Search for superheavy hydrogen in sea water

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Abstract: We report the first results of an experiment designed to search for superheavy hydrogen atoms, which started in Paris about two years ago. This search is based on the centrifugation of water, followed by atomic spectroscopy. It is sensitive to large masses from 10^4 GeV/c^2 up to 10^8 GeV/c^2, not accessible to accelerator physics or mass spectrometers.

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We report the first results of an experiment designed to search for superheavy isotopes of hydrogen, which started in Paris about two years ago. This search is based on the centrifugation of water (mostly sea water), followed by atomic spectroscopy [1]. It is sensitive to large masses from 10^4 GeV/c^2 up to 10^8 GeV/c^2 at least, not directly accessible to accelerator physics or to mass spectrometers, presently limited to about 10^4 GeV/c^2. At the present stage, still preliminary, this experiment provides an upper limit of about 10^{-13} for the relative abundance of superheavy hydrogen in sea water. While this is still above the possible limit ($< 10^{-15}$) given by P. Smith et al. from enriched D_20 density measurements [2], the present experiment relies largely on sea and ocean waters, in which superheavy hydrogen atoms could conceivably have accumulated, and be more abundant than in terrestrial waters.

Together with other arguments [3], this result may be used to test, and virtually eliminate, a scenario of charged dark matter recently proposed by A. de Rujula, S. Glashow and U. Sarid [4]. A significant improvement of the sensitivity is still expected by using centrifuged heavy water rather than ordinary water, and eliminating, in the atomic physics experiment, remaining backgrounds induced by ordinary hydrogen. For masses larger than about 10^9 GeV/c^2 the fate of superheavy hydrogen atoms - which would fall down to the bottom of the oceans [1], then get trapped inside sediments on the ocean floor to possibly reappear on land in sedimentary grounds - is still unclear, so that no reliable information on such extremely heavy particles may be given yet.

1. Motivations

But, at first, why should one consider the possibility of superheavy hydrogen isotopes? Particle physics theories, especially those which aim at the unification of the fundamental interactions, usually lead one to postulate the existence of a number of new particles. Most of them are predicted to be highly unstable, such as the $W^{\pm}$ and Z bosons of the electroweak unification discovered at CERN, or the much sought-after top quark or Higgs bosons. Some of them, however, might well be absolutely stable, their stability being guaranteed by the conservation of one or several new quantum numbers, other than the spin S, the electrical charge Q, and the baryonic and leptonic numbers, B and L. The conservation of the electrical charge guarantees the absolute stability of the electron. The - at least approximate - conservation of the baryonic number B ensures the stability of the proton (at least to a very good approximation, since no proton decay has been observed yet). As is well known the conservation of a new quantum number could lead to new, possibly charged, stable particles (for example new heavy leptons, or quarks), subsequently leading to heavy anomalous hydrogen-like atoms. Those have been searched for by P. Smith and his collaborators [2,5], who established very strong upper limits on their possible abundance in terrestrial waters ($\leq 3.\ 10^{-20}$), for masses smaller than 10^4 GeV/c^2.

What could be the motivations for considering such particles, and what masses could they have? Among various possible motivations we are particularly attracted by supersymmetric theories, which may well constitute a necessary step towards the unification of the fundamental interactions, including gravity. All presently known particles should then be associated with new superpartners, whose spin differ by 1/2 unit [6]. A multiplicatively conserved quantum number called R-parity distinguishes between ordinary particles, with $R_p = +1$, and new superpartners, which have $R_p = -1$. This definition is equivalent to

$$\text{R-parity} = (-1)^{2S}\ (-1)^{(3B+L)}$$

which relates the conservation of R-parity with those of baryonic and leptonic numbers. (The conservation of R-parity remains valid even if B and L are separately violated, as in grand-unified theories, as long as the difference B-L remains conserved, even only modulo 2.)

