A Study of the Registration of Arsenic and Iodine Ions in Olivine and Hypersthene Crystals

P. Horn, M. Maurette *, and W. von Oertzen

Max-Planck-Institut für Kernphysik, Heidelberg, Germany

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The registration of the trajectories of arsenic and iodine ions in olivine and hypersthene crystals, considered as "track chambers", is reported; these trajectories are revealed by chemical etching and observed as tracks in an optical microscope.

A brief discussion concerning the different criteria which have been proposed to explain the registration of heavy ion trajectories in solids is given. It is shown that the criterion which is presently accepted is limited in its application because of the uncertainty of two basic parameters in the equation which is used.

Since the work of Young [1], Silk and Barnes [2] and Price and Walker [3], it has been recognized that most solid insulators can be used as selective "track chambers" capable of registering the trajectories of heavy ions of appropriate masses and energies in the form of chemically etchable tracks.

It would be very interesting to assign to each of these solids the following characteristics: a) a critical nuclear charge, Z_c, below which the ions are not registered whatever their energies may be; b) the values of the range of energies, ΔE, over which an ion of nuclear charge $Z_i > Z_c$ gives an observable track. The knowledge of these characteristics for solids of different sensitivities would allow a better understanding of the nature of the damage that produces an "etchable track", and a better utilization of the fossil tracks recently observed in some constituent minerals of meteorites — tracks which are attributed to nuclei of the VH-group of primary cosmic radiation [4].

This paper considers the validity and the limits of application of different criteria proposed during the last few years to determine the values of Z_c and ΔE in solids. We have also attempted to apply the "best" of these criteria to deduce the values of Z_c in two minerals which are common in meteorites and which have not been calibrated until the present work by means of artificially accelerated heavy ions.

I. Experimental Results

Samples of olivine and hypersthene were exposed to a flux of accelerated arsenic and iodine ions. The crystals were then etched to reveal the tracks. By decreasing the energy of the ions, it was possible to measure the critical energy, E_c, below which the ions no longer produced etchable tracks. Then, by using methods which will be described later, we tried to deduce from E_c the value of the critical nuclear charge, Z_c.

The As and I beams were obtained from the Heidelberg Tandem Van de Graaff accelerator by single stripping of the ions in the terminal. For the determination of the beam energy, a surface barrier counter was used. The counter was calibrated with ^{16}O ions of 15, 20 and 25 MeV, scattered at a definite angle from a gold target. The energy of the As and I ions can then be derived with an accuracy of ± 1 MeV (As) and $^{+3}_{-1}$ MeV (I). The pulse height defect [5] between ^{16}O ions and As ions should be negligible, but for the I-ions a difference of $+3$ MeV relative to the ^{16}O calibration can be expected.

Because the beam was quite inhomogeneous and had only a small diameter, a special irradiation technique was employed:

a) Before each irradiation, we put into the sample holder a "positioner" sheet of glass which was placed normal to the beam and then etched 40

* Laboratory for Space Physics, Washington University, St. Louis, Missoure; currently on leave from: Laboratoire de Chimie-Physique, Faculté des Sciences de Paris, Centre d'Orsay, France.

[1] D. A. Young, Nature **182**, 375 [1958].
[2] E. C. H. Silk and R. S. Barnes, Phil. Mag. **4**, 970 [1959].
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seconds in 48% hydrofluoric acid. Tracks were always revealed, because the glass was sensitive to all of the ions we were using. It was possible to see the entire spot with the naked eye.

b) The crystal to be studied was then fixed at the proper place and covered with a thin sheet of mica in such a way that the edge XX′ of the mica covered half of the visible spot on the glass (Fig. 1). The positioner lamella was put back in the same position in the holder and irradiated at two angles of incidence: 30° and 90°.

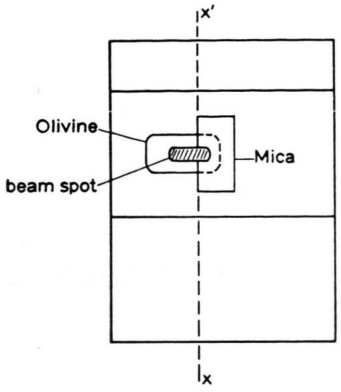

Fig. 1. Position of the beam spot relative to the position of the probes.

