# Search for supermassive Cahn-Glashow particles in lead

# Eric B. Norman, Robert B. Chadwick, K. T. Lesko, Ruth-Mary Larimer, and Darleane C. Hoffman Nuclear Science Division, Lawrence Berkeley Laboratory, 1 Cyclotron Road, Berkeley, California 94720

(Received 5 December 1988)

A radiochemical search has been made for negatively charged massive elementary particles  $(X^{-})$ bound to lead nuclei by utilizing the fact that such systems would have the nuclear properties of lead but the chemical properties of thallium. We looked for evidence of PbX<sup>-</sup> by searching for  $\gamma$ rays emitted following the  $\beta$  decay of <sup>206</sup>BiX<sup>-</sup> produced by PbX<sup>-</sup>(p,xn) reactions. No evidence of such decays was observed and a limit has been established on the possible concentration of  $X^-$  particles in lead of  $< 1.5 \times 10^{-13}$  per nucleon.

#### **I. INTRODUCTION**

It has been suggested that massive, long-lived, electrically charged elementary particles  $(X^{\pm})$  may have been produced in the big bang and have survived to the present day in concentrations small enough to have escaped detection.<sup>1</sup> If, as has been suggested, such particles have only electromagnetic interactions with ordinary matter, then an  $X^+$  particle would eventually capture an electron and then behave as an anomalously heavy isotope of hydrogen. An  $X^-$  particle would eventually become bound to an atomic nucleus and thereby reduce the effective nuclear charge by one unit. Thus, the chemical properties of an atom of  $(Z, N)X^-$  would be those of an ordinary atom of (Z-1, N) while its nuclear properties would be nearly the same as those of the nucleus (Z, N)(see Glashow and co-workers<sup>1,2</sup>). Estimates of the numbers of such massive elementary particles that were produced in the big bang depend sensitively on their assumed properties. If these objects are hadrons, then their abundances have been estimated to be in the range of  $10^{-12}-10^{-10}$  per nucleon.<sup>3,4</sup> If they are leptons, however, their concentration may be as high as  $10^{-5}$  per nucleon.4

Several previous searches have looked for anomalously heavy isotopes of hydrogen, which could correspond either to  $X^+$  particles or to He $X^-$  bound states.<sup>5-7</sup> These experiments have established that for masses <1 TeV, the concentration of such objects in terrestrial water is less than  $10^{-29}$  per nucleon.<sup>7</sup> While this limit appears to rule out the existence of  $X^+$  particles in anywhere near their predicted abundance, it does not necessarily preclude a much larger abundance of  $X^{-1}$ 's. Such an asymmetry might have been produced by the same type of process that led to the observed excess of baryons over antibaryons in the Universe. Searches in beryllium, boron, carbon, nitrogen, fluorine, magnesium, and curium have established upper limits on the possible concentrations of  $X^-$  particles in these elements that range from  $10^{-8}$  to  $10^{-20}$  per nucleon.<sup>8-12</sup> However, as discussed by Turkevich, Wielgoz, and Economou,<sup>9</sup> nucleosynthesis arguments suggest that these elements may not have been the most favorable places in which to look for  $X^-$  particles. Calculations by several authors<sup>9,13-15</sup> have shown that during charged-particle-induced nucleosynthesis,  $X^{-}$ 's will be preferentially concentrated in the heaviest element produced. Since iron is the element with the highest nuclear charge produced through such fusion reactions, an enhanced abundance of  $X^{-1}$ 's in iron was suggested by some of these investigators. A recent search by Norman, Gazes, and Bennett,<sup>13</sup> however, found no evidence for particles in iron and established a limit of X  $< 1.2 \times 10^{-12}$  per nucleon.

