

MASS-SPECTROMETRIC DETECTION OF COSMIC-RAY-PRODUCED  $\text{Kr}^{81}$  IN METEORITES  
AND THE POSSIBILITY OF Kr-Kr DATING\*

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$\text{Kr}^{81}$ , the first detected cosmic-ray-produced radioactive nucleus heavier than the Fe-Ni group, was found to be present in meteorites. The radiation age of the Macibini meteorite is calculated from the measured isotopic composition of krypton. This dating method should be applicable to most stone and stony-iron meteorites.

In recent investigations on the isotopic anomalies of meteoritic xenon<sup>1,2</sup> and krypton,<sup>2</sup> the excess concentration of the light isotopes of both rare gases was found to be correlated with the cosmic-ray exposure age of the meteorites. Furthermore, the xenon excess was found to be proportional to the abundances of barium and the rare-earth elements, and the krypton excess proportional to Sr, Y, and Zr. These elements have high production cross sections for Xe and Kr, respectively. Both observations strongly suggest an *in situ* production by recent cosmic-ray bombardment during a time interval given by the radiation age (for definition see Geiss, Oeschger, and Schwarz<sup>3</sup>). If this conclusion is correct, a small fraction of the  $2.1 \times 10^5$ -yr  $\text{Kr}^{81}$  isotope so produced should still be present. A measured isotope ratio of  $\text{Kr}^{81}$  and the spallation yield of a stable krypton isotope in a meteorite would allow one to calculate its cosmic-ray exposure age.

A mass-spectrometric study was made using a 60-deg sector-type instrument of 6-in. radius with a resolving power of about 450 (slit widths of 0.06 and 0.12 mm) and a detection limit of  $5 \times 10^{-15}$  cc STP per isotope. The experimental technique was similar to the one described by Marti, Eberhardt, and Geiss.<sup>2</sup> The Kr peaks could be resolved from the remaining small isobaric hydrocarbon background

in the Kr mass region while a possible interference by the bromine isotope  $\text{Br}^{81}$  could be monitored using mass 79. The extraction blanks were  $0.6 \times 10^{-12}$  cc STP of atmospheric Kr. A calcium-rich achondrite was used in this first investigation, because the elements Sr, Y, and Zr are enriched by about a factor of 7 in this meteorite class.  $\text{Kr}^{81}$  was found to be present in two analyses of 0.5- and 2-g samples. The  $\text{Kr}^{81}$  peak varied with the sample size, but the small isobaric background peak remained constant. A  $\text{Kr}^{81}$  concentration of  $(0.20 \pm 0.03) \times 10^{-12}$  cc STP/g was calculated. The results are given in Table I.  $\text{Kr}^{81}$  is the first cosmic-ray-produced radioactive nucleus heavier than the Fe-Ni group found so far. It is produced by spallation reactions mainly from Sr, Y, and Zr.

The relative production cross section for  $\text{Kr}^{81}$  can be obtained either from target experiments or by interpolating the spallation yields of the stable Kr isotopes. In Table II the calculated spallation yields of the Macibini achondrite are compared with the results from the Stannern achondrite<sup>2</sup> and the measured relative production cross sections from an Ag-target experiment.<sup>4</sup> The agreement is good and shows that neutron capture in Br contributes only little to the  $\text{Kr}^{80}$  and  $\text{Kr}^{82}$  yields.

An estimate from the Kr and Br concentra-

Table I. Isotopic composition and concentrations of Kr and Ar in the Macibini meteorite.

Isotope	Concentration	( $10^{-12}$ cc STP/g)	Isotope	Concentration	( $10^{-8}$ cc STP/g)
$\text{Kr}^{86}$	23 ± 3		$\text{Ar}^{36}$	8.0 ± 0.5	
$\text{Kr}^{78}$	0.388	±0.005	$\text{Ar}^{36}$	1.00	
$\text{Kr}^{80}$	1.288	±0.013	$\text{Ar}^{38}$	1.12	±0.02
$\text{Kr}^{81}$	0.0087	±0.0002	$\text{Ar}^{40}$	172	±8
$\text{Kr}^{82}$	2.31	±0.02	( $\text{Ar}^{38}$ ) <sub>spall</sub>	1.05	±0.02
$\text{Kr}^{83}$	2.85	±0.02			
$\text{Kr}^{84}$	4.32	±0.03			
$\text{Kr}^{86}$	1.00				

Table II. Relative Kr production cross sections in Ca-rich achondrites and in an Ag target.

