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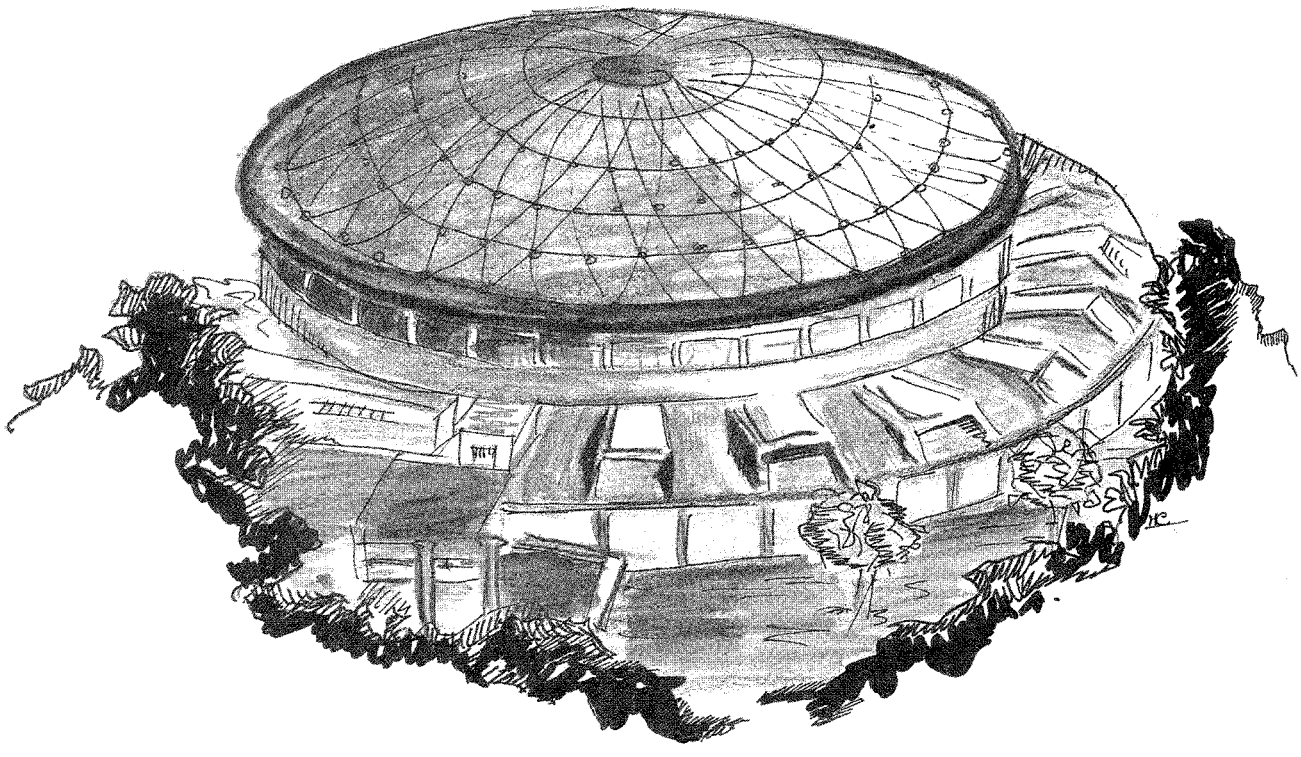

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PRELIMINARY MEASUREMENTS ON ELECTROLYTIC COLD FUSION AT UNDERGROUND GRAN SASSO LABORATORY

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ABSTRACT

Since April 15th '89, we have performed several measurements at the underground Gran Sasso Laboratory of nuclear radiation emission from an electrolytic cell of D_2O and LiOH using Pd or Ti(Al) as cathode electrodes, in the framework of the so called Nuclear Cold Fusion. The almost unique characteristics of such underground laboratory, located under about 3500 meter of water equivalent, is an extremely low value of thermal and fast neutron background. In a bunker built with low-activity lead bricks, to further reduce the gamma ray flux coming from the concrete and spurious materials of the experimental set-up, we allocated a set of different nuclear detectors in order to put in evidence gamma rays, neutrons and charged particles arising from the electrolytic cell. We report two emission events: a) both a "shot" signal and some bursts of signals, simultaneously in

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almost all of the detectors (may 2, at $\sim$ 23:30), from a just-prepared Pd electrode; b) burst signals from an over-stressed Ti(Al) electrode (may 1, at $\sim$ 13:30). The time evolution of these bursts signals is quite similar. A rough evaluation of the fusion rate, supposing that these emissions are due to cold fusions, is of the order of $10^{-19} \div 10^{-20}$ fusions/deuteron pair/second in the case of Pd electrode and still larger for the Ti(Al) one.

