Another point to emphasize is that in real fluids, as opposed to idealized models, there exists a maximum correlation distance  $\Lambda$  beyond which all correlations are wiped out by disordered collisions. For  $R \gg \Lambda$ , Blatt, Butler, and Schafroth<sup>5</sup> have proved that the moment of inertia necessarily approaches the rigid-body value:  $I = I_0[1 + O(\Lambda/R)]$ . How this affects the London equation (at large wavelengths) has been discussed by Schafroth and Blatt.<sup>12</sup> The results quoted above under (3) illustrate the fact that, for the BCS model,  $\Lambda$  is infinite. A realistic computation of  $\Lambda$  would be desirable, not only for superconductors but even more so for systems like He II and heavy nuclei.

 $^1R_{\circ}$  D. Amado and K. A. Brueckner, Phys. Rev. 115, 778 (1959).

- <sup>2</sup>R. M. Rockmore, Phys. Rev. <u>116</u>, 469 (1959).
- <sup>3</sup>R. M. Rockmore (to be published).
- <sup>4</sup>M. R. Schafroth, Phys. Rev. <u>100</u>, 502 (1955).
- <sup>5</sup>J. M. Blatt, S. T. Butler, and M. R. Schafroth,

Phys. Rev. 100, 481 (1955).

<sup>6</sup>J. M. Blatt and S. T. Butler, Phys. Rev. <u>100</u>, 476 (1955).

<sup>7</sup>M. R. Schafroth, Helv. Phys. Acta <u>24</u>, 645 (1951). <sup>8</sup>Unfortunately, Schafroth's paper<sup>4</sup> contains several bothersome misprints. On p. 504, all expressions for the magnetic moment should be divided by 2. Accordingly, in Eq. (13),  $2\chi'$  should be replaced by  $4\chi'$ .

<sup>9</sup>M. Gell-Mann and K. A. Brueckner Phys. Rev. <u>106</u>, 364 (1957); K. Sawada, Phys. Rev. <u>106</u>, 372

(1957); K. Sawada, K. A. Brueckner, N. Fukuda, and R. Brout, Phys. Rev. <u>108</u>, 507 (1957).

<sup>10</sup>G. Wentzel, Phys. Rev. <u>108</u>, 1593 (1957).

<sup>11</sup>P. W. Anderson, Phys. Rev. 112, 1900 (1958);

- G. Rickayzen, Phys. Rev. 115, 795 (1959).
- <sup>12</sup>M. R. Schafroth and J. M. Blatt, Phys. Rev. <u>100</u>, 1221 (1955), and Nuovo cimento 4, 786 (1956).

## ISOTOPIC COMPOSITION OF PRIMORDIAL XENON\*

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In a previous Letter,<sup>1</sup> we reported a large excess of Xe<sup>129</sup> in a chondritic stone meteorite, Richardton. We also reported secondary anomalous abundaces for many of the other isotopes in this xenon. We have now found similar secondary anomalies in xenon from a carbonaceous chondrite, Murray. Since the ratio of xenon to the cosmic-ray-produced nuclides Ne<sup>21</sup> and He<sup>3</sup> is very different for these two meteorites, we conclude that the anomalies in question are not due to nuclear reactions induced by cosmic rays. There appears to be an isotopic difference between primordial xenon-or at least xenon incorporated in stone meteorites-and xenon in the atmosphere. To our knowledge, xenon is unique among the elements in exhibiting an effect of this kind and magnitude.

