Upper Bound for Neutron Emission from Sonoluminescing Bubbles in Deuterated Acetone

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An experimental search for nuclear fusion inside imploding bubbles of degassed deuterated acetone at 0 °C driven by a 15 atm sound field and seeded with a neutron generator reveals an upper bound that is a factor of 10 000 less than the signal reported by Taleyarkhan *et al.* The strength of our upper bound is limited by the weakness of sonoluminescence, which we ascribe to the relatively high vapor pressure of acetone.

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Sonoluminescence [SL] is the phenomenon whereby pulsating bubbles focus diffuse sound energy by 12 orders of magnitude [1] to generate extremely short flashes of ultraviolet light [2–5]. For a bubble in water, the maximum observed photon energies of ~6 eV are limited only by the extinction coefficient of the liquid. So, for SL, what are the limits of energy focusing? If the energy density inside a collapsing bubble is concentrated by yet an additional factor of 1000 and if the bubble contained deuterium gas, then the kinetic energy of the individual ions would be sufficient to generate thermonuclear fusion [2,6]. Experimentally, the search for fusion from collapsing bubbles is facilitated by gating on individual flashes of light, which dramatically reduces background.

Observation of thermonuclear fusion generated by cavitation in deuterated acetone has been reported by Taleyarkhan and co-workers in papers [7–9] that appeared in 2002, 2004, and 2006. Here we describe our unsuccessful attempts to reproduce the claimed effect. Shapira and Saltmarsh [10], Tsoukalas *et al.* [11], and Saglime [12] have also reported null results.

Taleyarkhan's apparatus has the meritorious feature that the fluid was excited with sound fields that have a dynamic pressure of 15 atm, which is about 10 times stronger than in standard SL [3–5]. If a clean fluid is excited to a sound field amplitude of 15 atm [13,14], a small input of localized energy, such as a neutron knockon event or a pulse of light from a laser, is sufficient to generate a tiny [~100 nm] cavitation seed which can be expanded to ~1 mm by the rarefaction phase of the sound field. This bubble then implodes—supersonically—down to solid density, where it becomes so hot and stressed that a flash of light can be emitted. According to Taleyarkhan, this flash is sometimes accompanied by fusion when the host fluid is deuterated acetone at 0 °C under an ambient pressure determined by its vapor pressure.

A block diagram of the apparatus is shown in Fig. 1. An acoustic resonator was driven by a ring piezoelectric (PZT) [Channel Industries, Santa Barbara] glued to the Pyrex glass [Continental Glass, Los Angeles]. The resonator was filled with acetone-*d*6, cooled to ~0 °C, and the free surface was maintained at the vapor pressure. Inside the resonator were 2 plungers with thin glass walls [1 mm thick]; their interior was also maintained at vapor pressure. When ~500 V was applied to the ring PZT at the resonant frequency [~20 kHz, 0 °C, acetone-*d*6], the dynamic pressure reached 15 atm. A pulsed neutron generator (PNG) [thermoelectron P211] provided pulses of neutrons [nomi-

