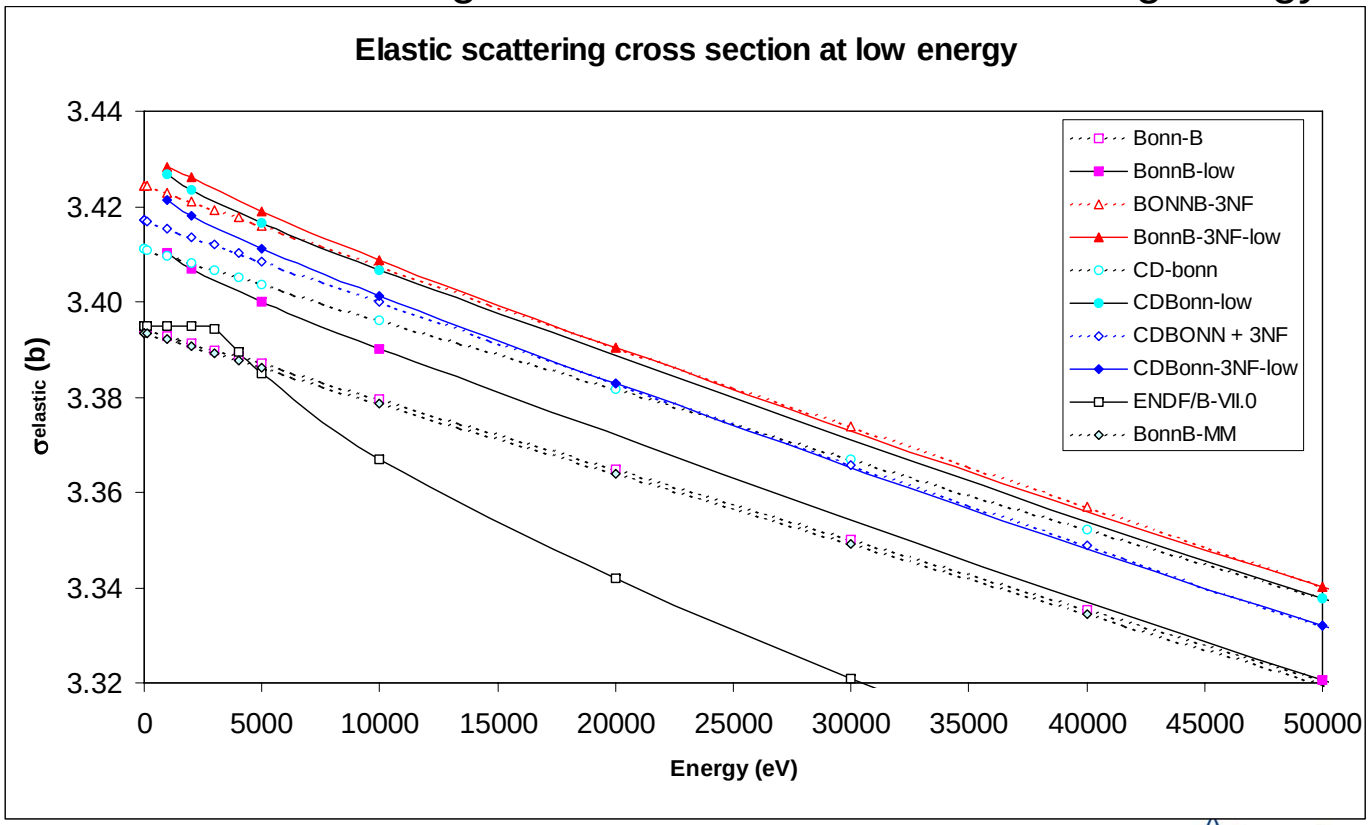
- Small differences among new data <~0.9% at 1 MeV but negligible (<0.002%) at 1 kev
- Noticeably more backscatter than ENDF/B-VII.0



# Low E scattering cross section (T=0 K)

- New theory results too high except for BonnB & BonnB-MM when using linear extrapolation of Ln($\sigma_{elastic}$) from 50 keV
- Supplementary results down to 1 keV higher still; *scaling* needed to correct for TM forces & get correct value for triton binding energy