We have made a comprehensive analysis by isotope dilution of all the rare gases in the two stones. The results are presented in Tables I, II, and III. The nuclides He<sup>3</sup> and Ne<sup>21</sup> are almost entirely cosmic-ray produced in both stones. We note from the abundances of these isotopes in Table I that the integrated cosmic-ray exposure for Richardton exceeds that for Murray by a factor of about 5. Data for argon and for the other isotopes of He and Ne show clearly that there is primordial gas in Murray. The isotopes  $A^{36}$  and  $A^{38}$  are more abundant in Murray than in Richardton by factors 30 and 9, respectively. The ratio  $A^{36}/A^{38}$  in Murray is 5.39 which is very close to that for the atmosphere (5.35) and very unlike that produced by cosmic rays in iron meteorites (0.6); for Richardton,  $A^{36}/A^{38}$  is 1.62 which is intermediate. That little of the argon in Murray can be from atmospheric contamination is seen from the  $A^{40}/A^{36}$  ratio which is 8.5, in contrast with 296 for the atmosphere. (The  $A^{40}/A^{36}$  ratio for Murray is the lowest that has been seen thus far in a stone meteorite.) Clearly argon was incorporated in the Murray stone at the time of its formation. Neon from Murray can be resolved into a cosmic-ray component (Ne<sup>20</sup> ≅ Ne<sup>21</sup> ≅ Ne<sup>22</sup>) plus a primordial component with  $Ne^{21}/Ne^{20}$  assumed to be that for the atmosphere and  $Ne^{22}/Ne^{20} = 0.111 \pm 0.005$  as compared with  $Ne^{22}/Ne^{20} = 0.097$  for atmospheric neon. It is very unlikely that this neon is from atmospheric contamination because Ne/A in Murray is 0.1 [which means that  $Ne/(A)_{atmos}$  in Murray is considerably greater than 0.1] while Ne/A in the atmosphere is 0.002. He<sup>4</sup> is also in

	Richardton	Murray	Pesyanoe
Sampling	Reduced to minus 20 m in diamond mortar. (Large malleable grains rejected.) 20-100 m used for analysis.	Samples crushed by several blows in diamond mortar. All material used.	
$\mathrm{He}^{3}$	$(2.88^{b} \pm 0.15) \times 10^{-7}$ $(3.30^{c})$	$(0.68 \pm 0.03) \times 10^{-7}$	<18 <sup>f</sup> ×10 <sup>-7</sup>
$\mathrm{He}^4$	$(12.0^{b} \pm 2.5) \times 10^{-6}$ $(15.1^{c})$	$(142 \pm 7) \times 10^{-6}$	7300 <sup>f</sup> ×10 <sup>-6</sup>
$Ne^{20}$	$(9.88^{b} \pm 0.6) \times 10^{-8}$	$(65.2 \pm 3.2) \times 10^{-8}$	1900 <sup>f</sup> ×10 <sup>-8</sup>
Ne <sup>21</sup>	$(11.52^{b} \pm 0.7) \times 10^{-8}$	$(2.22 \pm 0.11) \times 10^{-8}$	19 <sup>f</sup> ×10 <sup>-8</sup>
Ne <sup>22</sup>	$(11.82^{b} \pm 0.7) \times 10^{-8}$	$(9.07 \pm 0.45) \times 10^{-8}$	$171^{f} \times 10^{-8}$
A <sup>36</sup>	$(2.59^{b} \pm 0.4) \times 10^{-8}$ $(2.83^{d})$	$(76.5 \pm 3.8) \times 10^{-8}$	168 <sup>f</sup> ×10 <sup>-8</sup>
A <sup>38</sup>	$(1.60^{b} \pm 0.06) \times 10^{-8}$ $(1.16^{d})$	$(14.2 \pm 0.7) \times 10^{-8}$	36 <sup>f</sup> ×10 <sup>-8</sup>
A <sup>40</sup>	$(53.2^{b} \pm 2.1) \times 10^{-6}$ (54.0 <sup>d</sup> )	<6.53×10 <sup>-6</sup>	47.1 <sup>f</sup> ×10 <sup>-6</sup>
Total Kr	Within factor 3 of total Xe	$(27.3 \pm 1.3) \times 10^{-9}$	•••
Total Xe	$(1.04 \pm 0.05) \times 10^{-9}$	$(43 \pm 4) \times 10^{-9}$	•••
Anomalous Xe <sup>129</sup>	$(0.13 \pm 0.01) \times 10^{-9}$	$(1.09 \pm 0.11) \times 10^{-9}$	•••
Anomalous Xe other than Xe <sup>129</sup>	$(0.029 \pm 0.002) \times 10^{-9}$	$(1.24 \pm 0.12) \times 10^{-9}$	
K content	$(0.073^{b} \pm 0.009)\%$ $(0.083^{d})$	$(0.026^{b} \pm 0.010) \%$ $(0.038^{e})$	?
K-A age <sup>a</sup>	$(4.47 \pm 0.2) \times 10^9$ years	<2.77×10 <sup>9</sup> years	$4.2^{f} \times 10^{9} \text{ years}$

Table I. Rare gas analyses of the meteorites. (All gas contents are in cc STP/g.)

