

tive conversations, to E. Helfand for a critical reading of the manuscript and for important suggestions, and to L. Kopf for technical assistance.

¹Second-harmonic generation has been studied in ferroelectrics at temperatures both above and below the ferroelectric transition temperature by J. P. van der Ziel and N. Bloembergen, *Phys. Rev.* **135**, 1662 (1964); R. C. Miller and A. Savage, *Appl. Phys. Letters* **9**, 169 (1966); and R. C. Miller, *Phys. Rev.* **134**, A1313 (1964). These authors, however, do not discuss the harmonic scattering by fluctuations described here.

²H. A. Levy and S. W. Peterson, *Phys. Rev.* **86**, 766 (1952).

³F. Simon, *Ann. Physik* **68**, 241 (1922); A. W. Lawson, *Phys. Rev.* **57**, 417 (1940).

⁴A. Savage, *J. Appl. Phys.* **36**, 1496 (1965).

⁵A number of workers have suggested that the NH_4Cl lattice becomes unstable just below T_c and undergoes a first-order transition at this point: D. G. Thomas and A. K. Stanley, *J. Chem. Soc.* **1951**, 1420; C. W. Garland and C. F. Yarnell, *J. Chem. Phys.* **44**, 3678 (1965). In addition, a narrow hysteresis region has been found around T_c . The sharp decrease in σ at T_c is consistent with, but does not necessarily imply, a

first-order transition region. Nmr [S. Ueda and J. Itoh, *J. Phys. Soc. Japan* **22**, 927 (1967)], infrared [C. W. Garland and N. E. Shumaker, *J. Phys. Chem. Solids* **28**, 791 (1967)], and piezoelectric [S. Bahr and J. Engl, *Z. Physik* **105**, 470 (1937)] measurements all demonstrate that for $T > T_c$, long-range order vanishes for this system.

⁶This value may be in error by several hundredths of a degree since our primary concern has been to achieve a high degree of temperature stability and reproducibility, rather than great absolute accuracy.

⁷R. Bersohn, Y. H. Pao, and H. L. Frisch, *J. Chem. Phys.* **45**, 3184 (1966).

⁸L. S. Ornstein and F. Zernike, *Z. Physik* **19**, 134 (1918).

⁹M. E. Fisher and R. J. Burford, *Phys. Rev.* **156**, 583 (1967).

¹⁰Refractive indices were obtained from the data of G. Poincot and J. P. Mathieu, *Ann. Phys. (Paris)* **10**, 481 (1955).

¹¹Measurements of molecular-reorientation relaxation times in simple liquids using harmonic scattering have been reported by P. D. Maker and C. M. Savage, in Abstracts of the Symposium on Molecular Structure and Spectroscopy, Ohio State University, Columbus, Ohio, 1966 (unpublished), p. 63.

TIME VARIATION OF THE CHARGE OF THE PROTON

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From the terrestrial occurrence of the nuclei Re^{187} and Os^{187} , it is deduced that the elementary unit of charge e cannot have varied by as much as one part in 1600 during the history of the earth.

Gamow¹ has recently proposed the hypothesis that the elementary unit e of charge should increase with time according to the law

$$e^2 \sim t, \quad (1)$$

where t is the time elapsed since the beginning of the universe. This proposal was made in order to revive the old idea of Dirac² according to which the two dimensionless numbers

$$e^2/Gm^2, \quad tmc^2/\hbar, \quad (2)$$

which are presently both of the order 10^{38} , should increase proportionally as the universe evolves. Here G is the constant of gravitation and m is proton mass. Dirac supposed that e^2/Gm^2 would increase through a decrease of the strength of gravitational forces according to

$$G \sim t^{-1}. \quad (3)$$

However, various geophysical and astrophysi-

cal considerations³ argue against (3), and so Gamow suggested (1) as a substitute. Independently, but impelled by the same desire for economy of physics, Teller⁴ has suggested that e might decrease with time according to the law

$$(\hbar c/e^2) \sim \ln(tmc^2/\hbar), \quad (4)$$

so that "137" would be roughly equal to the logarithm of the big numbers (2). I present here a simple argument from nuclear systematics which excludes both possibilities (1) and (4).

