

Probing Nuclear Structure with Neutrons

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Association for Research at University Nuclear Accelerators







Fig. 4-5 Excitation functions for proton-induced reactions on ⁶³Cu. [From J. W. Meadows, *Phys. Rev.* 91, 885 (1953).]







Neutrons

| Neutron energy | Energy range |
|----------------|-----------------------|
| 0.0–0.025 eV | Cold neutrons |
| 0.025 eV | Thermal neutrons |
| 0.025–0.4 eV | Epithermal neutrons |
| 0.4–0.6 eV | Cadmium neutrons |
| 0.6–1 eV | EpiCadmium neutrons |
| 1–10 eV | Slow neutrons |
| 10–300 eV | Resonance neutrons |
| 300 eV–1 MeV | Intermediate neutrons |
| 1–20 MeV | Fast neutrons |
| > 20 MeV | Ultrafast neutrons |



UK.

n + 113Cd

K



UK.

n + 235U



n + 238U 10-10 10-5 1 ╡**10**4 104 **103** 10³ F 10² 10² Ξ E **10** 10 Ē 1 1 Ē 10-10 10-5 1

K

Incident Energy (MeV)

Cross Section (barns)

n + 239Pu

K





Incident Energy (MeV)

Cross Section (barns)





NEUTRON CROSS-SECTIONS FOR FISSION OF URANIUM AND PLUTONIUM



Neutron Reactions





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≜E A - i Z X \mathbf{S}_{n}

From Marcus Scheck

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EXILL: EX(ogam at) ILL high statistics (n_{th},γγ)







 $\sigma_{n_{th}} \cdot m_{target} \approx 250 \ mg \cdot b$ (good HPGe counts rates)





A.Blanc et al., Eur. Phys. Jour. Conf. 93 (2015) 01015

¹⁴³Nd($n_{th},\gamma\gamma$) and ⁹⁵Mo($n_{th},\gamma\gamma$): (only) 8 Clover detectors at $\Theta = 90^{o}$ octagonal symmetry \Rightarrow 4 angular groups (45^o, 90^o, 135^o, and 180^o)

| Nucleus | m _{target} [mg] | t _{mess} [hours] |
|-------------------|--------------------------|---------------------------|
| ⁹⁵ Mo | 17 | 22 |
| ¹⁴³ Nd | 0.8 | 20 |

⁹⁵Mo(n_{th},γγ)⁹⁶Mo projection total γγ matrix



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⁹⁵Mo(n_{th},γγ)⁹⁶Mo projection total γγ matrix



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Neutron Reactions

AZ

(n,n'γ) reaction

neutron scattering





 $\Gamma_n > \Gamma_v$

^{A+1}Z



UK

UK Accelerator Laboratory

- 7-MV single-ended Van de Graaff; rf ion source
- Gas targets: 0.5 to 10 MeV neutrons with ³H(p,n) and ²H(d,n); extended up to 25 MeV with ³H(d,n); monoenergetic (ΔE_n < 100 keV) neutrons
- Nuclear spectroscopy with neutron time-of-flight and γ-ray (HPGe/BGO) detection
- Nuclear structure and reaction mechanisms
- Femtosecond nuclear level lifetimes
- Neutron detector development with collaborators
- Various applications of fast neutrons
- Research program supported by the NSF since accelerator installation in 1964; upgrade in 1990s
- A hands-on, student-run facility
- www.pa.uky.edu/accelerator/







Miscellaneous Facts and Highlights

- Research program funded continuously by NSF for over 55 years!!
- About 10 publications per year result from work performed at the UK accelerator.
- In recent years, the accelerator has operated 24/7 for greater than half of the days of the year.
- For external (non-UK) non-collaborators, beam time is sold (at a bargain rate).
- For collaborations led by external users, beam time is considered an in-kind contribution.
- About half of a technician's time is in support of the accelerator facility.

University of Kentucky Accelerator www.pa.uky.edu/accelerator/







 HVEC 7-MV Model CN
rf ion source
pulsing and bunching to <1 ns pulse widths
Terminal and postacceleration bunching



Ion Source and **Beam Pulsing/ Bunching Unit**





Neutron Production

 $^{3}H(p,n)^{3}He Q = -1 MeV ^{2}H(d,n)^{3}He Q = 3 MeV$

Neutron Energies (Accelerator Voltage: 1.5 – 7.0 MV)

 3 H(p,n) 0.5 < E_n < 6 MeV 2 H(d,n) 4.5 < E_n < 10 MeV

• n' Mo foil ²H or ³H gas Target Pulsed p or d beam from VdG accelerator







(n,n'γ) Singles Measurements

IK





(n,n'γ) Singles Measurements



Compton suppression



TOF gating









Neutron Energy (MeV)


Angular Distribution $W(\theta) = 1 + a_2 P_2(\cos \theta) + a_4 P_4(\cos \theta)$

Comparison with statistical model calculations (CINDY)