# Cross section values calculated from nuclear-theory based scattering lengths

- H. Wita$\l$a, A. Nogga, H. Kamada, W. Glockle, J. Golak & R. Skibinski, ***Modern nuclear force predictions for the neutron-deuteron scattering lengths***, PHYSICAL REVIEW C, **68**, 034002 (2003)
- neutron-deuteron (*nd*) doublet ($^2a_{nd}$) & quartet ($^4a_{nd}$) scattering lengths calculated based on nucleon-nucleon (*NN*) interactions: CD Bonn 2000, AV18, Nijm I, II, & 93, alone & in selected combinations with Tucson-Melbourne (TM), a modified version thereof, TM99, & Urbana IX three-nucleon (3*N*) forces
- *48 combinations* involving: CD Bonn 2000, CD Bonn 2000+TM, CD Bonn 2000+TM99, CD Bonn 2000+Urb; AV18, AV18+TM, AV18+TM99, AV18+UrbIX; Nijm I, Nijm I+TM, Nijm I+TM99; Nijm II, Nijm II+TM, Nijm II+TM99; Nijm 93, Nijm 93+TM

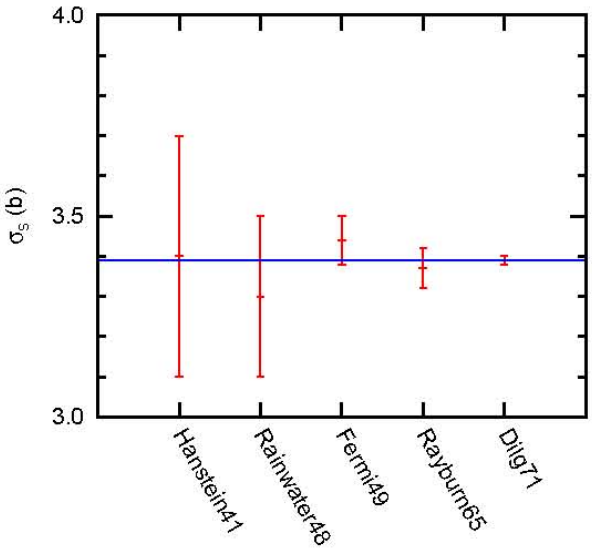
**Minimum: 3.3638 b (-0.6%)**

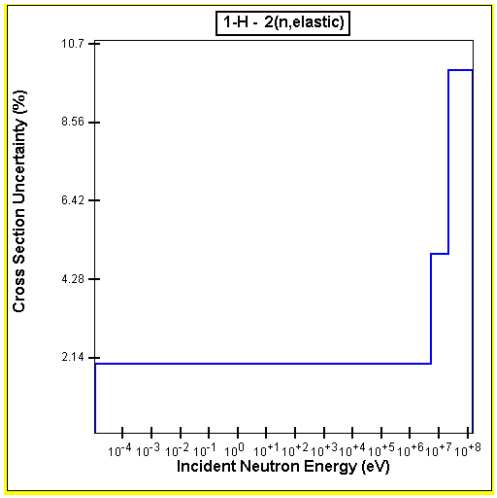
**Average: 3.3899 b**

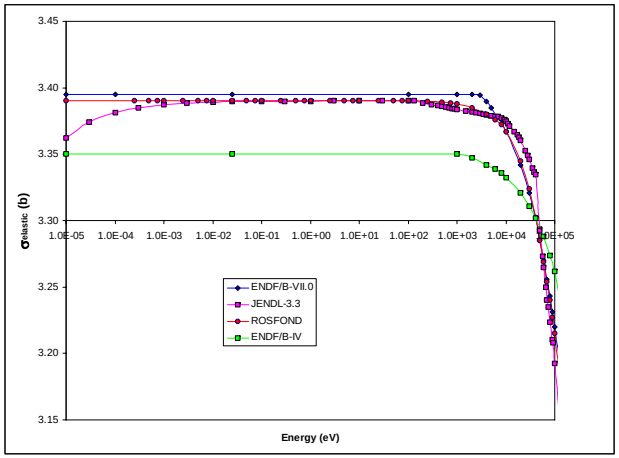
**Maximum: 3.4450 b (+1.6%)**

# Low E scattering cross section (T=0 K)

- A. Plompen review of expts. (2009): weighted average **3.390(11) b** [±0.3%]; identical to Dilg et al. (1971)
- But 'low fidelity' uncertainty ~2%; would allow TSUNAMI to adjust it
- *Other libraries* at $10^{-5}$ eV:
  - **ENDF/B-VI-VII.0: 3.395 b**
  - **ROSFOND (2008): 3.390 b**
  - **JENDL-3.3: 3.36234 b**
  - **ENDF/B-IV (IAEA Archive): 3.350 b [-1.2%]**
    - EVAL-JUN67 B.R.LEONARD,JR. AND K.B.STEWART; REV-NOV74
    - ENDF file used Legendre polynomials for MF4, MT2

