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Search for anomalously heavy hydrogen in deep sea water at 4000 m

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A search was made with a sea water sample taken at a depth of 4000 m for anomalously heavy hydrogen dating from the early Universe. A technique of accelerator mass spectrometry involving a time-of-flight spectrometer was used. A new upper limit for the concentration of heavy particles in hydrogen is set around 4×10^{-17} in the mass range of 5–1600 u at a 95% confidence level.

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Motivated by the speculations of the technicolor theory [1] as well as of the supersymmetry theory [2], many efforts to search for heavy particles with masses of more than 10 u have been directed at natural substances [3–13]. Indeed, a search for the heavy particles in nature seems to be feasible in view of their wide mass range ($10-10^5$ u), long lifetime (10^{16} yr or longer), and high concentration [14–17] ($10^{-10}-10^{-12}$ heavy particles/nucleon). Such light elements as H, He, Li, Be, and B seem to be particularly interesting since their origin dates back to the early Universe, in which the heavy particles were possibly present as a tiny fraction of the building blocks for nucleosynthesis [14,17].

The element studied most among natural substances with their origin in big-bang nucleosynthesis is hydrogen. Using highly enriched samples of heavy water, Smith *et al.* [6] and Hemmick *et al.* [12,13] separately set the following most stringent upper limits for the concentration of the heavy particles: $10^{-28}-10^{-29}$ heavy particles/nucleon (X/p) in the mass range of 8–1200 u, and $10^{-24}-10^{-20}$ X/p in the mass range of 100–10 000 u. Their results seem to impose serious doubt on the theoretical predictions on the presence of anomalously heavy hydrogen, at least in water at the terrestrial surface. However, as Smith *et al.* suggested [6], the gravitational effect, that is, the effect of the gravitational forces on the concentration of heavy particles, may be large. In this regard, it is crucial to examine deep sea water in search of exotic hydrogen. Very recently, Hemmick *et al.* [12,13] have extended a search to a variety of materials, including a sea water sample taken at a depth of 3000 m. The study of the deep sea water resulted in setting an upper limit at one of highest concentration levels ($10^{-15}-10^{-10}$) in the mass range of 10^2-10^4 u. According to naive theoretical speculation, this result alone may limit the possible existence of heavy hydrogen in the mass region above 3000 u. However, as discussed in Ref. [13], geochemical processes [18] can lower the theoretical concentration rate by about a factor of 100, making it premature to draw an unambiguous conclusion on the presence of anomalously heavy hydrogen in the mass region even below 3000 u. A more sensitive measurement with deep sea water is clearly needed.

In this paper, we report on the result of a new experiment with a sample of deep sea water at 4000 m. At 4000 m below sea level, the gravitational concentration, which is governed by $\exp(-Mgh/kT)$ [6], can be enhanced by a factor of about 1000 compared to that at 3000 m. A new upper limit is presented. The result seems to exclude the possibility that the heavy particles in the mass range of 5–1600 u are found in hydrogen with the concentration rate theoretically speculated.

The experiment was performed with the 1.5-MV tandem Van de Graaff [19] accelerator of Konan University. Figure 1 schematically shows the experimental setup. Approximately 0.25 l of deep sea water sampled at a

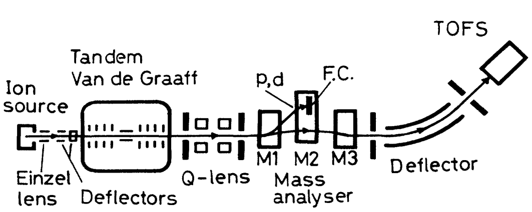

FIG. 1. A schematic of the experimental setup.

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depth of 4000 m near Japan was distilled into fresh water. After adding 0.025 g LiOH to the fresh water, the solution was electrolyzed with a pair of nickel electrodes. The generated hydrogen gas was collected into a glass bottle of 2.4 l.

The hydrogen gas was fed into a Duoplasmatron ion source [19]. The extracted negative ions were injected into the Van de Graaff accelerator through an einzel lens and a pair of electrostatic deflectors. The negative ions were accelerated at a terminal voltage of 1.5 MV and were converted into positive ions by colliding with a nitrogen gas in the stripper channel. The converted positive ions were reaccelerated up to the energy of 3 MeV.

The beam extracted from the Van de Graaff accelerator was led to a magnetic mass analyzer through an electrostatic quadrupole lens. The magnetic mass analyzer consisted of three dipole magnets ($M1$, $M2$, and $M3$) that were arranged symmetrically. The maximum deflection of the ions from the straight-line trajectory occurred in the middle of $M2$. Such ordinary particles as protons and deuterons were completely collected by a Faraday cup at the location of the maximum deflection. The number of beam particles was monitored by means of integrating the electric current from the Faraday cup. The beam intensity was typically 0.5 μA.

