Characterization of a Tunable Quasi-Monoenergetic Neutron Beam from Deuteron Breakup

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Abstract: A neutron irradiation facility is being developed at the 88-Inch Cyclotron at Lawrence Berkeley National Laboratory for the purposes of measuring neutron reaction cross sections on radioactive targets and for radiation effects testing. Applications are of benefit to stockpile stewardship, nuclear astrophysics, next generation advanced fuel reactors, and cosmic radiation biology and electronics in space. The facility will supply a tunable, quasi-monoenergetic neutron beam in the range of 10-30 MeV or a white neutron source, produced by deuteron breakup reactions on thin and thick targets, respectively. Because the deuteron breakup reaction has not been well studied at intermediate incident deuteron energies, above the target Coulomb barrier and below 56 MeV, a detailed characterization was necessary of the neutron spectra produced by thin targets.

Neutron time of flight (TOF) methods have been used to measure the neutron spectra produced on thin targets of low-Z (titanium) and high-Z (tantalum) materials at incident deuteron energies of 20 MeV and 29 MeV at 0°. Breakup neutrons at both energies from low-Z targets appear to peak at roughly half of the available kinetic energy, while neutrons from high-Z interactions peak somewhat lower in energy, owing to the increased proton energy due to breakup within the Coulomb field. Furthermore, neutron spectra appear narrower for high-Z targets. These centroids are consistent with recent preliminary proton energy measurements using silicon telescope detectors conducted at LBNL, though there is a notable discrepancy with spectral widths.

Prospects for producing a tunable, quasi-monoenergetic neutron facility of $10^6$-$10^8$ n/cm$^2$/s at LBNL are promising.

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INTRODUCTION

Dissociation, or “breakup,” of deuterons in the Coulombic field of a heavy nucleus is being investigated to produce a tunable, high-flux quasi-monoenergetic neutron source at the 88-Inch Cyclotron at Lawrence Berkeley National Laboratory (LBNL). This neutron irradiation facility will eventually be used to perform direct cross section measurements on unstable targets in the range of \( E_{\text{neutron}} = 10-32 \text{ MeV} \) for isotopes of importance to stockpile stewardship, advanced fuel cycle reactor physics, and stellar nucleosynthesis. In addition, the facility will be available to study radiation effects on microelectronics, radiation biophysics, materials studies, and other related neutron applications.

Deuteron breakup is a very old and basic nuclear physics problem, dating from speculations by Oppenheimer and Phillips [1] in 1935. More recent treatments of the Coulombic component of the breakup have been given by Liu and Thaler [2] in 1990 and by modern quantum mechanical 3-body theories [3]. In the energy range of the 88-Inch Cyclotron \( (E_d \leq 65 \text{ MeV}) \), these theoretical calculations have not been proven valid, and the viability of deuteron breakup as a source of quasi-monoenergetic neutrons in this energy range has not yet been proven. In fact, very few experimental studies of deuteron breakup exist in this energy range, and none of them have attempted to measure neutron spectra from thin targets at forward angles.

Assuming pure Coulombic breakup in the field of the heavy target nucleus, in the most simplistic model the deuteron deaccelerates on the way in to the breakup radius \( R_B \), and the proton and neutron each take one-half of the available energy at that point. Then the protons are accelerated on the way out, leaving

\[
E_n = \frac{1}{2} \left( E_d - \frac{Z e^2}{R_B} - 2.22 \text{ MeV} \right) \quad [1a]
\]

and

\[
E_p = \frac{1}{2} \left( E_d + \frac{Z e^2}{R_B} - 2.22 \text{ MeV} \right), \quad [1b]
\]

where 2.22 MeV is the binding energy of the deuteron. Several early experiments [4-6] studied the deuteron breakup phenomenon at low energies \( (E_d = 8-17 \text{ MeV}) \) on a range of heavy targets. The value of \( R_B \), calculated from the measured proton energies using equation 1b, showed that in most cases breakup occurred well outside the nuclear radius. The main conclusion of this body of work at lower energies was that Coulombic breakup dominated at forward angles for heavier targets, but nuclear breakup played a role at all energies for lighter targets like Be and C, and became more important for all targets as the deuteron energy was raised.

Contradictory to earlier low-energy studies, a recent experiment by Okamura et al [7] at 56 MeV concluded that Coulombic breakup was not important at this energy for the heaviest targets, providing a stringent test of modern theories and the conclusions are still being debated [8]. Clearly, the relative roles of Coulombic and nuclear breakup are changing in this region and simple predictions of the neutron energy spectra on thin targets cannot be relied upon and experimental neutron beam characterization at these energies is necessary. Characterization has been performed by a variety of methods, including direct measurement using time-of-flight, complimentary measurements of the proton energy using a silicon telescope array, and the use of activation foils. The direct time-of-flight measurement results only are presented here.

