Letters to the Editor

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The Radiations of RaD

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WITH a view to establishing the decay scheme of RaD,[1] the radiations of this element have been studied using a scintillation spectrometer. A summary of previous work on RaD up to 1949 has been given by Feather,[2] and more recent work has been carried out by Cranberg,[3] Frilley *et al.*,[4] and Butt and Brodie.[5]

The scintillation spectrometer resembles in principle the gas-filled proportional counter, which has previously been applied to the examination of the soft γ- and x-radiations of RaD.[6] However, it has a nearly 100 percent efficiency for the harder γ-rays[2] (37, 42.6, and 46.7 kev), which is a real advantage in view of the very low intensity of these radiations (<3 quanta per 100 disintegrations). If we assume that the main mode of decay consists in the emission of soft β-particles (estimated limiting energy[7,8] between 10 and 40 kev) followed by de-excitation from a 46.7-kev level of RaE, a coincidence system consisting of two scintillation spectrometers (Fig. 1, inset) offers a method of isolating and examining the β-particles of the primary spectrum.

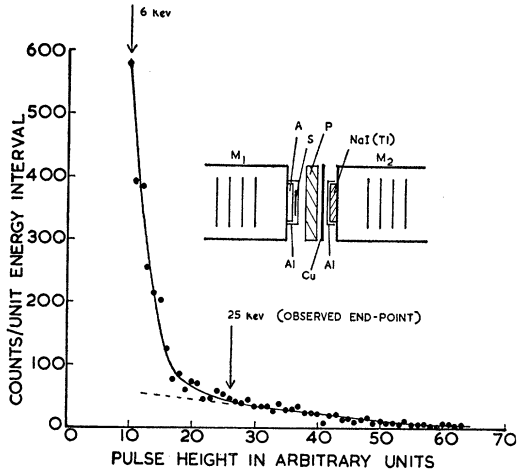

FIG. 1. The primary β-spectrum of RaD, isolated by coincidence with γ-rays of energy >40 kev. Inset: The coincidence arrangement employed in obtaining the primary β-spectrum. M_1, M_2: photomultiplier tubes, E.M.I. type VX 5032. A: thin anthracene crystal used as β-particle detector. S: separated source of RaD, mounted on Al foil and placed in direct contact with A. Al: aluminium reflector surrounding phosphor crystals. P: $\frac{1}{8}$-inch sheet of polythene to absorb β-particles from RaE. Cu: 2-mil copper sheet to reduce intensity of L x-rays of RaD. NaI(Tl): thin crystal of NaI(Tl) used to detect RaD γ-rays.

A separated source[3] of RaD was used which contained less than 5 percent RaE and RaF contaminant.

The spectrum of pulses from counter M_2 showed some of the known γ-rays of RaD, and the peak corresponding to the radiation of energy 46.7 kev stood out strongly (see Fig. 2 (A)). Careful search for harder radiations,[2] extending up to 140 kev, failed to reveal the presence of such in any significant intensity.

The output from counter M_1 was gated by those pulses from M_2 which corresponded to a γ-ray energy greater than 40 kev, and the resultant distribution analyzed with a single channel kick-sorter. The curve obtained is shown in Fig. 1. The region of the primary β-spectrum from 6 kev to the end point is clearly observed, and the coincidence control eliminates the photoelectron and Auger electron peaks which would otherwise complicate the results. A somewhat intense tail is found, the origin of which is not obvious but which is probably associated with the cosmic radiation and with bremsstrahlung effects from the slight admixture of RaE.

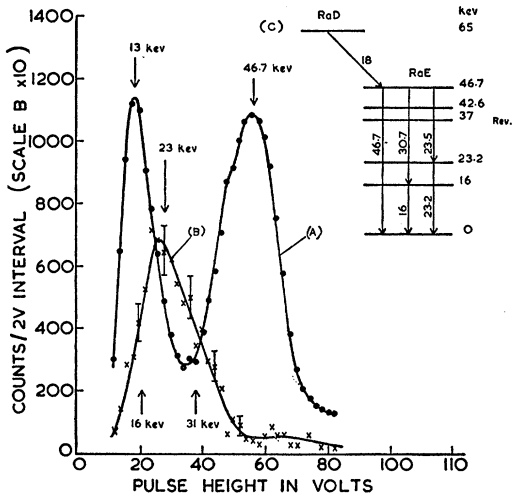

FIG. 2. (A) γ-ray distribution from RaD as observed in either counter of the $\gamma-\gamma$ coincidence arrangement, showing main γ-ray peak at 46.7 kev and x-ray peak at ~13 kev. (B) Pulse height distribution curve of γ-rays producing $\gamma-\gamma$ coincidences. (C) Suggested decay scheme for RaD, showing main transitions involved in $\beta-\gamma$ and $\gamma-\gamma$ coincidences. Other γ-ray transitions are not shown for sake of clarity.

