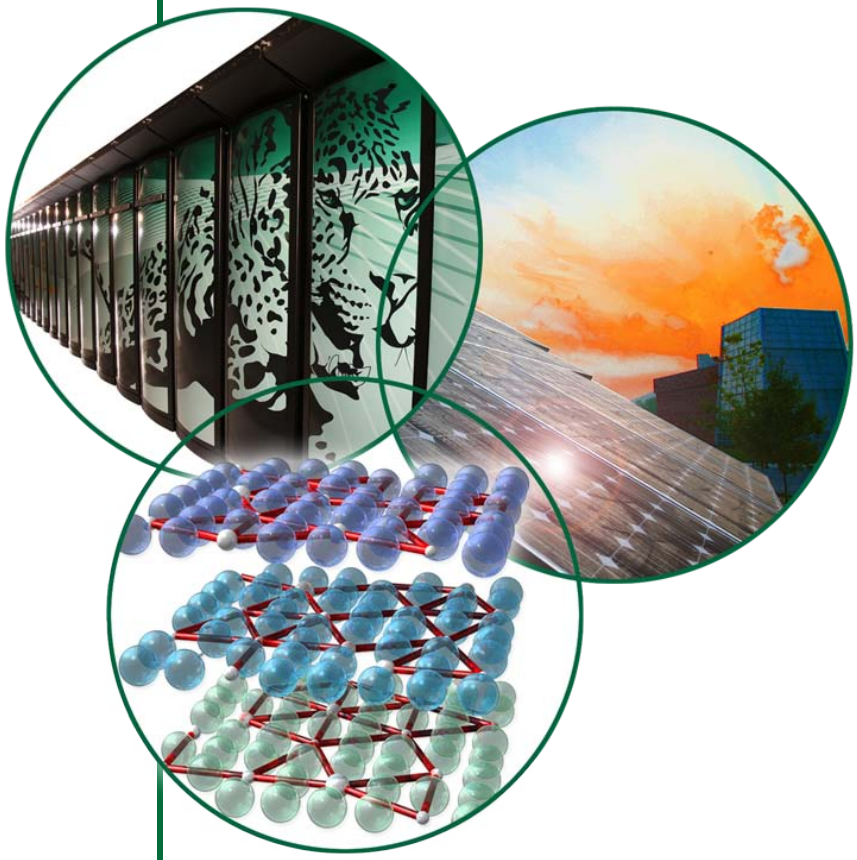
# Identification of Lithium Isotopes Using Time-Tagged Neutron Scattering

**Brandon Grogan, John Mihalczo**

**Oak Ridge National Laboratory**



**53rd Annual Meeting of the INMM**

**Orlando, FL**

**July 17th, 2012**

U.S. DEPARTMENT OF ENERGY

**This research is supported by the US Department of Energy under Contract No. DE-AC05-00OR22725 with UT-Battelle, LLC**

OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE DEPARTMENT OF ENERGY

# Background

- Lithium enriched in the $^{6}Li$ isotope is used for tritium production and as a component in nuclear weapons
- Natural lithium contains only 7.5% of the $^{6}Li$ isotope; the other 92.5% is $^{7}Li$
- The lithium is typically in a hydride form, with the lithium bound to either light hydrogen ($^{1}H$) or deuterium ($^{2}H$)
- The U.S. has had no lithium enrichment capability since 1963, so it is essential that existing stocks be properly characterized

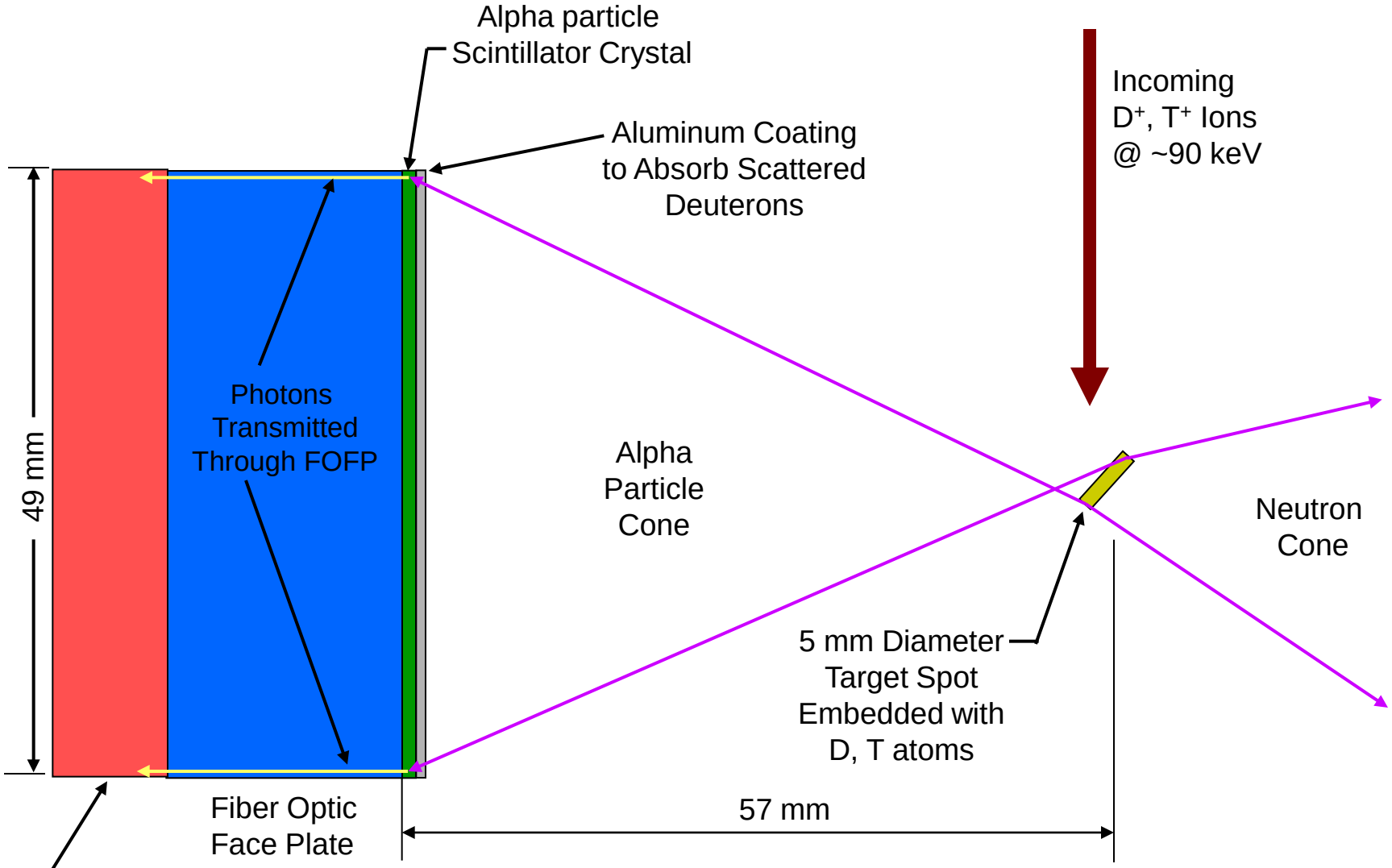
# Purpose

- Traditional destructive assay techniques can determine the isotopics, but the procedure can be time-consuming and hazardous since Li salts tend to be highly reactive.
- This work will demonstrate how time-tagged neutron scattering measurements can distinguish the Li and H isotopes non-intrusively.
- Two primary techniques:
  - Neutron time of flight
  - Active gamma spectroscopy

# API D-T Neutron Generator

- A deuterium-tritium neutron generator produces 14.1 MeV neutrons via the $^{3}H(^{2}H,n)^{4}He$ reaction
- An integral alpha particle detector time-tags and electronically collimates the neutrons (the associated particle technique)
- The neutron start time is recorded with ± 1 ns accuracy
- Because the neutrons are monoenergetic, their time of flight to an object at a fixed distance is easily calculated

