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Further Experiments on the Surplus Gamma-Radiation from Granite

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Field experiments in Massachusetts (1945) and laboratory experiments with a bulk sample of granite (300 lb.) from Quincy, Massachusetts, showed that the ionization produced by gamma-rays from this type of rock is about twice as great as the one computed from their content of uranium, thorium, and potassium. In order to find out the origin of this unexpected surplus radiation, new experiments were carried out, using essentially the same technique, but shielding the ionization chambers with lead cylinders about 1 cm thick. The radium content of the bulk sample of Quincy granite was re-checked in the National Bureau of Standards and found, on the average, only 7 percent above the figure given by Evans and Goodman.

These new experiments show again a surplus of penetrating radiation of about 100 percent above the computed value, exceeding by far all possible errors in the determination of the uranium, thorium, and potassium content of Quincy granite. It is shown that neither the production of photons in transmutations induced by alpha-rays within the granite, nor spontaneous fission of uranium and thorium in this rock by cosmic rays or by neutrons from cosmic rays could account for the observed effect.

Preliminary experiments with dunite (South Carolina), a mineral containing almost no uranium, thorium, or potassium showed a complete absence of any radiation from this type of rock.

It must be concluded that a hitherto unknown penetrating radiation is given off by granite. Further experiments are in progress.

IN May 1946, I found a very interesting discrepancy in the amount of observed ionization produced by gamma-rays from thick layers of granite, and the quantity computed from the known amounts of uranium, thorium, and potassium contained in it. The latter amounted to only about 50 percent of the total ionization actually found by measurements in a quarry (Quincy, Massachusetts), and in laboratory experiments with large quantities (300 lb.) of the same sort of granite. I reported on these experiments at the Annual Meeting of the American Geophysical Union in Washington, D. C. on May 30, 1946.[1]

A detailed description of the experiments and of the Gish-ionization meter used has been given in this paper (1946). It was concluded that it was very unlikely that the uranium, thorium, and potassium content given by R. D. Evans and Clark Goodman for the Quincy granite could be too low by such an extent. Therefore, it seemed necessary to investigate this discrepancy more rigorously by repeating all experiments with much heavier screening of the ionization chambers. These new experiments were carried out in the winter of 1946–47, and this paper presents the results.

[1] V. F. Hess, Trans. Am. Geophys. Union **27**, 670 (1946); V. F. Hess, Norsk Geologisk Tidsskrift **27** (Jan. 1947).

I. EXPERIMENTAL PROCEDURE AND COMPUTATION OF THE IONIZATION IN THIS ARRANGEMENT

The Quincy granite was recommended to me by Robley D. Evans and Clark Goodman as one of the most homogeneous and best analyzed types of igneous rocks. These authors[2] give the following figures of uranium content: $M_1=(2.7\pm0.5)\cdot10^{-6}$ gu per g of granite (taken from Swingles quarry, Quincy, Massachusetts). This in equilibrium with radium corresponds to $0.95\cdot10^{-12}$ g Ra per g of rock. For the thorium content, they gave, $M_2=(8.1\pm2.0)\cdot10^{-6}$ g Th per g of rock. The potassium content is not given in their paper; however, it can be estimated to be between 3 and 4 percent. Warren's[3] figure (3.8 percent) is certainly reliable enough for this purpose.

If an ionization chamber is either placed directly above a large slab of the rock, or surrounded by a thick layer of crushed rock, the ionization produced by the hard gamma-rays from Ra (RaC″), Th (ThC″), and potassium in the rock can be computed by Eve's formulas. The formula for a point-shaped source of radiation is $q=Km/r^2e^{-\mu r}$. Here q is the number of pairs of ions produced per cm^3 and per sec.; m is

[2] Robley D. Evans and Clark Goodman, Bull. Geol. Soc. Am. **52**, 459 (1941).
[3] C. H. Warren, Proc. Am. Acad. **49**, 227 (1913).

