

SEARCH FOR NEUTRON EMISSION FROM DEUTERIDED HIGH TEMPERATURE SUPERCONDUCTORS IN A VERY LOW BACKGROUND ENVIRONMENT.

F.Celani,¹ A.Spallone,¹ L.Liberatori,¹ F.Croce,² L.Storelli,³ S.Fortunati,⁴
M.Tului,⁴ N.Sparvieri.⁵

¹ INFN, Laboratori Nazionali Frascati, Via E.Fermi, 00044 Frascati (Italy).

² Department of Chemistry, Univ. "La Sapienza", Roma (Italy).

³ Faculty of Engineering, Univ. "La Sapienza", Roma (Italy).

⁴ CSM-ILVA (IRI), Via di Castel Romano 100, 00129 Roma (Italy).

⁵ ALENIA (IRI), Research Dep., Via Tiburtina Km 12.400, 00131 Roma (Italy).

ABSTRACT

Following the experiments performed with deuterided High Temperature SuperConductors (HTSC) at underground Gran Sasso Laboratory, we have learnt the capacity to absorb Deuterium (D) by these materials and the role played by non-equilibrium conditions to get neutron burst emissions in the framework of Cold Fusion.

So far, some $Y_1Ba_2Cu_3O_{7-\delta}$ (YBCO) pellets and high pressure D_2 gas were enclosed in a stainless steel vessel and a charging-up procedure was performed. The vessel was put in a thermal neutrons field and some thermal cycles (300-> 77-> 300 K) were performed; moreover, for comparison, background and blank runs were performed. A specific acquisition system, able to detect multiple neutron signals in defined time-windows ("time-correlated events"), was set-up.

One thermal cycle run showed a large increase of time-correlated events in respect to the blanks; one other run, although with no relevant mean-value increase of events detected, showed, on the other hand, one interesting multiple neutron signal (triple); other similar runs produced no relevant values.

One-other kind of experiment, at constant temperature (300 K), characterized by a heavy D_2 gas refill, showed both some increase of time-correlated events and few 'triple' neutron signals.

MOTIVATIONS

According both to our opinions [1], mainly based on some similarities between the behaviour of H or D doped Pd and $\text{Re}_1\text{Ba}_2\text{Cu}_3\text{O}_7$ (RBCO) in the superconducting state, and to some theoretical suggestions and considerations [2,3], we found some interests in studying compounds of deuterated rare-earths. Moreover our first tests were independently confirmed by S.E.Jones at BYU [4] who operated with deuterided YBCO in thermal cycles and mechanical stresses.

There are several similarities between the behaviour of heavily hydrogenated Palladium [1] and heavily oxygenated $\text{Re}_1\text{Ba}_2\text{Cu}_3\text{O}_{(6.5+\delta)}$ compounds (Re is any trivalent rare-earth apart yttrium). Among these we note:

- 1) As indicated in our previous paper [1] and experimentally found in [5], the absorption of H in RBCO occurs only if the RBCO is a superconductor material otherwise the H degrades the RBCO giving CuO, Cu_2O , Cu and BaO as final products.
- 2) The RBCO are superconducting only when Oxygen content is larger than 6.5. The critical temperature (T_c) depends steeply on Oxygen content (e.g. $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_7$ $T_c=92$ K, $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{6.5}$ $T_c\sim 60$ K).
- 3) The HTSC are easy to loose the Oxygen in excess of 6.5, i.e. these are intrinsically not stable.
- 4) It is possible to add H or D (nobody tried T) to superconductors even increasing T_c despite initial content of Oxygen (but at least larger the 6.5) [6].
- 5) There is no clear evidence for the usual isotopic effect.
- 6) Several authors found the rare property of inverse pressure coefficient in $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$.
- 7) By NMR measurements [7] has been found that H in $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{(7-\delta)}\text{H}_{0.2}$ occupies sites in the Cu-O planes and that it diffuses or moves dynamically in the crystal above 170 K but below 150 K it is trapped. In other words, after cooling to 77 K, during warming up we might have a moving of trapped H. Similar effects can be expected with D at similar temperatures.
- 8) There exist a particular value of H doping that increases [6] the critical temperature. The particular value seems to depend on sample preparation and H loading procedure [8,9].

