

# SEARCH FOR ENHANCEMENT OF NEUTRON EMISSION FROM NEUTRON-IRRADIATED, DEUTERIDED, HIGH-TEMPERATURE SUPERCONDUCTORS IN A VERY LOW BACKGROUND ENVIRONMENT

COLD FUSION

TECHNICAL NOTE

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Following experiments performed with deuterided high-temperature superconductors (HTSCs) at the underground Gran Sasso Laboratory, the capacity of these materials to absorb deuterium and the role played by nonequilibrium conditions in neutron burst emissions in the framework of cold fusion have been determined.

Taking into account that HTSC materials such as  $Y_1Ba_2Cu_3O_{7.8}$  (YBCO) are able to absorb deuterium without destroying the crystalline structure, deuterided YBCO pellets were placed in a neutron radiation field, and thermal cycles were operated. In this double nonequilibrium condition, neutron rate enhancement was sought by selecting "time-correlated" burst-like events. The pellets and high-pressure  $D_2$  gas were enclosed in a stainless steel vessel, and thermal cycles (300 to 77 to 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, was set up.

One thermal cycle run showed a large increase (seven times more, corresponding to  $>30$  standard deviations) of time-correlated events with respect to the blanks. In another run, although no relevant mean value increase in events was detected, one interesting multiple (triple) neutron signal occurred at a temperature ( $\sim 95$  K) close to the transition from superconducting to the normal state. These multiple events were sporadic (detected twice during four thermal cycles lasting  $\sim 3$  h), although the probability that these events were simulated by the background was quite low (one incident expected in 80 h). Similar runs produced no relevant values.

Another experiment, at constant temperature (300 K), characterized by a heavy  $D_2$  gas refill, showed both some increase in time-correlated events and a few triple neutron signals.

## MOTIVATIONS

According to both our opinions,<sup>1</sup> based mainly on similarities between the behavior of hydrogen- or deuterium-doped palladium and  $Re_1Ba_2Cu_3O_7$  (RBCO) in the superconducting state, and some theoretical suggestions and considerations,<sup>2,3</sup>

we found some interest in studying compounds of deuterided rare earths. Moreover, our first tests were independently confirmed by Jones at Brigham Young University,<sup>4</sup> who operated with deuterided,  $Y_1Ba_2Cu_3O_{7.8}$  (YBCO) in thermal cycles and under mechanical stresses.

There are several similarities between the behavior of

heavily hydrogenated palladium<sup>1</sup> and heavily oxygenated  $\text{Re}_1\text{Ba}_2\text{Cu}_3\text{O}_{6.5+5}$  compounds (Re is any trivalent rare earth except yttrium):

1. As indicated in Ref. 1 and experimentally found in Ref. 5, the absorption of hydrogen in RBCO occurs only if the RBCO is a superconductor material; otherwise, the hydrogen degrades the RBCO, giving  $\text{CuO}$ ,  $\text{Cu}_2\text{O}$ , copper, and BaO as final products.

2. The RBCO is superconducting only when the oxygen content is  $>6.5$ . The critical temperature  $T_c$  depends strongly on oxygen content (e.g.,  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_7$ ,  $T_c = 92$  K;  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{6.55}$ ,  $T_c = 60$  K).

3. High-temperature superconductors (HTSCs) easily lose oxygen in excess of 6.5; i.e., they are intrinsically unstable.

4. It is possible to add hydrogen or deuterium (nobody has tried tritium) to superconductors, even increasing  $T_c$ , despite the initial content of oxygen (as long as it is  $>6.5$ ) (Ref. 6)

5. There is no clear evidence of the usual isotopic effect.

6. Several authors found the rare property of an inverse pressure coefficient in  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{7.3}$ .

7. Nuclear magnetic resonance measurements<sup>7</sup> show that hydrogen in  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{7.5}\text{H}_{0.2}$  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 heating trapped hydrogen could move. Similar effects can be expected with deuterium at similar temperatures.

