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[8] See Ref. 2 for a comparison of preliminary data from Reaction (1) with the excitation function calculated in the Adler model. Theoretical curves are not shown in Fig. 3 since the model only describes πN masses below 1.4 GeV.

Search for Stable, Abnormal (Collapsed) Nuclei in Nature*

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Very tightly bound, abnormally-high-density states of nuclei, suggested as a possibility by Bodmer and by Lee and Wick, have been searched for as an anomalous isotope of radon. This was done by looking for high-energy γ rays following thermal-neutron capture. The limit that may be set for the atomic concentration of such nuclides on Earth is $\sim 10^{-29}$ atom of Si.

The possible existence of a very tightly bound, stable, and hitherto unobserved state of nuclear matter was suggested some thirty years ago.[1] More recently Bodmer[2] discussed this possibility in more detail and very recently Lee and Wick[3] suggested a specific model for such a state. This model leads to the expectation of abnormally dense nuclei with large A for which the $N=Z$ isotope would be stable with binding energy of perhaps several hundred MeV per nucleon. Several searches are under way to detect such nuclei in the backscattering of high-energy protons at Fermi National Accelerator Laboratory and to produce them in relativistic heavy-ion collisions at Berkeley.[4] The present experiment was designed to set some limits on the terrestrial concentration of such material.

The most obvious limit one may set for an isotope of small abundance and anomalous mass is from a comparison of the values of atomic masses determined directly by a mass spectrometer and those deduced from stoichiometric chemical measurements. In the case of Bi, for instance, the mass from chemical measurements[5] is 208.976 while the direct measurement[6] is 208.980 401. From this one may conclude that the abundance of a Bi isotope with anomalously low mass, $A = 166$ (if $N \approx Z$) and mass $\lesssim 150$, is $\lesssim 6\times 10^{-5}$. One could probably lower this limit by specific searches but not by much more than three or four orders of magnitude.

If there are stable nuclei with binding energies much higher than in normal nuclei, then the capture of an additional neutron should lead to the release of photons with anomalously high energy. If the energy available is greater than ~ 140 MeV, pion emission is likely, and these will likewise lead to high-energy photons, with fair probability.

In order to estimate the capture rate for such a process we first need to estimate the capture cross section at thermal energies. Assume that the level density is sufficiently higher than in normal nuclei so that many resonances occur in the thermal region (the level spacing $D \ll 25$ meV), and assume that the decay width Γ_c of the capturing state (whether it be pure photon decay or pion decay) is larger than the neutron width Γ_n. With these assumptions one gets[7] $\sigma_c \approx 4\pi k^{-1}K^{-1}$ where k and K are the wave numbers of the neutron at infinity and inside the nucleus, respectively. If the potential inside is approximately ten times deeper than in normal nuclei, one gets $\sigma_c \approx 10^{-21}$ cm^2. The analogous situation exists in normal heavy nuclei: By adding a thermal neutron one is above the fission threshold, and $\Gamma_f \gg \Gamma_n$; the fission cross sections are between 0.3 and 3 times 10^{-21} cm^2.

In an experimental search for high-energy γ rays resulting from thermal-neutron capture in the hypothetical tightly bound states, an array of NaI detectors was used to observe γ rays from several samples of material placed 7.3 m away in an 11.4-cm-diam tube passing near the core of the reactor CP-5, where the thermal-neutron flux is about 3×10^{13} neutrons cm^{-2} sec^{-1} (for details see Bollinger and Thomas[8]). The photon beam from the sample was collimated to 2.5 cm in diameter on the axis of a NaI scintillator 5 cm in diameter and 15 cm long. This central detector was surrounded by a large annular NaI detector.[8]

A γ-ray incident on the axis of the central 5×15-cm detector interacts with high probability. Moreover, if the γ-ray energy is less than a few hundred MeV, it will usually deposit at least as much energy in the central detector as in the annulus. In contrast, a cosmic-ray event almost always deposits much more energy in the thick annulus than in the central detector. Thus, the ratio of pulse heights in the two detectors gives a good indication of the nature of the radiation, and the sum of the pulse heights is a measure of the energy of an incident γ ray. The energy-response function of the detector system was found[9] to be a peak with a width (full width at half-maximum) of 14% for 20.6-MeV γ rays from the reaction $^3\mathrm{He}(n,\gamma)^4\mathrm{He}$.

Largely as a test of the detector system, a search for γ rays of anomalously high energy from a large sample of reactor-grade graphite was carried out early in 1973, after Bodmer suggested the possibility of collapsed nuclear states. These measurements showed that selection of the ratio of pulse heights in the central and outer detectors permitted the cosmic-ray background to be reduced to ~ 1 count/min, about two orders of magnitude lower than without the pulse-ratio requirement. The measured intensity of γ rays in the energy range 30–250 MeV was zero within the statistical error, and this result leads (under the assumptions specified below for another sample) to a limiting concentration of $\sim 10^{-14}$ collapsed nuclei per normal carbon nucleus in the sample.

