Characteristics of Ni^{59} and Ni^{63}

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Separated stable isotopes of Ni have been used to assign a 67-kev maximum energy beta-emitter which decays with an 85-yr half-life to Ni^{63}. Ni^{59} was found to decay by K electron capture with a half-life of 7.5×10^4 yrs.

I. INTRODUCTION

CAMAC and Brown[1] produced long-lived nickel activities by (d,p) and (n,γ) radiations on nickel and by an (α,n) reaction on iron. They identified the radiations as 50-kev electrons and x-rays. From yield data the radiations were estimated to decay with a half-life of 12 ± 6 yrs. Assignment of both radiations to Ni^{59} was based partly on production by an (α,n) reaction on iron and partly on the previous assignment of a 2.6-hr period to Ni^{63}. Later Swartout[2] showed by (n,p) reactions on separated copper isotopes that the 2.6-hr period should be assigned to Ni^{65}. This mass assignment was confirmed by Conn[3] from (n,γ) yields on separated nickel isotopes. In confirming the mass assignment of the 2.6-hr isotope, evidence was found for assignment of the x-radiations reported by Camac and Brown to Ni^{59} as they had proposed. The (n,γ) yields indicated, however, that the soft beta-radiation should be assigned to Ni^{63}.

An attempt has been made to determine the decay constants of Ni^{59} and Ni^{63} by measuring the x-radiation and the soft beta-radiation separately over a period of 3 yrs. Since these measurements were started, Pomerance[4] has measured the isotopic absorption cross sections of the nickel isotopes. These cross sections were used to make preliminary estimates of the Ni^{59} and Ni^{63} half-lives for the Table of Isotopes.[5] Further work has been done to eliminate a large and uncertain correction for self-absorption of the Ni^{63} beta-ray. Quantitative data are presented on (n,γ) yields from which mass assignments have been made and half-lives calculated.

II. MASS ASSIGNMENT OF Ni^{63}

When nickel with the natural isotopic composition is activated in the Oak Ridge National Laboratory reactor, two types of radiation are found associated with the long-lived nickel activities. Using a G-M counter with a mica end window several mg/cm² in thickness, an x-ray component only is observed. When a windowless counter is used, a soft beta-component also is observed with a counting rate several hundred times that of the x-ray component as shown by the aluminum absorption curve in Fig. 1.

In order to assign these radiations to the proper isotopes, (n,γ) yields from separated nickel isotopes** have been measured. In preliminary work it was found that repeated purification of the nickel was necessary in order to remove iron and cobalt activities formed by (n,α) and (n,p) reactions, respectively. It was also found necessary to prepare very thin uniformly spread samples in order to eliminate errors caused by absorption of the soft beta-radiation within the samples. Samples prepared by electrodeposition of nickel on platinum gave reproducible data as shown by the self-absorption curves for three different specific activities in Fig. 2. Further evidence that errors from self-absorption could be made small was obtained by deposition of a mass of nickel equivalent to a monatomic layer on a platinum plate. This sample gave the same value for the counting

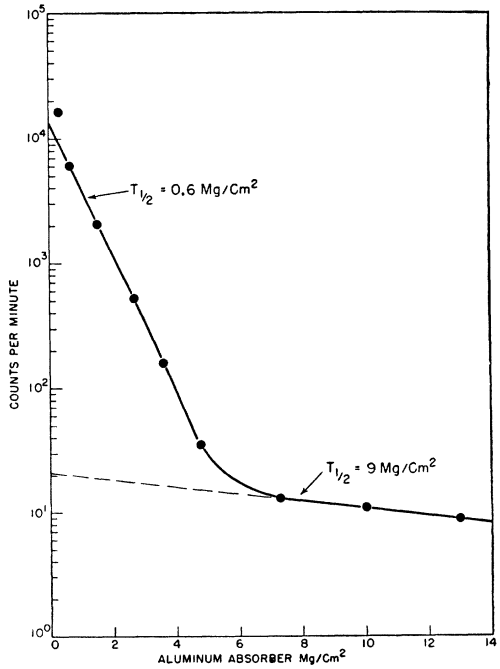

FIG. 1. Aluminum absorption curve of Ni^{63} beta-radiation.