METHOD AND EXPERIMENTAL SETUP

A series of experiments was performed in the 88-inch cyclotron “Vault” to directly measure neutrons produced from deuteron breakup reactions on two different thin targets, a relatively lower-Z target of 17.0 \( \text{mg/cm}^2 \) titanium and a higher-Z target of 42.2 \( \text{mg/cm}^2 \) tantalum. Beams
of two different incident deuteron energies, 20 MeV and 29 MeV, impinged on each target at about 10 nA for a total of four target/energy combinations. At each respective energy, the beam loses 0.5 or 0.4 MeV in the titanium target and 0.8 or 0.6 MeV in the tantalum target. Neutrons along the 0° axis are then measured using a Stilbene detector located about 3.75 m from the targets, while the deuteron beam was guided out of the Vault with a bending magnet as shown in figure 1. The beam pulse length was narrowed to about 2.5 ns by radially oriented phase slits to improve the time-of-flight energy resolution. A Faraday cup installed in cave 2 and another on a beam stop at the exit of the cyclotron permitted normalization of each measurement.

The Stilbene detector is sensitive to both photons and neutrons. However, neutrons deposit energy in the detector through proton recoil rather than direct interaction with electrons [9]. Therefore, neutrons can be discriminated from photons by their longer pulse shape. Because any amount of energy, up to the full neutron energy, can be transferred in proton recoil, the only direct means of measuring the energy of the neutron is through time-of-flight (TOF) to the detector relative to the radiofrequency (RF) of the cyclotron. The start time is given by the arrival of the photon burst from the beam passing through the target.

BACKGROUND SOURCE SUBTRACTION

Thin target time-of-flight measurements at 0° can only be made at a location where the neutrons exit through a thin window, mostly transparent to neutrons, along the 0° axis just after a bending magnet, so that the neutrons can travel unfettered to the detector while the deuteron beam is steered out of the room. Currently, the only location at the 88-inch cyclotron where such a geometry can be implemented is in the “Vault,” which also houses the cyclotron. Unfortunately, this location contains a number of background neutron sources, which must be subtracted from the target source.

An automated iron “shadowbar,” as shown in figure 1, was raised between the target and the detector to allow measurement of primarily background events from deuteron interactions in the beam pipe downstream of the target after the switching magnet. This shadowbar blocks all particles along the 0° axis so that the detector only measures particles coming from other sources in the room. Borated polyethylene blocks were placed between the detector and the end of the switching magnet to reduce the primary downstream source of these background neutrons when beam particles which have scattered to larger angles in the target encounter the narrow beam pipe exiting the magnet.

Furthermore, for each energy, we measured particles produced when no target was in place, both with and without the shadowbar, so that upstream sources along the 0° axis could similarly be subtracted from the particle spectra. The primary background source from upstream beam interactions appears to be from the deflector channels as the beam exits the cyclotron. While the gamma sources from upstream, target, and downstream reactions often appear roughly equivalent in strength, the forward-directedness of breakup neutrons versus the isotropic emission of photons leads to a much larger effect on the neutron spectra for upstream background interactions.

The gamma sources, from both general background and from the target, are shown in figure 2 for each deuteron energy and target. Figure 2 plots the time elapsed between an event in the detector and the next RF pulse. Thus, the time axis is from right to left, with the target peak plotted “first.” Subtracting time-of-flight spectra from a run with the shadowbar inserted removes the effect of downstream interactions, while subtracting a shadowbar-subtracted run without a target removes the effect of upstream interactions, ideally leaving only a gamma peak...
from target interactions. Unfortunately, the upstream background source appeared to change significantly at 20 MeV using a tantalum target (case c) compared to the run with no target. This led to a less-than-ideal upstream background subtraction, as seen in figure 2c.

Furthermore, the tantalum target tends to scatter the deuteron beam more, resulting in the beam scraping a vertical surface inside the magnet on the 0° axis about 60-70 cm downstream of the target, producing a background source which cannot be subtracted. Its proximity to the detector leads to a larger effect on the gamma source than the neutron contamination. Since the interaction material in the magnet is primarily iron, which has a Z-value similar to titanium, neutrons contaminating the spectrum are expected to be similar to those measured from the titanium targets.

Subtracting neutron time-of-flight spectra in the same way leaves only neutrons from target interactions, as seen in figure 3. It is clear that upstream interactions dominate the results, especially at higher deuteron energies and for the heavier tantalum target. In most cases, this upstream neutron source was, in fact, larger than the target source.

