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Distinguishing spontaneous fission neutrons from cosmic-ray background

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ABSTRACT

We have measured the neutron spectra of cosmic-rays and a spontaneous fission emitting source (Cf-252) using a neutron double scatter spectrometer. The energy range of measurements was 0.1-10 MeV where the spectrometer efficiency is determined to be up to 8.7%, depending on the separation between detection planes. Our cosmic-ray neutron spectrum measurement is in good agreement with the sea-level data reported by Goldhagen and his co-workers. In the energy range 0.1-1.0 MeV, the cosmic-ray and Cf-252 spectra are different and separable. This difference is expected from the applicable models that describe the phenomena, 'equilibrium slowing down' (cosmic-rays) and 'Maxwellian kinetic temperature' emission (spontaneous fission). We show that >80% of Cf-252 neutrons and <25% of cosmic-ray related neutrons are emitted in this energy range of measurement, and conclude that neutron spectroscopy provides effective ways to distinguish a fission source from the cosmic-ray background.

Key words: Fast neutron; time-of-flight; neutron spectra; cosmic-ray.

1. Introduction

We have constructed a neutron double-scatter spectrometer for spectral measurements optimized to operate in the energy range of 0.1-6 MeV (1), suitable for neutron emission of spontaneous fission sources, e.g., plutonium. When these sources are weak or distant, detection is often limited by background neutrons produced by cosmic-ray interactions in the air or in structural materials. Reduction of background can be accomplished in a number of ways. First, the neutron energy range of source emission is a small portion of that of cosmic-ray induced neutrons in air, 0.025eV - 1 GeV, so that the number of background neutrons included can be minimized by the spectrometers restricted energy range. When there are sufficient counts to actually measure spectra, we will show that the source and the air background spectra are fundamentally different and separable in the 0.1-1.0 MeV range. Also, when there are sufficient counts to actually measure spectra, we will show that the source may have an identifiable signature caused by interaction with surrounding materials that can be used for target analysis. Importantly, source neutrons are received from a point source in a single direction, whereas cosmic ray generated neutrons are reasonably isotropic. Therefore a directional detector can improve the signal-to-noise ratio. The neutron double-scatter spectrometer measures neutron spectra and does so directionally. The determination of source direction has been demonstrated with high energy neutrons by Preszler *et al.* (2,3). Another example of a neutron time-of-flight apparatus is described by Elevant *et al.* (4).

In this paper, we shall compare the instrument response to Cf-252 neutrons (source on axis) to that of cosmic-rays (more nearly isotropic). It appears that higher energy cosmic-ray events in the time-of-flight data are not likely from fast neutrons, but are most probably from high energy protons and muons that are observed in the individual detector response to background. However, cosmic-ray data obtained from the spectrometer with a detector separation of 30 cm appears to be useable to 4 MeV and the data are in agreement with Goldhagen *et al.* (5,6) from the Environmental Measurement Laboratory (EML).

2. Experimental methods

In Ref. 1, we have described the theoretical response of the instrument. The electronics system is designed to measure physical parameters given by the equations (1-3), where ϕ is the angle between incident neutron of energy (E_n) and the scattered neutron of energy (E_s). E_p is energy deposited by proton recoil in Detector 1, and θ is the angle of the scattered neutron relative to the axis of the double scatter detector. When the distance between the detectors, L , is expressed in cm, time-of-flight, T_{of} , is in units of nanoseconds, and energy expressed in MeV the relevant equations are:

$$E_n = E_s + E_p \quad (1)$$

$$E_s / E_p = \tan^2 \phi \quad (2)$$

$$E_s = (0.723 L / \cos \theta T_{of})^2 \quad (3)$$

Figure 1. Electronic components of the neutron double scatter spectrometer.

The electronic components used by the double-scatter spectrometer are shown schematically in Fig.1. The electronics are connected to two photomultipliers (PM) that detect light pulses from the scintillators. The light pulse from the first detector can be analyzed for the energy deposited from the scattered proton, E_p . Detector signals are sent to constant fraction discriminators which output 1 ns logic pulses, and the time difference between detector logic pulses is measured with a time/amplitude converter, which sends output pulses proportional to the delay to be recorded in a histogram by a multichannel analyzer.