Moreover, we note that the HTSC have perovskite structure, the same structure of most abundant minerals of the Earth's lower mantle ($\text{Mg,Fe})\text{SiO}_3$; the pressure of the core-mantle boundary is as large as $1.4 \cdot 10^6$ Bar [10]. Then, we can suppose that a proper perovskite

structure can be efficient like the "Mother-Earth-Soup" used by S.E.Jones [11] to search for geological-like Nuclear Fusions.

In other words, among the large unusual properties of HTSC there is their ability to absorb a large amount of hydrogen [6,7,12,13,14]. The possible sites, where the H or D are located, can be identified by Mossbauer spectroscopy as recently suggested by E.Kuzmann et al. [15,16].

Most of these properties have induced us to use high quality HTSC like YBCO in a gaseous system, instead of "standard" Ti or Pd, to research for Cold Fusion anomalous phenomena.

APPARATUS AND ACQUISITION SYSTEM

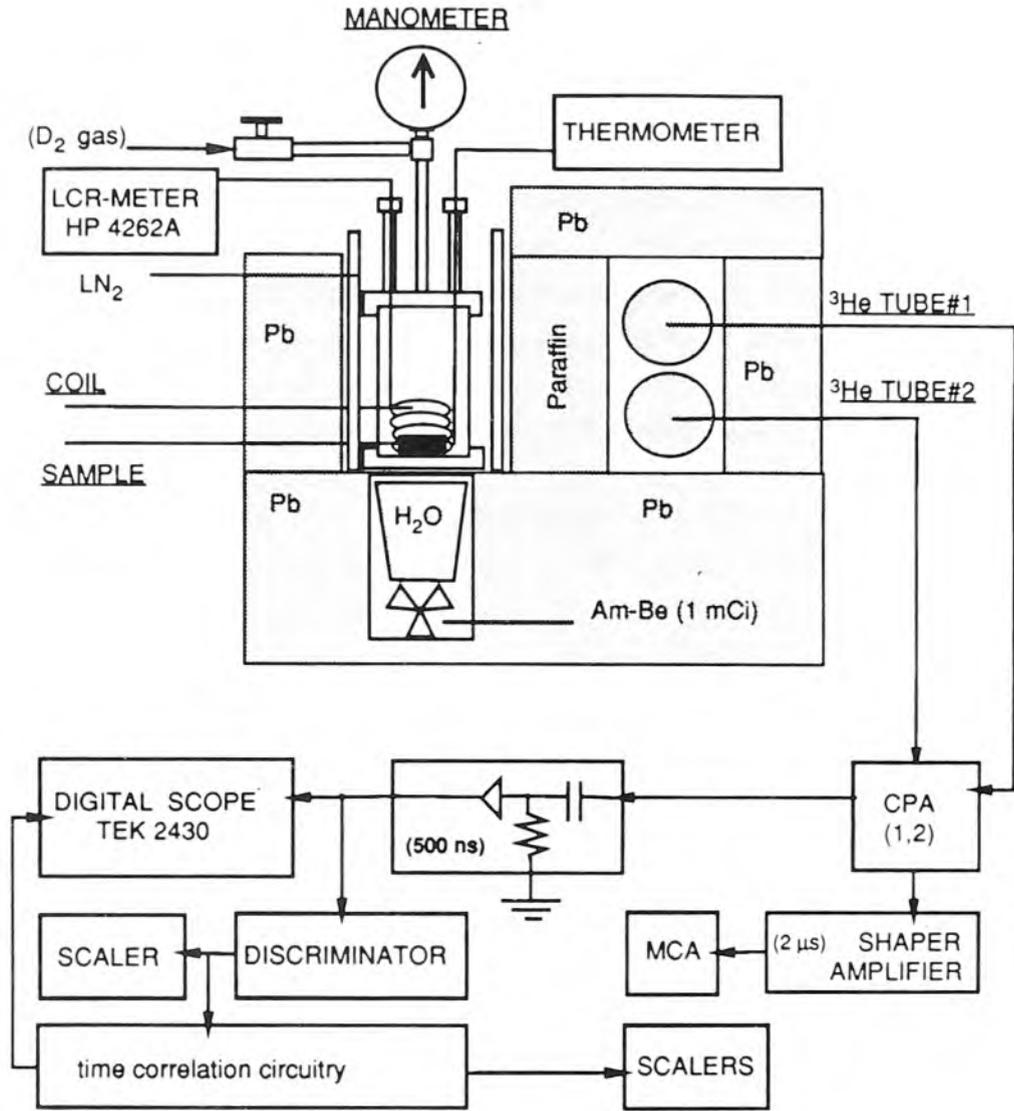
The apparatus is shown in (fig. 1). It consists of two ^3He tubes surrounded by paraffin and lead bricks to detect neutrons and a lead well where a weak (2200 n/s of ~ 4 Mev and 1500 γ /s of 4.43 MeV) Am-Be neutron source is located; the source is covered by 8 cm of water, in a plastic container, in order to moderate the neutrons. We put 6 YBCO pellets (disk shaped: diameter ~ 19 mm, thickness 4-6 mm, density 5.5-5.9 g/cm^3 , total weight ~ 50 g) in a PTFE coil (about 1 mH) enclosed in a cylindric stainless-steel vessel for high pressure gas.

