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# Identification of Very Heavy Cosmic-Ray Tracks in Meteorites

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Ions of Fe, Br, and I from the Oak Ridge tandem van de Graaff accelerator have been used to form charged-particle tracks in crystals of the minerals hypersthene and diopside, two of the most common minerals in meteorites. The results establish that the thresholds for track formation in these minerals, like those previously measured in more sensitive track-storing materials, are consistent with a primary specific ionization criterion. The direct calibration obtained for Fe ions confirms that the most abundant cosmic-ray tracks found in meteorites are due to nuclei of the Fe group ($Z=20$ to 30). Together with the use of the primary ionization criterion, this calibration provides improved relations between observed track length and atomic number of slowing-down cosmic-ray ions in hypersthene and diopside. The minimum atomic numbers for track formation are $Z=25$ for hypersthene and $Z=24$ for diopside. Fe nuclei should leave tracks up to $\sim 8\ \mu$ long in hypersthene and up to $\sim 15\ \mu$ long in diopside. From the present work and a recent study of cosmic-ray tracks in meteorites, we calculate that the abundance of cosmic rays with $Z>30$, relative to the abundance of Fe-group nuclei, is 1/4550.

## I. INTRODUCTION

It has been known for almost 20 years[1,2] that cosmic radiation consists primarily of energetic nuclei with atomic number $Z$ ranging from 1 up to at least 26 (Fe), but because of their extreme rarity it has only recently been established that cosmic rays considerably heavier than Fe exist. The first evidence for the existence of extremely heavy cosmic rays ($Z$ appreciably $>26$) was provided by studies of fossil particle tracks in meteorite crystals,[3] followed soon after by observations in emulsion stacks of unusually large area.[4] It is expected that detailed studies of abundances of individual nuclei by these two detection techniques will yield valuable information about the origin and propagation of extremely heavy cosmic rays. In addition the fossil tracks can be used to investigate the long-term time dependence of the charge spectrum.

Because of an inadequate knowledge of the response of meteoritic crystals to heavily ionizing particles, it was possible in the first study of fossil cosmic-ray tracks to set only very crude limits on the charge distribution of the very heavy nuclei.[3] In the present paper we describe new calibrations obtained by bombarding various meteoritic minerals with ions of Fe, Br, and I with energies up to $\sim 70$ MeV. These calibrations establish that the majority of cosmic-ray tracks in meteorites are produced by Fe ions. They also allow us to derive a more accurate relation between track length and atomic number than was previously available, thus making possible a more definite identification of the tracks of ions heavier than Fe observed in meteorites.[3]

## II. REGISTRATION OF COSMIC-RAY TRACKS IN METEORITES

Particle tracks in dielectric solids[5,6] consist of linear regions which are so intensely radiation-damaged that they can be preferentially dissolved by a suitable chemical reagent. In many of the useful meteoritic minerals, solutions of KOH or NaOH at ≥200°C are used to "develop" tracks.

Extensive calibration studies of several solids have shown that a "latent track" (one that is optically invisible but which can be revealed or "developed" by etching) is formed wherever the primary ionization rate $J$ of a slowing-down particle exceeds a critical value $J_c$ that is characteristic of the particular solid.[7] In such solids as mica, glasses, and various plastics, the critical ionization rates are sufficiently low that they can be determined without difficulty by calibration bombardments with ions available in the linear accelerators at Berkeley and Yale. Abundant meteoritic minerals such as olivine, hypersthene, and diopside are more resistant to ionizing radiation and have critical ionization rates that can be exceeded only by particles considerably heavier than Ar ions, which are the heaviest ions presently available in the Yale and Berkeley linear accelerators. This low sensitivity is, in fact, the reason meteorite crystals that have been exposed to the cosmic radiation for as long as $10^8$ years are not completely "fogged" by tracks, but instead record only the last

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[1] P. Freier, E. J. Lofgren, E. P. Ney, and F. Oppenheimer, Phys. Rev. **74**, 1818 (1948).