Sample	Kr <sup>78</sup>	Kr <sup>80</sup>	Kr <sup>81</sup>	Kr <sup>82</sup>	Kr <sup>83</sup>	Kr <sup>84</sup>
Macibini <sup>a</sup>	0.167	0.527	0.61 ± 0.03 <sup>e</sup>	0.754	1.00	0.48
Macibini <sup>b</sup>	0.161	0.515	0.61 ± 0.03 <sup>e</sup>	0.766	1.00	0.68
Stannern <sup>c</sup>	0.179 ± 0.008	0.495 ± 0.020	0.60 ± 0.03 <sup>e</sup>	0.765 ± 0.025	1.00	0.63 ± 0.17
Ag target <sup>d</sup>	0.24	0.51	0.64	0.80	1.00	0.1

(540-MeV protons)

<sup>a</sup>Spallation yields calculated from Table I. (Kr<sup>86</sup>)<sub>trapped</sub>/Kr<sup>86</sup> = 1.0 was assumed.<sup>b</sup>Spallation yields calculated from Table I. (Kr<sup>86</sup>)<sub>trapped</sub>/Kr<sup>86</sup> = 0.846, (Kr<sup>86</sup>)<sub>fission</sub>/Kr<sup>86</sup> = 0.039 {calculated from (Xe<sup>136</sup>)<sub>fission</sub> using Wetherill's data [G. W. Wetherill, Phys. Rev. 92, 907 (1953)] for the spontaneous fission of U<sup>238</sup>}, and (Kr<sup>86</sup>)<sub>spall</sub>/Kr<sup>86</sup> = 0.115 [(Kr<sup>86</sup>/Kr<sup>83</sup>)<sub>spall</sub> = 0.05] were assumed.<sup>c</sup>Ref. 2.<sup>d</sup>Ref. 4.<sup>e</sup>Interpolated value, assuming an isobaric fraction yield of  $g = 0.95 \pm 0.05$ .

tions, using resonance integrals for the epithermal region and the energy interval determined by the large resonances,<sup>2,5</sup> shows that ( $n, \gamma$ ) reactions on Kr<sup>80</sup> contribute very little to the Kr<sup>81</sup> production. For Macibini the contribution is less than 10<sup>-3</sup>%, and even in meteorites with large amounts of trapped Kr and large Kr<sup>80</sup>, Kr<sup>82</sup> anomalies it is less than 1%.

Assuming constant production rates  $P_M$  for the Kr isotopes, the concentration of Kr<sup>81</sup> as a function of time is given by

$$\text{Kr}^{81}(t) = (P_{81}/\lambda_{81})(1 - e^{-\lambda_{81}t}),$$

and the spallation yield of a stable isotope, e.g., Kr<sup>83</sup>, by (Kr<sup>83</sup>)<sub>spall</sub>( $t$ ) =  $P_{83}t$ . Therefore, at the time of fall we expect a ratio

$$\frac{(\text{Kr}^{83})_{\text{spall}}}{\text{Kr}^{81}}(T_r) = \frac{P_{83}}{P_{81}} \lambda_{81} \frac{T_r}{1 - \exp(-\lambda_{81}T_r)}$$

and obtain the radiation age

$$F(T_r) \equiv \frac{T_r}{1 - \exp(-\lambda_{81}T_r)} = \frac{(\text{Kr}^{83})_{\text{spall}}}{\text{Kr}^{81}} \frac{P_{81}}{P_{83}} \frac{1}{\lambda_{81}}.$$

For  $T_r \gg 1/\lambda_{81}$ ,  $F(T_r) \cong T_r$ . For Macibini we found (Kr<sup>83</sup>)<sub>spall</sub>/Kr<sup>81</sup> = 258 ± 10. Using  $P_{81}/P_{83} = 0.61 \pm 0.03$  and  $\lambda_{81} = 3.3 \times 10^{-6} \text{ yr}^{-1}$ ,<sup>6</sup> the radiation age is  $T_r = 48 \pm 4 \text{ Myr}$ .

For comparison, from (Ar<sup>38</sup>)<sub>spall</sub> using Eq. (4) given by Marti, Eberhardt, and Geiss,<sup>2</sup> we obtain  $T_r = 51 \text{ Myr}$ . The reported He<sup>3</sup> concentration<sup>7</sup> indicates a 45% smaller value, but this primarily reflects diffusion losses from the feldspar, as Megrue<sup>8</sup> has shown for other Ca-rich achondrites. More recent results from

other meteorites will be reported elsewhere. This result thus confirms the conclusion that meteoritic spallation krypton is entirely due to the same irradiation that produced He<sup>3</sup>, Ne<sup>21</sup>, and Ar<sup>38</sup> by spallation.

The advantages of Kr-Kr dating of stone and stony-iron meteorites are similar to those of K<sup>41</sup>-K<sup>40</sup> dating of iron meteorites developed by Voshage and Hintenberger<sup>9</sup>: (1) The age determination is based on the isotopic composition of one element. (2) The relative Kr spallation yields are expected to depend only slightly on the energy spectrum. In favorable cases the relative production cross sections can be determined. (3) Diffusion losses are negligible. The Kr-Kr dating method, however, is restricted to meteorites in which the spallation component is not masked by trapped krypton.