the amount of radioactive material per cm³ of rock; and μ is the absorption coefficient of gamma-rays in the rock. This expression gives, when integrated for an infinite thickness of radioactive rock all around the ionization vessel, the total ionization $q=q_1+q_2+q_3$ where $q_1=(4\pi K_1 m_1/\mu_1)$, $q_2=(4\pi K_2 m_2/\mu_2)$, $q_3=(4\pi K_3 m_3/\mu_3)$ are the components of the ionization produced by the radium and thorium products and potassium (subscript 1 pertaining to radium, 2 to thorium, 3 to potassium). In these formulas, m_1, m_2, m_3, denote the amounts of radium, thorium, and potassium per cm³ of rock. Dividing by the density (ρ) of the rock, we obtain $M_1=m_1/\rho$, $M_2=m_2/\rho$, $M_3=m_3/\rho$, which are the figures for the radium, thorium, and potassium content per gram of rock listed above.

For an infinite layer of rock surrounding an ionization chamber, the formulas for the components of ionization to be expected in the chamber are, therefore: for radium $q_1=4\pi K M_1/(\mu_1/\rho)$, for thorium $q_2=4\pi K_2 M_2/(\mu_2/\rho)$, $q_3=4\pi K_3 M_3/(\mu_3/\rho)$. This way of writing the formulas has the further advantage that one needs only the mass absorption coefficients (μ_1/ρ), (μ_2/ρ), (μ_3/ρ) of the hard gamma-rays of Ra, Th, and potassium which are much better known than μ_1, μ_2, μ_3, and are practically constant for all materials.

The constants K_1, K_2, K_3 (Eve's constants) differing slightly for vessels of different size and materials, were redetermined by the author and Eva M. Balling[4] for the chambers actually used for the present study. For a brass vessel, filled with nitrogen at normal pressure, we obtained 4.9×10^9 I (ion pairs/cm³ sec.) per gram of radium, in a chamber of 4888-cm³ volume.

In a monograph[5] on the ionization balance of the atmosphere, I have shown that the gamma-ray effects of radium itself, of radium B, uranium X, etc., are negligible, and therefore a simplified treatment with only one significant absorption coefficient for each component is permissible.

It is obvious that μ_1/ρ, μ_2/ρ, μ_3/ρ must be known for the hardest component of gamma-rays in each case (RaC″, ThC″). The absorption coefficient of the gamma-rays from potassium is, according to L. H. Gray and C. T. P. Tarrant,[6] practically equal to the one of the gamma-rays from RaC″.

Consideration of the softer components of gamma-radiation (larger coefficients μ) would give only small and insignificant increases in the values of q_1 and q_2. However, in order to eliminate completely the possibility of interference from the softer gamma-radiation, the present experiments were carried out with the ionization chambers shielded with 0.98 cm of lead on all sides and on the bottom. This, in addition to the $\frac{1}{8}$-in. brass wall of the chamber, excludes all gamma-rays of less than 1-Mev energy. Eve's constant for this arrangement was found to be

$$K_1=4.9\times10^9\times\exp[-\mu_{Pb}d]=4.9\times10^9\times0.592=2.93\times10^9 \text{ I}$$

per gram of radium ($\mu_{Pb}=0.533$ cm^{-1}).

Eve's constant for thorium (K_2) has not as yet been determined because of the lack of a radium-free thorium standard old enough to be in equilibrium with mesothorium and radiothorium. Finally I decided to use a powdered sample of a thorium mineral, practically free of uranium (thorite) as a standard. This sample (102 grams of thorite) in a sealed glass tube, was put at my disposal by Dr. L. F. Curtiss, National Bureau of Standards and I reported on these determinations last year.[1]

For the heavily screened ionization chambers (9.8-mm Pb) Eve's constant for thorium was determined anew last winter and found to be $K_2=440$ I/g Th/cm behind 9.8-mm lead (without lead the value is 742 I). For potassium the value of Eve's constant (K_3) computed by two different methods (0.12) gives, after reduction to the present experimental conditions (filtered through 9.8-mm Pb), $K_3=0.071$ I/g K/cm.

