$K/(L+M)$ Internal Conversion Ratios for $M4$ Transitions*

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(Received June 27, 1952)

A high resolution magnetic spectrometer has been used to study the internal conversion lines of magnetic four-pole transitions. The $K/(L+M)$ internal conversion ratios fall lower than those predicted from the empirical curve of Goldhaber and Sunyar in the region of low values of Z^2/E. A revised curve is presented. The following values are obtained for the energy of the transition and for the $K/(L+M)$ ratio: Y^{91}, 551.2 kev, 6.00; Sr^{87}, 388.2 kev, 5.79; Y^{87}, 381.3 kev, 5.41; Ba^{137}, 661.4 kev, 4.64; In^{113}, 391.7 kev, 4.21; and In^{115}, 334.6 kev, 3.76. Values are also given for lines in Cd^{111} and In^{114}.

INTRODUCTION

THE classification of an isomerism is usually accomplished either in terms of the half-life and the energy separating the states, by measuring the internal conversion in the K-shell, or by evaluating K/L internal conversion ratios.

In employing the first of these methods, it is usually not difficult to measure the transition energy quite well. However, the half-life which one uses for comparison with the theory must be the half-life for the radiation alone and not the total half-life of the state. This means that, if there are branching transitions, a detailed knowledge of these branches is required. In addition, it often happens that the half-life for the transition is one which is difficult to measure—e.g., it may be very short. When one employs the second method, one again needs some knowledge of the decay scheme, if one determines the gamma-ray intensity indirectly by finding the intensity of the beta-rays which feed the level. However, if one obtains the internal conversion and the gamma-ray intensity directly, or if one measures the K/L internal conversion ratio, an identification of the isomerism can be made without a complete knowledge of the decay scheme.

In a recent paper,[1] Goldhaber and Sunyar have attempted to formulate a scheme for the classification of nuclear isomers by identification of the type of radiative transition between the states. These authors employed a semi-empirical approach, relying chiefly on experimental results and on well-founded theoretical calculations such as the relativistic internal conversion coefficients of Rose *et al.*[2] One result of their investigations was the publication of curves of empirical K/L internal conversion ratios for various known transition types.[3] These curves can aid in the identification of other unknown transitions for which this ratio can be measured. In addition, they can be of assistance in determining the total conversion coefficient for a given type of transition. This quantity is valuable in analyzing complex decay schemes because it is needed for a comparison of intensity between the various beta- and gamma-decay branches.

The object of this paper is to present the results of a group of experiments which have indicated that a portion of the published empirical curve for $M4$ transitions should be substantially modified. A new curve for such transitions is proposed. The modifications are based largely upon $K/(L+M)$ ratios measured in the isomeric transitions In^{113}, In^{115}, Sr^{87}, Y^{87}, Y^{91}, and Ba^{137}, and to some extent on new results in the literature.

EXPERIMENTAL PROCEDURES

All measurements were made upon chemically separated, relatively thin and uniform, electrically grounded sources in a high resolution, magnetic spectrometer of 40-cm radius of curvature.[4] The momentum spread of

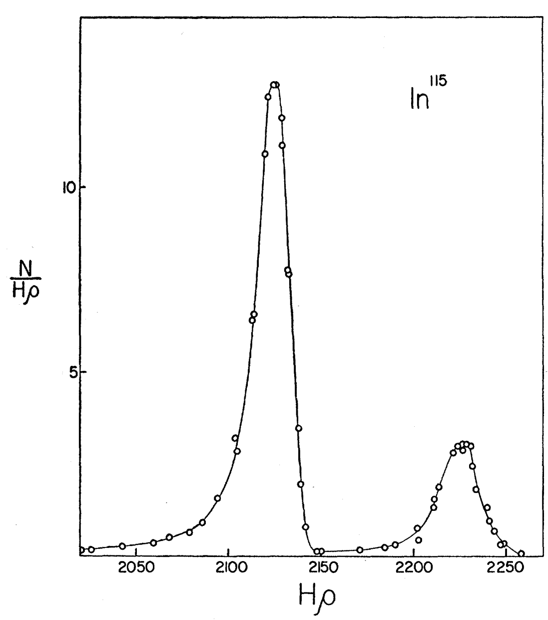

FIG. 1. Conversion electron spectrum of In^{115}.

* Assisted in part by the joint program of the ONR and AEC, and by a grant from the Frederick Gardner Cottrell Fund of the Research Corporation.