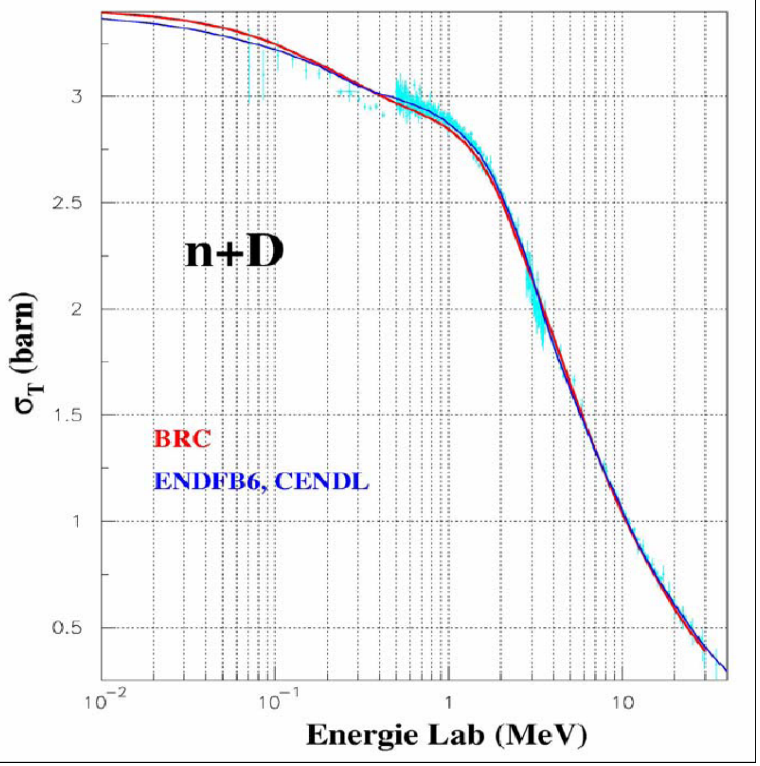


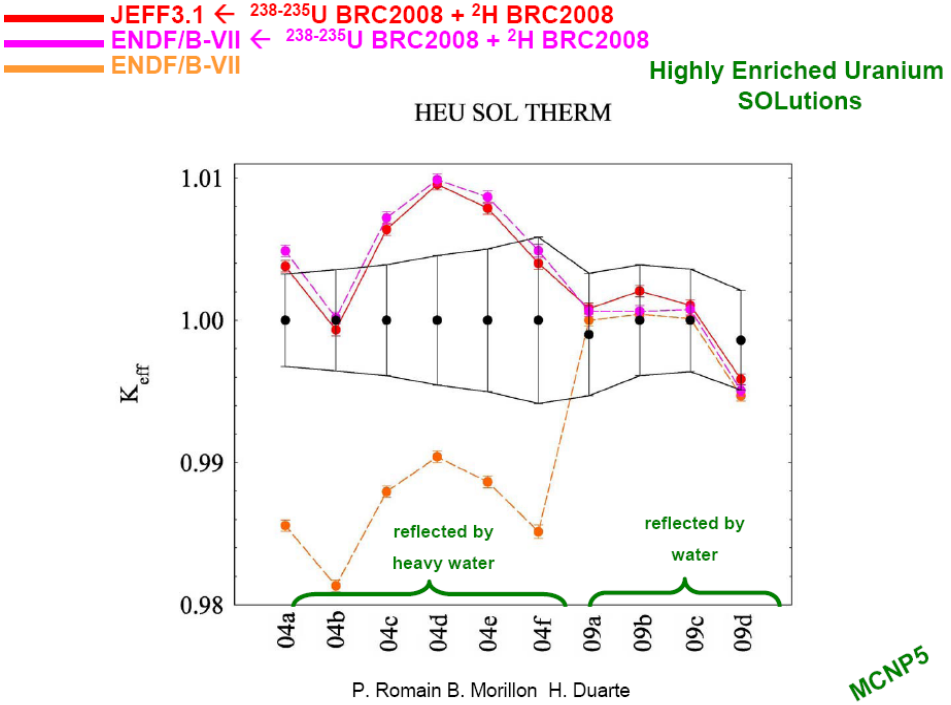



# BRC-2008 $^2$H evaluation & reactivity impact

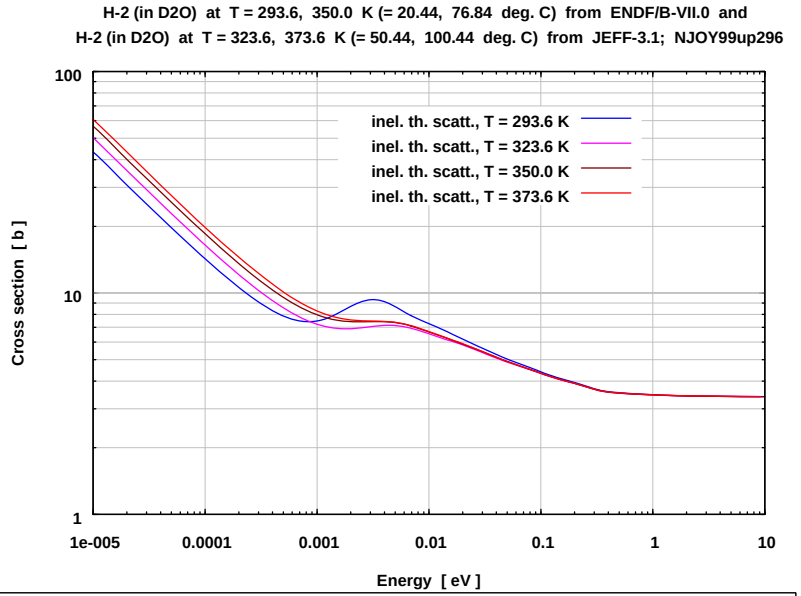
- R. Lazauskas & B. Morillon, CEA-Bruyéres-le-Chatel obtained similar nuclear-theory results using AV18 (see JEFFDOCs-1088 & 1266); may be adopted for JEFF
- $k_{eff}$ increases by **~16 mk** for HST-04

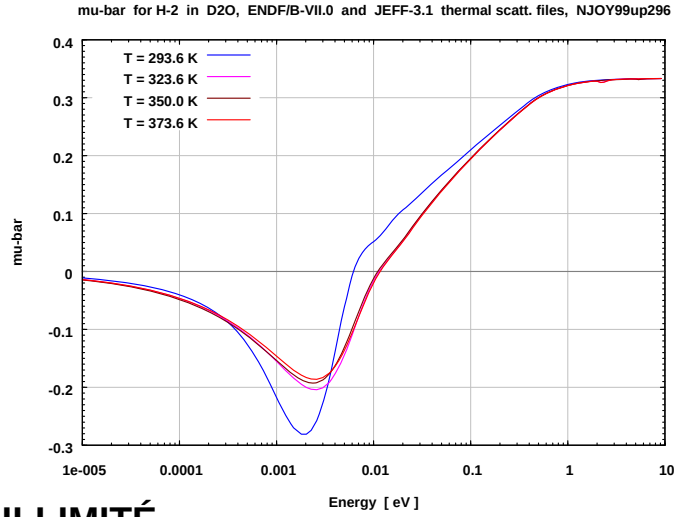


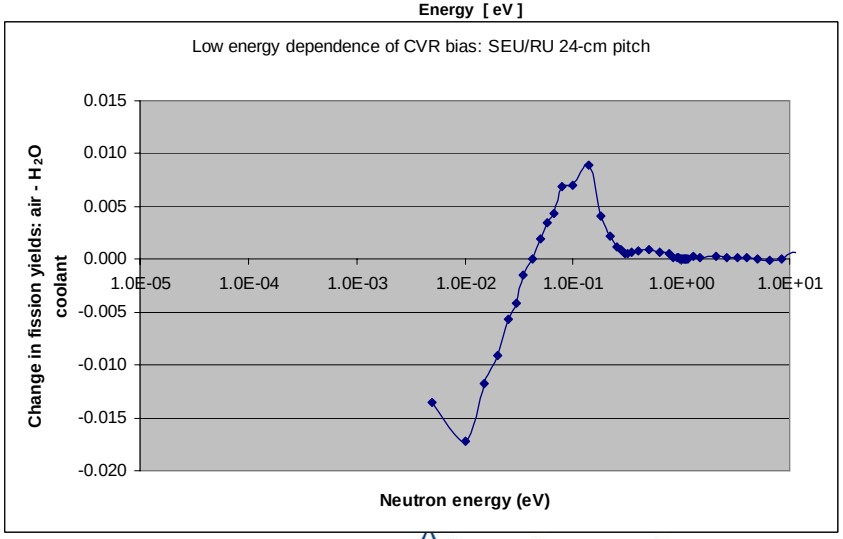



# ZED-2 CVR validation testing refinements: thermal energies

- D-in-$D_2O$ Thermal Scattering Law (TSL; $S(\alpha,\beta)$) data
  - R. MacFarlane, LANL **LEAPR** input file *interpolated* (phonon spectrum & s(kappa)) *to nearest ºC* [4ºC degree range for ZED-2 expts. studied]; $\sigma_{elastic}$(0.025 eV) changed for comparison libraries
  - 'Fast' H2 & DD2O-TSL ACE files prepared with NJOY for each T & $^2H$ ENDF file; tol.=0.0001, 64 energies, 16 angles
  - Strong anisotropy for DD2O at ~$10^{-3}$ eV
- Other data ENDF/B-VII.0 at 300 K



