Electromagnetic Spectrum of Radium D*

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(Received August 25, 1952)

The electromagnetic spectrum of RaD has been studied by the proportional counter spectrometry technique. The work of the Curie Laboratory on the gamma-ray spectrum has been generally confirmed. However, the presumed K x-ray lines are of much lower intensity and occur at a lower energy than previously reported. The L x-ray intensity is 22 per hundred disintegrations, indicating 0.63 conversion per disintegration and a conversion coefficient of 18.3 for the principal gamma-ray.

I. INTRODUCTION

THE radiations from RaD have been reported to include numerous gamma-rays of very low intensity as well as the more intense 46.7-kev γ, 7.9 kev γ, and the L x-rays of bismuth (Table I).[1–18] It was assumed until 1939 that the decay of RaD consisted of simple beta-decay followed invariably by de-excitation of a 46.7-kev state. In 1939, however, Amaldi and Rasetti reported the 43-kev gamma-ray.[7]

To date only one weak conversion line has been clearly demonstrated to be associated with a gamma-ray other than the 46.7-kev gamma.[1,6,13,14] Therefore, until further evidence is obtained, it must be assumed that the L-emission radiation observed results primarily from the internal conversion of the 46.7-kev gamma-ray.

Reviews of recent work can be found in papers by Feather,[6] Kinsey,[19] and Cranberg.[13]

TABLE I. Gamma-ray spectrum.

Gamma-ray line	Energy from previous determinations (kev)	Intensity from previous determinations (%)	Energy this paper (kev)	Intensity this paper (%)	References
A	46.7±0.1	3.5±0.4			1–14
B	43 ±1	0.2±0.1	41.5±1	0.1	7–9
C	37 ±1	0.2±0.1	37.0±0.5	0.1	8, 9, 12, 13
D	31.3±1	0.4±0.2	30.7±0.5	0.4	8, 9, 12
F	23.2±0.6	1.0±0.5	24 ±0.5	<0.3	12, 15, 16
G	16.1±0.4	0	16.1±0.3	~0.5	18
H	7.8	8 ±2			5, 15–17
J	65 ±5	0.2	62.5±1.0	0.02	12

* This work was supported in part by the U. S. Atomic Energy Commission.

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II. EXPERIMENTAL APPARATUS AND PROCEDURE

The detector used in this experiment was a brass proportional counter (18 in. long 4 in. cathode, 4-mil tungsten anode) filled with 62.3 cm of argon and 10.7 cm methane. The radiation was passed through Lucite collimators and entered the tube through a 0.005-in. beryllium window. For samples immediately after RaE removal no absorbers were used. For older samples a magnetic field or polystyrene absorber was used to eliminate RaE betas. The Lucite collimation served to reduce bremsstrahlung and secondary x-radiation from the surroundings.

After amplification the pulse spectrum was analyzed by three techniques: (1) Polaroid Land camera time exposures, of varying duration, of the oscilloscope pulse display; (2) sweep type, single channel differential discriminator, integrator, and Brown 10-millivolt recorder; (3) single channel differential discriminator, scaler and message register.

The energies of all lines were determined relative to the energy of the 46.7-kev gamma and the most prominent L peaks ($L\alpha = 10.8$, $L\beta$ 13.0). The intensities of all lines were determined relative to the 46.7-kev gamma.

The intensity of the 46.7-kev gamma is well known, having been studied by a number of different experimenters (Table I). The intensity given in Table I was determined by Von Droste.[5] The deviation is the unweighted average deviation of all determinations since 1930.[20] In the case of the L x-rays and 7.9-kev gamma-ray, the area under the peak was measured with a planimeter and compared with the 46.7-kev gamma-ray after making appropriate corrections for external absorption and detection efficiency. The low intensity gamma-rays were estimated by direct counting and comparison of the corrected count rate with the corrected count rate due to the 46.7-kev gamma-ray.

