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Quantification of the light output anisotropy in deuterated stilbene

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Abstract

Deuterated stilbene is an organic scintillator that is a desirable material for fast neutron spectroscopy using spectrum unfolding techniques without requiring time-of-flight information. Due to the crystal structure of the material, some anisotropy of the light output exists, which is dependent on the direction of heavy charged particle recoil relative to the crystal structure. The anisotropy of trans-stilbene (hereafter referred to as stilbene) has been well characterized in previous work, but for deuterated stilbene, the anisotropy has only been partially characterized along the a and b crystal axes, while the artificial c' axis, which shows the largest anisotropy in stilbene, has not been characterized until this publication. In this work, two deuterated stilbene crystals were characterized with neutron energies up to 35 MeV at the Los Alamos Neutron Science Center. For one of the crystals, the response is characterized along the a , b , and c' axes. This characterization shows a distinct anisotropy along the axes in deuterated stilbene, which is very similar to that found in regular stilbene, such that the a axis is the brightest, while the b and c' axes are approximately 3% and 20-35% lower relative to the a axis.

Keywords: passive interrogation, neutron, spectroscopy, unfolding, spectrometer, scintillation, deuterated stilbene

1. Introduction

Deuterated stilbene (d-stilbene) is considered to be a promising material for fast neutron spectroscopy [1]. Organic scintillators, like d-stilbene, have had much interest in recent years for use with spectrum unfolding techniques to do spectroscopy without the need for time-of-flight (TOF) information. Some examples of applications where interest has been found are cross section measurements [2], background characterization [3], source discrimination [4, 5, 6], and neutron leakage spectrum of critical assemblies [7], among others.

Spectrum unfolding requires a well-characterized response matrix, which defines the probability of emitting a given amount of scintillation light, which is the measured signal, when a given energy of neutron interacts with the detector material. The response matrix can be either measured or simulated, and in general measurement of the response matrix is preferred and initially needed to determine several key parameters for the simulations, such as the light output as a function

of energy deposition and, for crystals like stilbene, any anisotropies that exist due to the crystal structure.

Prior to the publication of this article there has been limited data for the quantification of the anisotropy of d-stilbene. The light output has been measured up to 14.1 MeV neutron energy [8, 9], but the data is sparse between the lower energies and the highest energies. The anisotropy of d-stilbene was originally found to be minor [1], though a more thorough investigation [10] has shown some anisotropy exists between the a and b crystal axes of d-stilbene. The greatest asymmetry in regular stilbene, however, is found along the artificial c' axis [11, 12], which needs to be characterized in order to better understand and simulate d-stilbene detectors.

In this work, two d-stilbene detectors have been characterized from 1-35 MeV along their b axes. The detectors were characterized at the Los Alamos Neutron Science Center (LANSCE). For one of the d-stilbene detectors, the detector response matrix (DRM) and light output have been characterized along three different axes (a , b , c') in order to examine the anisotropy.

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2. Experimental Methods

Crystal Growth

For this work, two d-stilbene crystals were solution grown [13] at Lawrence Livermore National Laboratory and were cut into 2" cubes (See Figure 1). The crystals were grown with commercial d-styrene from Sigma Aldrich which contained 98 atom % deuterium. Due to the high strength of the covalent C-D bond that is difficult to break during synthesis or crystal growth, the d-stilbene cubes are expected to contain a similar 98 atom % of deuterium. Although deuteration can cause changes to the crystal structure [14] (and therefore the anisotropy) of a material, X-ray diffraction analysis of the first d-stilbene crystals showed no difference between regular trans-stilbene and d-stilbene structures belonging to the same monoclinic space group $P2_1/a$ [13]. The crystal was wrapped by 3 layers of PTFE tape followed by a layer of the aluminum foil and finally by the black duct tape, leaving one open face such that the b axis was pointed into the photomultiplier tube (PMT). The detectors had their a , b , and c axes marked on the wrapping around the crystal.