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[1] M. Camac and L. Brown, Metallurgical Project Report CP-2407, Oct., 1944, unpublished.

[2] J. A. Swartout *et al.*, Phys. Rev. **70**, 232 (1946).

[3] E. E. Conn *et al.*, Phys. Rev. **70**, 768 (1946).

[4] H. S. Pomerance, Phys. Rev. **76**, 195 (1949).

[5] G. T. Seaborg and I. Perlman, Rev. Modern Phys. **20**, 585 (1948).

** Obtained from the Y-12 plant, Carbide and Carbon Chemicals Division, Oak Ridge, Tennessee.

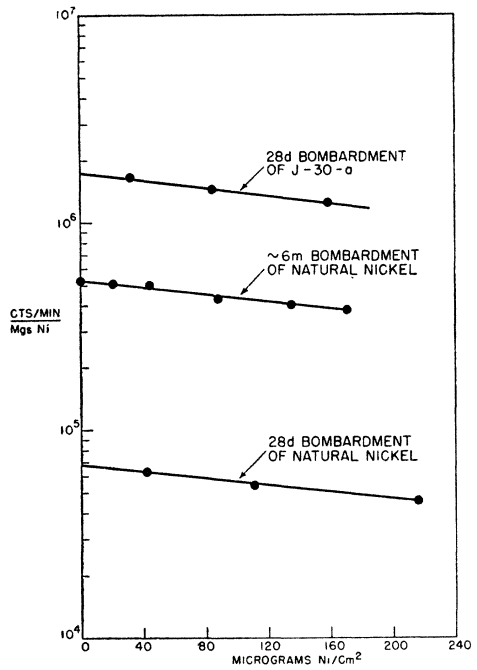

FIG. 2. Self-absorption curves of Ni^{63} beta-radiation.

rate per mg of nickel as the extrapolation to zero mg/cm² of the self-absorption curve of nickel with the same specific activity.

When preliminary data indicated that errors from self-absorption could be made negligibly small, three nickel samples were bombarded with cobalt flux monitors in the Oak Ridge reactor for 28 days. One of these (K-33a) was enriched in Ni^{58}, a second (J-30a) was enriched in Ni^{62}, and a third had the natural isotopic composition. After bombardment the samples were purified by the removal of iron and cobalt activities and finally by repeated precipitations of nickel dimethylglyoxime. Various thicknesses of each nickel sample were then electroplated onto platinum discs. These samples were counted in a flow type proportional counter[6] and the activity with no self-absorption was obtained by extrapolation to zero sample thickness as shown in Fig. 2. Since the counting rates of samples prepared from nickel depleted in Ni^{62} (K-33a) were too low for measurement of thin sources, these were compared with natural nickel using "infinitely thick" sources. Specific activity data along with mass assay data furnished by the Y-12 laboratory are given in Table I. Within the limits of error of the isotopic assay data, the soft beta-counting rate per mg of Ni^{62} bombarded is constant. Since the activity is formed by an (n,γ) reaction, the soft beta-radiation is associated with the decay of Ni^{63}.

III. Ni^{63} RADIATION

Magnetic deflection experiments showed that the radiation from Ni^{63} was soft electrons. The absence of annihilation radiation from 100 microcurie sources showed that negatrons rather than positrons were emitted by Ni^{63}. Feather analysis of aluminum absorption curves gave a range of 6.6 mg/cm² corresponding to an energy of about 65 kev.

[6] C. J. Borkowski, Anal. Chem. **21**, 348 (1949).

TABLE I. Comparison of (n,γ) yields of soft beta-emitter with Ni^{62} assay.

Nickel sample	Ni (β counts/ min/mg)	Ni^{62} (%)	Ni^{62} (β counts/ min/mg)
J-30a	1.75×10^6	94.25 ± 0.5	1.86×10^6
Natural	6.80×10^4	3.66 ± 0.01	1.86×10^6
K-33a	3.08×10^3	0.13 ± 0.05	$2.37\pm0.9\times10^6$

