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Energy and flux limits of cold-fusion neutrons using a deuterated liquid scintillator

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Deuterated liquid scintillator detectors (NE230), which give excellent n/γ discrimination and provide a direct measure of the neutron-*energy* spectrum, have been used to search for 2 to 3 MeV neutrons produced in $d+d$ cold fusion. The apparatus consisted of an electrolytic cell using high-efficiency inverted-well geometry. Several samples of annealed Pd wire and a Pd casting (up to 13 g) were studied over a period of several weeks. An upper limit of $\leq 10^{-3}$ fusion (n/s)/g Pd was obtained corresponding to $< 7\times10^{-24}$ fusion (n/s)/dd pair in our samples which excludes most of the reported positive results. Several sources of spurious signals, which could closely mimic signals from fusion neutrons, were also observed.

Shortly after the announced observation of cold fusion[1,2] by two groups in Utah claiming to have identified fast neutrons produced by $d+d\rightarrow {}^3\mathrm{He}+n$ ($E_n \cong 2.5$ MeV) we began a series of measurements to detect these neutrons. It is known that Pd (and Ti) can be heavily loaded with hydrogen or deuterium absorbed in the metal lattice and hence susceptible to fusion[3] if the normal ${}^1\mathrm{H}+{}^1\mathrm{H}$, ${}^1\mathrm{H}+{}^2\mathrm{H}$, or ${}^2\mathrm{H}+{}^2\mathrm{H}$ repulsion can be overcome. Like muon-catalyzed fusion,[4] an electron in the metal with a heavy effective mass, $m \geq 5m_e$, could induce fusion although such an effective mass is thought to be unreasonable for a Pd lattice. However, several mechanisms for deuteron acceleration in Pd are possible including deuteron emission with keV energies resulting from fracturing,[5] and high-electric-field "patch effects" due to local variations of conductivity. Such effects could perhaps cause acceleration of the deuterium nuclei to keV energies which is well above the threshold needed to induce measurable fusion. There is evidence that such cells exhibit electrical discharges (see below). Also associated with such a discharge might be the acceleration of electrons and, with them, deuterium ions. This collective acceleration mechanism can produce MeV-energy ions. Also, high-density electron discharges can cause the well-known ζ-pinch effect which in plasma-fusion devices can produce a large burst of neutrons. Alternatively, structural changes in the Pd lattice during Pd-^{2}H phase transitions can cause nonthermal vibrations which could conceivably lead to compression[3] of the absorbed deuterium. Since the type of cells used in our experiments often exhibit both electrical discharge and acoustical phenomena (see below), one cannot dismiss the possibility of detecting enhanced $d+d$ fusion in such a cell resulting from one or more of these or other mechanisms.

The $d+d$ fusion channel cannot be *a priori* excluded for very-low-energy fusion since the nucleus ^{4}He (Ref. 6) may have a broad, highly excited $J^\pi=0^+$ state above the $d+d$ threshold ($E_x=23.8$ MeV) which could be primarily a $d+d$ "breathing" mode. Such a level could mix with the known[6] $J^\pi=0^+$ level in ^{4}He at $E_x=20.1$ MeV, which is thought[7] to be mainly ${}^3\mathrm{He}+n$ and $t+p$, to provide enhanced $d+d$ fusion cross sections as $E_d\rightarrow 0$. Enhancement in $d+d$ fusion might occur since the configuration $d+d$ in ^{4}He, due to antisymmetrization, would have a *very* large rms radius, i.e., radial extent.[7] Enhancements due to interference of levels near threshold have been observed in many other low-energy nuclear-fusion and radiative-capture reactions, particularly those relative to stellar nucleosynthesis[8] such as ${}^{12}\mathrm{C}(\alpha,\gamma){}^{16}\mathrm{O}$.