[1] M. Goldhaber and A. W. Sunyar, Phys. Rev. 83, 906 (1951).

[2] Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report No. 1023 (unpublished); also Phys. Rev. 83, 79 (1951).

[3] Actually, these curves are a mixture of K/L and $K/(L+M)$ ratios, since conversions in the M, N, etc., shells were neglected where no separate measurements of them were available. Only for very high Z and very low energies are the L and M lines resolved, with present day techniques.

[4] L. M. Langer and C. S. Cook, Rev. Sci. Instr. 19, 257 (1948).

the lines at half-maximum intensity ranged from 0.5 percent to 1.1 percent, depending mostly on the thickness of the source. In all cases, after the background level appropriate to any beta-spectrum present had been subtracted, the data were corrected for decay and exhibited on plots of $N/H\rho$ *vs* $H\rho$. The ratio of the areas of the internal conversion lines yields the $K/(L+M)$ ratio.[5] The ratios quoted below are believed to be correct to within 5 percent, with the possible exception of that for In^{113}, where an arbitrariness in the subtraction of a complex beta-background might introduce as much as 5 percent additional error.

The particular experimental procedure used with the various isotopes is described in the separate discussions

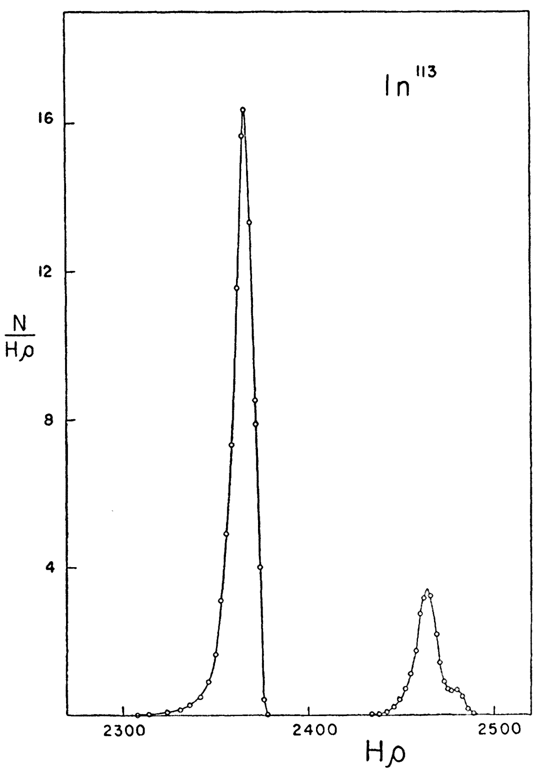

FIG. 2. Conversion electrons of 391.7-kev transition in In^{113}.

below, and these individual results are collected in a summary curve.

RESULTS

In^{115}

The study of this isotope provided one of the first indications that the $M4$ curve of Goldhaber and Sunyar might require a change.[6]

The source material was grown and extracted as

[5] The practice of evaluating conversion ratios from the ratios of the peaks in a plot of N *vs* $H\rho$ is only valid if the geometrical resolution is poor compared to the natural line width. If the sources are not thin or if the lines are not completely separated, such a procedure may lead to erroneous ratios.

[6] Langer, Moffat, and Graves, Phys. Rev. **86**, 632 (1952).

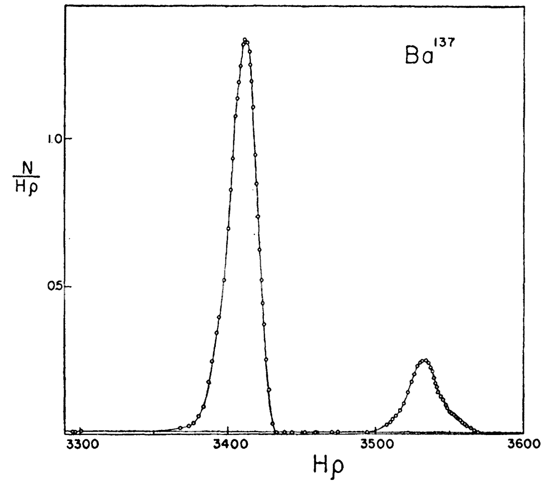

FIG. 3. Momentum distribution of conversion electrons of Ba^{137}.