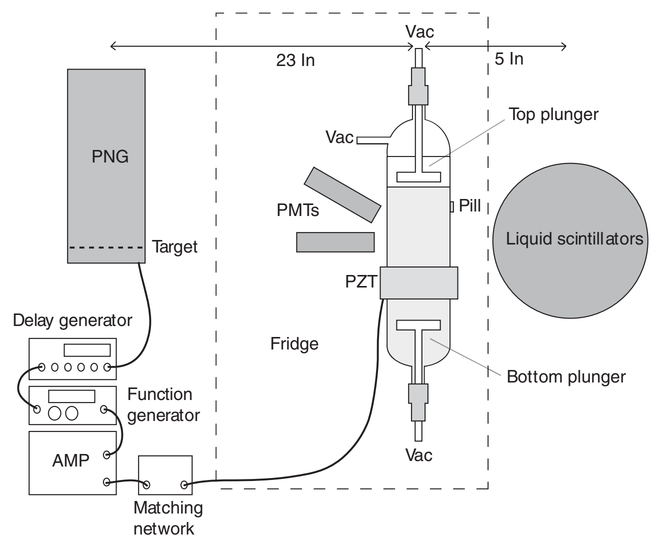

FIG. 1. Block diagram of the apparatus used to study neutron generated cavitation in acetone-*d*6. Length, width, thickness, and curvature of the acoustic resonator and plungers were chosen to match blueprints provided by Taleyarkhan [29]. Accordingly, in early runs the top plunger was suspended with a thin wire so that it touched the free surface. In most runs, however, the top plunger was near to but not in contact with the free surface. This resulted in more stable acoustic modes and avoided excessive breakage. Metal press vacuum seals were used top and bottom. Wax tape and vacuum grease were used to seal the upper and lower glass pieces. Acetone was degassed *in situ* with the sound field on using a rotary vein vacuum pump. During the last stage of the degas procedure, the PNG generated cavitation in the fluid. Cooling and degassing could lower liquid level by 4 cm. Depending on the gas concentration, neutron seeded cavitation could result in streamers or popping. All of the data presented herein are from the popping mode.

nal width 10 μs] which seeded cavitation. The flux of neutrons [1000 n/pulse at 30 Hz on the lower setting] could be varied by adjusting the accelerating potential for the deuterium ions inside the PNG tube which create 14.1 MeV neutrons as a result of striking a tritium target.

Data were acquired on 4 channels with a digitizer board [Acqiris DC270] with a bandwidth of 250 MHz [1 GSa/s]. Two channels recorded the response of 5″ photomultiplier (PMT) tubes [Hamamatsu RA2350] that read out the response of 5″ diameter liquid scintillators [Bicron 501A]; one channel read out the signal from a PMT [Hamamatsu R2027] that recorded SL from the collapsing bubble; the fourth channel recorded the sound wave or shock wave with a pill microphone mounted on the outside wall as seen in Fig. 1. For some data runs, the fourth channel was used for a second SL PMT. All data were stored to disk for offline analysis, following techniques described in Ref. [15].

Figure 2 shows the sequence of events underlying attempts to observe fusion in a collapsing acetone-$d6$ bubble. The PNG neutron-induced scintillation events in the top trace are seen to be coincident with the seeding of the bubble, which expands during the rarefaction portion of the acoustic cycle as recorded by Mie scattering [labeled "radius" on the y axis]. At the lower PNG setting, about 10% of the PNG pulses seeded a cavitation event with a collapse