The acquisition system (fig. 1) was based on digital counter and analog acquisition by a fast digital scope (TEK 2430) of the signals coming from the ^3He detectors; moreover the independent detector's signals were acquired by a MCA. All neutron signals are independently counted by proper counters (scalers), some of these were gated. Each signal from the proper detector is charge-amplified, shaped (6-8 μs of duration) and discriminated. From the discriminator one output is straightforward counted by a scaler, a second output, throw a specific circuitry that inhibits any other signal arriving within 10 μs , is counted by an other scaler. These last signals are used to get the "time correlated events" as below detailed. Each single signal coming from any detector opens for a defined time window the gate to the proper scaler which counts all the other signals (except the first) arriving during this time window.

The counts in a time window come from the logic OR pulses of the 2 detectors. We used 3 different contiguous and separated time windows:

- a) from 0 to 10 μs
- b) from 10 to 110 μs
- c) from 110 to 1110 μs

The time window in 10-110 μs was used as trigger to the digital scope in order to acquire and visualize the analog signals from both detectors (to check for the correct neutron signal shape). We consider the events in the time window b) the most significant events because the expected 2.45 MeV extra-neutrons, due to deuteron-deuteron reactions, have an expected thermalization time of the order of few tenth of μs in our experimental set-up.



[Fig. 1] Experimental set-up. The total neutron detection efficiency, in respect to the Am-Be source, is 0.4%. The underground Laboratory neutron flux is about $10^{-6} \text{ n cm}^{-2} \text{ s}^{-1}$. At the bottom of the figure is shown schematically the electronics set-up.

EXPERIMENTAL PROCEDURES AND MEASUREMENTS PERFORMED

The experimental procedure adopted to deuteride the superconductor material is summarized as following:

- 1) We put the superconductor inside the coil in the stainless steel vessel and we measured the inductance variation of the sample vs temperature ($300 \rightarrow 77 \rightarrow 300 \text{ K}$) with a AC magnetic field of about 1 Gauss at 1 KHz.

- 2) We filled with deuterium gas the vessel at a typical pressure of 35 Bar at room temperature.
- 3) The vessel was warmed-up to about 370 K and hold at this temperature for about 1 hour, after the temperature was decreased to about 360 K and held for 3 hours.
- 4) We cooled, in few minutes, the vessel from 360 K to about 300 K.
- 5) We refilled the D₂ gas to the vessel up-to 36 Bar.
- 6) We started the neutron detection performing thermal cycles (300->77->300 K). In order to check for the D absorption, because the low accuracy of manometer, we measured at 77 K the inductance value due to the diamagnetism of the sample [1] and the value of superconducting transition temperature during the warming up.