We measured the following quantities for each of the energies used: a) the value of the ratio R between the density of tracks in the sample to be studied and in the mica (the counting of the tracks was done as closely as possible to the XX′ edge in order to reduce any effects due to inhomogeneities of the beam); b) the value of the ratio L between the lengths of the tracks in the samples and in the mica. It was known from other studies that the mica was sufficiently sensitive to register with unit efficiency the tracks of all the ions we have used. Therefore, it was expected that the values of R and L would decrease rapidly as E approached E_c.

The experimental results are given in Table 1. It can be seen that, in olivine, tracks are not registered for 12 MeV $< E$(As) $<$ 16 MeV, and 20 MeV $< E$(I) $<$ 40 MeV. In hypersthene, the value of E_c for iodine ions must be near 18 MeV; however, the beam was so inhomogeneous during this irradiation that we cannot be sure that the observed decrease of R is real. We are only certain that hypersthene is more sensitive than olivine. Fig. 2 ** gives an example of tracks of 20 MeV As ions in mica and in olivine.

Type and Energy (in MeV)		R	L
	26	1	0.64
$^{33}_{75}$As	20	1	0.55
	16	0.70	0.30
$^{53}_{127}$I	40	1	0.61
	18	0	0

Table 1.

II. Discussion of the Results

The mechanism of damage which leads to the formation of etchable tracks in insulators is poorly understood. The formation of such tracks was first attributed to a displacement spike [2], then to a thermal spike [6] and, more recently, to an ion explosion spike by Fleischer et al. [7] and Maurette [8]. This latest model explains the greatest number of experimental facts. The succession of models has led to modifications of the criteria proposed to predict, whether an ion of given energy and mass can give an etchable track.

After a brief description of these criteria we specify in the next section the limits of application of the one recently proposed by Fleischer et al. [9]. To illustrate these limitations we discuss the application of the present experimental work to the problem of detecting tracks due to nuclei of the VH-group of the primary cosmic radiation in olivine crystals of meteoritic origin.

II.1. Description of the criteria

As a consequence of the work done by Fleischer et al. [10] it was first thought that only ions with an average energy loss per unit of path length greater than a critical value, $(dE/dX)_c$, could be registered. In Fig. 3, we plotted the curves which give, for ar-

** Fig. 2 on p. 1794 a.

[6] G. Bonfiglioli, A. Ferro, and A. Mojoni, J. Appl. Phys. **32**, 2499 [1961].

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

[8] M. Maurette, J. Phys. Radium **27**, 505 [1966].

[9] R. L. Fleischer, P. B. Price, R. M. Walker, and H. Hubbard, Phys. Rev. **156**, 353 [1967].

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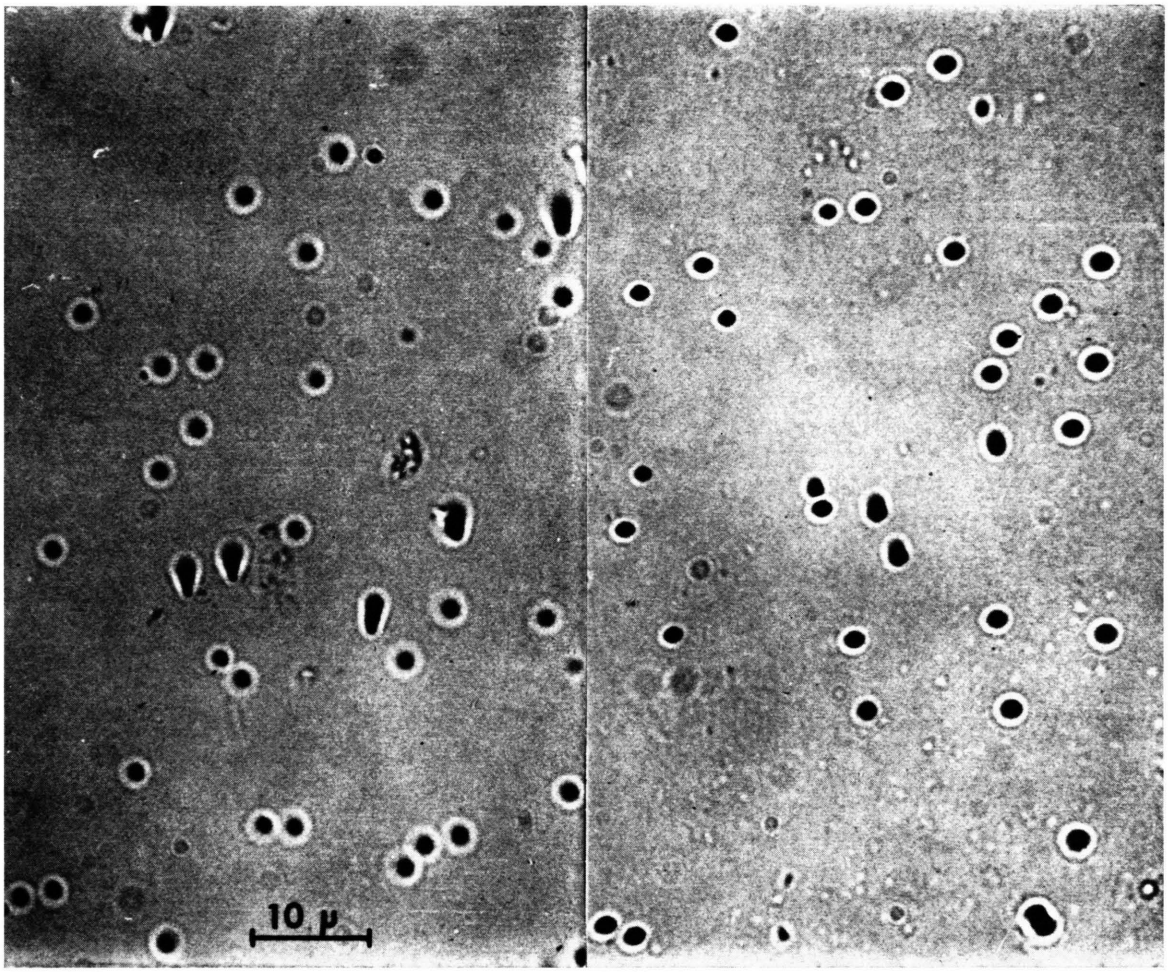