The lightest superpartner, with R-parity (-1), is then stable and may be neutral, or charged. In that case it could be, for example, a spin-0 lepton $\tilde{l}^{\pm}$, a spin -1/2 wino $\tilde{w}^{\pm}$, or a charge 2/3 spin-0 top quark $\tilde{t}$. Actually the latter possibility appears favored if the top quark, as it seems, turns out to be rather heavy - i.e. about 100 to 200 GeV/c^2 -, with the ordering of masses being reversed when going from ordinary particles to superpartners [7]. As discussed in [1] $(\tilde{t}ud)^+$ and / or $(\tilde{t}\bar{u})^\circ$ -proton, for example, could then be stable, and combine with electrons to form superheavy hydrogen isotopes.

These particles are usually expected to have masses of the order of m_W, i.e. $\sim$ 100 GeV/c^2 or so. Their masses, however, may well be considerably higher if the mass gap associated with supersymmetry breaking can be suitably decoupled from the electroweak scale $\sim m_W$. It is interesting to note, in that respect, that in higher-dimensional supersymmetric theories one may relate translations along an extra compact dimension, of size L, with R-symmetry transformations, so that [8] :

$$m\ (\text{superpartner}) \sim \hbar / Lc$$

This mass scale, fixed by the compactification scale (or one of them if there are several), could be as "low" as ~1 TeV/c^2 or even less; or, conversely, as high as the Planck scale $m_P \approx 10^{19}$ GeV/c^2, in the vicinity of which all four fundamental interactions might get unified; or also, be somewhere in the vast region in between...

Even in the absence of supersymmetry, the various continuous or discrete symmetries of higher dimensional theories are still expected to lead us to one or several new stable particles at the compactification scale(s) $\sim \hbar / Lc$. The mass interval below 10^4 GeV/c^2 has already been well explored experimentally, at least for charged stable particles [2]. But the whole domain between 10^4 and 10^8 - 10^9 GeV/c^2 , in which our experiment is sensitive to superheavy hydrogen, remains a promising field of investigations, which might reveal signs of the existence of supersymmetry, extra dimensions, or new interactions...

Stable particles should be present in the Universe as relics from the big bang. Under the optimistic assumption that one understands well enough the evolution of the early Universe, including the generation of the baryon asymmetry, one can give crude estimates of the abundances of relic heavy particles in the whole Universe, relatively to nucleons. The abundance n_X is essentially proportional to ($\sigma_{\text{annihilation}}\, m_X$) $^{-1}$, with $\sigma_{\text{annihilation}}$ varying like $1/m_X^2$ (see e.g. Ref.[9] and references therein) With $\Omega_b = \rho_{\text{baryonic}} / \rho_{\text{critical}} \sim 10^{-2}$ to 10^{-1}, this leads to the rough order of magnitude estimates:

$$\frac{n_X}{n_B} \sim (10^{-8} - 10^{-10})\ m_X\ (\mathrm{GeV/c^2})$$

for stable particles which interact strongly ($\sim \alpha^2{}_s$ or very strongly (~ 1)

$$\frac{n_X}{n_B} \sim (10^{-5} - 10^{-6})\ m_X\ (\mathrm{GeV/c^2})$$

for stable particles which interact electroweakly ($\sim \alpha^2$)

The constraints from the total energy density of the Universe ($\Omega = \rho/\rho_c \leq 1$, with Ω baryonic $\sim 10^{-2}$ to 10^{-1}) should then in principle require, in a very crude approximation, $m_X \leq 10^{(3 \text{ or } 4)}$, 10^5 or 10^6 GeV/c^2 at most, for particles which interact only electroweakly, strongly or very strongly (case of a new color-like interaction with a very large energy scale $\sim m_X$).