$In(OH)_3$ from the 54-hour Cd^{115} parent, which had been produced by deuteron bombardment on cadmium in the cyclotron. It was deposited as a slurry on a 0.0002-in. aluminum backing. The internal conversion lines of interest are shown in Fig. 1. The energy of this 4.5-hour transition is 334.6 kev,[7] for which the corresponding value of Z^2/E is 7.12. The $K/(L+M)$ ratio is 3.76.[8] This transition presumably occurs between an excited $p_{1/2}$ state and the $g_{9/2}$ ground state, whose spin has been measured. This assignment is in agreement with the level predictions of the nuclear shell model.

In^{113}

In^{113} was prepared by a short deuteron bombardment on cadmium in the cyclotron. The source was deposited as an $In(OH)_3$ slurry upon a 0.0002-in. Al backing. The internal conversion lines from this source were superimposed on a weak beta-ray background whose decay indicated that it might arise from a mixture of 54-min In^{116} and 117-min In^{117}. From an empirical plot of this background against time, the background level existing at the time of measurement of each of the points was determined. This level was subtracted from the measured counting rate, and the residual rates were corrected for a decay of half-life 104 minutes.

The internal conversion lines are shown in Fig. 2. For this transition, the measured energy is 391.7 kev, yielding Z^2/E as 6.13, and the $K/(L+M)$ ratio is 4.21. Shell model predictions ascribe $p_{1/2}$ to the upper state and $g_{9/2}$ to the lower state of this isomeric pair.

Ba^{137}

The $M4$ transition in Ba^{137} follows a beta-decay from Cs^{137}. The source of Cs^{137} was separated from the fission products at Oak Ridge. Since the period of the isomeric

[7] Because of a typographical error, this energy was erroneously given as 333.7 kev in reference 6.

[8] This value is somewhat lower than that previously given by J. L. Lawson and J. M. Cork, Phys. Rev. **57**, 982 (1940).

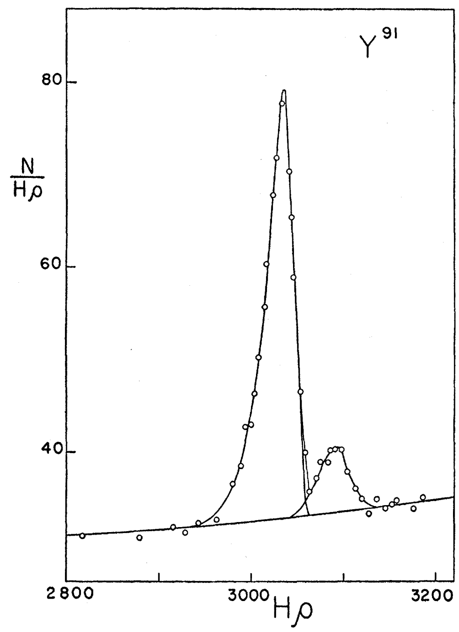

FIG. 4. Electron line spectrum in Y^{91}.

transition is very short (2.62 minutes) compared to that of the beta-decay which feeds it, its internal conversion lines are measured in equilibrium with the 37-year parent activity and no decay correction need be made. The momentum plot shown in Fig. 3 yields a $K/(L+M)$ ratio of 4.64. The gamma-ray energy is 661.4 kev, for which Z^2/E is 4.74. The decay is presumably from an excited state of $h_{11/2}$ to a $d_{3/2}$ ground state whose spin has been measured.

Y^{91}

The internal conversion lines in Y^{91} arise from a 51-minute metastable state which is fed by the 9.7-hour beta-decay of Sr^{91}. The Sr^{91} was separated from the fission products at Los Alamos, following a short neutron bombardment of fissionable material in a pile. The internal conversion electron spectrum, corrected for the 9.7-hour equilibrium decay, is shown in Fig. 4. The energy of the unconverted gamma-ray is 551.2 kev, Z^2/E is 2.76, and $K/(L+M)$ is 6.00. For this transition, the shell model predicts $g_{9/2}$ for the upper level and $p_{1/2}$ for the lower level.

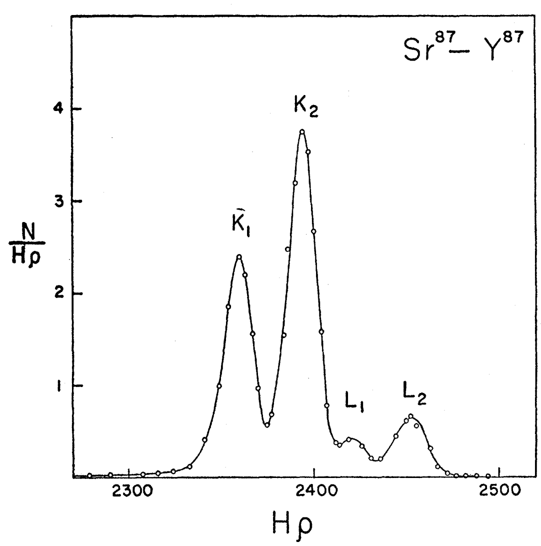

FIG. 5. Momentum plot of conversion electrons in $Sr^{87}-Y^{87}$. This curve is not decay-corrected and is introduced just to show the relative separation of these lines.