The PMT (Model 9821KB from Electron Tube Enterprises) had a silicone (EJ-560) rubber interface attached to it using a thin layer of silicone grease (EJ-550). The crystals were directly attached to this rubber interface without any silicone grease. A schematic of the crystal mounted to the PMT with crystal axes labelled can be seen in Figure 2. The PMTs differed with one having a standard voltage divider and the other having a transistorized voltage divider.

Measurements

The methods used for the measurements were the same as in the previous work by the authors [16]. Detectors were placed directly in the neutron beam at flight path 15R of LANSCE. The neutron beam is pulsed with

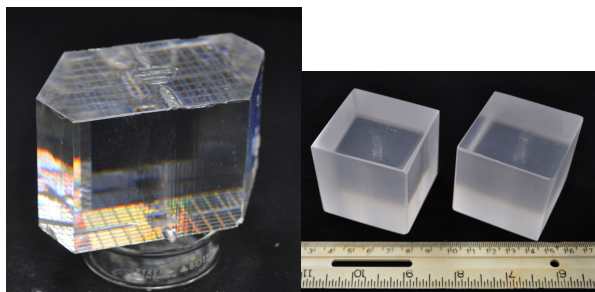


Figure 1: Raw d-stilbene crystal (left) and d-stilbene crystals cut into cubes (right).

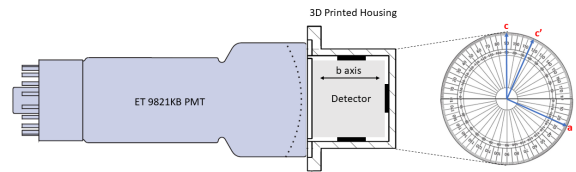


Figure 2: Schematic of d-stilbene mounted to PMT with crystal axes labelled. PMT schematic modified from [15].

a macropulse period of $625 \mu\text{s}$ and repetition rate of 100 Hz. Within each macropulse, hundreds of micropulses with a period of $1.8 \mu\text{s}$ are contained. The distances between the start of the beam and the detectors were approximately 21 m. A CAEN DT5730 was used to digitize the beam start (T_0) signal and the signal from the detector placed in the beam, which utilized a constant fraction discriminator for triggering. The onboard time coincidence (1144 ns window) feature of the digitizer was used to collect only events in the detector that were in coincidence with the T_0 signal. Due to the short period of the micropulses, some lower energy neutrons near the end of the pulse window had gamma-rays from the next micropulse pile-up in the signal, though this background is small relative to the actual signal and can mostly be removed with post-processing. Other than this source, the background has been found to be negligible [17].

The detectors were approximately gain matched and run at -1200 and -1400 V, and the integration length was set to 300 ns. Four total measurements were done on two different d-stilbene detectors with the beam parallel to a given axis: d-stilbene #1 b , a , c' axes (3.5, 0.3, 0.3 hour count times) and d-stilbene #2 b axis (5.5 hour count time).

Calibrations were done using the fitting methods of [18], which has been used in previous work by the authors [16, 19]. Pre- and post-calibrations were done using a ^{137}Cs source and the variation in the calibration was found to be approximately 1% for each measurement. Previous work [16] with the same type of PMT at similar voltages showed negligible difference between using a one point calibration with ^{137}Cs with no offset and a two point calibration with ^{137}Cs and an AmBe (4.4 MeV) source. The data presented here extends well past the 4.4 MeV line, so future work needs to be done characterize the response of the PMT past that point.

3. Analysis and Results

The DRM was generated from the data by converting the TOF between the detector and the T0 signal into an incident neutron energy. The energy uncertainty due to uncertainty in the TOF (flight path length and timing) is approximately 1% [16] for the energy range examined in this work. Therefore, the uncertainty in energy is taken as the bin width (± 100 keV). The data were binned with bin widths of 200 keV for the neutron energy, while conformal binning [20] was used for the light output. The resulting DRM can be seen in Figure 3.

