The Disintegration of Radium D

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The disintegration of radium D has been investigated using a gaseous source of lead tetramethyl in a proportional counter. The form of the low energy β-spectrum coincident with the 46.7-kev γ-ray has been determined. The end point has been found to be 15.2 ± 1 kev. The shape of the spectrum shows good agreement with the Fermi theory for an allowed transition, in which the effect of screening on the Coulomb factor is neglected. The results indicate that the number of β-particles below 3 kev is somewhat less than than predicted by theory, neglecting screening. It is also concluded that the change in binding energy of the atomic electrons (about 10 kev) released in the decay, is included in the energy of disintegration of the β-particles, and is shared between the β-particle and neutrino.

INTRODUCTION

THE disintegration of radium D has been investigated by many observers.[1] The exact measurement of the shape of the β-spectrum has, in particular, posed a difficult experimental problem, because of the extremely low energy of the β-particles. The form of a low energy spectrum for the high value of atomic number involved, i.e., $Z=82$, is also of considerable theoretical interest. Recently Insch, Balfour, and Curran[2] have examined the β-spectrum using proportional counters and a solid source, and have obtained the value of 18 ± 2.5 kev for the end point. In the work to be described, we have examined the disintegration using RaD, which is an isotope of lead, in the form of lead tetramethyl as a gaseous constituent in a proportional counter. (In one of the earliest investigations carried out on RaD, Richardson and Leigh-Smith[3] used radioactive lead tetramethyl in a cloud chamber.) This method clearly obviates the difficulties of self-absorption and backscattering and, as will also be seen, enables a very accurate energy calibration of the β-spectrum to be made and gives the form of the β-spectrum directly, in contrast with experiments using solid sources. The feasibility of the experiment was demonstrated by the preliminary verification of the following facts: (a) a counter filled with argon, methane, and including a few mm pressure of pure dry lead tetramethyl shows excellent proportionality; (b) the amount of sticking of the lead tetramethyl vapor to the counter walls, during the time of the experiment, was negligible; (c) the amount of RaE present in the counter during the experiment could be kept very small.

DESCRIPTION OF METHOD AND APPARATUS

Other work[4,5] has shown that a 46.7-kev γ-ray occurs in about 65 percent of disintegrations and is highly converted ($\sim$96 percent). If this γ-ray is emitted within a time short compared with the resolving time of the counter (a few microseconds) one would expect that 65 percent of the pulses obtained in a proportional counter containing RaD in the gas phase should correspond to the sum of the energies of the γ-ray and the β-particle, if all the secondary radiations are absorbed in the counter gas, giving rise to a peak in the region of 46.7 kev, and as the β-spectrum would be superposed on this gamma-line, one could get the shape of the β-spectrum from an analysis of the high energy side of the observed peak. An identical pulse distribution would also be produced by low energy β-particles coincident with cascade converted γ-rays, produced by indirect de-excitation of the 46.7-kev level, in some of the remaining 35 percent of the disintegrations. The counter was therefore designed to give the shape of this profile accurately. One might also expect, in relatively few cases, to get the pure low energy β-spectrum when all rays coincident with the decay are unconverted and escape from the counter, or if all the accompanying γ-rays are delayed due to metastability.

In order to minimize the wall effect and thus to diminish the distortion in the pulse distribution at low energies, the counter was made so that the range of the ionizing particles should be small compared with its dimensions. Most of the 46.7-kev γ-rays are converted in the L shell, resulting in conversion electrons of 30.5-kev energy and of maximum range (in the gas filling used, which was 60-cm argon+14 cm methane) about 1.2 cm. Only 12 percent of the γ-rays are converted in the MNO shells, giving rise to 44-kev conversion electrons of maximum range about 2.4 cm. The end effect, which causes distortion in the regions extending up to a distance of the order of a radius of the counter from each end, was kept small by making the counter long. The counter dimensions were thus chosen to be 9-cm in diameter and 72-cm long. The counter wall was made of brass and provided with a polythene window to permit energy calibrations using external sources of γ- and x-rays. The anode was a 4-mil diameter tungsten wire attached at each end to tungsten rods which were sealed into double metal-to-glass seals, the intermediate metal seals forming guard rings. The wire was shielded

