Searches for Supermassive X^- Particles in Iron

Eric B. Norman, Stuart B. Gazes,[a] and Dianne A. Bennett
Nuclear Science Division, Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720
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Two searches have been made for negatively charged massive elementary particles (X^-) in iron nuclei by utilizing the fact that such systems would have the nuclear properties of iron but the chemical properties of manganese. We have looked for $^{56,58}\mathrm{Fe}X^-$ by searching for γ rays emitted following the β decays of $^{56}\mathrm{Co}X^-$ and $^{59}\mathrm{Fe}X^-$ produced by (p,n) and (n,γ) reactions, respectively. No evidence of such particles was observed in either experiment, and a limit has been established on the possible concentration of X^- particles in iron of $<1.2\times10^{-12}$ per nucleon.

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As the energies available in accelerators have increased, new particles have been discovered with ever higher masses, and there are theoretical reasons[1] to suspect that very massive (100 GeV–100 TeV) particles actually exist. Such particles would have been created in the "big bang" and, if we assume that they carry some exotic quantum number (technicolor ?), could be sufficiently long lived to have survived to the present day. Among these may be the so-called $X^{\pm}$ particles.[1] If, as has been suggested, such particles have only electromagnetic interactions with ordinary matter, then an X^+ particle would capture an electron and then behave as a heavy isotope of hydrogen. An X^- particle would bind 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[1,2]). Calculations of the expected abundances of massive elementary particles depend sensitively on their assumed properties. If these objects are hadrons, then their abundances are estimated to be in the range of 10^{-10}–10^{-12} per nucleon.[3,4] 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 to either X^+ particles to $\mathrm{He}X^-$ bound states.[5–7] It is now known that for masses $\lesssim 1$ TeV the concentration of such objects in terrestrial water is less than about 10^{-29} per nucleon.[7] While this limit seems to rule out the existence of X^+'s in anywhere near their predicted abundance, it does not necessarily preclude a much larger abundance of X^-'s. Such an asymmetry might be the result of the same sort of process that led to the observed baryon excess in the universe. Searches in boron, nitrogen, fluorine, magnesium, and curium have established upper limits on the possible abundances of X^- particles in these elements to be in the range of 10^{-8}–10^{-16} per nucleon.[8–12] However, as discussed by Turkevich, Wielgoz, and Economou,[9] nucleosynthesis arguments suggest that these elements may not have been the most favorable places in which to look for X^- particles.

The presence of an X^- in a nucleus decreases the effective nuclear charge by one unit and hence lowers the Coulomb barrier for charged-particle-induced reactions on that nucleus. Furthermore, the Coloumb binding energy of an X^- to a nucleus grows with Z, so that the Q values for fusion reactions on a nucleus containing an X^- will be larger than those for ordinary nuclei. On the other hand, if the mass of an X^- is much greater than that of a typical nucleus, then in stellar environments nuclei containing such particles will have much lower thermal velocities than normal nuclei do. We have investigated the influences of these three effects and find that the increase in reaction rates due to the lowering of Coulomb barriers is essentially canceled out by the lower thermal velocities. However, the increase in the Q values does lead to substantially larger reaction rates for nuclei containing X^- particles. We find that the net result of these effects is that during each major stage of nucleosynthesis X^-'s will be preferentially concentrated in the heaviest element produced, in agreement with two previous studies of nucleosynthesis involving X^- particles.[13,14]

Turkevich, Wielgoz, and Economou,[9] and Short[14] have found that virtually all X^-'s will emerge from big-bang nucleosynthesis bound to ^{7,8}Be or still heavier nuclei. If this material becomes incorporated in stars, then charged-particle–induced reactions will again push X^- particles toward the highest-Z nucleus produced. We have investigated the evolution of X^- particles in the nuclear statistical equilibrium which processes matter from silicon into the elements up to iron. Assuming that any X^- particles present at this stage of nucleosynthesis are initially bound to silicon, we find that over a wide range of temperatures, the final abundance of $\mathrm{Fe}X^-/\mathrm{Si}X^- > 2\times10^4$. While only the innermost regions of stars reach high enough temperatures and densities for the nuclear statistical equilibrium to occur, iron is the major final product of this type of nucleosynthesis. Therefore, there should be a large enhancement in the

concentration of X^- particles bound to iron nuclei.