The light output was derived from the measured response matrix using the methods of [21]. The method takes the derivative of each quasi-mono-energetic light output curve and finds the minimum. The derivative was calculated using a symmetric difference quotient. It should be noted that this method has known sources of bias [21, 22] on the scale of 1-5%, mainly due to the resolution of the detector, which can vary due to crystal size, PMT response, optical quality of the crystal (hazing), and quality of reflective wrapping. This bias, which increases with energy, causes a systematic lowering of extracted light output values, due to increased multiple scattering, which worsens the resolution, as detector size is increased. It was also found that the determined light output was dependent on the width of the light output binning, particularly at lower energy values, so in addition to the calibration uncertainty, an uncertainty of one bin width was added to the results. This method was used in the previous studies of d-stilbene to extract light output curves and was used here to allow for a direct comparison, which can be seen in Figure 4. The extracted light output data agrees well with the existing literature [8, 9]. The two crystals show consistent light outputs along the b-axis that match within $1\text{-}\sigma$ up until approximately 9 MeV, after that point they differ by up to $2\text{-}\sigma$ (see supplemental material). The extracted light output, particularly at higher energies, was also found to vary with total number of counts. The second d-stilbene detector had approximately 50% more counts than the first d-stilbene along the b-axis. The light output was extracted using a subset of both sets of data with the same number of counts. The resulting light outputs agree within $1\text{-}\sigma$ for all energies with an average of $0.6\text{-}\sigma$ for the difference. The second d-stilbene detector shows a consistently higher value of the extracted light output, which is attributed to small variations in the aforementioned factors that can result in bias from the method.

The d-stilbene crystal that was measured along all

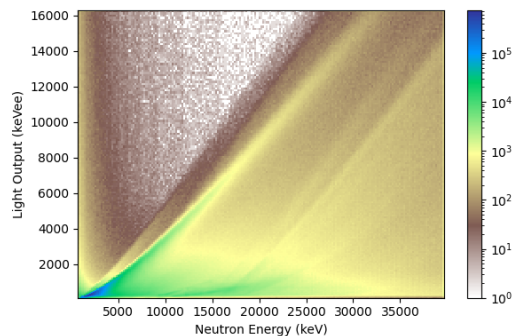


Figure 3: Detector response matrix of d-stilbene as measured without any processing. Apart from the light output edge due to deuteron recoils, a similar feature for proton recoils is also seen due to the 2% of residual hydrogen from the composition of the raw material used for crystal growth. Some background is seen as a vertical feature at the lowest neutron energies which corresponds to pile-up of particles from different pulses of the beam and is easily removed with post-processing.

three axes shows an anisotropy that is similar to regular stilbene. The a crystal axis shows the highest light output, while the artificial c' axis shows the lowest light output. The anisotropy is a function of neutron energy, but is approximately 3% lower along the b axis and 20-35% lower along the c' axis in the energy range examined in this work. For determining the ratios, a subset of 7 million counts from each measurement were taken for extracting the light outputs, in order to remove any potential bias from differences in counts. The ratios for the data points with respect to the a axis were taken and averaged over five data points. The resulting light output ratios, compared to those of stilbene can be seen in Figure 5.

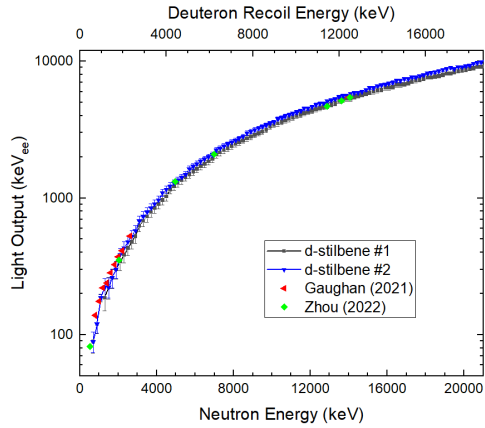


Figure 4: Light output (200 keV steps) of the two d-stilbene crystals measured along the b axis compared to literature [8, 9].

