Compton Backscattered Photons in Precision Proportional Counter Spectrometry*

R. W. Fink† and B. L. Robinson‡
University of Arkansas, Fayetteville, Arkansas
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The importance of Compton backscattering in gas proportional spectrometry in the energy range 10 to 100 kev is pointed out. If the backscattered photons are unresolved from the main peak, a large error may be introduced under certain conditions in absolute and relative intensity measurements based on theoretical quantum efficiency (geometry x absorption); if the backscattered peak is resolved, failure to recognize its origin may result in the "discovery" of a new gamma ray. The effect is especially pronounced when a collimator is used to establish geometry; for example, the intensity of backscattered photons can amount to as much as half the intensity of the main peak at 46.5 kev with a $\frac{1}{2}$-in. diameter collimator.

Although backscattering and wall effects in proportional counters have been carefully considered in the spectrometry of beta particles, similar effects in the spectrometry of photons seem to have been neglected except for guarding against excitation of characteristic fluorescence x-rays from the walls.

Backscattered Compton photons from outside and inside (far wall) of proportional counters can amount to as much as half of the intensity of the peak due to the direct beam at 46.5 kev with a $\frac{1}{2}$-in. diam collimator. If the peak due to Compton scattered photons is unresolved, failure to recognize its origin may result in the "discovery" of a new gamma ray. The 41-kev "gamma ray" once attributed to RaD is most likely due to this phenomenon. Even if the backscattered photons are unresolved, they contribute to the total intensity of the peak and may give rise to considerable error in absolute and relative intensity determinations based on theoretical quantum efficiency (geometry x absorption). As shown in Fig. 1 (for Cs$^{137}$ decay), the effect is especially pronounced when a collimator is used to establish geometry, and increases as the diameter of the

![Graph](image-url)

**Fig. 1.** Effect of increasing collimation on K x-ray peak in Cs$^{137}$ decay. Geometry is shown in Fig. 2(d). The intensity of the Compton backscattered photons increases relative to the main peak as the collimation becomes narrower. The intensity of the backscattered peak is of the order of 12% of the main peak for the $\frac{1}{4}$-in. diam collimator. Thus, when a collimator is used to establish geometry, an appreciable error is introduced when the theoretical detection efficiency is taken as the product of solid angle x gas absorption.

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†Department of Chemistry.
‡Present address: Department of Physics, Western Reserve University, Cleveland 6, Ohio.
Gamma Rays in the Decay of Barium-131*

WILLIAM C. BEGGS, BEROL L. ROBINSON, AND RICHARD W. FINK
University of Arkansas, Fayetteville, Arkansas
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Gamma rays accompanying electron capture of 12-day Ba$^{131}$ have been studied with a single-channel scintillation spectrometer. Relative intensities have been determined for the 122 kev (198), 214 kev (485), 372 kev (100), 496 kev plus satellite (360), and 620 kev (33) gamma-rays. Three new gamma-rays have been found at 83±20 kev (2.0), 917±15 kev (7.2), and 1632±15 kev (11.0), and their assignment to Ba$^{131}$ has been established by chemical purification and half-life. Crystal summing studies confirm that the 620-kev transition is the crossover of the 122-kev and 496-kev transitions, and no significant crystal summing is found for the three new gamma rays. A peak corresponding to 83 kev was observed, which appears to be due to two or more unresolved gamma rays, but no gamma ray could be found at 100 kev. The decay of Ba$^{131}$ was followed for 9 half-lives using an end-window beta proportional counter, and the half-life was found to be 11.5±0.08 days. The ratio of the pile neutron cross section for the activation of Ba$^{131}$ to that of long-lived Ba$^{133}$ has been found to be $\sigma(Ba^{131})/\sigma(Ba^{133}) = 1.2±0.3$.

INTRODUCTION

The complex gamma-ray spectrum associated with the electron capture of 12-day Ba$^{131}$ has been studied by a number of investigators. Table I summarizes the results of previous investigations and present findings. The gamma rays at 122, 214, 244, 372, 496, and 620 kev are well established.

Weak high-energy radiations were found in a rough scintillation spectrometer survey of a radiobarium source, which came to hand in connection with other studies. It seemed of interest to study these hard gamma rays, whose assignment had been in question.

Because of its inherent stability, our automatic recording scintillation spectrometer is well suited to the study of low-intensity radiation.