# Time Tagging D-T Neutrons




$^{2}H + {}^{3}H \rightarrow {}^{4}He + n + 17.6$ MeV

# Neutron Time of Flight

# Neutron Scattering

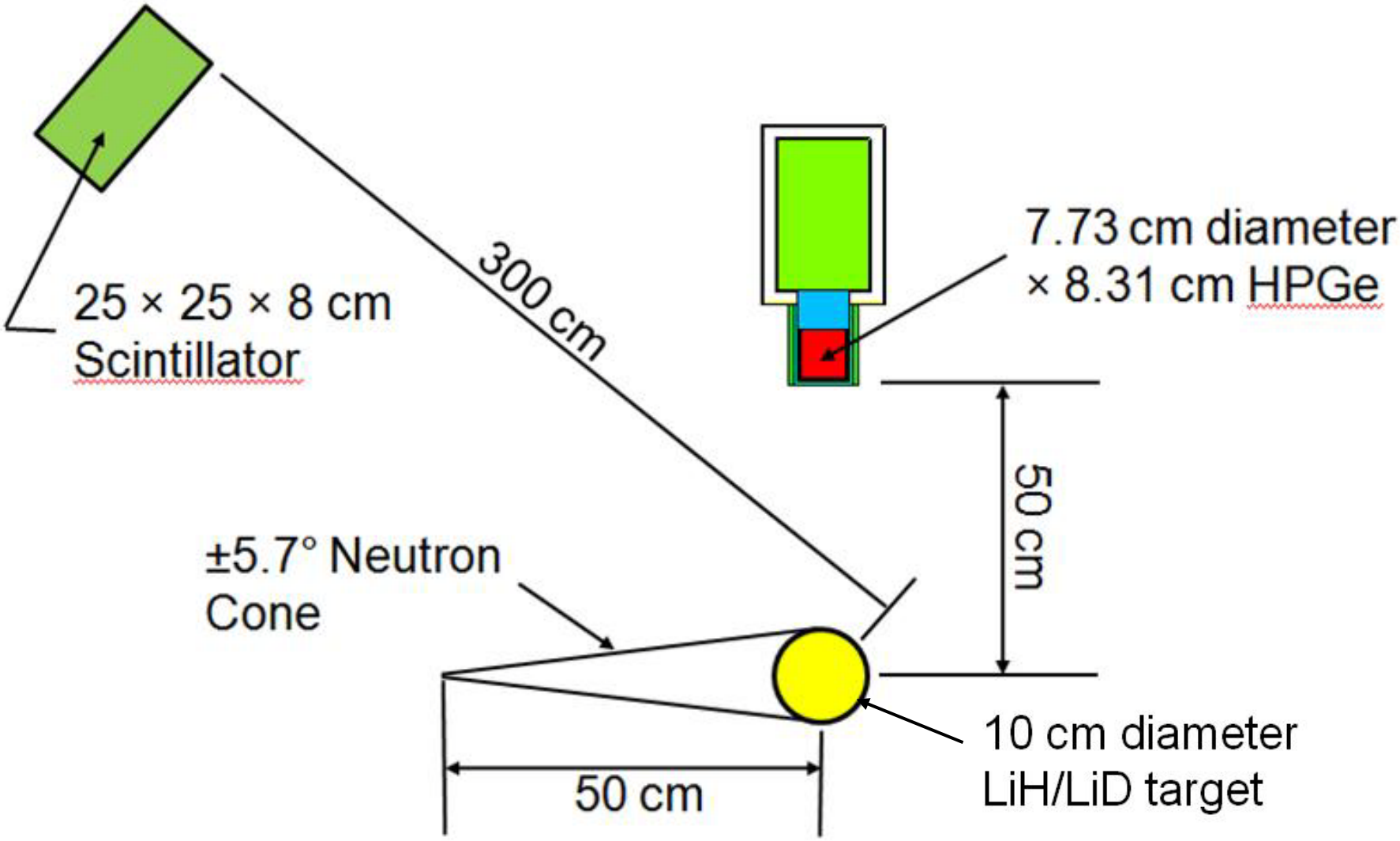
- Fast neutrons interact with light isotopes primarily through elastic and inelastic scattering
- Elastic scattering conserves kinetic energy in a billiard-ball collision
- Inelastic scattering transfers some of the kinetic energy to the target nucleus, leaving it in an excited state
  - The excited state produces characteristic gammas when it returns to the ground state
- For a single collision, the initial neutron energy, target nucleus mass, energy level, and scattering angle exactly determines the final energy

# Elastic/Inelastic Scattering Time of Flight: 50 cm source-to-target; 300 cm to detector

| Mass Number (Q Value) | t @ 45° (ns) | t @ 90° (ns) | t @ 135° (ns) | t @ 180° (ns) |
|---|---|---|---|---|
| 1 | 91.9 | -- | -- | -- |
| 2 | 77.5 | 110.2 | 158.9 | 183.2 |
| 7 (-4630 keV) | 84.8 | 95.1 | 106.8 | 112.0 |
| 6 (-3562 keV) | 81.2 | 92.1 | 104.6 | 110.2 |
| 6 (-2186 keV) | 76.7 | 86.0 | 96.6 | 101.4 |
| 6 | 71.0 | 78.6 | 87.2 | 91.1 |
| 7 (-478 keV) | 71.7 | 78.3 | 85.7 | 89.0 |
| 7 | 70.6 | 77.0 | 84.1 | 87.2 |
| 16 | 69.2 | 71.8 | 74.6 | 75.8 |
| 40 | 68.6 | 69.6 | 70.6 | 71.1 |
| 100 | 68.3 | 68.7 | 69.1 | 69.3 |
| 208 | 68.2 | 68.4 | 68.6 | 68.7 |

$$E_f = \frac{1}{(A+1)^2}\left[\cos(\theta_L)\sqrt{E_i} + \sqrt{E_i\left(\cos^2(\theta_L) + A^2 - 1\right) + A(A+1)Q}\right]^2$$

