

(3) Evaluation of $M_{M.D.}$.

$$M_{M.D.} = -\frac{b(\beta/2)^p}{|J_{iq}(\beta')| |\Gamma(1+iq)| \Gamma(1+p)} \times \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} \sum_{l=0}^{\infty} Im \left[\frac{A_l A_m A_n}{p+2n+2m+iq} \right],$$

$$A_l = \frac{(-1)^l (\beta'/2)^{2l} \Gamma(1-iq)}{l! \Gamma(l+1-iq)}, A_m = \frac{(-1)^m (\beta'/2)^{2m} \Gamma(1+iq)}{m! \Gamma(m+1+iq)}, A_n = \frac{(-1)^n (\beta/2)^{2n} \Gamma(1+p)}{n! \Gamma(n+1+p)}.$$

AUGUST 15, 1937

PHYSICAL REVIEW

VOLUME 52

Note on Nuclear Photoeffect at High Energies

F. KALCKAR, J. R. OPPENHEIMER AND R. SERBER
University of California, Berkeley, California

(Received May 19, 1937)

In I we consider nuclear transmutations when the energy is so high that the levels of the compound nuclei formed have a spacing smaller than their breadth, and investigate the relations between the cross sections for the various possible reactions and the decay constants characteristic of the compound nuclei. When the collision may be treated as a resonance effect, these relations take a simple form. In II we apply these considerations to the photoelectric disintegration of nuclei of intermediate atomic weight by 17 Mev γ -rays, and suggest that the yields should show a marked increase for γ -rays of somewhat lower energy. In III we study the formal connection between the evaluation of transmutation probabilities here given and the resonance formulae appropriate for lower excitation energies, and show that both descriptions may be derived as limiting cases from the same formalism.

I

BOHR'S analysis¹ of nuclear phenomena has shown the great importance, for an understanding of the processes of nuclear disintegration, of the capture of the incident particle with the formation of an intermediate nucleus having a considerable excitation energy, and so long a lifetime that the subsequent transformation of this unstable structure by the emission of particles (or γ -rays) may be treated as an independent process. A formalism consistent with these ideas, and especially appropriate to the study of the behavior of slow neutrons, has been developed by Breit and Wigner,² who have considered the case that the energy levels of the compound nucleus lie far apart compared to their breadth, and have applied the familiar quantum mechanical dispersion formulae to this problem. For sufficiently heavy nuclei and sufficiently high excitation energies the intermediate nucleus will however no longer have well-defined energy levels; instead the states of the system, and even the states of a particular kind, e.g., of a given angular momentum, will form a continuum. One

might hope that the disappearance of any characteristic level structure would lead to a simplification in the description of the probabilities of nuclear disintegration, and that the only quantities which then determine these probabilities would be the rates at which a wave packet representing the intermediate nucleus comes apart into the various possible residual nuclei and emitted particles.

This expectation is not however in agreement with the result given by a simple application of the principle of detailed balancing to the processes of formation and disintegration of the compound system. Thus we may divide the states of the compound nucleus into sets (i) each of which is characterized by decay constants which in the limit of close lying levels may be regarded as slowly varying functions of the energy, and by a density of levels $1/s^i$ per unit energy. Then we obtain a relation between the cross section σ_c for the capture of an incident particle and the decay constants Γ_{a0^i}/\hbar for the reemission of this particle with its original energy :

$$\sigma_c = (\lambda^2/2w_A) \sum_i \Gamma_{a0^i}/s^i, \quad (1)$$

¹ N. Bohr, Science, to be published.