EXPERIMENTAL

Forty-four grams of reagent grade barium nitrate were irradiated in the Oak Ridge reactor for 28 days. The irradiated sample was dissolved in water and the cesium daughter of Ba$^{133}$ was separated by a chemical procedure involving the addition of sodium and cesium carriers and the precipitation of barium chloride with a concentrated (5:1) hydrochloric acid diethyl ether mixture. The precipitate was evaporated almost to dryness three times to expel NO$_3^-$ and C$^4$ as CM$\text{O}_3$.

La$^{14+}$ and Fe$^{3+}$ carriers were added to an aqueous solution of the barium chloride, and ferric hydroxide and lanthanum hydroxide were precipitated with excess ammonium hydroxide. The scavenging was repeated three times. Barium was precipitated in final form as barium chromate in acetate-buffered acetic acid by the addition of potassium chromate. The barium chromate was filtered, washed, dried, and mounted.

Isomeric states of Ba$^{133}$ (39 hr) and Ba$^{131}$ (29 hr) were allowed to decay before proceeding with the scintillation spectrometry. Pulse-height spectra from chemically purified and unpurified sources were identical.

An automatic-recording single-channel scintillation spectrometer was used for all experiments reported here. The detector consisted of a cylinder of NaI(Tl) 1 1/2 in. diameter X 1 in. high mounted on a DuMont 6292 photomultiplier tube, with a mixture of MgO and Mg(ClO$_4$)$_2$ for diffuse reflector. It was encased in a tight-fitting thin brass can and sealed with Scotch electrical tape. The detector was operated in a graded shield.

RESULTS

Typical pulse-height spectra with source distances of 7.6 cm and 1.8 cm are shown in Fig. 1. Peaks corresponding to gamma rays of 122, 214, 372, 496, and 620 kev are clearly resolved. The 244-kev gamma ray is not clearly seen, but its presence can be inferred from the skewness of the 214-kev peak. No peaks are seen at 196 or 585 kev. A peak appears at approximately 83 kev, but none was found at 100 kev. Under closer examination, the 83-kev peak appears considerably broader than the 87-kev line of Cd$^{110}$; this peak is probably due to two or more unresolved gamma rays. The 496-kev peak was consistently broader than the full energy peak in the pulse-height spectrum of the Ba$^+$ gamma ray (478 kev), indicating the existence of an unresolved satellite.
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L/K-Capture Ratios, Mean L-Fluorescence Yields, and Transition Energies in Orbital Electron-Capture

BEROL L. ROBINSON† AND RICHARD W. FINK‡

University of Arkansas, Fayetteville, Arkansas

INTRODUCTION

ADVANCES in the technique of proportional and scintillation spectrometry have extended our knowledge of orbital electron-capture. In a study of comparative half-lives (β+ values), Major and Biedenharn have summarized the data existing up to the middle of 1954.

The theoretical work of Marshak and Rose and Jackson has been extended by Brysk and Rose in the light of present knowledge of beta-decay theory of forbidden transitions, with particular reference to capture of L-shell electrons.

In the interpretation of radiative electron-capture (inner bremsstrahlung) spectra, the capture of p-electrons appears to be significant. This review comprises a summary and analysis of the existing data on electron-capturing nuclides (up to May, 1955) whose decay schemes are relatively simple and well established. In particular, the primary concern is with the ratio of L-capture to K-capture both as a test of the theory of Marshak and of Brysk and Rose and as an application of the theory to the determination of transition energies in electron-capture and of L-fluorescence yields.

EXPERIMENTAL TECHNIQUES

Essentially three techniques have been applied to the determination of x-ray intensity and capture ratios. These are described below.

(A) Internal Source Spectrometry

The radioactive material is contained within the sensitive volume of the detector. Gas proportional counters have been used for the study of A²⁷, Kr⁹¹, and Ge⁷¹ (in the form of germane, GeH₄). In this method the

prompt cascade of x-rays and Auger electrons, which follows each K-capture event, is integrated by the detector to give a single "K-line" in the pulse-height spectrum. An "L-line" arises from L-captures and from K-captures which are followed by escape of the K x-rays from the sensitive volume. The correction for K x-ray escape may be made small by suitable choice of detector material and size, and it can be calculated quite accurately from x-ray absorption data and K-fluorescence yields. Since L x-rays and L-Auger electrons are totally absorbed, no correction need be made for L-fluorescence yield.