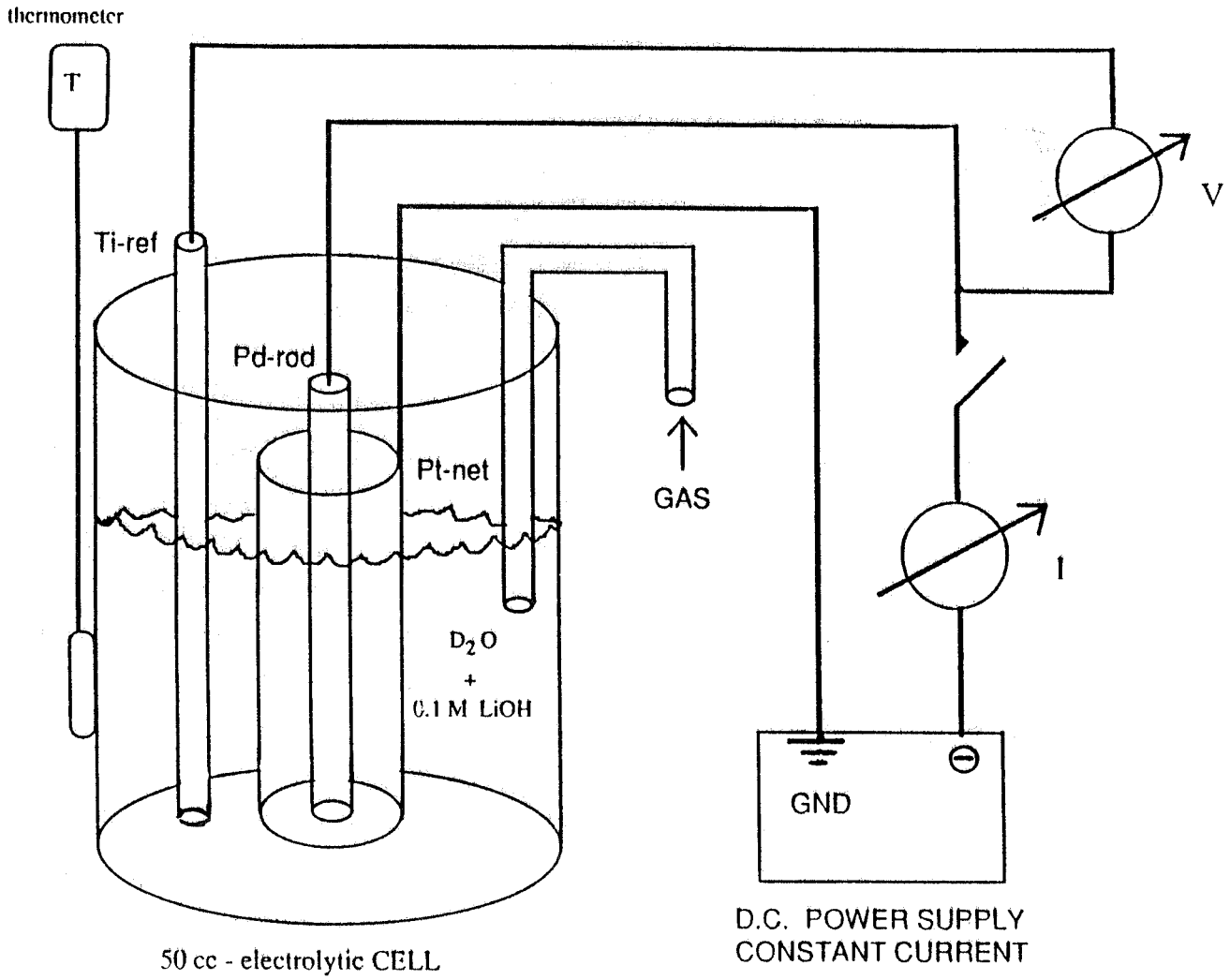
INTRODUCTION

It is still an open question whether the kind and the intensity of signals arising from reactions in experiments of so called "cold nuclear fusion" [ref.1,2], during electrolytic infusion of Pd or Ti in D_2O, are really related to any known nuclear phenomena. A similar experiment, using thermal stressed titanium in contact with pressurized deuterium gas, sometimes produced a neutron flow [ref.3]. After some preliminary experiments, performed in the Frascati National Laboratory of the Istituto Nazionale di Fisica Nucleare, with an electrolytic cell [ref.4], we realized that low level and unpredictable signals coming from such kind of cell needed a very quiet and "clean" laboratory to be detected clearly and undoubtedly. Taking into account the special requirements and cares to detect neutrons, we decided to move our experimental apparatus inside the underground Gran Sasso Laboratory because in this "special" laboratory the neutron flux is extremely low. The flux, for both fast and thermal neutrons, is of the order of $10^{-6}\ n \cdot cm^{-2} \cdot s^{-1}$, i.e. about 1000 times lower than normal environments [ref.5]. In this kind of experiment it is important to detect gamma rays too, because they give useful indications on the kind of reaction, or sequence of reactions involved.The total background flux of gamma rays measured with our experimental set-up is only 10 times lower than a "normal" laboratory, so that a further reduction was performed.

THE ELECTROLYTIC CELL

All our experiments were performed on electrolytic infusion of D_2O + 0.1 M LiOH with a Pd-Pt or Ti-Pt system and a small regular addition of a proper gas. The geometry of electrolytic cell is coaxial with the Pt (the nominal composition is 95 % Pt and 5 % Ir) used as cylindrical external net (diameter = 15 mm, l = 50 mm) and connected to the electrical ground in all the measurements. The Pd or Ti working electrode has been placed at the center of the Pt net electrode axis by proper teflon disks (fig. 1).