The mass absorption coefficients used in the following computations are: for radium (RaC″) $\mu_1/\rho=0.045$ cm²/g (the same value holds for potassium); for thorium (ThC″): $\mu_2/\rho=0.041$ cm²/g.

The average radium content of a number of samples from the bulk sample of Quincy granite was recently re-determined by L. F. Curtiss in the National Bureau of Standards and given to

[4] V. F. Hess and Eva M. Balling, Trans. Am. Geophys. Union **26**, 237 (1945).

[5] V. F. Hess, "Jonisierungsbilans der Atmosphaere," Ergebn. Kosm. Physik **2**, 95 (1934).

[6] L. H. Gray and C. T. P. Tarrant, Proc. Roy. Soc., London **A145**, 681 (1934).

be 1.02×10^{-12} g radium per gram, corresponding to 2.9×10^{-6} g uranium per gram, in substantial agreement with Evans' and Goodman's value (0.95×10^{-12} g Ra/g). In the following computation I used Curtiss' value.

a. Radium component: $K_1=2.90\times10^{9}$, $M_1=1.02\times10^{-12}$, $\mu_1/\rho=0.045$

$$q_1=\frac{12.56\times2.90\times10^{9}\times1.02\times10^{-12}}{0.045}=0.832 \text{ I.}$$

b. Thorium component: $K_2=440$, $M_2=8.1\times10^{-6}$, $\mu_2/\rho=0.041$

$$q_2=\frac{12.56\times440\times8.1\times10^{-6}}{0.041}=1.094 \text{ I.}$$

c. Potassium component: $K_3=0.071$, $M_3=0.038$, $\mu_3/\rho=0.045$

$$q_3=\frac{12.56\times0.071\times0.038}{0.045}=0.753 \text{ I.}$$

The total ionization to be expected from (a), (b), and (c) is, therefore,

$$q=q_1+q_2+q_3=2.68 \text{ I.}$$

II. LABORATORY EXPERIMENTS WITH HEAVILY SCREENED IONIZATION CHAMBERS

For these experiments, a quantity of 300 lb. of crushed granite (grain size from $\frac{1}{4}$ to 1 in.) was shipped by Swingle Company quarries to Fordham University in bags. The "iron house" was built up from iron bars on the bottom and on all sides 10 cm thick, with the top left open. The medium or the small ionization chamber (4.9- and 1.6-liter volume, respectively) was placed in the center of the hollow space on a lead plate 1 cm thick, and the side wall of the chamber was surrounded with a lead cylinder of 0.98-cm thickness. The top of the iron house was left open so that the electrometer and its housing could be easily placed on or removed from the top of the ionization chambers. Local gamma-rays from the ceiling therefore had access to the system, but their intensity was cut down by placing a large half-cylinder of lead (1 cm thick) above the electrometer head.

In all experiments, alternate readings were taken first with the iron house empty and with the space between the lead cylinder and the inner wall of the iron house filled completely with the crushed granite. Each series of readings continued for several days.

The ionization in the empty iron house (observed with the medium chamber) was 3.345 I; with the granite, 5.61 I. Thus the difference 2.26 I corresponds to the effect of a layer of granite of 15.6 cm average thickness.

A graphical integration of the "open cone" on top of the chamber (i.e., for the part of the space which was not covered by granite) showed that about 82 percent of the full solid angle 4π was covered with granite. A complete concentric layer (spherical shell 15.6 cm thick) therefore would give $2.265/0.819=2.76_5$ I. Since the mean absorption coefficient of gamma-rays from radium and thorium (filtered through 15-mm lead) in crushed granite was directly determined by myself[1] ($\mu=0.0563$ cm^{-1}), one can now extrapolate to infinite thickness by using the formula[7]

$$q_d=q_\infty(1-e^{-\mu d}),$$

and obtain

$$q_\infty=2.765/0.585=4.72 \text{ I.}$$

A final correction has to be made because the majority of the rays from the granite have to penetrate the lead cylinder and the wall of the chamber in an oblique direction. This correction was evaluated graphically and amounts to 3.5 percent. The final corrected value ($q_\infty=4.88$ I), therefore, is almost twice the value computed

TABLE I. Summary of results.