Fig. 2. Tracks of 20 MeV arsenic ions in olivine (right) and mica (left). Angle of incidence 30° and 90°.

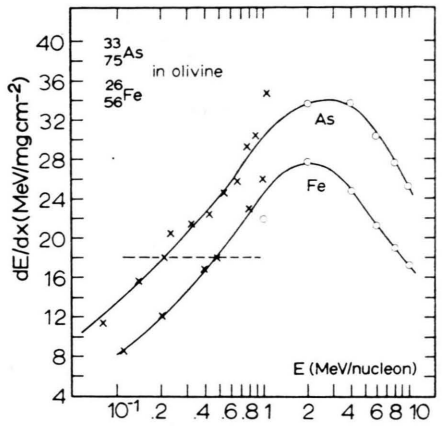

Fig. 3. Specific energy loss of arsenic and iodine ions in olivine and the critical energy loss of olivine as determined from the dE/dX criterion. + Calculated with the theory of LINDHARD et al. [11]. o Calculated with the method developped by NORTHCLIFFE [12] and HECKMAN et al. [13].

senic and iron nuclei, the variation of the average energy loss in olivine as a function of energy [14].

If this criteria were valid, a value of

$$(dE/dX)_c \approx 18 \text{ MeV/mg cm}^{-2}$$

would be obtained from the value of E_c for arsenic ions measured in the present work. Using this value, it would then be possible to deduce a value for Z_c of ≈ 23, by finding the ion whose maximum value of dE/dX is equal to 18 MeV/mg cm^{-2}.

However some recent work [9, 15] seems to show that the dE/dX criterion is not a proper description of track registration; therefore FLEISCHER et al. [4, 9, 15] proposed another one. They suppose that only those ions whose primary ionization rate $S(Z, E)$ (in number of ions/cm) is greater than a critical value, S_c, will be registered. To apply this criterion, they calculated the $S(Z, E)$ curves for different values of Z; by plotting experimental E_c values, it is possible to determine the critical primary ionization rate, S_c, below which tracks are not formed. To deduce the corresponding Z_c value, the curve $S(E, Z_c)$ whose maximum is equal to S_c must then be identified.

II.2. Limits of Application of the S_c Criteria

We will show that the limits of application of the S_c criteria result from the uncertainty of the values of two basic parameters in the equation which is applied. To illustrate this point, we use the S_c criterion to deduce (from our experimental results) the characteristics of the registration of ions whose nuclear charges Z are less than that of arsenic. We choose a value of $Z = 26$ because of the interest in verifying whether the iron nuclei of the primary cosmic ray flux are registered in some olivine and hypersthene crystals of meteoritic origin. FLEISCHER et al. [4] have observed fossil tracks in these meteoritic minerals and have attributed them to such ions.