A more precise measurement of the maximum beta-energy has been made using a proportional counter spectrometer.[7] For these measurements the nickel was electroplated on platinum to a thickness of 10 μg/cm². The pulse height distribution in the form of a Kurie plot is shown in Fig. 3. The energy scale was calibrated using the K x-radiations of Co, Xe, and Eu. Data on soft beta-emitters such as Tm^{171} and S^{35} that have been measured with both the thin lens spectrometer and the proportional counter spectrometer show that, although the shape of the Kurie plot is somewhat changed by the platinum backing, the end point is not appreciably affected. Hence no special significance is attached to the shape of the Kurie plot, but the maximum energy of 67 ± 2 kev is thought to be reliable and is in fair agreement with the value of 63 ± 2 kev given by Wilson and Curran.[8]

IV. Ni^{63} DECAY CONSTANT

In order to use the activation data in Table I to calculate a half-life from the neutron absorption cross section, it is necessary to know the counting yield of the counter. Since the counting yield might be expected to vary with maximum beta-energy the counting yields for several soft beta-emitters with known decay schemes

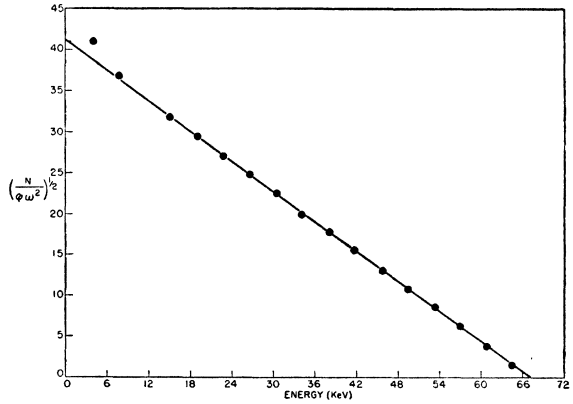

FIG. 3. Kurie plot of Ni^{63} beta-energy spectrum.

[7] C. J. Borkowski and E. Fairstein, Phys. Rev. **77**, 759 (1950).
[8] H. W. Wilson and S. C. Curran, Phil. Mag. **40**, 631 (1949).

were determined. These were Ru^{106}, Nb^{95}, and Co^{60} with maximum beta-energies[9] of 0.039 Mev, 0.147 Mev, and 0.31 Mev, respectively. Ru^{106} decays to 30 sec Rh^{106} which has a known decay scheme.[10,11] Co^{60} and Nb^{95} have been investigated by numerous[5] workers who are in essential agreement concerning the decay schemes. Since all these decay schemes have gamma-rays in coincidence with the beta-ray, it is possible to determine disintegration rates of these isotopes by coincidence methods. The counting yield is the ratio of the observed counting rate and the disintegration rate.

All sources were counted under the same conditions as was Ni^{63} using essentially weightless samples mounted on "infinitely thick" platinum. The cobalt and ruthenium sources were prepared by electroplating and the niobium sources were prepared by adsorption. In each case reduction in counting rate from self-absorption was shown to be negligible by counting samples which varied in thickness by a factor of five. The agreement in counting yield between sources of the same radiation was within the counting error of 2 percent. The counting yield data for the proportional counter used are given in Table II.

Although reproducible counting yields were found for the beta-emitters measured, there is still some uncertainty concerning the correct counting yield for Ni^{63} beta-radiation. Factors contributing to this uncertainty are the effect of differences in electron energy distributions and the correct method of interpolation between the beta-energies given in Table II. Graphical interpolation from a smooth curve gives a counting yield of 56 percent for beta-radiation with a maximum energy of 67 kev.

TABLE II. Counting yields of low energy beta-emitters.

Beta-emitter	Maximum energy (kev)	Counting yield (%)
Ru^{106}	39	53.5
Nb^{95}	147	62.1
Co^{60}	310	68.8

The activation data in Table I have been converted to disintegration rates using 56 percent as the counting yield. From these data and the disintegration rate of the Co^{60} activity induced in the neutron flux monitor the half-life of Ni^{63} can be calculated using the equation

$$T_1 = \sigma_1 N_1 D_2 T_2 / \sigma_2 N_2 D_1, \quad (1)$$

where T is the half-life, σ the cross section, N the number of atoms bombarded, and D the induced disintegration rate. The subscripts 1 and 2 refer to $Ni^{62}(n,\gamma)Ni^{63}$ and $Co^{59}(n,\gamma)Co^{60}$ reactions, respectively. Numerical values of the variables used to calculate a Ni^{63} half-life are given in Table III along with estimated probable errors.