Since low count rates were anticipated,[9-12] it was necessary to use a high-efficiency geometry, specifically a well-type geometry where the neutron detector can be placed in very close proximity to the sample (Fig. 1). Thus the Pd wire is wrapped around the inside glass well containing the neutron detector so that an emitted neutron has a high probability of hitting the central neutron detector. This also results in a minimum of fast neutron moderator (water) between the sample and the detector and hence permits the detection of the fast fusion neutrons directly. This geometry also allows one to use small detectors which minimize the cosmic-ray muon and other background.

The cell contained ca. 0.1M LiOD in $>99.8\%$ enriched D_2O (concentrations varied with evaporation and subsequent replenishment) contained within an annulus of inner diameter of 6.4 cm and outer diameter of 12.5 cm surrounding the detector (Fig. 1). A Pt wire anode wrapped the inner and outer walls in picket-fence fashion.

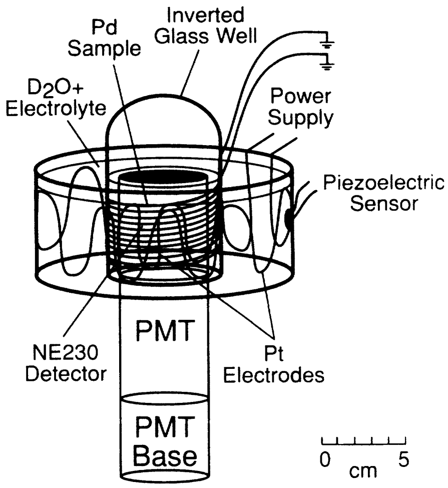

FIG. 1. Inverted-well geometry (muon-veto detectors not shown).

The Pd cathode was either extruded wire in a coil (1.25 mm thickness, $\approx$91 cm length, radius 9.2 cm) or a cast ring (2.14 mm thickness, radius 8.9 cm). Current densities were varied from 50 to 250 mA/cm^2. Piezoelectric sensors were attached to the outside of the cell to detect acoustical activity. The temperature varied in the cell from 33 to 49° C.

Since it was desired to measure a neutron-*energy* spectrum near $E_n \cong 2.5$ MeV, yet conventional time of flight (TOF) could not be easily employed, we opted to use Nuclear Enterprises NE230 deuterated liquid scintillator [manufactured by Nuclear Enterprises, Inc. (benzene-d6 solvent; 98.6% deuterated)] as the neutron detector. Like a conventional hydrogenated liquid scintillator, e.g., NE213, NE230 gives excellent pulse-shape discrimination to separate neutrons from γ rays. However, *unlike* the conventional liquid scintillators, e.g., NE213, fast neutrons are primarily detected using a recoil deuteron from $n+d$ elastic scattering. The cross section for the latter, even at $E_n \approx 2.5$ MeV in contrast to $n+p$ elastic scattering, is asymmetric[13] around $\theta = 90°$ with a large *backward* neutron-scattering peak which gives a peak in the *foreward* recoil deuteron spectrum (and hence in the scintillator) at nearly the full neutron energy. This feature has been exploited in a number of nuclear reaction studies[13–16] requiring measurements of neutron-energy spectra where TOF is not available or where it is desired to tag a recoiling deuteron from $n+d$ scattering.

The response function of NE230 to a neutron of energy E_n, i.e., light output due to the recoil deuteron from the $n+d$ reaction involves convoluting the appropriate $n+d$ differential cross section with the $n+d$ kinematics, the known response of the liquid scintillator to deuterons, and the detector geometry. Thus, Lister[16] has used NE230 to measure neutron-energy spectra from (d,n) reactions ($E_n \cong 5$–14 MeV) induced by 12.3 MeV deuterons incident on a variety targets. Unfortunately, our low-energy neutron calibration sources are not monoenergetic and hence produce, as calculated, only broad spectra. Fortunately, Chatelain *et al.* have measured the response of a scintillator equivalent to ours (called D-NE213 in their paper) for monoenergetic neutrons $E_n = 2.48$–3.28 MeV and observed, as expected from the asymmetric nature[13] of the $n+d$ cross sections, a distinctive peak in the scintillator output (see Figs. 2 and 3 of Ref. 13). Over a reasonably broad E_n range the exact deconvolution of the light-output spectrum is not critical on setting an upper limit on the number of neutrons detected in that energy range.