A) Determination of the registration characteristics of iron nuclei in olivine crystals

For this determination, we have calculated the value of the ratio $K(\text{Fe, As}) = S(\text{Fe}, E)/S(\text{As}, E_c)$ by applying a formula given by BETHE [16]. In the primary ionization model, the iron nuclei will not give tracks when $K < 1$, they will be "poorly" registered when $K \sim 1$, and they will be registered with an efficiency of 100% in the energy range for which $K > 1$. The general expression for K is given by the following formula:

$$K(\text{Fe, As}) = \frac{\beta_{\text{Fe}}^{-2}}{\beta_{\text{As}}^{-2}} \cdot \left(\frac{Z_{\text{Fe}}^*}{Z_{\text{As}}^*}\right)^2 \cdot \frac{\ln[\beta_{\text{Fe}}^2/(1-\beta_{\text{Fe}}^2)] - \beta_{\text{Fe}}^2 + B/A}{\ln[\beta_{\text{As}}^2/(1-\beta_{\text{As}}^2)] - \beta_{\text{As}}^2 + B/A}$$

where $\beta = v/c$.

Z_i^* is the effective charge of the ion of nuclear charge Z_i calculated by a formula proposed by HECKMAN et al. [13]

$$Z_i^* = Z_i[1 - \exp\{-125\,\beta_i/Z_i^{2/3}\}];$$

although it is known that this formula is only an approximation at low energies it is apparently the best analytical expression for Zi^* which is available. A and B are two constants in the BETHE formula. FANO [17] has discussed their physical meaning [18] and a method to calculate their values in the case of elements in the *gaseous* state. It is well known that these two constants can be calculated exactly only for hydrogen atoms; they appear only by their ratio, $X = B/A$, in the expres-

[11] J. LINDHARD, M. SCHARFF, and H. E. SCHIOTT, Kgl. Danske Videnskab. Selskab. Mat. Fys. Medd. **33**, No. 14 [1963].
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[18] A is the total dipole strength of ionizing transitions in the gas molecule and B is another molecular constant.

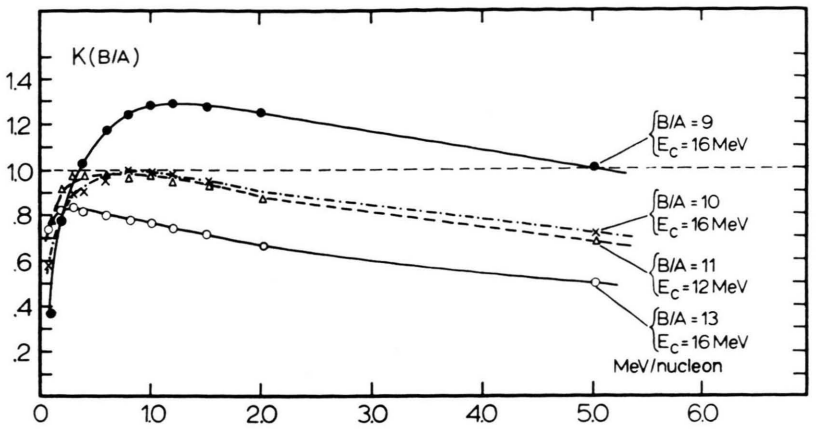

Fig. 4. Primary ionization rate of Fe nuclei relative to the primary ionization rate of As nuclei — (K) — as a function of energy for different parameters E_c and B/A.

E_c (in MeV)	X_c	$X(10\,\mu)$
20	9.5	9.2
16	10	9.7
12	10.7	10.0

Table 2.

sion of K and in the following we consider X as a variable parameter the values of which will be given later.

In Fig. 4 and Table 2 we have summarized the results of our calculations for olivine. In Fig. 4 the K(Fe, As) curves for 4 values of $X(9, 10, 11, 13)$ and 2 values of E_c (12 and 16 MeV) are plotted. In Table 2 we have listed different values of E_c for arsenic ions (column 1), the values of X_c such that for $X = X_c$ the iron nuclei are not registered (column 2), and the values of X for which the maximum track length of iron nuclei in olivine would be $\approx 10\,\mu$ [19].

Examination of the curves shows clearly that if the X values are not determined with very high accuracy it is not possible to use the present result to conclude very much about the registration of Fe ions in olivine. Let us consider, for e. g., the values $X = 9$ and $X = 10$. For the smaller X value, the iron nuclei would be registered from $E_{min} \approx 34$ MeV to $E_{max} \approx 190$ MeV whereas, in the second case, it would not be possible to see the tracks. Moreover, the values listed in Table 2 show that this conclusion cannot be modified even if different values of E_c are chosen.