We estimated a upper limit loading factor of 0.5 in D/YBCO ratio.

In this specific measurements we further investigated [1] the eventual neutron emission enhancement due to stimulation of the sample by a neutron source in order to detect time-correlated events; thermal cycles were performed during the stimulation in order to induce further non equilibrium states of the material.

We define the different kinds of measurements performed according to the following:

A - Background: measurement with neutron source but no vessel.

B - Blank: measurements with neutron source and vessel filled with HTSC samples in this sequence:

B1) no D₂, T= 300 K

B2) D₂ at 40 bar, T= 300 K

B3) D₂ at 36 bar, T= 300 K after the deuteration procedure.

C - Thermal cycle: measurements after the deuteration procedure with n-source, vessel filled with D₂ at 36 Bar (300K) and thermal cycles (300->77->300 K) operations.

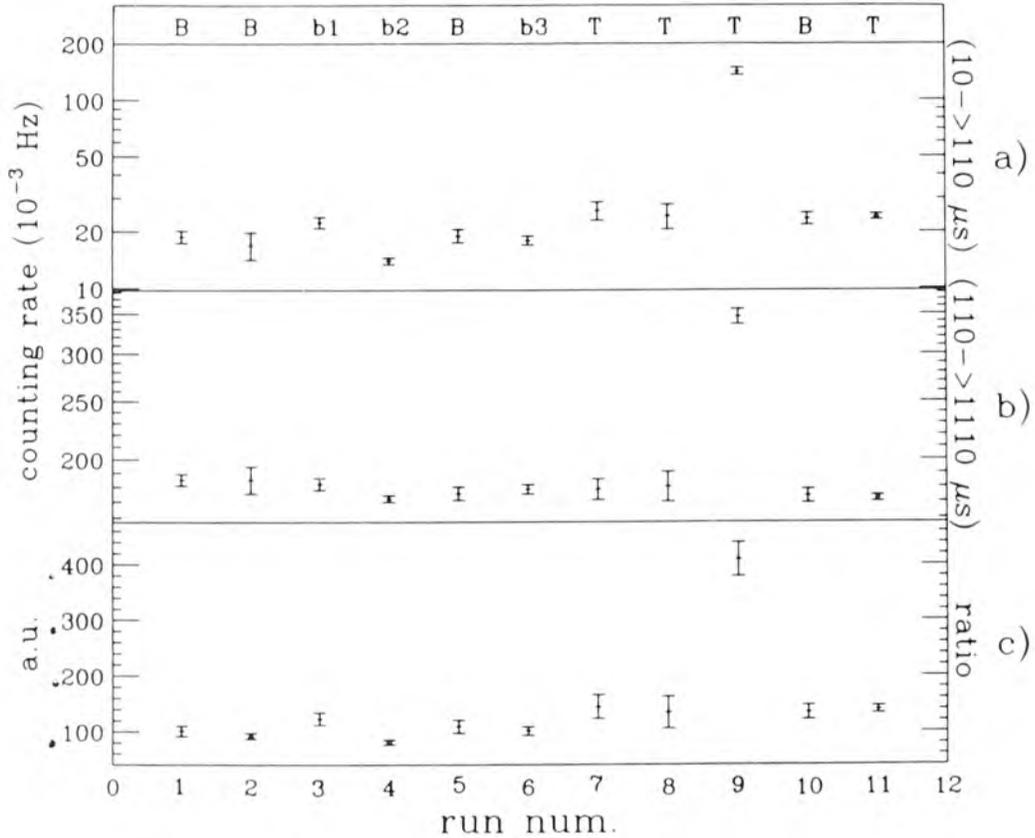
RESULTS

As shown in fig. 2, we made different tests, as above defined, performed in sequential independent runs. Typical Background (1,2,5,10) and Blank (3,4,6) runs ranged from 0.5 to 10 hours of acquisition time, while the Thermal cycle (7,8,9,11) runs ranged from 0.5 to 1 hours. As it results from fig. 2, no significant statistic differences come out from Background and Blank runs (we adopt Gauss statistic for errors calculation).