[2] H. L. Bradt and B. Peters, Phys. Rev. **74**, 1828 (1948).

[3] R. L. Fleischer, P. B. Price, R. M. Walker, M. Maurette, and G. Morgan, J. Geophys. Res. **72**, 355 (1967).

[4] P. H. Fowler, R. A. Adams, V. G. Cowen, and J. M. Kidd, Proc. Roy. Soc. (London), **A301**, 39 (1967).

[5] R. L. Fleischer, P. B. Price, and R. M. Walker, Ann. Rev. Nucl. Sci. **15**, 1 (1965).

[6] R. L. Fleischer, P. B. Price, and R. M. Walker, Science **149**, 383 (1965).

[7] R. L. Fleischer, P. B. Price, R. M. Walker, and E. L. Hubbard, Phys. Rev. **156**, 353 (1967).

TABLE I. Measured track lengths $L$ of heavy ions in minerals.

| Ion | Energy (MeV) | $R$ (range in diopside or hypersthene[a]) ($\mu$) | $L$ (hyp.) | | $L$ (diop.) | | $R$ (range in mica[f]) ($\mu$) | $L$(mica) ($\mu$) | Average $R-L$ ($\mu$) |
|---|---|---|---|---|---|---|---|---|---|
| | | | b | c | d | e | | | |
| I | 60.0 | 8.0 | $3.5\,\mu$ | | $4.0\,\mu$ | | 8.3 | 7.2 | Hyp: 4.1 |
| I | 29.4 | 5.3 | 1.5 | *bkgd*[g] | 1.6 | | 5.2 | 4.0 | Diop: 3.5 |
| I | 21.6 | 4.3 | | *bkgd*[g] | 1.0 | | 4.3 | 2.8 | Mica: 1.2 |
| I | 15.0 | 3.4 | 0 | | 0 | $<0.5$ | 3.4 | 2.2 | |
| Br | 60.0 | 8.0 | 3.4 | 3.4 | 4.2 | 4.2 | 8.3 | 7.1 | Hyp: 4.6 |
| Br | 29.4 | 5.3 | $<1\,\mu$ | *bkgd*[g] | 1.0 | 1.5 | 5.2 | 3.5 | Diop: 3.5 |
| Br | 21.6 | 4.3 | 0 | 0 | 0.7 | 0.7 | 4.3 | 2.7 | Mica: 1.6 |
| Br | 15.0 | 3.4 | 0 | 0 | ··· | $<0.5$ | 3.4 | 1.5 | |
| Fe | 69.0 | 9.8 | 5.0 | 5.6 | | 5.8 | 9.9 | 8.5 | |
| Fe | 62.5 | 9.3 | 4.2 | 4.8 | | 4.5 | 9.1 | 7.5 | Hyp: 4.8 |
| Fe | 54.0 | 8.2 | | 2.8 | | 3.2 | 8.2 | 7.3 | |
| Fe | 41.3 | 6.9 | 2.2 | 2.2 | | 3.0 | 6.7 | 5.7 | Diop: 4.0 |
| Fe | 30.4 | 5.6 | | 1 | 1.4 | 2.1 | 5.5 | 4.2 | |
| Fe | 21.1 | 4.4 | 0 | 0 | 0.5 | 0.8 | 4.3 | 3.5 | Mica: 1.2 |
| Fe | 13.5 | 3.2 | 0 | 0 | | $<0.5$ | 3.2 | 2.0 | |

[a] Ranges in hypersthene and diopside were calculated for Br and I from Ref. 16 and are essentially the same for each solid and for both ions at the same energy (not the same velocity). Data for use in the relations given in Ref. 16 were obtained from Ref. 17. Reference 17 gives the same ranges for Br ions as Ref. 16 and was therefore used for ranges of Fe ions.

[b] Hypersthene from Estherville, annealed so that fossil tracks were erased.

[c] Hypersthene from Estherville, containing a background of fossil tracks.

[d] Clear diopside from Calif., with low background of fossil tracks.