The jar used is conic, Erlenmeyer type of 50 ml, a borosilicate composition glass from Scott-Duran (West Germany). The Pd electrodes (99.98 % purity,Engelhard) used were cylindrical rods with a diameter ranging from 0.2 up to 8 mm and with a typical length of about 6 cm. These Pd electrodes were carefully vacuum degassed at 650 Celsius for 2



[Fig.1] Experimental set-up, not in scale, of electrolytic cell and power supply section. The DMM are FLUKE mod.77, the power supply is LARCET mod.3/60, the thermometer is J-type thermocouple from Keithley. All the connections between electrolytic cell and power supply or DMM are made by coaxial cable to reduce the possibility of noise pick-up. During the "nuclear" measuraments all the DMMs and the thermometer are switched-off to avoid spurious signals due to the sampling.

hours and slowly cooled down to room temperature under vacuum condition ($1.0 \cdot 10^{-3}$ mm Hg) just before being used in the experiments. The Ti electrode is a bar with dimensions of $\sim 5 \times 5 \times 50$mm (95.5 % Ti, 4.5 % Al nominal composition, ILVA Terni, Italy). The Ti electrode underwent an heavy treatment at a degassing temperature of 1050 Celsius in a purified Ar flux with Oxygen content < 1ppM for 2 hours and slow cooled to room temperature in Ar flux. Both the electrodes were kept in a dry box until their use. The bath volume of D_2O (99.8 % purity, Tritium < 74Bq/ml, Carlo Erba) was about 40 cc. The value of T < 74Bq/ml is just a legal limit according to radioprotection regulations on Tritium concentration in liquids.

We added a third reference electrode, a simple hyperpure Ti, to monitor the so called spontaneous voltage of working electrode. In the following the "voltage under charge" means the voltage between the reference electrode and the working electrode (Pd or Ti(Al)) when the power is supplied by the constant-current power supply. Similarly, "spontaneous voltage" is the same voltage when the power supply is in the open circuit state. The Ti used as reference electrode was 99.99 % purity, in the alfa phase, and was carefully cleaned, before being used, by the subsequent chemical treatments: HF 30 % concentration, 150 by volume H_2O_2, deionized H_2O, vacuum dried.

The cell was kept in the dark and the current density was about $60\text{mA} \cdot \text{cm}^{-2}$ related to the working electrode area as suggested by both Fleischmann and Jones [ref.1,2].

We added regularly some gaseous CO_2 to the electrolytic infusion in order to increase the deuterium intradiffusion in the Pd electrode which allows a faster diffusion of atomic deuterium into Pd [ref.6].

THE DETECTORS AND THE EXPERIMENTAL PROCEDURE

We further decreased the value of gamma ray background flux in Gran Sasso Laboratory (originally about 10 times lower then normal environments) by a factor of about $4 \div 5$, building a small local bunker made of low-activity lead bricks (from *Bolidn*, Sweden). All the detectors surround the electrolytic cell as close as possible, within a radius of less than 10 cm. The detection system was based on:

a) one cylindrical 5×5 in. NaI(Tl) gamma detector (*Quartz & Silice*, France);

b) two He^3 neutron detectors, cylindrical shape of 100 cm length and 5 cm diameter, 3 atm. of pressure, nominal thermal neutron sensitivity of $433 \cdot \text{cn}^{-1} \cdot \text{cm}^2$ (model 100 He3/228/50G from *Centronic*,England),the first one was bare and the second one was surrounded of 120 cm long, 30 cm diameter paraffin moderator,[ref.5];

c) one fast plastic scintillators optimized to detect charged particles, $1 \times 10 \times 10$cm.

We used a standard shaping time for NaI(Tl) (0.5μs) and He^3 (2μs) detectors (shaping amplifiers mod 7611L from *Silena*, Italy). The output (rise time $<$ 10ns) from the fast scintillator was sent directly to a level discriminator regulated at 100 ns pulse width.

We collect the energy spectrum from the NaI(Tl) detector by a multichannel analyzer, (fig. 2).

The threshold for neutron counters is set well below the capture peak. Two different ("low" and "high") thresholds are used for the NaI(Tl) detector, respectively corresponding to about 0.8 MeV and 2 MeV. The threshold for the plastic scintillator counter is set in order to reject low-energy gammas, while it is fully efficient for " minimum ionizing particles".

The data acquisition system is based on a standard *CAMAC* crate, which houses the modules required for acquisition from digital counters. It is controlled by a *CAVIAR* microcomputer [ref.7]. We acquired counts coming from all detectors, read by the afore mentioned devices.

At the beginning we measured the radiation background, for some hours, and checked it before and after any changing of electrode or apparatus environment. We often repeated this background measurements by putting the cell into the apparatus detection system and very far from it. We also have performed a series of tests on the apparatus to avoid any kind of false "nuclear" signals coming from natural radioactivity of each component of our measurement area or any kind of electrical (dc and ac) thermal or mechanical realistic perturbations. In details, with related results, the tests we performed are the following:

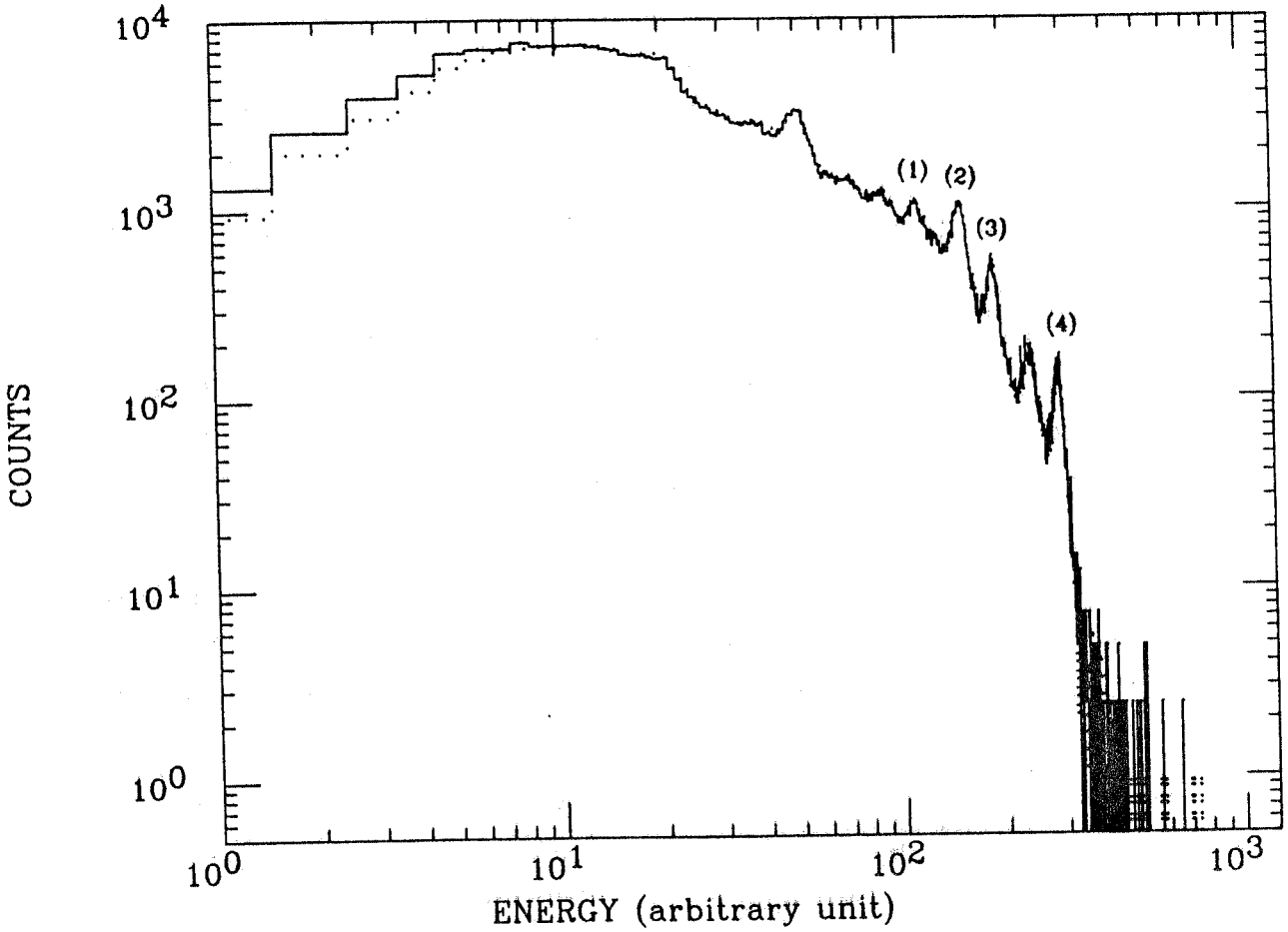
a) we checked each component of electrolytic cell one by one with null results;

b) we sometimes noted a very slight increase of counts when we assembled all the component of the electrolytic cell without external current flow;

c) the effect reported in b) disappeared when we replaced the Pd electrode with a 5 mm diameter 6 cm long iron nail;

d) we supplied the usual dc current of $60mA/cm^2$ to the electrolytic cell using the iron nail electrode with null results. This test has been extended up to 3A of total current and 3 hours of running time;

e) we noted a slight increase of counts when the cell is powered at the usual $60mA/cm^2$ current density;



[Fig.2] Log-Log energy spectra of signals coming from NaI(Tl) detector. The background is dotted line, the data from Pd electrode are in solid line. The data have been normalized to the background acquisition time (12268 s).
The numbers (1),(2),(3),(4) correspond to the most intense natural radioactive lines, respectively at (1120,1460, 1764,2615) KeV.

f) we ran, in standard "conditions", for more than 4 hours another complete electrolyte cell (with obviously a new Pd electrode, 4mm thick, as prepared) using deionized "light" water obtaining null results.

Single counts are acquired from all detectors. Because of the limited mass storage capability of our system, we use an on-line reduction of data. Single counts integrated over 0.6 s are acquired from all counters. If any of them exceeds a software threshold, the entire set of frequencies is written on the disk, otherwise it is ignored. Moreover, counts from all detectors integrated over 1 min. are always recorded.

Typical frequencies of count for the detectors (putting the working cell far away from the detection apparatus), and their respective software threshold for data reduction are:

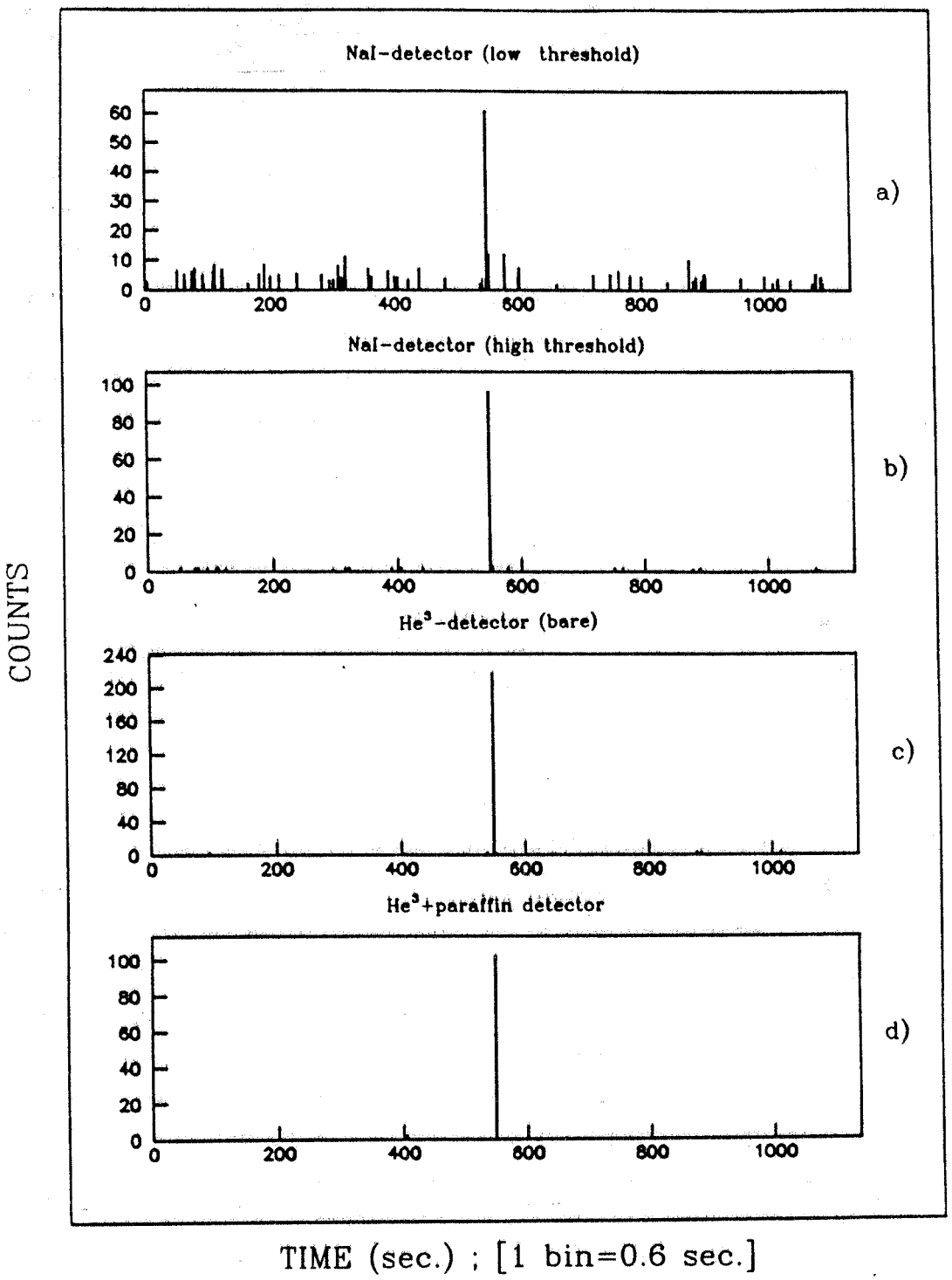
NaI(Tl) low-threshold	= 6 Hz		(12 counts/0.6 s)
NaI(Tl) high-threshold	= 0.3 Hz		(2 counts/0.6 s)
He^3 bare	= 0.3 Hz		(2 counts/0.6 s)
He^3 with paraffin	= 0.1 Hz		(2 counts/0.6 s)
Plastic scintillator	= 25 Hz		(25 counts/0.6 s)

