Nuclear Recoils Resulting from the Decay of Be^{7}*

PHILIP B. SMITH† AND JAMES S. ALLEN
Physics Department, University of Illinois, Urbana, Illinois
(Received October 3, 1950)

Be^{7} was deposited as a thin film on a Ta foil. The energy spectrum of the Li^{7} recoils from the reaction $Be^{7}+e_k \rightarrow Li^{7}+\nu+Q$ was studied. The observed spectra did not exhibit the monoenergetic character predicted by theory. The maximum energy of the recoils was 56.6 ± 1.0 ev, corresponding to a $Be^{7}-Li^{7}$ mass difference of 860 ± 8 kev. This mass difference, in combination with the best value of the $Li^{7}(p,n)Be^{7}$ threshold, yields 785 ± 8 kev for $n-H$. Coincidence measurements were made which indicated that monolayer surfaces were probably produced. Auger electrons following the K-capture were observed. The variation of the shape of the recoil energy spectrum was studied as a function of the temperature of the backing foil.

I. INTRODUCTION

As was first pointed out by Wang[1] it seems likely that an investigation of the recoil energy spectrum in the decay of Be^{7} will yield an excellent verification of the neutrino hypothesis. Since the decay scheme of Be^{7} is well known and quite simple, the results of such an experiment should be relatively unambiguous. In the 90 percent branch of the decay scheme a neutrino carries off practically the entire energy difference between Be^{7} and the ground state of Li^{7}. In this case the recoiling Li^{7} nucleus should have a well-defined momentum, and a corresponding unique energy. In the 10 percent branch a gamma-ray of 478 kev follows K-capture to the first excited state of Li^{7}. The recoiling nucleus in this branch will not be monoenergetic; however, if a coincidence technique is utilized to define the directions of both the recoil and the gamma-ray, the recoil momentum can have one of two possible values. In the hope of verifying the aforementioned theoretical conclusions the earlier work on this problem by one of the authors[2] has been repeated under more favorable conditions. In the earlier work the recoil energy spectrum from a thin layer of Be^{7} was investigated using a retarding potential method with an Allen-type electron multiplier as detector. In the present work better separation techniques were available, and a more refined retarding grid structure permitted a more precise determination of the maximum energy of the recoils. Preliminary results of these measurements have been reported previously.[3]

II. PREPARATION OF Be^{7} SURFACES

Selective evaporation was used to achieve the high order of separation required to prepare sufficiently thin Be^{7} surfaces. Several of the sources were obtained from the University of Illinois cyclotron, and one from the University of Chicago cyclotron. These were produced by both the $Li^{6}(d,n)Be^{7}$ and $Li^{7}(p,n)Be^{7}$ reactions. In addition, a separated cyclotron produced source was purchased from the AEC. The unseparated sources were in the form of metallic Li. These were converted to dry LiCl and then separated by selective evaporation. The separated source was supplied as a solution which was then dried and further separated as already mentioned. The general method of separation was the same as that developed by Sherwin.[4] The first step in the process was to determine the rate of transfer of activity as a function of temperature. This was done in a vacuum system provided with means of monitoring the activity transferred from the initial "hot" foil (3 mil Ta) to a catcher. It was found in the case of the untreated sources that the activity transferred to about 1400°C, as measured with an optical pyrometer. This confirms the conclusion reached by Allen[2] that the Be^{7} remained in its elemental state. The separated source from the AEC unfortunately showed an appreciable transfer rate over a wide range of temperatures. A high temperature fraction (about 1200–1400°C) of this source was used in the experiment. Actually, to prepare a source, a catcher foil was placed near the initial foil when it had reached the predetermined temperature of transfer. In some cases this catcher foil was then transferred to the final system and the recoil energy spectrum observed. In several other cases the catcher foil was moved to the final system and some of its activity was then transferred by a second evaporation to another catcher foil which could be moved into position in front of the detector without breaking the vacuum. In all cases the final foil was $\frac{1}{2}$ mil Ta. Surprisingly enough, the quality of surfaces prepared in the more complicated manner was no better than that of those prepared by the simple one-step process.