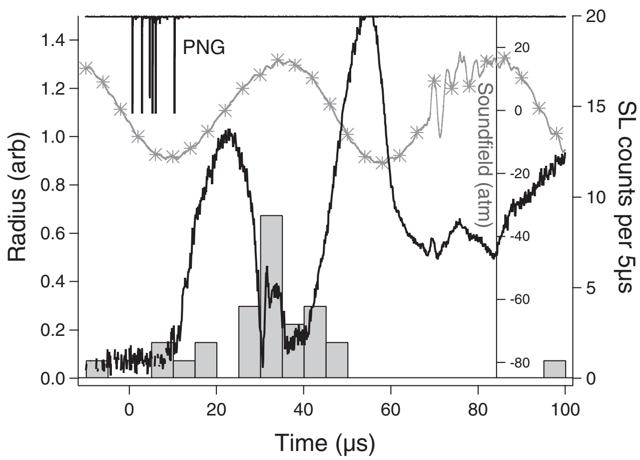

FIG. 2. Sequence of events leading to neutron seeded cavitation in a fluid driven by a 15 atm sound field. The top trace labeled PNG is from one of the two neutron detector channels. The starred trace is the pill microphone signal. The solid black line is the square rooted and normalized light scattering signal. The histogram of SL events was obtained with a bias of 1200 V applied to the SL PMT and corresponds to 1000 triggers, at this gain [single photon level ~50 mV] after pulsing with a 300 ns delay could reach 50% and was rejected out in software. The histogram width is due to the width of the PNG pulse as well as the range of times when the sound field is sufficiently negative to expand a seeded nanobubble. Light coincident with the PNG is due to Cherenkov radiation from electrons which are excited by gammas from nuclear interactions with 14 MeV neutrons. We ascribe the bounce in bubble radius, after the main bubble collapse, as being due to vapor trapped inside the bubble.

strong enough to make a shock wave, which appeared about 65 μs later in the sound field recorded by the pill microphone. The histogram shows the phasing of SL relative to the electronic trigger for the PNG, which is $t = 0$ for all traces.

In an experimental run, an 8 ms sweep was acquired under the condition that the PNG had fired and that a cavitation shock wave had been recorded within a 200 μs window. The firing of the PNG was synched to the phase of the sound wave. Background data were acquired 3 ms prior to $t = 0$ and signal data for 5 ms after $t = 0$.

Calibration of the single photon level [Hamamatsu R2027] at 1000 V bias reveals a peak at 15 mV, so the signal threshold was set at 10 mV. Neutrons were separated from gammas via pulse height discrimination. A scintillation event is deemed to occur from 100 ns before until 500 ns after the peak in the PMT response. The signal in the "tail" is integrated from 20 to 500 ns after the peak. The right inset in Fig. 5 displays gammas and neutrons, discriminated according to integrated light in the tail versus total light. The range [abscissa] of displayed scintillations corresponds to 300–2000 keV *e.e.* [light made from electrons with energy 300–2000 keV]. The 662 keV gamma ray emitted by Cs 137 creates, in a single collision, electrons with a maximum energy of 478 keV [Compton edge] which creates as much light as the peak of the spectrum of proton recoil from a 2.45 MeV neutron [Fig. 3(b) of Ref. [15]]. The window for acceptance of 2.5 MeV neutrons is drawn in the figure. Given our geometry, the detection efficiency for neutrons in this window is about 1%. The sound field was calibrated by noting the voltage at which neutrons [from a 1mCi AmBe source] could be observed to seed cavitation [13,14,16] at rates in excess of 1/minute. At twice this voltage, the sound field was approximately 15 atm. At this drive level, degassed acetone kept at its vapor pressure [70 torr at 0 °C] produced very scarce SL. For about 7300 cavitation events [2300 at the low PNG setting and 5000 at a neutron flux 100 times higher], about 87 SL events were observed, all occurring during the first collapse of a neutron seeded bubble. Adding small amounts of air increases the SL yield. At about 20 torr of air, the observed SL signal was similar to that reported by Taleyarkhan *et al.* [8]; see Fig. 3. In this case about 58 000 SL coincidences between 2 PMT's were observed for 1500 8 ms sweeps. Most of the SL occurred in a temporal region beginning a few acoustic periods after the first collapse [10]. At about 40 torr and higher air pressures, some neutron seeded events resulted in "streamers" such as those reported by Taleyarkhan *et al.*

In none of the cases where 2 PMT's recorded an SL event was that event coincident with a neutron within a 1 μs window. There is only one event [Fig. 4] where a neutron was coincident with the response of a single SL PMT within the ~10 ns window that would characterize a bubble fusion event. We propose that claims of new routes to fusion should be backed up with coincidence data of the type presented in this figure.