RESULTS

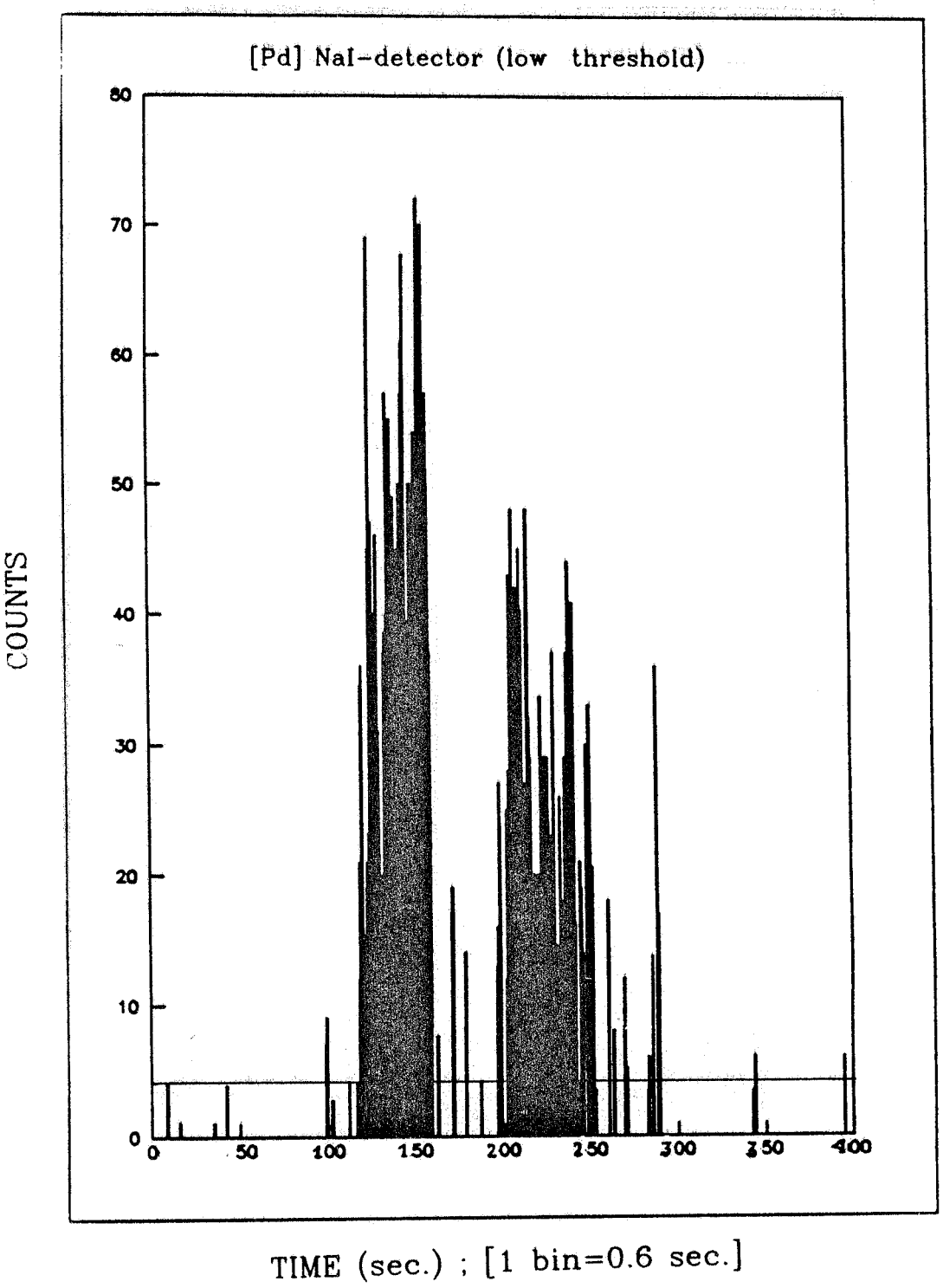
From the beginning of the first charging-up of the 8 mm diameter Pd electrode, we collect, in the multichannel analyzer (MCA), the signals coming from the NaI(Tl) detector at an energy threshold (about 200 KeV) as low as possible compatibly with the dead-time limit of MCA. Due to the long acquisition time adopted (5116 s) for the MCA, the energy spectra collected during the short emission time of the Pd electrode,i.e. shot and burst events,showed signals with low statistical significance. Anyway, in fig. 2, we report in log-log scales both the background (taken just about 3 hours before the beginning of the experiment), in dotted lines, and the data during the charging-up state, in solid lines. The data are normalized to the acquisition time of background (12268 s) and for a better visual readability we don't report statistical errors. Furthermore, we mark with numbers four of the most intense natural radioactive lines present in our experimental set-up (Bi^{214}, 1120KeV; K^{40}, 1460KeV; Bi^{214}, 1764KeV; Tl^{208}, 2615KeV).

In this paper we report two kinds of observed phenomena. The first one is a so called "shot" event (time duration $\leq$ 0.6s) (fig. 3), followed about 40 minutes later from several bursts (total time $\approx$ 4minutes) on a 8 mm diameter Pd electrode, (fig. 4). The second phenomena are several bursts (total time $\approx$ 6minutes) coming from an over-stressed Ti(Al) electrode, (fig 5).