The L x-ray spectrum was observed from RaD

20 *Note added in proof:*—Re-examination of the spectrum with a glass proportional tube with aluminum cathode confirms the report of Wu, Boehm, and Nagel (Bull. Am. Phys. Soc. **28**, No. 1, 59, 1953) that the 7.9-kev radiation is Cu x-radiation excited in brass tube walls. Dr. Wu reports (private communication) an intensity of 7±2 percent for the 46.7-kev gamma-ray. Note that intensity values reported above are based on an intensity 3.5±0.4 for this gamma. The new value would indicate 0.8 to 1.6 conversions per disintegration.

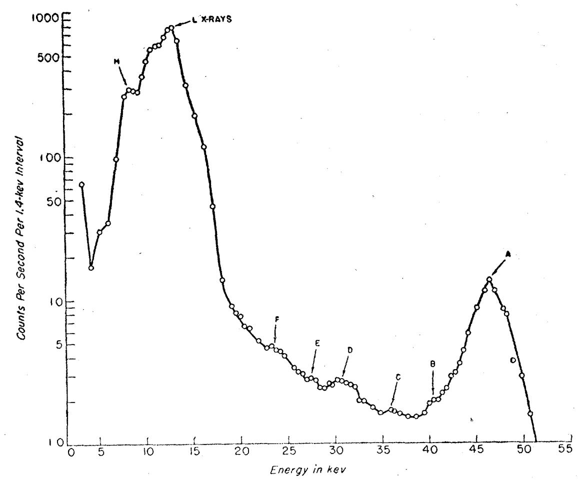

FIG. 1. Gamma-ray spectrum. 20-microcurie RaD–E–F source (spent radon seed) with polystyrene absorber.

samples of about 3 microcuries activity immediately after separation from RaE. Two different samples were used for the measurement of the gamma-ray spectrum. One RaD–E–F sample of 20-microcuries activity was separated from spent radon seeds, and the second 100-microcurie sample was prepared from natural "radiolead nitrate." The radon-produced sample was assumed to be free of contamination. Emanation measurements on the 100-microcurie source set limits of 0.15 and 0.001 percent, respectively, on radium and mesothorium I contamination.

III. EXPERIMENTAL RESULTS

A. Gamma-Ray Spectrum

The gamma-ray lines listed in Table I have all been determined previously by the method of crystalline diffraction or by the observation of cloud-chamber tracks. We have verified their existence by the proportional counter technique.

Gamma-ray A (46.7 kev) is shown in Figs. 1–3 and in the photographs (Fig. 4). As previously mentioned, the energy and intensity of this line were used as a standard.

Gamma-ray B (41.5 kev), shown in Figs. 1–3, occurs at a lower energy than previously reported.

Gamma-rays C (37 kev) and D (30.7 kev) occur at the same energy and intensity as previously reported.

Gamma-ray F (24 kev) is quite distinct in Fig. 1 and may also be seen in Figs. 2 and 5. We find the intensity to be closer to the value given by Curran[16] than to that of San-Tsiang Tsien.[15] Our early work indicated two gamma-rays in the region between 20 and 25 kev. However, subsequent work has failed to yield sufficient evidence to confirm this. Coincidences between L x-rays would produce a tendency to peak in this region.

Gamma-ray G (16.1 kev) is resolved from the $L(\gamma)$ x-ray in Figs. 5 and 7. We have estimated its intensity to be 0.5 percent in contrast to the very low intensity previously reported.

Gamma-ray H (7.9 kev) stands out prominently in Figs. 5 and 6. It is also distinctly resolved as the lower of four lines in the left-hand set of photographs (Fig. 4). The precise position of this line relative to the L x-rays was determined from ten separate observations. It is difficult to determine the intensity precisely since the Ll x-rays of bismuth which are not resolved fill the gap between this gamma-ray and the $L\alpha$ x-rays and obscure the shape of the line. There will be a small contribution to this peak from $L\alpha$ x-rays when the K x-ray of argon escapes without absorption in the counter gas. However, it seems fairly certain that the correct intensity is not as high as was previously reported.[20]

Gamma-ray J (62.5 kev) was reported in 1945 by San-Tsiang Tsien[12] and has never been confirmed. This gamma-ray is distinctly resolved in Fig. 3. Brown recorder traces invariably show a bulge if not a distinct peak in the region between 60 and 65 kev.