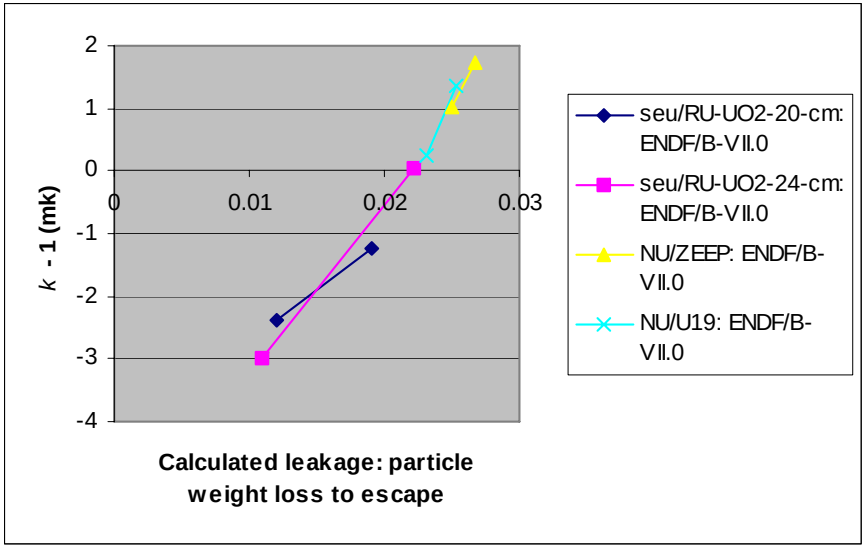



# ZED-2 CVR validation testing refinements: photonuclear data

- *Photonuclear data* (LANL-T2 ENDF/B-VII.0 g-files & IAEA-KAERI) added to account for ($\gamma$,n) reactions
  - Increases *k* by ~**0.4-0.7 mk** (94% due to $^2$H); little impact on CVR bias
- MCNP5 statistical uncertainty: *k* <±0.039 mk; CVR <±0.051 mk
- **'Average' CVR bias:** simplistic figure of merit used to gauge systematic trends
  - Typically *k* bias increases linearly with calculated leakage **[CVR bias = $k_{air} - k_{cooled}$]**
- Overall CVR bias reduced by ~0.3 mk, but may need to refine $H_2O$ TSL also

| MCNP5 CVR simulation bias (mk) | | | |
|---|---|---|---|
| ZED-2 Case | ENDF/B-VII.0 (CSEWG 2008) | ENDF/B-VII.0 reference (multiple $D_2O$ TSL, photoneutron) | ENDF/B-VII.0 reference (multiple $D_2O$ TSL, NO photoneutron) |
| SEU/RU $UO_2$: 20-cm air/$H_2O$ | 1.59 | 1.13 | 0.98 |
| SEU/RU $UO_2$: 24-cm air/$H_2O$ | 3.73 | 3.06 | 3.18 |
| NU-$UO_2$/ZEEP: air/$D_2O$ | 0.68 | 0.70 | 0.62 |
| NU-$UO_2$/U-19: air/$D_2O$ | 1.19 | 1.12 | 0.96 |
| Average | 1.80 | 1.50 | 1.43 |



# ZED-2 CVR bias $^2$H validation testing results

| MCNP5 CVR simulation bias (mk): New nuclear-theory P(E,μ) + ENDF/B-VII.0 $^2$H comparison | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| ZED-2 Case | BonnB | BonnB-3NF | CD-Bonn | CD-Bonn-3NF | BonnB-MM | AV18-Fad. | BonnB-Fad. | CD-Bonn-Fad. |
| SEU/RU $UO_2$: 20-cm air/$H_2O$ | 1.48 | 1.24 | 1.33 | 1.41 | 1.32 | 1.41 | 1.53 | 1.38 |
| SEU/RU $UO_2$: 24-cm air/$H_2O$ | 3.45 | 3.50 | 3.49 | 3.50 | 3.47 | 3.43 | 3.62 | 3.58 |
| NU-$UO_2$/ZEEP: air/$D_2O$ | 1.01 | 1.10 | 1.13 | 1.01 | 1.05 | 1.01 | 1.04 | 1.01 |
| NU-$UO_2$/U-19: air/$D_2O$ | 1.45 | 1.43 | 1.42 | 1.35 | 1.44 | 1.33 | 1.49 | 1.47 |
| Average | 1.85 | 1.82 | 1.85 | 1.82 | 1.82 | 1.80 | 1.92 | 1.86 |