² Breit and Wigner, Phys. Rev. 49, 519 (1936).

where $\lambda = h/p$ is the wave-length of the incident particle, and w_A is the combined statistical weight of the initial states of incident particle and bombarded nucleus. If then Γ_k^i/\hbar is the rate of emission of a particle of type k , Γ^i/\hbar is the total decay constant of a state (i), then the cross sections for transmutation are just

$$\sigma_k = \frac{\lambda^2}{2w_A} \sum_i \frac{\Gamma_{a0^i} \Gamma_k^i}{s^i \Gamma^i}. \quad (2)$$

This formula can also be brought into connection with the results derived by Breit and Wigner for the case of well separated levels ($\Gamma^i \ll s^i$):

$$\sigma_k(E) = \frac{\lambda^2}{4\pi w_A} \sum_i \frac{\Gamma_{a0^i} \Gamma_k^i}{(E - E_p^i)^2 + \frac{1}{4}(\Gamma^i)^2}, \quad E \sim E_p^i. \quad (3)$$

Here E is the total energy of the system, and E_p^i the energy of one of its quasi-stationary states. As Bethe and Placzek³ have pointed out, from (3) we can obtain a cross section, averaged over a range of energy large compared to spacing of the levels, which agrees with (2). One might thus suppose that although for the derivation of (3) the condition $\Gamma^i \ll s^i$ is essential, no such restriction limited the validity of (2).

Nevertheless, if one formulates the problem for the case $\Gamma^i \gg s^i$ in terms of the same dispersion-theoretic formalism as leads in the other limiting case to (3), and assumes as before that the collision may be treated as a resonance effect, one is led, not to (2), but to the radically different

$$\sigma_k = \frac{\lambda^2}{\pi w_A} \sum_i \frac{\Gamma_{a0^i} \Gamma_k^i}{(\Gamma^i)^2}, \quad (4)$$

where the summation \sum_i is to be taken over all sets of noncombining states of the compound nucleus, and where, as before, the Γ 's for each set are supposed to vary slowly with energy. Since (4) differs from (2) by the substitution for $1/s^i$ of $2/(\pi\Gamma^i)$, and since $\Gamma^i \gg s^i$, the cross sections given by (4) are smaller than those given by (2). As we shall see in III, the formal reason for this, and the reason for the failure of the statistical argument leading to (2), is that the "combining" states of the compound nucleus lying within a line breadth of each other do not

at all act independently, and that strong destructive interference is involved in the probability of their excitation [cf. (12)]. The coherence of phases implied by this interference is itself a consequence of the assumption that the processes involved in the collision may be adequately described in terms of resonance between states representing the incident particle (which may be elastically scattered at the surface of the nucleus), and other states of the compound system which have a very long life ($\sim \hbar/\Gamma^i$). This in turn implies that it is not necessary to include in the description those wave packets built up from the long lived states of the compound system, which represent short lived compound nuclei, and which correspond physically, on the one hand, to "surface effect" inelastic scattering and transmutation, and on the other to the possibility of forming the intermediate nucleus by a sequence of processes involving compound systems of increasingly long life and more complete energy degradation.

In the problem of the radiative capture of slow neutrons to which the Breit-Wigner formula was first applied, the characteristic energy dependence of the observed cross sections itself indicates the appropriateness of describing the collision as a resonance effect. But in the other limiting case, where $\Gamma^i \gg s^i$, neither (2) nor (4) may be applied without a careful examination of the physical problem. Since Γ^i increases, and s^i decreases, with the excitation energy, (2) and (4) differ more and more as the energy of the bombarding particle increases. It is in any case clear that the factor Γ_{a0^i}/Γ^i which occurs in (4) will fall off rapidly with energy because of the increasing improbability of a complete concentration of the excitation energy, and for sufficiently high energies the description of the collision as a resonance effect must in general be completely inappropriate.

A striking illustration of this is afforded by the impacts of quite fast neutrons on nuclei. In this case, as one may see from the simple mechanical model discussed by Bohr,¹ it will be extremely unlikely that the energy of the incident neutron will at once be divided among all the nuclear particles; rather the normal course of events will involve an inelastic impact near the surface of the nucleus, in some cases leading merely to the

³ Bethe and Placzek, Phys. Rev. 51, 450 (1937).

ejection of a particle with reduced energy, but quite often followed by a series of further impacts which lead ultimately to the complete degradation of the energy and the formation of a compound system of very long life. Such a situation cannot be formally described in terms of a simple resonance effect, nor can the formation of the compound system be treated without considering the wave packets which represent the surface disturbances and short lived intermediate states. On the other hand just the complication of the mechanisms actually involved in the formation of the compound nucleus may offer some justification, in this case, for the assumption of random phases⁴ involved in the derivation of (2).