[e] Dark green diopside from Madagascar, with low background of fossil tracks.

[f] Ranges of Fe and Br in mica were taken from Ref. 17. Since I ranges were not tabulated, we assumed $R(E)$ was the same for Br and I.

[g] If tracks were formed, they are not discernible above the background features.

few microns of the ranges of slowing-down heavy nuclei.

## III. PREVIOUS EVIDENCE ON THE THRESHOLD FOR TRACK REGISTRATION IN METEORITIC MINERALS

Reasoning based entirely on previous work[8] suggested very strongly, but did not prove, that the critical ionization rate $J_c$ for common meteoritic crystals has a value such that Fe ions form tracks[9] in these crystals over a distance of a few microns near the end of their range, where $J$ is highest. As a consequence, it was inferred that the majority of the cosmic-ray tracks observed in meteorites,[3] those with lengths up to $\sim 8\,\mu$, were made by Fe ions, whereas the few tracks ($<10^{-3}$ of the total) longer than $8\,\mu$ were made by ions heavier than Fe.

A lower limit for $J_c$ was set by noting that ions with $Z<18$ do not create tracks at any energy[8]; an upper limit for $J_c$ was set by noting that if the cosmic-ray tracks observed[3] in meteorites were due to ions with $Z>30$, their flux would be so great that they should have been detected in previous cosmic-ray studies using nuclear emulsions.[10,11] Since the most abundant charge of cosmic rays in the range $18<Z\leq 30$ corresponds to Fe,[12] these nuclei were inferred to be the major contributor to the abundant cosmic-ray tracks with lengths up to $\sim 8\,\mu$ that were observed in meteorites.

Registration experiments using spallation recoil nuclei from Ag, Nb, Zn, Cu, and Fe and fragments from $\mathrm{Cf}^{252}$ spontaneous fission gave results that were consistent with the hypothesis of Fe tracks $\sim 8\,\mu$ in length, but because of the wide range of masses and energies and unknown effective charges at low energy, no definitive threshold could be calculated.

Price *et al.*[13] and Cantelaube *et al.*[14] have recently found that the absolute values and depth dependence of the cosmic-ray track densities in two meteorites are consistent with their having been formed by Fe nuclei, but their observations were not sufficiently restrictive to further narrow the permissible values of $Z$ between $\sim 18$ and $\sim 30$.

The new experiments to be presented directly confirm that Fe nuclei can form tracks in meteoritic crystals. By using the new effective-charge data of Cumming

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[8] R. L. Fleischer, P. B. Price, R. M. Walker, and M. Maurette, J. Geophys. Res. **72**, 331 (1967).

[9] Tracks are defined here as linear, etchable features of length $\sim 1\,\mu$ or greater.

[10] This inference assumes a constant cosmic flux in the past within a factor of 2, an assumption for which there is evidence.

[11] J. R. Arnold, M. Honda, and D. Lal, J. Geophys. Res. **66**, 3519 (1961).

[12] For the most recent and comprehensive measurements of the composition of heavy cosmic rays, see O. Mathiesen, C. E. Long, P. S. Freier, and C. J. Waddington, paper OG-VII-56 in Proceedings of the Tenth International Conference on Cosmic Rays, Can. J. Phys. (to be published).

[13] P. B. Price, R. S. Rajan, and A. S. Tamhane, J. Geophys. Res. **72**, 1377 (1967).

[14] Y. Cantelaube, M. Maurette, and P. Pellas, *Proceedings of the Conference on Radioactive Dating and Methods of Low Level Counting* (International Atomic Energy Agency, Vienna, 1967), p. 215.

and Crespo,[15] we can show that a primary ionization rate criterion fits the track-formation behavior (as had been found previously for mica and plastics[7]). From this criterion and the observed threshold ionization rate for Fe, Br, and I ions we will compute an improved relation between track length and atomic number with which to infer the heavy cosmic-ray composition.