The Thermal cycle runs showed different cases: the runs 8 and 11 had no significant statistic differences in respect to Blank runs while the runs 7,9 presented different peculiarities. In the run 7, although we did not record large excess counts, anyway we recorded, on the digital scope (fig. 3), one "three neutron signal" (triple event). This triple event, recorded from both detectors, occurred in the 10->110 μs time window during a superconductive temperature transition. By statistic considerations, we expect such event is

occurring once in about 80 hours and we have to consider that all Thermal cycle runs lasted only about 2.4 hours.

We like to note that one other triple event occurred in further thermal cycles performed (3 cycles 150->77->150 K performed in 30 min of measurement).



[Fig. 2] Time correlated events vs run type. Run type symbols:
 B = Background;
 b (1,2,3) = Blanks in three different conditions;
 T = Thermal cycles.

[.a.] counting rate (Log) in time window 10->110 μs

[.b.] counting rate (Log) in time window 110->1110 μs

[.c.] ratio (Lin) between a) and b) counting rates.

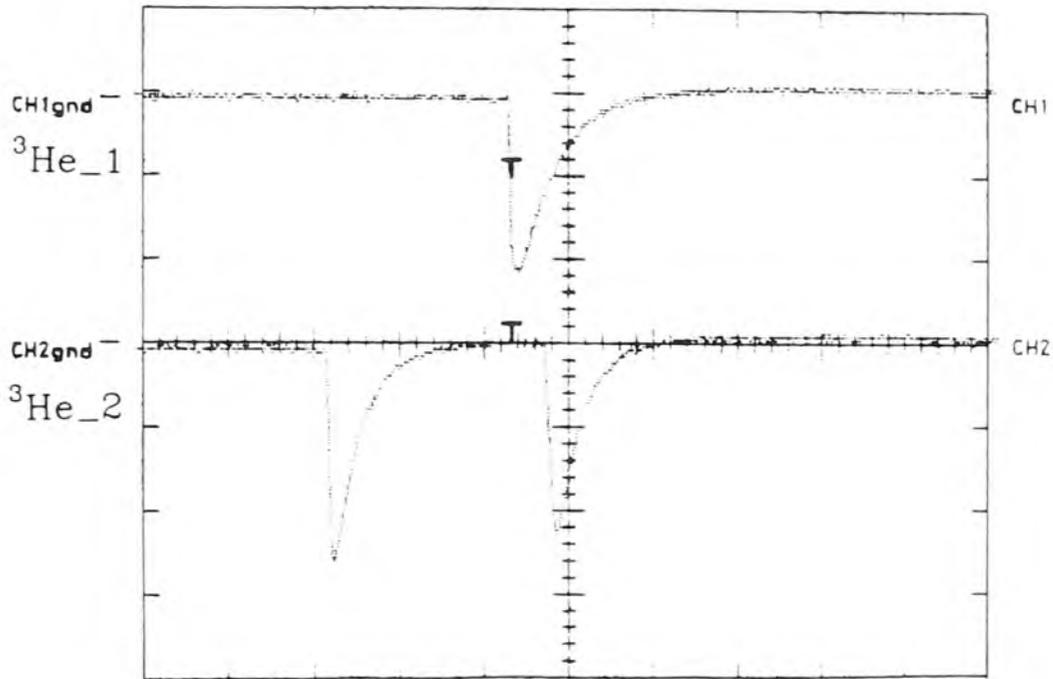
In the run 9, a large excess of time-correlated events occurred, over 7 times larger in respect to the blank and corresponding to over 30 standard deviations. Most of them were observed at the scope and they look like 2 neutrons in 10->110 μs; no spurious signals were observed. This excess was recorded, with different relative intensity, as in 10->110 μs as in 110->1110 μs time windows (fig. 2.a,b).