According to the above formulae, for $m_X \geq 10^4$ GeV/c^2 the new particle contribution to the energy density of the Universe, relatively to baryons, would be very significant or even dominant : i.e. $\geq 10^{-2}$ at least, up to about 10^2 at most, corresponding to $\Omega \sim 1$. (Indeed in the latter case the Universe might be closed by heavy dark matter particles of masses $\sim 10^{(3 \text{ or } 4)}$, 10^5 or 10^6 GeV/c^2, depending on whether they have electroweak, strong or very strong interactions). Given these values, and the low limits already available for the abundance of superheavy hydrogen in terrestrial water, the existence of such atoms seems somewhat unlikely. The various uncertainties which weaken the above arguments, however, justify an experimental search for these particles. In particular, we have noted in ref. [1] that the above "standard" abundance estimates could be significantly reduced if the relics having survived annihilation (if sufficiently heavy) were diluted by an inflation phase, prior to the generation of the baryon asymetry. Moreover, two years after the beginning of this search, De Rujula, Glashow and Sarid (in the framework of the above conventional abundance estimates), discussed a scenario according to which heavy charged stable particles ("champs") might be dark matter candidates and close the Universe [4]. Their expected abundance on Earth, difficult to evaluate in a reliable way, was estimated by these authors to be $\sim 10^{-7}$, at least, in mass, corresponding to abundances $\geq 10^{-11}$ - 10^{-13} for masses $\sim 10^4$ - 10^6 GeV/c^2. This is well within the range of sensitivity of our search for superheavy hydrogen in ocean and terrestrial waters, and motivated us to present here the first results of this search.

2. Preparative centrifugation of water:

In a previous paper [1] , we described the basic principles of a method that should lead to concentrate superheavy water molecules, possibly contained in water, by ultracentrifugation. This method involves the centrifugation, in a swinging-bucket rotor, of water samples layered over a small volume of a 20% (w/v) solution of sucrose (pure sugar) in water. The superheavy molecules that might have sedimented into the sucrose layer during the centrifugation would be prevented from diffusing back into the overlay of water by the high viscosity of the sucrose solution. Though this method is routinely used by biologists to purify macromolecules, it seemed important to check that the viscosity of the sucrose solution also suffices to trap small water molecules. This was verified as follows: in a polycarbonate tube identical to those used for the centrifugation experiments (see below), held vertical in a tube holder maintained at room temperature, 20 ml of distilled water were introduced first. 4.5 ml of a 10% sucrose solution in water were then slowly (1 ml/minute) deposited under the water layer, by means of a capillary tube introduced through the water down to the bottom of the tube and connected through a plastic tubing to a Minipuls 2 (Gilson) peristaltic pump. 0.5ml of a water solution containing 20% sucrose and tritiated water ($5x10^6$ dpm total) were then deposited in the same way under the 10% sucrose layer. The capillary tube was gently removed, and the filled centrifugation tube was left for one hour at room temperature on the bench to allow for the diffusion of the tritiated water molecules, which should be the same as that of putative superheavy water molecules concentrated in the lower 0.5 ml of the centrifugation tube. The capillary tube was gently introduced again into the tube, and the lower 5 ml of liquid were slowly (1 ml/mn) pumped out, collected in a test tube, and diluted with water to a total of 20 ml, the same volume as the water overlay. Samples of 0.5 ml of the overlay remaining in the tube, of the diluted lower layer, and of pure distilled water were then introduced in vials containing 10 ml of a water compatible scintillation cocktail (Ready Safe - Beckman) and the tritium radioactivity of each sample was counted during 5 minutes in a Tri-Carb 4530 scintillation counter (Packard). While the water blank showed 60 counts per minute (cpm), the sucrose layer showed 22,004 cpm, and the water overlay only 460 cpm. Since the total volumes of the overlay and of the diluted sucrose were the same, it can be concluded that only 1.8% of the tritiated water initialy contained in the 20% sucrose layer were, because of diffusion or mixing during the introduction and removal of the samples, recovered in the overlay. 98.2% stayed in the sucrose solution. Thus, one can estimate at 1.8% the loss of superheavy water molecules at each centrifugation.

