Ionization Yield of Radiations. II. The Fluctuations of the Number of Ions

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The ionization produced by individual fast charged particles is frequently used as a measure of their initial energy; fluctuation effects set a theoretical limit to the accuracy of this method. Formulas are derived here to estimate the statistical fluctuations of the number of ions produced by constant amounts of radiation energy. The variance of the number of ionizations is found to be two or three times smaller than if this number were governed by a Poisson distribution. An improved understanding is gained of the statistical treatment of fluctuation phenomena.

1. INTRODUCTION

THE number of ion pairs J produced in a volume of gas following the absorption of ionizing radiation is closely proportional to the amount V of energy absorbed. The ratio V/J is generally of the order of magnitude of 30 to 35 ev; it depends very little on the quality of the ionizing radiation and comparatively little on the nature of the gas. This question has been discussed in a recent paper,[1] which will be referred to as (I).

The constancy of the ratio V/J is frequently relied upon to obtain an experimental estimate of V by measuring J and multiplying it by a factor ϵ. This factor is equal to the ratio of the mean values of V and J obtained from a large number of experiments carried out under comparable conditions.

Even when all pertinent physical factors, including V, are kept constant, the number J is subject to statistical fluctuations. Knowledge of the extent of these fluctuations, which set an upper limit to the accuracy of experimental techniques, is of interest, e.g., when the ionization produced by individual particles serves as a measure of their initial energy; this was kindly pointed out to the author by Dr. R. G. Sachs. A rough theoretical estimate of this effect is derived in the present paper. Specifically, it is proposed to estimate the variance of J under the condition that the energy loss V has a fixed value V_0, i.e., to calculate the expected value of $(J-J_0)$,[2] where $J_0 = V_0/\epsilon$ is the expected value of J under the stated condition.

2. STATISTICAL TREATMENT

The production of a measurable number of ionizations involves a large number of elementary processes. Hence, the first objective is to express the variance of J in terms of quantities pertaining to individual elementary processes. For example, one may consider as an elementary process the impact of a fast charged particle against a gas molecule.

The probability that J assumes any given value after a *fixed number of impacts* has taken place, or after a particle has covered a *fixed length of track*, can be easily obtained; under these conditions the total *energy loss* V would be subject to *fluctuations*, just as is J. We are interested, however, in the opposite case of a *fixed energy loss*, $V = V_0$, with *fluctuating length of track* and *number of impacts*; in this case the probability distribution of J requires further investigation.

This problem is analogous to that of the straggling in range of α-particles[2] which is solved through the following consideration: If a particle happens to lose an amount of energy V instead of the expected amount V_0 along a length of track l, it is expected to lose the amount V_0 along a length of $l+\Delta l$, where $\Delta l = l(V_0 - V)/V_0$. The fluctuation of energy loss along Δl is disregarded here, but this fluctuation tends to average out since Δl may be positive or negative. Similarly, if the particle happens to have produced J ionizations along l while losing the energy V, it will produce $\Delta J = (V_0 - V)/\epsilon$ ionizations along Δl. Thus the eventual fluctuation of J for fixed V_0, namely, $(J + \Delta J) - J_0$ turns out to be equal, at least approximately, to the fluctuation $J - V/\epsilon$

[1] U. Fano, Phys. Rev. **70**, 44 (1946).

[2] N. Bohr, Phil. Mag. **30**, 581 (1915).

for fixed l; the probability of the latter can be easily calculated. The essential point is that the quantity $J - V/\epsilon$:

(a) Coincides with $J - J_0$ when $V = V_0$.

(b) Has the *mean value zero* and hence its variation over short sections of track can be disregarded so that its mean square should have, at least approximately, the same value for fixed l (or for fixed number of impacts) as for fixed $V = V_0$.