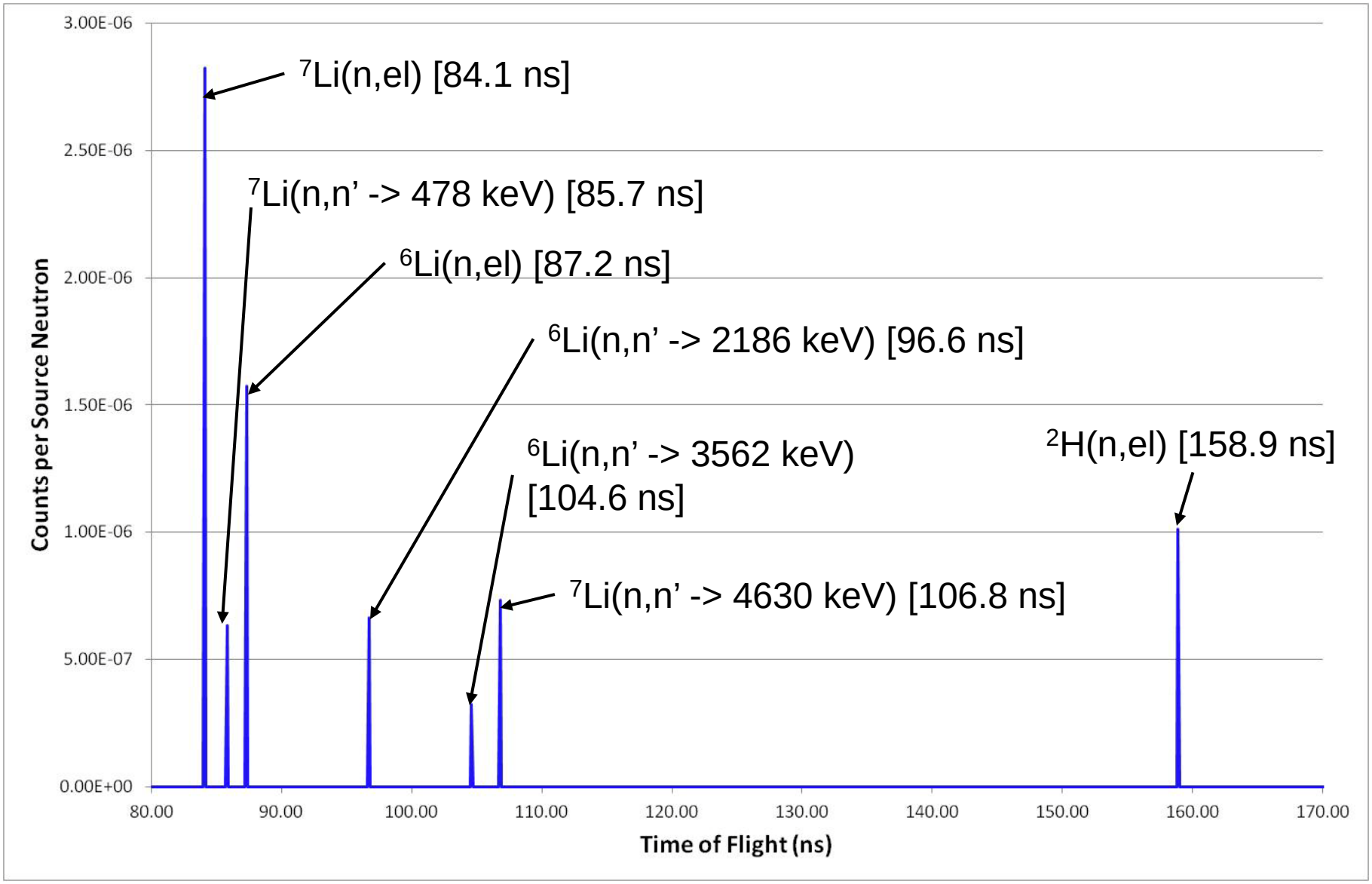
# Modeled Neutron Scattering Geometry



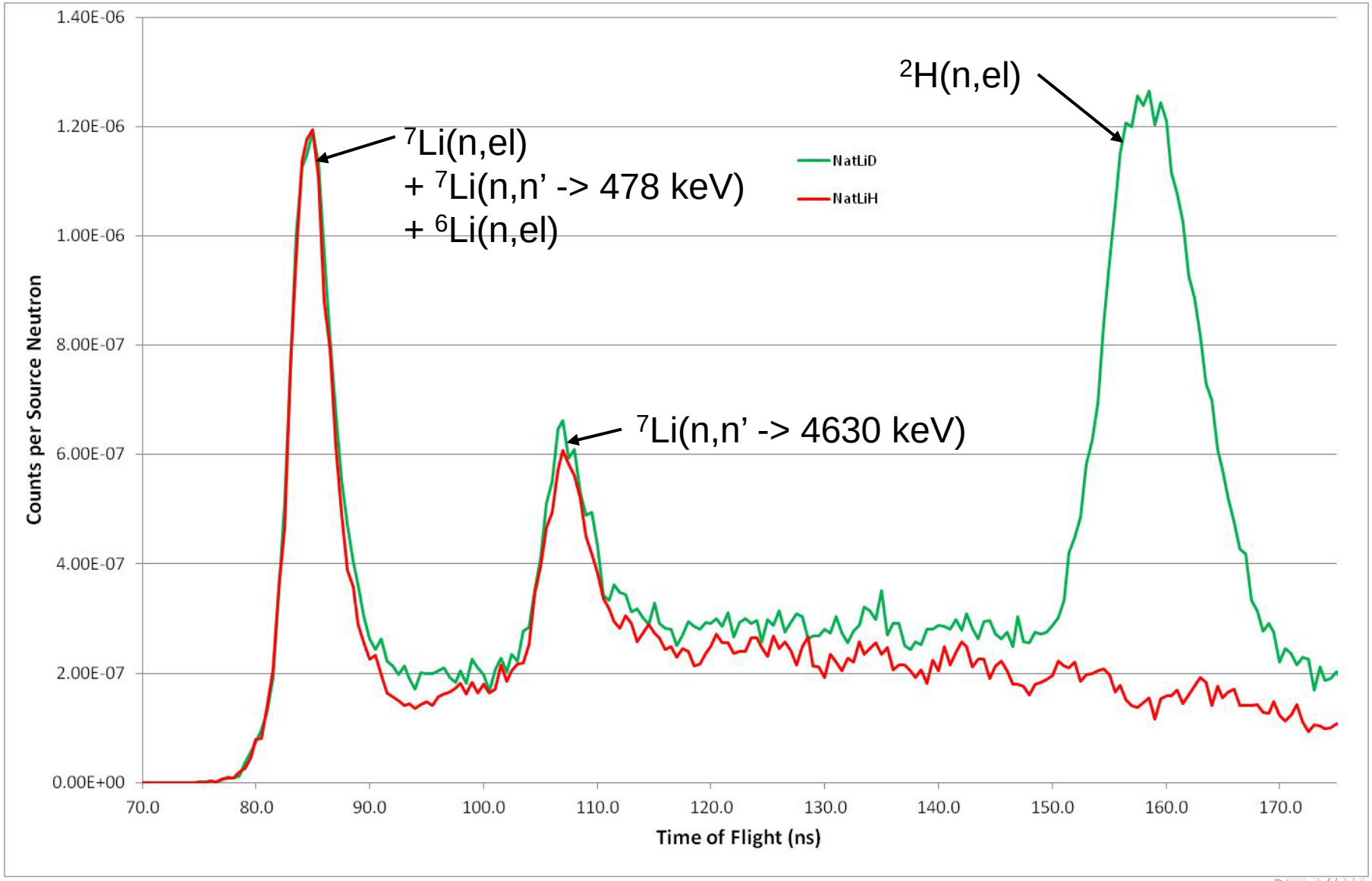
# Simulation Procedures

- Neutron time-of-flight measurements were simulated using MCNPX-PoliMi v 1.4.3
- Active gamma spectroscopy simulations used MCNPX v 2.6.0 with an F8 GEB tally to simulate HPGe resolution
- Each simulation represents a two-hour measurement time with a $10^8$ n/s generator
- $^6$Li enrichments of 0%, 7.5%, 20%, 40%, 60%, 80%, and 100% were tested.
- For natural (7.5%) Li, both LiH and LiD were tested

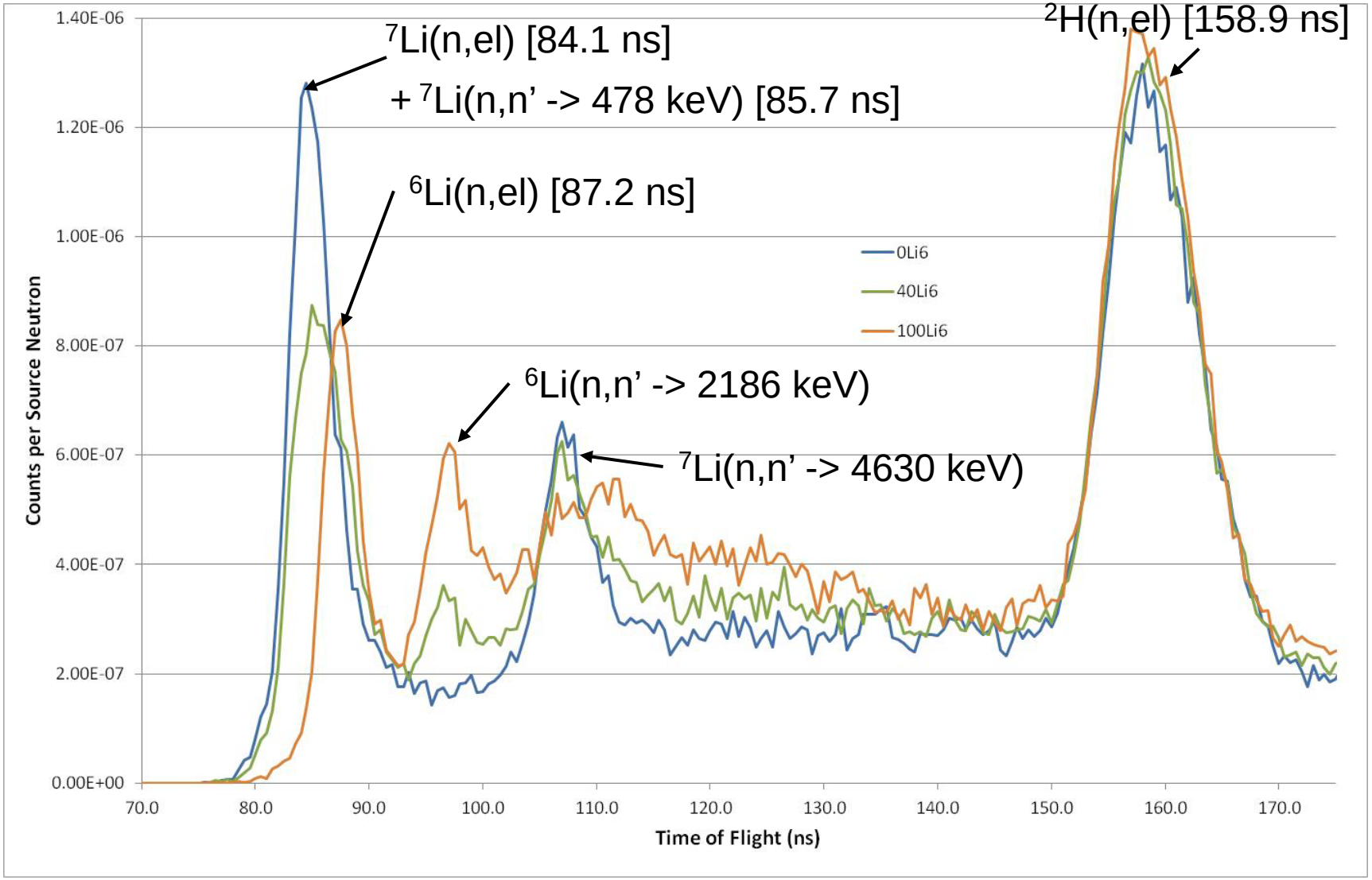
# Ideal Neutron Scattering Time of Flight



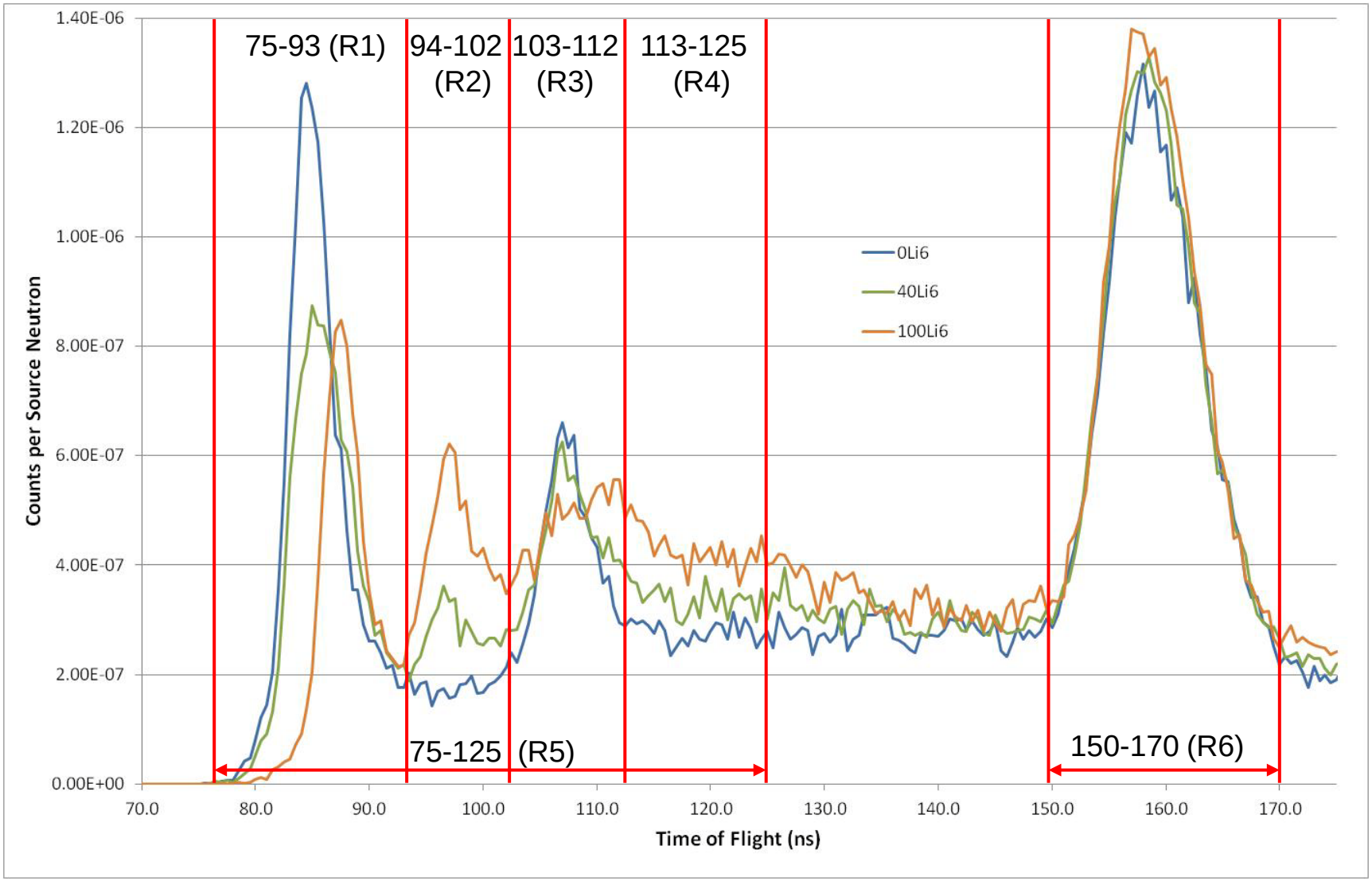
# Natural (7.5% $^{6}$Li) LiH/LiD Time of Flight