Samples of water from 6 different origins were first submitted to successive centrifugations in the three buckets of a SW 23.5 rotor (3x70 ml; maximum speed 23,500 rpm) of a Superspeed 65 ultracentrifuge (MSE - Crawley - England). The rotor speed was 21,000 rpm; the centrifugation time was always between 14 and 18 hours; the temperature was 15 °C. In each of the three tubes (65 ml; polycarbonate), 60 ml of water (diluted when needed with an equal volume of pure water to ascertain that the relative viscosity of the solution was below 1.1) were introduced. This will hereafter be refered to as the "overlay" solution.Then, 5 ml of a 20% sucrose solution in the same water (the "underlay") were gently pumped in, as described above, below the overlay. The time of injection of the underlay was 5 minutes per tube. The sucrose solution also contained a colored dye (bromophenol blue) to help in visualizing the interface between the two layers, and thus check on the absence of mixing artefacts and excess diffusion of the sucrose. The three tubes were submitted to centrifugation as described above. The centrifugation was terminated by letting the rotor slow down with the brake off, which took about 25-30 minutes. The lower 5 ml contained in each tube were then collected (in 5 minutes per tube) and the 60 ml of overlay were discarded. New 60 ml samples of water of the same origin were put into three tubes, and 5 ml of the sucrose solution recovered from the previous run were again carefully pumped to the bottom of each tube, under the new water layer. The centrifugation and collection of the lower layer were repeated as above, and so on. Since the centrifugation field was too low to significantly concentrate the dye and sucrose in the bottom of the tube, significant diffusion of these molecules took place during the successive centrifugation runs. To keep the underlay viscosity high enough, the 3x5 ml underlays collected after 3-4 runs were pooled, diluted to 180 ml with the water sample under study, and used again as overlay for further centrifugations. The last 3x5 ml of sucrose underlay thus obtained when the initial water sample was exhausted were pooled and the resulting 15 ml were kept frozen at -32 °C until further use.

When each of the 6 initial water samples had been "concentrated" to 15 ml, the 6 sucrose solutions were pooled, diluted two fold with 90 ml of distilled water, and submitted to a centrifugation in 3 tubes, over 5 ml underlays of 20% sucrose as above. To take care of the increased viscosity of the overlay (10% sucrose), the centrifugation was carried out at 23,000 rpm during 20 hours. The 5 ml lower layers in each tube were collected, pooled, diluted to 60 ml with distilled water, and introduced into one centrifugation tube. 5 ml of 20% sucrose were underlayed, and a centrifugation was run at 15,000 rpm during 42 hours . The 5 ml of underlay were pumped out, diluted with 13 ml of distilled water, and introduced in a centrifugation tube of a SW 30 swinging bucket rotor (MSE). 1 ml of 20% sucrose in distilled water were very slowly (0.2 ml/mn) pumped into the tube, under the overlay. Centrifugation was achieved in the SW 30 swinging bucket rotor spinning at 24,000 rpm for 18 hours in the Superspeed 65 preparative ultracentrifuge. At the end of the centrifugation, the lower 1 ml was pumped out, diluted with 3 ml of distilled water and introduced into a 4.4 ml centrifugation tube of a SW 60.Ti swinging bucket rotor (Beckman). 0.15 ml of 20% sucrose were pumped in under the overlay, and centrifugation was performed in the SW60.Ti swinging bucket rotor, for 5 hours at 40,000 rpm in a L8-70M (Beckman) preparative ultracentrifuge. At the end of the centrifugation, the lower 0.15 ml was pumped out, and kept frozen at -32 °C.

The origin of the six water samples was as follows:

1-snow was collected in the French Alps (Les Arcs) in January 1987, and melted (1260 ml). It has then been concentrated with 7 successive centrifugation steps.

2-water from the Indian Ocean was collected in February 1984 near the Kerguelen Islands, and stored for 3 years in an immobile tank. Water from the bottom of the tank was carefully collected (720ml). It has then been concentrated with 9 successive centrifugation steps (720 ml of distilled water were added before the first centrifugation to reduce the viscosity).

3-water from rest of the tank above (Indian Ocean - Kerguelen Islands) was randomly collected (2430 ml). 28 centrifugation steps were needed (2430 ml of ordinary water were added before the first centrifugation).

4-water from the Dead Sea was collected near the north-western shore (615 ml). 620 ml of ordinary water were added and 7 centrifugation steps were needed.

5-surface water from the Indian Ocean, near the Comores Islands (12° 5' S; 45° 10' E) was collected at 6 m below the surface (870 ml). 5 successive centrifugation steps were needed.

6-deep sea water from the Mediterranean (40° N ; 6° 30' E) was collected at a depth of 2800 m, 2 m above the sea bottom (4170 ml). 25 successive centrifugation steps were used.