<sup>a</sup>Using  $\lambda_{\kappa} = 0.585 \times 10^{-10} \text{ yr}^{-1}$ ;  $\lambda = 5.30 \times 10^{-10} \text{ yr}^{-1}$  for K<sup>40</sup> decay. <sup>b</sup>Correction for blank run has been made.

<sup>c</sup>P. Eberhardt and D. Hess (to be published), with different sampling. <sup>d</sup>J. Geiss and D. Hess, Astrophys. J. <u>127</u>, 224 (1958), with different sampling. <sup>e</sup>G. Edwards, Geochim. et Cosmochim. Acta <u>8</u>, 285 (1955). <sup>f</sup>E. Gerling and L. Levskii, Doklady Akad. Nauk S.S.S.R. <u>110</u>, 750 (1956).

Table II. Isotope abundance data for krypton.

Isotope ratio	78/84	83/84	86/84
Richardton	•••	$0.211 \pm 0.007$	$0.318 \pm 0.003$
Murray	$0.0062 \pm 0.0006$	$0.201 \pm 0.0025$	$0.311 \pm 0.006$
Atmosphere	0.00622	0.203	0.305

Isotope	Richardton	Murray	Murray/Richardton
124	$0.33 \pm 0.09$	$0.64 \pm 0.05$	$1.94 \pm 0.54$
126	$0.23 \pm 0.02$	$0.56 \pm 0.10$	$2.43 \pm 0.50$
128	$2.51 \pm 0.32$	$6.52 \pm 0.25$	$2.60 \pm 0.35$
129	$81.5 \pm 3.5$	$46.6 \pm 3.4$	$(0.572 \pm 0.048)$
130	$2.27 \pm 0.39$	$7.33 \pm 0.81$	$3.23 \pm 0.66$
131	$8.18 \pm 2.0$	$25.12 \pm 2.7$	$3.07 \pm 0.81$
132	$3.63 \pm 2.3$	$11.59 \pm 3.4$	$3.19 \pm 2.21$
134	$1.35 \pm 1.0$	$1.69 \pm 1.3$	$1.25 \pm 1.36$
136	≡ 0	$\equiv 0$	• • •
			Average 2.53
		V	Veighted average 2.53

Table III. Percentage composition of anomalous xenon.

excess abundance in Murray. If the uranium content of Murray is 0.01 ppm, as for Richardton and other chondrites,<sup>2</sup> then the large difference in He<sup>4</sup> content between Murray and Richardton must be attributed to primordial helium in the former. Richardton may contain a little primordial A<sup>36</sup>. We calculate an upper limit of 1.8  $\times 10^{-8}$  cc STP/g primordial A<sup>36</sup> in Richardton by assuming that  $A^{36}$  and  $A^{38}$  in this stone is a mixture of primordial argon with  $A^{36}/A^{38} = 5.35$  and cosmogenic argon with  $A^{36}/A^{38} = 0.6$ . Thus Murray has more primordial argon than Richardton by a factor of at least 42. There is one other stone meteorite known to contain primordial gas. This is Pesyanoe, which is variously described as a howardite or aubrite achondrite and which has been studied by Gerling and Levskii.<sup>3</sup> Their data are included in Table I for comparison.

K-A ages for the stones are given in Table I. The upper limit for the K-A age of Murray of  $2.8 \times 10^9$  years results from assuming that all of the A<sup>40</sup> is radiogenic. This low K-A age probably means that Murray has lost much (about 2/3) of its radiogenic argon. There may have been loss of primordial gas as well, although such loss would not alter markedly any of the conclusions reached in this Letter. Los of cosmic-ray-produced gas should be much less since these gases were produced in the last few tens of millions of years and, furthermore, cannot be concentrated at grain boundaries. A partial isotopic analysis of the krypton from the stones is presented in Table II. These data are averages from about 16 krypton runs. Other isotopes could not be studied due to memory problems in the mass spectrometer. It is seen

that krypton in the stones is of normal isotopic composition except for a slight excess of  ${\rm Kr}^{86}$  in Richardton.