3. NEUTRON DOUBLE SCATTER SPECTROMETER RESPONSE MEASUREMENT

coincidence time-of-flight data Vs. time

Figure 2. The spectrometer response to Cf-252 spontaneous fission and Cs-137 sources.

The spectra are count rates as a function of the time differences of events relative to the start time in detector 1. Sources were located 18 cm from the first detector on the axis passing through the center of both detectors. A delay in the signal line of the second detector causes the timing signal to be registered at about 45 ns when the detector signals start at the same time, or nearly the same time for gamma/gamma double scatter. If the second detector fires first, the time between pulses is less than 45 ns. Cf-252 emits fission gamma rays and neutrons that are correlated in time, and photons from other reactions, such as radioactive decay of fission products, that are not correlated with neutron emission. Note that the count rates are normalized to the peak response, which is the gamma/gamma coincidence. A number of features in these spectra are apparent:

1. The gamma/gamma peak has about the same half-width for the Cs-137 and Cf-252 sources. This time width is relatively independent of the spacing between detectors.
2. Within 5 nanoseconds of the peak, the Cs-137 signal becomes negligible relative to the Cf-252 neutron signal.
3. The shapes of the double-scatter Cf-252 timing spectra, shown to the right of the gamma/gamma peak, are quite different because the flight path of scattered neutrons between detectors is either 8 cm or 45 cm.
4. The ratio of the neutron double-scatter signal to the gamma-ray background ratio is greater when the detectors are widely separated. In general, this is explained by the wider dispersion of neutrons at the longer flight path.
5. Coincidence events to the left of the gamma/gamma peak are primarily from fission gamma rays creating a start pulse in the second detector, g2, followed by neutron time-of-flight detection from the source to the first detector, n1. The spectral response function of the detector used in this mode, g2n1, is different from the neutron double scatter mode. For example, the g2n1 mode detection efficiency is proportional to the product of solid angles of both detectors as seen from the source, whereas the double-scatter efficiency is proportional to the solid angle of the first detector as seen from the source times the solid angle of the second detector as seen from the first.
6. The shapes of g2n1 timing spectra in figure 2 are relatively independent of detector separation. This is because changing the position of the second detector affects the gamma flight time by less than a nanosecond and leaves the neutron flight time unchanged.

4. Detection efficiency

Detector efficiency was determined using a Cf-252 source consisting of 13 Cf-252 needles fabricated in 1974. The BNL Radiological Control Division used a SWENDI 2 detector (calibrated at the NIST, Test number 269453-04/44060C) to determine the absolute neutron intensity. It was found that the neutron output was within 5% of that calculated at the time of calibration from the original Cf-250 and Cf-252 masses provided by the manufacturer. The source intensity applied to the calibration is that calculated at the times of measurement.

Figure 3. Cumulative fraction of Cf-252 neutron spectra, source on axis, as a function of neutron energy for two detector separation distances. The spectrometer has an increasing efficiency with lower neutron energy. The calculated time-of-flight energies account for the fact that the most probable scattering probability for the detectors in close proximity, 4 cm separation, is at a neutron scattered angle of 45° .

Detection efficiency is a function of the distance between the detectors. The efficiency is also a function of photomultiplier gains and the threshold settings on the constant fraction discriminators. However these parameter values were held constant after the system was optimized using Bicron 501A scintillators, 2.5 and 5.1 cm thick respectively, viewed by linear focus photomultipliers obtained from Bicron.

The efficiency of the detector configurations for Cf-252 neutrons over the useful detection range is:

Detector configuration	Intrinsic efficiency
Double scatter - 4 cm separation	0.087
Double scatter - 15 cm separation	0.017
SWENDI 2 calibrated by NIST*	0.0019

Table 1. Intrinsic detection efficiencies of the double scatter neutron spectrometer with the Bicron type 501A scintillators separated with center to center distances of 4 and 15 cm. The scintillators were 6 cm and 13 cm in diameter, and 2.5 and 5.1 cm thick, respectively.