A still more striking case of the inadequacy of (4) to describe transmutations we find in reactions initiated by bombardment with high energy deuterons, where, in spite of the fact that the emission of a deuteron of high energy from a compound nucleus must be extremely rare, the cross section for deuteron induced transmutations can be of the order of magnitude of the area of the nucleus. It is clear that here in the course of the formation of the compound nucleus, the original coupling between proton and neutron will be completely dissolved, and that the formation of the compound nucleus, whether it involves the capture of proton or neutron or both, may surely not be considered as a single process.

As an example of a problem to which we may make a tentative application of (4), we may consider the nuclear photoeffect in complex nuclei, for γ -ray energies high enough to make $\Gamma^i \gg s^i$, and yet low enough so that the absorption of the radiation can be treated as a resonance effect. That such a treatment can remain valid for higher excitation energies for γ -rays than for neutrons depends upon the fact that the interaction between a γ -ray and a nuclear particle is smaller in order of magnitude than that between the particles themselves: whereas the waves representing incident neutrons of high energy will be very rapidly damped out at the surface of the nucleus, those representing γ -rays will be practically undamped, and therefore far more

effective in exciting oscillations of the nucleus as a whole. Apart from a necessarily rough estimate of the contribution of the nonresonance effects, to which we shall refer again in II, and which shows that it may well be smaller than that given by (4), there is some experimental evidence in favor of the applicability of (4) to this problem, in that the smallness of the ratio of the effects observed for light nuclei (N^{14} , O^{16}) to those observed for nuclei of intermediate atomic weight is most easily interpreted in terms of the far shorter lifetime of the lighter compound nuclei.

II

It is, of course, not possible at present to calculate the Γ 's on the basis of any complete nuclear theory; and, particularly in those problems to which classical arguments cannot be simply applied, one must resort to the transmutation experiments themselves, and may hope to use (4) and (2) in some cases to correlate the values of the Γ 's so obtained. It is from this point of view that we shall discuss the photo-disintegration of nuclei by high energy γ -rays. For nuclei of intermediate weight we would expect that the compound nucleus formed by absorption of the γ -ray will ordinarily dissipate its energy by the emission of neutrons, since the emission of a charged particle would require a much greater concentration of energy in the escaping particle than is required by a neutron, because of the necessity of its clearing the Coulomb barrier. This argument may not however be applied to those nuclei, relatively common for atomic weight below 20 and above 150, for which the emission of a charged particle is energetically far more favorable than that of a neutron.

Experiments on the capture of thermal energy neutrons (nuclear excitation energy ~ 8 Mev) give neutron widths $\Gamma_n \sim 10^{-4}v$. As the excitation energy is increased, the neutron width will at first be proportional to the velocity of the neutron; hence for an energy a million volts higher we would estimate $\Gamma_n \sim 1v$. For still larger excitation energy the neutron width will increase very rapidly, because of the rapidly increasing number of probable modes of disintegration. For several million volts additional excitation energy

⁴ Compare the discussion of W. Pauli, Sommerfeld Festschrift, p. 30, 1928.

we would therefore expect Γ_n of the order of some tens or hundreds of volts. The spacing between levels is between 10 and 100 v for 8 Mev excitation energy, decreases rapidly with increasing energy, and should become less than Γ_n for several million volts additional energy. Thus when a nucleus is raised by absorption of a γ -ray to an excitation energy greater than this, the level breadth will surely be wider than the spacing between levels, and we may try to use (4) to discuss the problem. Since in all probability $\Gamma \sim \Gamma_n \gg \Gamma_\gamma$, the cross section for neutron emission is

$$\sigma_n = (\lambda^2/w_A\pi) \sum_i \Gamma_{\gamma_0}^i / \Gamma_n^i.$$

We may expect that the number of noncombining sets (i) which contribute appreciably would be comparable with w_A , and shall thus write

$$\sigma_n \sim (\lambda^2/\pi) \Gamma_{\gamma_0} / \Gamma_n, \quad (5)$$

where here the Γ 's may be regarded as appropriate averages over the contributing noncombining sets (i).