Turning to the xenon data, we first assume, rightly or wrongly as in the previous Letter,<sup>1</sup> that Xe<sup>136</sup> is an index to the xenon of terrestrial composition in the sample; this is assumed so as to be able to resolve the xenon into a terrestrial and an anomalous component. The isotopic compositions of the anomalous components for Richardton and Murray are compared in Table III. For every isotope other than Xe<sup>129</sup> the ratio of the Murray percent abundance to the Richardton percent abundance is consistent within experimental error with the average value for this quantity. In other words, within experimental error the isotopic composition of the secondary anomalous component-i.e., excluding mass 129-is the same for both meteorites. The anomalous Xe<sup>129</sup> is, on the other hand, very definitely an independent component, in agreement with our assignment of the origin of most of it to extinct I<sup>129</sup> decay.<sup>1</sup>

The secondary anomalous component is more abundant in Murray than in Richardton by a factor 1.24/0.029 = 43. We have already seen that contrasted with Richardton, Murray is poorer by a factor 5 in cosmic-ray-produced nuclides and richer by a factor of at least 42 in a primordial nuclide. We take these ratios to be strong evidence that the secondary anomalous xenon component in the meteorites is primordial in origin-i.e., was incorporated in the stones at the time of their formation. The only alternative possibility, seemingly, is that Murray is uniformly richer in the heavy nuclides serving as cosmic-ray targets for Xe production by a factor  $5 \times 43 = 215$ , which seems most unlikely.

There seems to be no simple explanation for the secondary anomalies. If there is more than one mechanism responsible, it is going to require xenon analyses for other meteorites to distinguish among the mechanisms, since the anomalies in Richardton and Murray are so similar. In the meantime, the following speculative comments are offered.

The xenon in meteorites may have been augmented by nuclear processes between the time it was separated from the xenon now on earth and the time the meteorites were formed. The mass spectrum of the added xenon peaks at mass 131. The contribution at mass 129 is most probably comparable to the nearly equal contributions at masses 128 and 130. This means that  $Xe^{129}$  from I<sup>129</sup> decay is more abundant in Murray than in Richardton by a factor 7.4. If the stones have the same I<sup>127</sup> content, this would imply that Murray cooled 2.9 half-lives of I<sup>129</sup> or 50 million years before Richardton.

On the other hand a strong mass-dependent fractionation may be responsible for most of the anomalies. This is suggested by the regular nature of the isotope abundances of meteoritic xenon when plotted normalized to terrestrial abundances as in Fig. 1. Except at masses 129, 134, and 136, the normalized abundances conform well to a straight line on a logarithmic plot against mass. With this interpretation, there is excess  $Xe^{134}$  and  $Xe^{136}$  in meteoritic xenon which we cannot explain in any really satisfactory manner. If, nonetheless, the mass fractionation is real, Murray is deficient in Xe<sup>129</sup>. This would mean that  $I^{129}$  decay has contributed to  $Xe^{129}$  in the terrestrial atmosphere, as suggested by Katcoff, Schaeffer, and Hastings.<sup>4</sup> For illustrative purposes we assume that the meteorites were all formed 200 million years after the last nuclear synthesizing event. $^{5}$  Then we find from differences between Murray and Richardton that  $(I^{129}/I^{127})$  immediately after the event was 0.0021 and that  $(Xe^{129}/Xe^{132})_{\text{cosmic}}$  is 0.93. This means that about five percent of terrestrial Xe<sup>129</sup> is due to I<sup>129</sup> decay. Repeating the calculation of Katcoff, Schaeffer, and Hastings,<sup>4</sup> we find that the earth was isolated from cosmic xenon 250 million years after the last nuclear synthesis. The 50-million year interval thus found for the period between meteorite and earth formation does



FIG. 1. Isotopic composition of xenon from the meteorites.

not depend upon the 200-million year value assumed for  $(\Delta t)_{meteorites}$ .

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<sup>&</sup>lt;sup>1</sup>J. H. Reynolds, Phys. Rev. Letters  $\underline{4}$ , 8 (1960). <sup>2</sup>H. Hamaguchi, G. Reed, and A. Turkevich, Geo-

chim. et Cosmochim. Acta <u>12</u>, 337 (1957).

<sup>&</sup>lt;sup>3</sup>E. K. Gerling and L. K. Levskii, Doklady Akad. Nauk S.S.S.R. <u>110</u>, 750 (1956).

<sup>&</sup>lt;sup>4</sup>S. Katcoff, O. A. Schaeffer, and J. M. Hastings, Phys. Rev. <u>82</u>, 688 (1951).

 $<sup>{}^{5}</sup>$ G. J. Wasserburg, W. A. Fowler, and F. Hoyle, Phys. Rev. Letters 4, 112 (1960).