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DANCE: A 4π BARIUM FLUORIDE DETECTOR FOR MEASURING NEUTRON CAPTURE ON UNSTABLE NUCLEI

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Measurements of neutron capture on unstable nuclei are important for studies of s-process nucleosynthesis, nuclear waste transmutation, and stewardship science. A 160-element, 4π barium fluoride detector array, and associated neutron flight path, is being constructed to make capture measurements at the moderated neutron spallation source at LANSCE. Measurements can be made on as little as 1 mg of sample material over energies from near thermal to near 100 keV. The design of the DANCE array is described and neutron flux measurements from flight-path commissioning are shown. The array is expected to be complete by the end of 2002.

1. Introduction

Precise measurement of neutron capture cross sections in the electron-volt and keV regions on radioactive targets is needed for several applications, including nuclear astrophysics and stockpile stewardship. Capture cross sections are difficult to calculate accurately because they depend on fine details of nuclear structure and level densities at 5 to 10 MeV in excitation. A recent compilation of Maxwell-averaged capture cross sections using the

Non- Smoker statistical model code[1] showed that the calculated cross sections differed from measured cross sections by up to a factor of two for masses between A=25 and A=210. While there are capture measurements on most stable nuclides, there are very few measurements on unstable nuclides.

One of the main applications for capture cross sections is in understanding s- process nucleosynthesis[2]. The s-process occurs by sequential neutron capture along the line of beta stability. The interesting energy range for these cross sections is over Maxwell distributions centered at 25 keV and at about 10 keV. A second application is in Stewardship Science, where accurate capture cross sections on unstable nuclides are needed to interpret "rad-chem" diagnostics from past nuclear tests.

Three separate experimental components are needed to make these measurements: An intense neutron source, facilities to fabricate and handle radioactive targets, and an efficient, well characterized gamma detector. The source and detector are discussed in this paper; target preparation is done using the facilities of the Isotope and Nuclear Chemistry Group at Los Alamos.

2. Neutron Source

The DANCE is being constructed on Flight Path 14 at the Manuel J. Lujan, Jr. Neutron Scattering Center at LANSCE[3]. Flight Path 14 views the upper-tier "backscatter" water moderator. This moderator is 13 cm X 13 cm by 4 cm thick, and is surrounded by a Be and Pb neutron reflector. This "partially coupled" configuration maximizes the thermal neutron flux but at the expense of a broadened time distribution. At energies of a few eV and above, these effects become increasingly less signficant. It is anticipated that this backscatter configuration will reduce the beam contamination from high-energy neutrons, charged particles, and gammas. The Lujan moderators are discussed more thoroughly in Ref. [4].

The bulk shielding surrounding the spallation target and moderator is 4.72 m in radius. The sample is positioned 20 m from the moderator and the beam stop is at 30 m. A box for remotely inserting various filters is at 7 m. Filters used are Bi and S for black resonances, and Cd to reduce the flux of thermal neutrons. Four discrete collimators are located outside the bulk shield, each constructed of about a meter of copper, brass, and 5% borated polyethylene. The collimation was designed to produce a uniform 1 cm diameter beam spot at the target location with minimal penumbra outside the central beam. The last collimator has a r = 0.3 cm opening with the downstream edge at 18.88 m. This tight collimation limits the beam intensity, which depends on the area of the moderator that is viewed. To reduce gamma backgrounds, the beam pipes and flanges were constructed of aluminum and the use of iron in components outside of the bulk shield was kept to a minimum.

The flight path shielding was designed to limit the total external radiation dose to less than 1.0 mrem/hr along the first 10 m of the flight path and 0.5 mrem/hr beyond 10 m. Magnetite-loaded concrete blocks were used to shield the beam pipes and the walls of the detector area. Only polyethylene and borated polyethylene were used for shielding the roof of the detector area. Monte Carlo shielding calculations predicted significant high-energy gamma-ray production from neutron capture in the polyethylene and concrete, so the interior walls of the detector area were faced with 2.54 cm thick 5% borated polyethylene, which yields lower energy gamma rays following neutron capture.

During the 2001 run cycle, the spallation source was operated at a proton current of 55 μA. The neutron flux on FP14 was measured by three different techniques. First, a calibrated ^{3}He tube was used[5]. Next, a fission chamber with 286 μg/cm^2 of ^{235}U was used. Lastly, a neutron monitor consisting of a 546 μg/cm^2 thick, 1 cm diameter deposit of ^{6}LiF, on an Al foil backing viewed by a Si surface barrier detector, was employed to detect neutrons via the ^{6}Li(n,αt) reaction. The measured flux is shown in Fig. 1. The three measurements were each made at a different flight path length, and were converted to moderator surface current to correct for this geometric difference. The three measurements are not consistent, and all were considerably below the anticipated flux. This discrepancy is not understood and is still under study. The ^{3}He and ^{6}Li measurements can be fit to a surface current of the form I = A/E with E in eV and A = 1.10 x 10^9 N/cm^2/sr/eV/sec at 55 μA, where the area refers to the viewed area of the moderator, about 24 cm^2, and the solid angle refers to the solid angle of the sample. At the 20 m target location, this yields a flux Φ = (3.70 x 10^3 N/cm^2/eV/sec)/E.