RESULTS

Neutron energy spectra, calculated from the time-of-flight spectra in figure 3, are shown for each energy and target in figure 4. It appears that with the low-Z titanium target, the spectra peak at roughly half the deuteron energy minus the Q-value of the reaction. With the high-Z tantalum target, the peak is at a lower energy. This is as expected from the simple Coulombic approximation presented in equation 1a. Furthermore, at both energies, the width of the spectrum is narrower for the higher-Z target, consistent with the 56 MeV data of Okamura et al [7]. At 20 MeV, the full width at half maximum (FWHM) is reduced from about 7.1 MeV for the titanium target to about 5.6 MeV for the tantalum. At 29 MeV, the FWHM is reduced from about 8.1 MeV to about 4.8 MeV.

The absence of a titanium-like bump in the 29 MeV tantalum spectrum suggests that there may not be significant neutron contamination due to the 0° downstream background source, which cannot be measured independently from the target source, from beam scraping in the magnet. It is unclear if such contamination is present in the 20 MeV tantalum spectrum, though the larger FWHM suggests this may be the case. Moreover, due to the overwhelming strength of the subtracted upstream background source compared to the target source, especially at 20 MeV, the tantalum spectra must be viewed with some suspicion.

For the foil thicknesses used in this experiment, the total fluxes calculated for a 10 μA beam at 1 m from the target, a typical expected neutron target irradiation distance, were 2x10^5, 2x10^5, 5x10^5 and 7x10^5 n/cm^2/s respectively for 20 MeV deuterons on tantalum and titanium targets and for 29 MeV deuterons on the same targets. Due to the large background subtractions, the uncertainties on these fluxes are expected to be large, but provide a general guideline for the capabilities of a future irradiation facility. While fluxes fall slightly short of the 10^6-10^8 n/cm^2/s goal, the desired range can be easily achieved by moving the irradiation target closer to the neutron production target, or by using thicker foils. Comparing the energy loss in the foils versus the width of the spectra in Figure 4, a doubling or tripling of foil thickness would not significantly affect the width of the energy spectra. Moving the irradiation target inside the switching magnet to within 30 cm of the neutron production target can furthermore achieve another order of magnitude increase in flux.

CONCLUSIONS
These experiments have revealed important characteristics of the neutron spectra produced by thin target deuteron breakup at intermediate energies on both light and heavy targets.

The need for large background source subtractions, particularly from upstream beam interactions in the cyclotron extractor, leads to low confidence in the extracted widths. These widths are larger than what was expected from a simple extrapolation of results of Ref 7 to thinner targets; however, these measurements were performed at substantially lower energies. In the near future, we plan to extend our measurements to 56 MeV, to enable a direct comparison with Ref. 7, and also will measure the neutron energy spectra at other forward angles.

The results have been very important for guiding further development of the neutron irradiation facility. The beam optics must be carefully considered, even for targets as thin as those used in this study. Neutron flux will be limited by the target thickness, so it is necessary to weigh these factors together with the needs of the diverse applications in the beamline design.

Measurements on thick targets in caves outside of the Vault have demonstrated that these background sources can be reduced substantially. We are currently in the process of retrofitting and installing a switching magnet in cave 2, which will eliminate most of the upstream interactions and allow finer tuning to reduce downstream interactions, thereby improving the accuracy of these measurements dramatically. Despite the present limitations, the outlook for producing a tunable, quasi-monoenergetic neutron source using the deuteron breakup reaction is good. In the near future, the absolute neutron flux will be measured for the present configuration using the activation foil technique. It is expected that neutron fluxes of $10^6$ to $10^8$ n/cm$^2$/s at energies of interest are reasonably achievable.

REFERENCES
Figure 1: Experimental setup. The deuteron beam exits the cyclotron, travels down the beam pipe and strikes a thin target, then is bent 40° through the bending magnet into Cave 2. Neutrons travel from the breakup target along the 0° axis to the Stilbene detector. A shadowbar can be raised or lowered to block 0° particles from the target and from upstream background reactions.
Figure 2: Gamma ray time-of-flight (TOF) spectra. Targets and deuteron energies are (a) titanium at 20 MeV, (b) titanium at 29 MeV, (c) tantalum at 20 MeV, and (d) tantalum at 29 MeV. The rightmost peak represents gamma rays from the target. The two leftmost peaks are from upstream background reactions while the middle peaks are from downstream background reactions. Shaded regions represent gamma ray TOF spectra after background subtraction from “target out” and “shadowbar” runs.
Figure 3: Neutron time-of-flight spectra. Targets and deuteron energies are (a) titanium at 20 MeV, (b) titanium at 29 MeV, (c) tantalum at 20 MeV, and (d) tantalum at 29 MeV. Background subtraction is the same as in Figure 2.
Figure 4: Neutron energy spectra produced by 20 MeV and 29 MeV deuterons on thin titanium and tantalum targets. Lines are added only to guide the eye. Error bars represent statistical uncertainly only.