[1] N. Feather, Nucleonics **5**, 22 (July, 1949).
[2] Insch, Balfour, and Curran, Phys. Rev. **85**, 805 (1952).
[3] H. O. W. Richardson and A. Leigh-Smith, Proc. Roy. Soc. (London) **A160**, 454 (1937).
[4] L. Cranberg, Phys. Rev. **77**, 155 (1950).
[5] D. K. Butt and W. D. Brodie, Proc. Phys. Soc. (London) **A64**, 791 (1951).

at each end of the counter by glass sleeves 5 mm in diameter. Electrostatic screening for the whole of the counter was provided by a large wire cage. The high voltage (working voltage was usually about 2400 volts) was obtained from a stabilized power supply, and the linear amplifier (E. K. Cole Ltd. Type 1008) was adjusted to work at a differentiation time of 10 microseconds and an integration time of 0.7 microsecond. The pulse distribution obtained from the counter could be recorded with an oscillograph equipped with a moving film camera, or by use of a single channel pulse analyzer. Good agreement was obtained using both methods, and the final results were obtained using the pulse analyzer.

The method of preparation of the radioactive lead tetramethyl is described in the appendix. Preliminary experiments in which nonradioactive lead tetramethyl was introduced in various amounts into a counter, together with argon and methane, showed that in order

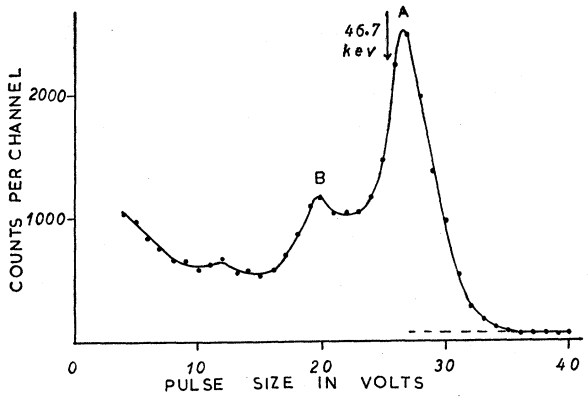

FIG. 1. Pulse-height distribution from proportional counter containing radioactive lead tetramethyl vapor, for the energy range 7 to 75 kev.

to obtain good proportionality (as checked by the line width obtained from external x-rays and γ-rays) the sample of lead tetramethyl must be thoroughly dried This was best achieved by desiccation with phosphorous pentoxide for periods of about 24 hours. It was at first thought that owing to the low energy of recoil of the Bi formed by the decay of RaD to RaE (0.08 ev for most of the disintegrations), there was some danger that volatile radioactive bismuth trimethyl might grow in the sample of liquid lead tetramethyl with a period of five days. Since, however, the age of the sample (from 1 to 3 days) produced no detectable difference in the pulse distribution obtained, it may be concluded that bismuth trimethyl is either not formed at all in the liquid lead tetramethyl or at the most in a very few percent of the disintegrations of the RaD. After introducing the lead tetramethyl vapor into the counter, however, measurements must be made within a few hours, because RaE, which has a half-life of 5 days, will grow inside the counter. Further preliminary experiments showed that when a few mm pressure of radioactive lead tetramethyl is introduced into the counter, only about 1 percent of it adheres to the walls, during times of the order of those used (not more than four hours).

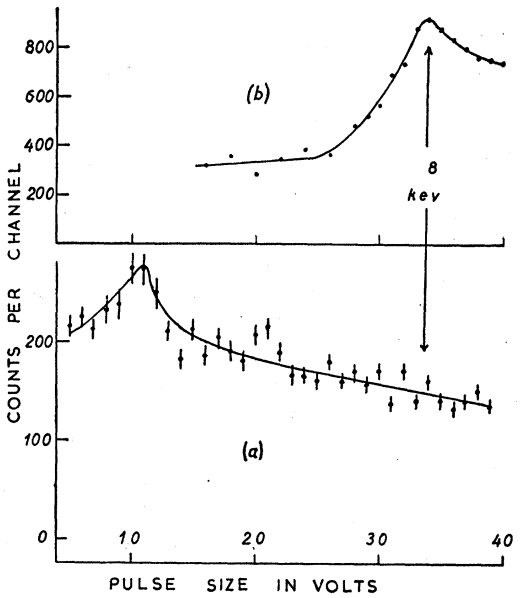

FIG. 2. (a) Pulse-height distribution from counter containing radioactive lead tetramethyl vapor, for the energy range from 1 to 9.5 kev. (b) Pulse distribution from a superimposed external source of RaD, taken with the same counter filling, to provide an energy calibration.