TABLE III. Calculation of a Ni^{63} half-life.

Variable	Numerical value	Probable error
σ_1	14.8 barns	20%
σ_2	34.5 barns	5
D_1	3.32×10^6 d/m/mg Ni^{62}	5
D_2	1.31×10^8 d/m/mg Co^{59}	2
T_2	5.3 yrs	5
T_1	85 yrs	20 yrs

The half-life of 85 yrs for Ni^{63} calculated from the data in Table III is considerably shorter than values previously estimated[12] from activation data. Since the neutron energy distribution for the activation experiments was somewhat different from that used in measuring the cross sections, resonance absorption could lead to an error in the half-life. Therefore, nickel enriched in Ni^{62} and a Co^{59} flux monitor were activated in the pile oscillator where the cross sections had been measured. The uncertainty with respect to the correct cross section was removed since these activation data also gave a half-life of 85 yrs for Ni^{63}.

Ionization currents, produced by two electroplated nickel samples containing Ni^{63} and by uranium and RaD standards, have been measured over a period of 3 yrs. Other Ni^{63} samples have been counted along with standards on a proportional counter for shorter periods. Although the relative probable errors in the half-life measurements of individual samples are of the order of 20 percent, the apparent half-lives vary from 35 to 60 yrs. Because of the low half-thickness of the Ni^{63} beta-particle (0.5 mg/cm^2) oxidation of a few hundred lattice layers or diffusion through twenty or thirty lattice layers over a 3-yr period could reduce the apparent activity by the amount observed.

Comparison of the beta-spectrum of Ni^{63} when first electroplated with the spectrum after the sample had stood in air for several months has shown that the relative number of low energy electrons decreases with time. Because of this evidence that changes occur in electroplated nickel samples, it is believed that the apparent decay gives only minimum values for the half-life. It is concluded, therefore, that the best value for the half-life of Ni^{63} is 85 yrs as given by the activation experiments.

V. MASS ASSIGNMENT OF Ni^{59}

When reactor activated nickel samples are counted with a G-M tube, radiation with an 8.8 mg/cm^2 half-thickness in aluminum is observed after the 2.6-hr Ni^{65} and 36-hr Ni^{57} have decayed. Introduction of a magnetic field between the sample and the G-M tube has no effect on the counting rate. X-rays with an energy of about 7 kev are therefore being counted. These could be Co K x-rays from K capture decay, Ni K x-rays from internal conversion in a metastable state or Ni x-rays

[9] H. M. Agnew, Phys. Rev. **77**, 655 (1950).
[10] W. C. Peacock, Phys. Rev. **72**, 1049 (1947).
[11] E. T. Jurney, Phys. Rev. **76**, 290 (1949).
[12] G. T. Seaborg and I. Perlman, Revs. Modern Phys. **20**, 585 (1948); G. Friedlander, BNL-AS 2, p. 49 (1949) (to be published).

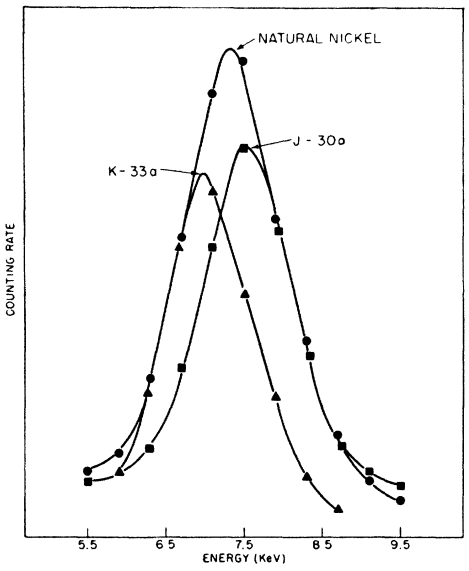

FIG. 4. Automatically recorded proportional counter spectrometer curves of the x-rays emitted by neutron bombarded nickel samples of different isotopic composition. Points are indicated for identification only.

produced by bombardment of Ni in the sample with the soft Ni^{63}-electrons.