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### EXCITED HYPERON OF MASS 1680 MeV\*

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Evidence is presented for a new  $Y_1^*$  of mass 1680 MeV and width 120 MeV. It is produced in  $K^-p$  interactions at 5.5 GeV/c and decays mainly into  $\Lambda\pi^+$  and  $Y_1^*(1385)\pi$ . The decay rate into  $\bar{K}^0p$  is  $0.3 \pm 0.2$  of the rate into  $\Lambda\pi^+$ .

We have analyzed 370 000 pictures taken in the 30-inch hydrogen bubble chamber exposed to the 5.5-GeV/c separated  $K^-$  beam<sup>1</sup> at the Argonne zero-gradient synchrotron (ZGS). Light particle contamination in the beam was  $\leq 5\%$  when calculated from a  $\tau$  scan of the film. A Čerenkov counter operated during the exposure indicated the same high purity. About 5000 two-prong events with an associated  $\Lambda$  decay have been found. After kinematic analysis, these yielded 328 fits to the final state  $\Lambda\pi^+\pi^-$  and 89 fits to  $\Sigma^0\pi^+\pi^-$ .

When more than one hypothesis produced a fit to a given event, a  $\chi^2$  probability cut was used to exclude all fits having a probability 5 times less than that for the best fit. After this selection, all events fitting the hypothesis  $\Lambda\pi^+\pi^-$  with a  $\chi^2$  probability  $>1\%$  were classified as  $\Lambda\pi^+\pi^-$  events. Of the remaining events, all those fitting the  $\Sigma^0\pi^+\pi^-$  hypothesis with a  $\chi^2$  probability  $>1\%$  were classified as  $\Sigma^0\pi^+\pi^-$  events. These criteria yield a rather pure sample of the four-constraint  $\Lambda\pi^+\pi^-$  final state.<sup>2</sup> The two-constraint fits to  $\Sigma^0\pi^+\pi^-$  have some (unknown) contamination from the  $\Lambda\pi^+\pi^-\pi^0$  events, but this uncertainty is not important for the present results. Because of the high-beam purity, contamination due to events originating from pions can be neglected.<sup>3</sup>

The Dalitz plot and the mass-squared projection for the events classified as  $\Lambda\pi^+\pi^-$  are shown in Fig. 1. Strong production of  $Y_1^*(1385)^+\pi^-$  and of  $\Lambda+\rho^0$  can be seen. In addition, a signif-

icant enhancement is observed at  $M^2(\Lambda\pi^+) \sim 2.8$  GeV<sup>2</sup>. For the rest of this Letter, we use the symbol  $Y_1^*(1680)$  for this enhancement. The peak region contains 79 events with an estimated background of 26 events and so is not likely to be a statistical fluctuation. The events in this peak are not associated with the crossing  $\Lambda\rho^0$  and  $\Lambda f^0$  bands, as can be seen from the shaded events in Fig. 1(b), which have  $M^2(\pi^+\pi^-) > 1.75$  GeV<sup>2</sup>. The  $\Lambda\pi^+$  decay mode establishes the isospin to be one.

To rule out an interpretation of this peak as the well-established  $Y_1^*(1660)$  ( $M=1660 \pm 10$  MeV,  $\Gamma=44 \pm 5$  MeV), we show in Fig. 2(e) the  $\Sigma^0\pi^+$  mass distribution from the 89 events fitted to  $\Sigma^0\pi^+\pi^-$ . No  $\Sigma^0\pi^+$  enhancement can be seen, and with a 90% confidence level we obtain an upper limit of 0.25 for the ratio  $[Y_1^{*+}(1680) \rightarrow \Sigma^0 + \pi^+]/[Y_1^{*+}(1680) \rightarrow \Lambda + \pi^+]$ . For the  $Y_1^*(1660)$  this ratio is  $3.4 \pm 1.5$ ,<sup>4</sup> a gross difference from the present result. A small signal for the  $Y_1^*(1660)$  has been observed in this exposure through the decay  $Y_1^*(1660) \rightarrow Y_0^*(1405) + \pi$ .<sup>5</sup> This result, together with the current best estimates of the  $Y_1^*(1660)$ -decay branching ratios,<sup>6</sup> leads to the prediction that only two events of  $Y_1^*(1660)^+$  should be present in our  $\Lambda\pi^+\pi^-$  sample. We therefore conclude that the observed  $\Lambda\pi^+$  enhancement is not due to the  $Y_1^*(1660)$ .

Figure 2(f) shows the  $\bar{K}^0p$  mass distribution for the 473 events satisfying the hypothesis  $K^- + p \rightarrow \bar{K}^0 + p + \pi^-$  on this film. Only a small enhancement is seen in the 1680-MeV mass