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MEASUREMENTS IN THE GRAN SASSO LABORATORY: EVIDENCE FOR NUCLEAR EFFECTS IN ELECTROLYSIS WITH Pd/Ti AND IN DIFFERENT TESTS WITH DEUTERATED HIGH TEMPERATURE SUPERCONDUCTORS

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ABSTRACT

Several experiments were performed at the INFN Underground Gran Sasso Laboratory (Italy) in order to detect nuclear signals, if any, coming from deuterated materials in the framework of so called "Cold Fusion". We followed three, quite different, lines of experimental researches.

In the first set of experiments we used electrolytic solutions of heavy water and 0.1 M LiOH with some addition of gasseous CO_2, using as cathode a rod of pure Pd or Ti alloys and as anode a Pt net. We detected several intense bursts of gamma rays, lasting up to 15 minutes of duration time, with energy less than 300 KeV.

In the second set of experiments we used pellets of sintered High Temperature Superconductors (HTSC) that underwent deuteration by high pressure (~30 Bar) and temperature (~370 K) gas loading procedure. The deuterated samples were several times cycled down to liquid nitrogen temperature and we detected by two indipendent ^3He moderated neutron detectors (1.2 % total efficiency), sporadic emission of signals mainly during these thermal cycles.

Later on similar tests were performed independently by S.E.Jones and collaborators (Provo Univ., Utah) in a deep underground mine and a very intense and short neutron burst was detected by 16 independent ^3He moderated neutron detectors (32 % efficiency).

In the third set of experiments some additional tests were performed using a low activity Am–Be neutron source, in order to study possible phenomena of enhanced neutron emission by radiation stimulation in these HTSC deuterated compounds. We detected excess counts of 2 statistic standard deviations, pointing to increased D–D cross section due to the effect of ceramic lattice.

INTRODUCTION

It is still an open and controversial question whether the kind and the intensity of signals arising from reactions in experiments of so called "cold nuclear fusion"[1,2], during electrolytic infusion of Pd or Ti in D_2O, are really related to any known nuclear phenomena.

Some experiments obtained results which can be explained with a sort of nuclear reactions [3,4] some other give null results[5,6] and BARC Laboratory published the results of more than hundred experiments, both positive and null[7].

A similar experiment, using thermal stressed titanium in contact with pressurized deuterium gas, sometimes produced a neutron flow[8].

After some preliminary experiments, performed in the Frascati National Laboratory (LNF) of the Istituto Nazionale di Fisica Nucleare (INFN), with an electrolytic cell[9] , 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 doubtless. 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 cm^{-2} s^{-1} , i.e. about 1000 times lower than in normal environments[10,11] .

ELECTROLYTIC TESTS

Since April 15th till the end of June '89, we have performed at the underground Gran Sasso Laboratory several measurements of nuclear radiation emission from an electrolytic cell of D_2O and LiOH, with addition of gaseous CO_2, and using Pd or Ti(Al) as cathode electrodes[12,13] .

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, without lead screening, is roughly 5 – 10 times lower than the LNF laboratory[14,15,16].

In a bunker built with low–activity lead bricks, to further reduce the previous gamma ray flux coming from the concrete and the 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 coming from the electrolytic cell.

We used in this test as cathode electrodes a Ti(Al) bar and later a Pd rod.

- The Ti electrode produced, after 3 days of charging–up, a intense burst–shaped gamma emission. No other emissions were detected up to 1 day later.
- The Pd electrode produced 7 different burst emissions along a month of discontinuos charging. Two of these emissions occurred after a few hours of the first charging up. During one of these emissions a gamma rays spectrum was collected using a MCA; the comparison with background spectra excludes that these gamma can have energy greater than 300 KeV. Taking into account the limit of electronics allowable and their set up, we can hypothesize that these signals were of low energy and high intensity origin.

A rough evaluation of the fusion rate, supposing that these emissions are due to cold fusions, gives about $4 \bullet 10^{-20}$ fusions/(deuteron pair)/second in the case of bulk effect in Pd

electrode; for the Ti(Al) electrode we have $5 \bullet 10^{-20}$ fusions/(deuteron pair)/second in the case of bulk effect or, in the case of surface effect, as large a $1.3 \bullet 10^{-17}$ fusions/(deuteron pair)/second.

SUPERCONDUCTIVITY TESTS

From May '90 to October '90, we explored a new way on the research of radiation emission from unconventional deuterated compounds. We used sintered pellets of deuterated High Temperature Superconductors (HTSC).

The loading was performed by putting D_2 gas and these materials, at high pressure and temperature, into a stainless steel vessel. We tried to load D_2 into these pellets varying the temperature (350 – 450 K), the pressure (20 – 50 Bar) and the time (1 – 24 hours). Then we subjected the vessel, kept at high pressure (at least 20 Bar at 300 K), to thermal cycles (300 $\rightarrow$ 77 $\rightarrow$ 300 K) and we looked for neutron emission, if any.

We were aware of the D_2 loading by the variation of the diamagnetic values, at 77 K, and of some increasing of Tc of these superconductor materials. After these tests we found, by accurate diamagnetic measurements, a significative increase in the critical temperature of these particular deuterated samples.

NEUTRON IRRADIATION OF DEUTERATED HTSC

Further tests were performed by putting a 0.7 mCurie (1.5 KBcq over 4 π of neutrons) Am–Be source close to the vessel. The aim of these tests was to explore the possibitity of enhanced stimulated neutron emission under irradiation of neutrons and gamma rays on deuterated superconductors. These tests were performed only at room temperature.

THE ELECTROLYTIC MEASUREMENTS

APPARATUS AND TESTS

The experimental apparatus[12,13], Fig. 1, is based on an electrolytic cell (50 cm^3 of D_2O on 0.1M LiOH and addition of gasseous CO_2) using as cathode a Pd rod or a Ti(Al) alloy bar and as anode a Pt net in a cylindrical coaxial configuration.

The detectors system was based on:

a) 2 ^{3}He neutron detectors;
b) 1 NaI(Tl) (5x5 inches) low–noise gamma detector;
c) 1 plastic scintillator for charged particles.

All the detectors were arranged around 10 cm close to the cell. We surrounded the cell and detectors with bricks of paraffin as inner shielding and lead as outer shielding.

The electronic acquisition system is based on 1 Multichannel Analyzer (MCA; mod. CATO from SILENA, Italy) used to collect signals from the gamma detector, some discriminator–counters (scalers) to count signals coming from neutrons and gamma detectors ($E\gamma > 800$ KeV).

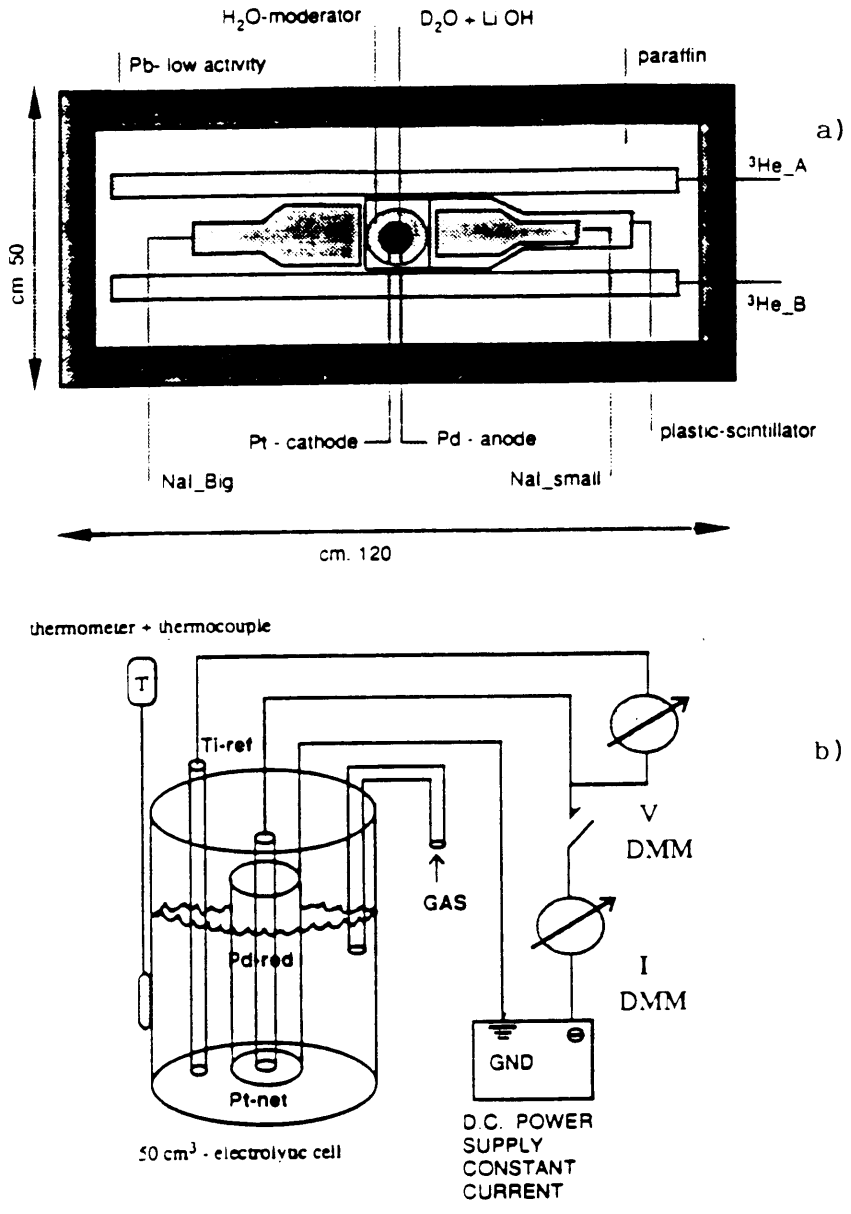

FIG. 1 - a) Electrolytic cell and detection apparatus set–up. The external shielding is made by, at least, 5 cm thick low activity lead bricks that completely surround the detectors. Paraffin bricks are enclosed between the neutron detectors and the external lead wall. The electrolytic cell, bunged by a rubber plug, is in a water bath of about 4 cm of thickness; **b)** A detail of electrolytic cell and power supply section. The Digital Multimeters (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 the electrolytic cell and the power supply or DMM are made by coaxial cables in order to reduce the possibility of noise pick–up.