On the earth there exist⁵ substantial quantities of the stable isotope Os^{187} and of the β -active isotope Re^{187} , which decays to Os^{187} with a half-life of 4×10^{10} y and a decay energy $\Delta = 2.6$ keV. According to the semiempirical mass formula,⁶ the energies of the two atoms contain

a Coulomb term

$$E_C = 0.6Z^2A^{-1/3} \text{ MeV} \quad (5)$$

which is proportional to e^2 , together with other terms independent of e^2 which represent nuclear binding. The energies will not be exactly linear functions of e^2 ; in particular the neutron-proton mass difference must be assumed to vary with e^2 in some unknown way. However, the term E_C is so large, and so well in agreement with observed nuclear binding energies, that any nonlinear electromagnetic effects must be comparatively insignificant. I therefore assume that the variation of Δ with e^2 comes entirely from the variation of E_C . Thus

$$e^2(d\Delta/de^2) = (E_C)_{\text{Re}} - (E_C)_{\text{Os}} = -15.8 \text{ MeV}, \quad (6)$$

with a theoretical uncertainty which may be conservatively estimated as $\pm 10\%$. The essential point is that Δ is a difference between two large quantities, the Coulomb energy and the nuclear binding, and the difference can be small only for a narrow range of values of e^2 .

The Re^{187} on the earth would not have survived if the half-life for its decay had been as short as 2×10^9 y during the early history of the earth, say 3×10^9 y ago. But the half-life for β decay between given nuclear states decreases at least as fast as $\Delta^{-2.835}$ as Δ increases.⁷ Therefore, the value of Δ 3×10^9 y ago cannot have been greater than $(200)^{0.353} = 6.50 \times$ its present value. This implies that

$$(d\Delta/dt) \geq -4.75 \times 10^{-9} \text{ keV/y}, \quad (7)$$

and hence by (6)

$$(1/e^2)(de^2/dt) \leq 3 \times 10^{-13} \text{ y}^{-1}. \quad (8)$$

The growth of e^2 according to (8) has been at least 300 times slower than Gamow's proposal (1).

In this derivation of (8) I assumed that the Fermi interaction constant g did not vary. A numerological argument based on the magnitude of g^2 and (2) suggests the possibility

$$g^2 \sim t^{1/2}, \quad (9)$$

while Gamow^{1,6} suggests that perhaps

$$g^2 \sim t^4. \quad (10)$$

Even the extreme assumption (10) does not change (8) significantly. If (10) were valid, the decay rate of Re^{187} 3×10^9 y ago would be reduced by a factor 4, and this would allow Δ to have been

greater only by a factor $4^{0.353} = 1.63$. The upper bound (8) would consequently be increased by less than a factor 2.

Another weakness of my argument is that I assume an age for the earth which is based on the assumed constancy of radioactive decay rates. I chose 3×10^9 y as a safe lower limit for the age, allowing for some variation of rates. Even if one takes a lower limit 1×10^9 y set by purely stratigraphic considerations, the effect on (8) is only a factor 3 on the right-hand side.

In discussing the possibility (4), one has to consider the electron-capture decay of Os^{187} which would occur if Δ became negative. When Δ is minus a few kilovolts, the capture occurs only from the outer shells of the osmium atom, and the theory of it is complicated. A comparison of the electron density in the osmium atom with the density in phase space of the continuum electrons emitted by the rhenium gives the relation⁸

$$(w_{EC}/w_\beta) \sim 1 \quad (11)$$

between the electron-capture and beta-decay rates at the same value of $|\Delta|$, provided that $|\Delta|$ is small compared with the K -electron binding energy (76 keV). The osmium decay at $\Delta = -2.6$ keV would thus be about as fast as the rhenium decay at $\Delta = +2.6$ keV. Since the osmium has survived for 3×10^9 y,⁹

$$(1/e^2)(de^2/dt) \geq -4 \times 10^{-13} \text{ y}^{-1} \quad (12)$$

by the same argument which led to (8). The hypothesis (4) would imply that

$$(1/e^2)(de^2/dt) = -11 \times 10^{-13} \text{ y}^{-1}, \quad (13)$$

and is therefore excluded, although the margin of safety is not so wide as in the case of (1).

Both of the inequalities (8) and (12) could be improved substantially by using accurate information concerning the natural occurrence of rhenium and osmium isotopes. For example, suppose that it is found that in some suitably old rocks Re^{187} occurs with less than 10% of radiogenic Os^{187} , and that Os^{187} sometimes occurs with less than 10% of Re^{187} . Then one could use 2×10^{10} y instead of 2×10^9 y as a lower limit to the half-lives of the two isotopes 3×10^9 y ago. Consequently (8) would be improved by a factor 20, and (12) by a factor 3. Information more precise than this concerning the isotopic composition of old rocks may well be

available.

