Inner Beta Spectra of Ag^{111} and Rb^{86}†

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The relatively high comparative half-lives of the low-energy, inner beta-ray group of Ag^{111} and the inner group of Rb^{86} suggest that their spectra may have nonstatistical shapes. The spectra were investigated with a 4π beta-ray scintillation spectrometer which had been adapted for beta-gamma coincidence studies. Both spectra are found to exhibit nonstatistical shapes.

INTRODUCTION

THE low-energy, inner beta-ray group in Ag^{111} and the inner beta-ray group in Rb^{86}, which are classified as once-forbidden, nonunique transitions, each have a log*ft* value of 7.9. This value is relatively high for transitions in this class of forbiddenness.[1] This suggests that there may be destructive interference occurring between the radiations generated by the nuclear matrix elements. Because of the usually dominant energy-independent terms in the once-forbidden, nonunique shape factor, most transitions in this class have been found to exhibit spectral distributions which are not measurably different from a statistical shape. If these energy-independent terms in the shape factor are sufficiently suppressed by the destructive interference, a nonstatistical shape may result. The low-energy, inner group of Ag^{111} and the inner group of Rb^{86} have been investigated to determine if they have nonstatistical shapes.

Although present interest lies mainly in the study of the above mentioned groups, it is also of interest in the case of Ag^{111} to examine the other beta-ray groups. Such a study provides a check of the decay scheme proposed for this isotope.

EXPERIMENTAL

The beta spectra were investigated with a 4π beta-ray scintillation spectrometer which had been adapted for beta-gamma coincidence studies. The spectrometer has been described in a previous paper.[2] The 4π-steradian geometry was obtained by sandwiching the source of activity between two cylindrical plastic phosphors which were each optically coupled to a 6292 Dumont phototube. The resolution of the instrument for the 624-kev internal-conversion electron line in Cs^{137} was 13 to 14%. The linearity of the relation between the observed pulse height and the beta-ray energy was checked in the energy region between 40 and 976 kev by the measurement of six internal-conversion electron lines. The capability of the instrument to measure both single and coincidence beta spectra was verified in the energy range from 36 kev to 1 Mev through the study of six known spectra.

The source preparation was essentially the same for the silver and the rubidium isotopes. Each source was mounted on a 10-μg/cm^2 Zapon film. Insulin was used to aid in the uniform spreading of the source material[3] over an area of $\sim$0.5 cm^2. A 7-μg/cm^2 Zapon film was placed over the source. The mounted source was centered on the face of one phosphor. The two phosphors were then brought into contact.

Ag^{111}

The source material was obtained from the Oak Ridge National Laboratory where it was produced by slow-neutron irradiation of Pd^{110} and subsequent 22-minute decay of Pd^{111} to Ag^{111}. The silver was separated from the palladium by means of an anion exchange procedure.[4]

The decay scheme as proposed in earlier investigations[5] is shown in Fig. 1. The energies, intensities, and

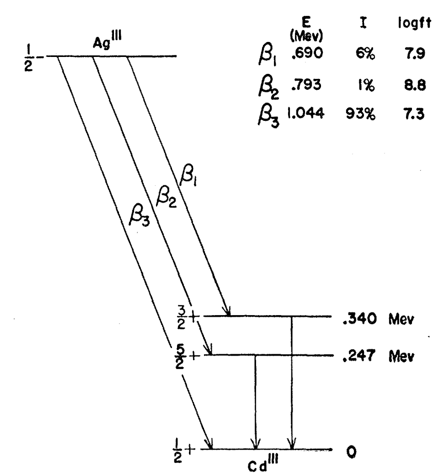

FIG. 1. Decay scheme of Ag^{111}. The energies, intensities, and log*ft* values are from the present study.