Nucleosynthesis of the elements more massive than iron occurs via the s (slow) and r (rapid) processes in which successive neutron captures are interspersed with  $\beta$  decays. Neutron-capture cross sections should not be affected by the presence of an  $X^-$  in a nucleus. However, as a result of the variation in the Coulomb binding energy with nuclear charge, the presence of  $X^-$  particles in nuclei alters the relative masses of all members of a given isobar. This changes the energies available in  $\beta$  decay and, hence, the  $\beta$ -decay half-lives. The  $\beta^-$  decay rates of nuclei on the neutron-rich side of the stability line will be increased by this effect. Since lead is a major product of such neutron-capture nucleosynthesis, an enhanced concentration of  $X^-$  particles in lead has been suggested.<sup>1</sup>

We have performed a radiochemical search for  $PbX^{-}$ systems, whose chemical properties would be those of thallium. As in all previous searches of this type, the critical assumption we made is that  $X^{-}$  particles have only electromagnetic interactions with nuclei. We calculated the Coulomb binding energy of an  $X^-$  particle to a nucleus using the expression derived by Cahn and Glashow<sup>1</sup> for spherical nuclei with uniform charge distributions. This calculation shows that an  $X^-$  particle is bound by 302 keV more energy to a bismuth nucleus than to a lead nucleus. Thus, the energy available for the nuclear  $\beta$  decay of <sup>206</sup>BiX<sup>-</sup> is 302 keV less than that for the decay of <sup>206</sup>Bi. The two principal decay modes of <sup>206</sup>Bi to the levels at 3.403- and 3.279-MeV excitation energy in <sup>206</sup>Pb have  $\log ft$  values of 6.5 and 6.9, respectively.<sup>16</sup> We assume that the log ft values for the  $\beta$  decays of <sup>206</sup>BiX are the same as those for <sup>206</sup>Bi, because nuclear matrix elements should not be affected by the presence of an  $X^{-}$ . An  $X^{-}$  bound to a nucleus could also shift the energies of the excited states of that nucleus. As a result of the collective nature of the excited states of <sup>206</sup>Pb, such shifts in

2499 39

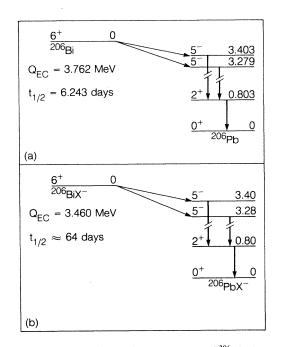


FIG. 1. (a) Known principal decay modes of  $^{206}$ Bi. (b) Calculated decay modes of  $^{206}$ BiX<sup>-</sup>. Note that the spins shown for the levels in  $^{206}$ BiX<sup>-</sup> and  $^{206}$ PbX<sup>-</sup> neglect the possible spin of the X<sup>-</sup> particle.

the positions of these levels are expected to be quite small<sup>17</sup> and are neglected in our analysis. The known<sup>16</sup> major decay modes of <sup>206</sup>Bi and the results of our calculations for the <sup>206</sup>BiX<sup>-</sup> decay scheme are shown in Fig. 1. We find that the  $\beta$ -decay half-life of <sup>206</sup>BiX<sup>-</sup> is approximately 64 days. The primary  $\gamma$  ray from <sup>206</sup>BiX<sup>-</sup> decay will be essentially the same as that from <sup>206</sup>Bi decay.

## **II. EXPERIMENTAL PROCEDURE**

We looked for evidence of  $PbX^-$  by searching for characteristic  $\gamma$  rays emitted following the  $\beta$  decay of <sup>206</sup>BiX<sup>-</sup>. We attempted to produce <sup>206</sup>BiX<sup>-</sup> via the  $PbX^{-}(p,xn)$  reaction. A thick disk containing approximately 23 g of 99.9999% pure thallium metal (which could contain  $PbX^{-}$  bound systems) was bombarded for 4.5 h with a 3- $\mu$ A beam of 25-MeV protons from Lawrence Berkeley Laboratory's 88-Inch Cyclotron. The bombarding energy was chosen to be just below the threshold for scattering an  $X^-$  particle out of a lead nucleus. After the sample was allowed to cool for approximately one month, the lead fraction, which would contain any  $^{206}\text{Bi}X^-$  present, was separated from the target. To do so, the target was first dissolved in  $12M H_2SO_4$ . In order to determine the lead recovery efficiency, a small amount of <sup>212</sup>Pb tracer was added to the solution. The lead fraction was precipitated as PbSO<sub>4</sub> and subsequently dissolved in a mixture of 6M NH<sub>4</sub>COOCH<sub>3</sub> and 6M CH<sub>3</sub>COOH. Na<sub>2</sub>CrO<sub>4</sub> was then added to precipitate PbCrO<sub>4</sub>. This precipitate was dissolved in a mixture of 6M HCl and 3%  $H_2O_2$  and sorbed on a 5-mm  $\times$  100-mm column of AG1-X10 anion-exchange resin. The lead