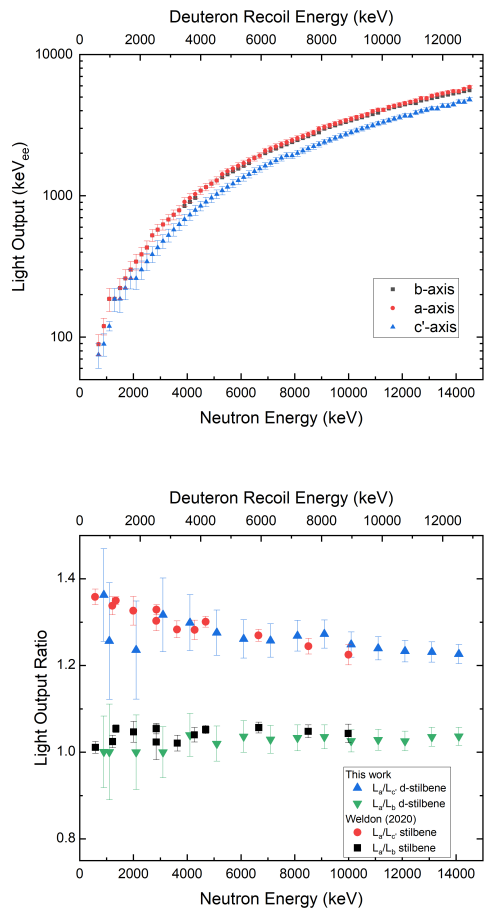


Figure 5: Top: Light output of d-stilbene along the a , b , and c' axes. Bottom: The ratio of the light output along the b and c' axes relative to the a axis. The ratios are calculated for each individual pint, then five points are averaged for each displayed point. The light output ratios of stilbene are taken from [12].

4. Conclusions

Two d-stilbene crystals had their detector response matrices characterized along the b axis from which a light output curve was extracted. For one of the crystals, the response was measured along three different (a , b , and c') axes to characterize the anisotropy. The light output along the b axis agrees with the current data available in literature. The d-stilbene material shows an anisotropy in the light output as a function of the direction relative to the crystal axes that the heavy charged particle recoils. The a axis is brighter than the b axis by approximately 3%, while the c' shows approximately 20-35% lower light output relative to the a axis. These results are similar to the anisotropy seen in regular stilbene crystals.

Future work should include double time-of-flight experiments [23] or scatter experiments to fully characterize the anisotropy as a function of angle relative to each axis as was done in [22] for regular stilbene in order to reduce the uncertainty of the light outputs and corresponding ratios, particularly at lower energies, and to reduce systematic bias from the methods used in this work. In particular, since the resolution is the main driver of the bias, the lower light output from the c' axis may change the resolution and add a bias into the result presented here on order of a few percent, so a less biased method would be ideal to verify the values in this work and reduce the uncertainties. For the application of spectrum unfolding, the effect of the anisotropy on the results should also be explored. This can be done by developing simulated response matrices with and without the anisotropy accounted for, which are then used with standard spectrum unfolding techniques.

CRediT authorship contribution statement

T.C. Borgwardt: Conceptualization, Investigation, Formal analysis, Methodology, Software, Writing - original draft **K.D. Bartlett:** Software, Investigation, Data curation, Writing - review and editing **K. Smith:** Software, Investigation, Writing - review and editing **K.C. Meierbachtol:** Conceptualization, Investigation, Supervision, Project administration, Funding acquisition, Writing - review and editing. **R.A. Weldon Jr** - Conceptualization, Investigation, Methodology, Writing - review and editing **N. Zaitseva** - Investigation, Methodology, Writing - review and editing **M. Febbraro:** Conceptualization, Investigation, Methodology, Supervision, Writing - review and editing.

227 Declaration of Competing Interest

228 The authors declare that they have no known com-
229 peting financial interests or personal relationships that
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