Search for Superheavy Hydrogen in Sea Water

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We report the results of an experiment designed to search for superheavy isotopes of hydrogen. Based on the centrifugation of sea water, followed by atomic spectroscopy, it is sensitive to large masses from 10^4 up to 10^8 GeV/c^2. The relative abundance of such hypothetical atoms, compared to ordinary hydrogen, is found to be less than about 6×10^{-15}. This may be used to exclude charged dark matter particles between 10^4 and 10^7 GeV/c^2.

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Particle physics theories, especially those which aim at the unification of the fundamental interactions, usually lead one to postulate the existence of new particles. While most of them are predicted to be highly unstable, some might be absolutely stable, as a result of the conservation of new quantum numbers. Among possible motivations for considering such particles, we are particularly attracted by supersymmetric theories, which may well constitute a necessary step towards the unification of the fundamental interactions. All presently known particles should then be associated with new superpartners, whose spins differ by $\frac{1}{2}$ unit [1]. A new quantum number called *R parity*, equal to $(-1)^{2S}(-1)^{3B+L}$, distinguishes between ordinary particles, with $R_p=+1$, and new superpartners, which have $R_p=-1$. R parity (which follows from the conservation of baryon number B and lepton number L, or only of $B-L$, even modulo 2) is a natural invariance in supersymmetric theories. The lightest superpartner in an R-parity-conserving theory should remain stable, and may be neutral or charged. In that case it could be, for example, a spin-0 lepton, $\tilde{l}^{\pm}$, a spin-$\frac{1}{2}$ W-ino, $\tilde{W}^{\pm}$, or a charge $-\frac{2}{3}$ spin-0 top quark, $\tilde{t}$ [2]. As discussed in Ref. [3], a $(\tilde{t}ud)^+$ and/or $(\tilde{t}\bar{u})^0$ proton could then be stable. Such particles, which would combine with electrons to form superheavy hydrogen isotopes, might be very heavy, especially in higher-dimensional theories, in which the scale of supersymmetry breaking may be fixed by the compactification scale $\hbar/Lc$ (L denoting the size of an extra dimension) [4]. 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. In brief, new conservation laws could lead to new stable charged particles and, subsequently, to heavy anomalous hydrogenlike atoms.

Smith and collaborators have already established very strong upper limits on their possible abundance in terrestrial waters ($\lesssim10^{-28}$–10^{-29} up to 3×10^{-20}), for masses between 10 and 10^4 GeV/c^2 [5]. Above the latter value there exists only a possible upper limit, smaller than 10^{-15}, obtained from enriched D_2O density measurements. In contrast, our experiment relies largely on sea water, in which superheavy hydrogen atoms could have accumulated and, owing to their greater difficulty to evaporate, be more abundant than in terrestrial waters.

Stable particles should be present in the Universe as relics from the big bang [6]. Standard abundance estimates, however, in principle, exclude the mass range above 10^6 GeV/c^2, and strongly disfavor the one between 10^4 and 10^6 GeV/c^2: In the latter range one would have to trust that such heavy particles might exist relatively abundantly in the Universe (i.e., between about 10^{-6} and 1 in abundance, relative to nucleons) while having escaped detection. This made their existence rather improbable. Nevertheless, in view of all the uncertainties inherent to abundance estimates (with, in particular, the possibility of dilution due to inflation), we decided to go ahead with the experiment in 1987 [3]. Moreover two years later De Rújula, Glashow, and Sarid [7] revived the idea that dark matter in the Universe might be made of charged (very) massive particles like the ones we were looking for, rather than of neutral particles; this scenario, however, is plagued with difficulties, although the general idea has not been totally ruled out [8–10]. Strongly interacting dark matter [11] could also lead to superheavy hydrogen atoms.

Our experiment is sensitive to superheavy hydrogen in the mass range 10^4 to about 10^8 GeV/c^2 [3]. The lower figure is due to the efficiency of the centrifugation process used to concentrate superheavy atoms possibly contained in water, which gets too slow if the masses are not large enough. The upper figure has two different origins. First the atomic physics experiment probes atoms in the central region of a ~1-cm-diam cell; because of the Boltzmann factor, the mean density in this region becomes significantly ($\geq15\%$) smaller than the average density for atoms heavier than 6×10^7 GeV/c^2. Second, this figure simply expresses that for larger masses superheavy particles would tend to sink to the bottom of the ocean, thereby decreasing the significance of the experimental limit. The fall velocity of superheavy hydrogen in still water (at 4 °C), (20 m yr^{-1})$[M/(10^8\ \mathrm{GeV}/c^2)]$, is derived from the measurement of the diffusion coefficient of heavy water in water, $D \cong 1.4\times10^{-5}$ cm^{2}s^{-1} ($v_{\rm fall} \cong MgD/kT$). If the fall velocity is smaller than the upwelling rate of the ocean in the equatorial zones (around 13 m yr^{-1} [12]), the sedimentation effect can be neglected [13], so that, for particles lighter than 6×10^7 GeV/c^2, currents keep the ocean well mixed.