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[18]. We acquired counts coming from all detectors, read by the afore mentioned devices.

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

The threshold for neutron counters is set over the electronics noise signal and well below the expected signals from the energy peak (764 KeV) due to the thermal–neutron capture by the ^{3}He gas of the detector; over this threshold we expect to detect, at low efficiency, gamma–rays at the left side (low energy) of neutron signal peak spectrum.

Two different ("low" and "high") thresholds are used for the 5x5 inches 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. Single counts are acquired from all detectors.

Because of the limited mass storage capability of our system, we used 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.

We just notify that during the measurements we added an other NaI(Tl) gamma detector (3x3 inches) set at a 1.2 MeV discrimination threshold and improved the overall shielding.

The Pd and Ti(Al) cathode electrodes underwent severe treatments before the use[12].

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 [19] .

Several blank tests were performed at the beginning of the measurements and periodic background runs were made before and after each set of tests.

The cell was kept in the dark and the current density was about 60 mA cm^{-2}, related to the working electrode area as suggested by both Fleischmann and Jones[1,2] ; some tests were performed superimposing a weak a.c. current to the d.c. current at a frequency from 1 Hz up to 10 KHz.

RESULTS AND CONCLUSIONS

We observed gamma emissions from electrodes made of Pd and Ti(Al) during non–equilibrium conditions due to electrolysis of D_2O and LiOH: all the emissions (1 for the Ti and 6 for the Pd) detected had a bunched structure ("bursts").

a) Ti:

The bursts coming from Ti (Fig. 2a) electrode arose after a long restoring cycle of this electrode (from 77 to 473 K thermal cycling, 2 times), subsequent to a few severe stresses : cathodic and anodic current up to 3A, 4 times.

We didn't get any significative result during all these tests.

We get the burst signals after 3 days of low current infusion at 50 mA cm^{-2}. The distance between electrode and NaI(Tl) was about 10 cm, so that the overall efficiency for gamma signals is less than 1 % in the energy range recorded.

We collected on MCA the gamma spectrum for the whole time of measurement (Fig. 3a); because of the long acquisition time and of the few signals counted by the scaler we do not have enough resolution to evidence the energy of these radiation emissions (Table I).

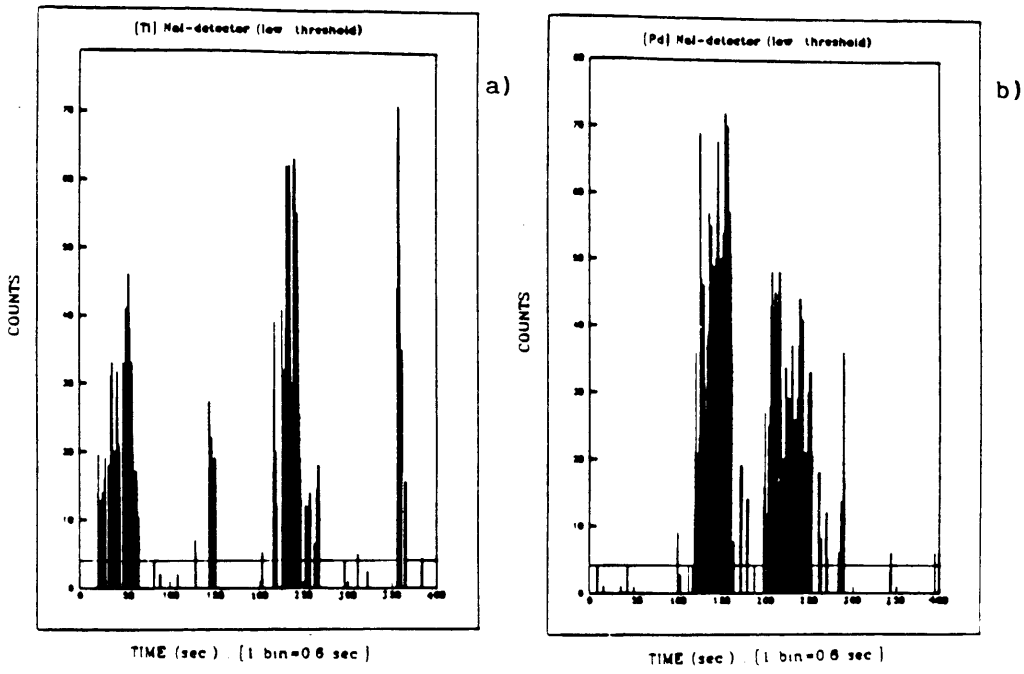

FIG. 2 – Detail of some tipical "burst" events: **a)** Ti electrode, burst event. For comparison the intensity of the signals about 30 s before and after the burst are shown. The total time of the event is about 6 min. The background level is shown by an horizontal straight line; **b)** Pd electrode, burst event. For comparison the intensity of the signals about 120 s before and after the burst are shown. The total time of the event is about 4 min. The background level is shown by an horizontal straight line.

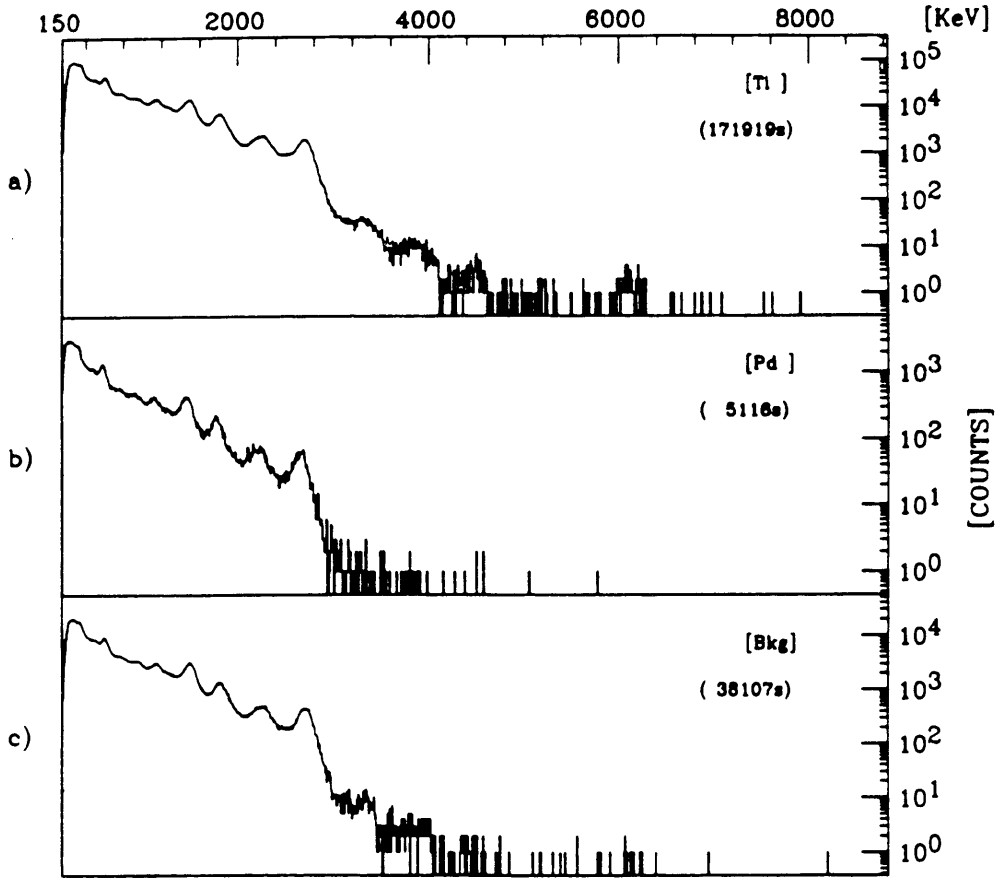

FIG. 3 – Gamma spectra, collected during the bursts, and background, are shown: **a)** Ti electrode, the acquisition time (171919 s) is much longer than the effective emission time (120 s); **b)** Pd electrode, the acquisition time (5116 s) is quite longer than the effective emission time (157 s); **c)** Background spectrum, collected (38107 s) after the Pd electrode emission, is reported as comparison.

TABLE I – Summary of some significative data of Fig. 3: 1) sectors of figure, electrode in use during the emission and date of acquisition; 2) acquisition time, total counts recorded by MCA and calculated rate; 3) counts recorded by MCA in the energy range 300–800 KeV, and calculated rate; 4) counts recorded by MCA in the energy range > 800 KeV, and calculated rate; 5) excess counts recorded by the scaler during the effective burst emission and effective burst emission time; 6) percent ratio calculated between the excess counts recorded by the scaler and the counts recorded by MCA in the energy range 300–800 KeV; 7) percent ratio calculated between the excess counts recorded by the scaler and the counts recorded by MCA in the energy range >800 KeV.

Figure 3 (sector) ====== electrode type ====== [date]	Tot. time (sec) ====== MCA (total counts) ====== rate [Hz]	MCA counts (<800 KeV and >300 KeV) ====== rate [Hz]	MCA counts (>800 KeV) ====== rate [Hz]	SCALER excess count ====== urst time (sec)	(excess count) / counts (<800 KeV and >300 KeV) [%]	(excess count) / counts >800 KeV [%]
a) ====== Ti ====== 1/May/89	171919 ====== 5195984 ====== (30.2)	3095980 ====== (18.0)	1564475 ====== (9.1)	2800 ====== (120)	0.09	0.18
b) ====== Pd ====== 2/May/89	5116 ====== 153883 ====== (30.1)	83745 ====== (16.4)	43712 ====== (8.6)	4092 ====== (157)	4.89	9.36
c) ====== Bckg ====== 3/May/89	38107 ====== 1062428 ====== (27.9)	621191 ====== (16.3)	320458 ====== (8.4)	0 ====== (0)	0	0

The calculation of Λ_f on Ti(Al) electrode is quite complicated when compared with Pd one (see later), even ignoring completely the effects due to 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[20].

The maximum measured concentration of deuterium in Ti at 296 K is 1.73, determining an oversimplified mean value of 0.85 in our calculations (this condition is called the $\alpha + \gamma$ phase)[20].