In order to establish the origin and the energy of the x-rays, proportional counter spectrometer measurements were made on nickel samples of different isotopic composition after a 28-day bombardment in the ORNL reactor. The spectrometer curves in Fig. 4 show that the x-rays in Ni^{58} rich sample K-33a were almost pure Co K x-rays. Both Co and Ni K x-rays were found in the natural nickel sample, while the x-rays in Ni^{62} rich J-30a were Ni x-rays plus a continuum of harder x-radiation.

The Ni K x-rays probably originate from the bombardment of nickel by the Ni^{63} beta-radiation. The yield of about 2 Ni K x-ray quanta per 1000 Ni^{63} β disintegrations is consistent with yields quoted by Compton and Allison[13] for the efficiency of x-ray production by electron bombardment.

The Co x-rays are associated with Ni^{58} in the bombarded sample and must therefore result from K capture decay of Ni^{59}. Additional evidence that Ni^{59} decays by K capture was found with the nickel fraction from a group of essentially nickel-free cobalt targets bombarded with 14-Mev deuterons in the MIT cyclotron.† This nickel is of interest because a $d,2n$-reaction on Co^{59} gives Ni^{59}. The x-rays from this nickel were found to be essentially pure Co K x-rays.

VI. Ni^{59} DECAY CONSTANT

If it is assumed that the x-radiation is produced by the beta-radiation from Ni^{63} and by K capture decay of Ni^{59} and by no other means, the observed counting rate will be given by the relationship,

$$\text{x-ray counts/min} = A_1 k_1 (\beta^- \text{ d/m}) + A_2 k_2 (\text{mg Ni}^{58}). \quad (2)$$

Here A_1 and A_2 correct for the absorption in the nickel sample of Ni K x-rays and Co K x-rays, respectively. The term k_1 is the product of the Ni K x-ray production efficiency of Ni^{63} beta-particles and the counting yield of Ni K x-rays. The term k_2 is the product of the K capture disintegration rate per mg of Ni^{58} bombarded, the fluorescent yield of Co K x-rays and the counting yield of Co K x-rays. In cases where the samples are thin, an additional term correcting for the fraction of Ni^{63} beta-energy absorbed in the sample is required.

X-ray counting data on nickel samples of different isotopic composition which were bombarded in the ORNL reactor for 28 days, then purified and counted after decay of the short-lived nickel isotopes, are given in Table IV. The value of k_2 obtained from these data using Eq. (2) is 6.0 with a probable error of about 10 percent because of uncertainties in the analyses of absorption curves.

Since the fluorescent yield of Co K x-rays is known, it is necessary to know only the counting yield of the counter for Co K x-rays to determine the K capture disintegration rate from k_2. This counting yield was measured using a standard sample of Ni^{59} produced in the MIT cyclotron. It was prepared by electroplating about 10 mg/cm² of nickel onto platinum and was covered with a one mil aluminum foil. From the ionization current measured when the sample was placed at the center of a spherical ionization chamber, from the chamber radius, gas density, mass absorption coefficients, x-ray energy and the energy to produce an ion pair, the number of quanta per minute radiated from the surface was calculated. The probable error in the number of quanta calculated from measurements with different gases at several different pressures was about 5 percent.

TABLE IV. X-ray counting data.

Sample	β^- d/m/mg Ni	Amount of Ni (mg)	Amount of Ni^{58} (mg)	A_1	A_2	X-ray (counts/min)
K-33a	5.49×10^3	23.45	23.2	0.79	0.74	106
Natural Ni	1.21×10^5	29.1	19.7	0.74	0.69	185
J-30a	3.12×10^6	22.2	0.5	0.80	0.76	2100

Another determination of the number of quanta per minute leaving the source was made with a 2π geometry proportional counter filled with xenon to a pressure such that 95 percent of the x-rays were absorbed. The counting rate was in good agreement with that expected from the ionization chamber measurements. The Gold integral[14] was used to make corrections for absorption in the one-mil aluminum and self-absorption in the

[13] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935).

† The authors wish to thank Prof. J. W. Irvine for this sample of nickel activity.

[14] E. Gold, Proc. Roy. Soc. (London) **A82**, 62 (1908).

nickel to obtain the total Co K x-ray emission rate in the standard.