## IV. BOMBARDMENTS WITH BEAMS OF Fe, Br, AND I IONS

Fifteen identical sets of samples were mounted on a rotatable target wheel and exposed in the Oak Ridge tandem van de Graaff accelerator to beams of Fe, Br, and I ions of various energies. Large, flat surfaces of the samples were oriented at 45° to the beam. The following minerals were irradiated: (1) Muscovite mica; (2) dark green diopside from Madagascar; (3) clear diopside from California; (4) hypersthene from the Estherville stony iron meteorite, containing fossil cosmic ray tracks; and (5) hypersthene from the same meteorite, annealed so that tracks were erased.

Estimates of ion doses were made by exposing a silicon fission-particle detector to the beam just before and just after an irradiation of a set of samples and averaging the two counting rates. The target holder was sufficiently far from the final quadrupole magnet so that uniform doses of $10^5$ to $10^6$ ions/cm$^2$ were achieved over an area of about 2 cm$^2$. Final measurements of doses were made by counting the tracks in the mica samples, which recorded ions with 100% efficiency.

In normal operation the beam of ions emerges from the stripper gas canal of the tandem accelerator with a continuous spectrum of energies peaked at a value that depends on the accelerating voltage $V$. A bending magnet allows only ions with a discrete set of energies to reach the target. These energies $E$ depend on the charge states $q$ through the relation

$$ME/q^2 = kH^2, \tag{1}$$

where $H$ is the field of the bending magnet, $M$ is the mass of the ion, and $k$ is a constant. By suitably adjusting $H$ and $V$ we could usually arrange for the majority of the ions to have the desired charge state and energy, with the remainder of the ions having a lower charge state and energy. We could then attribute the longest tracks in the samples to the desired ions. Pulse-height spectra were recorded immediately before and after each exposure to make sure that the beam of interest was the most intense one during the exposure.

The production and identification of beams of $^{79}$Br, $^{81}$Br, and $^{127}$I ions of any desired energy are straightforward.[16] $^{56}$Fe ions had never before been successfully accelerated. Working with us, Wells of the accelerator staff succeeded in producing FeCl$^-$ ions in the accelerator ion-source exchange canal by bombarding FeCl$_2$ with He ions. A 20° bending magnet ensured that only FeCl$^-$ ions reached the accelerator. After the first stage of acceleration, the FeCl$^-$ beam was stripped into multiply charged Fe and Cl ions, which were then accelerated along the stripper gas canal. From the spacing of the various charge states in the pulse height spectra and from Eq. (1), it was possible to select Fe ions of the desired energy from the melange of beams of various impurities that passed through the 90° bending magnet.

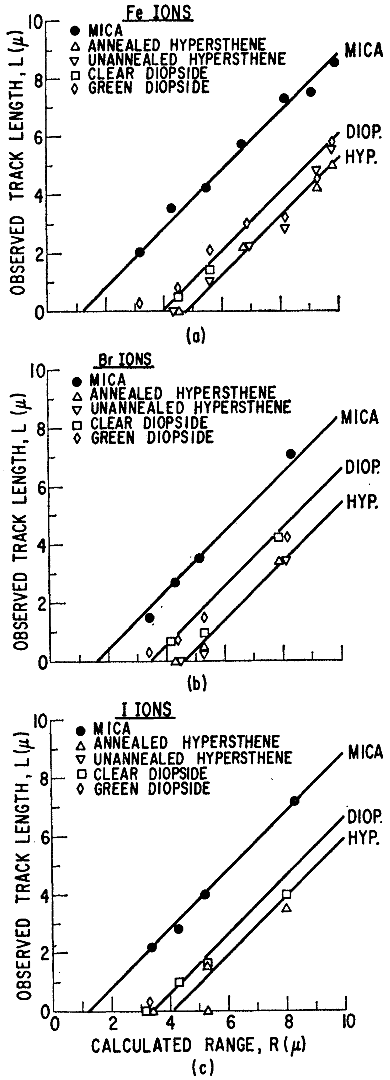

FIG. 1. Measured etched track lengths as a function of calculated range for (a) Fe ions, (b) Br ions, and (c) I ions in several solids. The uncertainty in measured length is ~±0.5 μ and in calculated range <±0.5 μ.