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[1] The log*ft* values of most once-forbidden, nonunique transitions with spin change of 0 and 1 are centered around 6.5 and 7.5, respectively. R. W. King and D. C. Peaslee, Phys. Rev. **94**, 1284 (1954).
[2] R. L. Robinson and L. M. Langer, Phys. Rev. **109**, 1255 (1958).
[3] L. M. Langer, Rev. Sci. Instr. **20**, 216 (1949).
[4] Schindewolf, Winchester, and Coryell, Phys. Rev. **105**, 1763 (1957).
[5] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

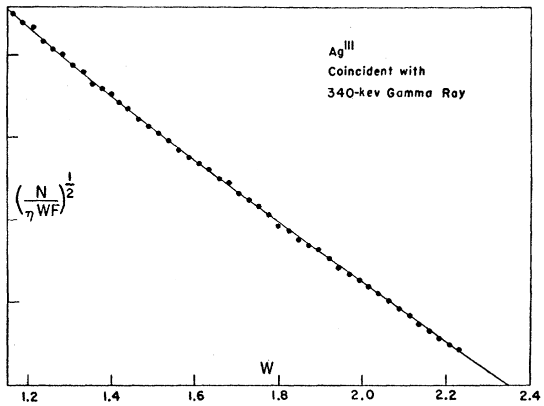

FIG. 2. Fermi-Kurie plot of the low-energy, inner beta spectrum of Ag^{111} measured in coincidence with the 340-kev gamma ray.

comparative half-lives which are given in the figure are those found in the present study.

The gamma spectrum of the silver isotope was studied with a NaI scintillation spectrometer. Photopeaks were observed at 247±5 and 340±7 kev. From the number of counts in the photopeaks, the gamma-ray counting rates were obtained by correcting for the efficiency of the NaI crystal,[6] for the photofractions,[7] for the absorption in $\frac{1}{8}$-inch Lucite (used to prevent beta rays from reaching the NaI crystal) and in the canister in which the crystal is packaged, and for the solid angle at the source subtended by the NaI crystal. The gamma-ray intensities were obtained from the comparison of the corrected gamma-ray counting rates with the total beta-ray counting rate which was measured for the same source with the 4π scintillation spectrometer. The intensities are 1.0±0.5% and 6.0±1.5% of the total decays for the 247− and 340-kev gamma rays, respectively. Since the internal conversion coefficients are small,[8,9] these intensities are approximately the intensities of the two inner beta-ray groups.

To observe the low-energy, inner beta-ray group separately, the beta rays in coincidence with the 340-kev gamma-ray photopeak were recorded. Three runs were made over a period of eleven days (approximately one and a half half-lives of Ag^{111}). The masses of the three sources differed by a factor of approximately seven. For two runs, the resolving time of the coincidence circuit was 0.9 μsec; for the third run the resolving time was 0.4 μsec. Chance counts and background counts were subtracted. Corrections for decay and for finite resolution were applied.[10] The corrections other than the decay correction were small. For example, in the third run the real to chance ratio was greater than 400, the resolution correction was less than 1%, and the background correction was less than 0.5% for more than half of the points. The maximum decay correction for this run was 9.7%. Also for more than half of the points, the mean statistical error was less than 3%.

No difference was observed in the spectrum shape obtained in the three runs. Fermi-Kurie plots of the data exhibit nonlinear shapes. The end-point energies, as determined by visual fitting of the best curve to each of the three Fermi-Kurie plots, are 687, 693, and 690 kev. Our best estimate of the end point is 690±13 kev.

To minimize the mean statistical error, the experimental data of the three runs were combined. The data were normalized in the energy range of 87 to 613 kev. The Fermi-Kurie plot of the combined data is shown in Fig. 2.

A sensitive method of distinguishing spectrum shapes is to plot the experimental shape factor C against the relativistic energy W of the electron. C is given by the quantity $N(W)/\eta WF(W_0-W)^2$, where η is the relativistic momentum of the electron, F is the Coulomb function, and W_0 is the maximum energy of the beta spectrum. For a spectrum with a statistical shape, such a plot is a straight line parallel to the energy axis. A plot of the experimental shape factor of the transition in Ag^{111} (Fig. 3) indicates clearly its nonstatistical shape. The shape factor decreases 17% between 80 kev and 600 kev. The error flags in the figure represent the mean statistical errors.