Scintillation crystals have been grown containing radioactive I¹²⁵ and Cd¹⁰⁹. In this type of experiment the amount of L-capture is obtained from the difference between the number of K x-rays and the total number of gamma-ray transitions on the assumption that one gamma ray accompanies each decay.

(B) External Source Spectrometry

The radioactive substance is placed outside of the sensitive volume, and corrections must be applied for source self-absorption and self-scattering, for differential air and window absorption, and for K and L fluorescence yields. One must also consider that a K-shell hole may be filled by an L-electron either by radiative transition (Kα = K − L₁, L₁) or by Auger transition (K → L₁L₂). The number of L-shell vacancies produced in this manner, nKL, ranges from 1.36 at Z = 29 to 0.75 at Z = 90, as will be discussed below.

In case electron capture is followed by gamma emission, the conversion of the gamma rays must be taken into account.

Many electron-capturing nuclides of the heaviest elements have been investigated. The intensities of the L x-rays have been measured carefully by use of proportional counters and a bent-crystal x-ray spectrometer. However, most of these nuclides have low-energy gamma transitions which are highly converted in the L-shell so that the interpretation of x-ray intensities in terms of electron-capture ratios is tenuous. No attempt has been made to correlate these data with theory in this paper.

† Supported in part by the U. S. Atomic Energy Commission.
‡ Department of Physics.
§ Department of Chemistry.

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INTRODUCTION

MARSHAK, Rose and Jackson, and Brysk and Rose have computed theoretical values of the \( L_1/K \)-capture ratio as a function of atomic number and disintegration energy. From the curves of Brysk and Rose, the value of this ratio is computed to be 0.14 for \( \text{Cs}^{131} \).

Saraf has found an anomaly in the shape of the inner bremsstrahlung spectrum of \( \text{Cs}^{131} \) (which we have confirmed independently); similar anomalies have been reported in other electron capturers. Glauber and Martin have suggested that the observed shape can be explained by taking into account the capture of \( p \)-electrons. Since the relative contribution of \( p \)-electron capture, it seemed of interest to measure the ratio of the intensities of \( K \) and \( L \) x-rays following electron capture in \( \text{Cs}^{131} \).

EXPERIMENTAL METHOD

A source was prepared by chemical separation of cesium from 44 grams of barium nitrate irradiated for 28 days in the Oak Ridge reactor. After precipitation with \( 5:1 \) HCl-diethyl ether mixture, the supernatant cesium fraction was scavenged extensively with barium chloride precipitations, followed by lanthanum hydroxide, ferric hydroxide, and silver chloride scavengings. Separation from other alkali metals was accomplished by precipitation as the silicotungstate. Scintillation spectrometer studies of the final cesium fraction detected no trace of the intense gamma rays of \( \text{Ba}^{133} \), indicating a decontamination factor from barium of better than 10^4.

Traces of 6.2-day \( \text{Cs}^{133} \) and 13.6-day \( \text{Cs}^{137} \) were found in the first cesium fraction from pile-irradiated barium nitrate, presumably by \((n,\alpha)\) reactions.

The \( L \) and \( K \) x-ray intensities were measured using an x-ray proportional counter filled with 2.1 atmos argon—0.2 atmos methane mixture. The counter was constructed of brass with an aluminum liner to eliminate fluorescent radiation from the brass. The side window was a 164 mg/cm^2 disk of Brush beryllium. The x-ray path length in the counter was about 10 cm. A potential of 3,850 volts was applied to the 0.004-inch diameter stainless steel wire, and the amplified output was fed through a single-channel pulse analyzer into a recording ratemeter or scaler. A typical spectrum is shown in Fig. 1. The observed ratio of x-ray intensities, \( (N_L/N_K) \), was obtained by graphical integration using a planimeter. This ratio is 0.334 ± 0.01, representing an average of many determinations. The same value was also determined by the method of integral-bias counting, giving an independent check on the result. The intensity ratio may be expressed by Eq. (1)

\[
\frac{N_L}{N_K} = \frac{\left[L_1 + L_{11,111} + nK\right]}{K} \times \left(\frac{\omega_{L1}}{\omega_{K}}\right) \left(\frac{T_L}{T_K}\right) \left(\frac{T_L}{T_K}\right)\left(\frac{\omega_{L1}}{\omega_{K}}\right) \left(\frac{S_L}{S_K}\right),
\]

where \( K \) is the probability that a disintegration will...
Letters to the editors

nitrate according to the procedure of Werner.\(^{(a)}\) Anal. Caled.: Cl 13.95, N 13.78. Found: Cl 13.77, N 14.08. We have also made the \(\beta\) - and \(\gamma\) -picoline analogues.