According to the uncorrected energy scale, the end point appears at 25 kev. Calibration was performed with known x- and γ-radiations, and from the observed widths of the peaks for such homogeneous sources it was possible to correct for the energy resolution of the counter. This procedure established that the limiting energy of the β-rays was

$$E_0 = 18 \pm 2 \text{ kev}.$$

This indicates a total disintegration energy of 46.7+18, i.e., 65 kev approximately, for the transition RaD→RaE. The form of the spectrum was found to agree, within the rather wide experimental limits of accuracy, with that expected for an allowed β-transition.

Similar work, with the aforementioned system modified to observe $\gamma-\gamma$ coincidences, showed that the γ-rays in coincidence formed a rather broad distribution extending from about 16 kev to just over 30 kev, with a maximum in the region of 23 kev (Fig. 2 (B)).

Further, the integrated spectrum[9] was studied with a split crystal of NaI(Tl), within which a thin source of RaD was held. The form of this curve showed clearly that all the transitions from the level at 46.7 kev were not direct to ground nor of the straightforward cascade type. Indeed it seemed necessary to assume that at least one of the intermediate levels was metastable, with a lifetime greater than 1 μsec, the resolving time of the circuit. Further work aiming at the use of thinner sources is in progress.

The decay scheme which appears to be consistent with previous results on RaD and with our own findings is indicated in Fig. 2 (C). All previously observed γ-rays can be fitted into the

scheme, while those transitions shown account for the $\beta-\gamma$ and $\gamma-\gamma$ coincidence results described above. Either one or both of the 42.6- and 37-kev levels is probably metastable.

[1] Detailed examination, by means of proportional counters of the form of the β-spectrum, is described by Insch, Balfour, and Curran of this Department, in a paper to be published in *The Physical Review*.
[2] N. Feather, Nucleonics **5**, No. 1, 22 (1949).
[3] L. Cranberg, Phys. Rev. **77**, 155 (1950).
[4] Frilley, Gokhale, and Valadares, Compt. rend. **232**, 50 (1951); **232**, 157 (1951).
[5] D. K. Butt and W. D. Brodie, Proc. Phys. Soc. (London) **64**, 791 (1951).
[6] Curran, Angus, and Cockroft, Phil. Mag. **40**, 36 (1949).
[7] *Nuclear Data*, National Bureau of Standards Circular No. 499 (1950).
[8] H. O. W. Richardson and A. Leigh-Smith, Proc. Roy. Soc. (London) **A160**, 454 (1937).
[9] Bannerman, Lewis, and Curran, Phil. Mag. **42**, 1097 (1951).

Information and Thermodynamics

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THE connection between information as defined in communication theory and statistical entropy (essentially negative or missing information) has been clarified by the measurement-communication analogy,[1] leading to the following statements which include the second law. Our information about an isolated system can never increase (only by measurement, i.e., interaction with the system, can new information be obtained). Reversible processes conserve information, irreversible ones lose it. Theoretically maximal information, corresponding to a pure state, has zero entropy; minimal information, corresponding to equilibrium, has maximum entropy. If one considers measuring apparatus and system of interest as a compound isolated system, then by the first statement information (obtained by measurement) is amply compensated by entropy increase in the compound system, no measurement is possible for equilibrium between the two parts, and the positive time direction can be taken equally as that corresponding to entropy increase of the compound system or of information increase regarding the system of interest. The last is closer to psychological or subjective time and also corresponds to time measurement (clock is object of interest). Past and future are symmetrical for equilibrium and for the purely mechanical case (zero entropy, pure case; note use of mechanical to include electrical, magnetic, etc., classical or quantal).