fraction was eluted with 6*M* HCl and then evaporated to dryness. 6*M* NH<sub>4</sub>COOCH<sub>3</sub> and 6*M* CH<sub>3</sub>COOH were added and the solution was brought to the boiling point. Na<sub>2</sub>CrO<sub>4</sub> was again added to reprecipitate PbCrO<sub>4</sub> which was filtered and then mounted on a planchet for counting. From measuring the <sup>212</sup>Pb activity before and after the chemical separation, we determined that the lead recovery efficiency was 96%.

Approximately two months after the proton bombardment, the lead fraction was counted for a period of 24 h at a distance of 1 cm from the front face of a 100-cm<sup>3</sup> intrinsic germanium detector. The detector was shielded on all sides with 10 cm of low-activity lead. The detector efficiency was measured in the same geometry using standard calibrated  $\gamma$  sources. The photopeak efficiency at 803 keV was found to be  $2.46 \times 10^{-2}$ .

### **III. RESULTS AND DISCUSSION**

The  $\gamma$ -ray spectrum below 1 MeV observed in counting this lead fraction is shown in Fig. 2. The major peaks seen in the spectrum are due to the decay of <sup>203</sup>Pb, which is produced by the <sup>203</sup>Tl(p, n) reaction and was obviously recovered in the lead chemistry, and the decay of <sup>202</sup>Tl, which is produced by the <sup>203</sup>Tl(p, pn) reaction and was not completely removed by the chemistry. All of the ob-

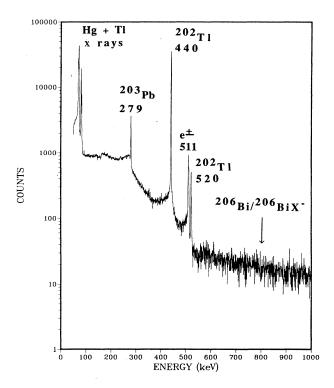


FIG. 2.  $\gamma$ -ray spectrum below 1 MeV observed in 24 h of counting the lead fraction extracted from the proton-irradiated thallium sample. The expected position of the <sup>206</sup>Bi $X^- \gamma$  ray (which is essentially the same as that for <sup>206</sup>Bi) is indicated by an arrow. Unlabeled peaks are due to background activities. All  $\gamma$ -ray energies are given in keV.

served peaks could be assigned to the decays of known nuclei. From the measured widths of known  $\gamma$ -ray peaks and from the measured background counting rate near the expected <sup>206</sup>BiX<sup>-</sup> photopeak position, a 1 $\sigma$  upper limit of 17 counts was established for the <sup>206</sup>BiX<sup>-</sup> decay  $\gamma$  ray. It should be noted that because there are no unidentified peaks between 525 and 1000 keV, this limit is not very sensitive to the exact position chosen for the <sup>206</sup>BiX<sup>-</sup> decay  $\gamma$  ray. From this limit and the measured efficiencies, we placed a limit on the number of <sup>206</sup>BiX<sup>-</sup> nuclei present in the irradiated sample of < 1.3 × 10<sup>5</sup>.

In order to convert this limit on the number of  $^{206}\text{Bi}X^-$  nuclei into a limit on the number of PbX<sup>-</sup> nuclei initially present in the thallium target, we needed to determine the thick-target yield for the  $Pb(p,xn)^{206}Bi$  reaction. To do so, we bombarded a thick disk of 99.95% pure lead metal of natural isotopic abundance with a 115-nA beam of 25-MeV protons. After bombardment, the sample was counted with a 100-cm<sup>3</sup> germanium detector. From the measured yield of  $^{206}$ Bi  $\gamma$  rays, we determined that the  $^{206}$ Bi yield per incident 25-MeV proton on a thick target of natural isotopic composition lead is  $(1.0\pm0.2)\times10^{-3}$ . The presence of a X<sup>-</sup> particle in a lead nucleus should increase this yield as a result of the lowering of both the Coulomb barrier and the reaction threshold energy. However, in order to establish a conservative limit, we have assumed that this yield is unaffected by the presence of a  $X^{-}$ .