These three compounds were found by titration to release ionic chloride in a first-order process and with identical rates at \(pH\) 1-2 and 9-18 (borne buffer). For comparison, trans-dichlorobis-(ethylenediamine) cobalt(III) chloride forms ionic chloride 1400 times faster at pH 9-18 than at pH 1-2. Similar large ratios have been found for other complexes containing replaceable protons.\(^{(a)}\)

The first-order rate constants found above were

\[
\begin{array}{ll}
\text{HL} & \text{(sec)} \\
\text{pH 1-2} & \text{pH 9-18} \\
[\text{Hal,Pic},\text{Cl}]_4^+ & 4.9 \times 10^{-4} & 5.0 \times 10^{-4} \\
[\text{Co}(\text{Pic})_2\text{Cl}]_4^+ & 15 \times 10^{-4} & 15 \times 10^{-4} \\
[\text{Co}(\text{Pic})_3\text{Cl}]_4^+ & 1.8 \times 10^{-3} & 9.1 \times 10^{-4} \\
\end{array}
\]

\(^{(a)}\) \(pH\) 8.5 because of decomposition at 9-18.

Higher \(pH\)'s could not be studied because of the complete decomposition of the complex ions.

Trans-dichlorobis-(dipyridyl) cobalt(III) chloride was prepared by the method of Jacob and Van Dijk.\(^{(a)}\) This compound equilibrated instantaneously, as does the cis isomer, and was not suitable for kinetic studies. It was converted into the dinitro/bis(dipyridyl) cobalt(III) chloride by treating with two equivalents of sodium nitrite in hot water. On cooling, a yellow salt separated which was recrystallized from absolute alcohol and dried at 115\(^{\circ}\) for twenty-two hours. Anal. Caled.: Cl 6.92. Found: 6.77, as ionic chloride.

The first nitro group in this complex was very labile, coming off in acid and in alkaline solution in a few minutes. The acid solution was not convenient for kinetic study because of the further reactions of nitrous acid. The rate of reaction in the presence of hydroxide ion was studied conductometrically at 25\(^{\circ}\). In four runs with complex ion and hydroxide ion equimolar between 1 \(\times 10^{-4}\) M and 1 \(\times 10^{-8}\) M, the nitrite ion was released in a first-order process with identical rate constants of 9.1 \(\times 10^{-2}\) min\(^{-1}\). Hence hydroxide ion had no effect on the rate.

The three pyridine complexes\(^{(a)}\) were also found to show a normal enhancement of rates of chloride ion release in the presence of Hg\(^{++}\). The second-order rate constant for the tetrakis(pyridine) complex was 11 M\(^{-2}\) min\(^{-1}\) at a concentration of 0.001 M in complex ion and mercuric nitrite.

These results are believed to provide strong evidence that the normal mechanism for the rapid basic hydrolysis of cobalt(III) complexes involves prior formation of the conjugate base of the complex ion.

Chemistry Department,  
Northwestern University,  
Evanston, Illinois

R. G. Pearson  
R. E. Meiker  
F. Basolo

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The Half-life of Emanation-220\(^{22}\)

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Currently the accepted value for the half-life of thoron (\(\text{Em}^{220}\)), 54.5 seconds, was chosen\(^{(a)}\) by the International Radium Standards Commission in 1930, based on measurements of Perkins and Schmid.\(^{(a)}\)

Previous measurements have been made, using a leaf electroscope method involving the insertion of...
(s axis) in the paramagnetic salt used to cool the nuclei. Separate observations were made on the angular distributions of the 530-kev and 92-kev gamma radiations. For the 530-kev gamma ray, an anisotropy $s = -0.29$ was observed at the lowest temperature reached (9903°C). Assuming spin $\frac{9}{2}$ for $\text{Nd}^{147}$ we conclude that the gamma transition is $E2$, and that the spins involved in the decay chain are $9/2^- \rightarrow 7/2^- \rightarrow 5/2^-$. No significant anisotropy was observed for the 92-kev gamma ray and it is concluded that this transition is a mixture of $M1$ and $E2$, and that the spins involved in the decay chain are $9/2^- \rightarrow 7/2^- \rightarrow 5/2^-$. From the temperature dependence of the anisotropy we deduce a value $0.22 \pm 0.05 \text{nm}$ for the magnetic moment of $\text{Nd}^{147}$. This rather small value (together with our value of $0.16 \pm 0.06 \text{nm}$ for $\text{Cs}^{133}$) may not be meaningful because of possible internal magnetic field effects. This, however, does not affect our other conclusions.