We have, therefore, undertaken two different searches for FeX^- systems, whose chemical properties would be those of manganese. We have looked for evidence of both $^{56}FeX^-$ and $^{58}FeX^-$ by searching for characteristic γ rays emitted following the β decays of $^{56}CoX^-$ and $^{59}FeX^-$ produced by (p,n) and (n,γ) reactions, respectively. As in previous radiochemical searches of this type,[9,12] the critical assumption that we make in all that follows is that X^- particles have only electromagnetic interactions with nuclei. We have calculated the Coulomb binding energy of an X^- particle to a nucleus by use of the expression derived by Cahn and Glashow[1] for spherical nuclei with uniform charge distributions. As a result of the variation in this binding energy with Z, the presence of X^- particles in nuclei alters the relative masses of all members of a given isobar, which then changes β-decay energies and, hence, half-lives. We assume that the logft values for the β decays of $^{56}CoX^-$ and $^{59}FeX^-$ are the same as those for ^{56}Co and ^{59}Fe, respectively. The presence of an X^- in 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 ^{56}Fe, such shifts in the positions of these levels are expected to be quite small[15] and are neglected in our analysis. On the other hand, the low-lying levels in ^{59}Co are well described as proton single-particle states[16,17] and are thus more strongly affected by the presence of an X^-. Assuming the mass of the X^- is much larger than that of the nucleus, and using harmonic-oscillator wave functions, we have calculated the expected Coulomb shifts for the $1f_{7/2}$ and $2p_{3/2}$ proton orbitals in ^{59}Co.

The known[18] major decay modes of ^{56}Co and ^{59}Fe are shown in Figs. 1(a) and 1(b). The results of our calculations for the $^{56}CoX^-$ and $^{59}FeX^-$ decay schemes are shown in Figs. 1(c) and 1(d). We find that the β-decay half-life of $^{56}CoX^-$ is approximately 1 yr, and that of $^{59}FeX^-$ is 1.5 d. The primary γ rays from $^{56}CoX^-$ decay will be essentially the same as those of ^{56}Co, while those from $^{59}FeX^-$ will be shifted down in energy by approximately 120 keV with respect to those from ^{59}Fe.

In the first experiment, we attempted to produce $^{56}CoX^-$ via the $^{56}FeX^-(p,n)$ reaction. A thick disk containing 28 g of 99.9%-pure manganese was bombarded for 6 h with a 2-μA beam of 10-MeV protons from Lawrence Berkeley Laboratory's 88-in. cyclotron. After the sample cooled for one month, it was dissolved in concentrated HCl and passed through a column of AGI-X8 anion-exchange resin. The iron fraction extracted from this sample, which would contain any $^{56}CoX^-$ present, was stripped from the column with use of water. The principal activity produced in the target was ^{55}Fe. Through measurements of the yields of Mn K x rays produced by the decay of this isotope, we determined that the chemical-recovery efficiency for iron was approximately 50%. The iron fraction was then counted in close

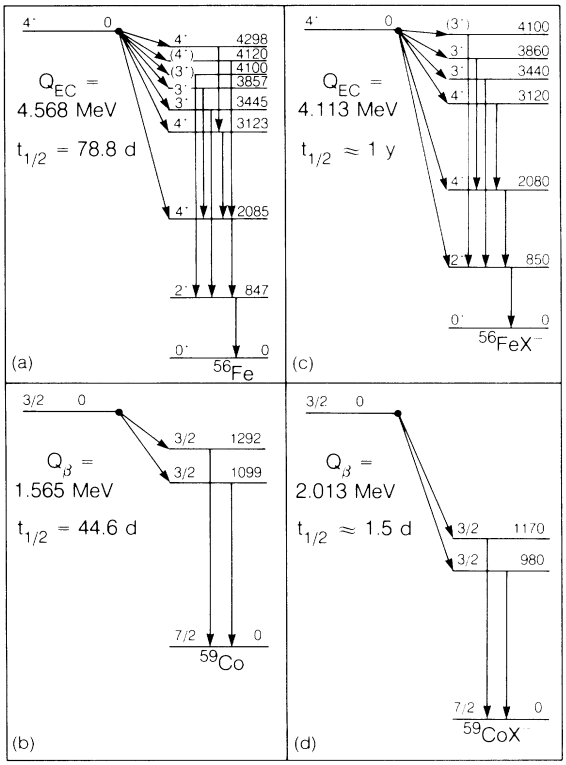

FIG. 1. Known principal decay modes of (a) ^{56}Co and (b) ^{59}Fe, and calculated decay modes of (c) $^{56}CoX^-$ and (d) $^{59}FeX^-$. Note that the spins shown for the levels in $^{56}CoX^-$ and $^{59}FeX^-$ neglect the possible spin of the X^- particle.

geometry with a 110-cm^3 high-purity Ge detector shielded with 10 cm of lead.