Two detectors were used. Detector "*A*" was 3.8 cm diam × 3.8 cm long, attached to an RCA 8575 photomultiplier tube (PMT) and an ORTEC 265 PMT fast-timing base running negative HV (ca. −1900 V). A second detector ("*B*") was 3.8 cm diam × 3.8 cm long, mounted on an RCA 880 PMT with a homemade base and ran positive bias (ca. + 1900 V). N_2 gas cooling of the PMT was used for several runs to minimize temperature drifts. The estimated overall efficiency for detecting neutrons $E_n = 2$–3 MeV is 1% which includes the cell geometry, the detector efficiency, and the convolution of the known $n+d$ cross sections[13] at $E_n = 2.5$ MeV. The detector efficiency and response was calibrated using AmBe and ^{252}Cf low-energy neutron sources of known strengths.

Although our detectors were designed to minimize their cross section to cosmic-ray muons, large (ca. 10×60×1.2 cm^3 thick) plastic scintillators were employed as cosmic-ray veto (or coincidence) shields for selected runs. These could also be used as a veto for muon-catalyzed fusion events although in D_2O these should be a minimum, since the muon will capture on the oxygen rather than the deuteron. Likewise, muon-catalyzed $d+d$ fusion in the deuterated scintillator is not expected to be large, since the scintillator does not have free D_2 in it and, in any case, the rate of stopping muons is expected to be about 0.4/h in our apparatus. Other groups[17] have specifically looked for stopped-muon catalyzed fusion and have found it to be negligible in cells similar to those used here, in particular D_2O loaded cells.

The electronics setup consisted of stabilized HV PMT supplies isolated via transformers, and standard NIM-type fast-timing-filter amplifiers, NIM constant-fraction discriminators and logic modules. Pulse-shape discrimination (PSD) for n/γ was done using an ORTEC 552 module. Scintillator light output ($L \equiv$ energy signal) and PSD signals were recorded via appropriate CAMAC modules using an IBM-PC/AT computer.

The PSD circuitry was adjusted and calibrated for n/γ discrimination using neutrons and γ rays from ^{252}Cf ($\bar{E}_n \approx 2$ MeV) and AmBe ($\bar{E}_n \approx 4$ MeV) neutron sources. The neutron-energy scale (± 0.2 MeV) was determined from the latter two sources together with γ-ray sources and the known ratio of γ ray (recoil electron) to neutron (recoil deuteron) scintillator light output.[18,19] A typical ^{252}Cf neutron PSD spectra is shown in Fig. 2. PSD spectra for low E_n often exhibit the slight distortion exhibited in Fig. 2. Therefore, event-mode recording was used to permit direct two-dimensional (2D) projections from the light output (L) versus PSD spectrum to minimize γ-ray contamination of the neutron spectra.

Runs typically were 10–20 h with the cell power on and then a background run of equal length with the cell turned off or in some cases removed completely. Due to spurious

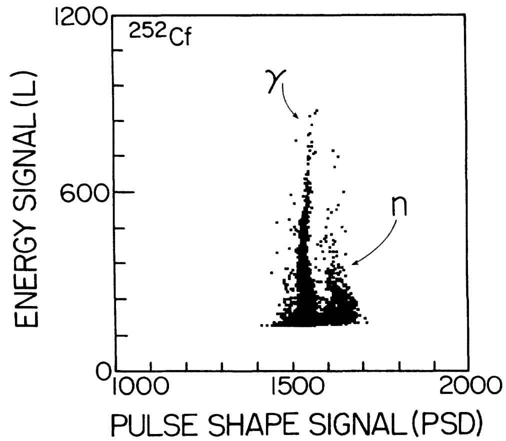

FIG. 2. Pulse shape vs energy signal (L, light output) observed in detector A using an ^{252}Cf n/γ source.