## V. EXPERIMENTAL RESULTS

Table I gives the energies of the major charge groups for the 15 bombardments and the calculated ranges of

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[15] J. B. Cumming and V. P. Crespo, Phys. Rev. **161**, 287 (1967).

[16] C. D. Moak, J. H. Neiler, H. W. Schmitt, F. J. Walter, and G. F. Wells, Rev. Sci. Instr. **34**, 853 (1963).

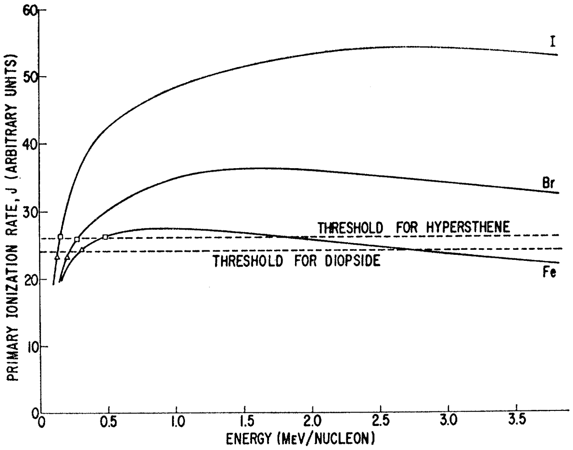


FIG. 2. Relative primary ionization rates of Fe, Br, and I ions in hypersthene and diopside, calculated from Eq. 3 with $K=9.7$. For hypersthene we observe that Fe tracks are formed above 0.48 MeV/$N$, and from the curve we conclude that track formation should cease above 1.77 MeV/$N$. The range over which Fe tracks can be formed is calculated to be 7.6 $\mu$. In diopside the limiting energies are 0.29 and 2.7 MeV/$N$ and the range over which Fe tracks can be formed is calculated to be 15 $\mu$.

the most energetic group in each bombardment. The ranges of Fe, Br, and I ions in hypersthene, diopside, and mica were obtained from the empirical relation of Bridwell and Moak[17] or from the range-energy tables of Henke and Benton.[18] The observed maximum lengths of the etched tracks are given in the table and are also plotted in Fig. 1 as a function of calculated range. One can see that in every case the observed etched track length $L$ is less than the calculated range $R$ by an amount that is approximately constant for a given ion in a given solid, independent of bombarding energy. The reason for this difference between calculated and observed lengths is that, near the end of the range of a slowing-down heavy particle, its ionization rate drops below the critical rate for track formation. Values of the difference between calculated and observed ranges, $R-L$, appear in the last column of Table I.

We note that clear and green diopside appear to have practically the same sensitivity. Annealing does not appear to change the sensitivity of hypersthene.

## VI. IONIZATION CURVES AND CRITICAL IONIZATION RATES FOR TRACK FORMATION

If the primary ionization rates were known as a function of velocity, we could use the residual ranges at which slowing-down I, Br, and Fe ions cease forming etchable tracks (last column of Table I) together with range-energy curves, to calculate $J_c$ for each ion and learn whether the same value results for each ion. Unfortunately, values of primary ionization rate for ions in solids are not known. However, Fleischer *et al.*[7] recently showed that in the case of mica, Lexan polycarbonate, and cellulose nitrate, the equation of Bethe[19] for primary ionization rates in atomic hydrogen correctly predicts the dependence of track formation on atomic number and velocity of the slowing-down ion, provided a physically realistic choice is made of the constant $I_0$, which represents the energy required to eject the most loosely bound electrons in the medium. The primary ionization rate, according to Bethe,[19] is

$$J=(aZ_e^2/\beta^2 I_0)\{\ln[2mc^2\beta^2/(1-\beta^2)I_0]+3.04-\beta^2\}, \quad (2)$$