# LiD Time of Flight vs $^{6}$Li Enrichment



# Neutron Time of Flight Regions of Interest



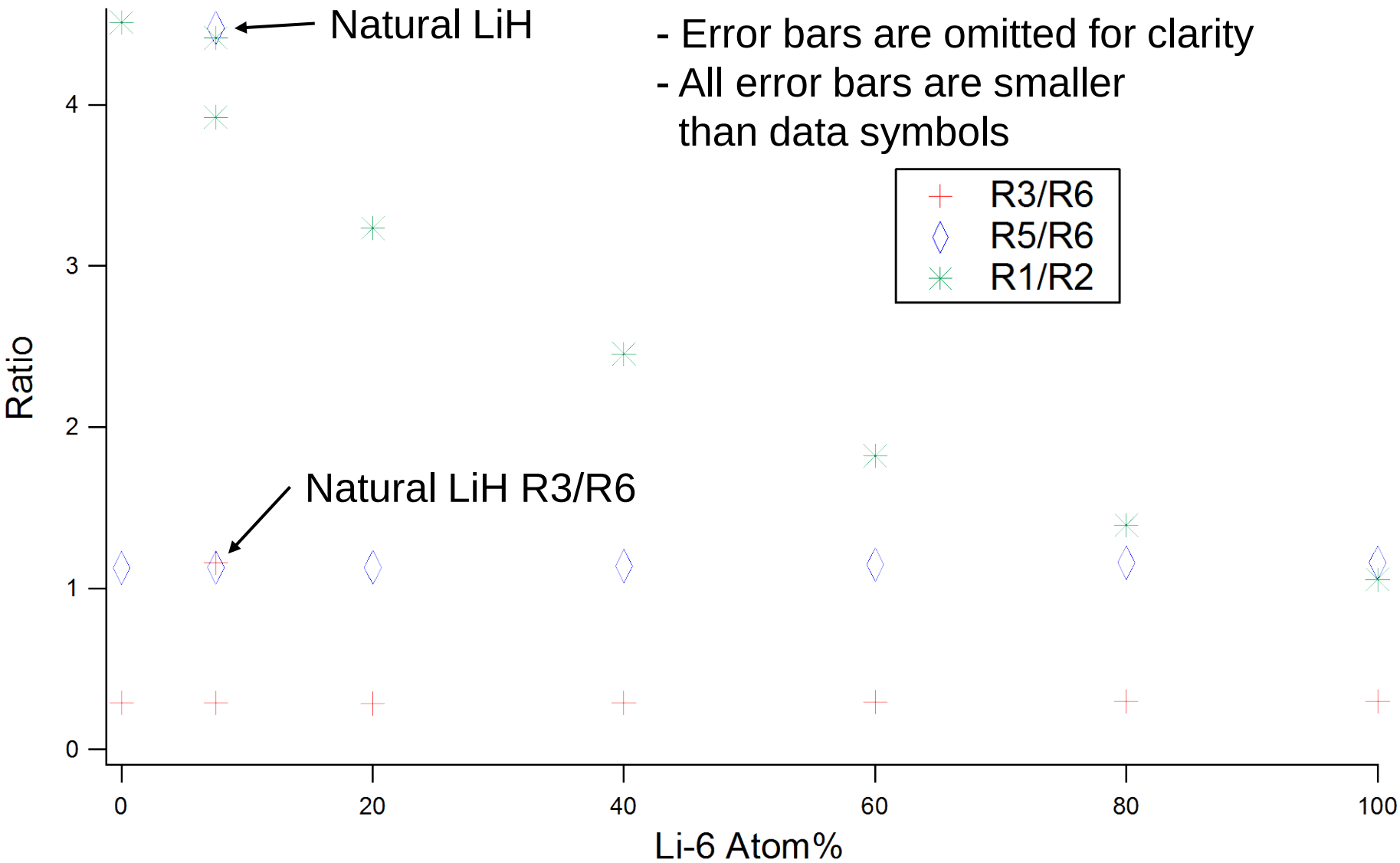
# Regions of Interest and ROI Ratios

ROI Values are detected counts per $10^6$ source neutrons

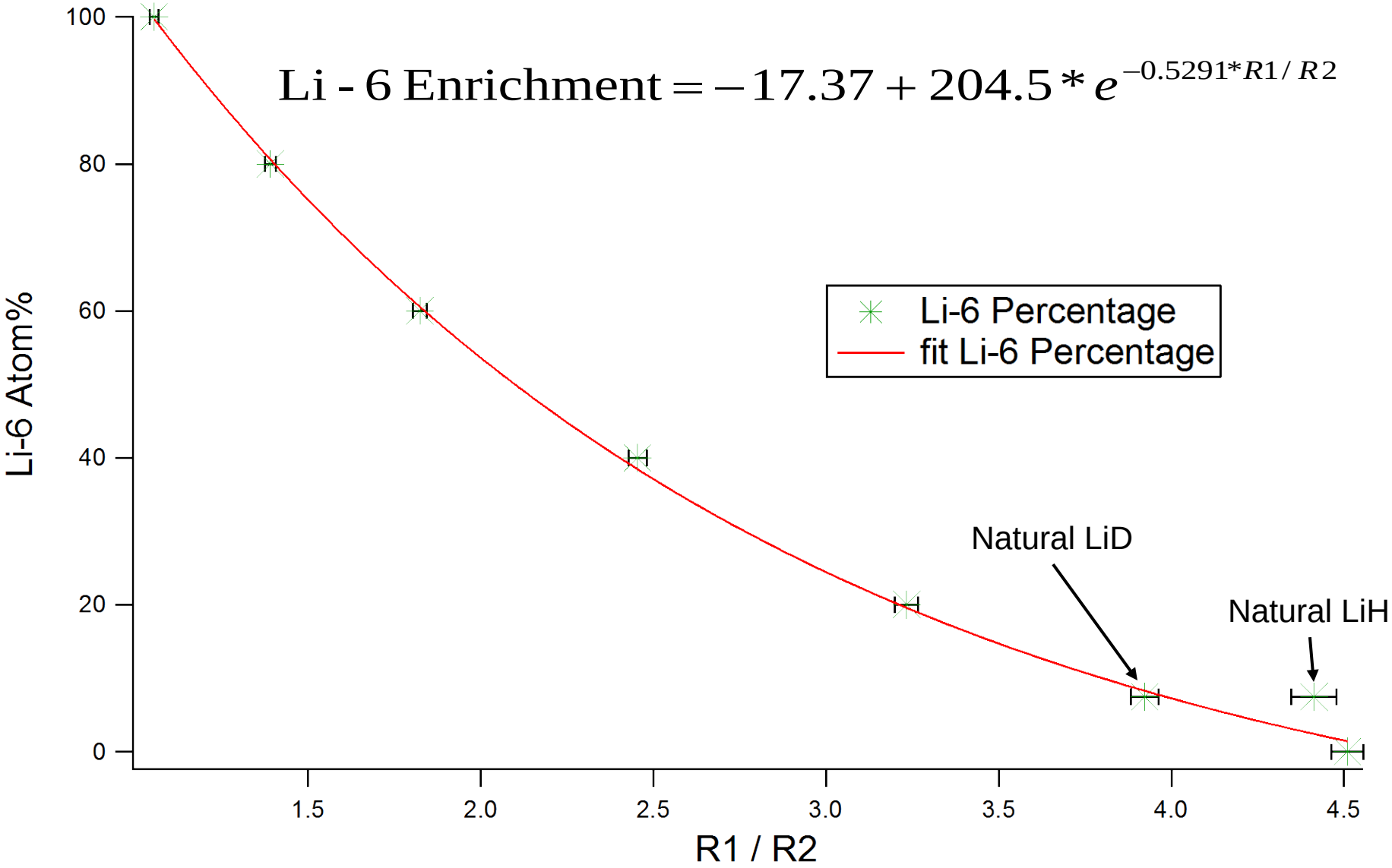
| Region | Time Range | 0Li6 | 20Li6 | 40Li6 | 60Li6 | 80Li6 | 100Li6 | NatLiD | NatLiH |
|---|---|---|---|---|---|---|---|---|---|
| R1 | 75.0-93.0 | 14.50 | 13.34 | 12.21 | 10.96 | 9.67 | 8.38 | 14.01 | 13.13 |
| R2 | 93.5-102.0 | 3.22 | 4.13 | 4.98 | 6.01 | 6.95 | 7.95 | 3.57 | 2.97 |
| R3 | 102.5-112.0 | 8.63 | 8.74 | 8.96 | 9.11 | 9.39 | 9.59 | 8.69 | 7.93 |
| R4 | 112.5-125.0 | 7.23 | 8.03 | 8.78 | 9.59 | 10.38 | 11.16 | 7.62 | 6.58 |
| R5 | 75-125.0 | 33.57 | 34.23 | 34.92 | 35.67 | 36.39 | 37.07 | 33.89 | 30.61 |
| R6 | 150.0-170.0 | 29.82 | 30.33 | 30.67 | 31.09 | 31.37 | 31.95 | 30.02 | 6.84 |