B. X-Ray Spectrum

The $L\alpha$, β, γ x-rays of bismuth are shown in Figs. 5 and 6. These lines are also clearly discernible in three of the four sets of photographs (Fig. 4).

The intensity of the L x-rays was carefully compared to the intensity of the principal gamma (3.5/100). From this we conclude that about 22 x-rays are emitted per 100 disintegrations. Using Kinsey's[19] figures for the fluorescence yield and L-conversion fraction, the total number of conversions per disintegration is 0.63.[20] This is in good agreement with the work of Stahel,[2] Butt and Brodie,[14] and Cranberg[13] but does not agree with the value obtained by Curran *et al.*[16] (33/100). Assuming that most of the L x-rays result from conversion of the principal gamma (A), the conversion coefficient is 18.3.

The ratios of the $L\alpha$, $L\beta$, $L\gamma$ are 1.0/0.88/0.15. These ratios are subject to some error since the Ll and $L\eta$ x-rays are not resolved. The estimated intensity of the 16.1-kev gamma-ray was subtracted from the $L\gamma$ peak. In those cases where these two were not resolved, the spread and high intensity of the unresolved peak was

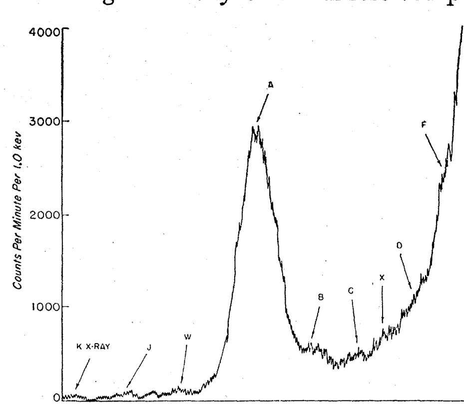

FIG. 2. Gamma-ray spectrum. 100-microcurie RaD–E–F source (natural radiolead nitrate) with polystyrene absorber.

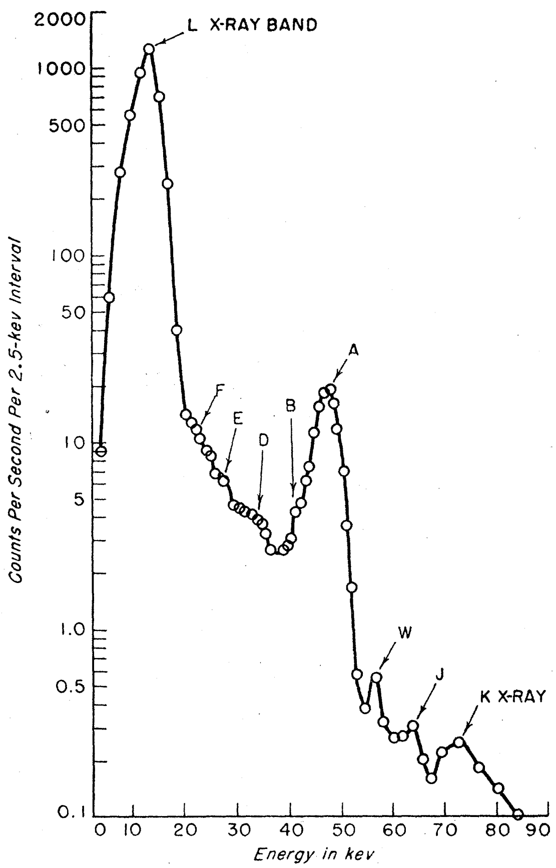

FIG. 3. Gamma-ray spectrum. 20-microcurie RaD–E–F source (spent radon seed) with polystyrene absorber.

an indication of complex structure. If no allowance is made for complexity, the $L\alpha$, $L\beta$, $L\gamma$ ratios are 1.0/0.88/0.23.