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:

(Ti–surface hypotesys): $\Lambda_f = 1.3 \bullet 10^{-17}$ fusion/(deuteron pair)/s.

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

When the whole Ti electrode was infused with deuterium, (it may have occurred in some hours, because the electrolysis can increase of a few order of magnitude the diffusion coefficient) we can estimate the fusion rate:

(Ti – volume hypotesys) : $\Lambda_f = 5.2 \bullet 10^{-20}$ fusion/(deuteron pair)/s

This is the same value stimated by S.E.Jones[2]

b) Pd:

The Pd electrode (8 mm diameter) was under test along about 40 days. At the beginning of the first charging up (about 30 min.) a very short and intense emission (less then 0.6 s), figure 2b was detected by all detectors (except the plastic scintillator).

After this event (about 40 min. later) a gamma burst emission occured and it was detected by the (5x5 inches) NaI(Tl) detector. The total duration was about 4 min. while the effective duration was almost 120 s.

We measured both the voltage–under–charge and the spontaneous voltage just few minutes before and after emission and we noted that both voltages decreased after the emission (of the order of 1 %).

We collected, on the 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 (of the order of tenth of microseconds); in Fig 3 b,c we show both the gamma spectrum collected during the Pd burst emission and the event background spectrum for comparison.

By these spectra comparation, as summarized in Table I, no excess signals ($E\gamma > 800$ KeV), out of the system gain stability and statistic significance, are found with strong evidence.

This fact support the hypotesis of a quite intense emission at low energy. Observing the energy spectra we have to argue that most of the signals detected must be provided by gammas of energy less than 300 KeV.

We have to note that in case of bunched signals (peak rate > 1 MHz) small and very close signals can simulate a high energy signal (pile–up effect); that most probably happened in our experiment, although unexpected.

About the calculation of the "fusion rate" Λ_f, if we use the emission data of Pd during the burst (figure 2b) 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 4092 in 205 s, gamma detection efficiency 1 %, D/Pd concentration in β phase about 0.6, we should obtain

(Pd –volume hypotesys) : $\Lambda_f = 4.3\ 10^{-20}$ fusion/(deuteron pair)/s

Other 5 burst events were detected along the following 30 days, in spite of only 50% of operative measurements time. These events, although they look different in shape and intensity, show similar fusion rate values, all in the range from $3.2\ 10^{-20}$ to $9.2\ 10^{-20}$ fusion/(deuteron pair)/s.

Pd/Ti – CONCLUSION AND DISCUSSION

We observed gamma emissions from electrodes made of Pd and Ti(Al) during non–equilibrium conditions due to electrolisys of D_2O and LiOH: all the emissions (one for the Ti and several for the Pd) detected had a bunched structure.

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.

We don't exclude that fracto–emission phenomena [ref. A21] can have occured which could have given a part of the gamma counts detected.

Anyway we can exclude that natural radioactivity at LNGS can generate such processes in such a long time. Moreover we recall that rare "natural" gamma burst emissions have a time duration of the order of few seconds, not several minutes, as detected in our measurements.

We can exclude that an eventual relevant electromagnetic wave emission might be the origin of our signals at the NaI(Tl) because the ^3He detectors, more prone than NaI(Tl) to this effect, never gave signals with such behaviour.

In any case, we are 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.

EXPERIMENTS WITH HIGH TEMPERATURE SUPERCONDUCTORS

MOTIVATION

Following allowable literature [22,23,24] there are over 30 Hydrogen dissolving metals apart some of their binary and/or ternary alloys. Neglecting the well–known Pd, Ti and transition metals like Nb, Ta and V exist two other groups of elements that can absorb, in proper conditions, large amounts of H, D and T. These groups of elements are the rare earths and actinides. About the rare earths (Re) we have to add scandium and yttrium elements, the last one being of particular interest in high temperature superconductivity.

It has been experimentally shown that the addition of some impurities or the formation of alloys[22,23,24] can change, in a large quantity, the behaviour of the hydrogen uptake, i.e. the pressure–temperature–concentration (P–T–C) diagram.

According both to our opinion, mainly based on some similarities between the behaviour of H or D doped Pd and $Re_1Ba_2Cu_3O_7$ in the superconducting state, and to some theoretical suggestions and considerations[25,26], there is some interest in studying compounds of deuterated rare–earths showing high temperature superconductivity in the frame–work of cold fusion; this because they show reduced dimensionality[25,26] that can enhance fusion rate mainly by tunneling.

We examine below these hydrogenated materials from the superconducting point of view.

PALLADIUM AND SUPERCONDUCTIVITY

There are some unexpected peculiarities when Pd is heavily loaded with H, D or T from the point of view of the "classical" superconductivity [27,28]. Some of the pecularities are:

1) Pd is superconductor only when heavily loaded with H, D or T.
2) The minimum loading factor to get superconducting state and then a critical temperature Tc (T > 1.2 K) is a function of the H isotopes:

   H/Pd > 0.81 ; D/Pd > 0.77 ; T/Pd > 0.73 .

3) All the minimal loading factors are larger than the end point of the β phase of Pd (0.6), so these isotopes are in a metastable state inside the Pd lattice.
4) The H or D is moving in the lattice even at 77 K.
5) The critical temperature Tc rises as H, D or T loading increases (up to 8.8 K and 11.05 K at H or D ratio equal to 1, 4.2 K for T loading of only 0.82 , see Fig. 1 in ref. 28).
6) The superconducting compounds show the rare property of inverse isotopic effect.
7) The superconducting compound H/Pd and D/Pd (no data allowable for T/Pd) have the rare property of inverse pressure coefficient.

THE RARE-EARTHS HIGH TEMPERATURE SUPERCONDUCTORS IN RESPECT TO O CONCENTRATION AND H/D ADDITION

There are several similarities between the behaviour of heavily hydrogenated Palladium and heavily oxygenated $Re_1Ba_2Cu_3O_{6.5+x}$ (RBCO) compounds (Re is any trivalent rare–earth except yttrium). Among these we note :

1) The RBCO are superconducting only when O content is larger than 6.5. Tc depends steeply on O content (e.g. $Y_1Ba_2Cu_3O_7$ Tc=92 K, $Y_1Ba_2Cu_3O_{6.5}$ Tc ≅ 65 K).
2) The superconductors easily loose the O in excess of 6.0, i.e. these are intrinsically not stable.
3) It is possible to add H or D (nobody tried T) to superconductors even increasing critical temperature, despite their initial content of O (but at least larger than 6.5)[29] .
4) There is no clear evidence for the usual isotopic effect.
5) Several authors found the rare property of inverse pressure coefficent in $Y_1Ba_2Cu_3O_{7-\delta}$.
6) By NMR measurements [30] it has been found that H in .. $Y_1Ba_2Cu_3O_{7-\delta}H_{0.2}$ occupies sites in the Cu–O planes and that it diffuses or moves dynamically in the crystal above 170K, but that below 150K it is trapped. In other words, after cooling to 77K, during warming up we might have an abrupt moving of trapped H. Similar effects can be expected with D at similar temperatures.
7) There exist a particular value of H doping that increases[29] the critical temperature. This particular value seems to depend on sample preparation and H loading procedure[27,34].

In conclusion, among the many unusual properties of high temperature superconductors there is their ability to absorb a large amount of hydrogen[29,30,31,32,33]. It is experimentally established that the hydrogen loading in solids has noticeable effects on the electronics and vibrational properties[22,24] .

About the experiments of our interest, in the superconducting $Y_1Ba_2Cu_3O_{7-\delta}H_x$ a hydrogen solid solution is obtained for x up to about 0.5, but a hydride phase precipitates at higher hydrogen contents. It is still an open question where exactly the H is located but, according to most of the authors, for low concentrations of H, it is interstitial near Cu sites and, differently from what expected, Cu–H type bonds are created instead of O–H ones (Fig. 4).

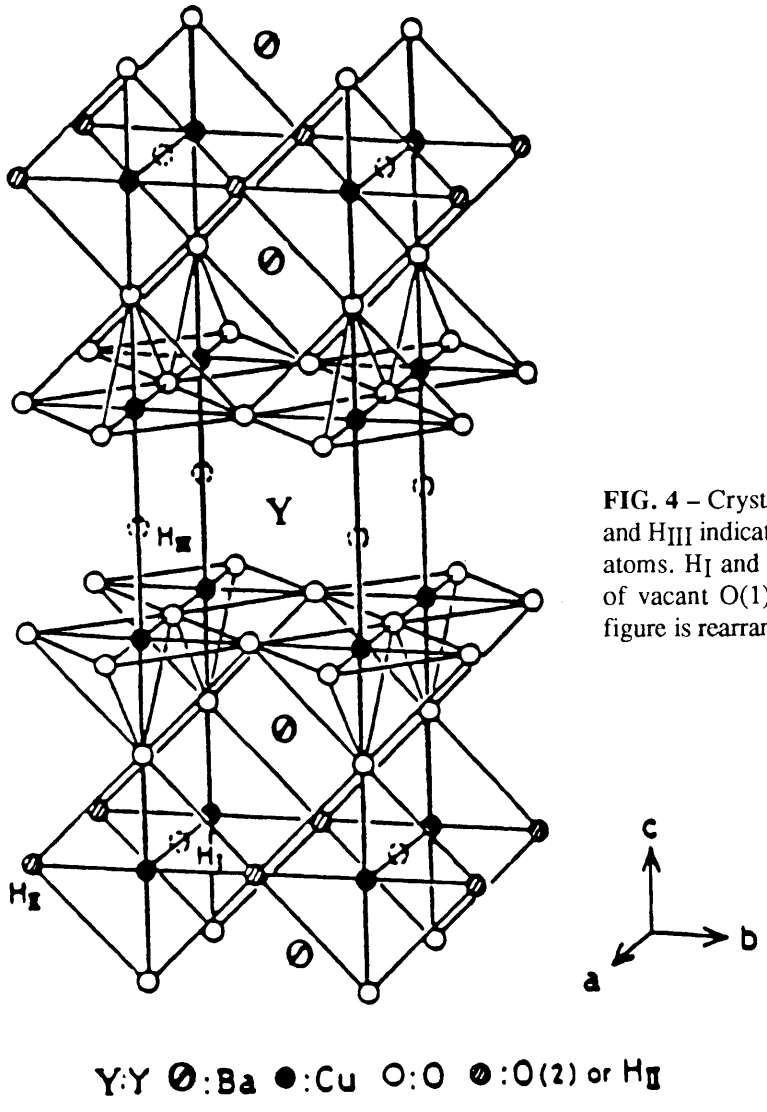

FIG. 4 – Crystal structure of $Y_1Ba_2Cu_3O_{7-\delta}$. H_I, H_{II} and H_{III} indicate the possible sites occupied by hydrogen atoms. H_I and H_{II} sites correspond to oxygen position of vacant O(1) and occupied O(2), respectively. The figure is rearranged from Fig. 2 in ref. 30.