The second inner beta-ray group in Ag^{111} was observed by recording the beta rays in coincidence with gamma rays whose energies fell between 220 and 280 kev. The average end-point energy of three such coincidence runs is 793±15 kev. Unfortunately, the gamma-ray energy interval between 220 and 280 kev included not only the photopeak of the 247-kev gamma ray, but also the low-energy side of the 340-kev gamma-ray photopeak and Compton electrons produced by scattered 340-kev gamma rays. Thus, some beta rays from the 690-kev group were also recorded. In some instances, the energy of a Compton electron produced by a 340-kev gamma ray which was scattered in a plastic

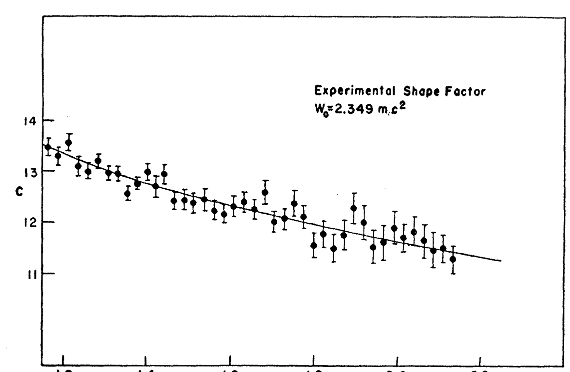

FIG. 3. Experimental shape factor of the low-energy, inner beta spectrum of Ag^{111}.

[6] P. R. Bell, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. 5.
[7] M. J. Berger and J. Doggett, Rev. Sci. Instr. **27**, 269 (1956).
[8] S. Johansson, Phys. Rev. **79**, 896 (1950).
[9] C. L. McGinnis, Phys. Rev. **81**, 734 (1951).
[10] J. P. Palmer and L. J. Laslett, U. S. Atomic Energy Commission, Bulletin ISC-174, 1950 (unpublished).

phosphor as well as in the NaI crystal would be added to the energy of a beta ray from the 690-kev transition. The resultant spectrum is therefore distorted. Because of this distortion, it is not possible to determine whether or not the spectrum has a once-forbidden, unique shape, as the spin and parity assignments indicate.

The total spectrum of Ag^{111} was also observed. The maximum energy is 1044±20 kev. To check the decay scheme, the end-point energy of each inner group was added to the energy of its coincident gamma ray. The two sums are 1030±20 kev and 1040±20 kev. The agreement between these two sums and the maximum energy of the ground-state transition indicates that both gamma rays terminate at the ground state. This is consistent with the earlier reported decay scheme.[5]

Rb^{86}

The one known inner beta-ray group of Rb^{86} decays from a 2− level[5,11] to a suggested 2+ level[12] in Sr^{86}. The intensity of this group, as reported by Lyon and Strain,[13] is 9%.

A number of investigations[12,14–18] have been made of the inner beta-ray transition. The experimental measurements of the spectrum shape are not in agreement for energies less than 230 kev. However, the sources used in previous studies were relatively thick (~0.1 mg/cm^2 to ~2 mg/cm^2). This may explain the reported differences in the observed spectrum shape. It seemed desirable to re-examine this spectrum with thin sources. The 4π beta-ray scintillation spectrometer is ideally suited for this. With its high transmission, it is possible to obtain reasonable coincidence counting rates with thin sources.

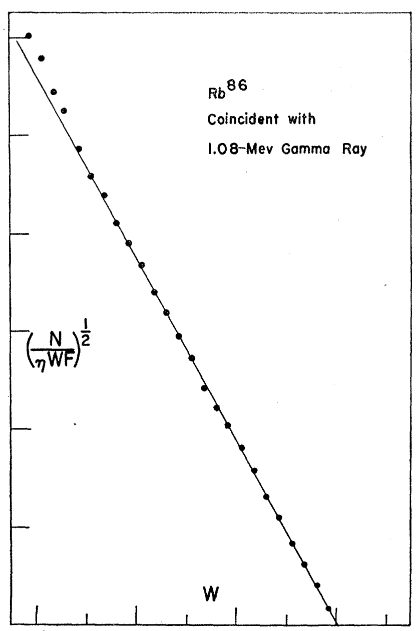

FIG. 4. Fermi-Kurie plot of the inner beta spectrum of Rb^{86}, measured in coincidence with the 1.08-Mev gamma ray.