Energy calibrations were made by superimposing the L x-rays and γ-rays from an external source of RaD. The energies of the most intense of these radiations have already been investigated accurately, and we have also checked the values of these energies against the K x-rays of Fe^{55} which decays by K capture. As will be seen from the discussion of the results, the unconverted γ-ray at 46.7 kev provides a very useful calibration and defines the zero of the β-spectrum, since (as suggested above and, in fact, as confirmed) in the majority of the disintegrations the β-particles appear simultaneously with the quanta of 46.7 kev. At the same time the line widths obtained with the superimposed sources give a check of the proportionality of the gas mixture during the actual experiment.

EXPERIMENTAL RESULTS

The pulse distribution for the energy range of 7 to 75 kev is shown in Fig. 1. (This was obtained in an experiment in which 2 mm of lead tetramethyl vapor, together with 60-cm argon and 14-cm methane, gave a total counting rate of about 3000/sec.) The lower energies were examined at greater amplifications, and in Fig. 2(a) the range from 1 to 9.5 kev is shown. Figure 2(b) shows the distribution from the superimposed source of RaD (obtained by subtracting the pulse distribution without the external source from that ob-

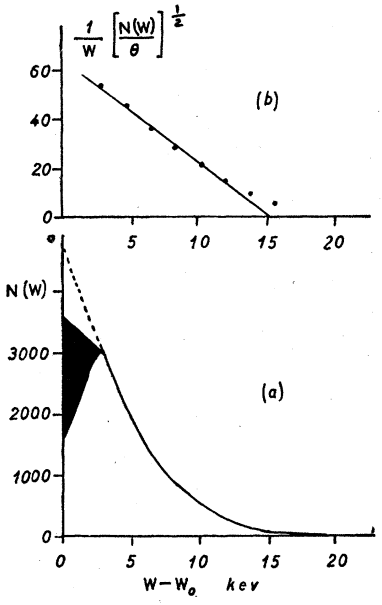

FIG. 3. (a) The solid curve shows the form of the β-spectrum found for RaD, the black area indicating the region of uncertainty below 3 kev. The dotted curve indicates the shape predicted by the Fermi theory, in the region where there is significant departure between the theoretical and experimental curves. (b) A Bleuler-Zünti plot of the experimentally found β-spectrum.

tained when the external source was superimposed) and displays the peak at about 8 kev, due to fluorescent x-rays of copper and zinc excited in the brass wall (see below).

Careful examination in the neighborhood of peak A, (Fig. 1), using the 46.7-kev γ-rays from an external source of RaD of approximate strength 0.5 millicuries, showed that the peak A was shifted towards higher energies by 2.7 ± 0.5 kev with respect to the peak of the superimposed 46.7-kev line. A strong external source must be used in this case owing to the low efficiency of the counter gas for the γ-rays and the necessity of obtaining a counting rate comparable with the internal counting rate; most of the softer L x-rays were removed by the use of a suitable filter.

THE SHAPE OF THE LOW ENERGY β-SPECTRUM

The width of the peak A (Fig. 1) is considerably greater than that obtained with an external source of 46.7-kev γ-rays (which has a semihalf-width of 1.7 kev as measured in the same counter with nonradioactive fillings). It is therefore to be interpreted as being due to the low energy β-spectrum coincident with quanta of total energy 46.7 kev as suggested above, and confirms that the lifetime of the 46.7-kev transition is less than the resolving time of the counter. Sometimes, however, the bismuth L x-rays produced after internal conversion in the L shell (mainly 10.8 and 13.0 kev) may escape from the counter. This gives rise to the peak B which occurs at 36.7 kev. The average escape probability has been very roughly estimated as 0.6–0.7, and using the value obtained by Kinsey[6] for the fluorescent yield of the L x-rays (0.47) it is possible to calculate the expected ratio of the areas of the two peaks; the experimental results are found to be in good agreement. The number of pulses corresponding to β-emission coincident with absorbed quanta of total energy 46.7 kev is found to be 73 percent of the number of disintegrations. The residual tail of low intensity on the high energy side of peak A can be accounted for by the random superposition of pulses due to the finite pulse duration and high counting rate, and probably also to the presence of a very small amount of RaE, perhaps in the form of bismuth trimethyl.

