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TITLE: SENSITIVITY STUDIES OF THE ORTHO/PARA H2 CONCENTRATION ON THE NEUTRONIC PERFORMANCE OF THE LUJAN CENTER LIQUID H_2 MODERATOR USING STATE-OF-THE-ART COMPUTATIONAL TOOLS

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Sensitivity Studies of the Ortho/Para H_2 Concentration on the Neutronic Performance of the Lujan Center Liquid H_2 Moderator Using State-Of-The-Art Computational Tools

Gary J. Russell, Phillip D. Ferguson, and Eric J. Pitcher

Abstract

A liquid H_2 moderator has been operational at the Manuel Lujan Jr. Neutron Scattering Center (Lujan Center) since 1985. Detailed Monte Carlo calculations have been made to estimate the neutronic performance of the liquid H_2 moderator cold source, using the state-of-the-art Los Alamos LAHET Code System and liquid H_2 scattering kernels. The absolute neutron spectrum leaking from the moderator has been measured. In thermal equilibrium at 20 K, the moderator would convert to 100% para-hydrogen on some time frame. We show that the performance of the Lujan Center cold neutron source is sensitive to small fractions of ortho-hydrogen, and display these results as a function of the ortho/para H_2 concentration. Integral data from the measured neuron spectrum have been combined with calculated predictions to estimate the ortho/para H_2 concentration in the moderator. From these analyses, we estimate the ortho-hydrogen concentration in the liquid H_2 moderator to be 6-9% at the time the neutron spectrum was measured.

Introduction

A liquid H_2 moderator cold neutron source has been in operation at the Lujan Center since 1985.[1] While the cold source is operating, we have no method of directly measuring the ortho/para H_2 concentration. Employing the Los Alamos LAHET Code System (LCS)[2] and the Los Alamos scattering kernels for liquid H_2,[3,4] we have used an "indirect method" to estimate the ortho/para H_2 concentration in the moderator at the time the neutron spectrum leaking from the moderator was measured by Brun.[5,6]

The neutronic performance of a liquid H_2 moderator at a spallation neutron source depends on the ortho/para H_2 concentration.[7] One reason for this is due to the different cross sections between ortho- and para- hydrogen. Figure 1 shows the calculated cross sections for liquid ortho- and para-hydrogen of MacFarlane compared to measured data.[3] The differences in these cross sections translate into distinct neutron leakage spectra from a liquid H_2 moderator. In Fig. 2, we illustrate this effect for the Lujan liquid H_2 moderator, where we compare calculated neutron leakage spectrum from the moderator with a (normal) ortho/para H_2 concentration of 75/25% to that with a (pure) 100% para-hydrogen concentration. The differences in the spectra start to manifest themselves below about 10 meV.

For a fixed moderator thickness (the as-built Lujan liquid H_2 moderator is 4.45-cm thick), the ortho/para H_2 concentration affects the average moderator brightness. We illustrate this effect in Fig. 3 for the Lujan cold source for two different energy intervals ($E < 5$ meV and $E < 10$ meV). The inclusion of para-hydrogen (with its falling cross section below about 10 meV) in a liquid H_2 moderator makes the moderator "neutronically reentrant". As can be seen in Fig. 3, the highest integrated intensity (for the 4.45-cm-thick Lujan liquid H_2 moderator) occurs for an ortho-hydrogen concentration between 35-40%. Note that an ortho-hydrogen concentration of 10% can affect the integrated neutron leakage intensities ($E < 5$ meV and $E < 10$ meV) at the level of 15-25%. Also, the integrated intensity below 5 meV is more sensitive to the ortho-hydrogen concentration than the integrated intensity below 10 meV.