| Ratio | 0Li6 | 20Li6 | 40Li6 | 60Li6 | 80Li6 | 100Li6 | NatLiD | NatLiH |
|---|---|---|---|---|---|---|---|---|
| R1/R6 | 0.49 | 0.44 | 0.40 | 0.35 | 0.31 | 0.26 | 0.47 | 1.92 |
| R2/R6 | 0.11 | 0.14 | 0.16 | 0.19 | 0.22 | 0.25 | 0.12 | 0.43 |
| R3/R6 | 0.29 | 0.29 | 0.29 | 0.29 | 0.30 | 0.30 | 0.29 | 1.16 |
| R4/R6 | 0.24 | 0.26 | 0.29 | 0.31 | 0.33 | 0.35 | 0.25 | 0.96 |
| R5/R6 | 1.13 | 1.13 | 1.14 | 1.15 | 1.16 | 1.16 | 1.13 | 4.47 |
| R1/R2 | 4.51 | 3.23 | 2.45 | 1.82 | 1.39 | 1.05 | 3.92 | 4.41 |
| R1/R3 | 1.68 | 1.53 | 1.36 | 1.20 | 1.03 | 0.87 | 1.61 | 1.66 |
| R1/R4 | 2.01 | 1.66 | 1.39 | 1.14 | 0.93 | 0.75 | 1.84 | 1.99 |
| R1/R5 | 0.43 | 0.39 | 0.35 | 0.31 | 0.27 | 0.23 | 0.41 | 0.43 |