*The SWENDI 2 intrinsic efficiency only applies to the geometry used for calibration by NIST where the detector area is calculated from its diameter times its height.

5. Cosmic-ray detection

We first compare Cf-252, Cs-137 gamma ray, and cosmic ray detection for the detector planes in contact with each other where the center to center distance between them is about 4 cm. This configuration is instructive because it is clearly the most efficient for double scatter (most of the scattered neutrons will travel from the first to second detector), Cs-137 source photon double scatter should be well defined (flight path differences of scattered photons are sub-nanosecond in time), and cosmic ray background will be efficiently detected. Results are shown shown in Figs. 4 and 5.

Figure 4. Response for detection planes in contact. Cf-252 source 30 cm from first detector. The ordinate is the detector count rate, where the Cf-252 neutron count rate was 209,000 counts/sec. Note that the Cs-137 gamma-gamma data becomes dominated by the cosmic ray signal at times far from the coincidence peak.

Figure 5 . Cosmic-ray and Cs-137 data taken 4 and 30 cm separation distances, Cs-137 source located on axis.

It is first observed that there is little difference in dispersion of the Cs-137 data with separation distance and that difference, > 1 ns, is consistent with the flight time of a photon over 30 cm. It is also observed that there is a modest dispersion of the cosmic ray data with distance of separation, which is inconsistent with that expected for high energy neutrons (close to the peak), see Fig. 1. We believe that a significant portion of the high energy signal is due to detection of muons; we observe muon signal in each detector. We will show later in this manuscript that the derived neutron spectra at lower neutron energy are in agreement with each other and Goldhagen ().

6. Cf-252 Neutron spectroscopy:

The response of the neutron double scatter spectrometer is complicated by the effects of multiple scattering in the first detector and the change of path length in the second with scattering angle. The energy response can be approximated by computing the probability of proton scatter in both detectors of the form $(1 - \exp(-x_i/\lambda_p(E)))$, where x_i refers to the detector thickness and $\lambda_p(E)$ is the mean free path for proton scattering as a function of energy. For axial Cf-252 response, we have chosen data from our longest flight path separation distance, 45 cm as shown in Fig. 6.

Figure 6 . Neutron fluence ($\text{n}/\text{cm}^2 \cdot \text{sec}$) data compared with the calculated Cf-252 Maxwellian distribution spectrum for a kinetic temperature of 1.43 MeV. Neutron energy derived by time-of-flight has been multiplied by 1.1 to approximate corrections for angular effects and for energy loss from the neutron collision in the first detector.

The agreement between the data corrected for detector efficiency (using first order theory) and normalized theory is encouraging, although Monte Carlo calculations need to be done to quantify the results. At lower energies, the spectrometer response is less certain because of statistical fluctuations in the data and because the efficiency is strongly influenced by the first detector PM gain and by the constant fraction discriminator threshold.

Neutron transmission spectroscopy can also be used to identify materials, or at least their signatures, located between the neutron emitter and the detector. The obvious example is high explosive surrounding plutonium. Low Z materials, such as carbon, nitrogen, and oxygen have defined neutron resonance structure in the 0.1-10 MeV energy range. In Figure 7, there is a comparison of double-scatter data taken from Cf-252 with a sugar target about 10 cm thick (a weapon surrogate) and the time-of-flight spectrum of an underground nuclear detonation measured at 20 meters.

Figure 7. Comparison of the neutron double scatter derived spectrum with the time-of-flight measurement of a nuclear detonation. The depression of the weapon data from 2-6 MeV is due to resonance structure in oxygen and carbon. The double scatter derived spectrum of the weapon surrogate is in good agreement with the published weapon data.