We already described elsewhere [3,14] the basic principles of a method which should lead to increased concentrations of superheavy water molecules possibly contained in water. It involves the centrifugation of water samples layered over a small volume of 20% (weight/volume) of sucrose solution in water. The superheavy molecules that would 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, we checked that the viscosity of the sucrose solution also suffices to trap small water molecules. A preliminary experiment [14] done with tritiated water showed that 98.2% of the radioactive element initially present in the sucrose solution remained in this solution. Because of the almost perfect efficiency of the centrifugation for sufficiently long times and masses larger than 10^4 GeV/c^2, we can estimate the loss of superheavy water molecules to be less than 1.8% at each centrifugation.

Six samples of water from different origins (Indian Ocean near the Kerguelen Islands, Indian Ocean near the Comoro Islands, deep sea water from the Mediterranean collected at a depth of 2800 m, 2 m above the sea bottom, Dead Sea, and snow from the French Alps) were submitted to successive centrifugations and ultimately reduced to 0.15 ml (see Ref. [14] for details). From the exact sequence of runs undergone by each water sample, and from the probability of loss at each centrifugation, one ends up with a concentration factor of at least 0.45×10^5 for the 8.8 l of sea water, and of at least 0.7×10^5 for the total initial volume of about 12.4 l (including the snow and the distilled water used for dilutions).

The water obtained by ultracentrifugation is then reduced in a uranium oven to obtain hydrogen gas. The mass of this gas practically coincides with the mass of hydrogen in our water sample, which indicates that almost all the water is reduced. This hydrogen gas is used to fill the experimental cells, consisting of Pyrex tubes with glued quartz windows, connected to a pumping line. The hydrogen molecules at a pressure of about 0.1 Torr are dissociated by a pulsed radio-frequency discharge. In the afterglow, a powerful ($P \approx 0.1$ MW), narrow-band ($\Delta\nu \approx 100$ MHz), pulsed (pulse duration 10 ns, repetition rate 10 Hz) light source tuned around 205 nm excites the hydrogen atoms from the ground state to the $n=3$ excited state by a two-photon transition [Fig. 1(a)]. The light source comes from an amplified continuous pulsed laser working at 615 nm whose output is frequency tripled by two successive nonlinear crystals (potassium dihydrogen phosphate and beta barium borate) [15]. The atoms excited in the $n=3$ level are detected by monitoring the fluorescence on the Balmer α transition due to the spontaneous emission from the $n=3$ to the $n=2$ level (an interference filter centered at $\lambda=656$ nm having a resolution of $\Delta\lambda=2.5$ nm full width at half maximum is placed in front of the photomultiplier). Because of the isotropic mass shift, the excitation energy for ordinary hydrogen differs from that of a hypothetical superheavy hydrogen by 53 cm^{-1}, which is large compared to the Doppler width of the transition (≈ 1 cm^{-1} for hydrogen).

To calibrate the signals, we studied the fluorescence when the light source is scanned around the deuterium resonance frequency. The experiment was done first with a standing wave (obtained by reflecting the incident wave back on itself with a mirror). In such a case, one can obtain a Doppler-free spectrum [16]. An example of recording is shown in Fig. 1(b). One observes the two fine-structure components of the $1S$-$3D$ two-photon transition, corresponding to the $3D_{3/2}$ and $3D_{5/2}$ final states [17]. The theoretical curve, including the effects of hyperfine structure and collisional broadening, and obtained for a full width at half maximum $2\Gamma=480$ MHz, is shown in Fig. 1(c). A second set of experiments uses a traveling wave to excite the deuterium atoms. In that case, only a fraction of the atoms are excited. The comparison of the total intensity $I_{\rm D}$ of the different com-