- <sup>1</sup>R. N. Cahn and S. L. Glashow, Science 213, 607 (1981).
- <sup>2</sup>P. H. Frampton and S. L. Glashow, Phys. Rev. Lett. **44**, 1481 (1980).
- <sup>3</sup>C. B. Dover, T. K. Gaisser, and G. Steigman, Phys. Rev. Lett. **42**, 1117 (1979).
- <sup>4</sup>S. Wolfram, Phys. Lett. 82B, 65 (1979).
- <sup>5</sup>T. Alvager and R. A. Naumann, Phys. Lett. **24B**, 647 (1967).
- <sup>6</sup>R. A. Muller *et al.*, Science **196**, 521 (1977).
- <sup>7</sup>P. F. Smith et al., Nucl. Phys. **B206**, 333 (1982).
- <sup>8</sup>T. K. Hemmick *et al.*, Nucl. Instrum. Methods **B29**, 389 (1987).
- <sup>9</sup>A. Turkevich, K. Wielgoz, and T. E. Economou, Phys. Rev. D 30, 1876 (1984).
- <sup>10</sup>R. Middleton et al., Phys. Rev. Lett. 43, 429 (1979).
- <sup>11</sup>W. J. Dick, G. W. Greenlees, and S. L. Kaufman, Phys. Rev.

We then established an upper limit on the concentration of  $PbX^{-}$  nuclei in thallium. Using the fact that the solar-system abundance of lead is 13.7 times that of thallium,<sup>18</sup> we find that the concentration of  $X^-$  particles in lead is  $<1.5\times10^{-13}$  per nucleon. This limit is well below the level expected if  $X^{-}$ 's were distributed uniformly in all matter. This is in spite of the enhanced concentration of  $X^{-}$ 's in lead suggested by nucleosynthesis arguments. As in all previous searches of this type, the present search was carried out on a terrestrial sample of unknown geochemical theory. While the possibility of significant mass fractionation cannot be completely ruled out, the studies of Jørgensen<sup>19</sup> and Bigeleisen<sup>20</sup> indicate that such effects should not have greatly depleted the abundance of  $PbX^{-}$ in terrestrial thallium. On the basis of the possible mass fractionation during our laboratory chemical processing of the sample, the present limit should be applicable for  $X^-$  masses up to about 100 TeV (Ref. 9).

### ACKNOWLEDGMENTS

Valuable discussions with R. N. Cahn, W: Swiatecki, and J. Bigeleisen are gratefully acknowledged. This work was supported by the Director, Office of Energy Research, Office of High Energy and Nuclear Physics, Nuclear Physics Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

Lett. 53, 431 (1984).

- <sup>12</sup>R. Brandt et al., Radiochim. Acta 34, 163 (1983).
- <sup>13</sup>E. B. Norman, S. B. Gazes, and D. A. Bennett, Phys. Rev. Lett. 58, 1403 (1987).
- <sup>14</sup>C. K. Au, Phys. Lett. **120B**, 31 (1983).
- <sup>15</sup>K. M. Short, Senior thesis, University of Rochester, 1985.
- <sup>16</sup>C. M. Lederer and V. S. Shirley, *Table of Isotopes*, 7th ed. (Wiley, New York, 1978).
- <sup>17</sup>W. Swiatecki (private communication).
- <sup>18</sup>A. G. W. Cameron, in *Essays in Nuclear Astrophysics*, edited by C. A. Barnes, D. D. Clayton, and D. N. Schramm (Cambridge University Press, Cambridge, England, 1982), p. 23.
- <sup>19</sup>C. K. Jørgensen, Nature (London) **292**, 41 (1981).
- <sup>20</sup>J. Bigeleisen (private communication).