# Selected ROI Ratios vs $^{6}$Li Enrichment

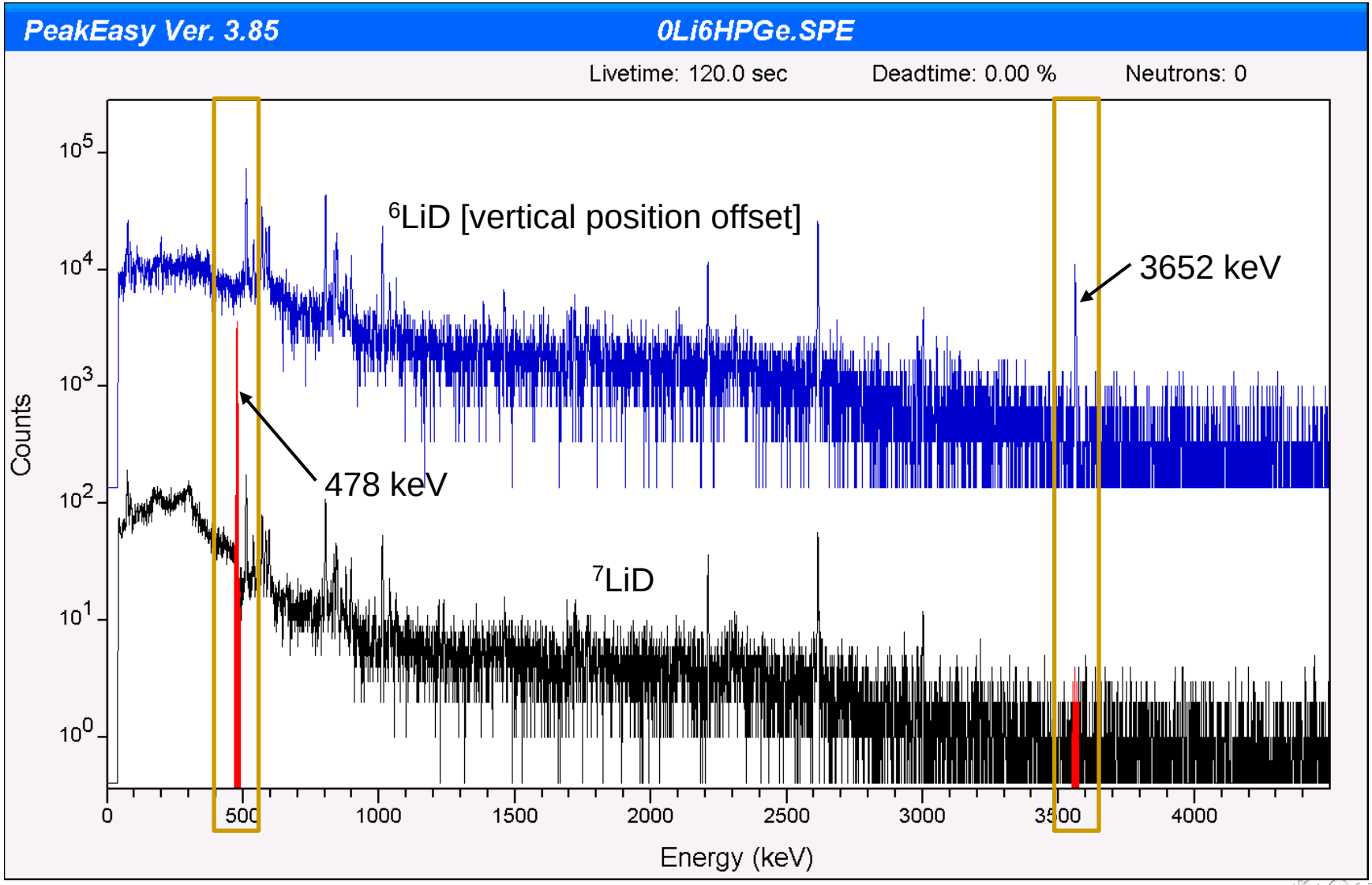


# Fit of R1/R2 Ratio vs $^{6}$Li Enrichment

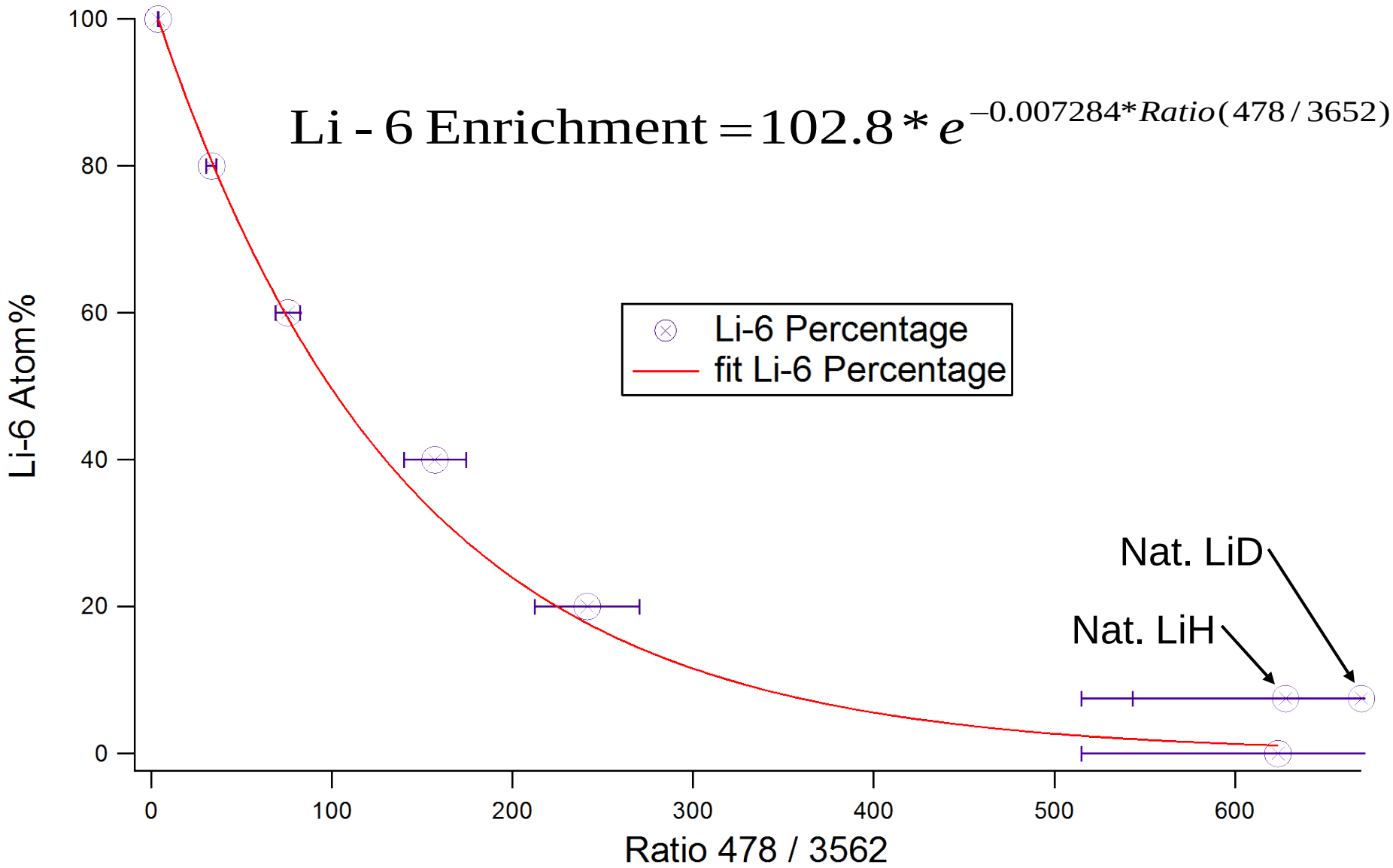


# Active Gamma Spectroscopy

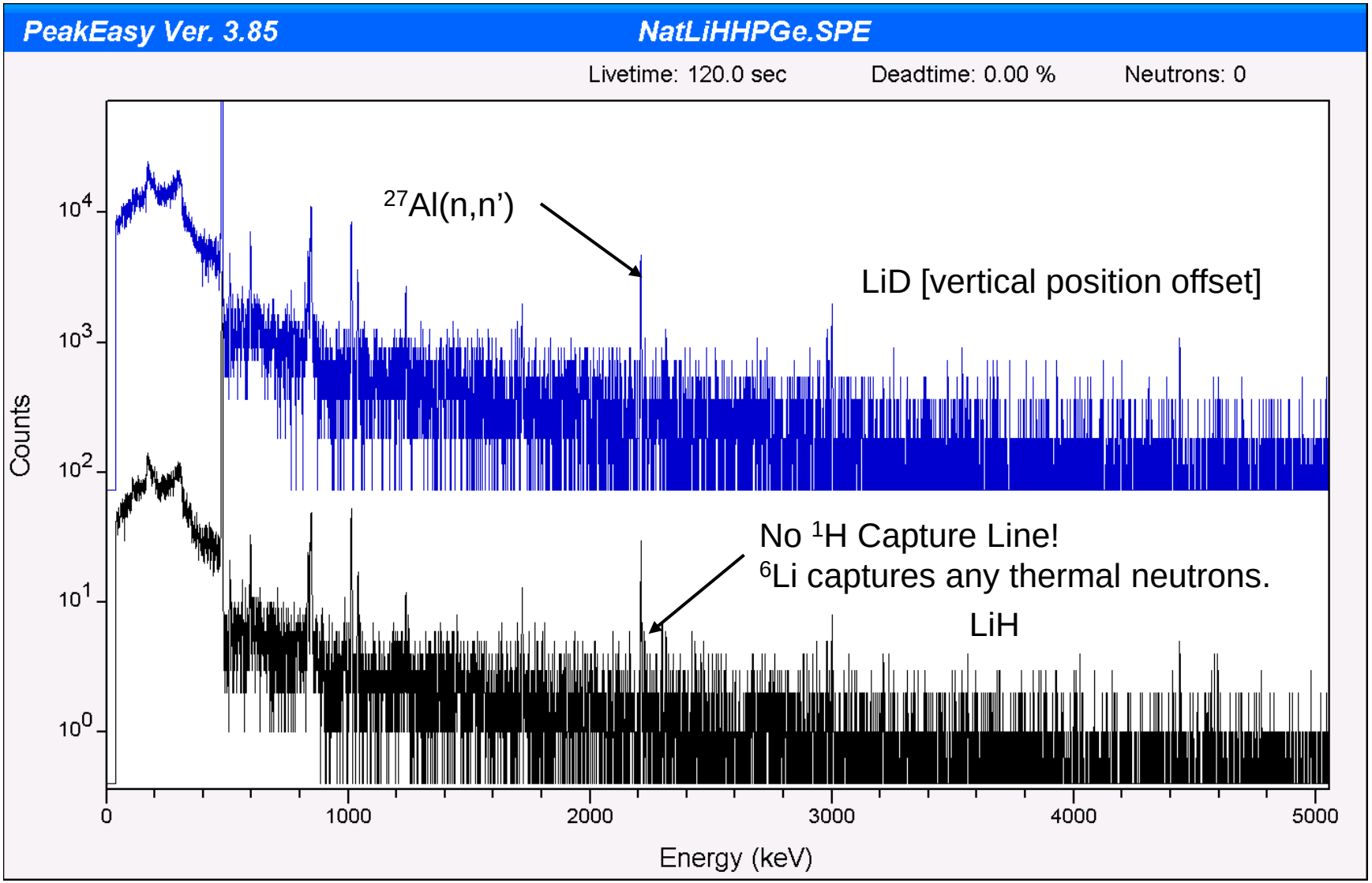
# Comparison of $^{6}$LiD and $^{7}$LiD Gamma Spectra



# Fit of $^{7}Li/^{6}Li$ Gamma Peak Ratios



# Comparison of LiH and LiD Gamma Spectra



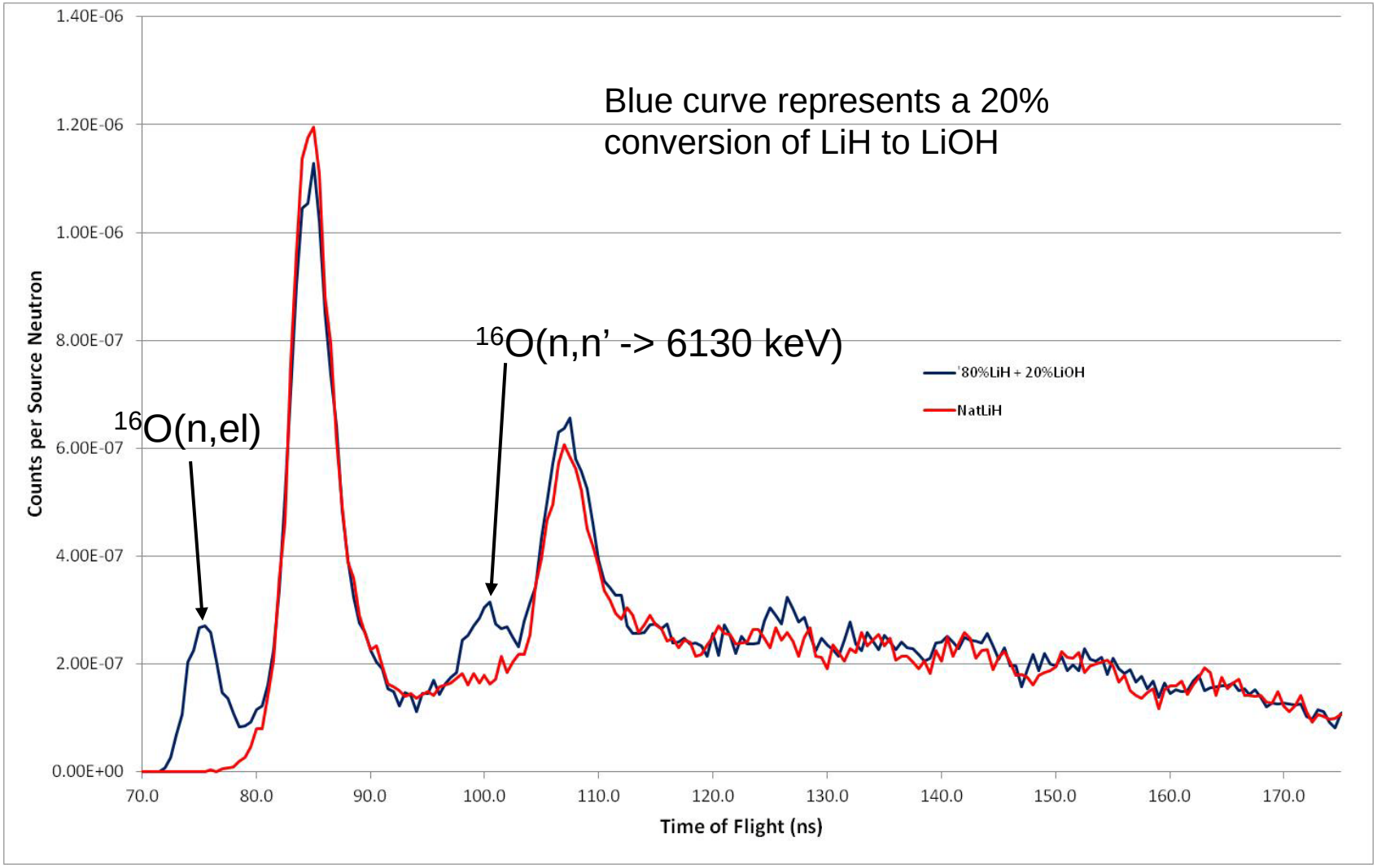
# Conclusions

- These simulations have shown that time-tagged neutron scattering can effectively characterize a light material such as LiH/LiD.
- The $^{6}$Li enrichment can be measured using scattered neutron time of flight or gamma spectroscopy
- Gamma spectroscopy is inaccurate at $^{6}$Li enrichments below 20%
- Deuterium is easily distinguishable from light hydrogen using the neutron time of flight, but not by gamma spectroscopy