The probability of loosing a superheavy water molecule from the underlays in the centrifugation steps used to concentrate these samples was estimated as follows: we first estimated the probability of loosing the molecule during the n successive centrifugations required to concentrate the initial sample to 15 ml. If a molecule was introduced in the n_ith overlay, it will undergo $n-n_i+1$ runs and therefore have a $(n-n_i+1)\times 1.8\%$ chances of being lost (1.8% corresponds to the loss per run estimated in the diffusion experiment described above). The average chance of loss during this series of n runs will thus be, at most:

$$P_1 = n^{-1}\Sigma_i(n-n_i+1)\times 1.8\% = 1/2 \times (n+1)\times 1.8\%$$

The superheavy water molecules that would have been effectively recovered in the 15 ml of concentrated sample have to undergo 4 other successive centrifugations, two of which were performed in tubes identical to those used for the diffusion experiment reported above. The two last centrifugations were performed in smaller tubes but, because of the geometry of the tubes, the thickness of the underlay was practically the same. It therefore can be inferred that the probability of loss by diffusion was the same in the two last runs as in the previous ones. Hence, during the last four runs, the chances of loss of superheavy water can be estimated to:

$$P_2 = 4 \times 1.8\%$$

Finally, the total probability of loss is, at most:

$$P_1+P_2 = 1/2 \times (n+9)\times 1.8\%$$

From this and from the number of runs indicated above, it can be concluded that there is an average 30% chance of loosing the superheavy water molecules during the centrifugation process, and then that molecules initially contained in a total of 8.8 liters of sea water (finally reduced to .15 ml) have effectively been concentrated 4 x 10^4 fold. If one also considers the fresh water (molten snow, or distilled water used for dilutions) the total initial volume was about 12.4 liters and the concentration factor 6 x 10^4.

3. Laser spectroscopy

The laser spectroscopy experiment is slightly different from the experiment described in our proposal [1]. Instead of using a stepwise excitation to bring the hydrogen atoms from the n=1 to the n=3 level, we perform a direct two-photon absorption (fig.1). The details of the experimental set-up have been described previously [10] and we just recall here the essential features. The hydrogen gas obtained by reduction of the centrifuged sea water is enclosed in a quartz cell. The pressure of hydrogen is of the order of 0.1 Torr. Hydrogen molecules are dissociated by a pulsed r.f.discharge. In the afterglow, a powerful ($P \sim 0.1$ MW) narrowband ($\Delta\nu \sim 100$ MHz) pulsed ($\tau = 10$ ns, repetition rate 10 Hz) light source tuned around 205 nm excites the hydrogen atoms from the $1S_{1/2}$ ground state to the $3S_{1/2}$, $3D_{3/2}$ and $3D_{5/2}$ sublevels of the second excited state by a direct two-photon absorption. The excited atoms are detected by monitoring the fluorescence on the Balmer α transition due to the spontaneous emission from the n=3 level to the n=2 level. By reflecting the incident beam into the cell, we can obtain a Doppler-free two-photon spectrum of hydrogen or deuterium (fig.2). The typical width of the lines obtained in these conditions is of the order of 300 MHz which is much smaller than the Doppler width of the transition (30 GHz for hydrogen and 20 GHz for deuterium).

Because of the large mass shifts of the superheavy hydrogen (the energy levels of atomic hydrogen are proportional to the reduced mass of the electron), its excitation energy differs from that of deuterium by 26 cm^{-1} and 52 cm^{-1} from that of ordinary hydrogen(isotopic mass shift). We have tuned our light source in the range of frequency corresponding to the excitation of this exotic hydrogen atom. To find the correct wavelength we have used a home-made lambdameter. The exactitude of the wavelength is confirmed by scanning the iodine absorption .

To excite the hypothetical superheavy hydrogen atoms we have used standing wave and travelling wave excitations. The travelling wave excitation is particularly interesting since the Doppler width of the superheavy hydrogen atoms is smaller than the instrumental width for the range of mass considered here. The instrumental width comes from the laser linewidth and from collisional broadening and can be estimated from the Doppler-free spectrum of deuterium shown in fig.2 . In a travelling wave three lines separated by the fine structure of the n=3 level and having a width of the order of 300 MHz would thus constitute an unambiguous identification of the presence of superheavy hydrogen since any impurity would give rise to an absorption spectrum having a Doppler width of the order of a few GHz.