The photoeffects produced in many nuclei by the 17 Mev γ -ray of $\text{Li}^7 + \text{H}^1$ have been studied by Bothe and Gentner,⁵ who do in fact find that the typical reaction involves the ejection of a neutron. Their estimate, $\sigma_n \sim 10^{-27} \text{ cm}^2$, gives at once, since $\lambda^2/\pi \sim 10^{-23} \text{ cm}^2$,

$$\Gamma_{\gamma_0} / \Gamma_n \sim 10^{-4}.$$

If we combine with this the estimate of some 100 v for Γ_n at these energies, we find that Γ_{γ_0} must be of the order of 1/100 of a volt, about a tenth of the total radiative breadth of the resonances found in slow neutron capture. In fact one might expect that Γ_{γ_0} would not change very much in going from excitations of 8 to 17 Mev; for, on the one hand, it will increase with a high power (probably the fifth power characteristic of electric quadrupole and magnetic dipole radiation) of the frequency; on the other, it will decrease exponentially very roughly with the square root of the energy because of the smaller probability of finding all the excitation energy in a single mode of high frequency.⁶ Thus if one supposes that the electric moments asso-

ciated with these oscillations of varying frequency are of the same type and order of magnitude at 8 and at 17 Mev, the variation of the two factors on which Γ_{γ_0} depends will tend to cancel. We should then expect that in this range σ_n should increase with decreasing γ -ray energy, and continue to increase until Γ_n becomes equal to the spacing between levels. At this point (γ -ray energy ~ 10 –12 Mev) we should have $\Gamma_{\gamma_0} / \Gamma_n \sim 10^{-2}$ – 10^{-3} , and a cross section, σ_n , between 10^{-25} and 10^{-26} cm^2 . For still lower energies (5) is no longer valid; we must then, as pointed out by Bethe and Placzek, apply (2), i.e., in (5) replace Γ_n by $(2/\pi)s$. In this range the cross section will decrease with decreasing energy, as s increases.

It would thus be interesting to see if such an increase in yield occurs when photodisintegrations are produced by γ -rays of lower energy. Two points must, however, be kept in mind. In the first place there are in the Li spectrum⁷ γ -rays of about 14 Mev which may contribute as much to the photoeffect as the 17 Mev line. Moreover, since the yields from 10–12 Mev radiation may be much larger than those at the higher energies, the presence of any appreciable contamination of such degraded radiation in the experiments we have quoted would render their interpretation uncertain, and the maximum cross sections we suggest might be much too large.

In the second place, our discussion has been based on the use of (4); and we know that the validity of this is conditioned by the possibility of disregarding short-lived intermediate states and their contribution to the formation of the compound nucleus. It is clear that as the energy of the γ -ray is indefinitely increased, processes of absorption involving the dipole moment associated with the acceleration of a single particle, will be essential even for those impacts in which a long lived compound nucleus is ultimately formed. To estimate the order of magnitude of the relative contribution of these processes compared to the resonance effects described by (4), one may in (5) replace Γ_{γ_0}/\hbar by the radiative transition probability to the normal state for a wave packet representing a concentration of all

⁵ Bothe and Gentner, *Naturwiss.* **25**, 90, 126, 191 (1937).

⁶ A discussion of these questions is to be published by Bohr and Kalckar.

⁷ Delsasso, Fowler and Lauritsen, *Phys. Rev.* **51**, 391 (1937).

the excitation energy in one particle, and further replace \hbar/Γ_n by the nuclear collision time, and must include a factor to take into account the fact that several wave packets of this type can take part in the absorption process. It seems difficult to obtain a good enough evaluation of the quantities involved to tell us for what energies (4) will become inapplicable. As already pointed out however, the fact that the photoelectric yields for 17 Mev radiation from O^{16} and N^{14} are only about one percent of those from heavier nuclei⁸ is a strong indication that resonance effects predominate at these energies, since the lifetime of an intermediate state of a light nucleus is so much smaller than that of a heavy nucleus with the same excitation energy that on the basis of (4), and even allowing for a considerable increase in $\Gamma_{\gamma 0}$, one would anticipate much smaller photoelectric yields.