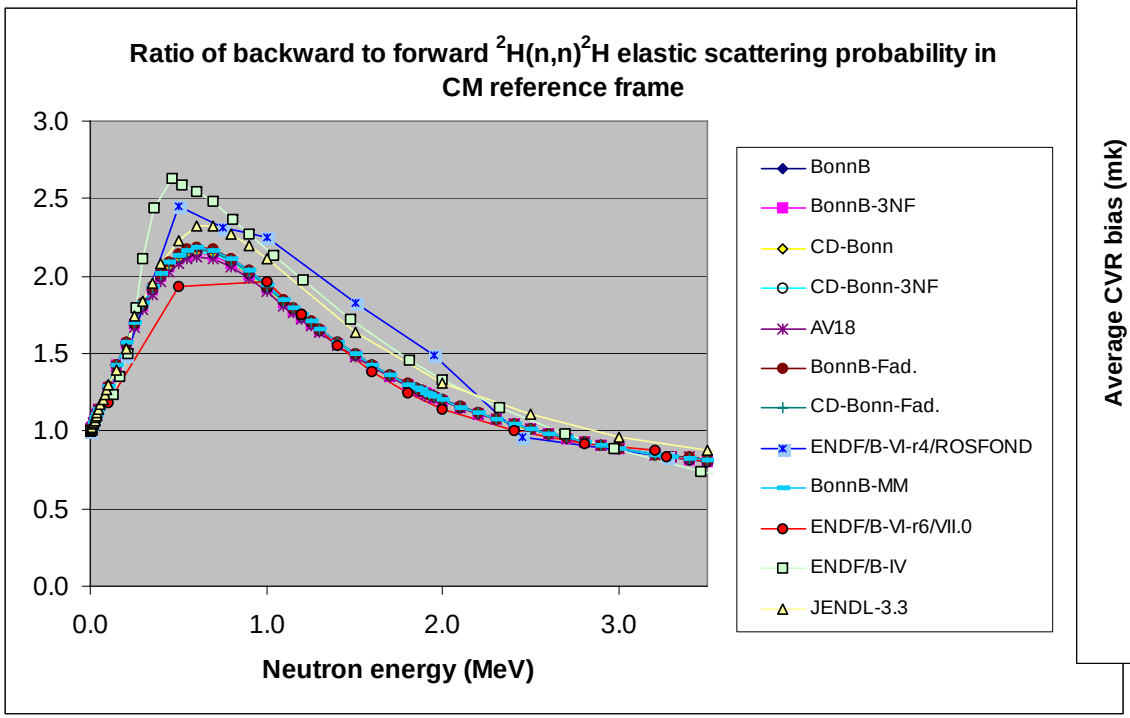
| MCNP5 CVR simulation bias (mk): $^2$H library comparison | | | | | |
|---|---|---|---|---|---|
| ZED-2 Case | ENDF/B-VII.0 reference | ENDF/B-VII.0 isotropic | ENDF/B-IV | JENDL-3.3 | ROSFOND |
| SEU/RU $UO_2$: 20-cm air/$H_2O$ | 1.13 | -0.15 | -0.42 | 0.74 | 0.97 |
| SEU/RU $UO_2$: 24-cm air/$H_2O$ | 3.06 | 1.70 | 2.04 | 3.24 | 3.36 |
| NU-$UO_2$/ZEEP: air/$D_2O$ | 0.70 | -0.40 | 0.98 | 1.27 | 1.18 |
| NU-$UO_2$/U-19: air/$D_2O$ | 1.12 | -0.10 | 1.23 | 1.49 | 1.67 |
| Average | 1.50 | 0.26 | 0.96 | 1.69 | 1.80 |