III. ENERGY SPECTRA OF NUCLEAR RECOILS

Figure 1 is a schematic diagram showing the arrangement of the grids and multiplier tube used to study the recoil energy spectra. The recoil counting rate was measured as a function of the potential of the retarding grid. The plot of these data is the integral energy spectrum of the recoils, except for the distortions introduced by the grid structure. Since these distortions

* This work was supported in part by the joint program of the AEC and ONR.
† Address: Physics Department, University of São Paulo, Caixa Postal 219-B, São Paulo, Brazil.
[1] K. C. Wang, Phys. Rev. **61**, 97 (1942).
[2] J. S. Allen, Phys. Rev. **61**, 692 (1942).
[3] P. B. Smith and J. S. Allen, Phys. Rev. **77**, 747 (1950).
[4] C. W. Sherwin, Phys. Rev. **73**, 216, 1173, 1799 (1949).

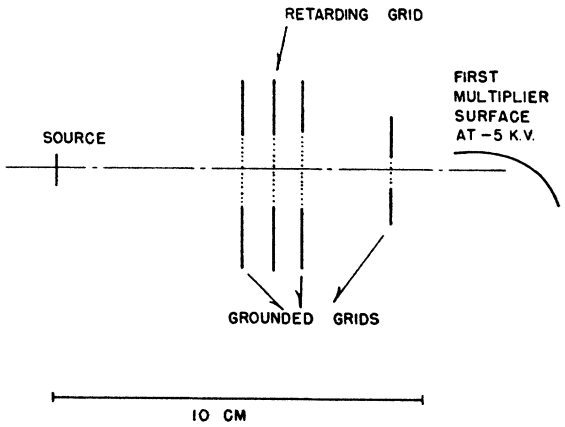

FIG. 1. Schematic diagram showing the location of the source, grids, and detector used to obtain the retarding potential curves for the Be^7 decay. The source was held at ground potential during the retarding potential measurements.

are almost impossible to calculate analytically, the grid system was calibrated by monoenergetic ions. A broad beam of ions was produced by surrounding the source holder with a cylindrical grounded grid and replacing the sources with a spodumene ion source on a Pt foil. The Pt foil was held at the desired positive voltage and then heated until the spodumene emitted ions.

Figure 2 shows the extreme high energy end of a retarding potential curve taken with a Be^7 surface and also a typical curve taken with a Pt-spodumene source. The curves were normalized at 58.75 retarding volts. This particular Pt-spodumene curve, corresponding to a positive ion energy of 56.6 ev, is shown because it matched the end point of the Be^7 curve so well. On the basis of this comparison, the value of 56.6 ± 1.0 ev was assigned to the upper end point of the Be^7 recoil spectrum. While it is clear from Fig. 2 that the end point could be assigned more accurately than ±1.0 ev, since there was no information on the difference between the contact potential of the Pt-spodumene source and the contact potential of the backing material of the Be^7 source, an error of ±1.0 ev was assigned which was thought to be large enough to take care of such a difference. The mass difference, Be^7-Li^7 calculated using this end point is 860 ± 8 kev. A neutrino mass of 10 kev would introduce an error of only 60 ev in this calculation, and, in view of the fact that recent measurements indicate that the neutrino mass is probably considerably less than 10 kev, it can be stated that the Be^7-Li^7 mass difference as measured by this method is essentially independent of neutrino mass. Using the Q value of $Li^7(p,n)Be^7$ as measured by Shoupp[5] *et al.* of $(-1.645\pm0.002$ Mev) and the reaction equation:

$$(n-H)=-Q-(Be^7-Li^7), \qquad (1)$$

$n-H$ is 785 ± 8 kev. In Table I are presented some recent values of $n-H$ with the corresponding values of the Be^7-Li^7 mass difference and the expected recoil energy. In each example the Be^7-Li^7 mass difference has been calculated using the Q value of the $Li^7(p,n)Be^7$ reaction as determined by Shoupp[5] *et al.*

Figure 3 presents the retarding potential curves for three surfaces and also the theoretically predicted curve D. This latter curve has not been corrected for grid distortion since the error introduced by the grids is very small compared to the difference between the theoretical curve and the measured curves. Table II summarizes the important characteristics of these surfaces. In the next section the method of obtaining the source strength and surface efficiencies is described.