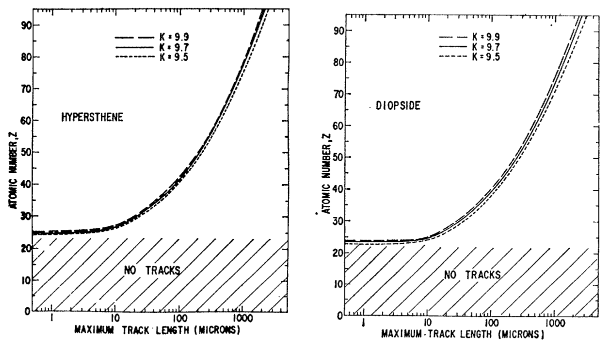
where $Z_e$ is the effective charge of the ion, $c$ is the velocity of light, $c\beta$ is the velocity of the ion, $m$ is the electron mass, and $a$ is a constant. All track registration data for mica were correlated with $J$ (i.e., tracks form when $J>J_c$ and do not form when $J<J_c$) when $I_0$ was chosen to be $\sim$13 eV and all data for Lexan polycarbonate and cellulose nitrate were correlated with $J$ when $I_0$ was taken to be $\sim$2 eV.

Actually, McClure[20] has found experimentally from measurements of $J$ in $H_2$, He, Ne, and Ar that the constant 3.04 in Eq. (2) applies only for atomic hydrogen. He rewrote Eq. (2) in terms of a new constant, $K=b+\ln(2mc^2/I_0)$:

$$J=(a'Z_e^2/\beta^2)\{\ln[\beta^2/(1-\beta^2)]+K-\beta^2\}. \quad (3)$$

He found values of $K$ ranging from 8.53 for Ne to 12.2 for $H_2$ (with $K$ for atomic hydrogen equal to 14.3). In terms of $K$, the constant for mica is 14.2, and for the two plastics $K$ is 16.1.

In the present paper we will choose a value of $K$ that yields curves of $J$ versus $\beta$ such that the value of $J_c$ is the same for I, Br, and Fe ions in a particular solid.[21]



(a) (b)

FIG. 3. Dependence of maximum etched track length on atomic number of slowing-down nucleus in (a) hypersthene and (b) diopside. The derivation of this curve is described in the text.

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[17] L. B. Bridwell and C. D. Moak, Phys. Rev. **156**, 242 (1967).

[18] R. P. Henke and E. V. Benton, U.S. Naval Radiological Defense Laboratory Report No. TR-1102, 1966 (unpublished).

[19] H. A. Bethe, Ann. Physik **5**, 325 (1930).

[20] G. W. McClure, Phys. Rev. **90**, 796 (1953).

[21] Values of $J$ in different solids with different values of $K$ can be directly compared only if $J$ is expressed in terms of some standard ion at a standard velocity; for example, a proton at the ionization minimum or maximum.

We will see that, for the right choice of $K$, Eq. (3) shows that the maximum etchable length $L$ of the track from an iron ion in hypersthene is consistent with the observed values of $\sim 8\,\mu$ reported for the vast majority of cosmic-ray tracks in meteoritic hypersthene.[3] Although this approach is empirical, we justify it on the grounds that the ion explosion spike model of track formation in inorganic materials predicts a primary ionization rate criterion,[22] that previous measurements are fully consistent with that criterion,[7] and that this procedure leads to no inconsistencies.

To evaluate (3) we need to know how the effective charge of an ion depends on velocity. Heckman *et al.*[23] found that an empirical expression for the effective charge,

$$Z_e = Z[1 - \exp(-125\beta/Z^{2/3})], \qquad (4)$$

fitted the data available at that time, which included ions with $Z$ up to 18. Cumming and Crespo[15] have recently measured values of effective charges for I and Br ions that are lower than would have been predicted by the Heckman relation. For I and Br we used the values measured by Cumming and Crespo, and for Fe we used Eq. (4).