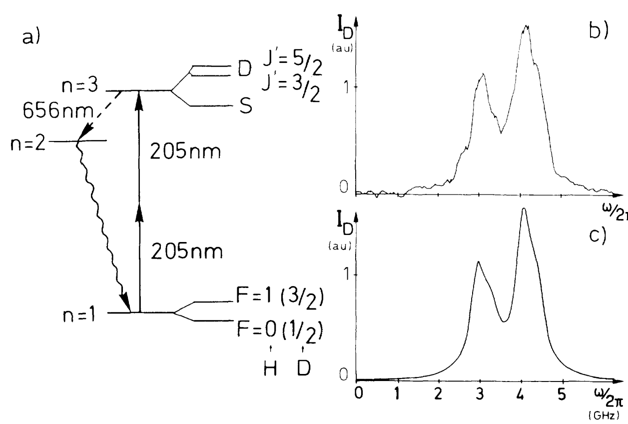

FIG. 1. (a) Two-photon transition from the $n=1$ ground state to the $n=3$ excited state in hydrogen. Excited atoms are detected by monitoring the fluorescence on the Balmer α line due to the spontaneous emission from the $n=3$ to the $n=2$ state; (b) recording of the Doppler-free spectrum for the deuterium present in the experimental sample; and (c) theoretical curve obtained for an effective full width $2\Gamma=480$ MHz. $\omega/2\pi$ is the frequency of the transition, which is twice the frequency of the light source at 205 nm.

ponents of the $1S$-$3D$ two-photon transition for stationary atoms and the maximum intensity $I'_{\rm D}$ in a traveling wave for a Doppler-broadened medium shows that $I'_{\rm D}=I_{\rm D}(\Gamma/\pi\Delta)$, where $\Delta=2ku/\sqrt{\pi}$ is the Doppler width of the two-photon transition in deuterium. Using a neutral filter which attenuates the fluorescence by a factor $F\cong 800$ (measured with an auxiliary He-Ne laser at 633 nm), we detect for $I'_{\rm D}$ about 10 fluorescent photons per pulse.

We have then tuned our light source in the range of wavelengths where superheavy hydrogen may be excited. To find the correct wavelength, we have primarily used a homemade lambda meter. The wavelength is confirmed first by a set of Fabry-Pérot etalons and secondly by monitoring the absorption of an auxiliary beam (at the fundamental wavelength 615 nm) in an iodine cell. The experiment is done with a traveling wave excitation. This is an interesting geometry for the search for superheavy hydrogen because the Doppler width for these atoms varies between 300 MHz for a mass equal to 10^4 GeV/c^2 and 3 MHz for 10^8 GeV/c^2. The excitation spectrum of impurities that could be present inside the cell would give a larger Doppler width and thus cannot lead to false signals. The observation, in the good frequency range, of two lines separated by 1.1 GHz (frequency interval between $3D_{3/2}$ and $3D_{5/2}$) and having a full width of the order of 400–800 MHz would thus be a clear identification of the presence of superheavy hydrogen atoms [18]. For each recording, the laser is scanned to cover a final energy range of 6 GHz, much larger than the fine structure of the transition and than the variation in isotropic mass shift, which is about 150 MHz for masses between 10^4 and 10^8 GeV/c^2. To estimate the maximum abundance of superheavy hydrogen, each recording is compared with the expected dependence [16] of the signal versus the frequency $\omega/2\pi$ of the transition (which is twice the frequency of the light source at 205 nm):

$$\frac{I_S}{I'_{\rm D}}=F\Delta\frac{N_S}{N_{\rm D}}\left[\frac{0.4\Gamma'}{\Gamma'^2+(\omega-\omega_{3/2})^2}+\frac{0.6\Gamma'}{\Gamma'^2+(\omega-\omega_{5/2})^2}\right].$$

In this formula, $N_S/N_{\rm D}$ is the ratio between the abundances of superheavy hydrogen and deuterium in the cell, $\omega_{3/2}$ and $\omega_{5/2}$ are the resonance positions for the $3D_{3/2}$ and $3D_{5/2}$ sublevels, and $2\Gamma'$ is an effective full width which includes the combined effects of the collisional broadening, instrumental width, and Doppler width. This full width varies typically between 800 and 400 MHz in the mass range 10^4 and 10^8 GeV/c^2. An example of curve, together with the "best fit" obtained with $2\Gamma'=600$ MHz, is shown in Fig. 2. For this curve and this value of Γ', we obtain $N_S/N_{\rm D}<8\times10^{-6}$, at the 95% confidence level. Taking into account the natural abundance of deuterium ($N_{\rm D}/N_{\rm H}\cong 1/6000$), this gives $N_S/N_{\rm H}<1.4\times10^{-9}$. The noise is associated with the fluctuations of a red fluorescence coming from the quartz windows. The average number of parasitic photons which reach the