After being energetically ($E_i = 3$ MeV) filtered by a 12° electrostatic deflector, the rest of the beam particles was analyzed with a time-of-flight spectrometer (TOFS). The TOFS consisted of two secondary electron detectors and a silicon solid-state detector (SSD) of 200 μm thickness as shown in Fig. 2. The incident particles passed through two carbon foils of 5 $\mu g/cm^2$ thickness each and were detected with the SSD. Two separate measurements of the TOF were carried out between the first and second foils (ΔT_1) and between the second foil and the SSD (ΔT_2). Secondary electrons from the carbon foil were guided to a Hamamatsu R595 electron multiplier tube through an electron collector with five grids, each consisting of gold-plated tungsten wires of 50 μm diameter and of 2–3 mm intervals. The first flight pass length was 32 cm and the second length was 36 cm, which provided the time resolution of 14 ns and 10 ns in full width at half maximum (FWHM), respectively. The resultant mass resolution was 65 u in FWHM at 1600 u.

The SSD was energy calibrated in a separate run with an ^{241}Am α source and checked with a proton beam. The absolute energy (E) of the particles was determined within the accuracy of 75 keV. A typical energy resolution was 70 keV.

The mass range covered in the present experiment was 5–1600 u. The lower and the upper limits were determined by the transmission of ions in the magnetic mass analyzer and by the resolving time (2 μs) of the TOFS, respectively.

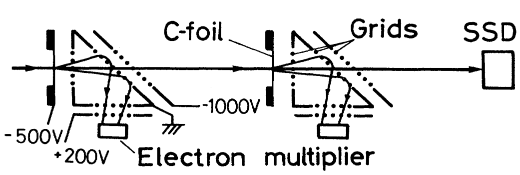

FIG. 2. A schematic of the time-of-flight spectrometer.

Figure 3 shows three two-dimensional spectra in the ΔT_1 vs ΔT_2 plane, in the E vs ΔT_1 plane, and in the E vs ΔT_2 plane, respectively. These are representative spectra obtained in the present experiment. One can see that there are three intense spots corresponding to small values of ΔT_1 and ΔT_2. From the kinematics, they were found to be 2.5-MeV protons (labeled by $p1$ in Fig. 3), 2.2-MeV deuterons ($d1$), and 1.1-MeV deuterons ($d2$), respectively. For reference, kinematical loci for protons and deuterons in the energy range from 0 to 3 MeV are indicated by the dashed lines in the individual spectra. These particles were most likely to arise from bouncing of beam particles (protons and deuterons), whose charge states were probably not consistent in the course of beam transport, off the beam pipe and/or slits. Accidental events built on these intense particles are seen along the vertical and horizontal axes in the spectra.

The solid lines in Fig. 3 indicate kinematical loci for 3-MeV particles with $Z=1$ and $Z=2$ in the mass range from 1 to 1600 u. Events corresponding to the heavy particles of interest could be identified, if there, along the kinematical loci for $Z=1$ particles as coincidental events in the three spectra. No such events were, however, found with a total of 3.2×10^{17} (N_p) incident protons.

The upper limit for the abundance of the heavy particles can be evaluated by $3\times(\varepsilon_I\cdot\varepsilon_{CE}\cdot\varepsilon_{TA}\cdot\varepsilon_{TT}\cdot\varepsilon_D\cdot N_p)^{-1}$, where a factor of 3 assures a 95% confidence level in the

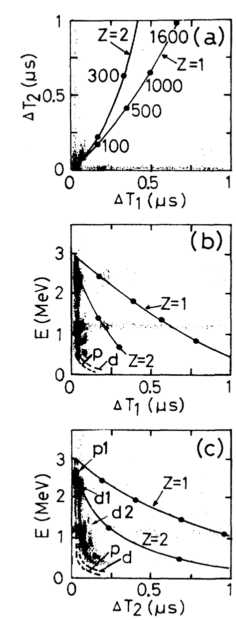

FIG. 3. Representative two-dimensional spectra obtained in a 3 h run in the ΔT_1 vs ΔT_2 plane (a); in the ΔT_1 vs E plane (b); and in the ΔT_2 vs E plane (c). The solid lines represent kinematical loci of $Z=1$ and $Z=2$ particles in the mass range of 1–1600 u at a fixed incident energy of 3 MeV. For the dashed lines and labels of $p1$, $d1$, and $d2$, see the text.

Poisson statistics, ε_I is the ionization efficiency in the ion source, ε_{CE} is the charge-exchange efficiency in the stripper channel, ε_{TA} is the transmission efficiency through the magnetic mass analyzer, ε_{TT} is the beam transmission after the magnetic analyzer, and ε_D is the detection efficiency of the TOFS.

The ε_I may be estimated from the period of time (τ) during which a gas molecule stays in plasma [6]. The diffusion velocity of a gas molecule in plasma at a fixed energy is inversely proportional to the square root of its mass m. That is, τ is proportional to $\sqrt{m}$. Therefore, ε_I for the anomalous hydrogen (HX) is considered to become $[(1+M_x)/2]^{1/2}$ times that for the ordinary hydrogen (H_2).