We found that for $K \approx 9.7$ the value of $J_c$ deduced from the data in Table I was closely the same for I, Br, and Fe ions in hypersthene. The value of $J_c$ for diopside (which was lower than that for hypersthene) was also constant for each of the three ions if we used the same value, $K \approx 9.7$. Figure 2 gives curves of $J$ versus the energy per nucleon $E/N$, calculated for I, Br, and Fe with $K = 9.7$. The data points are positioned on the ionization rate curves at energies corresponding to residual ranges where track formation stops. If $K$ is taken as large as 9.9, $J_c$ becomes considerably higher for I than for Br and higher for Br than for Fe. If $K$ is taken as small as 9.5, the situation is reversed and $J_c$ is highest for Fe and lowest for I. We shall adopt the value $K = 9.7 \pm 0.2$, with the values 9.5 and 9.9 representing the extreme values that can possibly be consistent with the present data. At this point it should be noted that the value of $K$ required to convert Eq. (3) into the Bethe-Bloch equation for *total energy loss* in hypersthene or diopside is 8.8. A total-energy-loss criterion for track formation in these solids is thus in conflict with observation. It has been pointed out previously,[22] but should be reemphasized here, that a model for track formation in terms of nuclear stopping or displacement spikes is also incompatible with observation since the maximum energy loss in hard-sphere collisions occurs at only $\sim 0.8$ keV/nucleon for Fe ions. At this energy the range is undetectable optically.

TABLE II. Comparison of old and new values of $Z$ for certain track lengths in hypersthene.

| Etched track length ($\mu$) | Old $Z$ | New $Z$ |
|---|---|---|
| 15 | 30 | 28 |
| 20 | 32 | 30 |
| 50 | 42 | 36 |
| 100 | 52 | 41 |
| 200 | 64 | 49 |
| 500 | 84 | 64 |

## VII. TRACK LENGTH AS A MEASURE OF ATOMIC NUMBER OF HEAVY COSMIC RAYS

Using $K = 9.7 \pm 0.2$ in Eq. (3), we have calculated the intervals of velocity over which $J$ exceeds $J_c$ for various ions in both hypersthene and diopside. From the appropriate range-energy relations we have converted velocity intervals into range intervals. The results (for $K = 9.5$, 9.7, and 9.9), displayed in Fig. 3, allow us to relate observed etched track lengths in meteoritic crystals to atomic numbers of slowing-down cosmic rays. For Fe ions in hypersthene, we get $L = 7.6$, 5.9, or 8.9 $\mu$, depending on whether we use $K = 9.7$, 9.9, or 9.5. The preferred value, $L = 7.6\,\mu$, agrees well with the value 8 $\mu$ experimentally observed in meteorites as the maximum track length of the majority of cosmic-ray tracks.

From Fig. 3 we also see that the minimum atomic number recordable in hypersthene is $Z = 25$ and diopside is $Z = 24$. Fe ions in diopside should be capable of forming tracks up to $\sim 15\,\mu$ long. This prediction should be testable in the near future.

These new relations between $Z$ and $L$, being based on direct measurements on Fe, Br, and I ions, are considerably more reliable than the curves shown in Fig. 7 of the paper by Fleischer *et al.*[3] Those curves were obtained by assuming that for Fe ions $L = 8\,\mu$ in hypersthene and 12 $\mu$ in another mineral (bytownite) and by assuming that the value $K = 14.2$ found for mica also holds for hypersthene and bytownite. The much lower value used for $K$ in the present work leads to a smaller dependence of $Z$ on $L$. The old and new values of $Z$ deduced for several track lengths in hypersthene are given in Table II. The difference in the value of $Z$ predicted by the old and new relations increases with increasing track length. The result of varying $K$ between the limits 9.5 and 9.9 is to change the predicted atomic number by no more than 1 unit of charge for all but the very heaviest nuclei.

Measurements of track-length distributions in meteorites were used by Fleischer *et al.*[3] to deduce that the ratio of cosmic rays with $Z = 20$ to 30 relative to those with $Z \gtrsim 32$, averaged over the last 50 million years, was $\sim 4550$. This ratio was derived by attributing all

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[22] R. L. Fleischer, P. B. Price, and R. M. Walker, J. Appl. Phys. **36**, 3645 (1965).