$Sr^{87}-Y^{87}$

The activity in these isotopes has a rather extensive history, to which references can be found in two recent articles.[9,10] Convincing evidence has been presented in these papers that the decay scheme, at least in part, is as follows: Y^{87} is formed in two isomeric states. The upper state undergoes a 14-hour transition of about 385 kev to the ground level. This lower state in turn decays with a period of 80 hours, mainly by K-capture to an excited state in Sr^{87} at about 875 kev above its ground level. This state then undergoes a 485-kev transition to a 2.8-hr isomeric level which is about 390 kev above the ground state. No crossovers have been definitely established in this chain, although some positrons as well as some internal conversion lines in the region of 1 Mev have been reported and sometimes attributed to this decay.

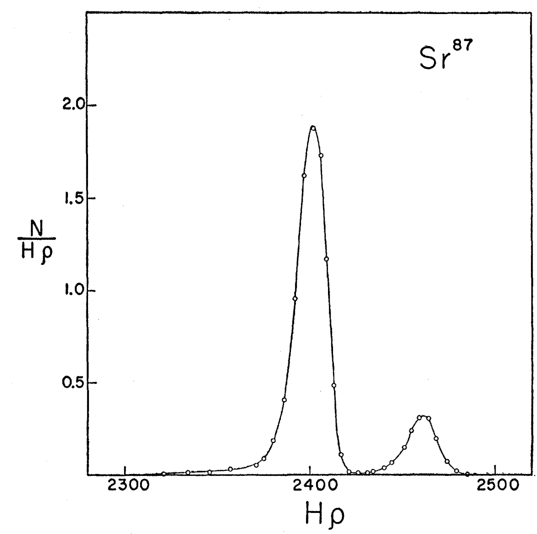

FIG. 6. Internal conversion electrons of the 388.2-kev transition in Sr^{87}.

Both the 14-hr and the 2.8-hr isomeric states appear to give rise to $M4$ transitions. $K/(L+M)$ ratios reported for these conversions have ranged from 6.0 to 8.3. About 0.4 percent of the 485-kev transitions occur by internal conversions, and no $K/(L+M)$ values have been previously reported for these lines.

9 E. K. Hyde and G. D. O'Kelly, Phys. Rev. 82, 944 (1951).
10 L. G. Mann and P. Axel, Phys. Rev. 84, 221 (1951).

Because of the small energy separation of the two $M4$ transitions, in the past there has been difficulty in resolving the conversion lines, which has complicated measurements on the $K/(L+M)$ ratios. The present experiment was performed with better resolution, and it was also arranged to measure the internal conversion lines of each transition with sources in which the other transition was essentially absent. This made possible a more accurate determination of the energies and of the $K/(L+M)$ ratios. Here the K and L lines are completely separated and no arbitrariness of extrapolation is necessary.

Radioactive yttrium was produced by bombarding strontium with deuterons in the cyclotron. The yttrium was separated as a hydroxide and deposited as a slurry onto a 0.0002-in. Al source backing. An examination of this source in the spectrometer about eighteen hours after the separation gave the results shown in Fig. 5. In the course of the different studies on this source, it was obvious that the lines K_1 and L_1 decayed with a 14-hr half-life, while K_2 and L_2 first grew with this half-life and later decayed with the 80-hr period. This lends support to the genetic relationships previously proposed[10] and summarized above.

One hundred and twenty-two hours after preparation of this source, its spectrum was again studied. By this time the 14-hr activity was completely gone, and the internal conversion lines of the 2.8-hour transition were easily measured in equilibrium with the 80-hr state into which the 14-hr activity had fed. These lines, which are in Sr^{87}, are corrected for the 80-hr decay and shown in Fig. 6. Our measurements ascribe 388.2 kev to the energy of the transitions, 3.72 to Z^2/E, and 5.79 to the $K/(L+M)$ ratio.