Surface efficiency is defined as the ratio of the number of recoils which escape from the surface as ions to the total number of recoils occurring from the source. The efficiency is then a measure of the quality of a surface. Presumably, if all the recoils are able to escape from the surface the source is a monolayer. Since in the case of surface C the surface efficiency is 93 ± 42 percent, it is presumed that this source was a monlayer. The shape of the retarding potential curve C above 20 v indicates the emission of relatively more high energy recoils from this surface than from either of the thicker surfaces A or B. However, the counting rates in the case of surface C were so small that the statistical accuracy of the data was very poor. Although addi-

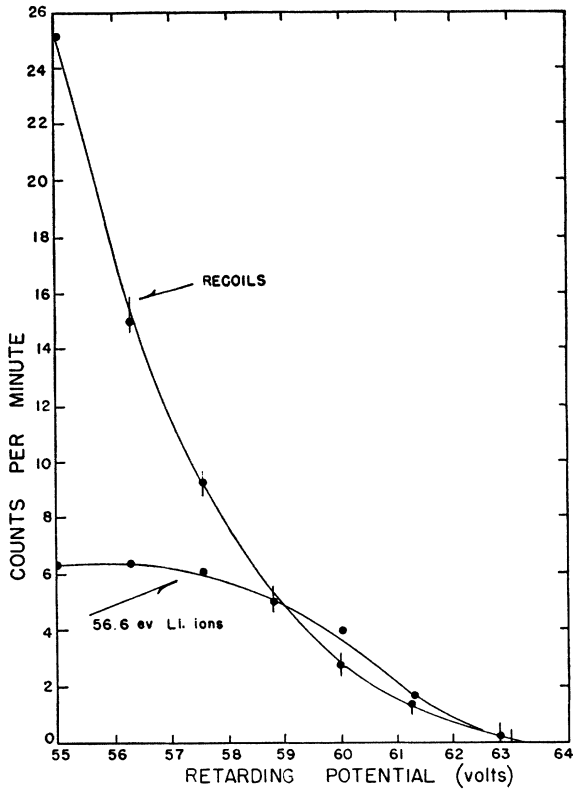

FIG. 2. The retarding potential curves for the recoils from the decay of Be^7 and for singly charged Li ions from a spodumene source.

[5] Shoupp, Jennings, Jones, and Garbuny, Phys. Rev. **75**, 336 (1949); Shoupp, Jennings, and Jones, Phys. Rev. **76**, 502 (1949).

tional data could have been obtained by counting for a longer period of time, the increasing importance of instrumental errors would have limited the accuracy of the data. We conclude from these results that very little improvement is to be expected if weaker sources on Ta substrata are used. It is of course possible that other substrata may produce less interference with the recoils. It seems unlikely that a very marked difference will occur, however, and thus the authors feel that the investigation of K-capture recoils with a gaseous source is a more fruitful field of investigation than the continuation of experiments with surface sources.[6]

TABLE I. Values of the n–H and Be^7–Li^7 mass differences.

$n-H$ (kev)	Be^7-Li^7 (kev)	Recoil energy (ev)	Reference
783±5	863±5	57.2±0.7	Stephens[a]
782±1	864±3	57.2±0.3	Taschek *et al.*[b]
800±4	846±5	55.0±0.5	Tollestrup *et al.*[c]
804±9	842±9	54.4±1.1	Bell & Elliott[d]
785±8	860±8	56.6±1.0	Present measurements

[a] W. E. Stephens, Phys. Rev. **76**, 181 (1949).
[b] Taschek, Jarvis, Argo, and Hemmendinger, Phys. Rev. **75**, 1268 (1949).
[c] Tollestrup, Jenkins, Fowler, and Lauritsen, Phys. Rev. **76**, 181 (1949).
[d] R. E. Bell and L. G. Elliott, Phys. Rev. **74**, 1552 (1948).

IV. COINCIDENCE MEASUREMENTS

For coincidence measurements the experimental arrangement was the same as for retarding potential measurements (Fig. 1) except that a scintillation crystal counter was placed behind the source. The crystal was so located that it detected the gamma-rays from the 10 percent branch at an angle of 180° from the direction of the recoils. The signal from the scintillation counter, indicating that a gamma-ray had been stopped in the crystal, was used as "zero-time" for the decay. The recoil traveled a known distance through field-free space to the grounded grid in front of the electron multiplier and was accelerated from there into the first dynode. The time of flight spectrum was recorded by a 20-channel delayed coincidence circuit. Details of this circuit will appear in another publication.