# Questions?

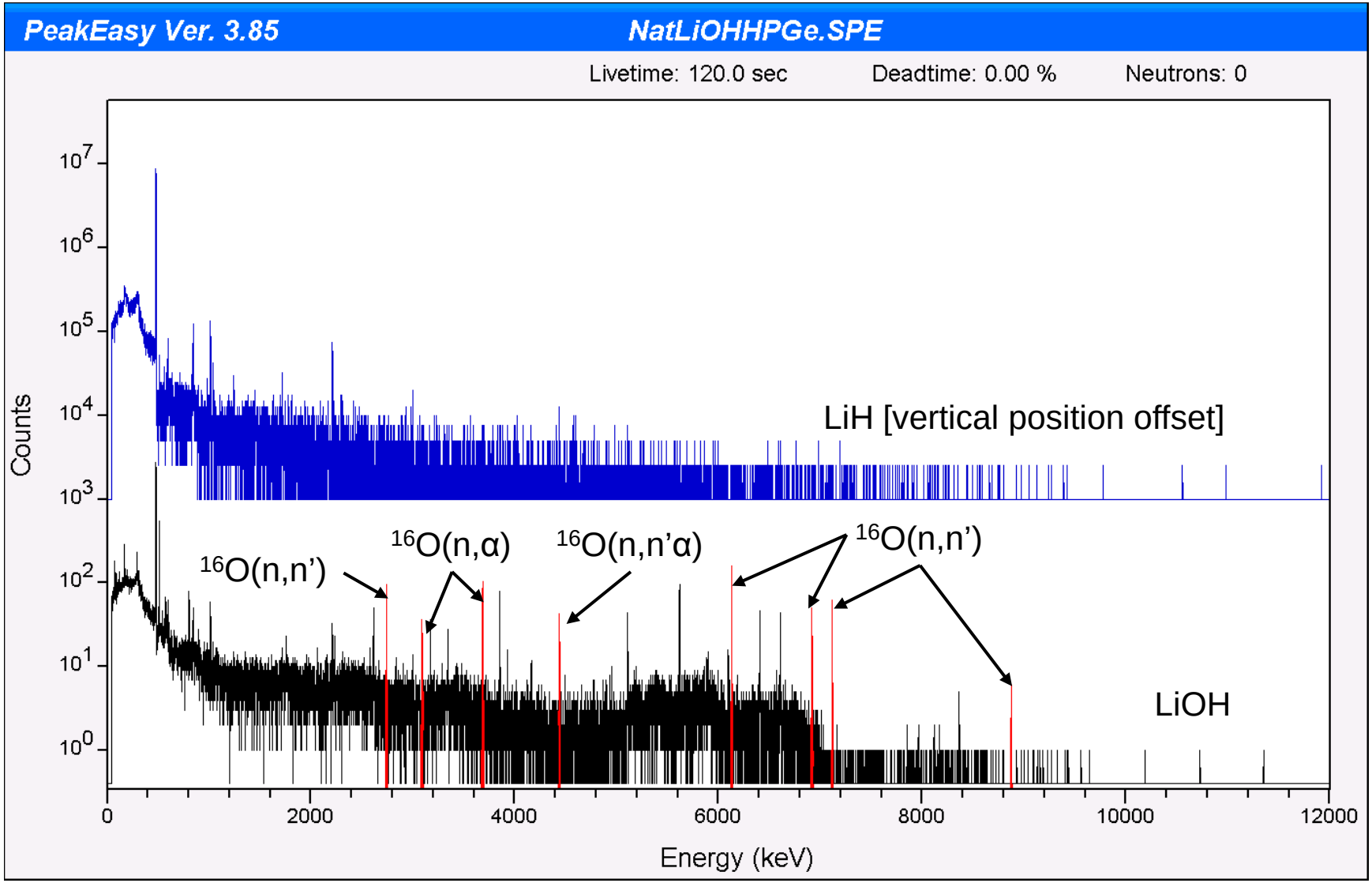
# Backup Slides

# Effect of LiH → LiOH Oxidation

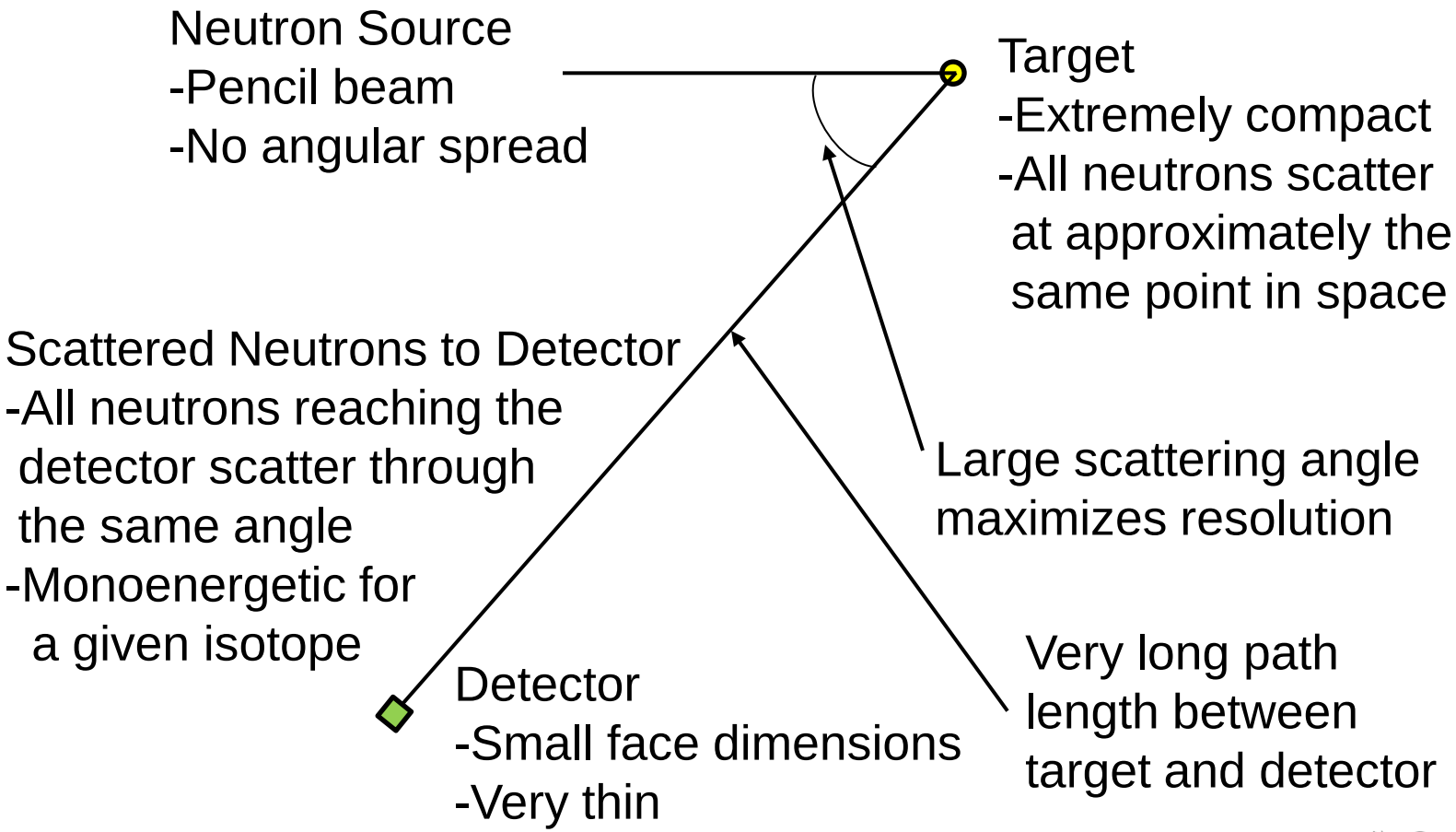



$LiH + H_2O \rightarrow LiOH + H_2$ (Highly Exothermic)