The considerations of Lee and Wick suggest that the hypothetical abnormal nuclei would have to be quite heavy. Consequently, we have attempted to find a sample from which possible heavy abnormal nuclei can be extracted to form an enriched sample. Such an enrichment procedure exists in nature for elements for which none of the normal isotopes is stable. The noble gas radon ($Z = 86$) is especially favorable since its longest-lived isotope has a half life of 4 days. Thus, any anomalous isotope of Rn, if stable, is likely to be the dominant one. Such a radon isotope might be expected to be present with Kr and Xe in the atmosphere, since Kr and Xe have similar histories and similar ratios of abundances in the atmosphere and in cosmic concentrations.[10]

A sample of the possible abnormal radon nuclei was obtained from Linde Air Corporation in the form of 335 l of the "bottom" fraction of their Xe distillation column, where any hypothetical Rn would remain. It represents the Rn fraction collected from at least 10 000 l (STP) of Xe, which corresponds to 0.12 km^3 (or ~ 150 000 tons) of air.

Chemical purification of the Rn was accomplished by adding 500 000 dis/min of ^{222}Rn tracer to the Rn-Xe mixture and allowing the mixture to flow at a rate of 15 l/hr through 1 g of $1\mathrm{F}_6\mathrm{Sb}_6$. This compound has been shown[11] specifically to oxidize radon to a nonvolatile fluoride, even in the presence of dry atmospheric gases. The compound was then hydrolyzed by dissolution in a 1*N* NaOH solution and the Rn gas was entrained from the solution by use of He gas. The He-Rn mixture was passed through a 1*N* NaOH, 1*N* $\mathrm{Na_2SO_3}$ solution to remove residual traces of iodine and fluorine, the gas was dried by use of a Drierite fitted *U* tube, and the Rn was condensed in a liquid-nitrogen trap. This condensate was recycled through a second purification train of identical chemical composition, recondensed, and sealed in a 2 mm× 1 cm quartz capsule for irradiation. The overall yield as indicated by the ^{222}Rn tracer was 80%.

Measurements to set limits for high-energy γ rays, using this radon sample, gave a null result; specifically, insertion of the sample into the reactor caused the counting rate to change by -0.012 ± 0.021 counts/min. The measurements with the radon sample were more than 1 week in duration, and the background level was measured by using a He-filled capsule with the reactor on and also with the reactor off.

Taking the detector efficiency as 0.25, the solid angle as 2×10^{-5} sr, and assuming a neutron-capture cross section of 10^3 b, the measurements imply that the radon sample contains $< 3\times 10^{10}$ of the abnormal nuclei. Corresponding concentrations are given in Table I. It should be noted that the radon sample should also contain stable, abnormal isotopes of the next heavier noble gas (eka-radon) expected[12] at $Z = 118$. For even heavier gases, the chemical properties become more dubious.

What concentration of abnormal nuclei might be present in the atmosphere? Under the worst assumption, that the *ab origine* concentration is zero, the abnormal nuclei might be produced by cosmic-ray nuclei with $A \gtrsim 150$ and very high energies ($E/A \gtrsim 1$ GeV).[3] From the known fluxes

TABLE I. Limits on radonlike abnormal matter.

Limit of observation (no. of atoms)	$<3\times 10^{10}$
Atomic concentration in xenon sampled	$<10^{-16}$
Present terrestrial concentration[a]	$<10^{-29}$ per atom of Si $<10^{-29}$ by weight[b]
Primordial concentration in solar system[c]	$<10^{-21}$ per atom of Si $<10^{-24}$ by weight[b]

[a] Assuming all Rn-like material on Earth is in the atmosphere and using 5×10^{-13} Xe atoms per atom of Si (a value obtained by taking Earth's mass to be 14% Si by weight) for the terrestrial abundance.

[b] Using atomic weight of ~ 150 for these radon and eka-radon nuclei.

[c] Assuming that any Rn-like material in Earth's atmosphere is lost from there at the same rate as Xe and Kr.

(10^{-8} cm^{-2} sec^{-1}) and concentrations (3×10^{-4} for $A>150$), one can calculate that in 10^9 y there are ~ 10^5 collisions per square centimeter between such energetic heavy projectiles and heavy nuclei on the moon and in meteorites or other objects without a light-atom atmosphere. If we make the, perhaps optimistic, guess that 10^{-5} of such collisions lead to an abnormal nucleus, then 1.0 cm^{-2} would be produced in 10^9 y. Averaged over the volume of meteorites of a few-cubic meters, this implies atomic concentrations of ~ 10^{-24}; and averaged over the volume of the moon it implies a concentration of ~ 10^{-30}. These concentrations are indeed rather low.