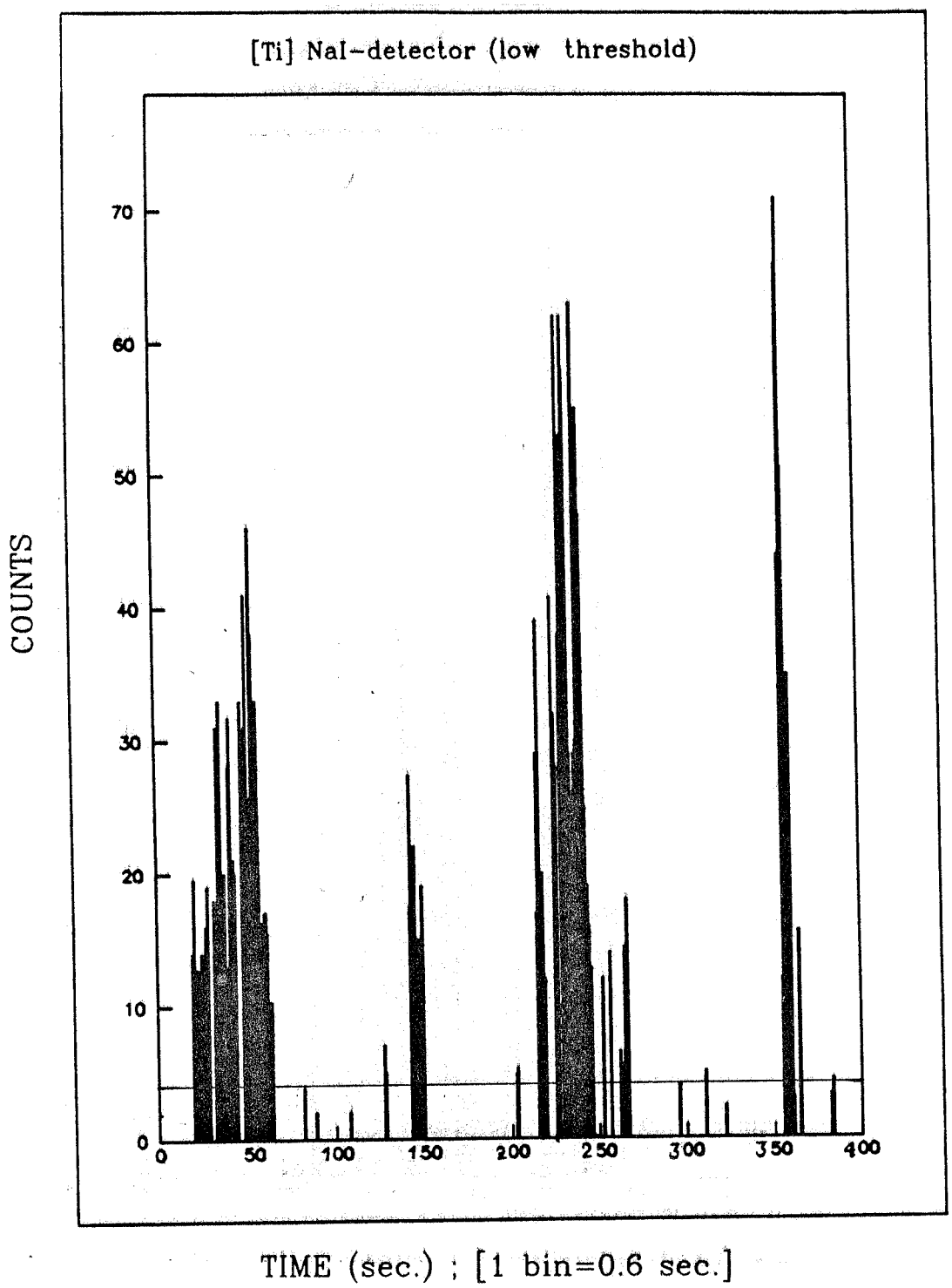
The shot event on Pd occurred about 30 minutes after the *FIRST* charging-up, at a constant current density of 66 mA/cm^2 (maximum allowed voltage $Pd - Pt < -3.1$volts).



[Fig.3] Pd electrode, SHOT event. Time evolution of the signal on NaI(Tl) gamma detector at a) low threshold and b) high threshold. In fig. c) it is shown the result with He^3 bare detector and in fig. d) it is shown the result with He^3 detector surrounded by 12 cm thick paraffin moderator. The bin of the signal is $\simeq$ 0.6 s . For comparison, it is shown the time evolution of the signal at about 550 s before and after the bang event.



[Fig.4] Pd electrode, BURST event. For comparison we show the intensity of the signal about 120 s before and after the burst. The total time of the event is about 4 min. The background is shown by an horizontal straight line.



[Fig.5] Ti electrode, BURST event. For comparison we show the intensity of the signal about 30 s before and after the burst. The total time of the event is about 6 min. The background is shown by an horizontal straight line.

The intensity of the shot was really impressive. We got, in an acquisition time of about 0.6 s, the following counts:

NaI(Tl) low thres.	=	61	counts	(mean value in 0.6 s $\leq$ 4)
NaI(Tl) high thres.	=	96	counts	(" " " " $\leq$ 1)
He^3bare	=	217	counts	(" " " " $\leq$ 1)
He^3moderated	=	102	counts	(" " " " $\leq$ 1)

The apparent incongruence on counts between low and high threshold of NaI(Tl) can be understood if we assume that the signal have a very intense burst behaviour:the low threshold retriggerable discriminator-shaper circuitry was "locked" because most of the following pulses arrived between a time shorter than $1\mu s$ (the time we chosed for the digital shaper of gamma signals). About neutron detectors, if we take into account the detector efficiency and unfavourable geometrical factors, we can roughly claim to have produced a number of neutron of the order of 10^4. This large value can be still higher if we take into account the intrinsic dead-time of the conventional shaping amplifier adopted ($\simeq 10\mu s$) in respect to the probable burst structure of the signal.We measured both the voltage-under-charge and the spontaneous voltage just few minutes before and after emission because it was our standard experimental routine during the first charging-up of the electrode: *we noted that both voltages decreased after the emission* (of the order of 1 %).

The bursts occurred after 73 minutes from the beginning of the first charging-up, about 1 minute after measuring the spontaneous voltage of the electrolytic cell. Such type of measurement, as pointed before, involves a little "stress" to the system because it is necessary to disconnect the power supply (floating condition) to measure the voltage between the Pd electrode and the pure Ti used as reference one.

As previously described, our "trigger" is any detector (OR condition) that overcomes the stated counts. The "typical" out-of-burst condition is shown with a continuous line in figs. 4 and 5, the amplitude is in counts/bin and one bin correspond to about 0.6 s, i.e. the minimum collecting time allowed from our acquisition system. We show our bursts together with the situation before and after the signal bursts. Fig. 4 shows two main bursts ($\simeq$ 50 s each) with some dead-time in between ($\simeq$ 40 s) and some after-main-burst pulses so that the total duration time of the event is of the order of four minutes.