In the following paragraph we will show that it is not posible at present to determine precise values of X. For this reason, the S_c criterion cannot be applied quantitatively and therefore the question of Fe ion registration in olivine is not settled by the present work.

[19] This value has been deduced from a length distribution study of fossil tracks in meteoritic olivine samples [4].

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

B) Estimation of X values

B) 1. Estimation of the X values for solids from measurements of X values in gases: Except for hydrogen atoms ($X \approx 14$) it is not possible to get an accurate value of X for elements in the gaseous state (and, a fortiori, for compounds in the *solid* state) by quantum mechanical calculations. For hydrogen molecules and some elements, the values of X have been deduced from ionization studies in gases. For molecular hydrogen, helium, neon, and argon McClure [20] reports the following values: 12.2, 9.65, 8,53, 9.70. To our knowledge, no measurement in solids has been as yet reported. This stems from the fact that it is not yet possible experimentally to evaluate the primary ionization from a measurement of total ionization.

It can be seen that, in general, the values of X for gases are nearly constant (≈ 9) when the atomic masses are high. If this value of 9 were also valid for solids [21], we would conclude that ions of $Z \approx 26$ would register in olivine. However the maximum length of iron tracks ($\approx 30\,\mu$) which can be deduced from this value of X is larger than the upper limit ($\approx 10\,\mu$) deduced by Fleischer et al. [4].

B) 2. Estimation of a lower limit for X from the study of track length in olivines of meteoritic origin: The maximum length of tracks in meteoritic olivine is $l_{max} \approx 10\,\mu$. These tracks could be due to VH nuclei ($Z \approx 26$) of the cosmic rays. This upper limit of the track length of an "iron" nucleus can be used to deduce a lower limit of $X \approx 9.6$ as follows:

[21] According to U. Fano (Ann. Rev. Nucl. Sci. **13**, 1 [1963]), a very small change is to be expected as one goes from the gaseous state to the solid state.

by successive approximations, the $K(X)$ curve which intersects the $K=1$ asymptote at $E_1=0.6$ and $E_2=2$ MeV/nucleon can be identified; for these values of E_1 and E_2, a value $l_{max}\approx 10\ \mu$ can be deduced by graphical integration of the dE/dX curves on Fig. 2 $\left[l_{max}=\int_{E_1}^{E_2} dE/(dE/dX)\right]$.

B) 3. Estimation of the X values by the study of registration of heavy ion tracks in solids: If the S_c criterion is valid, it is correct to assume that there is a single value S_c which is characteristic of a solid and which does not depend on the nuclear charge or on the energy of the ions used to determine its value. Thus, there ar at least two methods of estimating the value of X from track measurement in solids.

If heavy ions of $Z_i > Z_c$ are accelerated to high energies (≈ 10 MeV/nucleon), it should be possible, by decreasing progressively the ion energy, to measure successively the two values of the critical energies, E_{max} and E_{min}, which deliniate the energy range in which the ions are registered. The values of

$$K_1 = S(Z_i, E'_{max})/S(Z_i, E_{max})$$

and

$$K_2 = S(Z_i, E'_{min})/S(Z_i, E_{min})$$

can then be determined, where E'_{max} designates an energy slightly smaller than E_{max} and E'_{min} an energy slightly greater than E_{min} for which the ion is registered with an efficiency of 100%. Since $K_1>1$ and $K_2>1$ either an upper limit (for K_1) or a lower limit (for K_2) of X can be deduced and therefore an estimation of $X\approx\frac{1}{2}(X_{min}+X_{max})$.

It is also possible to measure the X value by using ions of different nuclear charges $Z_i > Z_c$ and measuring the E_c^i value at low or at high energy. Then for a couple of ions, Z_1, Z_2, we can write down the following identity:

$$S(Z_1, E_c^1) \approx S(Z_2, E_c^2) \approx S_c .$$

Therefore:

$$S(Z_1, E_c^1)/S(Z_2, E_c^2) \approx 1 .$$

By solving this identity, it is possible to get one value of X.

If the S_c criteria is valid, all the X values which can be obtained with different ions have to be compatible. For example, it would not be possible to get with the second method an X value which is outside the $X_{max}-X_{min}$ interval determined by the first method.

We have carried out such calculations for olivine and also for mica, using the experimental data of FLEISCHER et al.[10]. The results of our calculations are listed in Table 3, where we give in column 1 the values of $K=S(Z_i, E_i)/S(Z_j, E_j)$ and in column 2 the limits or values of X, as deduced from the relation $K \geqq 1$.