The charge-exchange efficiency ε_{CE} is sensitive to the velocity of the hydrogen ions. ε_{CE} was estimated from the equilibriated charge states (H^+, H^0, H^-) for hydrogen in a nitrogen gas [20].

The transmission efficiency through the magnetic mass analyzer, ε_{TA}, depends on the magnitude of the magnetic deflection as well as on the width of the beam. The calculated ε_{TA} quickly reaches 100% as the mass of the particle goes up from 5 to 10 u.

The beam *transmission* after the magnetic analyzer, ε_{TT}, was measured to be 80% for 3-MeV protons using two Faraday cups, one located in the middle of the magnetic analyzer and the other in front of the SSD of the TOFS. The beam transmission was essentially equal to the geometrical transparency of the total of the six grids of the TOFS. The transmission loss due to the beam optics and to multiple scattering in the two carbon foils of the TOFS was estimated to be negligibly small, at most a few percent.

The detection efficiency of the TOFS, ε_D, is defined as the ratio of the number of threefold events in the two secondary electron detectors and the SSD to the number of singles events in the SSD. Thus, the ε_D is considered to be essentially determined by the efficiency for production and collection of the secondary electrons. ε_D was measured for protons in the energy range from 0.08 to 3.0 MeV. ε_D was found to be constant at around 17.5%. In general, the production efficiency depends upon the stopping power, i.e., the particle velocity. The velocity range covered by these proton energies is equivalent to that for particles with the mass of 1–37 u at a fixed energy of 3 MeV. Scaling by velocity, the ε_D measured for protons was interpreted as the ε_D for hydrogen with different masses. In the scaling procedure, the collection efficiency of secondary electrons was assumed to be the same. The result was plotted with solid circles in Fig. 4. The extrapolation of the efficiency to the heaviest mass region, namely, to extremely small velocities, was made following the prescription of Smith *et al.* [6].

The various efficiencies, i.e., ε_I, ε_{CE}, ε_{TA}, and ε_D, are summarized in Fig. 4 as a function of the mass of the particle.

Figure 5 shows the resultant upper limit for the concentration rate of the heavy particles in a hydrogen atom. The upper limit turned out to be approximately 4×10^{-17} X/p in the mass range from 5 to 1600 u at a 95%

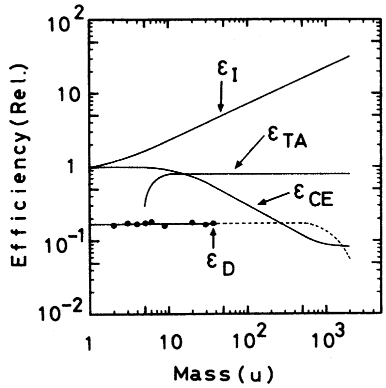

FIG. 4. The dependence of various efficiencies on the mass of the particle: ε_I, ionization efficiency; ε_{CE}, charge-exchange efficiency; ε_{TA}, transmission through the magnetic analyzer; and ε_D, detection efficiency of the TOFS. See the text for details.

confidence level. It is noted that the present measurement is at least 100 times more sensitive than that of Hemmick *et al.* [12,13]. More importantly, even for the deep sea water, the upper limit is much lower than the theoretical concentration rate (10^{-10}–10^{-12} X/p) based upon the scenario of the standard big-bang nucleosyntheses which incorporates heavy particles postulated by the technicolor and supersymmetry theories [14,17]. This conclusion holds even when the theoretical rate is lowered by a factor of 100 due to the geochemical effect [18].

It is instructive to compare our result with that of Smith *et al.* [6], taking into account the gravitational effect $\exp(-Mgh/kT)$, where $-Mgh$ is the gravitational energy and kT is the thermal energy. The present result at $h=-4000$ m is converted to about 10^{-28} X/p at $h=0$

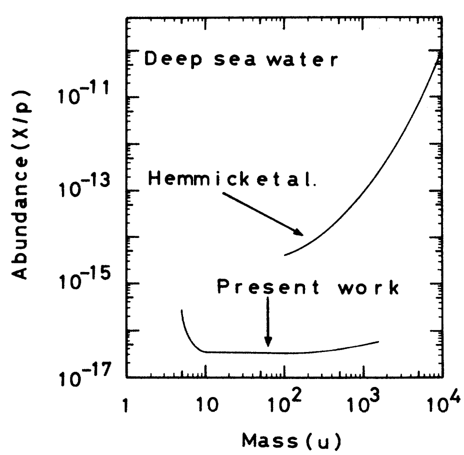

FIG. 5. The experimental upper limit for the concentration of heavy particles in hydrogen obtained for deep sea water at 4000 m. For comparison, the upper limit previously set by Hemmick *et al.* is also shown.

m. The converted result seems to be in good agreement with the result of Smith *et al.* despite the two different condensation effects: i.e., the gravitational and the electrolytic condensations.

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