If, however, some of the abnormal nuclei produced by cosmic rays are exposed to a large slow-neutron flux in a stellar interior, they would capture many neutrons[13] and could possibly multiply manyfold through fission. In fission the fragments would remain in the tightly bound state, since substantial additional energy would be required for them to re-enter the normal state. The concentration of abnormal matter might then be anything from 10^{-6} (the experimental limit from mass measurements) on down to 10^{-35}, though many of these nuclides could have atomic weights of 10^3 or more and unpredictable chemical properties. The abundance estimate for Rn or eka-Rn in the abnormal state has a similar range of uncertainties, perhaps 10^{-8}–10^{-40}. Our experimental limit, though probably fifteen orders of magnitude better than other existing evidence, is not a conclusive disproof of this fascinating hypothesis.

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[13] Using cross-section estimates similar to the ones made for thermal neutrons, but modified for appropriate stellar temperatures, one may estimate ~10^4 captures from the integrated flux of the s process and 10^{10} captures during the r process for each abnormal nucleus. If fission occurs after 10^3 captures the s process would increase the atomic abundance by a factor of 1000, if it occurs after 100 captures the abundance would increase by 10^{30}. In the r process almost all matter could be converted into the abnormal state even if fission occurred only after 10^6 captures. β decays are likely to be fast (~msec) because of the large symmetry energy terms expected, but since the duration of

the r process is days, this would still not be a very serious limitation. In any case the r process is likely to increase the amount of matter in the abnormal state very substantially. From terrestrial studies on the abundance of various isotopes above Fe, one may estimate that $\sim 10^{-3}$ of all matter has been processed through the s process and perhaps one quarter as much through the r process, as discussed in D. D. Clayton, W. A. Fowler, T. E. Hull, and B. A. Zimmerman, Ann. Phys. (N. Y.) 12, 331 (1961).

Evidence for Two-Electron Excitation Producing Li *K* Vacancies in Slow Li^+ + He Collisions

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Significant Li K excitation is observed in $Li^+ + He$ collisions at incident energies as low as 6 keV. To interpret the results a multistep process is proposed including two-electron excitation in the quasi molecule formed during the collision. The importance of this process is discussed for heavier collision systems.

In the past, K-shell excitation in slow ion-atom collisions has often been described in terms of the molecular orbital (MO) model.[1,2] According to this model available L-shell vacancies may be transferred to the K shell by rotational coupling of the $2p\pi$ and $2p\sigma$ MO's.[2,3] In the case of asymmetric collisions,[4] K vacancies are produced primarily in the particle with the lower atomic number Z. However, there is a finite probability for K excitation of the higher-Z particle as the $2p\sigma$ and $1s\sigma$ MO's couple. Meyerhof,[5] using the charge-transfer model by Demkov,[6] has derived a simple expression to calculate the $2p\sigma$-$1s\sigma$ transition probability in good agreement with a large number of experimental data.[5,7] Only recently, measurements[8] from our laboratory have shown discrepancies between experiment and Meyerhof's theory when light collision systems such as C + B are used.

In the present work we report on the even lighter system Li + He. Similar studies[9-14] have previously been made to investigate primarily excitation of He. Measurements of Li K excitation have been carried out at relatively high energies,[15,16] and corresponding cross sections have not been reported. In this work cross sections for Li K excitation were measured in $Li^+ + He$ collisions at energies as low as 6 keV for which the promotion model is expected to be applicable. It is found that Li K vacancies are produced with much higher probability than predicted from Meyerhof's theory. We suggest that the application of Meyerhof's formalism needs revision with respect to the possibility of multielectron excitation in the lighter collision partner. The significant double excitation of He in $Li^+ + He$ collisions has been studied extensively in the past.[9-14] Here, it is shown that double excitation of the lighter particle produces enhanced K-vacancy production in the heavier collision partner.

The experiments were carried out using the crossed-beam apparatus described in detail previously.[17] The chamber was operated at a 50-kV ion accelerator equipped with a universal ion source. Beams of 6- to 30-keV Li^+ were magnetically analyzed and, after the passage through 2-mm collimator slits, currents of typically 2 μA were obtained. In the center of the chamber the ion beam was crossed by an atomic He beam producing a gas target of $\sim 2\times10^{-3}$ Torr pressure and ~ 3 mm length. Electrons ejected from the target region were detected by an electrostatic parallel-plate spectrometer. In Fig. 1 a typical electron spectrum is shown for 10-keV $Li^+ + He$. The plot indicates two groups of peaks at about 35 and 65 eV originating from autoionizing He and Li, respectively. The two groups were separately integrated with respect to electron emission angle and energy to obtain total cross sections for electron production. Electron emission was found to be significantly anisotropic. Therefore, the angular integration was performed under the assumption[19] that the cross-section differential in electron-emission angle θ is given by $d\sigma/d\theta = a + b\cos^2\theta$, where a and b are constants determined from separate measurements at 30° and 90°.

With neglect of competing radiative transitions, the deduced electron-production cross sections were set to be equal to cross sections $\sigma_2(\mathrm{He})$ and