Furthermore, under some specific conditions, the hydrogen doping leaves unchanged, for a H concentration volume up to 2 respect to Y, both the basic chemical composition and the oxygen content[32,33]. Moreover, according to some authors[30,35] and taking into account the intrinsic property of Y to absorb H up to a value of 3 (but only at high temperature, like 800 C) and from NMR measurements they assert that at large doping values the H is bonded to Y. In other words, increasing H doping there are several sites where it is bounded.

Another important experimental evidence, although controversial from an author to another, is that the volume of elemental cells is decreasing up to some critical value of the H doping, after which it increases again. The typical values of H concentration X to get the minimal volume of the elementary cell is between 0.2 and 0.5.

All of these results point to the interest of connecting high temperature superconductors and deuteration for solid state fusion experiments, both for research and application purposes.

THE EXPERIMENTAL PROCEDURE

Following these ideas we loaded with D_2 gas $Y_1Ba_2Cu_3O_{7-\delta}$ (YBCO in short) and other materials. The sample, before deuteration, was characterized by a.c. magnetic susceptibility measurements, standard 2 coils measurement method, and X ray analysis. The X ray diffractogram showed that the samples are about 100 % pure "1–2–3" material. By magnetic susceptibility measurements we get, as expected from X–rays, a critical temperature of about 92.5 K and a very low value of intergranular materials at an oscillating a.c. field of 1 Gauss (Figs. 5 and 6). We used typically high quality disk shape superconductors (diameter =19 mm, thickness 3–5 mm) with an apparent density of 5.5–5.9 g/cm^3 in respect to a theoretical one of 6.3 g/cm^3. In other words, we have typically about 0.005–0.01 of mole of $Y_1Ba_2Cu_3O_7$ (molar weight = 666 g).

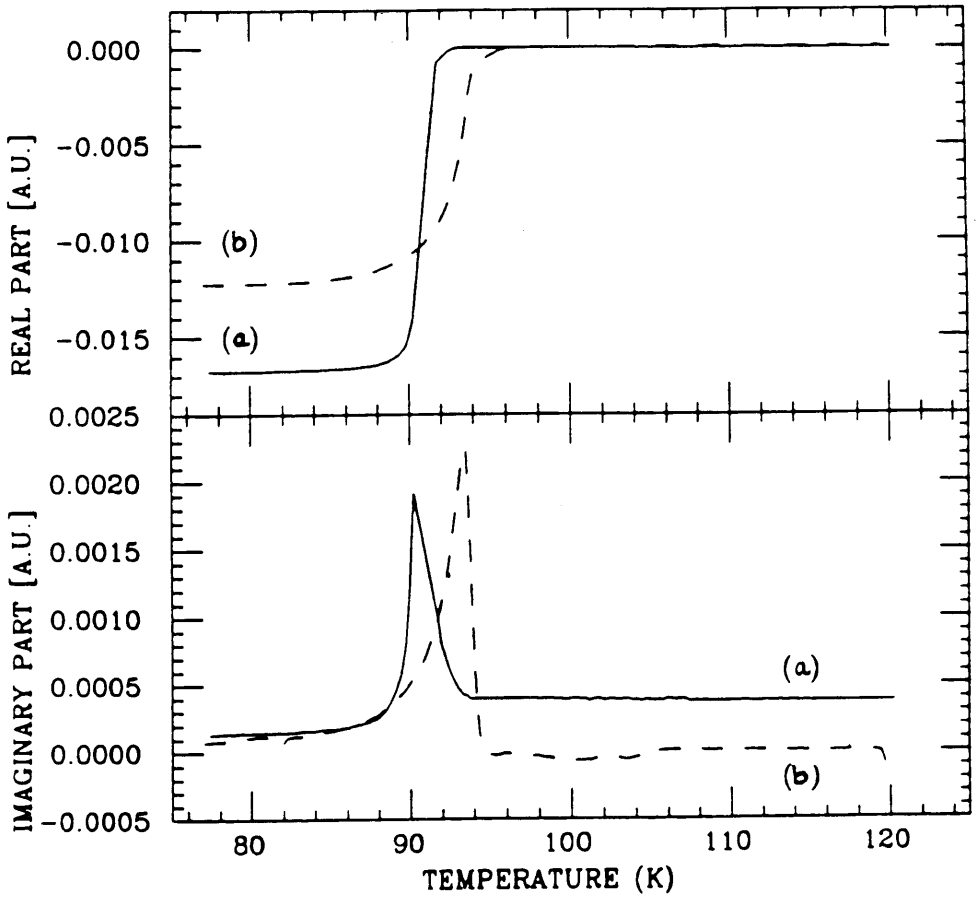

FIG. 5

FIG. 6

FIG. 5, 6 – A.C. magnetic susceptivity measurements (+- 1 Gauss,1KHz); two coils method,before (a) and after (b) deuteration versus temperature. Sample: $Y_1Ba_2Cu_3O_{7-\delta}$ H_x sintered with 5% of ozone;x=0 in (a) and x ≅ 0.5 in (b). In (5) the real part χ' and in (6) the imaginary part χ'' of the susceptivity are shown.

The deuterium loading procedure was to put the superconductor inside a high pressure stainless steel vessel (Fig. 7) and fill it with deuterium gas at a typical pressure of 35 Bar at room temperature. Later on we rise the temperature of the vessel up to 370 K by a standard

temperature–controlled heater. The thermal contact and temperature homogeneity is simply obtained surrounding the vessel with a chemical glass dewar (Pyrex type) filled with water or oil. An heavy–duty j–type thermocouple "touching" the superconductor monitorized the temperature inside the vessel. We leave the vessel at the temperature of 370 K until a pressure drop begins due to the absorbing of D_2 from YBCO. At this point we decrease the temperature to 360 K in a few minutes and wait at this temperature for 2–3 hours or more depending on the intrinsic characteristics of the sample (porosity, "degree of sinterization", thickness, area etc.) and the degree of loading planned. After the loading was terminated we cooled down to room temperature the vessel in a few minutes (just adding some liquid nitrogen to the water or oil), measured the final value of pressure (giving an estimate of the deuterium loading factor) and refilled again the vessel with deuterium at a pressure of 1 bar higher than the starting one. These precautions minimize the spontaneus degassing of the sample after high temperature loading. The loading factor is an average value, because generally speaking some gradients of concentration exist between the external and internal side of the sample. About the specific measurement reported in Figs. 5 and 6, performed at Frascati National Laboratory, we can estimate an average loading factor of 0.5 at maximum, due to the poor sensibility of the manometer allowable at that time (July 1990).

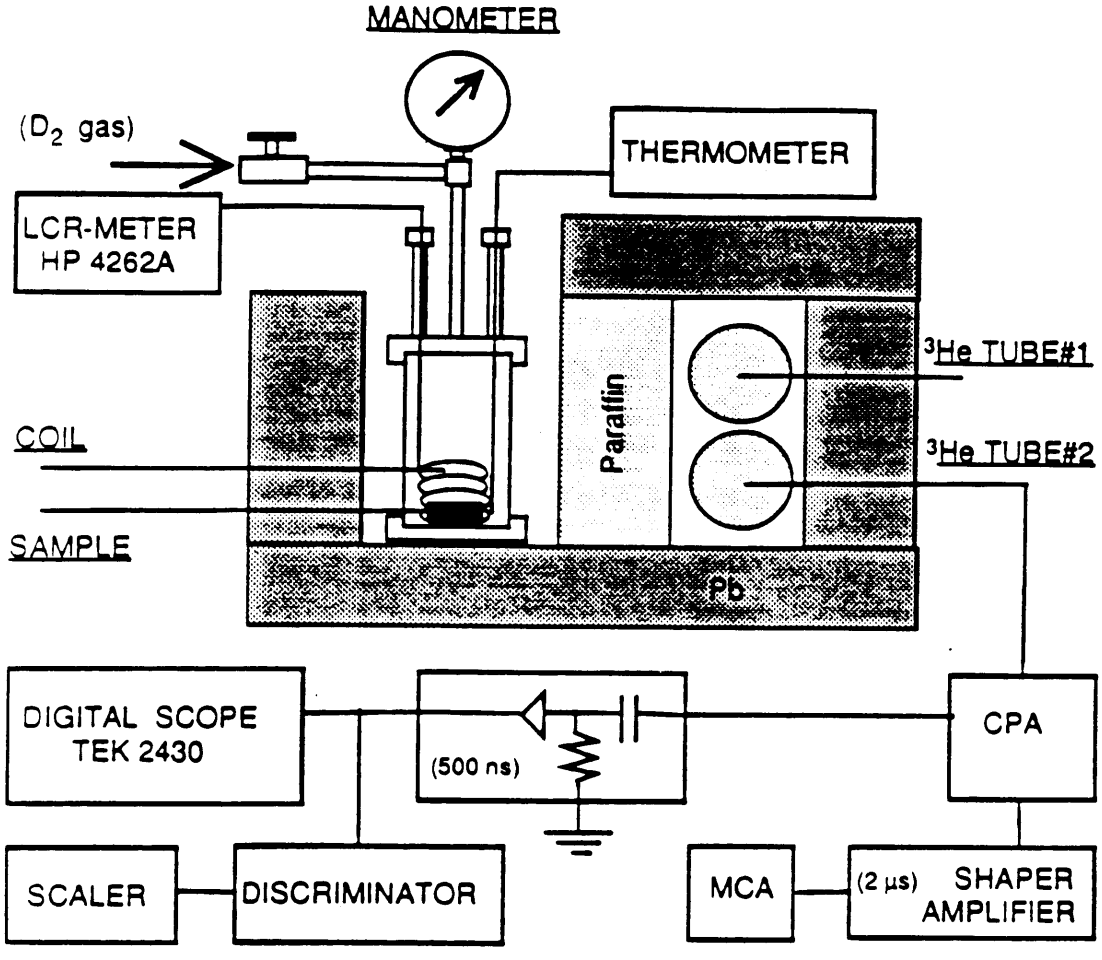

FIG. 7 – Experimental set up for neutron detection during thermal cycles 300→77→300 K.