If n_p and E_p are the number of ionizations and the energy loss resulting from the pth impact within a group of N impacts occurring under the same conditions, then the mean values $\bar{n}_p = \bar{n}$ and $\bar{E}_p = \bar{E}$ are independent of p and:

$$J = \sum_{p=1}^{N} n_p, \quad V = \sum_{p=1}^{N} E_p, \quad \epsilon = \bar{E}/\bar{n}. \tag{1}$$

Since the results of successive impacts are independent of one another, the mean of $(J - V/\epsilon)^2 = [\sum_p (n_p - E_p/\epsilon)]^2$ is obtained by adding the mean of $(n_p - E_p/\epsilon)^2$ in each impact, and this is also independent of p. Hence, for a fixed $N = V_0/\bar{E} = J_0/\bar{n}$, the desired variance is given by:

$$\langle (J - V/\epsilon)^2 \rangle_{Av} = N \langle (n - E/\epsilon)^2 \rangle_{Av} = F J_0, \tag{2}$$

where:

$$F = \langle (n - E/\epsilon)^2 \rangle_{Av}/\bar{n}. \tag{3}$$

Independent mathematical investigation[3] of a quantity

$$X = \sum_{p=1}^{N} x_p,$$

where the x_p's are independent random variables governed by the same probability distribution *and* $\bar{x}_p = 0$, has actually shown that $\bar{X} = 0$ and $\langle X^2 \rangle_{Av} = \bar{N} \langle x^2 \rangle_{Av}$ *even though N is not fixed but subject to fluctuations*, provided only that $\bar{N}$ and $\langle N^2 \rangle_{Av}$ are finite. This confirms that the variance of J for $V = V_0$ is given by Eq. (2) *exactly*, even when the number of elementary processes involved is not large. It also shows that Bohr's formula for the straggling in range has a similarly large range of application and can be derived directly by taking the variance of the quantity $l - V/B$ (B = stopping power) whose mean is zero.

[3] M. A. Gershick and D. Blackwell, Ann. Math. Statistics (to be published).

3. TREATMENT OF SECONDARY IONIZATION

The method introduced in (I), Section 4, is to consider every inelastic impact undergone by gas molecules as an elementary process, independently of whether the impinging particle is fast or slow, a "primary" or a "secondary."[4] The approximation involved is to assume that the ratios among the cross sections for different types of impact are independent of the speed and nature of the impinging particle. This assumption is not closely fulfilled but offers a convenient basis for discussion.

Call s_j the cross section for an impact which sends the molecule to its jth excited state; E_j is the energy absorbed by the molecule inclusive of all its electrons. Following the notation of (I), the state j is classified as:

(e) if $E_j < I$, where I is the first ionization potential (simple excitation);

($i1$) if $I \leqslant E_j < 2I$ (an electron is ejected which is incapable of further ionization);

($i2$) if $E_j \geqslant 2I$ (an electron is ejected with energy $E_j - I$, and is capable of further ionization).

The treatment in the preceding section can now be applied considering that:

(a) The number of ionizations per impact is $n_j = 0$ in case (e), $n_j = 1$ in cases ($i1$) and ($i2$).

(b) The energy actually lost by the ionizing radiation in case ($i2$) is only I, since $E_j - I$ can be utilized again.

Then:

$$F = [\Sigma_j^{(e)} s_j (E_j/\epsilon)^2 + \Sigma_j^{(i1)} s_j (1 - E_j/\epsilon)^2$$
$$+ \Sigma_j^{(i2)} s_j (1 - I/\epsilon)^2]/\Sigma_j^{(i1, i2)} s_j. \tag{3'}$$

4. DISCUSSION

For purposes of comparison it is pointed out that if the number of ionizations were governed by a Poisson distribution, then $\langle (J - J_0)^2 \rangle_{Av} = J_0$, that is $F = 1$.

Formula (3') is easier to evaluate numerically than (3). Experimental or theoretical data on the ratios among the s_j's can be used. The nature of the formula is such that the result is rather insensitive to moderate errors in the s_j's.

[4] The method might be extended to cover the impacts of ionizing (primary or secondary) photons against molecules. Another case which is not covered by the present theory is the double ionization caused, for example, by removal of an internal electron followed by an Auger process. These phenomena appear to involve only a small fraction of the ionized molecules.