A detailed analysis of the factors causing the true and chance coincidence rates shows that the absolute source strength is given to a high degree of accuracy by

$$N_0=(N_c/N_t)(\alpha/\tau), \qquad (2)$$

where N_0 is the source strength in disintegrations per sec and N_c/N_t is the ratio of chance to true coincidence rates, α is a constant of the apparatus which can be evaluated and τ is the resolving time of the delayed coincidence apparatus.

A 70-hr coincidence run was taken on surface B. The coincidence counting rate was so low that not much can be said about the shape of the energy spectrum, except that it is in rough agreement with the retarding potential measurements for this surface shown in Fig. 3. However, it was possible to obtain an estimate of the rates of the true to chance counting rates which is accurate to about ±35 percent. This ratio was 23 for surface B and the source strength computed from Eq. (2) was 0.25 μc. A recoil counting rate of 5.4/sec was expected if the surface efficiency were 100 percent. Since the observed recoil rate was 3.2/sec, the actual surface efficiency was 59 percent. The source strengths and surface efficiencies of surfaces A and C were determined by measuring their gamma-ray and recoil counting rates under the same conditions as used for surface B. The characteristics of these three surfaces are shown in Table II.

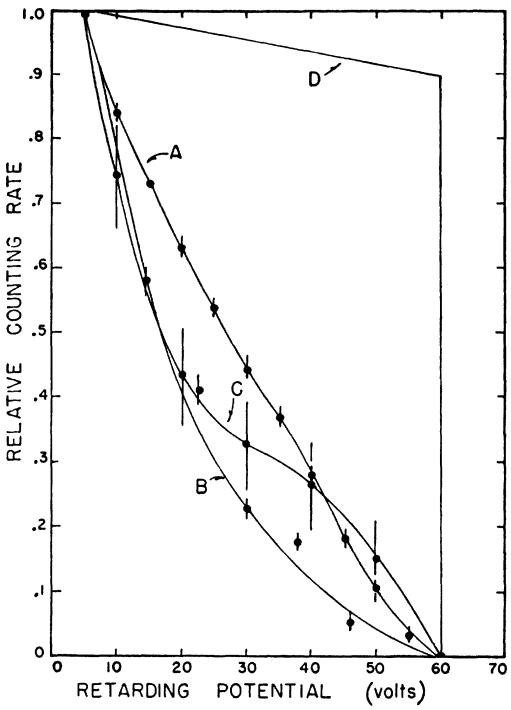

FIG. 3. A, B, and C are retarding potential curves for Be^7 surfaces of decreasing thickness. Curve D is the predicted curve for the recoils. The decrease in the relative counting rate as the retarding potential is increased is due to the distribution of recoil energies resulting from the nearly simultaneous emission of a gamma-ray and a neutrino.

TABLE II. Characteristics of the activated surfaces.

Surface	Gamma-count (counts/min)	Recoil count (counts/min)	Source strength	Source efficiency (percent)
A	3×10^4	2350	0.12±0.04 mc	1±0.35
B	61	189	0.25±0.09 μc	59±20
C	9	43.7	0.037±0.013 μc	93±32

V. HEATING OF SURFACES

In the earlier work of Allen[2] it was found that more recoils were observed and the maximum energy in-

[6] Since the conclusion of these experiments the authors have been informed that R. Davis of Brookhaven National Laboratory has obtained considerably better results with a somewhat different technique.

creased, for a short time after the backing foil was heated and allowed to cool. For this reason the present apparatus was designed in such a way that the backing foil could be kept warm continuously during observations.

It was found that the temperature of the foil during observation was not critical. Generally the foil was held at a temperature of about 500°C. The effect of allowing the surface to cool is shown strikingly in Fig. 4. Successive retarding potential curves were taken at intervals after the heating current had been turned off. The change was so rapid in the first few minutes that the first curve taken after shutting off the heating current is certainly distorted. It should be noticed that after an hour the upper energy endpoint had decreased by about 10 v, and the total number of recoils had decreased to one half of its initial value. This is in excellent agreement with the results of Allen.[2] It should also be remarked that the surface regained its original quality immediately after the heating current was again applied. It was impossible to get more accurate information because of the time (10 min) required to get even a rough retarding potential curve.

It is concluded that the remarkable change in the recoil energy spectrum previously described is due to the absorption of a layer of gas on the surface. Apparently, this layer forms very quickly when the surface is allowed to cool, and is rapidly driven off when the surface is reheated.