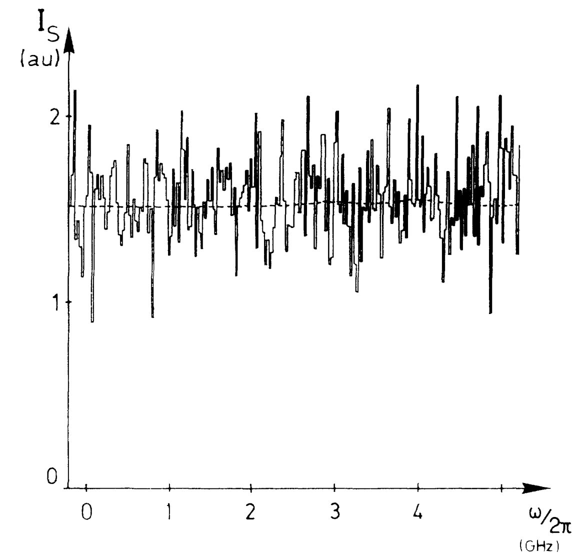

FIG. 2. Experimental curve, obtained with a traveling wave excitation, in the frequency range where the excitation of superheavy hydrogen atoms is expected. The dashed line shows the "best fit" given by the computer.

photomultiplier in the 30 ns following the laser pulse is of the order of 2. By averaging over 64 recordings, we find that the maximum abundance of superheavy hydrogen atoms in our experimental cell is equal to 2.6×10^{-10} for 10^5 GeV/$c^2<M<3\times10^7$ GeV/c^2, with the precision on the edges outside this domain being slightly lower. Taking into account the concentration factor obtained by ultracentrifugation, we find the corresponding upper limits (at the 95% confidence level) on the abundance: 5.8×10^{-15} for the superheavy hydrogen in sea water, and 3.7×10^{-15} in the water including snow and distilled water, for masses between 10^5 and 3×10^7 GeV/c^2. The limits for $M=10^4$, 6×10^7, and 10^8 GeV/c^2 are, respectively, 7.3×10^{-15}, 6.7×10^{-15}, and 9.2×10^{-15}, for superheavy hydrogen in sea water.

This result may be used, in particular, as a stringent test of charged dark matter models [7–10]. Let f_+ be the fraction of local dark matter density [$\rho_{\rm halo}\approx 0.3$ (GeV/c^2)cm^{-3}] due to charged massive particles (CHAMPs) of charge +1. The global flux of such particles may be written as

$$\phi_+\cong\frac{\rho_{\rm halo}}{M}v_0f_+\approx 10^7[M({\rm GeV}/c^2)]^{-1}f_+({\rm cm}^{-2}{\rm s}^{-1}),$$

with $v_0\cong 300$ km/s being an average particle velocity. These CHAMPs, which first interact in the atmosphere, are captured at the surface of the Earth and accumulate in the ocean (of average depth $d\cong 3.7$ km) in a typical time $t_{\rm acc}$, with a resulting density $\phi_+/4dt_{\rm acc}$. Indeed since those arriving above continents should also end up in the ocean, d may in fact be taken as the average ocean depth over all of the Earth's surface, $\cong 2.6$ km. This leads to a rough estimate for the relative abundance of superheavy

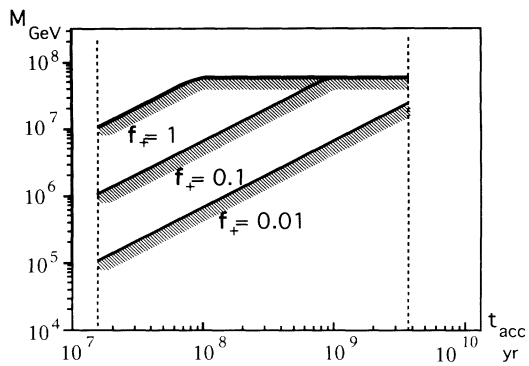

FIG. 3. Lower limits on CHAMP masses, depending on their accumulation time in the ocean, and on the fraction f_+ of dark matter due to positive CHAMPs. The $f_+=1$ limit excludes positive CHAMPs between 10^4 and 10^7 GeV/c^2 as unique dark matter constituents. For $M=10^5$ GeV/c^2, f_+ must be less than 10^{-2}–5×10^{-5}.

hydrogen in the ocean, relative to ordinary hydrogen: $n_X/n_H \approx 4\times10^{-15}[M(\text{GeV}/c^2)]^{-1}t_{acc}(\text{yr})f_+$.