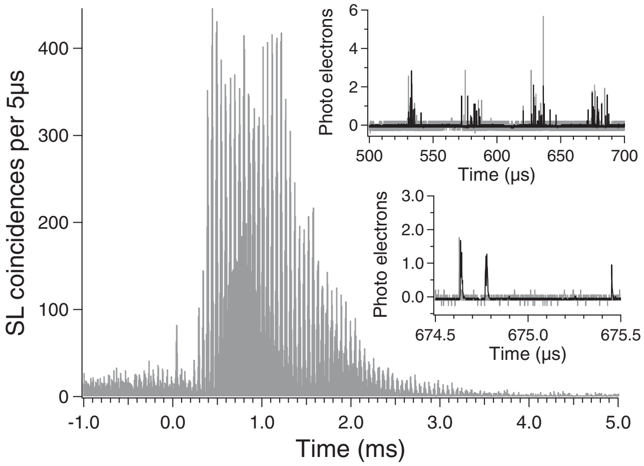

FIG. 3. Histogram of 58 000 SL coincidences for 2 PMT's excited above single photoelectron level, for 1500 8 ms sweeps. For a single tube there were 175 000 events above background. The insets show an example of single sweep data for the two SL PMT's (black and gray) on the scale of an acoustic cycle and within a single sound cycle.

The absence of neutron-SL coincidences means that at the 90% confidence level and for a 1% neutron detector efficiency there are less than 230 physical coincidences. For the 20 torr air run, this means less than 230 n/58 000 SL or less than ~1 n/250 SL. For the vapor pressure run, it corresponds to less than 3 n/SL. This relatively weak upper bound is due to the dearth of SL for well degassed acetone.

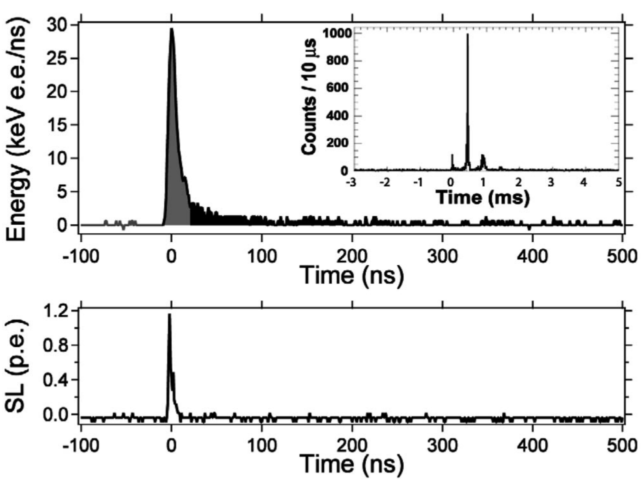

FIG. 4. Real time coincidence between neutron count and one SL PMT. The energy of the neutron scintillation corresponds to 582 keV *e.e.* and occurs 9.6 μs after the turn-on of the PNG. We interpret this event as a PNG neutron coincident with light from Cherenkov radiation in the acetone due to the same PNG neutron pulse. The neutron trace shows in black the region used to calculate the tail area for the pulse shape discrimination. The inset shows SL events for a cylindrical resonator filled with filtered [0.2 μm] degassed acetone-*d*6, pressure released to the vapor pressure, run at 15 atm sound, and seeded by an AmBe source.

Next we turn our attention from coincidences to the total neutron emission [Fig. 5]. For the degassed system with a low number of SL events, 2300 sweeps generated a total of 9 neutron counts in the 2.5 MeV acceptance box from the second acoustic cycle to the end of the sweep [~5 ms]. This is the same as the background of 5 neutrons in the 3 ms before the PNG seed for the same 2300 sweeps. Most of the background is at the lower threshold where gammas and neutrons blur together. In real time, the cosmic contribution to the background—above the window—yields ~1 count/8 sec per scintillator. At the 90% confidence level, the number of putative signal neutron counts which could generate a neutron count equal to the background is less than ~10 [17]. Given our 1% detector efficiency, this would correspond to an upper bound of less than 1 neutron per two 5 ms sweeps.