	$K(B/A) = S(Z_i, E_i/S(Z_j, E_j)$ (E_i and E_j in MeV)		$B/A = X$
olivine	S(As, 20)/S(As, 16)	>1	$<11{,}4$
	S(As, 20)/S(I, 18)	>1	< 9.3
	S(As, 20)/S(Ar, 40)	>1	> 1.0
	S(As, 16)/S(I, 18)	≈ 1	9.0
	S(Ar, 92)/S(Ar, 152)	>1	> 6.7
	S(Cl, 69)/S(Cl, 105)	>1	> 7.20
	S(S, 54)/S(S, 84)	>1	> 7.6
	S(Si, 28)/S(Si, 47)	>1	> 7.9
mica	S(Ne, 3.8)/S(Ne, 20)	>1	>10
	S(Ar, 152)/S(S, 84)	≈ 1	≈ 14
	S(Ar, 152)/S(Si, 47)	≈ 1	≈ 14.5
	S(Cl, 84)/S(Si, 47)	≈ 1	≈ 14

Table 3.

II.3. Discussion

The values of X listed in the Table 3 for olivine and mica show that the limits and values of X are compatible. From this it can be concluded that, within the experimental error, the primary ionization model is consistent with the results.

However, it can be seen that the spread in X values is rather large and this, in turn, implies that a precise determination of Z_c cannot be obtained from the studies of registration of heavy ions of $Z_i > Z_c$. In particular Z_c for olivine is uncertain to ± 5 charge units being equal to 26 ± 5.

Although the available experimental results with accelerated heavy ions are consistent with the primary ionization criteria, they do not constitute a definite proof of the validity of the model. For example, in olivine, the lower limits of X deduced from the length distribution of fossil tracks in meteorites is greater than some of the upper limits derived in other ways. Also, the X value for mica ($X\approx 14$) is much greater than the upper limit of X deduced for olivine. This observation is disturbing in light of the previously mentioned fact that the X values are relatively constant for elements in the gaseous state. The fact that mica and olivine have virtually the same average atomic number $\bar{Z}$ (≈ 12) and that their mean excitation potentials are both

nearly equal to 140 eV [22] makes it difficult to understand the difference in X values between these two materials [23].

Moreover, it appears to be difficult, for two independent reasons, to apply the S_e criterion in its actual form: a) according to WILLIAMS [24], the BETHE equation is valid only when $Z(e^2/\hbar v) \ll 1$; in our experiments and in those of FLEISCHER et al., the values of this parameter vary between 1/5 and 3/4, therefore, the interpretation of the experiments cannot be taken for granted; b) it is impossible to obtain accurate values of the effective charges of heavy ions at low energies (below 0.5 MeV/nucleon); this accentuates the inadequacy of the BETHE equation.

We conclude that the problem of the registration of VH nuclei in crystals of meteoritic origin cannot be solved by the study of the registration of accelerated heavy ions of $Z > 26$. Crystals for which $Z_e \approx 26$ will have to be exposed to a beam of accelerated iron nuclei whose energies are high enough (≈ 60 MeV) to produce a maximum value in the rate of energy losses, or another method for the study of the registration sensitivity of solids as given by HORN and v. OERTZEN may be used [25]. In this method elastically scattered nuclei of the relevant element are knocked into the material to be studied *.

Acknowledgement

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[22] In order to get these "mean" values we have used the following equation:

$$N\bar{Z} = \sum_i N_i Z_i \quad \text{and} \quad N\bar{Z}\cdot \ln \bar{I} = \sum_i N_i Z_i \ln I_i ,$$

where N is the total number of atoms per cubic centimeter and I_i are the relative number and the excitation potential of the i-th element.

[23] However, we cannot exclude the possibility that the difference in sensitivity between mica and olivine is due to a difference between the values of X. It would be interesting to measure the values of X in solids of different sensitivities in the manner we have described; in this way it could be seen whether there is a correlation between the sensitivities of the crystals and the values of X.

[24] J. WILLIAMS, Rev. Mod. Phys. **17**, 217 [1954].

[25] P. HORN and W. v. OERTZEN, Earth Planet. Sci. Letters **2**, 280 [1967].

* Note added in proof: The registration of Fe ions in olivine has now been demonstrated by HORN and VON OERTZEN using the method of ref. [25] (Z. Naturforschg. **22 a**, Heft 10).