To determine the shape of the β-spectrum from the profile of peak A, after subtracting the extrapolated low intensity tail, it is necessary to take into account the statistical spread of pulse sizes, corresponding to a given energy, which is a characteristic of the proportional counter. Using the pulse distribution experimentally obtained from the 46.7-kev γ-rays from an external source, it is possible to estimate the amount of distortion of the β-spectrum associated with A. Since the semihalf-width at 46.7 kev is only 1.7 kev, it is found that the experimental curve must closely approximate to the shape of the β-spectrum down to about 3.5 kev (i.e., 50.2 kev in curve of Fig. 1). Below 3 kev, however, the shape cannot be uniquely determined from the results. Figure 3 shows the form of the β-spectrum obtained after correction for the statistical spread by successive approximations, and allowing for the small contribution from the peak B; the region of uncertainty below 3.5 kev is indicated by the shaded area. Further information on the β-spectrum below 3 kev is provided by the pulse distribution at low energies, shown in Fig. 2(a). As was explained above it may be expected that this distribution contains the pure low energy β-spectrum, though there may also be other causes contributing pulses to this region. It is interesting to see that the curve has a maximum at 2.4 kev. The existence of such a maximum in the β-spectrum would be consistent with the curve of Fig. 3(a).

COMPARISON OF THE β-SPECTRUM WITH THEORY

The Fermi theory for an allowed transition gives the form of the spectrum as

$$N(W)dW \propto p(W-W_0)^2F(Z,W)WdW.$$

The Coulomb factor $F(Z, W)$, neglecting the effects of screening, for which no adequate calculations are available (for high Z and such low energies), is given to a good approximation, for RaD, by $1/p$. Also W, the sum of the rest energy and the kinetic energy of the particle, is approximately constant over the range of the spectrum. Therefore, the form of the Fermi function approximates a parabola, $N(W) \propto (W-W_0)^2$. The effect of the Coulomb field is to increase the number of electrons in the low energy region and thus to remove the maximum normally obtained in allowed spectra of higher energies. Figure 3(b) shows a plot of $(1/W)\times[N(W)/\theta]^{\frac{1}{2}}$ against $W-W_0$ where θ is the funtcoin

[6] B. Kinsey, Can. J. Research **26**, 421 (1948).

defined and calculated by Bleuler and Zünti[7]), and it is seen that a good straight line is obtained from 3 to 13 kev; extrapolated, it gives an end point of 15.2 kev, which is also the mean value found from several sets of measurements. The accuracy of the above value is estimated as ± 1 kev. In Fig. 3(a) the experimental curve closely coincides with the Fermi curve for $N(W)$ as a function of $W-W_0$, assuming an end point of 15.2 kev, down to 3 kev. The dotted curve below 3 kev shows the Fermi distribution in this region.

The results indicate that there are fewer electrons below 3 kev than predicted by the Fermi theory neglecting screening. It seems very likely that the departure can be explained by the effect of screening, since the latter reduces the effective Coulomb field and would thus tend to restore the maximum normally obtained.

As first pointed out by Schwartz and Edwards,[8] an experimental study of the β-spectrum of RaD should yield conclusive information on the effect of the change in binding energy of the atomic electrons on the shape of the β-spectrum. For RaD, calculations based on the Thomas-Fermi statistical model[9] lead to a value of about 10 kev for the change in atomic binding energy during the decay, and this is seen to be of the same order of magnitude as the maximum energy of the β-particles.

Since the above results show good agreement with the Fermi theory, at least down to 3 kev, it may be concluded that the relatively large change in atomic binding energy is included in the energy of the β-disintegration and is shared between the β-particle and the neutrino. This conclusion is in agreement with those recently obtained from theoretical considerations by Schwartz[10] and by Serber and Snyder.[11]

THE PULSE DISTRIBUTION IN THE LOW ENERGY REGION

In Fig. 1 the pulse distribution shows a rise towards low energies below 16 kev, and there is also an indication of a maximum at about 23 kev. A disturbing contribution to the low energy region is made by the wall and end effects which cause some of the high energy pulses to be thrown down into lower energies. The fraction of pulses thus distorted and their approximate distribution has been estimated theoretically (about 20 percent of the total number of pulses). Figure 4 shows the distribution obtained after subtracting this correction, the points below 7 kev being obtained from the results at higher amplifications [as shown in Fig. 2(a)] after normalizing the channel width. The correction hardly affects the pulse distribution at high energies, already discussed.