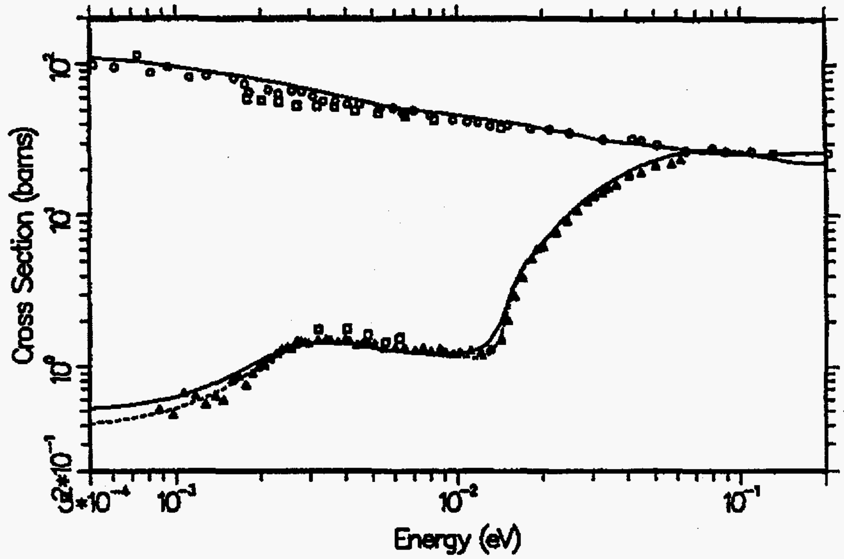


Figure 1. The cross section for liquid ortho-hydrogen (upper curve) and liquid para-hydrogen (lower curve) are compared with experimental data. The solid curves are at 20 K, and the dashed curve is at 14 K. The sharp drop in the para-hydrogen cross section below about 0.05 eV is due to spin coherence, and the second drop below 0.003 eV is due to intermolecular interference.

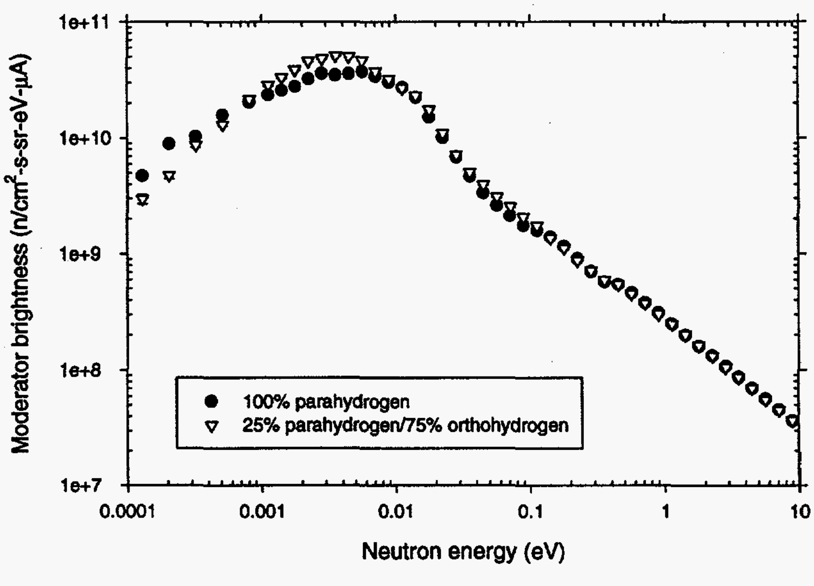


Figure 2. Calculated moderator brightness for a "pure" 100% para-hydrogen moderator compared to an ortho/para-hydrogen moderator with a concentration of ortho/para of 75/25% (a so-called "normal"-hydrogen moderator).