In the Gran Sasso Laboratory we have a simpler experimental set up (from the point of view of superconducting measurements, Fig. 7). It is based on a single coil, teflon, 190 turns,

linked to a LCR meter mod. HP4262A. The measurement is based on the "inductance variation method" and we used generally a measuring frequency of 1 KHz at 10 mA of current ($B_{max} = \pm$ 1 Gauss). The typical value of inductance at 77 K without superconductor is 990 μHenry. This value decreases up to ≅ 500 μHenry for large size high quality superconductor in the superconducting state.The vessel is put in a simple dewar made by expanded polystyrene to allow thermally decreasing cycles down to 77 K. Some care has been taken and specific long tests have been performed in order to avoid that the water condensation due to the use of liquid nitrogen (on the high voltage connector of the detectors and associated electronics) can fake our measurements

The nuclear measurements are performed during thermal cycling from 300 K to 77 K and viceversa. We have tested several sample of HTSC ($Y_1Ba_2Cu_3O_7$, $Bi_4Sr_3Cu_3O_4$, $Gd_1Ba_2Cu_3O_7$). The experiment that gave, up to now, the best results began on april 1990 and was based on a high density (5.9 g/cm^3) $Y_1Ba_2Cu_3O_7$ specially treated at 5 % ozone concentration during standard high temperature annealing and sinteration processes in order to increase the density and the strenght between superconductive grains. After deuteration and several thermal cycles the sample was measured again about superconducting parameteres and we found a large increase (up to 4 K) of the superconducting transition temperature (Figs. 5 and 6).

The samples gave a counting rate systematically larger than before deuteration, but the corresponding pulse height distribution (Fig. 8) was different from the expectation (compare the n–source spectrum in Fig. 14), by showing a big unexpected peak (around abscissa 600 in Figure 8) and a substantial excess at very high pulse heights.

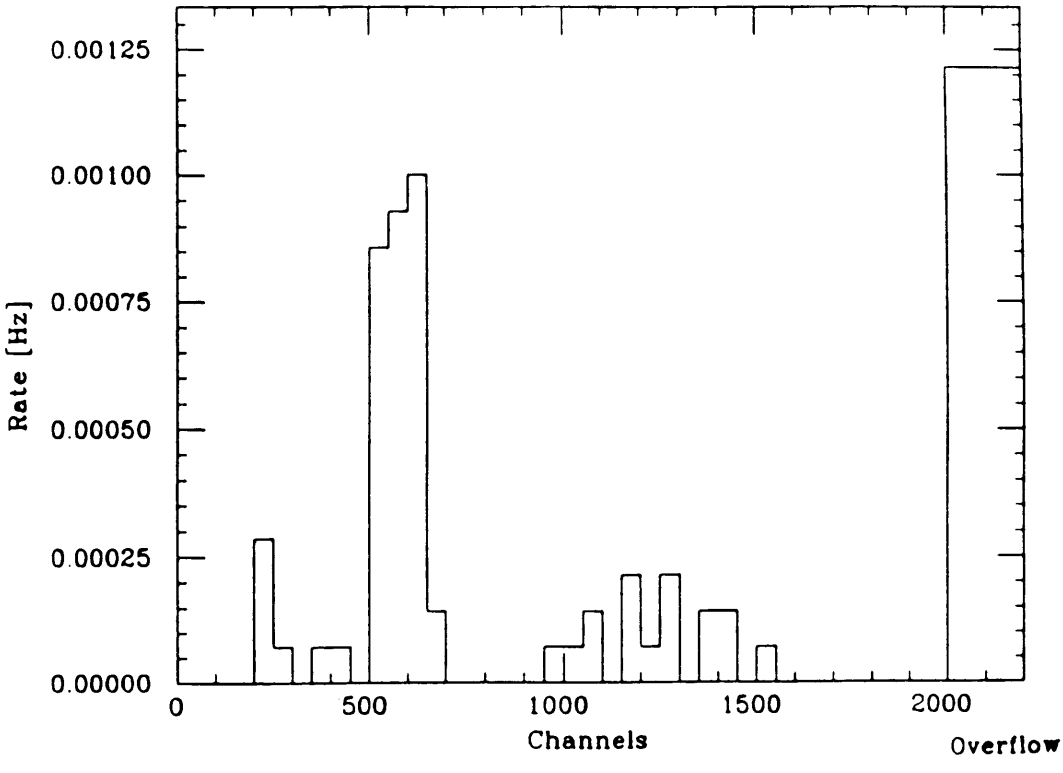

FIG. 8 – Pulse amplitude spectra acquired by QVT analizer from ^{3}He neutron detector, during the YBCO thermal cycling tests. A unexpected 300 KeV energy peak appears at the left side of the 764 KeV thermal captured neutrons.

We have not been able to explain this phenomenon.

Once limiting ourselves to the pulses in the region of thermal neutron capture peak (764 KeV), the rate is only two standard deviations higher than background. If we try to correlate these neutrons with the superconductive transition, we get the result shown in Fig. 9: here three neutrons showed up in a very short time during the superconductor–normal transition. This correlation happened only once in 6 runs, but the phenomenon indicated in Fig. 8 repeated itself in two other different experiments.

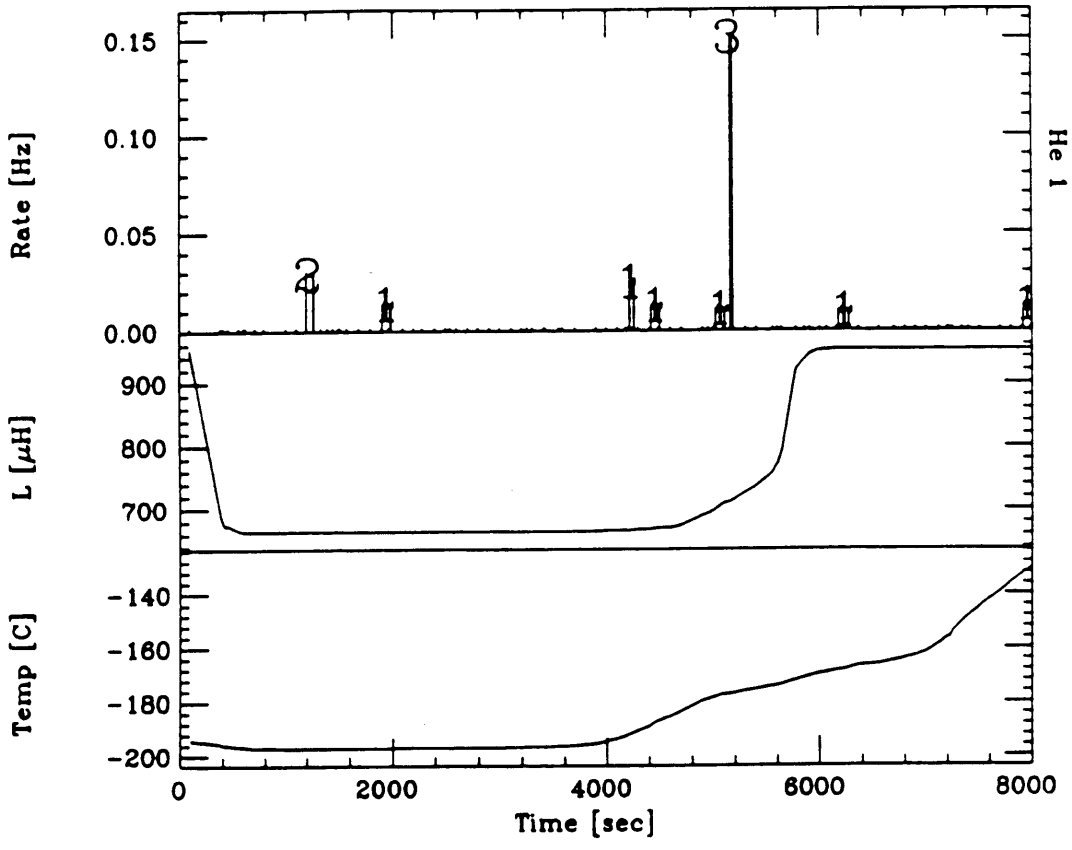

FIG. 9 – Time distribution of counts in the energy region of thermal captured neutrons. The windows report: **a)** counting rate; numbers on peaks indicate the amount of counts; **b)** magnetic susceptivity of the superconductor pellet as measured by the coil inductance; **c)** temperature of the pellet during the thermal cycle.

With samples slightly contaminated with Bismuth (we call it BYBCO here), by using an energy window of 3 times the FWHM around the thermal neutron peak, we have got 3 coincidences in our pair of ^{3}He detectors in 11 runs (about 40 hours), a rate much higher than measured by other groups in the Gran Sasso laboratory. Two of these coincidences happened close in time and are shown in Fig. 10.

Also with BYBCO we found some examples of correlation of the neutron emission with the superconductive transition. An example is shown in Fig. 11. By averaging the counting rate of 11 runs as a function of the susceptivity of the sample (normalized to the minimal susceptivity, which varies for subsequent runs) we summarize the results in Fig. 12.