In fig. 2.c we put in evidence that in the run 9 the ratio between 10-110 μs time-window and 110-1110 μs time-window is about 4 times larger than the Blank runs. This can be consistent with several neutron bursts (separated in time much more than 1 ms) having burst

intrinsic time duration $\ll 110 \mu\text{s}$. We recall that the expected thermalization time of 2.45 MeV neutrons lies in the 10-110 μs time-window.

It is quite improbable that some persistent disturbances can reproduce this kind of behaviour.

In an other test performed at room temperature (one month later, at about 20 Bar D_2 gas pressure) we increased the pressure to 42 Bar and after about 10 hours we put the vessel into the source well. The measurement, starting a few second later, gave immediately a large excess in 0-110 μs time correlated events (fig. 4) for few minutes. Moreover, we observed at the scope at least 3 "triple events" (similar to fig. 4) occurring in a time window of 200 μs during the excess counts.



[Fig. 3] The triple event occurred in run 7 of fig.2.and acquired by digital scope. X-scale = 20 $\mu\text{s}/\text{div}$, Y-scale = 100 mV/div. The event occurred in correspondence of superconducting transition temperature, around 95 K.

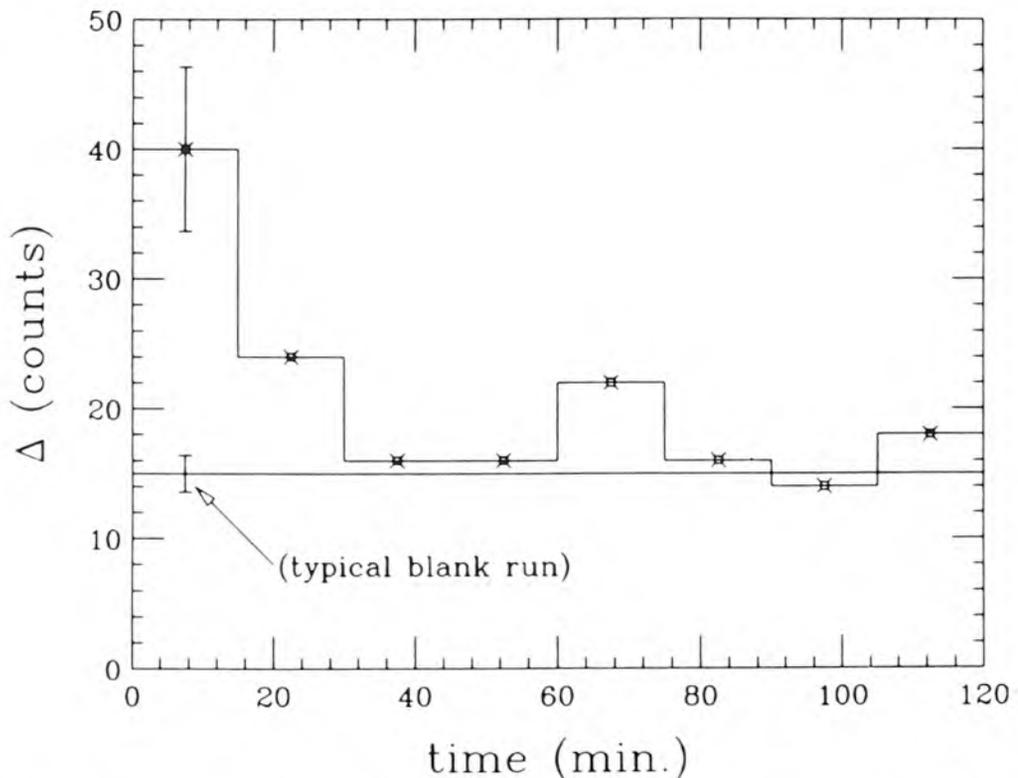
CONCLUSIONS

Taking in account that HTSC materials (only "high quality" and perovskite structure) as YBCO are able to absorb Deuterium, without destroying the crystalline structure, we put deuterated YBCO pellets in a neutron radiation field and we operated thermal cycles. In this double non-equilibrium condition we looked for a neutron rate enhancement selecting "time-correlated" events, burst-like.