Two peaks in the K x-ray region have been observed. One distinct peak (71 kev) may be seen in Fig. 3. The other peak, which is of much lower intensity, occurs at about 85 kev. The energy of the 71-kev peak corresponds to the $K\alpha$ x-radiation of thallium rather than bismuth as previously reported. The total K x-ray intensity (around 0.1) is only 10 percent of the value previously reported (1.0).[8]

The M x-rays of bismuth may also be seen in Fig. 5. We have as yet made no effort to determine their intensity.

C. Additional Lines in the Proportional Counter Spectrum

Line W (57 kev) is clearly resolved in Fig. 3. On Brown recorder tracings (Fig. 2) a bulge or sometimes a distinct peak is invariably observed at this energy. This peak could result from nonrandom coincidences between L x-rays and the 46.7-kev gamma-ray.

Line X (34.5 kev) occurred only in the 100-microcurie RaD–E–F sample (Fig. 2), which was obtained from natural "radiolead nitrate." Measurements on the 20-microcurie radon produced sample indicate that radiation of this energy does not follow radon in the radium decay series.

Line Y (18.6 kev) is a broad, low intensity peak which may be seen most clearly in Fig. 7. This broad peak could result from nonrandom coincidences between the 7.9-kev gamma-ray (H) and L x-rays.

Line Z (17.3 kev) was recently reported by Cohen and Jaffe.[17] There appears to be an indication of a line of this energy in Fig. 7 and perhaps Fig. 6. However, its existence cannot be asserted from these data.

The line in Fig. 5 designated "x-ray?" occurs at an energy slightly greater than the energy of the gamma-

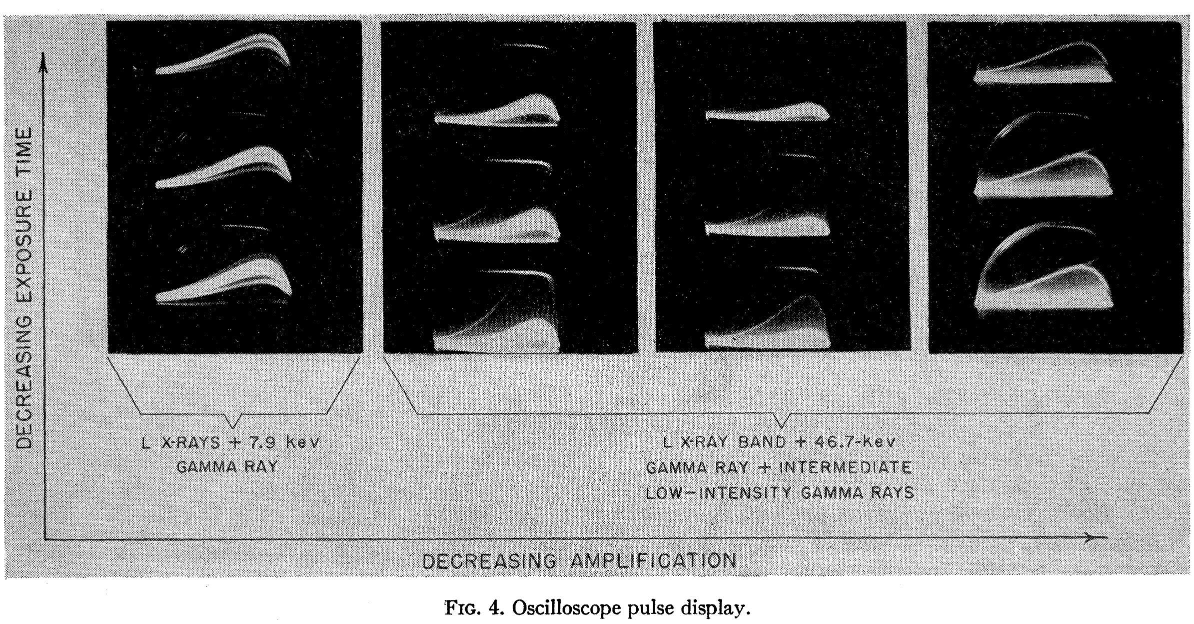

FIG. 4. Oscilloscope pulse display.