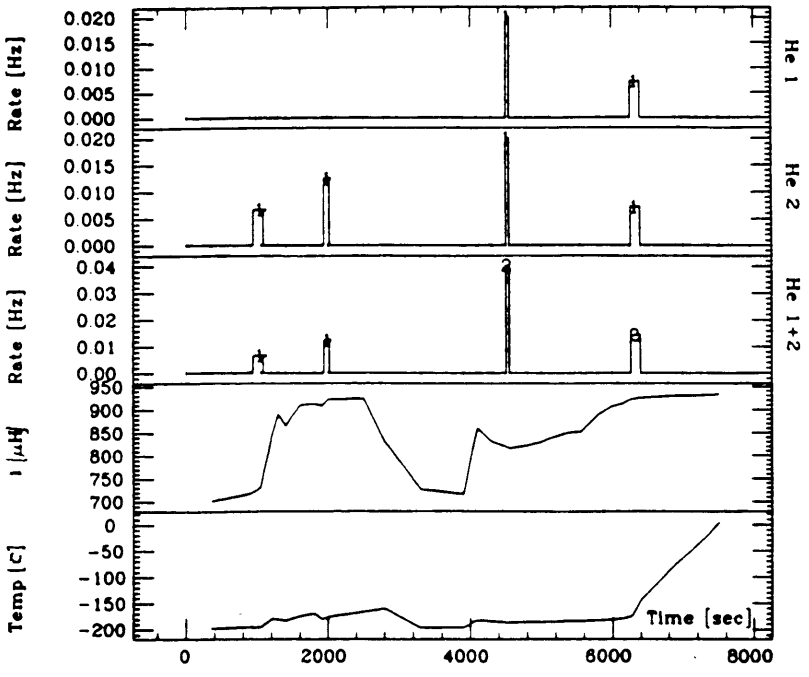

FIG. 10 – An additional ^{3}He neutron detector is added respect to measurements reported in Fig. 9; windows a), b), c) report the counting rate of the two ^{3}He and their sum. The counts are aquired in the 600→900 KeV range energy region. Two events are unexpectedly detected by both ^{3}He counters in very short time.

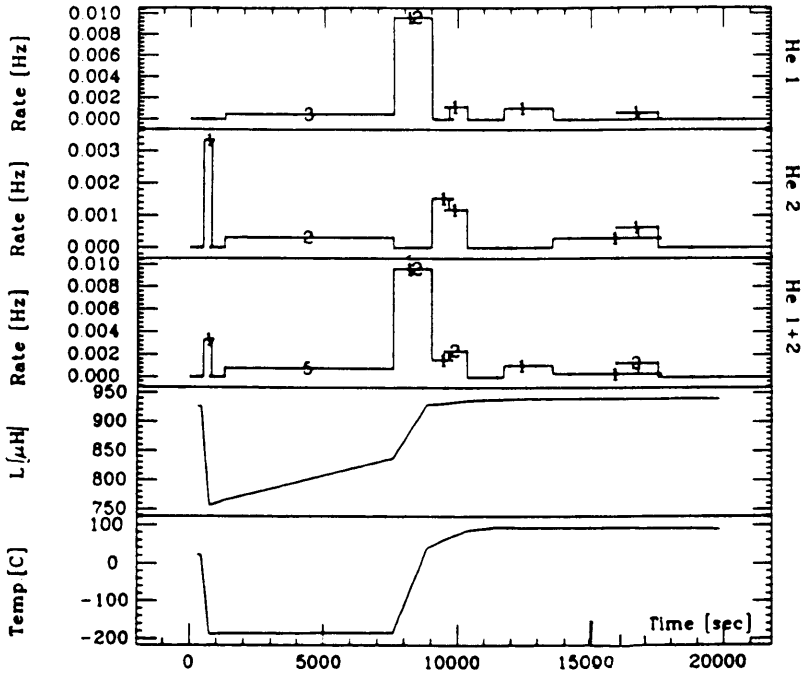

FIG. 11 – As in Fig. 10. Here a relevant amount of counts appears in one of the detectors during the superconductive transition of the sample.

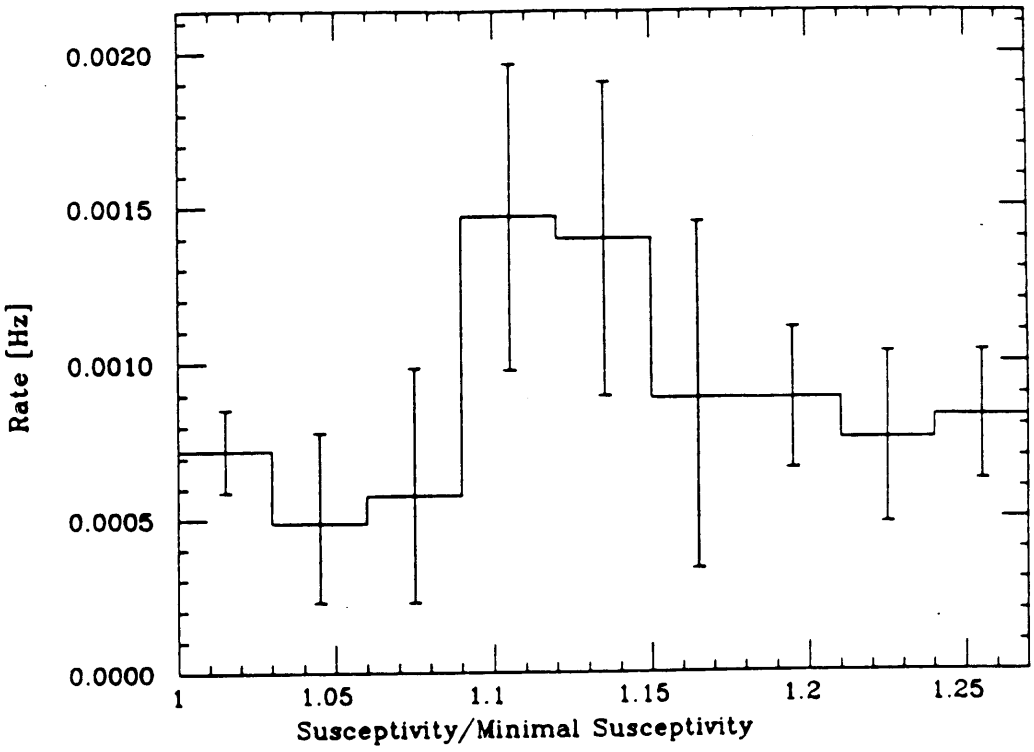

FIG. 12 – The average counting rate of both ^{3}He counters in function of susceptivity of the HTSC material is shown; a weak correlation between neutron emission and superconductive transition appears. This picture summarizes 37 thermal cycles. The speed of the transition might have smoothed this distribution, due to the uncertainty of the abscissa.

Here the transition correspond to the center of the figure, where at the boundaries one has the superconductive (left) and the normal (right) state. The central part shows a factor two in rate, with 2 to 3 standard deviations significance. (One has to remember that this is the sum of many runs where nothing showed up with a few with some emission).

As a conclusion, though weakly statistically significant (or partly not understood), an excess of neutron counts has been observed which has never been seen during very long background runs in the same conditions. Moreover our results indicate some correlation between the detection of neutrons and the superconductive transition of deuterated HTSC materials.

A large increase of Tc has been measured and the analysis of the imaginary part of a.c. magnetic susceptivity has clearly shown the formation of a new superconducting phase.

The interest of further studying these materials in connection with solid state fusion has finally been stressed and confirmed.

STIMULATION OF DEUTERATED $Y_1Ba_2Cu_3O_7$ BY NEUTRON IRRADIATION

INTRODUCTION

Cold fusion in deuterated solid state is meant to be a non–equilibrium process. After deuteration, the non–equilibrium situation can be realized by electrolysis, mechanical or thermal stresses, phase transitions etc.

A possibility we have been interested to explore is the perturbation through penetrating radiation.

Charged particles have been tried already[36] with no success by using alfa and beta sources: here the idea was to prove that the erratic cold fusion results could actually be normal fusion initiated by charged cosmic rays.

Neutrons should be of course much more effective, through elastic and inelastic interactions with deuterons, in perturbing the deuterated solid state[37]. Our aim will also be to show that the presence of the ceramic substantially changes the cross section for reactions producing neutrons in the final state, in order to stress, as done for Palladium, the role played by the solid lattice.

Both goals can be achieved in principle by fast or thermal neutrons. Here we have at first explored the effect of fast neutrons.

EXPERIMENTAL SETUP

Using about the same experimental device (except the coil) and similar set–up (Fig. 13) as the previous experiment (always located at the underground National Laboratories of Gran Sasso) we have irradiated deuterated high Tc superconducting materials with neutrons coming from a low activity Am/Be source (0.7 mCi of ^{241}Am total activity, giving a yield of 1.55 KBq over 4 π of neutron activity from Be–energy spectra ranging from 0.1 to about 10 MeV).

The only difference respect to the previous experimental setup was a supplementary Cd shielding of 2 mm thickness around the two ^3He counters used; we left unshielded only a "window" of about 40 cm at the center of the detector, placed behind paraffin blocks. This both to reduce the background level and ensure that only neutrons from the source or the cell were thermalized and revealed in the counters.

Using the known source activity we have determined the total detection efficiencies of the counters to be about 0.4 % for each ^3He tube in this experimental setup.

The neutrons impinging of the cell were not moderated, while the thermalization was achieved for neutrons before the counters by putting in front of them some paraffin bricks with a total thickness of 5 cm (Fig. 13).

The electronics (preamplifiers, MCA etc.) was the same used in the previous tests. The two counters were acquired using a MCA per detector, with slightly different amplification factors to better check the electronics stability.