In this particular experiment the Pd electrode was very close to the NaI(Tl) detector ($\simeq$ 2.5 cm) so that we can estimate the overall detection efficiency of the order of some percent. We like to note that after the end of the burst ($\simeq$ 5 minutes) we measured again the spontaneous voltage and we found it slightly lower as before stated. According to us, this fact is related to the coming-out of deuterium from Pd electrode. We didn't note any temperature change (stable at 17 ± 1 Celsius) before and after the event. The thermometer, a J type thermocouple from Keithley, was positioned *external* to the electrolytic cell

in open air, well bound to the jar.

The bursts coming from Ti (fig. 5) electrode arose after a long restoring cycle of this electrode subsequent to several severe stresses: -196 ÷ +200 Celsius thermal cycling, cathodic and anodic current up to 3A, mechanical stress. We get the signal after 3 days of low current infusion at 50mA/cm^2 and we can't say anything about spontaneous voltage or temperature conditions because we were absent during the emission and we hadn't automatic record of them. The distance between electrode and NaI(Tl) was about 10 cm, so that the overall efficiency for gamma signal is less than 1 % in the energy range recorded.

The time evolution of the signal is similar to that appeared for Pd electrode and, in this case, the total time of the event is about 6 *minutes.*

We never got statistic evidence of excess counts from the charged-particle detectors, i.e. the plastic scintillator, using both Pd or Ti(Al) electrodes.

CONCLUSION AND DISCUSSION

We observed gamma and neutron emissions from electrodes made of Pd and Ti(Al) during non-equilibrium conditions due o electrolisys of D_2O and LiOH: all the emissions detected had a bunched structure.

About the calculation of "fusion rate" λ_f, if we use the *emission data of Pd during the burst* (*fig.*4) *and we assume that all the excess gamma counts arise from any kind of fusion*, adopting a mass of Pd 30 g, excess gamma counts 4100 in 205 s, gamma detection efficiency 1 %, D/Pd concentration in beta phase about 0.6, we should obtain

$$\lambda_f = 4.3 \cdot 10^{-20} \text{ fusion/deuteron pair/s}$$

This value is more than 1 order of magnitude larger of results of P.Perfetti et al. [ref.8] and about 3 order of magnitude lower than the results of M.Fleischmann et al. [ref.1]. We remark that it is not clear, at this time of the research, if our assumption of excess gamma counts is equivalent, from the point of view of λ_f calculation, to the excess neutron counts reported from P.Perfetti or M.Fleischmann.

We note that the results by M.Fleischmann arise from *continuous* emission, not *burst* as in our or P.Perfetti calculation.

Moreover, if we consider the evaluation of λ_f during the shot, using the gamma count value at low threshold (still neglecting the saturation effect), we reach a fusion rate even larger then the value reported by M.Fleischmann [ref.1]. Furthermore, the neutron counts during the shot, corrected by the proper detection efficiency, is roughly the same value reported by M.Fleischmann. As before stated, the count of excess neutrons during the burst has not sufficient statistical significance because of our low sensitivity and non optimised energy threshold.

The calculation of λ_f on Ti(Al) electrode is quite complicated when compared with Pd one, even ignoring completely the effects due the heavy Al doping (4.5 %) and is less accurate because we adopted for the deuterium the hydrogen diffusion coefficient. At first, we remind that the diffusion velocity of the deuterium in Ti is very low, so in our experimental conditions, supposing a charging time as long as 48 hours, the penetration depth of deuterium in Ti is only 0.32 mm [ref.9]. The maximum measured concentration of deuterium in Ti at 23 Celsius is 1.73, determining an oversimplified mean value of 0.85 in our calculation (this condition is called the $\alpha + \gamma$ phase) [ref.9]. As a consequence, the effective active volume of the electrode is only 5 mm^3; taking the electrode density about 4.5 g/cm^3, the atomic weight 48, gamma detection efficiency about 0.5 % , excess count of 2800 counts in 360 s, we should obtain:

$$\lambda_f = 1.3 \cdot 10^{-17} \text{ fusion/deuteron pair/s}$$

This value is about three order of magnitude larger than the largest value estimated by S.Jones [ref.2] taking into account burst effect, but, again, we remark that, in this calculation, we consider a system composed by hydrogen and pure Ti instead of deuterium and Ti(Al).

The nuclear origin of the reported signals is however still an open question, since the gamma spectra collected by the MCA do not show peaks at the foreseen energies. Pile-up of signals and dead-time of the MCA during the very intense emissions could be an argument.

During the experiment we had some others, less intense, events with a behaviour very similar to these quoted.

In any case, we are strongly certain that almost all the possibilities of fake signals originated by electrical or mechanical perturbation and instability of the detectors and associated electronics, as well as any abrupt changing of environmental nuclear activity (thanks to the underground laboratory) were deeply investigated and eliminated if any. Obviously, there is a remote possibility that some source in the electrodes (ultrasounds, fractures, phonons, etc.) could originate some kinds of the signals reported.

We like to note about the hypothesis of muon-induced fusions that , due to the extremely low value of muon flux inside the Gran Sasso Laboratory ($\simeq 1.2 \cdot 10^{-8} \mu \cdot cm^{-2} \cdot s^{-1}$) [ref.10], i.e. an intensity about 1 million times lower than at sea level, it is very difficult to think it was the main cause of signals in our experiment. Obviously, further and more accurate measurements are in progress in the clean, from the point of view of nuclear detection, Gran Sasso Laboratory. Moreover, we plain to set-up a completely new experimental apparatus to detect gamma ray energy spectra with a short acquisition

time, of the order of 1 second, because of our experienced burst nature of the signals.

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