high-frequency electromagnetic interference (EMI) from cellular telephones, pagers, and other microwave transmitters, clean spectra could only be obtained during evenings and weekends. A high-writing speed oscilloscope (Tektronix 2467B) was used to monitor the PMT output signals and identify EMI pickup. EMI shielding was utilized for several runs but did not eliminate all of the pickup. Likewise, occasional pulses due to microphonic pickup were observed. In both cases pulse-shape discrimination eliminated most spurious events from being detected as neutrons. Lacking PSD, it would be difficult to distinguish true neutron events from these known, spurious sources of PMT signals, at least at low count rates. This also applies to large proportional counters such as those used for thermal neutron detection which are particularly susceptible to these effects. Many of the "neutron" events reported[20] using these types of detectors may be due to such spurious detector pickup[21] since, as is well known, both PMT's and large gas counters can act as sensitive rf and acoustical antennae.

Initial trials were done using Pd wire samples and detector A in a standard well geometry, i.e., the reverse of that shown in Fig. 1 with the detector situated on top. Surprisingly, this sample gave apparent "neutron" signals that were correlated with cell operation, in particular the D_2 loading and unloading phases, with rates that scaled approximately as $1/r^2$ from the cell. However, upon closer inspection, these signals did not appear to have a pulse shape consistent with known neutron signals. Nor were they observed in detector B when it was set below the cell. The piezoelectric sensor used to monitor microphonic pickup indicated that the sample was acoustically active, but no definite correlations with neutron activity were observed. Likewise, the cell was electrically active (arc discharges) but no reproducible correlations with neutron activity were observed. One possibility for these signals was electrical discharges of the PMT or base due to the evolution of gas and material from the cell. It was therefore decided to seal the PMT and base and use the inverted geometry shown in Fig. 1 to avoid such a problem. Measurements using the new setup did not produce any 2.5-MeV neutron signals above normal background levels, which were on the order of ~ 1 n/h at $E_n \approx 2$–3 MeV.

A cast sample was then prepared from 99.9% pure Pd in a phosphate base casting investment. Casting was done under an argon atmosphere. Measurements over several weeks using this sample (Figs. 3 and 4) yielded no net neutrons $E_n = 2$–3 MeV above background.[22]

In the course of the primary neutron measurements, attempts were made to measure other fusion products, specifically γ rays (using large NaI detectors) and slow neutrons (using activation foils). No events above normal background levels were observed. However, these types of detectors are quite sensitive[23] to common background sources including γ rays from U/Th, radon, and other trace elements contained in building construction materials, the cell, glass, and the chemicals used for the electrolyte, and hence would only detect a high flux of neutrons. Cosmic-ray showers are also a known source of slow/fast neutron bursts. Obviously, passive detectors such as activation foils cannot exploit cosmic-ray veto detectors to reduce such background. Hence, among our set of measurements the NE230 neutron-energy spectrometers gave the most accurate limits on the cold-fusion rates.

In order to determine the typical deuterium loading, a control sample of Pd was loaded in 0.1M LiOD in en-