7. Cosmic-ray spectra

Cosmic-ray spectra derived from the spectrometer response must include the isotropic nature of the source. The highest probability of scattering in the first detector plane resulting in detection at the second occurs at $\phi = 45^\circ$ (2,3). Thus, to a first approximation, the average energy of the scattered neutron obtained by time-of-flight is half the incident cosmic-ray neutron energy. Useful data were obtained up to 1 MeV and 3 MeV for the 15 cm and 30 cm detector separations, respectively, where the tof signals began to arise from physical processes other than neutron reactions, such as muons passing through both detectors. Detection count rates limited the data range at lower energies, where the count rates were so low that about one week data collection time was necessary, and the results still had significant statistical limitations.

A comparison between our measurements and EML's for the shape of the cosmic-ray spectra, shown in Fig.8, are quite good. Below 1.0 MeV, it is expected that the shape of the spectra should be similar as it transforms into the equilibrium slowing down spectra anticipated in homogenous scattering media of the air ($\Phi(E_n) = k/E_n$). There are clearly defined resonance structure "dips" from air transmission in the EML data not easily observed in ours where the statistical fluctuations are less than 15%. On the other hand, our experiments were carried out on the 5th floor of Stony Brook University's Chemistry building so there may be a significant effect of the concrete walls.

Figure 8. Neutron double scatter spectrometer results transformed to energy as the variable (rather than time) for cosmic rays and Cf-252 plotted and then compared with cosmic-ray measurements by EML, at sea level. The BNL data is normalized to the EML spectra at 1.0 MeV.

At lower energies, the measured and calculated Cf-252 spectra are clearly different from the cosmic-ray spectra. Cf-252 neutron emission by spontaneous fission, a spallation process, is generally described by the Maxwellian distribution, of the form:

$$\Phi(E_n) = C E_n^{0.5} \exp(E_n/kT) \quad (4)$$

Where the kinetic temperature, kT , has been determined to be 1.43 MeV. Below 1 MeV, the Cf-252 spectrum varies with the square root of neutron energy, whereas the cosmic ray data varies with inverse of neutron energy. Thus the spectra are separable. Above 1 MeV, the Cf-252 and cosmic ray data converge. This is because the dominant mechanism for producing cosmic-ray neutrons in the energy range 1.0-10 MeV is also a spallation process.

8. Cosmic-ray spectrum over full neutron energy range

The integral of the cosmic-ray spectrum over its measured range is shown below with the calculated Cf-252 integral: In the energy range 0.1-8 MeV, the cosmic-ray neutrons are perhaps 25% of the total, the calculated and measured Cf-252 neutrons are > 80% of the total. The calculated Cf-252 integral agrees with our measurement given in Figure 1. Thus it is observed that most of the cosmic-ray neutrons are not included in the energy range used for detection of the Cf-252.

Figure 9. Measured neutron cosmic-ray integral Vs. neutron energy (Goldhagen, private communication) at sea level, and that calculated for Cf-252 neutrons using a kinetic temperature of 1.43 MeV in a Maxwellian distribution.

9. Conclusions

We have shown that signal-to-noise for detection of spontaneous fission neutrons is favored by limiting detection response to a small fraction of the cosmic-ray background energy range. This can be achieved by using a neutron spectral measurement device such as the neutron double scatter spectrometer. . Our measurements demonstrate that when there are statistically significant count rates, the spectra of neutrons from a spontaneous fission source (Cf-252) and those of neutrons produced in the air by cosmic-rays differ in the neutron energy range 0.1-1.0 MeV. This difference may be used to separate the phenomena. The double scatter data may be more accurately unfolded into neutron spectra with the use of Monte Carlo calculations; however, our first order calculations provides an unfolded spectrum which matches the known energy distribution of Cf-252 neutron emission. Moreover, our unfolded spectra showed the proper transmission signature of a nuclear weapon surrogate source. These signatures are can contribute to assessing the nature of neutron emitting source. We have not completed the full spectrometer electronics package so have not implemented the direction measurements based on single event analysis of both the energy deposition in the first detector and the scattered neutron energy. However, feasibility for these measurements has already been demonstrated by Preszler et. al, (2,3) for higher energy fusion diagnostic experiments.

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