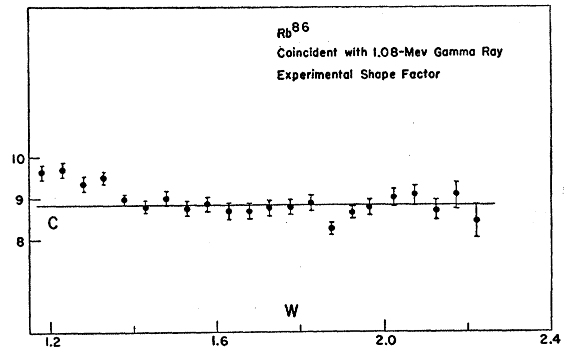

FIG. 5. Experimental shape factor of the inner beta spectrum of Rb^{86}.

The Rb^{86} activity was obtained from the Oak Ridge National Laboratory where it had been produced by neutron irradiation of Rb^{85}. The initial specific activity was given as 910 mC/g. A gamma spectrum of the source material showed a photopeak at 1.084±0.020 Mev. This agrees with the energy value of the gamma ray in Rb^{86} as measured by other investigators.[5] The half-life of the activity, which was obtained from measurements made over a period of five months, is 18.7±0.5 days.

Two experimental runs were made of the inner beta-ray group. This group was observed separately by recording only those beta rays which were in coincidence with gamma rays in the energy region between 1.03 and 1.25 Mev. From the assay of the source material given by the Oak Ridge National Laboratory, the source thicknesses were estimated to be 0.8 and 1.7 μg/cm^2 if one assumes that source material was uniformly distributed. The maximum decay correction was 6%. The background counts to total counts were less than 1%. For more than half of the points in both runs, the resolution corrections were less than 1%, the real to chance ratio was greater than 300, and the mean statistical errors were less than 3.5%. The Fermi-Kurie plots of the data appear linear for energies greater than 220 kev. Below 220 kev, both plots show a similar deviation away from the energy axis. The end-point energies, which are determined by a least-squares fitting of all the experimental points above 220 kev, are 718 and 716 kev. The data of the two runs were normalized between the energies of 80 and 618 kev. The Fermi-Kurie plot of the combined normalized data is given in Fig. 4. The end-point energy is 717±14 kev ($W_0=2.404m_0c^2$). At 100 kev, the deviation from a linear Fermi-Kurie plot amounts to an increase of 9.3% in the counting rate.

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[11] E. H. Bellamy and K. F. Smith, Phil. Mag. 44, 33 (1953).
[12] Pohm, Lewis, Talboy, and Jensen, Phys. Rev. 95, 1523 (1954).
[13] W. S. Lyon and J. E. Strain, Phys. Rev 95, 1500 (1954).
[14] H. R. Muether and S. L. Ridgway, Phys. Rev. 80, 750 (1950).
[15] A. G. Dmitriev and P. R. Zarubin, Izvest. Akad. Nauk S.S.S.R. Ser. Fiz. 18, 580 (1954).
[16] Macklin, Lidofsky, and Wu, Phys. Rev. 82, 334 (1951).
[17] R. S. Caird and A. C. G. Mitchell, Phys. Rev. 94, 412 (1954).
[18] J. Laberrigue-Frolow, J. phys. radium 16, 346 (1955).

A plot of the experimental shape factor of the combined data is shown in Fig. 5. The flags represent the mean statistical errors. For energies greater than 200 kev, it is possible within the limit of error to fit the data with a straight line parallel to the energy axis. However, the data are also consistent with a nonconstant shape factor which is a continuous function of the energy.

There are possible effects other than the 717-kev beta spectrum having a true nonstatistical shape which could give rise to the anomalous shape observed for energies less than 220 kev. Such effects are considered in the following paragraphs.

The deviation from a linear Fermi-Kurie plot could be obtained if a beta-ray transition with an end-point energy of 220 kev were present in the coincidence spectrum. If such a group exists, its intensity is $\sim$4.4% of the 717-kev beta-ray group. This was estimated by extrapolating the Fermi-Kurie plot to zero energy and by assuming that the 717-kev beta-ray group has an allowed shape.

If such a 220-kev beta-ray transition were due to an impurity, the impurity must have a half-life similar to that of Rb^{86}. Otherwise the two measurements made of the spectrum 22 days apart (more than one half-life of Rb^{86}) would have different shapes. Within the experimental errors, no such difference was observed. Also, the linearity of the exponential decay curve, followed for eight half-lives, indicates that there were no impurities with appreciably different half-lives. The impurity must also have a prompt gamma ray with an approximate energy of 1.1 Mev in coincidence with the 220-kev beta-ray transition. No likely impurity which meets these requirements could be found.