A. *Medium chamber:* (shielded with 0.98-cm lead)	
Ionization produced by a 15.6-cm layer of crushed granite	2.26_5 I
Corrected for open cone on top	2.76_5 I
Extrapolated for granite layer of infinite thickness	4.72 I
Corrected for oblique rays	4.88 I
B. *Small chamber:* (shielded with 0.98-cm lead)	
Ionization produced by a 15.6-cm layer of crushed granite	2.05 I
Corrected for open cone on top	2.44 I
Ionization extrapolated for infinite layer of granite	4.17 I
Corrected for oblique rays	4.32 I
C. *Value computed:* Taking uranium content of Quincy granite $(2.9\pm0.5)\times10^{-6}$ g/g; thorium, $(8.1\pm2.0)\times10^{-6}$ g/g; and potassium, 0.038 g/g	2.68 I

[7] V. F. Hess, Trans. Am. Electrochem. Soc. **41**, 287 (1922).

for this experiment from the uranium, thorium, and potassium content of the Quincy granite (2.68 I).

The experiments were repeated with the small (1.6-liter) ionization chamber, also screened with a 9.8-mm lead cylinder around. Table I gives a summary of the results with both chambers. It is evident that in both cases the ionization observed is almost twice as great as the computed value.

An independent, although only approximate, check on this discrepancy was made by another method: with the small chamber it was possible to perform an absorption experiment with the granite in place by inserting a second screen of lead (0.966-cm thickness) between the ionization chamber and the first lead screen. Taking a mean absorption coefficient $\mu/\rho=0.0433$ for the composite gamma-radiation of RaC″, ThC″, and potassium corresponding to $\mu=0.0563$ cm^{-1} in the crushed rock, and the thickness of the second lead screen, $d=0.966$ cm, we obtain for the transmitted fractions, $e^{-\mu d}=0.581$: the fraction of radiation absorbed in the second lead screen, therefore, is 0.419. This corresponds to the difference in the ionization actually observed with one and two lead screens, which was 1.10 I. This, therefore, is 41.9 percent of radiation which actually is emitted from the granite: $1.100/0.419=2.62$ I; the "open cone" correction in this case has to be doubled since the bottom of the chamber was not covered with a second lead plate. Therefore the open cone correction is accomplished by integrating the solid angle subtended by the contour of the second lead cylinder as seen from the center of the small chamber. This gives $2.62/(1-2\times0.0985)=3.26$ I and from a granite layer of infinite thickness $3.26/(1-e^{-\mu d})=3.26/0.585=5.6$ I, which again is about two times greater than the value computed from the uranium, thorium, and potassium content.

The experiments, previously reported upon[1] with the same material but without the 0.98-cm lead screen around the chambers, gave similar discrepancies: the "iron house" experiment with crushed granite surrounding the ionization chamber (brass wall $\frac{1}{8}$ in. thick) gave 9.5 I as compared with a value of 4.1 I computed from the value of the uranium, thorium, and potassium content. In an experiment in the quarry itself, where the apparatus was set up directly above a large block of solid granite, the observed ionization was 5 to 6 I, while for this case, the computed value was 2.06 I.

DISCUSSION

Since both the field experiments and the laboratory measurements show that the uranium, thorium, and potassium content of granite could account for only about one-half of the gamma-ray ionization observed in five independent experiments, one is now confronted with the existence of a "surplus penetrating radiation" of unknown origin.

At first one may be inclined to assume that the well-known "back scattering" of cosmic rays may be responsible for this surplus ionization. However, this effect would be expected to be of much smaller magnitude, since the total ionization produced by cosmic rays at sea level amounts to only 2 I. Furthermore, this intensity was considerably reduced in the case of all "iron house" experiments, since these were carried out in the basement of the Physics Building of Fordham University with four floors and ceilings above the iron house.