Using an acoustics resonator whose design is based upon the specifications of Taleyarkhan, we have found that fusion inside of a collapsing bubble in deuterated acetone is zero within our experimental accuracy. The upper bound established in runs reported here corresponds to a value which is over 4 orders of magnitude smaller than the 10 000 neutrons per each 5 ms sweep following a neutron seeded cavitation event as claimed by Taleyarkhan *et al.* [7–9]. Negative results were also found in runs [18] where neutron seeds, for cavitation in a filled cylindrical resonator running at 15 atm, were provided by an AmBe source.

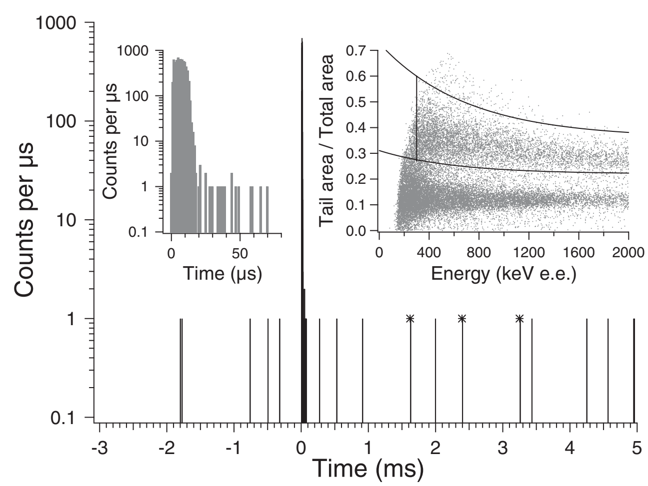

FIG. 5. Neutron counts received during data acquisition for 2300 neutron-induced cavitation events at low PNG setting for 0 °C degassed acetone-*d*6 excited with a 15 atm sound wave. The left inset shows the temporal structure of the PNG pulse. The right inset shows the gamma/PNG discrimination for all of the data in this figure. The upper branch is due to neutrons, and the lower branch is due to gammas. The experimental acceptance window for 2.5 MeV neutrons is indicated on the upper branch. The accuracy of the pulse shape discrimination is affected by overlap of the tail and rise of successive closely spaced neutron-induced scintillation events as well as the dc offsets due to cross talk with the high acoustic drive [30]. Events with an asterisk correspond to background neutrons with energy higher than the 2.45 MeV acceptance window.

These runs employed degassed acetone at the vapor pressure and 1 atm ambient pressure. In the former case, ~4000 SL were observed in 2200 triggers [Fig. 4 inset] and in the latter case over 50 000 SL were observed in 3000 triggers.

For degassed acetone [running at the vapor pressure], the SL signal is very weak, and we cannot rule out the possibility that in the limit of perfect degassing SL also vanishes. We ascribe the differences of the SL signal [Fig. 3] and the data reported in Ref. [8] to the amount of dissolved gas. In the limit of small amounts of gas, SL becomes particularly complex and even depends upon the resonator design. For the filled cylindrical resonator that is pressure released, we have observed a strongly peaked SL signal about 13 acoustic cycles after seeding [inset in Fig. 4]. This could be a signature of bubble cloud collapse [19,20] which has been cited [21] as a possible route to acoustic fusion.

The null result reported here does not change our opinion that the search for cavitation driven thermonuclear fusion is a worthwhile high risk endeavor. Although high vapor pressure fluids such as acetone are easily cavitated with neutrons, they are historically known to display very weak SL [22,23]. The parameter space for SL includes possibilities for energy density concentration that are *a priori* more promising than acetone to search for fusion: Single bubble SL from low vapor pressure fluids such as sulfuric acid can be driven so as to emit 1000 times more light than bubbles in water [24]. At low sound, fields where light emission is weaker than for bubbles in water spectral lines from Ar^+, with an upper level which is 37 eV above ground state, can be seen [25]. Furthermore, in water driven at 1 MHz, the spectrum matches thermal bremsstrahlung emission from a million degree plasma [26]. Also, in phosphoric acid [27] flashes with a peak power in excess of 100 W have been observed [28].