Ocean water continually circulates into the Earth's crust down to the magma and then again up to the ocean, in about 15 to 45×10^6 yr [19]. This yields a conservative lower value for the accumulation time, to be used if this circulation were to act as a perfect filter and trap all superheavy particles inside the magma. Otherwise the accumulation time may be as large as the age of the ocean, more than 3×10^9 yr. Our abundance limit may then be turned into an upper limit on the fraction f_+ of dark matter due to positively charged heavy particles, as shown in Fig. 3. In particular a scenario in which the dark matter would be totally constituted, in equal amounts, of positive and negative CHAMPs ($f_+=\frac{1}{2}$) lighter than about 5×10^6 to 6×10^7 GeV/c^2 (depending on t_{acc}) can be eliminated. Moreover for $M=10^5$ GeV/c^2, for example, the fraction f_+ is constrained to be less than about 10^{-2}–5×10^{-5}, depending on t_{acc}. The hypothesis of strongly interacting dark matter particles is, also, very severely constrained: The above results apply to the fraction, inside dark matter, of new heavy particles which either carry themselves $+1$ unit of charge or will do so after binding to protons.

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[a] Deceased.

[1] P. Fayet, Phys. Lett. **69B**, 489 (1977).

[2] The latter possibility appears favored if the top quark, as it seems, turns out to be rather heavy (about 100 to 200 GeV/c^2) with the ordering of masses being *reversed* when going from ordinary particles to superpartners; see P. Fayet, Phys. Lett. **125B**, 178 (1983).

[3] B. Pichard *et al.*, Phys. Lett. B **193**, 383 (1987).

[4] P. Fayet, Nucl. Phys. **B263**, 649 (1986).

[5] P. F. Smith *et al.*, Nucl. Phys. **B206**, 333 (1982); T. K. Hemmick *et al.*, Phys. Rev. D **41**, 2074 (1990); for earlier searches see P. F. Smith and J. R. J. Bennett, Nucl. Phys. **B149**, 525 (1979), and references therein.

[6] S. Wolfram, Phys. Lett. **82B**, 65 (1979); J. Ellis *et al.*, Nucl. Phys. **B238**, 453 (1984), and references therein.

[7] A. De Rújula, S. L. Glashow, and Uri Sarid, Nucl. Phys. **B333**, 173 (1990).

[8] J. L. Basdevant *et al.*, Phys. Lett. B **234**, 395 (1990); A. Gould *et al.*, Phys. Lett. B **238**, 337 (1990); S. W. Barwick *et al.*, Phys. Rev. Lett. **64**, 2859 (1991).

[9] S. Dimopoulos *et al.*, Phys. Rev. D **41**, 2388 (1990).

[10] The conclusion to essentially rule out (Ref. [9]) the possibility that the abundance of charged dark matter particles is determined by the usual freeze-out mechanism appeals to experiments whose interpretations depend crucially on extrapolations of the stopping power of the material above or inside the detector.

[11] G. Starkman *et al.*, Phys. Rev. D **41**, 3594 (1990).

[12] W. S. Broecker and T. H. Peng, *Tracers in the Sea* (Eldigio Press, New York, 1982), p. 425.

[13] P. Jean-Baptiste (private communication).

[14] B. Pichard *et al.*, in *New and Exotic Phenomena*, Proceedings of the 1990 Moriond Workshop, Les Arcs, France, edited by O. Fackler and J. Tran Thanh Van (Editions Frontières, Gif-sur-Yvette, France, 1990), p. 489.

[15] P. Verkerk *et al.*, Opt. Commun. **72**, 202 (1989).

[16] G. Grynberg and B. Cagnac, Rep. Prog. Phys. **40**, 791 (1977).

[17] Actually, at a lower pressure, each of these components is split into two components, corresponding to the hyperfine structure of the initial $1S_{1/2}$ state; see Ref. [15]. This shows that a substantial fraction of the width is due to collisional broadening.

[18] The range of widths considered here comes from an estimation of the width of the Voigt profile obtained by convolution of the instrumental and Doppler curves. When comparing with deuterium, we have allowed for the possibility of getting smaller width since the collision broadening might be smaller due to a smaller relative velocity between collision partners.

[19] P. Jean-Baptiste *et al.*, Earth Planet. Sci. Lett. **106**, 17 (1991).