III

We want now to look more closely at the formal derivation of the result (4), and at the relation of our treatment to the more familiar application of dispersion formulae to nuclear problems.

According to Bohr's picture the intermediate nucleus lasts for a time long compared to the mean time between collisions of particles in the nucleus. In attempting to take this fact into account in formulating a dispersion theoretic treatment of collision problems, certain simplifying assumptions must be introduced, which state that the incident particle may only be either elastically scattered at the surface of the nucleus, or captured to form a compound system of long life which subsequently disintegrates. This can be so only when the short-lived compound systems, which are characteristic of surface effects, are not involved in the inelastic scattering or transmutations. The formal consequence of this simplification is that we can then describe the collision in terms of resonance between two quite distinct types of states, each of which may be supposed to give an approximate description of a stationary state of the whole system. One of these sets, ψ_r , corresponds

just to discrete excited states of the compound nuclei; the other set, ψ_s , represents asymptotically a definite residual nucleus in a definite state, and definite free particles of given energies. In setting up these states, we may so choose the ψ_r and their energies E_r , that the coupling between the various r states is as far as possible eliminated, and the ψ_s so that we may neglect all terms in the Hamiltonian directly coupling the s states with each other. The long life of the compound nucleus now means that the coupling between the r and s states is so small that the decay time of the r states is very long compared to the nuclear collision time: the condition that in the description of the collision process short-lived compound nuclei do not appear means that the coupling between r and s states is effective only near resonance, and that we need not consider wave packets built up from the ψ_r whose lifetime is much smaller than that of the states ψ_r themselves.

If we now call the matrix elements of the total Hamiltonian $H_{rr'}$, H_{rs} , $H_{ss'}$, our conditions on the ψ_s mean that $H_{ss'}$ may be treated as a diagonal matrix:

$$H_{ss'} = E_s \delta(s - s').$$

The optimal elimination of the coupling between the r states means, that not $H_{rr'}$, but $H_{rr'}$ corrected for the "line shifts" due to the coupling with the s states, is diagonal:

$$H_{rr'} + P \int ds H_{rs} H_{sr'} / (E - E_s) = E_r \delta_{rr'}, \quad (6)$$

where E is the total energy of the system. We now try to find a wave function of the form

$$\psi = \psi_0 + \sum_r c_r \psi_r + \int ds c_s \psi_s,$$

where ψ_0 is that one of the ψ_s which represents asymptotically the bombarding particle and the bombarded nucleus, and is normalized so that the incident flux is unity. The wave equation for the c 's is then

$$E c_r = \sum_{r'} H_{rr'} c_{r'} + \int ds H_{rs} c_s + H_{r0}, \quad (7)$$

$$(E - E_s) c_s = \sum_r H_{sr} c_r. \quad (8)$$

⁸ We are indebted to Dr. Cockcroft for telling us of these experiments carried out by Goldhaber and his collaborators.

From this, substituting c_s from (8) in (7), and using (6),

$$\chi_r = H_{r0} - \frac{i}{2} \sum_{r' \neq r} \frac{\Gamma_{rr'} \chi_{r'}}{s_{r'} E - E_{r'} + \frac{1}{2} i \Gamma_{r'r'}}, \quad (9)$$

where $\Gamma_{rr'} = 2\pi \sum_s H_{rs} H_{sr'}$, and the sum \sum_s is taken over all states with $E_s = E$, and where we have introduced

$$\chi_r = (E - E_r + \frac{1}{2} i \Gamma_{rr}) c_r,$$

which varies smoothly with E .