Our experiments imply an upper bound that is 10 000 times smaller than Taleyarkhan's signal. We believe, however, that a neutron signal of only 1% of this upper bound would be very interesting, since a tantalizing and frustrating aspect of the search for new routes to fusion is discerning a weak signal in the tail of an exponential distribution. Small increases in bubble temperature would lead to huge increases in the rate of *D-D* fusion.

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[1] B. P. Barber and S. Putterman, Nature (London) **352**, 318 (1991).
[2] S. Putterman, Sci. Am. **272**, No. 2, 46 (1995).
[3] B. P. Barber *et al.*, Phys. Rep. **281**, 65 (1997).
[4] S. Putterman and K. Weninger, Annu. Rev. Fluid Mech. **32**, 445 (2000).
[5] M. P. Brenner *et al.*, Rev. Mod. Phys. **74**, 425 (2002).
[6] B. P. Barber *et al.*, Phys. Rev. Lett. **72**, 1380 (1994).
[7] R. P. Taleyarkhan *et al.*, Science **295**, 1868 (2002).
[8] R. P. Taleyarkhan *et al.*, Phys. Rev. E **69**, 036109 (2004).
[9] R. P. Taleyarkhan *et al.*, Phys. Rev. Lett. **96**, 034301 (2006).
[10] D. Shapira and M. Saltmarsh, Phys. Rev. Lett. **89**, 104302 (2002).
[11] L. Tsoukalas, T. Jevremovic, F. Clikeman, M. Bertadano, J. Walter, A. Bougaev, and E. Merritt (to be published).
[12] F. Saglime III, MS thesis, RPI, 2004.
[13] C. West and R. Howlett, Br. J. Appl. Phys., J. Phys. D **1**, 247 (1968); Nature (London) **215**, 727 (1967); Acustica **21**, 112 (1969).
[14] M. Greenspan and C. Tschiegg, J. Res. Natl. Bur. Stand. Sect. C **71**, 299 (1967).
[15] B. Naranjo, J. Gimzewski, and S. Putterman, Nature (London) **434**, 1115 (2005).
[16] D. Lieberman, Ph.D. thesis, UCLA, 1958.
[17] G. J. Feldman and R. D. Cousins, Phys. Rev. D **57**, 3873 (1998).
[18] "Horizon, An Experiment to Save the World," BBC2, February 17, 2005.
[19] T. Ikeda *et al.*, J. Acoust. Soc. Am. **116**, 2508 (2004).
[20] G. E. Reisman, Y. Wang, and C. E. Brennen, J. Fluid Mech. **355**, 255 (1998).
[21] I. S. Akhatov, R. I. Nigmatulin, and R. T. Lahey, Jr., Multiphase Sci. Technol. **17**, 225 (2005).
[22] A. Chakravarty and A. Walton, J. Lumin. **92**, 27 (2000).
[23] G. E. Vazquez and S. Putterman, Phys. Rev. Lett. **85**, 3037 (2000).
[24] D. J. Flannigan and K. S. Suslick, Nature (London) **434**, 52 (2005).
[25] D. J. Flannigan and K. S. Suslick, Phys. Rev. Lett. **95**, 044301 (2005).
[26] C. Camara, S. Putterman, and E. Kirilov, Phys. Rev. Lett. **92**, 124301 (2004).
[27] A. Chakravarty *et al.*, Phys. Rev. E **69**, 066317 (2004).
[28] B. Kappus *et al.*, in Proceedings of the Acoustical Society of America, Honolulu, 2006 (to be published).
[29] R. P. Taleyarkhan and C. West, U.S. Patent Application No. 20050135532, June 23, 2005; (private communication).
[30] See EPAPS Document No. E-PRLTAO-98-053706 for the neutron detector response to an AmBe source. For more information on EPAPS, see http://www.aip.org/pubservs/epaps.html.