VI. OBSERVATION OF AUGER ELECTRONS

Ta was used as the substratum for all the surfaces made in this experiment. It is easy to handle, easy to clean, and can be heated well above the Be melting point without danger of melting or evaporating. Since its work function is somewhat smaller than the first ionization potential of Li (the recoiling atom), there is a very small probability of surface ionization. It was therefore desirable to show that Auger emission followed K-capture, since this would provide a mechanism for ionization. By running the first dynode of the multiplier at +500 v it was possible to detect these very low energy electrons. A retarding potential curve was taken on these electrons and it was found that they had a maximum energy of 36±5 ev. This value agrees with the expected energy of Auger electrons from Li. In addition, the total number of electrons was in rough agreement with the number of recoils observed. It was also found, when counting either ions or electrons, that if a sufficient retarding potential was applied to stop the most energetic particles, the counting rate fell to the same value as when the source was completely removed. This result indicated that x-rays from Li were almost entirely absent, since the multiplier should count these with high efficiency.

We conclude that Auger emission occurs in almost 100 percent of the decays, and that this is the probable explanation of the existence of charged recoils when Ta is used as the substratum.

VII. CONCLUSION

This experiment has shown that the reaction energies of the K-capture type of decay can be determined with good accuracy by measurement of the maximum energy of the recoiling ions. We have succeeded in measuring the Be^7-Li^7 mass difference in this way, and have found the value 860±8 kev. From this and the Q value of the reaction $Li^7(p,n)Be^7$ we have arrived at a value of 785±8 kev for the $n-H$ mass difference which agrees closely with the value 782±1 kev of Taschek *et al.*[7]

The experiment would also have provided convincing corroboration of the neutrino hypothesis if monoenergetic recoils had been observed. The coincidence measurements described above prove that, although very nearly monolayer surfaces were produced, the recoil energy spectra observed were in sharp disagreement with the predictions of the neutrino hypothesis. It must be concluded that either the Ta substrata distorted the recoil energy spectra, or else that the predictions of

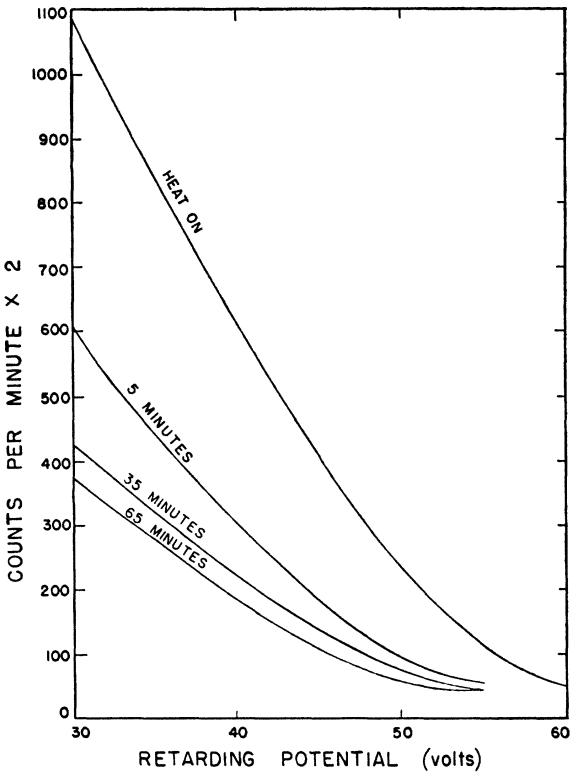

FIG. 4. Retarding potential curves for the recoils from Be^7. The top curve was obtained with the source at 500°C. The lower curves were taken at various intervals of time after the source had returned to room temperature.

[7] Taschek, Jarvis, Argo, and Hemmendinger, Phys. Rev. **75**, 1268 (1949).

beta-decay theory are not fulfilled in this decay process. Certainly all other experimental results support the former conclusion.

The authors wish to express their appreciation to the crews of the University of Illinois and the University of Chicago cyclotrons, who provided most of the Be^7 used in these experiments. They are also grateful for a very valuable discussion of the experiment with Dr. Raymond Davis of the Brookhaven National Laboratory, to a great deal of excellent advice on surface preparation given by Prof. C. W. Sherwin of the Department of Physics, University of Illinois.