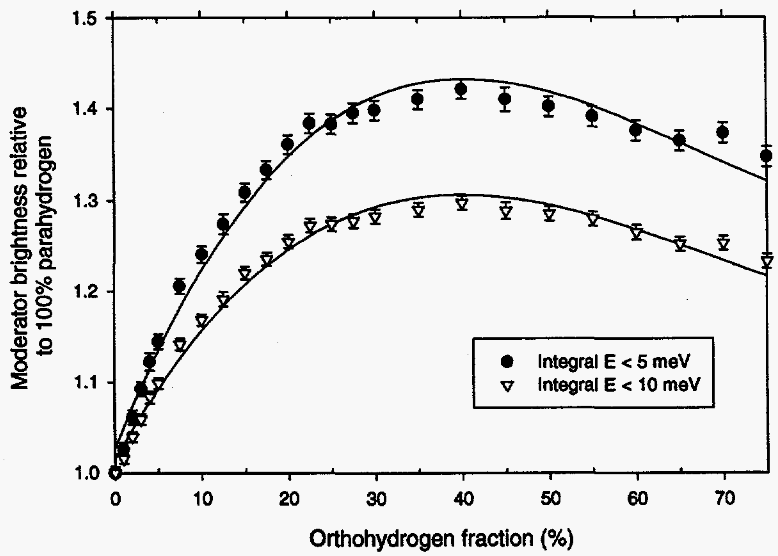


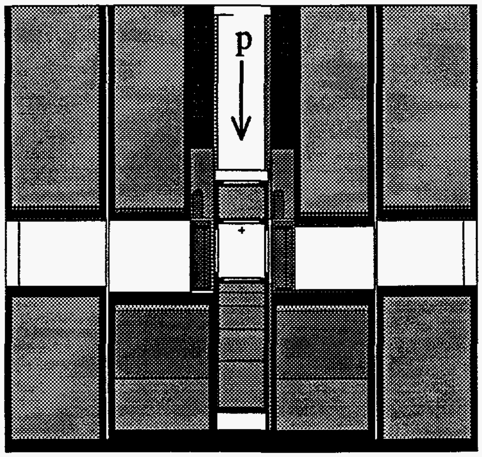
Figure 3. Relative moderator brightness as a function of ortho-hydrogen fraction for integrals below 5 and 10 meV.

Calculational Model and Procedures

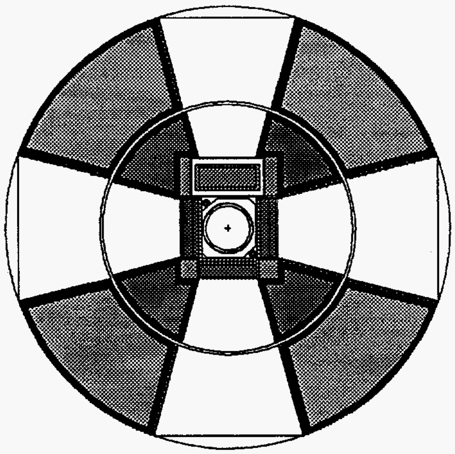
Hughes constructed a detailed Monte Carlo simulation of the as-built geometry of the Lujan Center target-moderator-reflector shield system (TMRS).[8] This geometric model captured the essence of target and moderator canisters and their relative positions, neutron flight-path penetrations, materials, etc. The Monte Carlo mockup of Hughes is depicted in Fig. 4, and this geometry was used in the present study. Another detail that is important when computing the absolute neutronic performance of a spallation target system is the size, location, and intensity of the proton beam on the spallation target. At the time the neutron spectrum from the Lujan cold source was measured, the proton beam spot was somewhat diffuse and non-symmetric. Our best estimate of the production proton beam spot at the time Brun made his neutron spectrum measurement from the Lujan cold source is illustrated in Fig. 5. Note the asymmetry with respect to the liquid H_2 moderator. This proton beam spot was utilized in the Monte Carlo calculations for this paper. All calculations were performed using the LAHET Code System[2] and the liquid H_2 scattering kernels of Bob MacFarlane[3,4].

Estimation of the Ortho/Para-Hydrogen Concentration in the Lujan Cold Source

We estimated the ortho/para-hydrogen concentration in the Lujan liquid H_2 moderator at the time Brun made his spectrum measurement as follows. We noted that the ratio of integrated neutron intensities divided by the 1-eV flux has been utilized extensively to characterize ambient-temperature moderator performance.[9,10,11] These early studies showed the dependency of this ratio with moderator material, geometry, and the presence or absence of a moderator poison. Pursuing this approach, we fit Brun's measured (n/eV-p) data from 0.1-10 eV (using a functional form of $C*E^{(1-\alpha)}$)to determine the value at 1 eV. We then numerically integrated the measured data below 5 meV and 10 meV using a simple Simpson's Rule approximation. The ratio of these integrated data to the value at 1 eV should be characteristic of the Lujan Center liquid H_2 moderator dimensions, should depend on the ortho/para H_2 concentration, and should reduce the number of uncertainties (e.g., number of incident protons, detector efficiency, etc.) when comparisons are made with the measured data.. We performed numerous Monte Carlo simulations using the Lujan target system geometry and proton beam profile discussed above. For the calculated data for the Lujan Center liquid H_2 moderator, we computed the same ratios as we did for Brun's experimental data but as a function of the ortho/para H_2 concentration in the moderator. The results are shown in Fig. 6 where we see that the calculated estimate of the ortho-hydrogen concentration for the Lujan moderator (as discussed above) would be in the 6-9% range depending on the upper energy limit on the integral. This approach provides the first estimate of the ortho-hydrogen concentration for the Lujan Center liquid H_2 moderator at the time Brun made his spectrum measurement.