A portion of the γ-ray spectrum observed in a period of 63 h is shown in Fig. 2(a). It can be seen that clear peaks are observed at the positions expected from ^{56}Co and/or $^{56}CoX^-$ decay. This could either signal the presence of X^- particles in iron, or be merely the result of imperfect chemical separation. These two possibilities can be distinguished by measurement of the half-lives of these γ rays. We followed their emission rates over a period of approximately two months. The results of these measurements are shown in Fig. 2(b). The composite decay curve obtained by the summing together of the yields of the 847- and 1238-keV γ rays is consistent with that expected from a single activity with the known[18] 78.8-d half-life of ^{56}Co. A least-squares fit to this decay curve allows an upper limit to be placed on a 1-yr component. This 1σ limit was then combined with the measured chemical-recovery and counting efficiencies to set an upper limit on the number of the $^{56}CoX^-$ nuclei present in the irradiated sample. In a separate experiment, we determined that the thick-target $^{56}Fe(p,n)$ yield for 10-MeV protons is $(3.1 \pm 0.5) \times 10^{-4}$ per in-

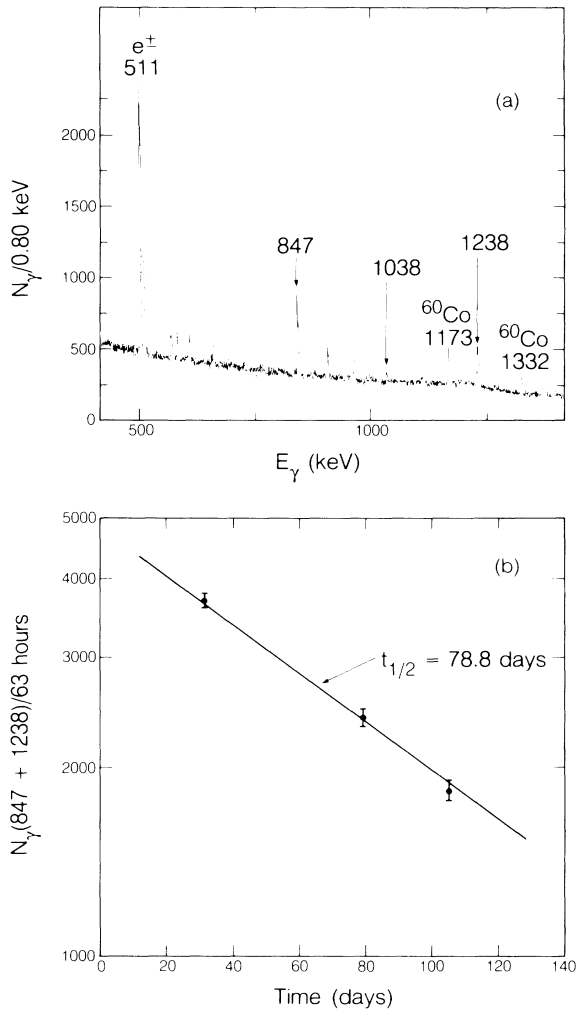

FIG. 2. (a) Relevant portion of the γ-ray spectrum observed from the iron fraction isolated from the proton-irradiated manganese sample. The counting period was 63 h. The positions of the known ^{56}Co γ rays (which are expected to be essentially the same as those of ^{56}CoX^-) are indicated by arrows. Unlabeled peaks are due to the decays of uranium and thorium contained in the shielding material. (b) Composite decay curve of the (847+1238)-keV γ rays. The line is a least-squares fit with assumption of a single activity with the known 78.8-d half-life of ^{56}Co.

cident proton. The presence of an X^- in a ^{56}Fe 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 an X^-. We then establish a limit on the concentration of ^{56}FeX^- nuclei in manganese. Using the fact that the solar-system abundance of iron is 97 times that of manganese,[19] we find that the concentration of X^- particles in iron is $< 1.1\times10^{-11}$ per nucleon.