The foregoing arguments have all assumed that the nuclear forces remained constant while the Coulomb forces varied. The arguments would fail if the nuclear forces had varied in precisely the right way to preserve the delicate balance between the Re^{187} and Os^{187} energies. There are two reasons why I do not believe in an exact compensation between varying e^2 and varying nuclear forces. (1) The nuclear binding energy is a strongly nonlinear function of the strength of the nuclear interaction, because of the short range of the forces, whereas the Coulomb energy is approximately linear in e^2 . (2) There are a number of other beta-active nuclei besides Re^{187} which could not have survived any strong variation of e^2 . Re^{187} gives the tightest numerical bounds, but each of the species⁵ V^{50} , Rb^{87} , Te^{123} , La^{138} , and Lu^{176} gives weaker inequalities similar to (8) and (12). It seems overwhelmingly improbable that a precise balance between varying nuclear and Coulomb energies could be preserved in all these cases simultaneously.¹⁰

I am grateful to Professor Gamow for a letter in which he discusses additional arguments that have been raised against his proposal (1).

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¹G. Gamow, *Phys. Rev. Letters* **19**, 759 (1967).

²P. A. M. Dirac, *Nature* **139**, 323 (1937); *Proc. Roy. Soc. (London)* **A165**, 198 (1938).

³P. Pochoda and M. Schwarzschild, *Astrophys. J.* **139**, 587 (1964); G. Gamow, *Proc. Natl. Acad. Sci. U. S.* **57**, 187 (1967).

⁴E. Teller, *Phys. Rev.* **73**, 801 (1948).

⁵C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes* (John Wiley & Sons, Inc., New York, 1967), 6th ed., p. 363; R. L. Brodzinski and D. C. Conway, *Phys. Rev.* **138**, B1368 (1965).

⁶G. Gamow and C. L. Critchfield, *Theory of Atomic Nucleus and Nuclear Energy Sources* (Oxford University Press, New York, 1949), p. 146.

⁷The exponent is $\{2 + [1 - (Z/137)^2]^{1/2}\}$ at $\Delta = 0$ and increases with Δ . See E. J. Konopinski, *Rev. Mod. Phys.* **15**, 209 (1943), Eq. (24c).

⁸The precise connection between w_{EC} and w_{β} may be written as

$$w_{\beta}(|\Delta|) = \int_0^{|\Delta|} d\epsilon (|\Delta| - \epsilon)^2 S(\epsilon),$$

$$w_{\text{EC}}(-|\Delta|) = \int_0^{|\Delta|} d\epsilon (|\Delta| - \epsilon)^2 S(-\epsilon).$$

Here the electron energy is $mc^2 \pm \epsilon$, with the plus sign for β decay, the minus sign for electron capture. The neutrino energy is $|\Delta| - \epsilon$ in both cases, and $S(\pm\epsilon)$ is the density of squared matrix elements for electron Coulomb wave functions with energy $mc^2 \pm \epsilon$. Although $S(x)$ is a continuous function for positive x and a discrete sum of δ functions for negative x , the correspondence principle ensures that its average strength is continuous across $x = 0$. Thus the average $w_{\beta}(|\Delta|)$ and $w_{\text{EC}}(-|\Delta|)$ will be approximately equal so long as the states contributing to $w_{\text{EC}}(-|\Delta|)$ are close to the continuum. The equality fails completely when $|\Delta|$ is so large that the K electrons contribute to $w_{\text{EC}}(-|\Delta|)$. An analogous situation exists in the theory of photoelectric excitation of electrons into bound and free states; see H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Academic Press, Inc., New York, 1957), pp. 308 and 335.

⁹I assume here that not all of the existing Os^{187} is radiogenic, an assumption which could easily be checked by isotopic analysis of osmium from various ores.

¹⁰Similar arguments have been applied to α -decaying nuclei by D. H. Wilkinson, *Phil Mag.* **3**, 582 (1958).

CONSTANCY OF THE FUNDAMENTAL ELECTRIC CHARGE

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In a recent Letter, Gamow¹ suggested that e^2 increases in time, in direct proportion to the age of the universe. The purpose of the present note is to rule out such a possibility on the basis of geological evidence.

If Gamow's hypothesis were true, then a pre-Cambrian chart of nuclides would have looked

very different from a modern one. The stable heavy elements would have N/Z ratios much closer to 1 (because the deviation of N/Z from 1 is due to the electrostatic repulsion between protons).² For instance, if e^2 were just a few percent lower, then U^{238} would be unstable against double beta decay to Pu^{238} , and a further decrease