In addition, the most recent experiments carried out at Fordham where the iron house was filled with 340 lb. of dunite from North Carolina (a mineral containing only 0.02×10^{-6} g uranium, 0.06×10^{-6} g thorium, and 0.0002 g of potassium per gram), showed no trace of such an effect. The dunite experiments will be reported on later, quantitatively, in another publication.

The possibility that the uranium and thorium content of Quincy granite (as reported by R. D. Evans and C. Goodman), are much too low can be dismissed, in view of the great care and improved technique employed by these authors. Furthermore, the value of the radium content of the Quincy granite was confirmed recently by L. F. Curtiss by direct analysis of the crushed granite used in my experiments. There are only a few other possibilities of explanation left, some of which can be investigated by further experiments.

1. Nuclear Processes Produced by Alpha-Rays in Granite, and Accompanied by Photons

This possibility was suggested by the Swiss geologist H. Hirschi in a private communication. There are a few (α, p) and (α, n) reactions where photons of >1.5 Mev are also emitted, and some of the elements involved are present in granite. A rough quantitative estimate can be made as follows: assuming that the thorium and uranium products in granite are together equivalent to 2×10^{-12} g Ra/g in their alpha-ray emission, 1 g of granite would produce $3.7\times10^{10}\times2\times10^{-12}\times8=0.6$ alpha-particles per second, and the 300 lb. of granite in the iron house would give off 90,000 alpha-particles per second. Effective nuclear transmutations would certainly not be accomplished by alpha-rays of low range and energy. Furthermore, the yield would be less than 1 in 10^5, so that much less than one transmutation per second would occur in 300 lb. of granite. Photons may be produced perhaps in $\frac{1}{10}$ of these cases, and less than half of them could go through the ionization chamber. Thus the effect would amount to much less than 0.01 I (per cm^3).

2. Nuclear Processes in the Granite Induced by Cosmic Rays

A. One might consider neutron-induced fission of uranium and thorium and the so-called spontaneous fission. G. N. Flerov and K. A. Petrshak[8] reported this first in 1940 and several authors have confirmed their findings.[9] The values of the "partial decay constant" for spontaneous fission can be taken as $\lambda_F=7$ to 9×10^{-24} sec.$^{-1}$ under the assumption that one neutron is emitted per fission, as compared with the "normal" alpha-ray decay of U_I with $\lambda_\alpha=5\times10^{-18}$ sec.$^{-1}$. The amount of uranium in 150 kg of Quincy granite $(2.9\times10^{-6}\times1.5\times10^{-5}=0.435$ g) corresponds to $0.44\times6.02\times10^{23}/238=1.1\times10^{21}$ atoms, and the number of spontaneous fissions in this material would be $n_f=8\times10^{-24}\times1.1\times10^{21}=8.8\times10^{-3}$ per sec. Doubling this amount because of the fissionable thorium in the granite, we would have about 18×10^{-3} fissions/sec. occurring in the bulk sample of 150-kg crushed granite. If the energy of photons in each fission is estimated as 10 Mev, we would have 18×10^4 ev of photon energy liberated, corresponding to a production of 5600 ion pairs/sec. for complete absorption all around. Taking the solid angle subtended by the cross section of the chamber as seen from an average point in the granite as $0.16/4\pi$ we would have an energy of 1×10^4 ev going through the chamber per second. Half of it will be absorbed in the granite before reaching the chamber. If now, the whole energy, arriving at the chamber (volume 1600 cm^3) were absorbed in it and used for productions of ions (32 ev per ion pair), we could expect $(5\times10^3)/(32\times1.6\times10^3)=0.1$ I produced in the chamber. Actually, of course, only a very small fraction of this energy would be used up in producing ions within the chamber, and so we see that spontaneous or cosmic-ray induced fission of uranium and thorium in the granite cannot produce the surplus ionization actually observed. Mr. David Hill (Palmer Physical Laboratory, Princeton University) arrived independently at the same conclusion, using a different method of computation.*