3. Preliminary Data with C_6D_6 Scintillators

Preliminary capture measurements were made using two C_6D_6 detectors, each 12.5 cm diameter by 7.5 cm thick, mounted adjacent to a 5 cm diameter target holder made from 0.16 cm Al tubing. A LeCroy Model 2367 Universal Logic Module was configured as a multi- hit long-range TDC with 100 ns/chan resolution. The targets studied were Au, ^{234,236,238}U powder, each contained in a small quartz tube, ^{150,152}Sm powder in quartz, and ^{151}Sm

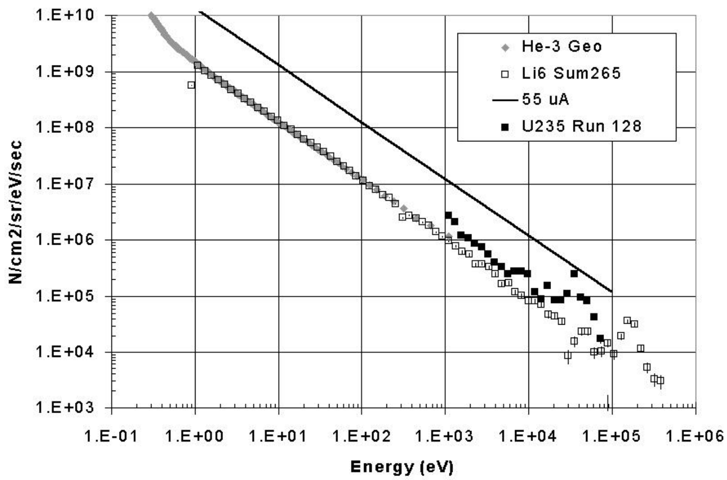

Figure 1. Surface current for FP14 at the Lujan Center, measured at 55 μA proton current. The predicted surface current is shown as the solid line.

electroplated on a thin (2.5 μm) Ti foil covered with a second Ti foil. These data are still being analyzed.

Figure 2 shows the time-of-flight spectrum from the ^{151}Sm target. The isotopic composition of this target is 71% ^{151}Sm, 13% ^{150}Sm, 7% ^{152}Sm, 6% ^{147}Sm, and 1% ^{149}Sm. The large resonance near channel 14000 is the 1.088 eV resonance in ^{151}Sm. Note that 1 keV corresponds to channel 500. The gray line is the spectrum from a Ti foil blank normalized to the same neutron fluence.

4. DANCE Design

The advanced gamma detector is being built to provide an increased and better- determined detection efficiency, and also to provide better background rejection. Backgrounds are due to capture in material surrounding the target, but also due to capture reactions in the detector from neutrons scattered in the target. The neutron scattering cross section is greater than the capture cross section in the kilovolt region for many materials. Three criteria were established for the detector:

- Calorimetric to measure the total gamma ray energy emitted
- Insensitive to neutrons
- Segmented and fast to handle radioactive targets (one Curie is 37

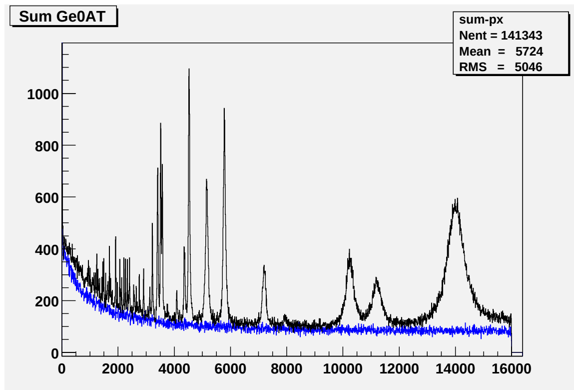

Figure 2. Time of flight spectrum for ^{151}Sm(n,γ). The contribution from a blank Ti foil sandwich is shown as the smooth gray curve. The time calilbration is 100 ns/channel.

decays/ns)

Extensive Monte Carlo calculations were made using GEANT-3 to design the detector[6,7]. Of the commonly available scintillator materials, BaF_2 was chosen because it has the smallest neutron capture cross section and a very fast (0.6 ns) component of light. It suffers from an internal alpha particle background due to decay of Ra and its decay chain products, but pulse-shape discrimination can be used if needed. The crystal array should completely cover 4π steradians with no gaps, and each crystal should have equal area and volume. The analysis of Habs[8] showed that 162 elements with 4 different shapes will meet this requirement. The inner radius of the ball is 17 cm and each crystal is 15 cm deep, 734 cm^3 in volume, and has an inner area of 22.9 cm^2. Each crystal is coupled to an Electron Tubes 9921 7.5 cm phototube with quartz window using RTV-615 for maximum UV transmission.