 \rightarrow multipole mixing ratio (δ) and spins





Kentucky Gamma-ray Spectrometer KEGS



KEGS







| Neutron energy at center of the gas cell | 1.75 MeV | 3.19 MeV |
|--|----------|----------|
| Straggling in 3.3-µm Mo entrance foil (keV) | 32 | 31 |
| dE/dgas, 3-cm tritium cell at 1 atm (keV) | 81 | 55 |
| $dE/d\theta$, outgoing neutron energy deviation over the sample (keV) | 23 | 40 |

Diagnostic MCNPX calculations of neutron production in gas cell

Doppler-Shift Attenuation Method



$\mathsf{E}(\theta) = \mathsf{E}_{\gamma} \left(1 + v/c \cos \theta \right)$

The nucleus is recoiling into a viscous medium.

$$v \rightarrow v(t) = F(t)v_{max}$$

 $E(\theta) = E_{\gamma} (1 + F(\tau) v/c \cos \theta)$





Level Lifetimes: Doppler-Shift Attenuation Method (DSAM)



Scattered neutron causes the nucleus to recoil. Emitted γ rays experience a Doppler shift. Level lifetimes in the femtosecond region can be determined.

T. Belgya, G. Molnár, and S.W. Yates, Nucl. Phys. A607, 43 (1996). E.E. Peters *et al.*, Phys. Rev. C 88, 024317 (2013).



DSAM



T. Belgya, G. Molnár, and S. W. Yates, Nucl. Phys. A607, 43 (1996).

University of Kentucky Experiments

- Inelastic neutron scattering
 - Monoenergetic neutrons
 - Allows determination of :
 - Level scheme
 - Transition multipolarities
 - Multipole mixing ratios
 - Level lifetimes
 - Transition probabilities



Inelastic Neutron Scattering with Accelerator-Produced Neutrons

- No Coulomb barrier/variable neutron energies
- rightarrow Excellent energy resolution (γ rays detected)
- Nonselective, but limited by angular momentum
- Lifetimes by Doppler-shift attenuation method (DSAM)
 T. Belgya, G. Molnár, and S.W. Yates, Nucl. Phys. A607, 43 (1996) (feeding-time problem minimized)
 E.E. Peters *et al.*, Phys. Rev, C 88, 024317 (2013)
- Gamma-gamma coincidence measurements
 C.A. McGrath *et al.*, Nucl. Instrum. Meth. A421, 458 (1999)
 E. Elhami *et al.*, Phys. Rev. C 78, 064303 (2008)
- Limited to stable nuclei
- Large amounts of enriched isotopes required

^{144}Nd 3_3^{-} octupole isovector candidate



Lifetime measurement using Gamma-Ray induced Doppler (GRID) Broadening Method





H.G.Boerner and J.Jolie, J.Phys.G. **19** (1993) 217 Literature: $au(3_3^-) = 94^{+75}_{-34}$ fs



PHYSICAL REVIEW C

VOLUME 59, NUMBER 5

MAY 1999

Ultrahigh resolution study of collective modes in ¹⁵⁸Gd

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PHYSICAL REVIEW C

VOLUME 47, NUMBER 3

MARCH 1993

Absolute B(E1) values in the shape transitional ^{148–152}Sm isotopes

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NUCLEAR INSTRUMENTS & METHODS

> IN PHYSICS RESEARCI

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Collective excitations of ⁹⁶Ru by means of $(p, p'\gamma)$ experiments

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Fig. 1. Typical γ -ray singles spectrum (upper panel) and proton singles spectrum (lower panel) obtained in the 96 Ru(p, p' γ) experiment at angles of $\theta_{\gamma} = 90^{\circ}$ and $\theta_{p'} = 131^{\circ}$, respectively. The former is dominated by the $2^+_1 \rightarrow 0^+_{gs.} \gamma$ -ray transition of 96 Ru (indicated with *) and transitions stemming from the 56 Co calibration source (c). The proton spectrum is dominated by protons scattered in the target and backing material, indicated with (t) and (b), respectively.





Fig. 3. (Color online) γ -ray spectra gated on the excitation energy of the 2_3^+ state at $E_x = 2283$ keV obtained for three different groups which are characterized by three different values of $\cos \Theta$. An unshifted peak is observed for $\cos \Theta = 0.00(7)$, while the peaks for $\cos \Theta = -0.84(5)$ and $\cos \Theta = +0.84(5)$ are shifted to lower and higher energies, respectively.



Fig. 6. Calculated Doppler-shift attenuation factor $F_{\text{calc}}(\tau)$ for the 2_3^+ state at $E_x = 2283$ keV as a function of the level lifetime τ . The experimental lifetime value τ_{\exp} is obtained by projecting the experimental Doppler-shift attenuation factor $F_{\exp}(\tau)$ to the *x*-axis. The gray-shaded area corresponds to the statistical uncertainties.