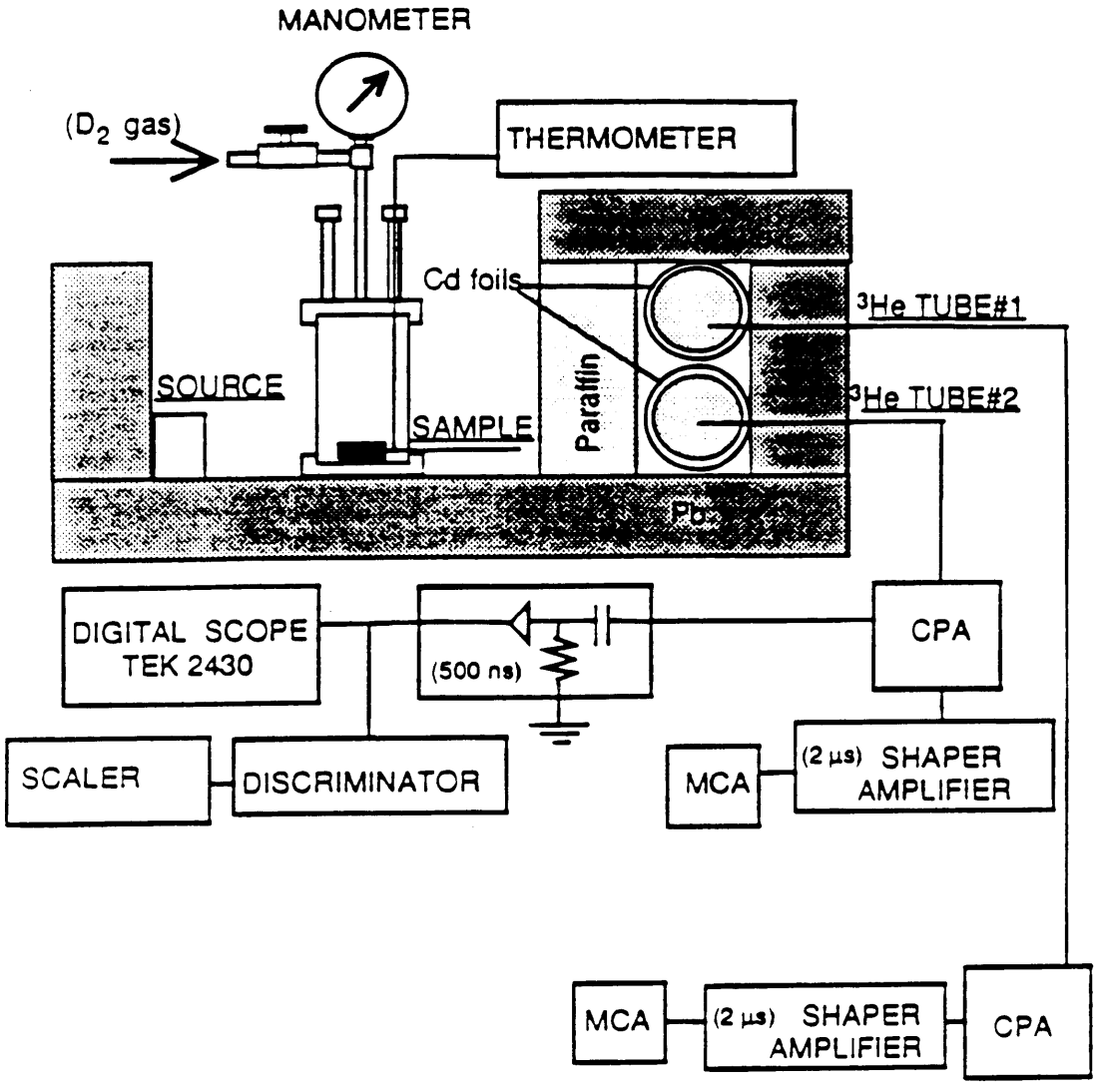

FIG. 13 – The experimental set up, showing the position of the Am–Be neutron source. Cadmium foils leave a open window in correspondence with the paraffin blocks.

TESTS

We have irradiated first a sample of high Tc superconducting material (2 "coins" of YBaCuO) at room temperated before it was deuterated; the material was placed inside its vessel, without any gaseous D_2 inside; this we will call the "reference blank".

Any difference respect to this sample will show only the effect of the D_2 absorbed in the material or present inside the cell volume.

We have then loaded the previous 2 samples and other 5 similar "coins" of the same material (got allowable in the mean time) with deuterium by following exactly the same procedure as explained in the previous section. We note that, by adding sample coins without the deuterium, the neutron counting rate is reduced: see figure 15 and Table II. By monitoring the pressure drop in the manometer after the material was brought back to room temperature, we have estimated the D_2 loading factor.

TABLE II – Schematic of the experiments performed by the Am–Be neutron, gamma source. It is reported the duration of each experiment as detected from the 2 detectors (Time1/Time2), the average rate of particles flux and some comments (Notes) on the kind of experiments performed.

Run nr.	Notes	Time1/Time2 (sec)	Avg. Rate +-Davg. (Hz)
793	Only source	6209 / 6153	5.543 +- 0.026
794	Source with		
	empty cell	4167 / 4197	5.321 +- 0.021
795	Source – cell with		
	HTSC only, i.e.		
	"reference blank"	4265 / 4269	5.298 +- 0.025
797	Deut. sample		
	Gas D_2 in cell	4341 / 4298	5.392 +- 0.025
798	as 797	4343 / 4369	5.378 +- 0.025
799	as 797	7394 / 7209	5.342 +- 0.019
800	as 797	4403 / 4313	5.376 +- 0.025
801	as 797	4232 / 4265	5.392 +- 0.025
802	as 797	4047 / 4025	5.368 +- 0.026
804	as 797	5863 / 5845	5.358 +- 0.021
–	Avg. 797 – 804	34623 / 34324	5.370 +- 0.009
805	Deut. sample		
	No gas D_2 in cell	4250 / 4201	5.379 +- 0.025
806	as 805	39228 / 39253	5.358 +- 0.007
807	as 805	4259 / 4242	5.339 +- 0.025
808	as 805	4278 / 4236	5.346 +- 0.025
809	as 805	4262 / 4301	5.368 +- 0.025
–	Avg. 805 – 809	56277 / 56233	5.358 +- 0.008
810	as 797	8687 / 8742	5.348 +- 0.018
–	Avg. 797 – 810	99587 / 99281	5.361 +- 0.006

Beginning with a pressure of 19.5 atm inside the vessel, after loading, only 6 atm of gaseous deuterium were left.

The total cell volume was 70.7 cm^3 , only 20% of it occupied by the HTSC material, so that 30.3 • 10^{-3} moles of D_2 were absorbed of the original 45.4 • 10^{-3} moles present at the beginning of the loading.

After loading, we have refilled the vessel with 19.25 atm of D_2 (at this time the used deuterium bottle was partially empty and we could not refill it at the pressure foreseen by our standard procedure) to minimize the material degassing during the experiment; this small pressure drop brings the content of gaseous D_2 in the cell to 44.8 • 10^{-3} moles.

The following measurements were made in several separate runs one after the other in order to obtain a better continuous monitoring of the signal stability on the electronics and MCA.

After a certain numbers of runs, we have opened the vessel valve to release all the gaseous D_2 inside the cell, so to obtain a separate estimate of the (n – gaseous D_2) contribution; at the end of these runs we have refilled again with gas the cell, to check once more both the system stability and the (n–gaseous D_2) cross section in our experimental conditions.

DATA ANALYSIS AND CONCLUSIONS

In the data analysis we have made use of the information coming from both detectors by scaling the two detector response at a common factor (by multiplying the second detector response by the known amplification percentage difference respect to the first one) and then averaging them for every run. We have checked for each run the signal shape and peak value and rephased them (were necessary) to the value of the "reference blank" run.

The maximum observed displacement of the signal was of 6 channels; the average one was 0.12 channels (over a peak amplitude of 1080 channels) (Fig. 14).

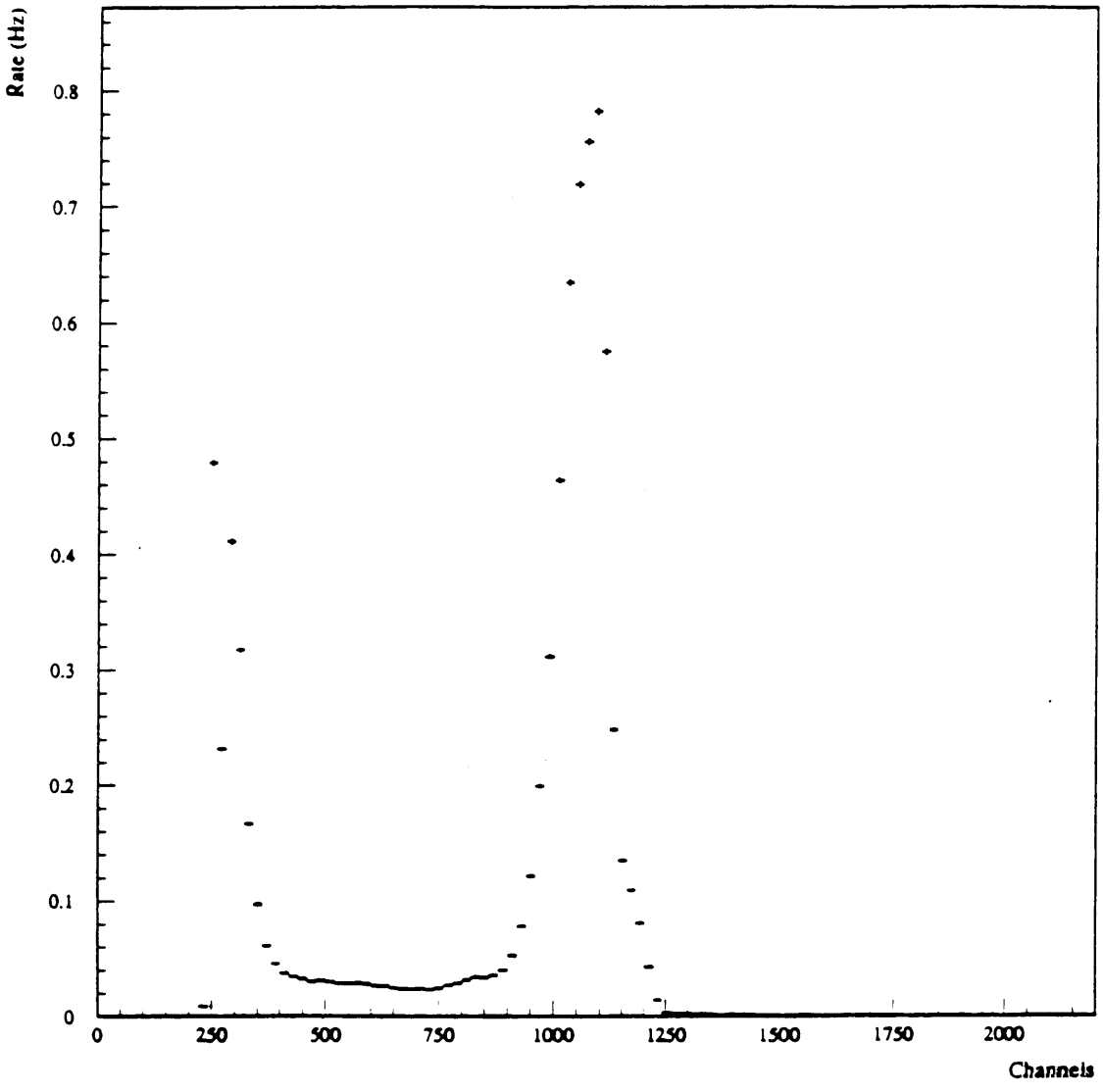

FIG. 14 – Tipical neutron counting spectrum from superconduting deuterated materials irradiated by a Am–Be source; the spectrum is cumulated over 7 runs with the sample under D_2 pressure.