The Monte Carlo calculations indicated that scattered neutrons could contribute a significant background, especially in the 10 to 100 keV range of interest. Several additional methods will be employed to decrease this background. First, the measured reaction Q value can be used in many cases to discriminate between true capture events and events induced by scattered neutrons[6,7]. Next, the Monte Carlo calculations predict that an 8 cm thick ^{6}LiH shell inside the array would reduce the scattered neutrons to 42% of the unattenuated number, in the 10 to 100 keV energy range[7]. However, because of space limitations we will use a 6 cm thick ^{6}LiH sphere surrounding the target. Finally, a "hit pattern" analysis of the crystals that fire in an event will be tried. The calculations[7] predict that true capture events will produce several clusters of detectors firing, each due to an individual gamma ray from the decay cascade. Events due to neutron capture in the BaF_2 tend to be grouped primarily into one cluster.

The data acquisition system will consist of two Acqiris DC-265 8-bit waveform digitizers on the anode signal of each phototube. The digitizers sample at 500 MHz. Each digitizer has a different voltage gain to match the dynamic range of the fast and slow components of the scintillation light. The slow component contains about 85% of the light, and has a decay time of 630 ns. The fast component, while providing only 15% of the light, has a decay time of 0.6 ns and is the dominant feature of the waveform. The front end software will initially return for each event only a time and pulse height number, which the analyzer routine will histogram and log.

Figure 3 shows a typical pulse from a completed crystal assembly, acquired with an Acqiris DC-270 1 GHz digitizer. Fig 4. shows a ^{60}Co spectrum obtained by simply adding the counts in a waveform from 10 ns before the trigger time to 1790 ns after. The average resolution of the 1332 keV peak is 11% fwhm.

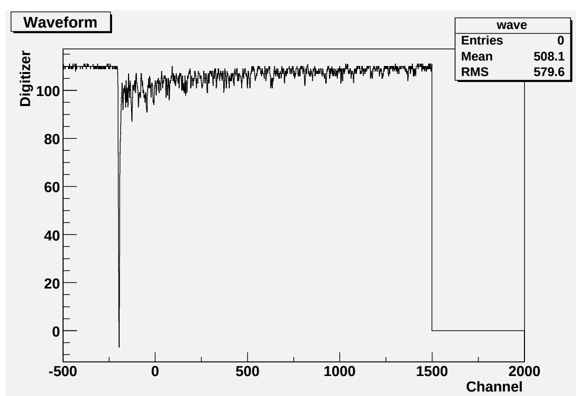

Figure 3. Typical waveform from a completed Ba_2 crystal assembly using a DC-720 1 GHz digitizer. One channel corresponds to 1 ns. The fast and slow components are easily recognized.

5. Summary and Future Plans

The DANCE array is under construction. As of August, 129 crystals have been delivered with delivery of all 169 crystals expected before the end of

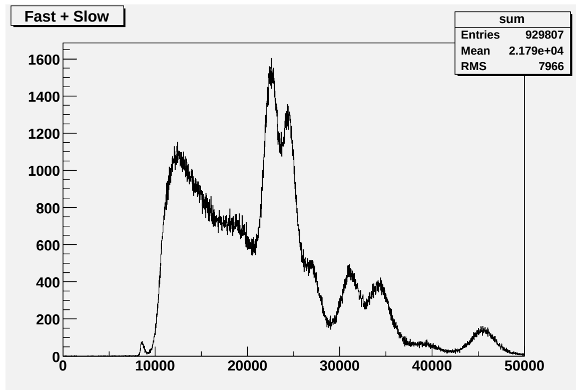

Figure 4. ^{60}Co spectrum obtained by simply summing waveforms from a 1 GHz digitizer. The peaks from the alpha-decay background in the crystal are readily seen.

2002. A multi-year program of targets to be measured for stockpile stewardship and s-process branch point studies has been mapped out. Initially, targets that can be chemically purified have been chosen, and ^{146}Nd, ^{154}Sm, and ^{170}Er were irradiated at the ILL in spring, 2002, to produce 4 to 10 mg of ^{147}Pm, ^{155}Eu, and ^{171}Tm. It is expected that these isotopes will be studied using the DANCE array in 2002 , along with a new measurement of ^{151}Sm.

References

1. T. Rauscher and F.-K. Thieleman, in *Atomic and Nuclear Astrophysics*, A. Mezzacapa, *ed.*, IOP, Bristol (1998), p. 519.
2. F. Käppeler, *Prog. Part. Nucl. Phys.* **43**, 419-483 (1999).
3. For more information http://lansce.lanl.gov.
4. G.J. Russell, et al., in *Fundamental Physics with Pulsed Neutron Beams (FPPNB)*, C.R. Gould, et al., *ed.*, World Scientific, Singapore, pp 19- 48 (2000).
5. L. Daemen, private communication.
6. M. Heil, et al., *Nucl. Instr. Meth.* **A459**, 229-246 2001). M. Heil, et al., Los Alamos National Laboratory Report LA-UR-99-4046 (1999).
7. R. Reifarth, et al., Los Alamos National Laboratory Report LA-UR-01-4185 (2001).
8. D. Habs, F.S. Stephens, and R.M. Diamond, Lawrence Berkeley Laboratory Report LBL-8945 (1979).