PHYSICAL REVIEW VOLUME 81, NUMBER 3 FEBRUARY 1, 1951

A Simplification of the Hartree-Fock Method

J. C. SLATER
Massachusetts Institute of Technology, Cambridge, Massachusetts*
(Received September 28, 1950)

It is shown that the Hartree-Fock equations can be regarded as ordinary Schrödinger equations for the motion of electrons, each electron moving in a slightly different potential field, which is computed by electrostatics from all the charges of the system, positive and negative, corrected by the removal of an exchange charge, equal in magnitude to one electron, surrounding the electron whose motion is being investigated. By forming a weighted mean of the exchange charges, weighted and averaged over the various electronic wave functions at a given point of space, we set up an average potential field in which we can consider all of the electrons to move, thus leading to a great simplification of the Hartree-Fock method, and bringing it into agreement with the usual band picture of solids, in which all electrons are assumed to move in the same field. We can further replace the average exchange charge by the corresponding value which we should have in a free-electron gas whose local density is equal to the density of actual charge at the position in question; this results in a very simple expression for the average potential field, which still behaves qualitatively like that of the Hartree-Fock method. This simplified field is being applied to problems in atomic structure, with satisfactory results, and is adapted as well to problems of molecules and solids.

I. INTRODUCTION

THE Hartree-Fock equations[1] furnish the best set of one-electron wave functions for use in a self-consistent approximation to the problem of the motion of electrons in the field of atomic nuclei. However, they are so complicated to use that they have not been employed except in relatively simple cases. It is the purpose of the present paper to examine their meaning sufficiently closely so that we can see physically how to set up a simplification, which still preserves their main features. This simplified method yields a single potential in which we can assume that the electrons move, and we shall show the properties of this field for problems not only of single atoms but of molecules and solids, showing that it leads to a simplified self-consistent method for handling atomic wave functions, easy enough to apply so that we can look forward to using it even for heavy atoms.

II. THE HARTREE-FOCK EQUATIONS AND THEIR MEANING

It is well known that the Hartree equations are obtained by varying one-electron wave functions $u_1(x)$, $u_2(x)$, $\cdots u_n(x)$, in such a way as to make the energy $\int u_1^*(x_1)\cdots u_n^*(x_n)Hu_1(x_1)\cdots u_n(x_n)dx_1\cdots dx_n$ an extreme, where H is the energy operator of a problem involving n electrons in the field of certain nuclei, and where the functions u_i are required to be normalized. Similarly the Hartree-Fock equations, as modified by Dirac,[2] are obtained by varying the u_i's so as to make the energy

$$\frac{1}{n!}\int \begin{vmatrix} u_1^*(x_1)\cdots u_1^*(x_n) \\ \cdots\cdots\cdots\cdots \\ u_n^*(x_1)\cdots u_n^*(x_n) \end{vmatrix} H \begin{vmatrix} u_1(x_1)\cdots u_1(x_n) \\ \cdots\cdots\cdots\cdots \\ u_n(x_1)\cdots u_n(x_n) \end{vmatrix} dx_1\cdots dx_n$$

an extreme, where in this latter expression the u's are assumed to be functions depending on coordinates and spin, and where the integrations over the dx's are interpreted to include summing over the spins. The Hartree-Fock equations can then be written in the form

$$H_1u_i(x_1)+\left[\sum_{k=1}^{n}\int u_k^*(x_2)u_k(x_2)(e^2/4\pi\epsilon_0 r_{12})dx_2\right]u_i(x_1)$$
$$-\sum_{k=1}^{n}\left[\int u_k^*(x_2)u_i(x_2)(e^2/4\pi\epsilon_0 r_{12})dx_2\right]u_k(x_1)$$
$$=E_iu_i(x_1). \quad (1)$$

Here H_1 is the kinetic energy operator for the electron of coordinate x_1, plus its potential energy in the field of

* The work described in this paper was supported in part by the Signal Corps, the Air Materiel Command, and the ONR, through the Research Laboratory of Electronics of M.I.T.

[1] J. C. Slater, Phys. Rev. **35**, 210 (1930); V. Fock, Z. Physik **61**, 126 (1930); L. Brillouin, *Les Champs Self-Consistents de Hartree et de Fock*, Actualités Scientifiques et Industrielles No. 159 (Hermann et Cie., 1934); D. R. Hartree and W. Hartree, Proc. Roy. Soc. **A150**, 9 (1935); and many other references.

[2] P. A. M. Dirac, Proc. Cambridge Phil. Soc. **26**, 376 (1930).