We have repeated the experiment of Taleyarkhan et al. [Science 295, 1868 (2002)] in an attempt to detect the emission of neutrons from $d$-$d$ fusion during bubble collapse in deuterated acetone. Using the same cavitation apparatus, a more sophisticated data acquisition system, and a larger scintillator detector, we find no evidence for 2.5-MeV neutron emission correlated with sonoluminescence form collapsing bubbles. Any neutron emission that might occur is at least 4 orders of magnitude too small to explain the tritium production reported in Taleyarkhan et al. as being due to $d$-$d$ fusion. We show that proper allowance for random coincidence rates in such experiments requires the simultaneous measurement of the count rates in the individual detectors.

The discriminator threshold for the sonoluminescence light detector was set up as specified in Ref. [6]. Our neutron detector threshold, set by a fast discriminator, was calibrated by comparison with the pulse-height spectrum from a Pu-Be source available during the experiment. The relative response to a Pu-Be source and a $^{60}$Co source was checked prior to the experiment. The $^{60}$Co Compton edge (equivalent to a 1-MeV electron) was found to be at a fraction 0.29 of the Pu-Be edge. The threshold used in the experiment was at an equivalent electron energy of 270 keV. Using published [8] curves of light output, we estimate our threshold to have been at an equivalent neutron energy of 1.2 MeV, well below the threshold for 2.5-MeV neutron detection. Approximating the geometry of our detector as a slab of area 285 cm$^2$ and average thickness 10 cm, we obtain a rough estimate of its intrinsic...
efficiency. The mean free path of a 2.5 MeV neutron in NE213 is 5 cm so the probability of an \( n-p \) scattering (0.57) is given by the total cross-section ratios for H and C corrected for their relative abundance in NE213 [9]. Correcting this probability to allow for our finite threshold (a factor of 0.52) gives an estimated efficiency of 0.3. We estimate the uncertainty in this figure to be about a factor of 2.

The data acquisition system recorded three types of events: \( n\gamma \) singles: any pulse from the \( n\gamma \) detector above the fast (lowest) threshold; SL singles: any pulse from the SL detector above the threshold; SL-\( n\gamma \) coincidences: defined by a \( \pm 10 \mu s \) window triggered by the SL signal.

For each event listed above, the following information was digitized and recorded: the time relative to the last SL and SL detector above the threshold; SL-\( n\gamma \) coincidences: defined by a \( \pm 10 \mu s \) window triggered by the SL signal.

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For each event listed above, the following information was digitized and recorded: the time relative to the last PNG trigger pulse using a 1-MHz clock (1 \( \mu s \) time resolution); the pulse heights from the SL and \( n\gamma \) detectors; an \( n\gamma \) discrimination signal; for each coincidence event the time difference between the \( n\gamma \) and SL signals was recorded with time resolution near 10 ns.

The nominal output of the PNG was \( 10^6 \) n/s (\( \sim 5000 \) n/pulse). The pulse width from the PNG was measured to be about 12 \( \mu s \). Thus, the event rate seen by the electronics during the PNG pulse was high and would have resulted in unacceptable counting losses in the period immediately after the start of the neutron pulse. To avoid this, a blocking signal was used to veto all counts from the \( n\gamma \) detector for 20 \( \mu s \) after the onset of the PNG trigger pulse. In addition, the event rate from \( n\gamma \) singles was reduced by looking only at every eighth pulse, thereby reducing the dead time for the data acquisition systems to \( \leq 3\% \). The division by eight was not applied to the pulses used to identify the coincidences.

Figure 2 shows the distribution of events recorded during the experiment, as a function of the time from the preceding PNG pulse. No \( n\gamma \) discrimination is activated for these plots.

The background seen by the \( n\gamma \) detector has three components, which have different time structure (Fig. 2) and pulse-height spectra (Fig. 3). In region A during the 12 \( \mu s \) PNG pulse, there is the isolated peak due to 14 MeV neutrons, which we normally block out. In the absence of blocking the instantaneous rate would have been \( \sim 10^6 \) s\(^{-1}\). The pulse-height spectrum from this component, taken with reduced PNG intensity, is shown in Fig. 3(a). Regions B and C, immediately after the PNG pulse, show an exponentially decaying tail (Fig. 2). These events we identify as essentially all gammas, resulting

![FIG. 2. Event occurrence time relative to PNG trigger time recorded for \( n\gamma \) singles, SL light emission singles, and the SL-\( n/\gamma \) coincidences. The data shown on the right are with an expanded time scale. Parts (a) and (b) show data taken with cavitation and (c) and (d) data taken without cavitation. The figure represents counts accumulated in \( \sim 1h \) runs. We look at the data rates in four distinct regions marked in the figure as A, B, and D and 62 \( \mu s \leq C \leq 539 \mu s \) (not marked)
from slow scattered (room return) neutrons being captured on various nuclei in the scintillator and surrounding material. The instantaneous count rate in this region is as high as 4000 s$^{-1}$ immediately after the PNG pulse, dropping off with a time constant $\sim 140 \mu$s. This component we expect to scale with the PNG output. Finally, in region $D$, beyond 620 $\mu$s there is constant ($\sim 150$ counts/s) background composed mostly of $\gamma$s ($\approx 96\%$) due to fixed contamination, etc, unrelated to the PNG output.