8. There is a particular value of hydrogen doping that increases<sup>8</sup> the critical temperature. The particular value seems to depend on sample preparation and hydrogen loading procedure.<sup>8,9</sup>

Moreover, HTSCs have a perovskite structure, the same structure as the 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  $1.4 \times 10^6$  bar (Ref. 10). Thus, we can suppose that a proper perovskite structure can be efficient like the "Mother-Earth-Soup" used by Jones et al.<sup>11</sup> to search for geological nuclear fusion.

In other words, among the unusual properties of HTSCs, there is their ability to absorb a large amount of hydrogen.<sup>6,7,12-16</sup> The possible sites where the hydrogen or deuterium is located can be identified by Mossbauer spectroscopy, as recently by Kuzmann et al.<sup>15,16</sup>

These properties have led us to use high-quality HTSCs such as YBCO in a gaseous system, instead of the "standard" titanium or palladium, to research for anomalous cold fusion phenomena.

As reported in Ref. 1, where we performed measurements with deuterated HTSC material under neutron radiation, collecting all the events accumulated over a long time ( $>10$  min), we obtained indications (although with poor statistical significance) of enhanced neutron emission compared with the blank runs. To exhibit this effect, we developed circuitry that can detect neutron emissions occurring in short time window.

## APPARATUS AND ACQUISITION SYSTEM

The apparatus is shown in Fig. 1. It consisted of two  $^3\text{He}$  tubes surrounded by paraffin and lead bricks to detect

neutrons and a lead well where a weak (2200 n/s of  $\sim 4$  MeV and 1500  $\gamma$ /s of 4.43 MeV) Am-Be neutron source was located. The source was covered by 8 cm of water, in a plastic container to moderate the neutrons. We put six YBCO pellets (disk shaped,  $\sim 19$ -mm diameter, 4 to 6 mm thick, density of 5.5 to 5.9 g/cm<sup>3</sup>,  $\sim 50$  g total weight) in a polytetrafluoroethylene coil ( $\sim 1$  mH) enclosed in a cylindrical stainless steel vessel for high pressure gas.

The acquisition system (Fig. 1) was based on a digital counter and analog acquisition by a fast digital scope (TEK 2430) of the signals coming from the  $^3\text{He}$  detectors; moreover, the independent detector's signals were acquired by a multichannel analyzer (MCA). All neutron signals were independently counted by proper counters (scalers), some of which were gated. Each signal from the proper detector was charge amplified, shaped (6- to 8- $\mu\text{s}$  duration), and discriminated. From the discriminator, one output was straightforwardly counted by a scaler; a second output, through a specific circuitry that inhibits any other signal arriving within 10  $\mu\text{s}$ , was counted by another scaler. These last signals were used to get the time-correlated events. Each signal coming from any detector opened for a defined time window the gate to the proper scaler that counts all the other signals (except the first) arriving during this time window.

The counts in a time window came from the logic OR pulses of the two detectors. We used three different contiguous but separate time windows:

1. from 0 to 10  $\mu\text{s}$
2. from 10 to 110  $\mu\text{s}$
3. from 110 to 1110  $\mu\text{s}$ .

The 10- to 110- $\mu\text{s}$  time window was used as a 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 time window b to be the most significant because the expected 2.45-MeV neutrons due to deuteron-deuteron reactions, have an expected thermalization time of the order of a few tenths of a microsecond in our experimental setup.

## EXPERIMENTAL PROCEDURES AND MEASUREMENTS PERFORMED

The experimental procedure adopted to deuteride the HTSC material is summarized as follows:

1. We put the HTSC inside the coil in the stainless steel vessel and measured the inductance variation of the sample with temperature (300 to 77 to 300 K) with an alternating current magnetic field of  $\sim 1$  G at 1 kHz.
2. We filled the vessel with deuterium gas at a typical pressure of 35 bar at room temperature.
3. The vessel was heated to  $\sim 370$  K and held at this temperature for  $\sim 1$  h, after which the temperature was decreased to  $\sim 360$  K and held for 5 h.
4. We cooled the vessel from 360 to  $\sim 300$  K in few minutes.
5. We refilled the  $\text{D}_2$  gas to 36 bar.