ray H (7.9 kev) minus the energy of the argon K x-ray. This peak could be due to the escape of argon K radiation. However, a K x-ray peak due to iron contamination in the beryllium window is also expected at nearly the same energy (6.4 kev).[21]

Line E (28.0 kev) visible in Figs. 1 and 3 occurs at the energy of the 30.7-kev gamma-peak (D) minus the energy of the argon K x-ray. This could also be an argon x-ray escape peak.

D. Nuclear Isomerism

A search was made for the existence of metastability using a variable resolution time method (to be described in a forthcoming paper) and by analysis of oscilloscope-motion picture photographs of pulse distributions. It has been shown that the half-period of any such state must be less than 2 microseconds or greater than 50 milliseconds, provided its intensity is not less than 10 percent. Bannerman and Curran[22] report a metastable state of half-period greater than 2 microseconds. However, they assign the metastability to the 37-kev (B) or 42-kev (C) low intensity gamma-ray.

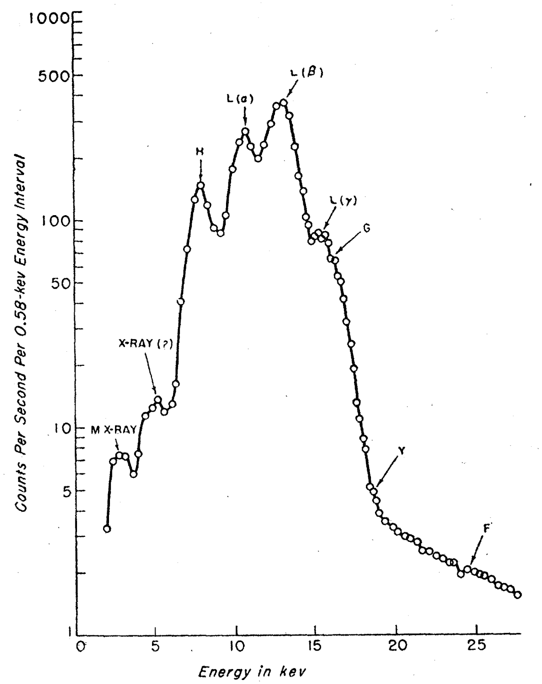

FIG. 5. L x-ray and low energy gamma-ray spectrum. 20-microcurie RaD–E–F source (spent radon seed) with magnetic field.

IV. CONCLUSIONS

The appearance of the 34.5-kev gamma-ray in the sample of RaD–E–F separated from uranium minerals, and its absence in the spent radon seed suggests that perhaps much of the variability in earlier results may be traced to the origin and purification of the source material. The wide variations in reported intensity of

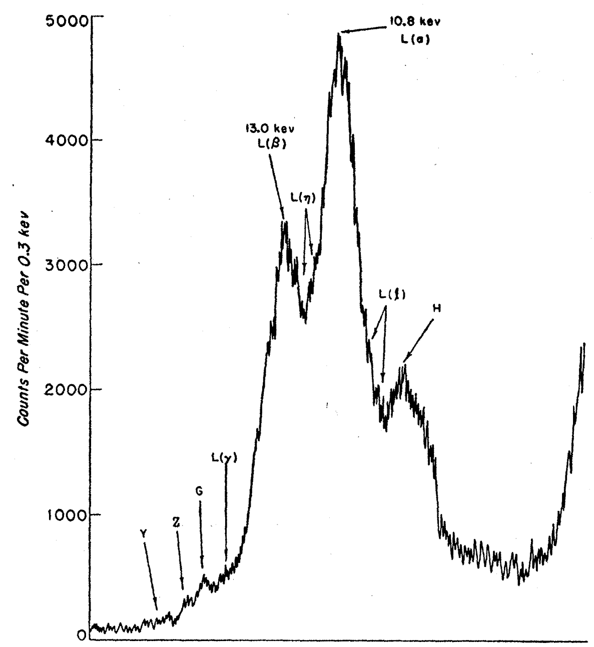

FIG. 6. L x-ray and low energy gamma-ray spectrum. 3-microcurie RaD source with minimum absorption.