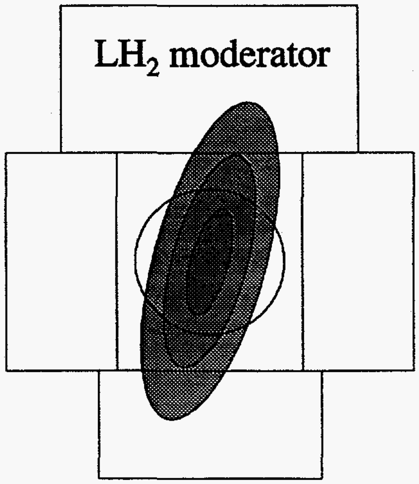


(a)



(b)

Figure 4. The Monte Carlo mock-up of the Lujan Center target system shown in elevation (a) and plan (b) views.



Production beam (for measurement)
Gaussian
$\sigma_x = 3.8$ cm $\sigma_y = 1.0$ cm
angle of rotation = 75°

Figure 5. Measured proton beam profile for the LH_2 spectrum measurement by Brun.

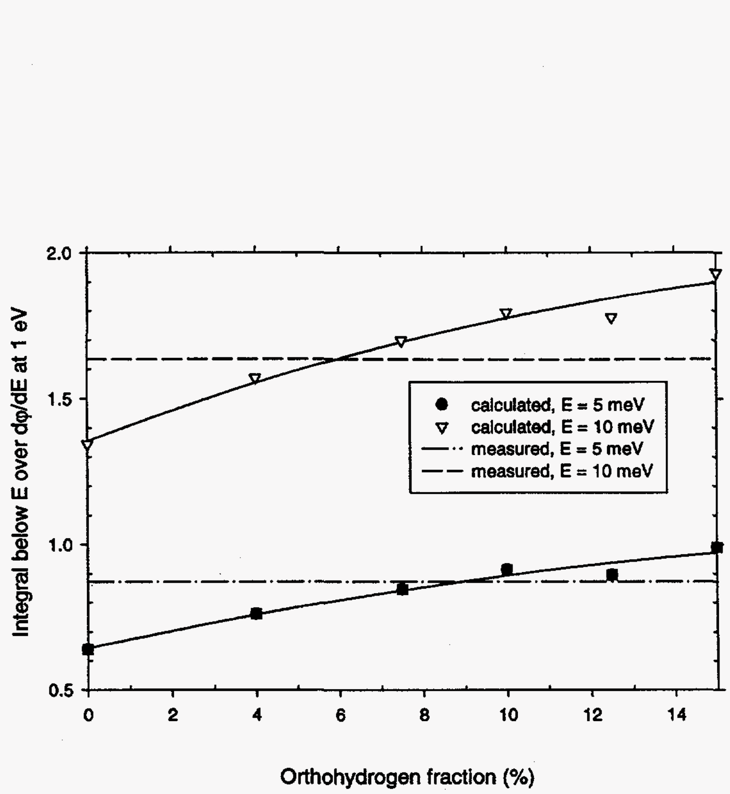


Figure 6. Comparison of the ratio of integrals over the low-energy spectrum (E < 5 and < 10 meV) to the 1 eV flux for Brun's measurement and the Monte Carlo calculations.

Effects of Neutron Apertures (Collimators)

The calculated spectra shown in Fig. 1 employed point detectors viewing the entire ($13x13\text{-cm}^2$) moderator surface. In Brun's measurement, there were collimating apertures that considerably reduced the moderator field-of-view. The estimated moderator field-of-view for his measurements was 9.34 cm^2. [5,6] Our ultimate goal is to compare calculated predictions of the neutron leakage spectrum from the Lujan cold source with measured data on an "absolute" basis. Therefore, for some of our Monte Carlo calculations, we included two collimating apertures (shown in Fig. 7) that were used in Brun's measurement. However, we assumed the apertures were neutronically "black".

To illustrate the effects of these apertures on the calculated neutron flux at the "detector" position, we placed point detectors at 865 cm from the moderator viewed surface (Brun's neutron detector location) as a function of x-z positions. The positions x=0 cm and z=0 cm correspond to the center of the liquid H_2 moderator. The z-axis is in the direction of the proton beam, and the x-axis is in the horizontal direction when viewing the moderator. The results of these calculations are shown in Figs. 8 and 9. Both distributions are relatively flat to about a position of ±0.35 cm and fall to zero at about ±0.5 cm.