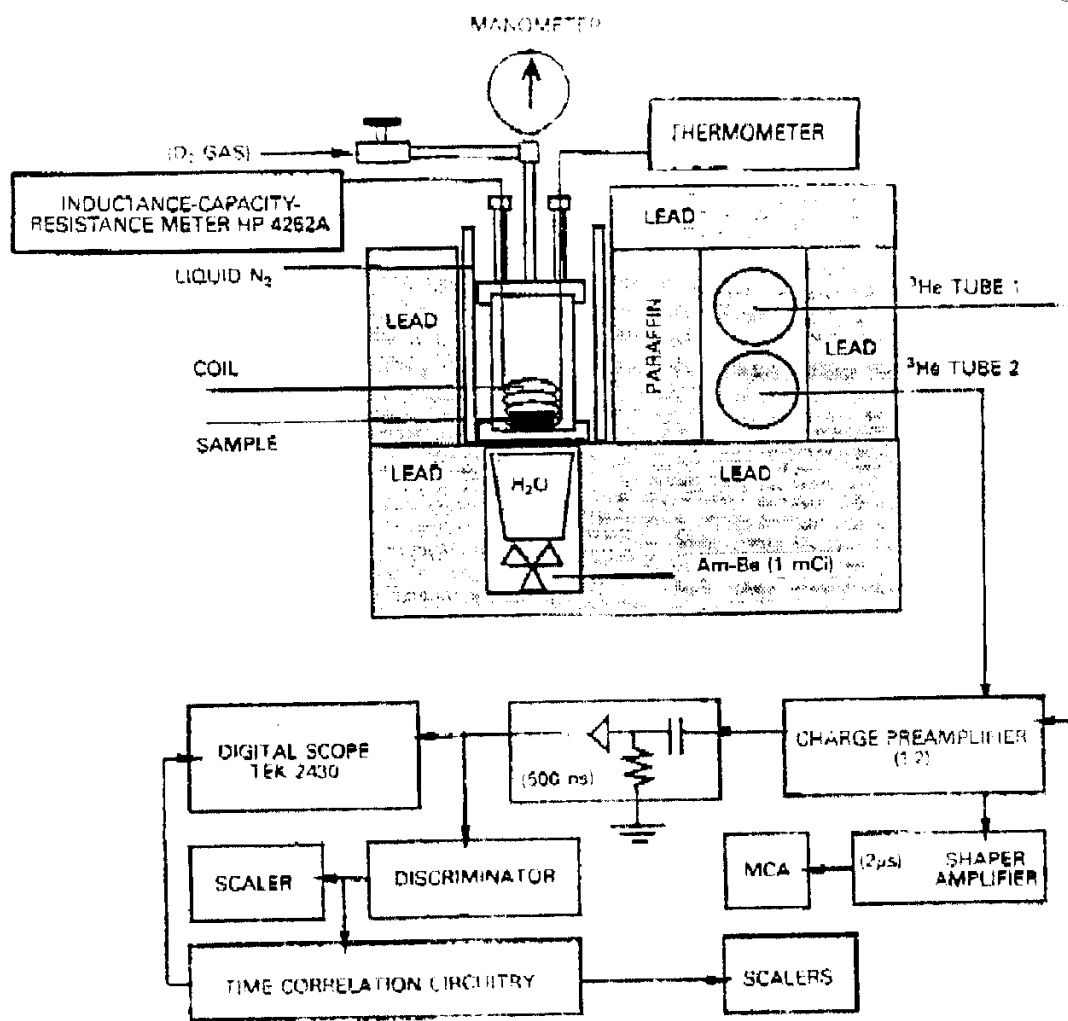


Fig. 1. Experimental apparatus and data acquisition system

6. We started the neutron detection, performing thermal cycles (300 to 77 to 300 K). To check for deuterium absorption, because of the low accuracy of the manometer, we measured the inductance value due to the diamagnetism of the sample at 77 K and the value of the superconducting transition temperature during the heating.

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

In these specific measurements, we further investigated 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 to induce further nonequilibrium states of the material.