In the case of atomic H, available theoretical numerical data[5] have been used with the result:

$$F \sim 0.46 \quad \text{for 100 kv impinging electrons}$$
$$F \sim 0.42 \quad \text{for 1 kv impinging electrons.} \quad (4)$$

The calculation has been repeated using the same simplified theory of impacts as in (I), Section 4, namely, by taking (with notation similar to that in (I)):

$$s_j = s_j^{(g)} + s_j^{(h)}, \quad s_j^{(g)} = (kx_{j0}^2 \log r)/\text{Ry},$$
$$s_j^{(h)} = 0 \text{ for } E_j < I, \quad s_j^{(h)} = k/E_j^2 \text{ for } E_j \geqslant I \quad (5)$$

where x_{j0} is the dipole matrix element for the transition from the normal to the jth state and $r = 2mv^2/I$. Hence:

$$F = \{\Sigma_j^{(e)}(x_{0j}^2/x_i^2)(E_j/\epsilon)^2$$
$$+ \Sigma_j^{(i1)}(x_{0j}^2/x_i^2)(1 - E_{0j}/\epsilon)^2$$
$$+ (x_{i2}^2/x_i^2)(1 - I/\epsilon)^2 + (\epsilon_1/I \log r)$$
$$\times [1 - (1 + 2 \log 2)I/\epsilon + 3I^2/2\epsilon^2]\}/$$
$$[1 + (\epsilon_1/I \log r)] \quad (3'')$$

with

$$x_i^2 = x_{i1}^2 + x_{i2}^2 = \Sigma_j^{(i1, i2)}x_{0j}^2, \quad \epsilon_1 = \text{Ry}/x_i^2,$$

Ry = Rydberg's energy. This formula holds for any substance; experimental or theoretical values of x_{0j} can be used. The results for H are:

$$F \sim 0.47 \quad \text{for 100 kv impinging electrons}$$
$$F \sim 0.43 \quad \text{for 1 kv impinging electrons.} \quad (4')$$

The agreement between (4) and (4'), even though it may be partly accidental, parallels the agreement obtained in (I) under the same circumstances and encourages the application of the simplified theory of impacts.

There remains to be discussed the effect of the assumption that the ratios among the s_j's are independent of the speed and nature of the impinging particles. This assumption was taken as a basis in (I), Section 4, as well as in Section 3 of this paper. The correct procedure in both cases should have been:

(a) To classify all impacts undergone by individual molecules according to the nature and speed of the radiation impinging on that molecule (e.g., "secondary electron of 47 ev");

[5] Geiger-Scheel, *Handbuch der Physik* (Julius Springer, Berlin, 1933), Vol. 24/1, pp. 517, 519.

(b) To calculate ϵ and F (as defined by (1) and (3), the former being equivalent to (3) of (I)) for each "kind" of impinging radiation;

(c) To calculate the final average values of ϵ and F, giving to each "kind" of impinging radiation a weight proportional to the number of ionizations that are expected to be produced *directly* by it.

From this standpoint it appears that the values of ϵ and F calculated in (I) and above in this paper should be intended, strictly speaking, to apply *only* to the ionization produced *directly* by electrons of the stated energy *excluding* that produced by secondaries or by the same electrons after considerable slowing down. In practice, however, since ϵ and F do not depend very greatly upon the kind of impinging radiation, the corrections required are not too large.

It is of particular interest to evaluate ϵ and F for the slow secondary electrons which produce a large fraction of the total ionization. To do this properly, experimental data on impact cross sections might be used, as it is difficult to obtain theoretical data owing to the lack of suitable approximation methods. However, the theory applicable to fast electrons indicates that the frequency of excitations decreases with decreasing energy as compared to that of ionizations. This has the effect of decreasing the theoretical estimate of F as well as that of ϵ. A crude estimate may be hazarded by extrapolating the results of the simplified theory down to $\log r = 2$, which would correspond to slow electrons not quite able to produce further ionizations. This yields (still for atomic H) $F \sim 0.33$. Since approximately equal numbers of ionizations are produced in H by primary and by secondary electrons, the over-all value for H might be expected to be $F \sim 0.4$. If the same method of crude estimation is applied to correct the second approximation value $\epsilon_2 = 36$ ev obtained in (I) for H, a new estimate ~ 32 ev is obtained.