Further evidence for the correctness of the previously proposed decay scheme was obtained by separating strontium from yttrium left over in preparing the above source. This second separation was performed 90 hr after the bombardment. One portion of this Sr was used as a spectrometer source; the other was monitored by a device which automatically recorded its decay. From this, the half-life of this material was found to be 173 ± 2 minutes. Two traverses of the K-line in the spectrometer fitted to within 2 percent at their peaks when both were corrected for this decay to the same initial time. The $K/(L+M)$ ratio obtained from this separated strontium source differed from that quoted above by only 1 percent.

To investigate the conversion of the $M4$ transition in Y^{87}, strontium was bombarded with eighty microamperes of deuterons for one hour. Yttrium was separated from the strontium, a spectrometer source prepared, and the scanning of the spectrum commenced within about one hour after the bombardment. The actual counting rates are shown in Fig. 7. It is obvious from this figure that there is only a very small contribution to the counting rate from internal conversions in strontium (K_2 and L_2).

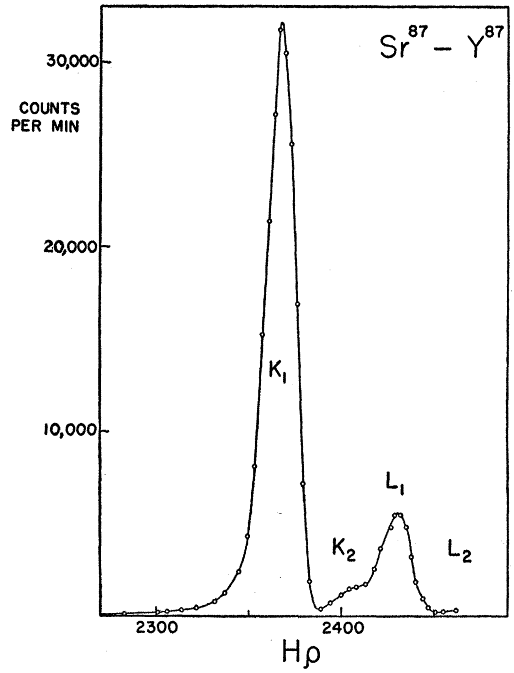

FIG. 7. Electron momentum distribution obtained shortly after separation from a source containing only yttrium at the initial time. These points have been corrected for the counter losses at high counting rates. No decay correction has been applied.

From our previous measurements on strontium it was easily determined where its spectrum had a negligible influence on these points. For these points, a 14-hr decay correction was applied, and the plot of Fig. 8 was made. The proper low energy extrapolation of the

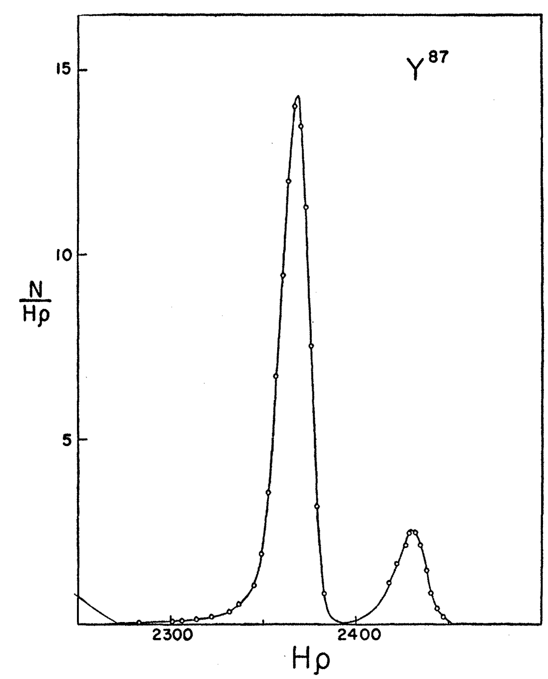

FIG. 8. Conversion elections from the 14-hr transition in Y^{87}.

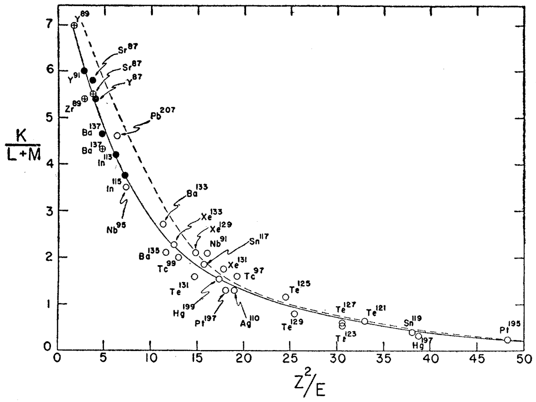

FIG. 9. $K/(L+M)$ internal conversion ratios for $M4$ transitions.