The neutron integrated rate was calculated by integrating the observed spectra between 880 and 1280 channels, that is only on the thermal capture peak.

The results for the different runs and the relative averages calculated over both detectors are summarized in Fig. 15 (real time sequence) and in Table II. An excess of neutron is observed with the deuterated sample, as to say more neutrons come out of the cell respect to the number entering it.

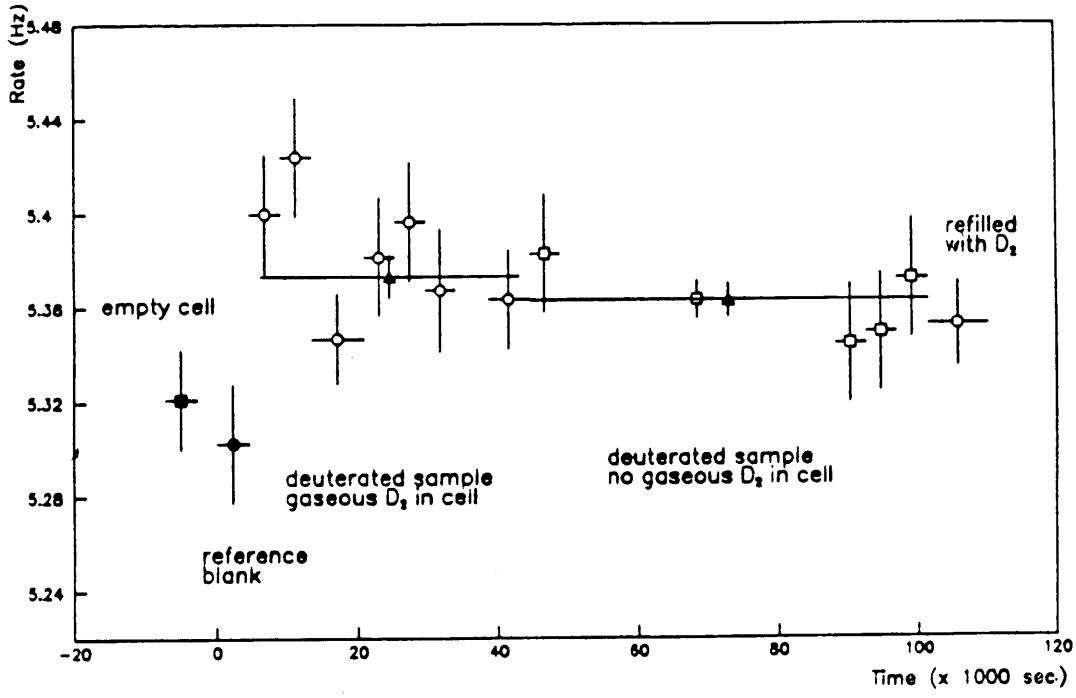

FIG. 15 – Sequence of the tests performed with a neutron source irradiating the sample. Blank tests, tests with D_2 gas and tests without gas are reported. (Here the reference blank is with only 2 sample coins, to be compared with the 7 coins of the deuterated cell). Triangle symbols correspond to the mean value of a set of similar runs spanned by the horizontal bar.

In Fig. 16 we show the difference between the spectra of the average of the runs using deuterated sample with gaseous D_2 in cell and the reference blank: the observed rate excess is clearly located in the thermal neutron peak zone.

The difference between the samples with gaseous D_2 in the cell and those without it is well inside the errors on the averages and shows that the contribution of the cross section (n – gaseous D_2) is less than the difference observed respect to the non–deuterated sample.

The excess of counts on the average of the deuterated sample runs amounts to about 2 σ, where the dominating error comes from the statistical error on the background reference run.

If we calculate the excess coming from the expected neutron cross section on D_2, using an average neutron cross section of 2 barn (average of total n cross section for energies from 0.1 to 10 MeV), it turns out that:

- the expected number of average hits per neutron on the gas inside the cell is $2.9 \bullet 10^{-3}$
- the expected number of average hits per neutron on the D_2 absorbed in the material is $7.8 \bullet 10^{-3}$
- the expected ratio is:

$$\frac{\sigma\ (\text{n–gaseous } D_2)}{\sigma\ (\text{n} - D_2 \text{ in solid phase})} = 1.5$$

- with our geometry we expect that only 0.5 % of the neutrons from the source arrive on the deuterated HTSC material and, if isotropically scattered or reemitted, only 14 % are visible from the counters; even assuming a total thermalization efficiency of 100 %, with 35 % detection efficiency we expect, with our source activity, an observed rate change, respect to the reference blank, of about $3 \bullet 10^{-3}$ Hz
- under the same conditions, we can estimate the (n–gaseous D_2) contribution to be about $10 \bullet 10^{-3}$ Hz.

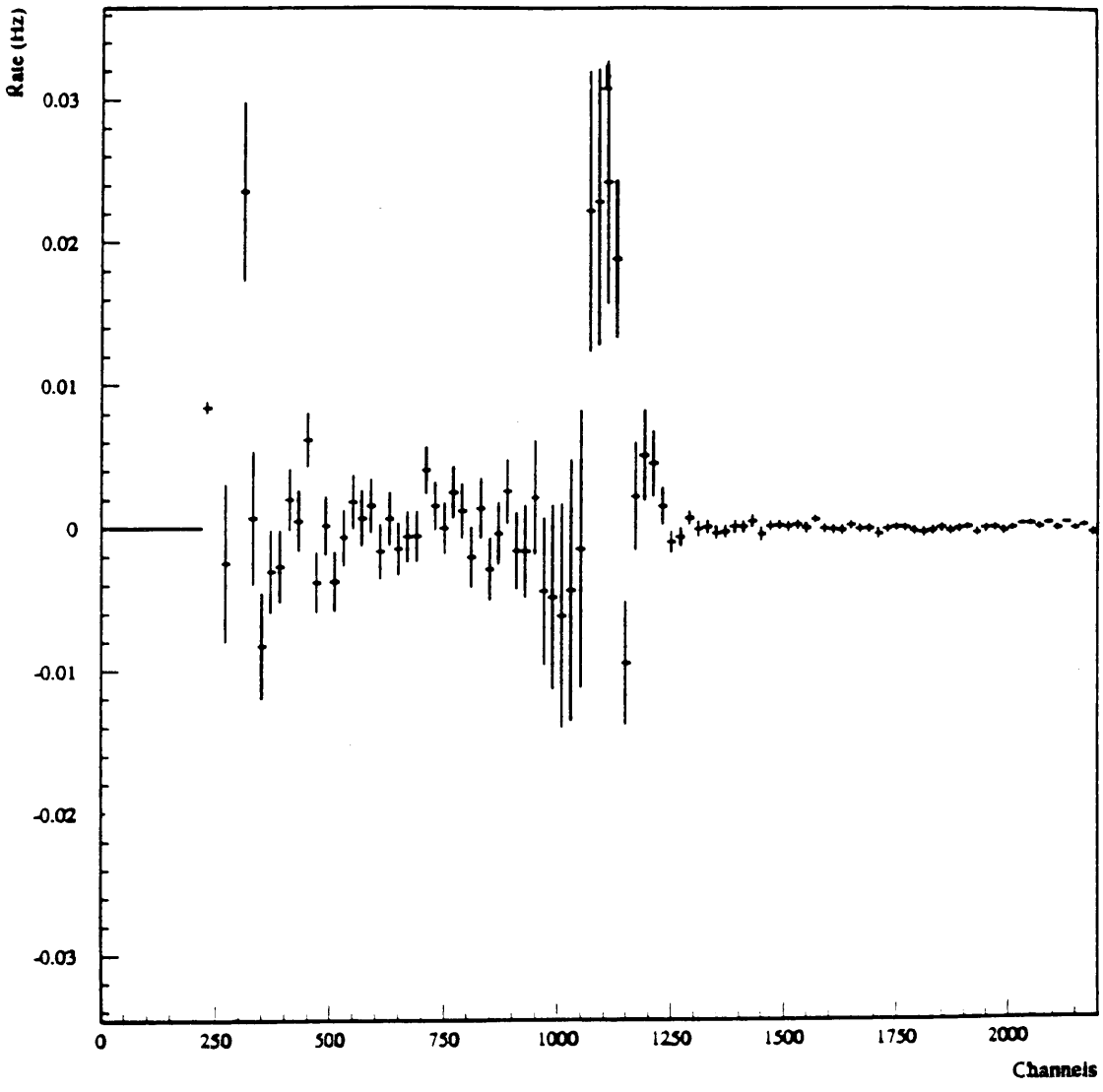

FIG. 16 – Subtracted spectrum between the mean values of deuterated sample under D_2 pressure and the reference blank run; if compared to Fig. 14, it shows an excess in the region of neutron capture peak.

The expected contribution from photodisintegration on D by the energetic gammas from the Am source (about 1050 photons/sec over 4 π) can be evaluated as the previous one, but with a total cross section of about 2.5 mbarn; that is this contribution should be about three orders of magnitude less than the above.

As we can see, the observed rate change between the deuterated sample and the reference blank is 60 $\bullet 10^{-3}$ Hz, quite above the estimated neutron contribution, which, we remind, has been calculated under conservative assumptions (total n–D cross section used, thermalization efficiency of 100 %).

The excess of neutrons (once experimentally subtracted the known sources (n–D interactions) is (53 ± 28) 10^{-3} Hz. Though statistically not very significant, the effect is understimated, because the reference blank had only 2 sample coins, compared with the 7 coins of the deuterated sample and the neutron rate is reduced by increasing the non deuterated material.

Our result underlines a role played by the solid state lattice in increasing the yield of neutrons, very likely through D–D fusion.

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