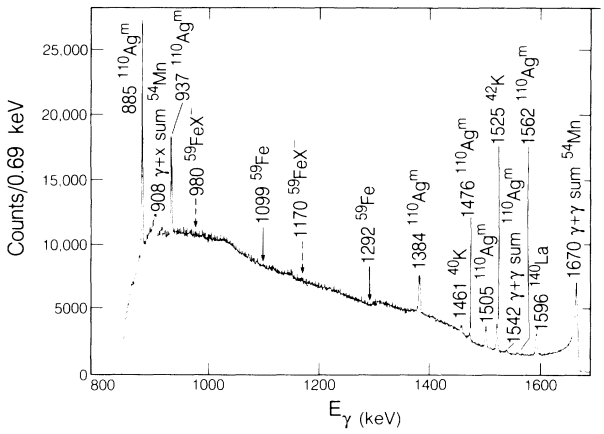

FIG. 3. Relevant portion of the γ-ray spectrum observed following the neutron activation and chemical purification of a 58.2-g sample of manganese. The counting period was 24 h. The positions of the known ^{59}Fe γ rays are indicated by solid arrows. The expected positions of the ^{59}FeX^- γ rays are indicated by dashed arrows.

In the second experiment, we attempted to produce ^{56}FeX^- via the ^{58}Fe$X^-(n,\gamma)$ reaction. 58.2 g of 99.995%-pure manganese were irradiated for 2 h in a thermal flux of 8.4×10^{12} n/cm^2-s at the University of California Berkeley TRIGA Mark III reactor. After the sample cooled for 3 d, the manganese was dissolved in concentrated HCl plus concentrated HNO_3 and then passed through columns of hydrated antimony pentoxide and AGl-X8 anion-exchange resin in order to remove ^{24}Na, ^{59}Fe, and ^{60}Co activities. Lanthanum carrier was then added to the sample, followed by concentrated HF. Rare-earth activities such as ^{140}La and ^{160}Tb were precipitated as fluorides, then centrifuged and discarded. The manganese solution was then boiled down to approximately 100 ml for counting. A minor product in the irradiated sample was ^{54}Mn. Through measurements of the yields of 835-keV γ rays produced by the decay of this isotope, we determined that the manganese chemical-recovery efficiency was approximately 62%. The purified manganese fraction was then counted for 24 h in close geometry with two 110-cm^3 and one 180-cm^3 high-purity Ge detectors, each shielded with 10 cm of lead.

A portion of the γ-ray spectrum observed in one detector over a period of 24 h is shown in Fig. 3. The major peaks seen in the spectrum, aside from ^{54,56}Mn-decay γ rays, are due to the decays of ^{110}Ag^m nuclei produced by <1-ppm silver impurity in the manganese sample. No previously unidentified γ-ray peaks were observed anywhere in the spectrum. From the observed widths of known γ-ray peaks and from the background counting rates measured in all three detectors near the expected ^{59}FeX^--decay peak positions, 1σ upper limits of approx-

imately 650 total net counts were established for each of the $^{59}FeX^-$ decay γ rays. It should be noted that because there are no other peaks seen between about 940 and 1380 keV, these limits are not very sensitive to the exact positions chosen for the $^{59}FeX^-$ decay γ rays. From these limits and the measured efficiencies, we place an upper limit on the number of $^{59}FeX^-$ nuclei present in the irradiated sample. Assuming that the 1.14-b thermal-neutron capture cross section of ^{59}Fe (Ref. 18) is unaffected by the presence of an X^- particle, we derive, as described above, that the upper limit on the concentration of X^- particles in iron is $<1.2\times10^{-12}$ per nucleon.

In conclusion, we have performed two different radiochemical searches for evidence of X^- particles in iron. No evidence of such particles was observed in either experiment. From these searches, we have established that the concentration of X^- particles in iron is $<1.2\times10^{-12}$ per nucleon. This is at or below the level expected[3,4] if X^-'s were distributed uniformly in all matter. This is in spite of the enhanced concentration of X^- particles in iron suggested by nucleosynthesis arguments. Like all previous experiments of this type, the present searches were carried out on terrestrial samples of unknown geochemical history. While the possibility of significant mass fractionation cannot be completely ruled out, the investigations of Jørgensen[20] and Bigeleisen[21] indicate that such effects will not greatly deplete the terrestrial abundances of FeX^- in manganese. On the basis of the possible mass fractionation during our laboratory chemical processing of the samples, the present limits should be applicable for X^- masses up to about 100 TeV.[9]

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[a]Present address: Department of Physics and Astronomy, University of Rochester, Rochester, NY 14627.

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