B. One could also think of artificial radioactivity induced by cosmic radiations in the granite. There are so many elements contained in volcanic rocks like granite that it is almost impossible to speculate on it. P. J. G. deVos and S. J. duToit[10] found that penetrating ionizing particles are produced by cosmic-ray neutrons, although the effect in a triple coincidence-counter telescope, using a paraffin layer of 1.3 cm, was very small (3 percent of the total number of coincidences). The possibility of such an effect of cosmic rays on granite can be tested by performing the "iron house" experiment in a mine or in a tunnel deep enough to exclude cosmic rays completely. Such experiments are planned for the immediate future.

Since the dunite in the iron house experiment did not show any penetrating radiation, although exposed to cosmic rays, it is rather to be expected that other rocks, even granite, will behave similarly.

[8] G. N. Flerov and K. A. Petrshak, J. Phys. U.S.S.R. **3**, 275 (1940).

[9] Mauer and Pose, Zeits. f. Physik **121**, 285 (1940); G. Scharff-Goldhaber and G. S. Klaiber, Phys. Rev. **70**, 229 (1946).

* Private communication from Professor J. A. Wheeler.

[10] P. J. G. deVos and S. J. duToit, Phys. Rev. **70**, 229 (1946).

If the granite, protected from cosmic rays by thick layers of rock, shows the same ionizing effect as in the laboratory, we will have to assume that a hitherto unknown penetrating radiation is given off by the granite itself. It will be interesting to investigate other volcanic rocks of different uranium and thorium content in order to see whether this effect is correlated with the content of radioactive materials in the rocks.

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The Electric Resonance Method of Radiofrequency Spectroscopy The Moment of Inertia and Electric Dipole Moment of CsF*

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This paper describes a new method of radiofrequency spectroscopy in which transitions between the energy levels of a single rotational state of a linear rotator in an electric field are caused by a superimposed radiofrequency field. The method utilizes a molecular beam and has been applied to the measurement of the moment of inertia, I, and the electric dipole moment, μ, of CsF. Values for these quantities are: $I=(187\pm22)10^{-40}$ g cm^2 and $\mu=7.3\pm0.5$ Debye. The internuclear distance, derived from the moment of inertia, is 2.60 ± 0.16 angstroms.

1. INTRODUCTION

THE purpose of this paper is to describe some experiments[1] on CsF, a linear polar molecule which is convenient to handle, in which the molecular beam resonance method was applied to molecules in a single rotational state and in which the fields were all electric instead of magnetic. With the demonstrated high resolving power of this method, it is possible to evaluate the parameters which enter into the orientation polarizability of a molecule, namely, the moment of inertia and the electric dipole moment. In addition, it is expected that the resolving power will be so high that the variation of these quantities with vibrational quantum number will be observed. From the standpoint of nuclear physics it is also interesting that the resolving power should be ample to measure accurately the interaction of the nuclear electric quadrupole moment[2] with the electric field gradient of the electrons, and the interaction of the nuclear magnetic moment with the rotation of the molecule. These latter points will be developed in another paper from this laboratory.

The molecular transitions which have been observed are not of the ΔR type (transitions between different rotational states) since the microwave techniques required for their observation were not developed at the time that the apparatus was designed.[3] Rather, the transitions are between states with the same value of R but

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[1] H. Hughes, Phys. Rev. **70**, 570 and 909 (1946).

[2] Dr. U. Fano, National Bureau of Standards, private communication.

[3] Recently Dakin, Good, and Coles, Phys. Rev. **70**, 560 (1946), have observed a ΔR-type transition of OCS in a wave guide at 24,320 Mc. The frequency for the $R=1\rightarrow R=0$ transition of CsF is 8830 Mc. This is a wave-length of 3.4 cm.