Two runs were made, one with cavitation (i.e., the acoustic drive applied to the acetone chamber), and one without. In the 65-minute run with cavitation, 51 coincidences between pulses from the SL detector and the $\eta\gamma$ detector were observed; in the 58-minute run without cavitation, four coincidences were observed. These numbers are consistent with the expected random rates. Figure 2 shows the time distribution of singles and coincidences relative to the start of the PNG trigger pulse for the two runs. The coincidence event timing is defined by the neutron signal. Note that even with no cavitation there are light signals seen during the PNG pulse (region $A$). These are therefore not true SL signals.

The SL events during cavitation occur in four distinct regions shown in Figs. 2(a) and 2(b). The observed singles count rates in the $\eta\gamma$ detector within these four regions are quite different and must be known to calculate the expected random coincidence rates within the 20 $\mu$s coincidence window. Table I shows the results of such calculations. The number and time distributions of observed coincidences agree with the calculated random coincidence rates.

Figure 4 shows the distribution of time intervals between SL and $\eta\gamma$ pulses that were in coincidence (within 10 $\mu$s of each other). As expected for random coincidences, there is no evidence for any clustering of events in time.

Note that the $\eta\gamma$ events in region $A$ (Fig. 2(b)) were almost entirely eliminated by the 20 $\mu$s blocking pulse. In the absence of the blocking, essentially 100% of the SL signals in region $A$ would fall within $\pm 10 \mu$s of an $\eta\gamma$ count occurring during the PNG pulse and register as an $\eta\gamma$-SL coincidence. These 400 coincidences would also exhibit a peaked time structure defined by the shape of the PNG pulse, unlike the others, shown in Fig. 4 that are scattered randomly throughout the 20 $\mu$s coincidence window. The coincidence technique used in Ref. [6] would be dominated by such events, which must be measured and then subtracted before any real coincidence events can be inferred from the data.

Taleyarkhan et al. [6] report seeing more neutrons in their singles data during runs with cavitation as compared to those without. We have examined our data to look for this effect and note a difference of 740 $\pm$ 290 counts in the 20 to 220 $\mu$s period following the PNG pulse in our $\eta\gamma$ singles during the run with cavitation as compared to the

![FIG. 3 (color online). Pulse-height spectra recorded with the same $\eta\gamma$ detector settings. (a) shows a 14 MeV neutron spectrum taken with reduced output from the PNG and no blocking signal. The 14-MeV edge is around channel 200, the threshold around channel 8/9. The pulse-height spectra taken during data runs with blocking turned on are shown in (b). The spectrum from all events (solid line) show a distinctive edge around channel 50, consistent with 2.2 MeV n-p capture $\gamma$s.](image-url)

![FIG. 4 (color online). Distribution of time intervals between SL and $\eta\gamma$ pulses. The original data had much higher dispersion, 5 ns/channel, and showed no structure on that time scale.](image-url)
run without cavitation. This difference amounts to about 2.5% of the room return background component and is significant if one considers only counting statistics as a source of uncertainty. A more detailed view of this difference is shown in Fig. 5(a). The statistical uncertainties are also indicated. The solid line shows the expected difference if the room return background during cavitation were to be increased by 2.5%, as might be expected from a variation in the PNG intensity (which was not monitored contemporaneously), or possibly a change in neutron scattering geometry due to people in the area being at different positions for the two runs.

The data shown in Fig. 5(a) were for our lowest threshold (~1.2 MeV neutrons). In Fig. 5(b) we show the difference spectrum for the data rescaned with a higher threshold of 2.8 MeV neutrons applied to the nγ pulse-height spectrum. The data show essentially the same features, implying that the excess counts are due to events that deposit considerably more energy than 2.5-MeV neutrons, and consistent with the background due to n-p capture gammas.

From these data, we can put an upper limit on the number of 2.5-MeV neutron events correlated with the SL light that might be in our data. This limit is approximately 100 counts (see Fig. 5(a)) accumulated in about 1 h. Using our estimate of detector efficiency for 2.5-MeV neutrons (0.3), applying the solid angle factor appropriate for the detector geometry (2.5 × 10^{-2}), and allowing for transmission through the acetone, refrigeration material, and enclosure walls (0.35), we estimate the efficiency for detection of 2.5-MeV neutrons emitted from the acetone to be 2.5 × 10^{-3}, which would correspond to a source rate ≤ 100 neutrons/s, about 4 orders of magnitude below the rate implied by the tritium production reported in Ref. [6].

In the period during the PNG pulse the time resolution of the n-γ electronics deteriorated due to the high instantaneous count rate. This resulted in poor n-γ pulse shape discrimination in the period immediately after the PNG pulse. This was not the case in the time region between 700 and 1700 μs (see Fig. 2(a)) where about 80% of the SL light pulses appear. They appear in our data as sharp peaks 5 μs wide (FWHM) separated by the acoustic wave period of 52 μs, and would be an interesting area for future study. In this time range no excess neutron yield was detected for runs with cavitation and we did not observe any correlation between the neutron yield and the emitted SL light pulse.

We conclude that there is no evidence of any real coincidences in this experiment. Furthermore, there is a substantial random coincidence rate, which must be properly allowed for in any attempt to identify real coincidences. The singles rates seen by both detectors must be measured contemporaneously in order to properly evaluate the random coincidence rate.

The observed excess of nuclear radiation seen during the run with cavitation appears to be due to a ~2.5% increase in room return background. It prevails at pulse heights corresponding to neutron energies in excess of 2.5 MeV, and is consistent with the characteristics of n-p capture gammas.

We also note the intriguing time dependence seen in the SL detector.

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