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

1. background (A): measurement with neutron source but no vessel
2. blank (B): measurements with neutron source and vessel filled with HTSC samples in this sequence:

- a. no D<sub>2</sub>, T = 300 K (B1)
  - b. D<sub>2</sub> at 40 bar, T = 300 K (B2)
  - c. D<sub>2</sub> at 36 bar, T = 300 K after the deuteration procedure (B3)
3. thermal cycle (C): measurements after the deuteration procedure with neutron source, vessel filled with D<sub>2</sub> at 36 bar (300 K), and thermal cycles (300 to 77 to 300 K).

## RESULTS

As shown in Fig. 2, the different tests were performed in sequential independent runs. Typical background (1, 2, 3, and 10) and blank (3, 4, and 5) runs ranged from 0.5 to 10 h of acquisition time, while the thermal cycle runs (7, 8, 9, and 11) ranged from 0.5 to 1 h. As can be seen from Fig. 2, no significant statistical differences result from the background and blank runs (we adopted Gaussian statistics for error calculation).

The thermal cycle runs showed different cases: Runs 8 and 11 had no significant statistical differences with respect

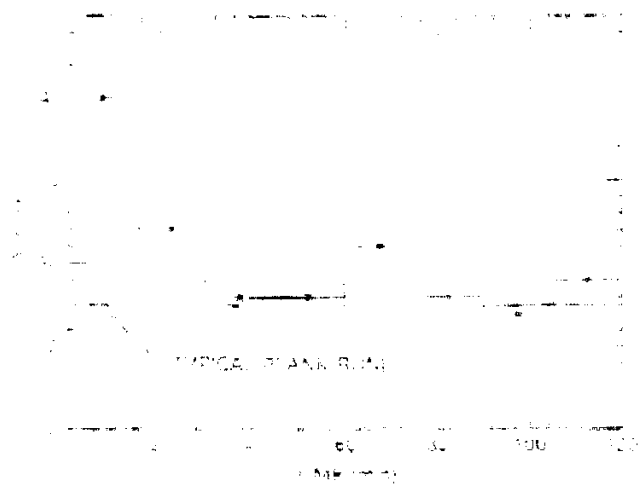


Fig. 4. Typical blank runs showing triple events.

was observed after 2.4 h. One other triple event occurred in another identical cycle (three cycles of 150 to 200 K below the expected transition temperature).

There is a large excess of time-correlated events recorded and it is about seven times more with respect to the blank run and corresponding to  $>3\sigma$  standard deviations. Most of these events were observed at the scope, and they looked like two neutrons at 10 to 110  $\mu$ s; no spurious signals were observed. The excess was recorded, with different relative intensity, as in 10 to 110  $\mu$ s as in 110 to 1110  $\mu$ s time window (see Fig. 3).

As shown in Fig. 3, we note that in run 9 the ratio between the 10 to 110- $\mu$ s time window and the 110 to 1110- $\mu$ s time window is about four times larger than the blank runs. This may be consistent with several neutron bursts (separated in time by  $>1$  ms) having burst intrinsic time duration  $\approx 110$   $\mu$ s. We recall that the expected thermalization time of 2.45-MeV neutrons is in the 10- to 110- $\mu$ s time window. It is quite improbable that some persistent disturbances can reproduce this kind of behavior.

In another test performed at room temperature (1 month later, at  $\approx 20$  bar  $O_2$  gas pressure), we increased the pressure to 42 bar, and after  $\approx 10$  h, we put the vessel into the source well. The measurement, starting a few seconds later, immediately gave a large excess in 0- to 110- $\mu$ s time-correlated events (Fig. 4) for a few minutes. Moreover, we observed at the scope at least three triple events (similar to Fig. 4) occurring in a 200- $\mu$ s time window during the excess counts.

## CONCLUSIONS

The tests performed show that neutron production from accelerated YBCO pellets can be real. The conditions to expect an effect, as we have shown, can be different, but they are different from equilibrium state. These states can be caused by external mechanical and/or thermal or mechanical stresses. Because this effect also occurred during the phase transition from superconducting to the normal state, we suppose that the superconducting phase transition can stimulate the effect.

Since the transition of HTSC materials can be used in different experiments and that nonequilibrium conditions can lead to anomalous eventual neutron bursts,

The full reproducibility of the phenomena detected is not yet under complete control.

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