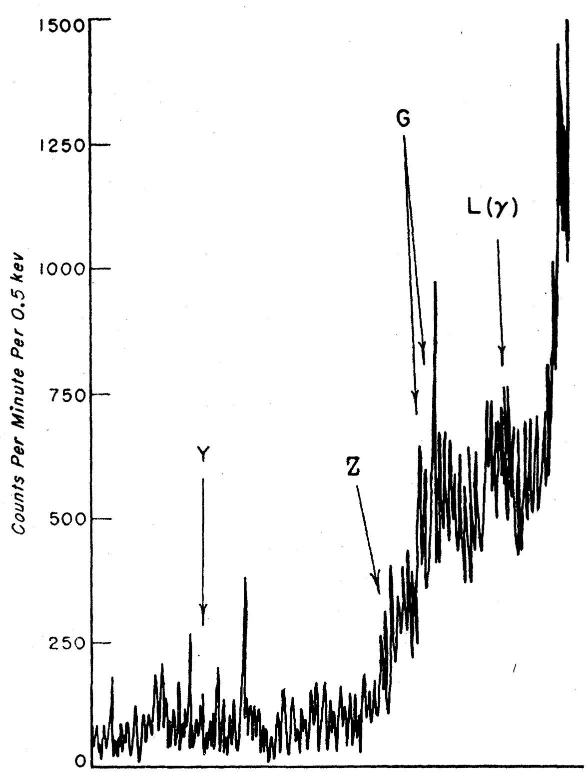

FIG. 7. $L(\gamma)$ x-ray region. 3-microcurie RaD source with minimum absorption.

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TABLE II. X-ray spectrum.

X-ray	Energy from previous determinations (kev)	Intensity from previous determinations (%)	Energy this paper (kev)	Intensity this paper (%)	References
$K\alpha$ x-ray	77	0.8±0.3	71±2	0.06	8
$K\beta$ x-ray	87	0.2	85±2	less than 0.02	8
L x-ray	9−16	20−35	9−16	22	2, 7, 16
M x-ray	...	...	1.9−3.4	...	...

lines in the K x-ray region is an additional illustration of the difficulty. A striking dearth of information on source materials exists in the published papers on both the β- and γ-spectra of RaD, and on the β-spectrum of RaE. The question of possible complexity in the latter must be left open until the complete decay scheme of RaD is elucidated, since the L x-ray intensity suggests that the 46.7-kev state is promptly traversed by less than 80 percent of the RaD transitions, and no high energy β-branch has been observed in RaD decay.[23]

TABLE III. Additional lines in proportional counter electromagnetic spectrum.

Line description	Energy this paper (kev)	Intensity this paper (%)	Remarks
W	57 ±1	0.02	Possible coincidence peak
X	34.5±1	0.1	Not associated with RaD
Y	18.6±1	0.3	Possible coincidence peak
Z	17.3±0.3	∼0.3	Peak reported by Cohen and Jaffe[a]
X-ray?	5.5±0.5	0.3	Fe x-ray or argon K x-ray escape peak
E	28.0±0.5	0.05	Possible argon K x-ray escape peak

[a] See reference 17.

The authors wish to express their appreciation to Mr. J. M. Day for source preparation and to Mr. C. R. Alls and Mr. H. I. Hyde for assistance on instrumentation.

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PHYSICAL REVIEW VOLUME 90, NUMBER 2 APRIL 15, 1953

A Nonperturbation Approach to Quantum Electrodynamics

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(Received December 31, 1952)

The equations governing the interaction of an electron with the electromagnetic fields are used in the form given by Schwinger to derive a linear integral equation for the function Γ, whose kernel is expressed as a power series in α. The first approximation to this kernel is used and the resulting integral equation solved without recourse to perturbation theory. With the aid of the solution first approximations can be found for self-energy effects, which are now finite, and some discussion of the analytic behavior of these and related quantities is given. An application of the method to meson theory illustrates the classification of the types of integral equation which arise. The possibility of extending the method is discussed.