Fig. 4. Centroid shifts of the peaks observed in the proton-gated γ -ray spectra as a function of the $\cos \Theta$ value characterizing the groups. The linear trend of the peak centroids is clearly visible. The error bars in horizontal direction include the standard deviation to the mean value of $\cos \Theta$ for each group as well as the uncertainty introduced by the opening angles of the particle detectors. The uncertainties of the peak centroids are dominated by the uncertainty in the energy calibration with the ⁵⁶Co source.

The ⁹⁴Zr Story: In the beginning...

Our studies of ⁹⁴Zr with the $(n,n'\gamma)$ reaction and an enriched ⁹⁴ZrO₂ scattering sample revealed an interesting and <u>unique</u> result — *i.e.*,





Measurements at TU Darmstadt

Studies of ⁹⁴Zr with electron scattering indicated a conflict with our results.

M. Scheck, N. Pietralla et al., Technical Univ. Darmstadt

 $B(E2; 2_2^+ \rightarrow 0_1^+) > B(E2; 2_1^+ \rightarrow 0_1^+)$

 $B(E2; 2_2^+ \rightarrow 0_1^+) = 0.9 \times B(E2; 2_1^+ \rightarrow 0_1^+)$

 $(\tau = 183^{+13}_{-12} \text{ fs}) \times \sim 2$



Is there a problem with our measurement of the 2^+_2 lifetime?

- Return to the analysis of Elhami's data: $\tau = 183^{+13}_{-12}$ fs
 - Revealed an error in the density used
 - Corrected result: $\tau = 202^{+16}_{-13}$ fs
- Re-analyze the data, starting from the spectra:

- Result: $\tau = 215^{+21}_{-19}$ fs

- Re-measure with a metallic Zr scattering sample of natural abundance:
 - Elhami *et al.* used an enriched (98.6%) ⁹⁴ZrO₂ sample.
 - Any problems which may arise due to sample composition should be eliminated.
 - Stopping powers are better understood for metals.
 - Validity of Bragg's Rule additivity of stopping powers
 - Natural sample allows comparison with other isotopes.

^{nat}Zr_{metal}(n,n'γ)



Is this difference due to the use of an oxide scattering sample by Elhami *et al*.?



they agree.

^{nat}ZrO₂(n,n'γ)



 $E_{n} = 2.0 \text{ MeV}$



Literature/Metal/Oxide Comparison





What is the source of the discrepancy?

• Stopping powers?

 No! The lifetimes from the metallic and oxide samples of natural abundance agree well.

- Enriched ⁹⁴ZrO₂ sample composition?
 - Is it really ZrO₂?
 - Is it crystalline material?



ZrO₂ exhibits polymorphism. Monoclinic phase - 5.850 g/cm³ Tetragonal phase - 6.045 g/cm³ Cubic zirconia - 5.500 g/cm³

⁹⁴ZrO₂ Sample
98.57% enriched, leased from ORNL
20 g in 2.6 cm x 3.9 cm container; compressed to 0.988 g/cm³
"This is much less dense than other ZrO₂ samples. The material was very grainy and did not compress well." — SWY 3/5/06



From X-ray Diffraction

⁹⁴ZrO₂







Powder X-ray Diffraction





SEM ^{nat}ZrO₂









SEM ⁹⁴ZrO₂







Is the difference in lifetimes due to the amorphous component?

Effect of Amorphous Material: ^{nat}Zr(OH)₄

IK

Powder X-ray Diffraction Spectrum



Effect of Amorphous Material: ^{nat}Zr(OH)₄



 $\tau = 198^{+24}_{-19}$ fs

Previous result: $\tau = 215^{+21}_{-19}$ fs

^{nat}Zr metal: $\tau = 388^{+42}_{-33}$ fs

^{nat}ZrO₂: $\tau = 343^{+38}_{-33}$ fs Why???

Effect of Particle Size V velocity velocity **ν(**τ) **ν(**τ)

time

Large Particle Size

- Recoil within a single particle
- Uniform stopping

time

Small Particle Size

- Recoiling through multiple particles
- Non-uniform stopping

^{nat}ZrO₂ Particles of Different Sizes

 $ZrO(NO_3)_2 \cdot nH_2O(s) + H_2O(l) + 2NH_4OH(aq) \rightarrow Zr(OH)_4(s) + 2NH_4^+(aq) + 2NO_3^-(aq)$

 $Zr(OH)_4(s) \xrightarrow{calcination} ZrO_2(s) + 2H_2O(g)$





with B.H. Davis University of Kentucky Center for Applied Energy Research

Lifetime as a Function of Domain Size


UK

Final Results





PHYSICAL REVIEW C 88, 024317 (2013)

Level lifetimes in the stable Zr nuclei: Effects of chemical properties in Doppler-shift measurements

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PHYSICAL REVIEW LETTERS

week ending 11 JANUARY 2013

Collective Structure in ⁹⁴Zr and Subshell Effects in Shape Coexistence

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