In Brun's measurement, he essentially counted all the neutrons passing through the final aperture in front of his detector. However, his detector was some 26 cm from the last collimating aperture, causing the "effective" area of his detector to be larger than the area of the aperture closest to the detector. In order to compare our point detector data directly with Brun's measurement, we need to integrate several x-z brightness distributions at the detector location to obtain the total number of neutrons at that location. These distributions will also be energy dependent. This work is in progress. Also, in our "aperture calculations", we assumed that the apertures were "black" to neutrons. We know that this is not so, and are in the process of including the entire collimation system used in Brun's measurement into our Monte Carlo simulation model. These details must be attended to before comparing calculated predictions to measured data on an "absolute" basis.

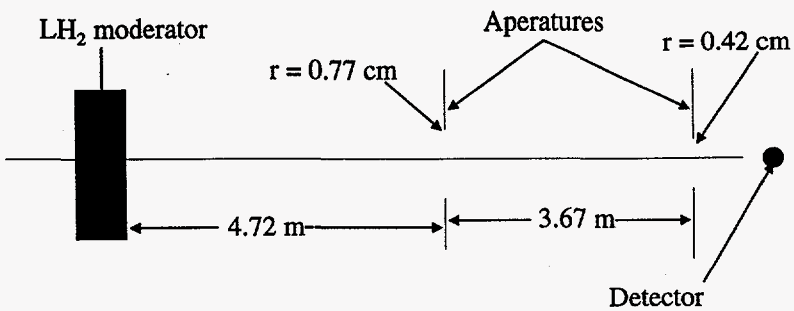


Figure 7. Simplified schematic of Brun's experimental set-up.

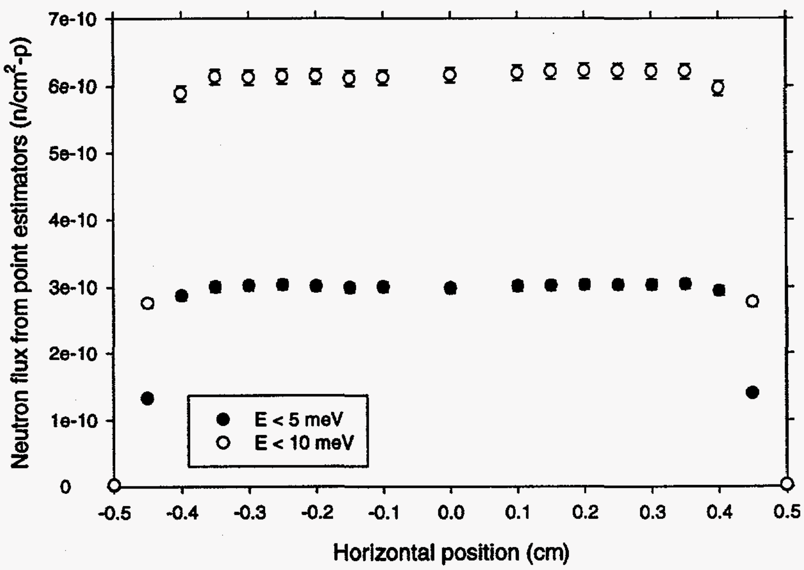


Figure 8. Horizontal neutron flux profile calculated at the position of Brun's detector assuming neutronically "black" apertures.

Specific Moderator brightness

In a previous study[12], we have shown that the neutronic brightness of neutrons leaking from a liquid H_2 moderator depends on the size of the moderator field-of-view. That is, there is an "average" moderator brightness (over the entire moderator surface) and a "specific" moderator brightness (which depends on the size of the moderator field-of-view). Figure 10 shows specific moderator brightness versus field-of-view for both decoupled and coupled liquid H_2 flux-trap moderators. Because of "edge effects" (changes in the spatial distribution of leakage neutrons), the specific moderator brightness of a decoupled moderator is more sensitive to moderator field-of-view than is a coupled moderator. Also, the moderator, whether coupled or decoupled, is "colder" at the center, i.e., at smaller fields-of-view.