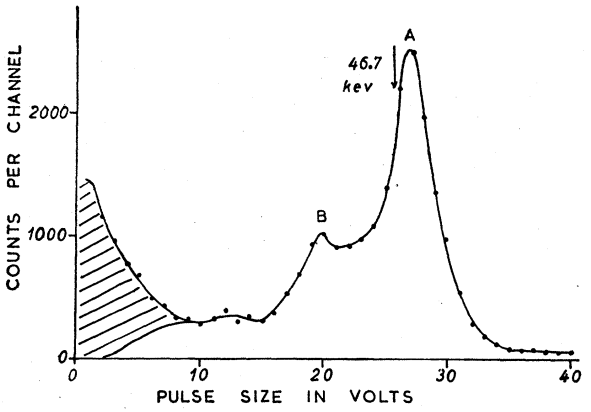

FIG. 4. The pulse-height distribution (obtained from results shown in Figs. 1 and 2) after correction for wall and end effects. The shaded area indicates the maximum possible contribution from the unaccompanied low energy β-spectrum.

The number of pulses below 15 kev in the corrected distribution amounts to about 13 percent of the total number of pulses. The low energy β-spectrum accompanied by unconverted 46.7-kev γ-rays (for which the counter efficiency is very low, and which therefore in most cases escape from the counter) should occur in only about 3.5 percent of the disintegrations.[1] This suggests that there is a contribution in this energy region from β-particles accompanied by metastable or unconverted γ-rays (the latter having a low efficiency for detection) other than the 46.7-kev γ-rays. The shaded area in Fig. 4 represents the largest contribution of the pure low energy β-spectrum which is compatible with the shape of the curve and also the form of the β-spectrum as previously found from the measurements at high energy described above. The exact significance of the remaining distribution (from 0–30 kev), obtained after deducting the shaded area, is not clear. It may include some low energy metastable γ-rays, in accordance with the hypothesis made above to account for the intensity of the pure low energy β-spectrum. In addition it is possible that a second more energetic β-spectrum exists of relatively low intensity; this would hardly affect the shape obtaining at high energies. This possibility has already been suggested by Cranberg[4] to explain his results using a magnetic β-ray spectrometer.

In a previous note[12] we have mentioned that there was evidence to believe that such a more energetic β-spectrum was in cascade with a γ-ray at 7.8 kev. A γ-ray of this energy has been reported by Curran, Angus, and Cockcroft[13] using proportional counters and was also apparently detected by the present authors.[14] Mr. D. West (of Harwell, England) has, however, kindly informed us in a private communication that he

[7] E. Bleuler and W. Zünti, Helv. Phys. Acta **19**, 375 (1946).
[8] H. M. Schwartz and R. R. Edwards, J. Chem. Phys. **19**, 385 (1951).
[9] Z. Hund, *Handbuch der Physik* (Julius Springer, Berlin), second edition, p. 621.
[10] H. M. Schwartz, Phys. Rev. **86**, 195 (1952).
[11] R. Serber and H. S. Snyder, Phys. Rev. **87**, 152 (1952).
[12] A. A. Jaffe and S. G. Cohen, Phys. Rev. **86**, 1041 (1952).
[13] Curran, Angus, and Cockroft, Phil. Mag. **40**, 36 (1949).
[14] S. G. Cohen and A. A. Jaffe, Phys. Rev. **86**, 800 (1952).

obtained no trace of a line at 7.8 kev using a proportional counter with *glass* walls and a thin mica window, and he suggests that the 7.8-kev line, found by the above-mentioned authors, has its origin in the fluorescent x-rays of copper (8.05 kev) excited in the wall of the counter by the L x-rays of RaD. We have since confirmed by absorption experiments, using the same *brass* walled counter with which we had previously observed the "7.8-kev line," that this line is in fact produced by fluorescent x-rays from the copper and zinc in the counter wall.

We wish to thank Dr. S. Israelshvili of the Department of Organic Chemistry, Hebrew University, for his advice on the synthesis of lead tetramethyl, and also Mr. S. Friedman, who built the pulse analyzer.