When we scan the light frequency we obtain on the detector an almost constant background as shown in fig.3. We have identified the origin of this background as due to the fluorescence of the quartz cell. This fluorescence has a very long lifetime ($\sim$ 300 ns), much longer than the fluorescence of the hydrogen atom ($\leq$ 30 ns). Thus we can have a fair estimate of the atomic fluorescence by subtracting the signals recorded in two separate windows.The first window is opened just after the light pulse and lasts 75 ns. The signal in this window contains both the noise and the hypothetical fluorescence of the exotic atom. A second window which is opened 75 ns after the light pulse and lasts 75 ns contains only the noise. It is the difference between the signals recorded in these two windows and averaged over 16 runs, compared for deuterium and for superheavy hydrogen, which gives the upper limit on the relative abundance of superheavy hydrogen atoms, taking into account the efficiencies of excitation (Doppler-free compared to travelling wave excitations). More precisely a numerical analysis of the signals shows that the abundance of superheavy hydrogen atoms compared to ordinary hydrogen is less than $4\ 10^{-9}$.

Taking into account the enrichment factor of $4\ 10^4$ due to the centrifugation process, we end up with a still preliminary limit for the relative abundance of superheavy hydrogen from 10^4 up to 10^8 atomic mass units in sea water of 10^{-13}.

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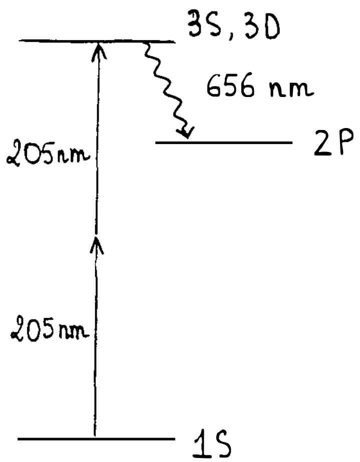

figure 1: two photon excitation scheme

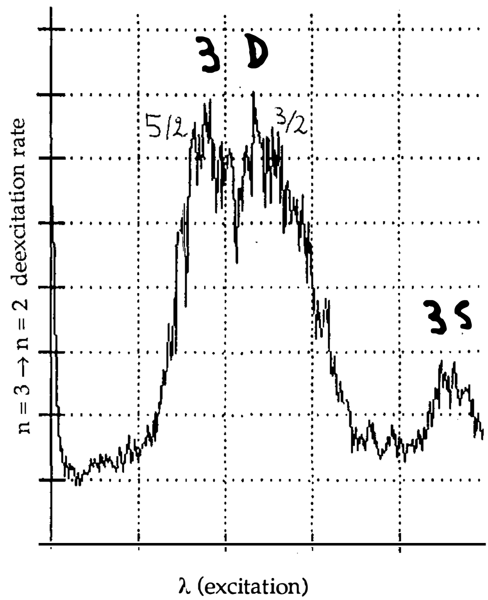

figure 2: excitation curve in the deuterium region

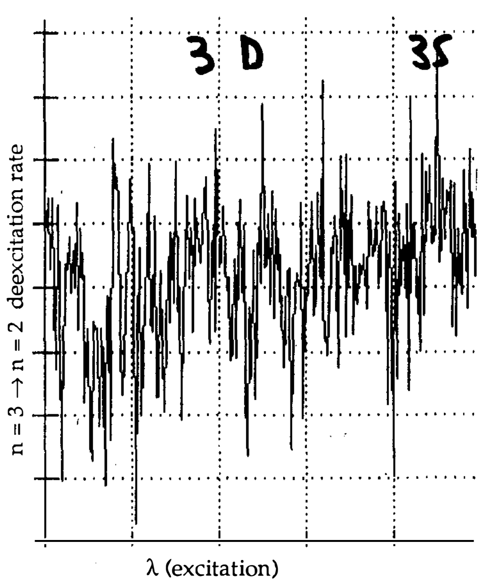

figure 3: excitation curve in the region where the presence of superheavy hydrogen is expected