Figure 10 illustrates the need to be careful when extrapolating measured "specific" moderator neutronic brightness (as in the case of Brun's measurement) to "average" moderator brightness over a larger area. There are also "aperture" effects that must be considered when scaling specific moderator brightness to average moderator brightness. For example, for Brun's measurement conditions, we estimate a scaling factor of ~11.7 be applied to scale his measured data for his 9.34 cm^2 (circular) field-of-view to a 144 cm^2 (square) field-of-view. To obtain this scaling factor we computed (for each field-of-view) the neutrons leaking from the moderator surface into 0.5 sr with

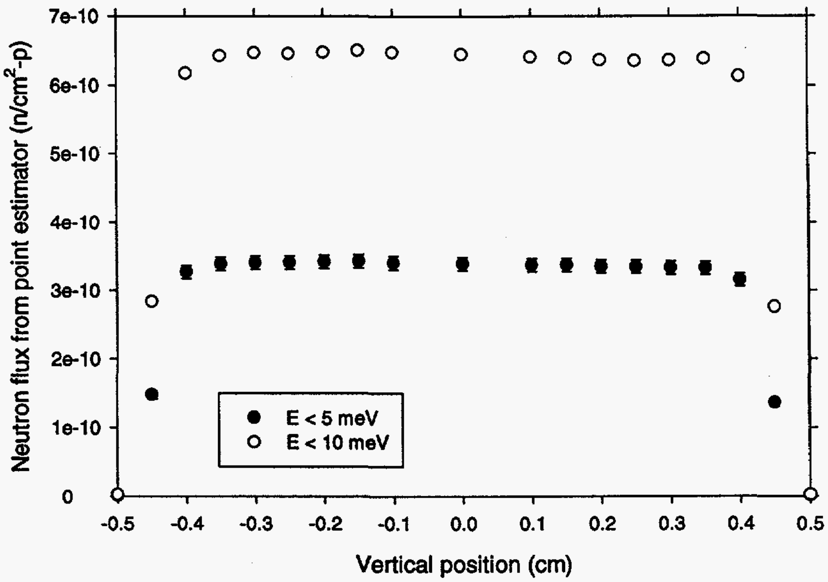


Figure 9. Vertical neutron flux profile calculated at the position of Brun's detector assuming neutronically "black" apertures.

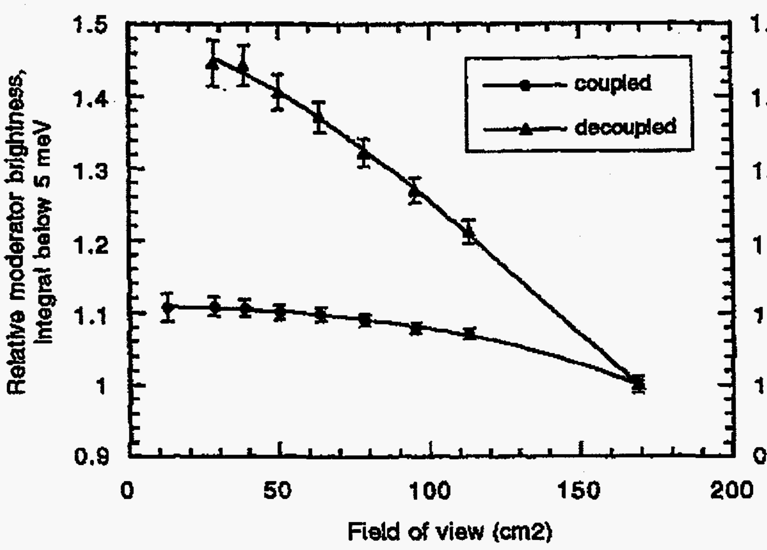


Figure 10. Specific brightness versus moderator field-of-view for decoupled and coupled liquid H_2 flux-trap moderators (ortho/para H_2 concentration of 50/50 %)

E < 10 meV. (Note that this intensity increase does not scale as the ratio of the areas associated with the respective field-of-view; this ratio would be 15.4.) Also, the scaling factor of ~11.7 does not account for any "aperture" effects.

Conclusions

The ortho/para H_2 concentration in a liquid H_2 moderator affects the neutronic performance of the moderator. We need a method to directly measure the ortho/para H_2 concentration in an operating liquid H_2 moderator at a spallation neutron source. We have estimated the ortho-hydrogen concentration in the Lujan cold source to be in 6-9% at the time when Brun made his neutron spectrum measurement from the cold source. When trying to compare "absolute" calculated predictions of neutron leakage spectra to measured data for the Lujan Center liquid H_2 moderator, we find that it is essential: a) to have a detailed Monte Carlo model of the target system; b) to know the proton beam parameters accurately; and c) and to pay attention to experimental details, including collimation systems. We are continuing to calculate precisely the quantity measured by Brun (n/p at the detector location) so that we can compare calculated predictions to measured data on an "absolute" basis. We must continue to improve and validate the scattering kernels for liquid hydrogen.

Acknowledgments

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