Consider now the informational interpretation of the first law and of quantity of heat. The first law can be expressed by mechanical concepts (in the general sense) alone, with quantity of heat then defined by the conservation of energy.[2] The first law asserts the existence of a state energy function, *viz.*: to bring a system adiabatically (i.e., via well-defined mechanical operations) from a definite initial state to a definite final state requires the same amount of mechanical work independently of the sequence of intermediate states. The state function U so defined (energy) is fixed to within an additive constant; for adiabatic change from state 1 to state 2,

$$U_2-U_1=\int^2 dW. \tag{1}$$

The quantity of heat given a body going from 1 to 2 over a specific, not necessarily adiabatic, path is now defined by

$$\int_1^2 dQ=(U_2-U_1)-\int_1^2 dW. \tag{2}$$

From an information viewpoint quantity of heat is thus energy transferred in a manner which has eluded mechanical description, about which information is lacking in terms of mechanical categories. Adiabatic changes make (2) vanish and thus for heat to be transferred the path must contain states which cannot be reached adiabatically from the initial state. Applied to (2) in differential form, namely

$$dQ=dU-dW, \tag{3}$$

the existence of adiabatically inaccessible states in the neighborhood of a given state follows. But this is precisely Caratheodory's principle, from which the existence of an integrating denominator for the Pfaffian (3) then follows, and from which in turn absolute temperature and the entropy function can be derived. It is noteworthy that even if the informational or statistical interpretation of entropy were not at hand, the first law with quantity of heat informationally characterized as above would lead to it. For all states adiabatically accessible (equivalent informationally) from a given state would generate a "surface" which would be a member of a one-parameter family. The value of the parameter would be a measure of missing information relative to some standard state; the parameter can be shown to be a function of the entropy only.

To sum up, the laws of thermodynamics can be stated as:

(a) The conservation of energy.

(b) The existence of modes of energy transfer incapable of mechanical description.

In a sense (b) is implied in (a), for (a) without (b) is a mechanical theorem devoid of thermodynamic content.

(c) The third law is true by definition, for in a perfectly ordered state at absolute zero there is no missing information, i.e., the entropy is zero (pure case).

[1] J. Rothstein, Science **114**, 171 (1951) and references cited.
[2] A. Landé, *Handbuch der Physik* (J. Springer, Berlin, 1926), Vol. IX, Chapter IV gives this formulation of the first law and a readable account of Caratheodory's axiomatic formulation of thermodynamics.

Slow Neutron Fission of Am^{242}, Am^{242m}, and Am^{243}*

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THIS letter is to report the pertinent results of some work which was done at scattered intervals during the years 1947 to 1949 and which may now be published. Independent measurements of the same cross sections by a Canadian team[1] gave somewhat different results.

Shortly after the discovery[2] of Am^{242} in a sample of Am^{241} irradiated with pile neutrons, it became apparent that this nuclide had a large destruction cross section. In a second Am^{241} irradiation of three times the intensity of the first, only 1.4 times as much Am^{242} was formed as determined[3] by counting the beta-particles of Am^{242}. This ratio gave a destruction cross section for Am^{242} of approximately 10^4 barns. (This result is very insensitive to the cross section for the formation of Am^{242} as long as it is much less than 10^4 barns.) That this destruction was at least partially due to neutron capture to form Am^{243} became apparent with the discovery of this nuclide.[3]

When a mass spectrographic analysis of this second americium sample became available in December, 1949, it was possible to estimate the distribution of the destruction cross section of Am^{242} between fission and neutron capture to form Am^{243}. The composition of the americium was shown to be Am^{241} (99 percent), Am^{242} (0.5 percent), and Am^{243} (0.5 percent). From these data one calculates the following approximate cross sections: cross section for the formation of Am^{242}, $\sim$50 barns; neutron capture cross section of Am^{242} to form Am^{243}, $\sim$2000 barns; and total cross section for the destruction of Am^{242}, $\sim$8000 barns. Thus the fission cross section of Am^{242} must be $\sim$6000 barns. The estimate of 2000 barns for the cross section of the reaction $Am^{242}(n, \gamma)Am^{243}$ is somewhat smaller than a previous estimate[3] and is still subject to large errors. The principal uncertainties are in the estimation of the neutron flux.

In 1949 the fissionability of americium samples containing various amounts of the nuclides Am^{242} and Am^{243} was measured with a fission counter[4] in the thermal neutron column of the Argonne heavy water pile. Measurements on americium of the composition given above gave a fission cross section for Am^{242} of