[23] H. H. Heckman, B. L. Perkins, W. G. Simon, F. M. Smith, and W. M. Barkas, Phys. Rev. **117**, 544 (1960).

tracks with length $>20\ \mu$ to cosmic rays with $Z>32$. Using the new relation between $Z$ and $L$, we find that their ratio 4550 should apply to the number of cosmic rays with $Z=20$ to 30 relative to those with $Z\gtrsim 30$.

## VIII. CONCLUSIONS

1. Fe ions form tracks in hypersthene at energies between $\sim$0.48 MeV/$N$ and some as yet undetermined higher energy; Fe ions form tracks in diopside between $\sim$0.29 MeV/$N$ and some undetermined higher energy. It will be very important experimentally to determine the high-energy threshold at which track formation ceases. We estimate that this energy is $\sim$1.8 MeV/$N$ for hypersthene and $\sim$2.7 MeV/$N$ for diopside, energies which are now attainable in the newest tandem van de Graaff accelerators.

2. Diopside is somewhat more sensitive than hypersthene. It should record tracks of ions as light as Cr. Tracks of Fe ions should have lengths up to $15\ \mu$ in diopside.

3. A primary ionization rate model of track formation fits the observations of Fe, Br, and I ion tracks in hypersthene and diopside, and this description is consistent with the observations of fossil cosmic-ray tracks in meteorites, provided the constant $K=9.7\pm0.2$ is used in Eq. (3).

4. A new relation between track length and atomic number has been derived which should be used to interpret future observations of cosmic-ray tracks in meteorites until further registration data can be obtained at higher energies and for other ions than Fe, Br, and I.

## ACKNOWLEDGMENTS

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# Irreversibility in Paramagnetic Spin Systems: Free Induction Decay and Spin Diffusion

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In a recent work, the authors derived kinetic equations for the spin autocorrelation function for a paramagnetic spin system using the resummation procedure introduced by Résibois and De Leener in the framework of the statistical mechanics of irreversible processes due to Prigogine and co-workers. These equations are non-Markovian and nonlinear in the high-field, high-temperature, and Weiss-limit approximations. In the present paper, methods of approximation are given to solve such kinetic equations and are applied to the study of two important NMR problems, namely, free induction decay (FID) and spin diffusion. The general characteristics of the FID are obtained even in the lowest order of approximation owing to the resummation procedure, whereas the next higher-order correction leads to very good agreement with the experimental results given by Barnaal and Lowe. The following asymptotic form is also derived:

$$\Gamma(t)=(a\cos\alpha t+b\sin\alpha t)e^{-\beta t}.$$

A diffusion equation is obtained for the magnetization. From this the diffusion coefficient is computed and is found to be in agreement with that proposed by several authors. However, consideration of higher-order corrections does not seem to explain the strong dependence on the orientation of the external magnetic field which was observed experimentally by Leppelmeier.

## 1. INTRODUCTION

In a recent work[1] (hereafter I), the authors derived kinetic equations for the two-spin autocorrelation functions in the case of paramagnetism in the high-field, high-temperature approximations. The method used was an extension of that of Résibois and De Leener[2,3] (hereafter RDL), based on the general theory of nonequilibrium statistical mechanics developed by Prigogine and co-workers.[4] They treated the case of a Heisenberg spin system by reorganizing the perturbation expansion of the spin autocorrelation function so that the final equations involve only quantities which

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[1] P. Borckmans and D. Walgraef, Physica **35**, 80 (1967).

[2] P. Résibois and M. De Leener, Phys. Rev. **151**, 305 (1966).

[3] M. De Leener and P. Résibois, Phys. Rev. **151**, 318 (1966).

[4] I. Prigogine, *Non-Equilibrium Statistical Mechanics* (Interscience Publishers, Inc., New York, 1962).