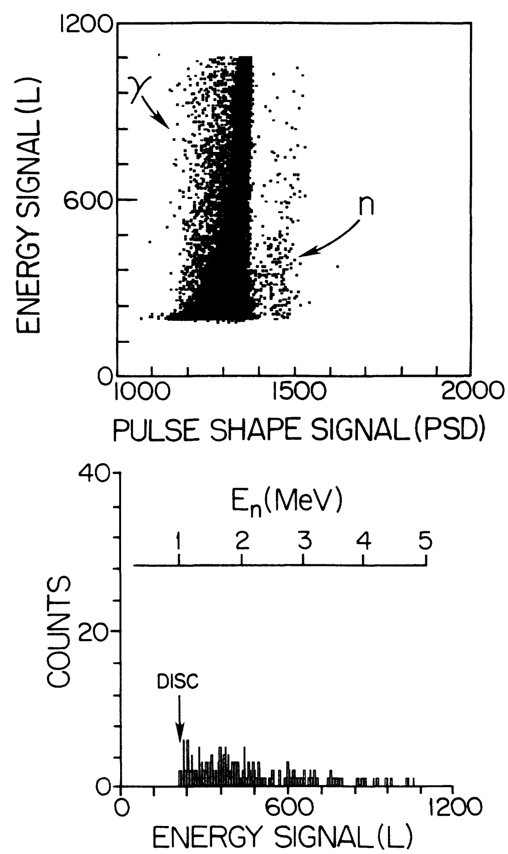

FIG. 3. Top: Typical data of PSD vs energy signal (L, light output) using inverted-well geometry (detector B and cast sample). Bottom: Projected neutron spectrum with cell on 200 h. DISC indicates the lower-level energy-signal discriminator setting.

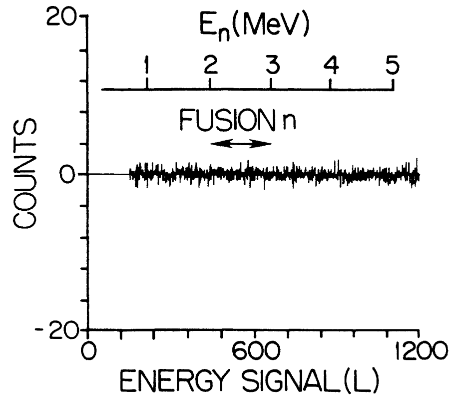

FIG. 4. Net neutron events (cell on − cell off) for a period of 200 h using inverted geometry and cast sample (see Fig. 3).

riched D_2O at 100 mA/cm^2 for a period of 48 h. The sample was then outgassed in a chamber equipped with an absolute-pressure capacitance manometer. The samples were found to be past the deuterium loading of the β-Pd hydride phase with the excess deuterium in solution in the Pd. We deduced a loading of at least 0.84 d/Pd.

Using this information, the observed neutron spectra together with the estimated neutron detector cell efficiency leads to limits of $\leq 10^3$ fusion (neutrons/s)/g Pd or more specifically $< 7\times10^{-24}$ fusion (neutrons/s)/d-d pair. These rates are averaged over 200 h of cell-on and 200 h of cell-off measurements using both extruded samples and the cast sample. Our observed limit rates are *far* below the cold-fusion rates reported by Pons and Fleischmann,[4] and are just below the results reported[3] by Jones *et al.* for Ti. Other groups[9–12,17] using more conventional detectors have reported similar low limits on electrolytic cold fusion. However, our detectors have the advantage of yielding neutron-energy spectra and hence direct limits for the fast 2.5-MeV neutrons produced from $d+d \rightarrow {}^3\mathrm{He}+n$. Should a reproducible method of generating such "cold-fusion" neutrons become publicly available, the techniques described here should prove advantageous.

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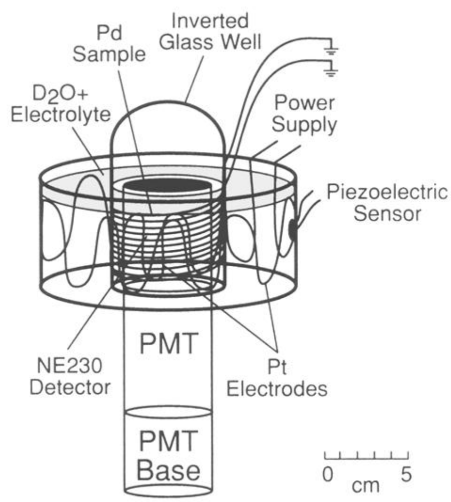

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