APPENDIX

Preparation of Radioactive Lead Tetramethyl

RaD (dissolved in nitric acid and in equilibrium with its products) was added to a carrier solution containing usually from about 5–10g of ordinary lead in the form of lead nitrate. By the addition of excess dilute HCl the RaD was precipitated with carrier as lead chloride, and this precipitate was washed several times with water and acetone and then thoroughly dried. It was found by preliminary experiment that the lead chloride so obtained contained less than 3 percent of the RaE and RaF originally in equilibrium with the RaD. This precipitate was refluxed with an ethereal solution of the freshly prepared Grignard reagent (methyl magnesium iodide) for about 3 hours. After destroying any excess Grignard by the addition of water, the resulting ethereal solution of lead tetramethyl was separated, dried with calcium chloride, and distilled, the lead tetramethyl (bp 110°C) being collected.

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Proof That Probability Density Approaches $|\psi|^2$ in Causal Interpretation of the Quantum Theory

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In two previous papers a causal interpretation of the quantum theory was developed which involved the hypothesis that a quantum-mechanical system contains a precisely defined particle variable $\mathbf{x}$ but that, at present, we are restricted to calculating the probability density $P(\mathbf{x}, t)$ that the particle is at the position $\mathbf{x}$. It was shown that the assumption that $P(\mathbf{x}, t)=|\psi(\mathbf{x}, t)|^2$ is consistent, in the sense that if it holds initially, the equations of motion of the particles will cause this relation to be maintained for all time. In this paper, we extend the theory by showing that as a result of random collisions, an arbitrary probability density will ultimately decay into one with a density of $|\psi(\mathbf{x}, t)|^2$. Since all quantum-mechanical experiments to date have been concerned with statistical ensembles of systems that have been colliding with other systems for a very long time, it is therefore inevitable that as we draw samples from such ensembles, the probability density of systems with particles at the point $\mathbf{x}$ will be equal to $|\psi(\mathbf{x}, t)|^2$.

In the previous papers we also pointed out that, within the conceptual framework of the causal interpretation, it was possible to suggest mathematical theories more general than are permitted by the usual interpretation and that these more general theories might be needed in the domain of 10^{-13} cm, where present theories seem to fail. However, if these more general theories should apply at the level of 10^{-13} cm, then there would be a tendency to create discrepancies between P and $|\psi|^2$, a tendency whose cumulative effects should be felt even at the atomic level, where the more general theory ought to approach the usual theory. However, because those discrepancies have been shown to die out as a result of collisions, we can expect that under normal conditions the difference between P and $|\psi|^2$ would be negligible. Conditions are suggested, however, in which this difference might be appreciable, and experiments are indicated which might be able to test for the existence of such discrepancies.

I. INTRODUCTION

IN two recent papers,[1] (to be denoted hereafter by I and II, respectively) the author has proposed a causal reinterpretation of the quantum theory, based on the following hypotheses:

(a) A quantum-mechanical system, such as an electron, consists basically of a particle having a precisely defined position, which varies continuously as a function of the time.

(b) This particle is acted on not only by the classical potential $V(\mathbf{x}, t)$ but also by an additional quantum-potential $U(\mathbf{x}, t)$, which is important at the atomic level but negligible at the macroscopic level.

(c) If we write $\psi=Re^{iS/\hbar}$, where ψ is the wave function and R and S are real, then the quantum potential is given by

$$U(\mathbf{x}, t)=-(\hbar^2/2m)\nabla^2R(\mathbf{x}, t)/R(\mathbf{x}, t). \quad (1)$$

The equation of motion of the particle then takes the form

$$md^2\mathbf{x}/dt^2=-\nabla\{U(\mathbf{x}, t)+V(\mathbf{x}, t)\}. \quad (2)$$

To obtain the same predictions for all experimental results as are obtained from the usual interpretation of the quantum theory it is necessary, however, to make the following additional special assumptions (see paper I, p. 171):

(1) The ψ field satisfies Schrödinger's equation.

(2) The particle velocity is restricted to $\mathbf{v}=\nabla S(\mathbf{x})/m$.

[1] D. Bohm, Phys. Rev. **85**, 166 (1952) (paper I); **85**, 180 (1952) (paper II). See also, Phys. Rev. **87**, 389 (1952).