A second inner beta-ray group in Rb^{86} with a maximum energy of 220 kev could also be a source of the deviation. The group, in order to be measured, must be in coincidence with the 1.08-Mev gamma ray. In coincidence with both the beta-ray group and the 1.08-Mev gamma ray, there would then have to be a second gamma ray with an energy of 0.5 Mev. It was estimated that a gamma ray of this energy would have been detected in the study of the gamma spectrum if its intensity were 1% of the 1.08-Mev gamma-ray intensity. Such an intensity of this gamma ray is a factor of four or more too small to explain the deviation in the beta spectrum at low energy. The possibility that the 0.5-Mev gamma ray is highly converted can also be eliminated. To obtain a reasonable coincidence counting rate, the gamma-ray transition must have a half-life of less than a few microseconds. This indicates that the transition must go by either dipole or quadrupole radiation.[19] From Rose's tables,[20] the internal conversion coefficients for such transitions are less than one percent.

The sources and source backings and covers were thin. They would not be expected to give any deviation down to the lowest energy measured (80 kev). It is unlikely that the deviation is because of an instrumental effect, since the experimental setup has been found to measure other beta spectra reliably. It therefore appears likely that the spectrum shape found for the inner group is real and may be a result of the forbiddenness of the transition.

DISCUSSION

If the nonstatistical shapes of the 690-kev beta-ray group in Ag^{111} and the 717-kev group in Rb^{86} are real, the shapes should be explainable by the theory of beta decay. A theoretical shape factor for a once-forbidden, nonunique transition similar to that used by Plassmann and Langer[21] in the fitting of the RaE spectrum was used in the present work to fit the nonstatistical shape of the inner group in Ag^{111}. For this shape factor it was assumed that only S and T interactions contribute. At the time this correction was applied, all experimental evidence was in agreement with S and T interactions. However, recent investigations[22,23] indicate that V and A are probably the correct interactions. This correction factor contains two parameters ξ and χ which depend on the ratio of nuclear matrix elements. The experimental shape factor with the end-point energy $W_0=2.349m_0c^2$ is compared with the theoretical shape factor in Fig. 6. The uncertainty of the end-point energy of the spectrum is another parameter. A small change in the end-point energy changes the high-energy portion of the experimental shape factor appreciably. The limits placed on the error in the end point relative to the rest of the spectrum is $\pm\frac{1}{2}$%. The experimental shape factor with

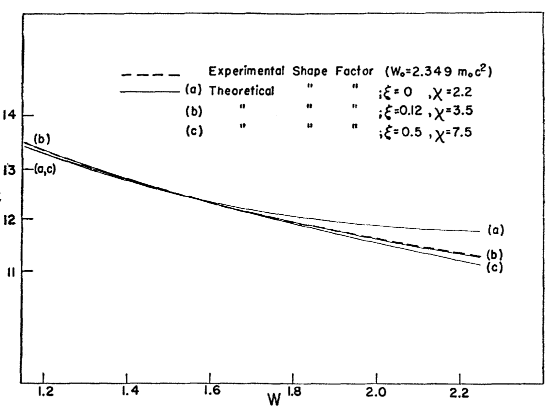

FIG. 6. Theoretical fits (in terms of ST) to the experimental shape factor of the low-energy, inner beta spectrum of Ag^{111} ($W_0=2.349m_0c^2$). Curves (b) and (c) are considered to be good fits. Curve (a) is the best fit with $\xi=0$.

[19] S. A. Moszkowski, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. 13.

[20] M. E. Rose, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Appendix IV.

[21] E. A. Plassmann and L. M. Langer, Phys. Rev. **96**, 1593 (1954).

[22] Herrmannsfeldt, Maxson, Stähelin, and Allen, Phys. Rev. **107**, 641 (1957).