1. INTRODUCTION

PERTURBATION theory has been extremely successful in electrodynamics in explaining experimental results since the renormalization program has been adopted. But almost all experimental results are associated with interactions in which the integrals concerned are quite convergent after renormalization, whereas for self-energy effects as well as the other renormalization coefficients there has been a complete failure to obtain finite results. At this juncture two alternative approaches suggest themselves: (1) the Lagrangian used is not adequate to handle self-energy effects, though accurate enough for interaction effects, and new ideas are required to attack these problems, (2) we may object that since only the simplest mode of solution has been used, that of straightforward expansion, some more powerful approach is required, which would also be superior to perturbation theory when the coupling constant was large. The self-energy of the electron, for example, is expressed in perturbation theory by

$$m=e^2(m_1+e^2m_2+e^4m_3+\cdots),$$

where all the m's are infinite. Such an expansion is only valid if me^{-2} is analytic in e^2 at the origin, and there is no *a priori* reason to believe this. A final point in favor of the second approach is that many theories, such as meson theory with gradient coupling, diverge still after renormalization, and it is not clear if this is not also due to the method employed.

It is this second approach which is considered here, and an attempt to solve the quantum electrodynamics of electrons without recourse to perturbation theory is presented below. The formulation of Schwinger,[1] in

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[1] J. S. Schwinger, Proc. Natl. Acad. Sci. 37, 452 (1951). We follow Schwinger's notation conventions.

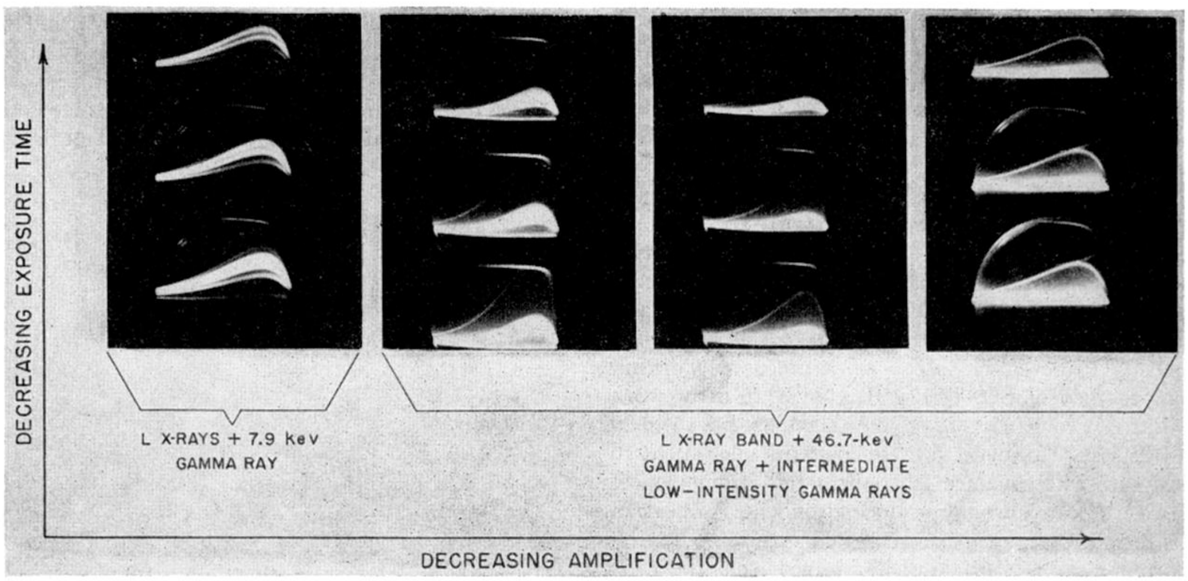

FIG. 4. Oscilloscope pulse display.