The assumption that no short-lived intermediate nuclei contribute now means that we may neglect the contribution to the sum in (9) of all states r' for which $|E_{r'} - E| \gg \Gamma_{r'r'}$. When the spacing of the levels, s_r , is large compared to Γ_{rr} we then get at once

$$\chi_r = H_{r0} \quad \text{when} \quad E_r \sim E. \quad (10)$$

In the other limiting case, with $s_r \ll \Gamma_{rr}$, many terms in the sum contribute. We may then suppose that the Γ 's vary smoothly from level to level. There may of course be various sets of intermediate states, for example states of given angular momentum, with quite different sets of Γ 's; but in general one will expect this only when the different sets of levels belong to different representations of a group of transformations which leave the Hamiltonian approximately invariant, and in this case each such set (i) may be considered separately; there will be no interference between different sets and their contributions to the cross section will be simply additive, so that from now on we shall consider only a single such set. The summation in (9) may thus be replaced by an integral

$$\chi_r = H_{r0} - \frac{i}{2} \int \frac{dE_{r'}}{s_{r'}} \frac{\Gamma_{rr'} \chi_{r'}}{(E - E_{r'} + \frac{1}{2} i \Gamma_{r'r'})}. \quad (11)$$

If then we neglect the contribution of levels $|E_{r'} - E| \gg \Gamma_{r'r'}$, and the variation over the line breadth of the matrix elements of H , we may replace the integral by its residue. The solution of (11) is then

$$\chi_r = H_{r0} - \frac{\pi \Gamma_{r\rho} H_{\rho 0}}{2s_\rho + \pi \Gamma} \sim \frac{2s_\rho H_{r0}}{\pi \Gamma} \quad (12)$$

except when $|E_r - E| \gg \Gamma$, and where $E_\rho \sim E$ and $\Gamma = \Gamma_{\rho\rho}$. Because of the assumed asymptotic form of ψ_s , one sees from (8) that the cross section for a process of type k involving the emission of a given particle and leaving the nucleus in a given state is

$$\sigma_k = (2\pi/\hbar) \sum_{s=k} \left| \sum_r H_{sr} c_r \right|^2, \quad (13)$$

where $\sum_{s=k}$ is to be taken over all states s , with $E_s = E$, and leading to a disintegration of type k . From (10), (12) and (13), and treating the sum over r as before, we then get

$$\begin{aligned} \sigma_k &= \frac{8\pi}{\hbar} \frac{|H_{\rho 0}|^2 \sum_{s=k} |H_{s\rho}|^2}{\Gamma^2}, & \Gamma \gg s_\rho \\ & & E_\rho \sim E. \quad (14) \\ &= \frac{2\pi}{\hbar} \frac{|H_{\rho 0}|^2 \sum_{s=k} |H_{s\rho}|^2}{(E - E_\rho)^2 + \frac{1}{4} \Gamma^2}, & \Gamma \ll s_\rho \end{aligned}$$

Because of the asymptotic form of ψ_s and ψ_0 , the matrix elements occurring in (14) are connected with the decay constant Γ_k for the disintegration of type k , and the decay constant Γ_{a0} for the reemission of the incident particle with its initial energy, by

$$\Gamma_k = 2\pi \sum_{s=k} |H_{s\rho}|^2, \quad \Gamma_{a0} = (4\pi w_A / \hbar \lambda^2) |H_{\rho 0}|^2.$$

We thus have

$$\begin{aligned} \sigma_k &= \frac{\lambda^2}{\pi w_A} \frac{\Gamma_{a0} \Gamma_k}{\Gamma^2}, & \Gamma \gg s_\rho \\ &= \frac{\lambda^2}{4\pi w_A} \frac{\Gamma_{a0} \Gamma_k}{(E - E_\rho)^2 + \frac{1}{4} \Gamma^2}, & \Gamma \ll s_\rho, \quad E_\rho \sim E. \end{aligned}$$

By summing over all sets of intermediate states (i) we get from this (3) and (4). It is clearly not possible by formal arguments alone to decide whether, in a given problem, the incisive conditions necessary for the validity of (3) or (4) are really fulfilled. For this, as in the case of the nuclear photoeffect, a detailed discussion of the physical problem is in general essential.