[23] Goldhaber, Grodzins, and Sunyar, Phys. Rev. **109**, 1015 (1958).

end-point energies of the upper and lower limits ($W_0=2.343m_0c^2$ and $W_0=2.355m_0c^2$) were also fitted with the theoretical shape factor. Figure 7 shows a plot of the values of the parameters ξ and χ which give a theoretical shape factor in good agreement with the experimental shape factor whose end-point energy lies between these limits. From this figure it can be seen that there is a wide range in the values of these parameters which will give good agreement between the theoretical and experimental shape factors.

It should be emphasized that even though good theoretical fits to the experimental shape are found when only S and T interactions are assumed to contribute, it does not necessarily mean that these are the correct interactions. It is likely that good fits could also be obtained when V and A are assumed to be the contributing interactions.[24]

In the case of Rb^{86}, since the spin change is zero, the theoretical shape factor required can be more complicated. There are more parameters which can be adjusted than in the theoretical shape factor applied to the transition in Ag^{111}. With the additional parameters, it is very likely that a good theoretical fit of the experimental data can be found. The limits placed on the parameters in the shape factor which will give a good fit would probably be even less definitive than the limits obtained when fitting the 690-kev beta-ray group in Ag^{111} because of the additional parameters. Thus, no attempt was made to fit the experimental shape factor of the inner group of Rb^{86} with a theoretical shape factor.

An empirical correction of the form $(1+b/W)$ has been found to be necessary to linearize the Fermi-Kurie plots in the case of the allowed transitions Na^{22}, P^{32}, and In^{114} and in the case of the once-forbidden, unique transition Y^{90} after a once-forbidden, unique shape correction has been applied.[25–27] All four spectra have

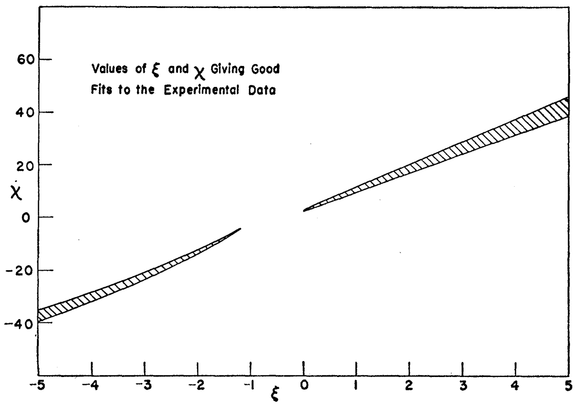

FIG. 7. Values of ξ and χ which give theoretical (ST) shape factors in good agreement with the experimental shape factor of the low-energy, inner beta spectrum of Ag^{111}.

nonstatistical shapes which cannot be explained by the present approximation of the theory of beta decay. For $0.20<b<0.35$, the empirical shape factor linearizes these Fermi-Kurie plots.

It is possible that such a correction should be included in the theoretical shape factor for once-forbidden, nonunique transitions. This correction with $0.20<b<0.35$ has a large effect on the nonstatistical shapes of the 690-kev beta spectrum in Ag^{111} and the 717-kev beta spectrum in Rb^{86}. Indeed, with a larger value of b this empirical shape factor will reasonably fit the experimental shape factor. Such a fit to the experimental shape is obtained for the 690-kev beta-ray group in Ag^{111} with $0.5<b<0.6$ and for the 717-kev group in Rb^{86} with $0.4<b<0.6$.

Nevertheless, these fits for Ag^{111} and Rb^{86} cannot be interpreted as evidence for the general applicability of the empirical factor $(1+b/W)$, since for a once-forbidden, nonunique transition, an approximation for the theoretical shape factor might be expected to have an energy dependence of just that form.

ACKNOWLEDGMENT

The authors wish to express their appreciation to Dr. W. G. Smith for his assistance in performing the chemical separation of the silver isotope.

[24] *Note added in proof.*—It is pointed out by T. Kotani and M. Ross [Phys. Rev. Letters 1, 140 (1958)] that the shape factor for a nonunique once-forbidden transition may be written in the form $1+aW+b/W$, where a and b are constants which may be interpreted ultimately in terms of the correct interaction forms.

[25] Hamilton, Smith, and Langer (to be published).

[26] O. E. Johnson, Ph.D. dissertation, Indiana University, 1956 (unpublished).

[27] L. M. Langer, *Proceedings of the Rehovoth Conference on Nuclear Structure* (North-Holland Publishing Company, Amsterdam, 1958), p. 437.