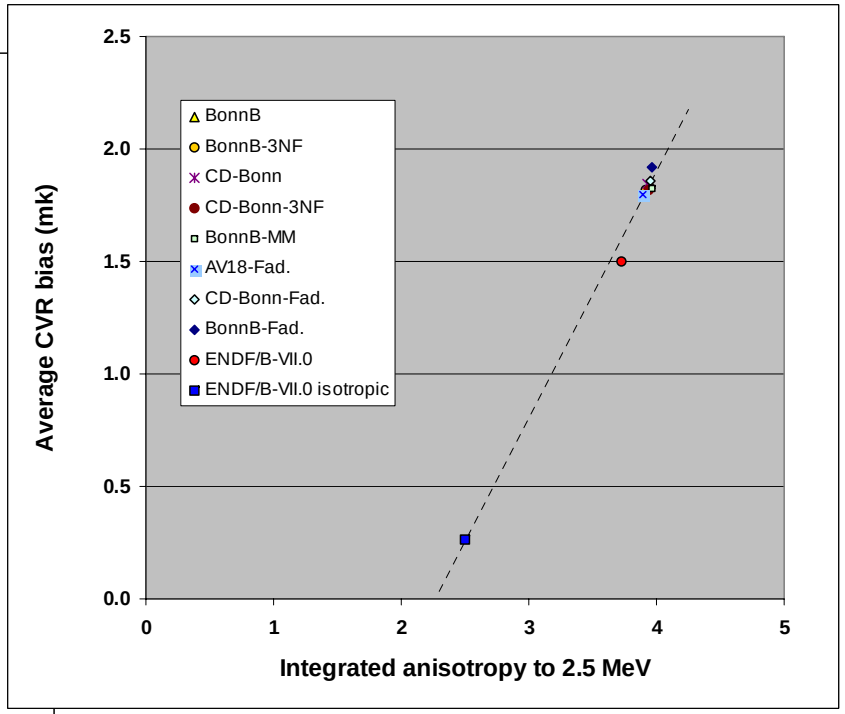
**Note:**

- Isotropic ENDF/B-VII.0 demonstrates sensitivity; right answer for the wrong reason?
- ENDF/B-IV, JENDL-3.3 & ROSFOND $^2$H include other changes besides elastic scattering
- MF6, MT16 from ENDF/B-VII.0 was used to process ROSFOND with NJOY

# Comparison of $^2H(n,n)^2H$ 'fast' anisotropy

- Used cumulative scattering probability tables from ACE files to compare *ratio of backward-to-forward scattering* in centre of mass reference frame
- Main problem with ENDF/B-VI.5 to VII.0 appears to be inadequate energy-angle numerical representation <1 MeV; ENDF/B-IV more refined
- Low CVR bias obtained for low 'integrated anisotropy' (with same $\sigma_{el}(E)$)



# GELINA $^{2}H(n,n)^{2}H$ measurements

(Arjan Plompen, Mihai Stanoiu, Nikolay Nankov & Chariklia Rouki)

- Initial measurements in July using **$C_6D_6$** demonstrated need for **$CD_2$**; sample container & high relative amount of C suppress D contribution significantly
- AECL radiochemist Raghu Rao guided synthesis of deuterated polyethylene (**$CD_2$**: 99.999% $^{2}H$) by Polymer Science Inc., Montreal & molding into a 3 mm disc at National Research Council of Canada's Industrial Materials Institute, Boucherville; arrived at IRMM Sept. 30
- Development of an MCNP model of GELINA set-up in progress (Paul Carlson, Acsion Industries Inc., Pinawa, MB)
- IRMM also considering $^{2}H$ recoil measurements using a Time Projection Chamber

Heavy hockey pucks anyone?