# Comparison of LiH and LiOH Gamma Spectra



# Ideal Neutron Scattering Geometry



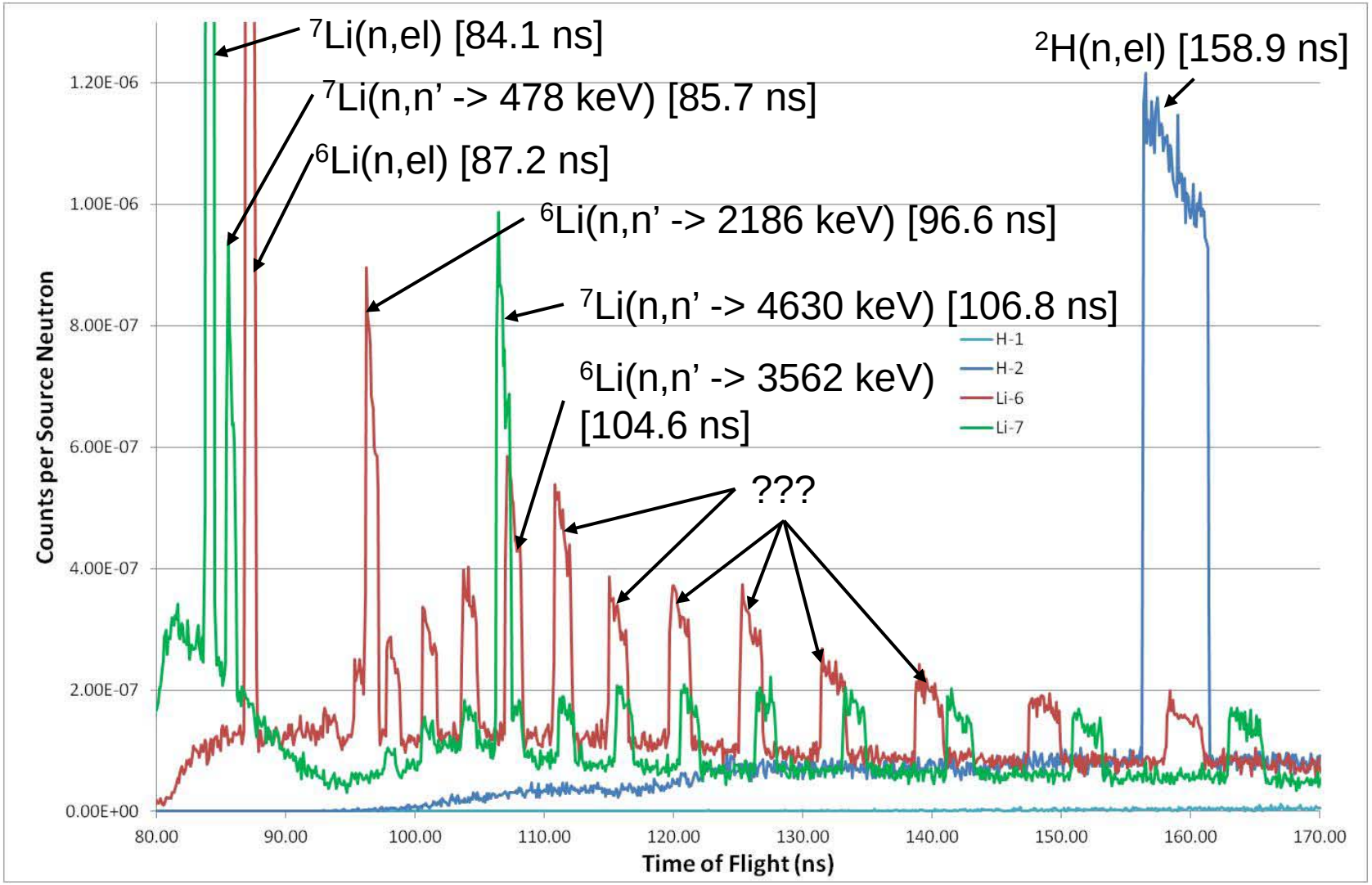
# Timing Uncertainty in Modeled Geometry

- Uncertainty — Source of Uncertainty
- $\sigma_\Phi = \pm 0.86$ ns — Initial neutron angle
- $\sigma_l = \pm 0.65$ ns — Neutron penetration depth in object
- $\sigma_\theta = \pm 0.48$ ns — Neutron scattering angle
- $\sigma_d = \pm 1.03$ ns — Interaction depth in detector
- $\sigma_t = \pm 0.50$ ns — Time resolution of electronics

$$\sigma_{tot} = \sqrt{\sigma_\phi^2 + \sigma_l^2 + \sigma_\theta^2 + \sigma_d^2 + \sigma_t^2} = \pm 1.64\,\text{ns}$$

This is probably sufficient to resolve the 3.1 ns difference between $^6$Li and $^7$Li elastic scattering

# Near-Ideal Neutron Scattering Time of Flight



# $^{7}$Li Energy Levels and ENDF Values

*Source: National Nuclear Data Center, http://www.nndc.bnl.gov*

| $E_{level}$ (keV) | XREF | Jπ | $T_{1/2}$ | $E_{\gamma}$ (keV) | γ mult. |
|---|---|---|---|---|---|
| 0.0 | AB D FGHIJKLMN PQRS UVWXYZa | 3/2- | STABLE | | |
| 477.612 *3* | A D FG IJKLMN PQRS UVWXYZa | 1/2- | 73 fs *2* | 477.595 | M1(+E2) |
| 4630 *9* | CD FG IJKLMNOP RSTUVW | 7/2- | 93 keV *8*<br>% 3H = ?<br>% α = ? | 4628 | E2 |
| 6680 *50* | C FG IJK NO T VW | 5/2- | 0.88 MeV *+20-10*<br>% 3H = ?<br>% α = ? | | |
| 7459.5 *10* | BC EFGHIJKL NO QRST V | 5/2- | 89 keV *7*<br>% n = ?<br>% 3H = ?<br>% α = ? | | |
| 9.67E+3 *10* | BC G J L N T | 7/2- | ≈ 400 keV<br>% 3H = ?<br>% α = ?<br>% n = ? | | |
| 9850 | E Q | 3/2- | ≈ 1200 keV<br>% n = ?<br>% α = ? | | |

| | | | | |
|---|---|---|---|---|
| 15 | Summary | ENDF-6 | σ | Plot |
| 16 | Summary | ENDF-6 | σ | Plot |
| 17 | Summary | ENDF-6 | σ | Plot |
| 18 | Summary | ENDF-6 | σ | Plot |
| 19 | Summary | ENDF-6 | σ | Plot |
| 20 | Summary | ENDF-6 | σ | Plot |
| 21 | Summary | ENDF-6 | σ | Plot |
| 22 | Summary | ENDF-6 | σ | Plot |
| 23 | Summary | ENDF-6 | σ | Plot |
| 24 | Summary | ENDF-6 | σ | Plot |
| 25 | Summary | ENDF-6 | σ | Plot |
| 26 | Summary | ENDF-6 | σ | Plot |
| 27 | Summary | ENDF-6 | σ | Plot |
| 28 | Summary | ENDF-6 | σ | Plot |
| 29 | Summary | ENDF-6 | σ | Plot |
| 30 | Summary | ENDF-6 | σ | Plot |
| 31 | Summary | ENDF-6 | σ | Plot |
| 32 | Summary | ENDF-6 | σ | Plot |

```
LI-7(N,N`)LI-7-L1,SIG     MT51  QM=0  QI=-477610  LR=0
LI-7(N,N`)LI-7-L2(T)HE-4,SIG MT52  QM=-2.4667e+6  QI=-2.75e+6  LR=33
LI-7(N,N`)LI-7-L3(T)HE-4,SIG MT53  QM=-2.4667e+6  QI=-3.25e+6  LR=33
LI-7(N,N`)LI-7-L4(T)HE-4,SIG MT54  QM=-2.4667e+6  QI=-3.75e+6  LR=33
LI-7(N,N`)LI-7-L5(T)HE-4,SIG MT55  QM=-2.4667e+6  QI=-4.25e+6  LR=33
LI-7(N,N`)LI-7-L6(T)HE-4,SIG MT56  QM=-2.4667e+6  QI=-4.63e+6  LR=33
LI-7(N,N`)LI-7-L7(T)HE-4,SIG MT57  QM=-2.4667e+6  QI=-4.75e+6  LR=33
LI-7(N,N`)LI-7-L8(T)HE-4,SIG MT58  QM=-2.4667e+6  QI=-5.25e+6  LR=33
LI-7(N,N`)LI-7-L9(T)HE-4,SIG MT59  QM=-2.4667e+6  QI=-5.75e+6  LR=33
LI-7(N,N`)LI-7-L10(T)HE-4,SIG MT60  QM=-2.4667e+6  QI=-6.25e+6  LR=33
LI-7(N,N`)LI-7-L11(T)HE-4,SIG MT61  QM=-2.4667e+6  QI=-6.75e+6  LR=33
LI-7(N,N`)LI-7-L12(T)HE-4,SIG MT62  QM=-2.4667e+6  QI=-7.25e+6  LR=33
LI-7(N,N`)LI-7-L13(T)HE-4,SIG MT63  QM=-2.4667e+6  QI=-7.75e+6  LR=33
LI-7(N,N`)LI-7-L14(T)HE-4,SIG MT64  QM=-2.4667e+6  QI=-8.25e+6  LR=33
LI-7(N,N`)LI-7-L15(T)HE-4,SIG MT65  QM=-2.4667e+6  QI=-8.75e+6  LR=33
LI-7(N,N`)LI-7-L16(T)HE-4,SIG MT66  QM=-2.4667e+6  QI=-9.25e+6  LR=33
LI-7(N,N`)LI-7-L17(T)HE-4,SIG MT67  QM=-2.4667e+6  QI=-9.75e+6  LR=33
LI-7(N,N`)LI-7-L18(T)HE-4,SIG MT68  QM=-2.4667e+6  QI=-1.025e+7  LR=33
LI-7(N,N`)LI-7-L19(T)HE-4,SIG MT69  QM=-2.4667e+6  QI=-1.075e+7  LR=33
LI-7(N,N`)LI-7-L20(T)HE-4,SIG MT70  QM=-2.4667e+6  QI=-1.125e+7  LR=33
LI-7(N,N`)LI-7-L21(T)HE-4,SIG MT71  QM=-2.4667e+6  QI=-1.175e+7  LR=33
LI-7(N,N`)LI-7-L22(T)HE-4,SIG MT72  QM=-2.4667e+6  QI=-1.225e+7  LR=33
LI-7(N,N`)LI-7-L23(T)HE-4,SIG MT73  QM=-2.4667e+6  QI=-1.275e+7  LR=33
LI-7(N,N`)LI-7-L24(T)HE-4,SIG MT74  QM=-2.4667e+6  QI=-1.325e+7  LR=33
LI-7(N,N`)LI-7-L25(T)HE-4,SIG MT75  QM=-2.4667e+6  QI=-1.375e+7  LR=33
```

The ENDF/B-VII file shows a uniform 500 keV spacing between energy levels that is not reflected in the actual energy levels of $^{7}$Li (and $^{6}$Li)

These pseudo-energy levels may be holdouts from a time when computers were much slower than today