The counter used to obtain the data in Table IV was found to have a counting yield of 0.016. Since the fluorescent yield of Co K x-rays is 0.33, the K capture disintegration rate per mg of Ni^{58} bombarded is 1130 $\pm$170 per min. Equation (1) can now be used to calculate the Ni^{59} half-life with subscript 1 referring to the $Ni^{58}(n,\gamma)Ni^{59}$ reaction and subscript 2 to the (n,γ) reaction on Co^{59} as before. The data used in the calculation are given in Table V along with estimated probable errors. These data give $7.5\pm1.3\times10^4$ yrs for the K electron capture half-life of Ni^{59}.

TABLE V. Calculation of Ni^{59}-half-life.

Variable	Numerical value	Probable error
σ_1	4.2 barns	5%
σ_2	34.5 barns	5
D_1	1130 d/m/mg Ni^{58}	15
D_2	1.31×10^8 d/m/mg Co^{59}	2
T_2	5.3 yrs	5
T_1	7.5×10^4 yrs	1.3×10^4 yrs

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Diffusion of High Energy Gamma-Rays through Matter. I. Fundamental Equations*

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The fundamental processes, photoelectric absorption, Compton scattering, and pair production, leading to the modification of the energy spectrum and angular distribution of gamma-rays diffusing through matter are discussed in relation to the problem of penetration of gamma-rays through matter. The transport equation describing the diffusion is derived in the approximation in which radiation from the secondary electrons produced by the photoelectric effect and pair production is neglected. By limiting one's interest to gamma-rays of energy greater than a few Mev, further approximations may be made which reduce the transport equation to a much more simple form. An elementary proof is given for the following theorem: If monoenergetic gamma-rays are incident normally on a slab of material, then after the gamma-rays have undergone many Compton scatterings, the gamma-rays of a given energy are distributed Gaussian-wise in angle about their original direction with the breadth of the distribution being simply related to the energy and increasing with decreasing energy.

I. INTRODUCTION

IN many experiments and applications involving gamma-rays one is confronted with the problem of how the energy spectrum of a beam of gamma-rays is modified in passing through a given thickness of a particular material.[1] Important examples of applications which may be quoted are the design of shields for gamma-rays, the determination of depth-dosage relations in radiology, and the design of gamma-ray filters (differential absorbers). The *elementary* processes[2] by which the spectrum is modified in a material are very well known. In the low energy region the principal process is the *photoelectric effect* in which the gamma-ray is absorbed by an atom with the ejection of an electron from the atom. For higher energies, *Compton scattering* of the gamma-rays by electrons surpasses the photoelectric effect in importance. In the Compton scattering process, the scattered gamma-ray has a lower energy than does the incident gamma-ray, with the remainder of the energy going to the electron. At still higher energies, the *production of positron-electron pairs* by gamma-rays in the Coulomb field of a nucleus surpasses the Compton effect in importance. The energy of the gamma-rays passes into the positron-electron pair. In both the Compton effect and pair production the secondary electrons resulting from the process may produce further gamma-rays by impact with nuclei (bremsstrahlung) before they are stopped by ionization energy loss.

The energy region in which each of these processes is dominant depends strongly on the atomic number of the material being traversed by the gamma-ray. In light materials (carbon, aluminum, water) the photoelectric effect becomes unimportant at relatively low energies and pair production does not become important until relatively high energies are reached, so that Compton scattering is the dominant process over a wide energy range. On the other hand, in heavy materials (lead) the energy above which the photoelectric effect

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[1] Previous studies of the diffusion of high energy gamma-rays through matter have been reported in the following papers: Hirschfelder, Magee, and Hull, Phys. Rev. 73, 852 (1948); J. O. Hirschfelder and E. N. Adams II, Phys. Rev. 73, 863 (1948); W. R. Faust and M. H. Johnson, Phys. Rev. 75, 467 (1949); U. Fano and P. R. Karr, Phys. Rev. 75, 1303 (1949); Bethe, Fano, and Karr, Phys. Rev. 76, 538 (1949); U. Fano, Phys. Rev. 76, 739 (1949); P. R. Karr and J. C. Lamkin, Phys. Rev. 76, 1843 (1949); L. V. Spencer and F. Jenkins, Phys. Rev. 76, 1885 (1949); Fano, Hurwitz, and Spencer, Phys. Rev. 77, 425 (1950).

[2] See, for example, W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, London, 1944).