In one thermal cycle we recorded a large increase, in respect to blank runs, of these events. Not all thermal cycle runs gave excess counts results although in some of them we visualized multiple neutrons (triple event) in a very short time-window. These multiple events were sporadic (typically during the superconductive temperature transition), although the probability that these events were simulated by the background was quite low.

In an other particular test, after a D₂ gas refilling at room temperature, we measured an increase of "time-correlated" events and we recorded some triple neutron events lasting to some minutes.

We have to conclude that HTSC materials can be used with a lot of interest in the Cold Fusion experiments and that non-equilibrium conditions are required to stimulate eventual neutron bursts. A full reproducibility of the phenomena detected until now is not under complete control.



[Fig. 4] Time correlated events occurred after long time D₂ low pressure loading and 10 hours high pressure refilling. The abscissa indicates the time elapsed from the initial irradiation. Several triple events were observed during this measurement.

ACKNOWLEDGEMENTS

We are indebted to Dr F.Ferrarotto and M.Corradi (Rome Un., Physics Dep.) for the time they spent with us during the experiment installation and data taking at G.Sasso Laboratory; moreover we thank Prof. B.Stella (Rome Un.) for his criticism on data interpretation.

We are grate for invaluable and skillful cooperation and technical assistance to F.Basti (INFN,Roma) and M.Giardoni (INFN,LNF).

We would like to thank prof. S.E.Jones (B.Y.U. Provo), prof. G.Preparata (Milan Un.) and prof. G.Bologna (Turin Un.) for their stimulating discussion, criticism and suggestion on nuclear aspects.

A special thank to prof. S.Pace (Salerno Un.) for stimulating discussions on superconducting materials.

This work is supported by grants from INFN and Italian Council of Research (CNR), "Comitato Nazionale per le Ricerche Tecnologiche e le Innovazioni".

REFERENCES

- [1] F.Celani et al.
S.E.Jones, F.Scaramuzzi, D.H.Worledge eds. "Anomalous Nuclear Effects in Deuterium/Solid System" AIP Conference Proceeding 228 pg. 62 (1991).
- [2] M.Rabinowitz, Modern Physics Letters B Vol. 4, No. 4 pg. 223 (1990).
- [3] G.Dattoli et al. preprint ENEA RT/89.49 Jul 6, (1989), Italy.
To be published by "Il Nuovo Cimento" (1991).
- [4] S.E.Jones et al.
S.E.Jones, F.Scaramuzzi, D.H.Worledge eds. "Anomalous Nuclear Effects in Deuterium/Solid System" AIP Conference Proceeding 228 pg.206 (1991).
- [5] M.V.Abrashev, L.N.Bozukov and M.N.Iliev. Physica C 178, 317 (1991).
- [6] J.J.Reilly et al., Phys. Rev. B 36, 5694 (1987).
- [7] H.Niki et al., Solid State Comm., vol.69, 547 (1989).
- [8] H.Hemmes et al. Phys. Rev. B 39, 4110 (1989).
- [9] T.Takabatake et al., Physica C 162-164, 9865 (1989).
- [10] E.Knittle and R.Jeanloz, Science vol. 251, pag. 1438, (1991).
- [11] S.E.Jones et al. Nature 338, 737 (1989).
- [12] C.Y. Yang et al., Phys. Rev. B 36, 8798 (1987).
- [13] H.Fujii et al., Jpn. J. Appl. Phys. 27, L525 (1988).
- [14] T.Kato et al., Jpn. J. Appl. Phys. 27, L564 (1988).
- [15] E.Kuzmann et al.
S.E.Jones, F.Scaramuzzi, D.H.Worledge eds. "Anomalous Nuclear Effects in Deuterium/Solid System" AIP Conference Proceeding 228 pg.262 (1991).
- [16] E.Kuzmann et al.
"MOSSBAUER SPECTROSCOPY CHARACTERIZATION OF SAMPLES FOR COLD FUSION EXPERIMENT" ACCF 2, June 29 - July 4, 1991 Como - Italy.
To be published as Conference Proceeding of "II Annual Conference of Cold Fusion" SIF, Bologna, (1991) Italy .