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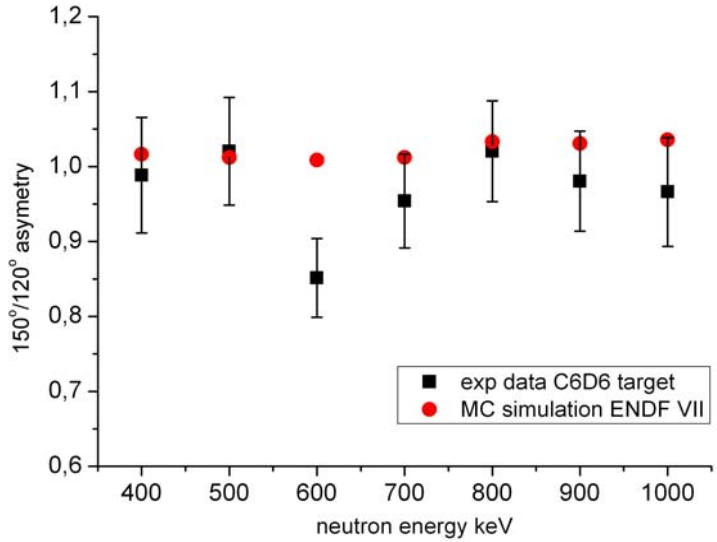
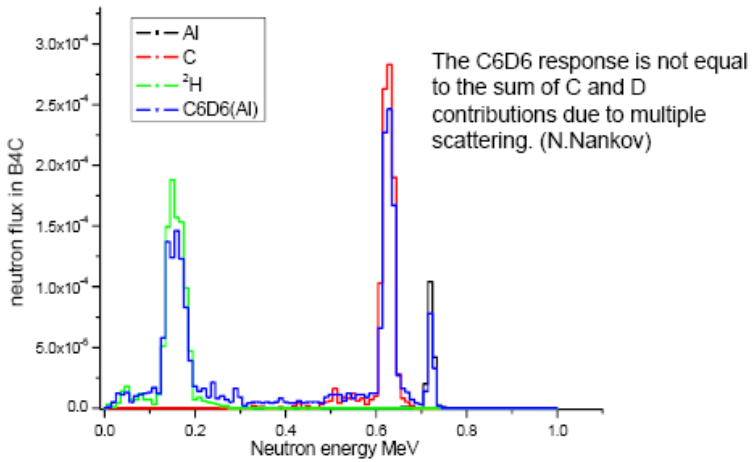
# Preliminary GELINA results using $C_6D_6$

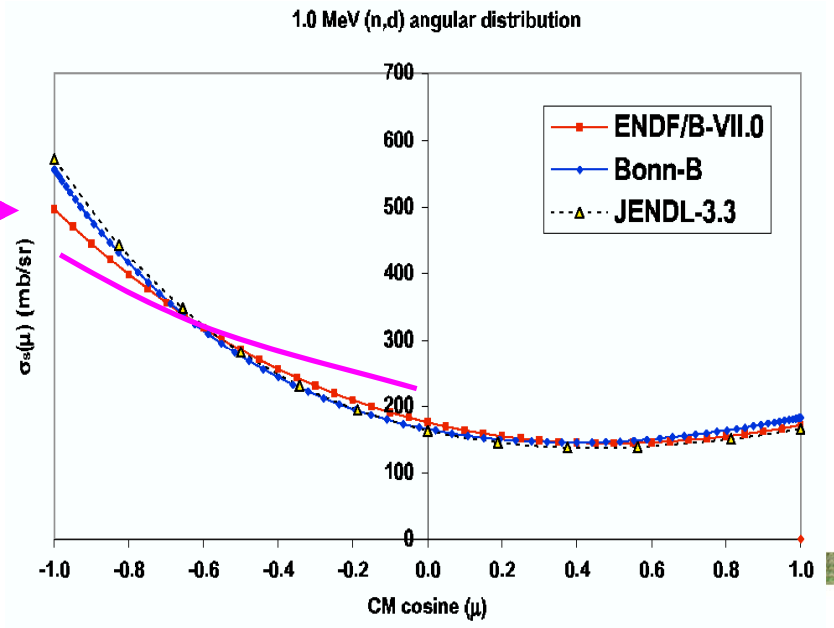
- Preliminary **$C_6D_6$** data show qualitatively *less* anisotropy than ENDF/B-VII.0 & nuclear-theory calculations

One week beam @300m




C6D6 target response at 120° for 800 keV neutron



# Summary for deuterium

- Nuclear-theory elastic scattering cross sections require scaling (in progress); sensitivity of CVR bias to P(E,$\mu$) from different potentials, 3NF & MM interaction is small
- Small reduction in $\sigma_{el}$ from 3.395 b to ~3.390 b at low E may be warranted from both theory & experiment
- ZED-2 simulation refinements ($D_2O$ TSL & photonuclear data) moderately significant (reduces CVR bias ~0.3 mk; increases $k_{eff}$ ~0.4-0.7 mk), but may need to refine $H_2O$ TSL similarly (increases CVR bias)
- CVR bias sensitive to *both* $^2H$ elastic scattering cross section value [prefer lower] & angular distributions [prefer isotropic], but there could be …
  - a systematic error due to some behaviour that is incorrectly modelled as the ZED-2 $D_2O$ moderator level changes
  - residual nuclear data issues with $^{235}U$ &/or $^{238}U$
- **Energy grid for $^2H$ MF4, MT2 inadequate in ENDF/B-VII.0**
- New measurements at GELINA using **$CD_2$** key to further progress
  - Preliminary results using **$C_6D_6$** show *less* anisotropy

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