**ROBINSON AND RICHARD W. FINK,**

Decay schemes based on the experimental results will be observed with the following energies and intensities: $1.83 \text{MeV}$ (0.02). The unconverted portion of a strongly converted $\text{K}$-electron capture are not adequate in interpreting the theoretical predictions, which take account only of the electromagnetic radiations of $\text{Cs}^{133}$ and $\text{Cs}^{137}$, and were found to be $0.930 \text{MeV}$.

No significant anisotropy was observed for the 92-kev gamma ray, and it is concluded that this transition is a mixture of $M1$ and $E2$, and that the spins involved in the decay chain are $9/2^- \rightarrow 7/2^- \rightarrow 5/2^-$. From the temperature dependence of the anisotropy we deduce a value $0.22 \pm 0.05 \text{nm}$ for the magnetic moment of $\text{Nd}^{147}$. This rather small value (together with our value of $0.16 \pm 0.06 \text{nm}$ for $\text{Cs}^{133}$) may not be meaningful because of possible internal magnetic field effects. This, however, does not affect our other conclusions.
Observations on the Smoke Trail of a Sky-Writer

WHILE driving on the highway recently my attention was drawn to a sky-writer whose trail fell between me and the sun and less, perhaps, than 30 degrees above the horizon. On first observation I had the feeling that the sunlight reaching me through the smoke trail was reddish-orange in color, and a moment's study confirmed the notion that the smoke possessed a predominantly reddish-grey hue. Within the next several seconds my own course was then brought in electrical contact. A discussion of the potential and charge-distribution changes which might be called to the attention of students. It is obvious that the observer need not change his position; all one needs is that the sky-writer should pass between him and the sun.

I am led to suggest also that some interesting observations on wind direction and velocity may be made on such smoke trails.

A Problem in Electrostatics

MANY college textbooks which cover the fundamentals of electrostatics contain a discussion of the following experiment. A large hollow conducting sphere with a small opening in its side has a charge q. A small conducting sphere with a charge q' is brought from infinity and inserted through the opening into the larger sphere. The two spheres are then brought in electrical contact. A discussion of the potential and charge-distribution changes which result seems to prove very enlightening to students and paves the way for a discussion of the principle of the Van de Graaff generator.

A few years ago a student asked a question which seemed to involve a paradox, and which caused so much class discussion and aroused so much interest, that the author has used it in all subsequent classes on the subject. Suppose the now uncharged small sphere is removed from the larger sphere and carried to infinity. The total energy of the system is the same as when the uncharged body was inside the larger sphere. Yet, because of the charge induced on the small sphere when it is outside of, but near to, the larger sphere, energy is required to withdraw it from this position to infinity. How can this be reconciled with the law of conservation of energy?

Concerning the Frequencies Resulting from Distortion

THIS note concerns the question raised recently by Maxwell concerning the physical origin of the harmonic frequencies which appear in the analysis of a rectifier circuit.

Van Name has suggested that these frequencies are not physically present at all. If this were the case, it would not be possible to operate an harmonic analyzer, which is essentially a tuneable highly selective amplifier and voltmeter. As a matter of fact, electronics-laboratory courses frequently include an experiment in which the amplitudes of the harmonics of a nonsinusoidal wave are measured and compared with those values calculated by a Fourier analysis.

Thus experiment shows that these frequencies are physically real and present in the system. They have been generated by the nonlinear response characteristic of the rectifier itself. In general, if one is given the input wave form and the characteristic of a nonlinear device, one may calculate the amplitudes and phases of the harmonics which will be generated. The simplest case is probably that of the amplifier with a quadratic plate-current characteristic and a sinusoidal input signal; that is, 

\[ v(t) = V_0 \sin \omega t \]

and

\[ e(t) = E \sin \omega t \]

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