L-line beyond these points was determined from the shape of the L-line in the strontium data. The energy of this transition was measured to be 381.3 kev, Z^2/E was 3.99, and $K/(L+M)$ was 5.41.

The higher energy conversion lines in strontium were also studied during this investigation. It was confirmed that they decay with 80-hr half-life and their $K-L$ energy difference shows that they are in strontium. The transition energy is 483.4 kev, Z^2/E is 2.99, and the $K/(L+M)$ ratio was measured to be 6.64. This last quantity might be in error by as much as 25 percent, since the counting rate on these lines is low. Mann and Axel[10] measured the internal conversion coefficient for this transition, from which they concluded it was either $M1$ or $E2$, and probably $M1$. Our value of the $K/(L+M)$ ratio seems to substantiate this assignment when compared with Goldhaber and Sunyar's curves,[1] although the results are not conclusive.

The spin and level assignments previously given in Sr^{87} would seem to be correct. The ground state is $g_{9/2}$, the first excited state at 388.2 kev would then be $p_{1/2}$, and the state at 871.6 kev could be $p_{3/2}$. For Y^{87}, the shell model would assign $p_{1/2}$ to the 80-hr level and $g_{9/2}$ to the level at 381.3 kev. These assignments lead to difficulties in that one then expects allowed positron emission from the upper state of Y^{87} to the ground state of Sr^{87}, and very few, if any, positrons are observed corresponding to this branch. This discrepancy would appear to indicate either an error in the predictions of the shell model, or the operation of some additional selection rule which forbids the transition.

A search was made for conversion lines previously reported[9] in the region above 1 Mev in the spectrum of freshly prepared Y^{87}, but these lines were not found.

Cd^{111} and In^{114}

These are not $M4$ transitions, but in the course of these experiments measurements were made on some of the internal conversion electrons of these isotopes, and the results are presented here.

The source was the one from which the In^{113} data had previously been obtained. Three sets of K, L, and M lines were measured. Two of these sets represented conversions following the 68-hr beta-decay of In^{111} to Cd^{111}, and one set resulted from a 50 day $E4$ transition between isomeric states of In^{114}. The $K-L$ energy differences support the assignment of these lines to these elements.

For the two lines arising between levels in Cd^{111}, the gamma-ray energies are 170.8 kev and 245.6 kev,[11] with $K/(L+M)$ ratios of 7.03 and 4.79, respectively. The distribution among K:L:M conversions for these transitions have been estimated as 875:105:20 and 827:146:24, respectively. For In^{114}, the transition energy is 189.8 kev,[11] with a $K/(L+M)$ ratio of 1.00, and with K:L:M equal to 500:425:75. There is the possibility of considerable error in our estimate of the relative strength of the L and M conversions, since the lines are multiple and are not completely resolved, and this makes it difficult to determine the true contribution of each. The estimates given above seem reasonable from the data.

CONCLUSIONS

The results for the $M4$ transitions presented in this paper are shown along with results of other investigations in Fig. 9. In this figure, Z is the nuclear charge of the isomeric pair; E is the transition energy in kilovolts. The dashed curve is the original empirical curve of Goldhaber and Sunyar;[1] the solid curve is the one proposed on the basis of the new data. The solid circles represent data presented in this paper. The barred circles represent results reported by Bendel *et al.*[12] The open circles represent estimations of the best values for $K/(L+M)$, as determined by Bendel from re-evaluation of the literature.[13]

Although this new curve of $K/(L+M)$ ratios should be useful for the identification of $M4$ isomers, a curve of true K/L ratios would be of more value for a direct comparison with theory when the new theoretical results for L conversion become available.

The authors take this opportunity to express their appreciation to Dr. Milo B. Sampson and the cyclotron staff for bombardments and to Mr. Arthur Lessor for performing many of the chemical separations. We are indebted to Dr. D. P. Ames for the chemical separation of the Y^{91} sample.

[11] These energies are about two kev lower than reported by Lawson and Cork (see reference 8). The energy of the transition in In^{114} agrees well with the 190-kev energy reported by Mei, Mitchell, and Zaffarano, Phys. Rev. **76**, 1883 (1949).

[12] Bendel, Shore, Brown, and Becker, Phys. Rev. **87**, 195 (1952).

[13] We are grateful to Mr. Bendel for very kindly supplying us with his results.