No attempt has been made to calculate F for substances other than H, the purpose of the paper being simply to estimate its order of magnitude. It appears, in view of the considerations presented in (I), that the influence of the factor s_j in the sum $\Sigma_j^{(e)}$ in (3') or (3'') should decrease sharply as I increases, the factor $(E_j/\epsilon)^2$ in the same summation should, however, counteract this effect strongly. The factors $(1 - E_j/\epsilon)^2$ and

$(1-I/\epsilon)^2$ in the remaining summations should act to reduce the value of F as I increases.

In conclusion, it seems rather likely that F should generally be of the order of magnitude of $\frac{1}{3}$ to $\frac{1}{2}$.[6]

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[6] The ionizing action of comparatively energy-rich but slow ions, of velocity close to that of molecular valence electrons, constitutes an exceptional case for the determination of both ϵ and F; ϵ may become much larger than 30–35 ev, and F may approach 1.

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Higher Angular Momenta and Long Range Interaction in Resonance Reactions

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The results of two preceding articles on the energy dependence of the reaction and scattering cross sections are generalized. As before, the configuration space is divided into two parts. In the internal region or reaction zone there is no restriction on the type of interaction between the particles. In the external region, on the other hand, the interaction takes place between the colliding or separating particles without changing their structure (wave function). The present article deals with a more general situation than the preceding ones, first by allowing for an interaction, although only of a restricted type, outside the reaction zone. The most common type of interaction which plays a role outside the reaction zone is the electrostatic interaction of the colliding or separating particles. In addition, the present article does not restrict the angular momentum of the particles to zero, but permits the treatment of arbitrary angular momenta. The cross sections are expressed in well-known fashion in terms of the collision matrix $\mathfrak{U}$ which is, of course, independent of the size of the internal region. $\mathfrak{U}$, in its turn, is expressed by the $\mathfrak{R}$ matrix ((35) and (38)) and the quantities ω, $\mathfrak{B}$, and $\mathfrak{C}$ describing the interaction in the external region. $\mathfrak{R}$ as function of energy is given by (24), and a number of properties of this function are enumerated. None of the quantities $\mathfrak{R}$, ω, $\mathfrak{B}$, and $\mathfrak{C}$ are strictly independent of the size of the internal region, although the combination (38), i.e., the collision matrix, of course, is.

I. INTRODUCTION AND SUMMARY

THE present treatment of the representation of the scattering and reaction cross sections differs from that given in two previous papers[1a, b] in that it divides the problem of finding a stationary solution of the quantum-mechanical equations into two steps. The first step is taken in Section III; Section II is devoted to the introduction of several definitions and a few mathematical preliminaries. In Section III an expression is obtained for the value which the wave function assumes on the surface S separating the internal and external regions,[2] in terms of the normal derivative of the wave function on that surface. The essential content of this section may be described as follows. A complete set of orthonormal functions X_λ is defined in the internal region by means of an Hermitean boundary value problem. The X_λ are those solutions of the wave equation which satisfy the boundary condition that their normal derivatives vanish at the boundary S of the internal region. If an arbitrary stationary-state wave function, φ, associated with the energy E, is expanded in terms of the X_λ the expansion coefficients are found to be $\gamma_\lambda/(E_\lambda - E)$ where $\gamma_\lambda = \int X_\lambda^*(\partial\varphi/\partial n)dS$. The E_λ is the characteristic value associated with X_λ, $\partial\varphi/\partial n$ denotes the normal derivative; the integral is to be extended over the boundary surface S. As a result, the value of φ is given by

$$\varphi = \sum_\lambda \frac{\gamma_\lambda}{E_\lambda - E} X_\lambda = \sum_\lambda \frac{\int X_\lambda^*(\partial\varphi/\partial n)dS}{E_\lambda - E} X_\lambda. \quad (1)$$

[1a] E. P. Wigner, Phys. Rev. **70**, 15 (1946).

[1b] E. P. Wigner, Phys. Rev. **70**, 606 (1946). Professors J. Schwinger and V. Weisskopf have kindly informed us that they have recently obtained results which closely parallel those of the present paper.

[2] Most of the notions (such as internal and external region, etc.) used in this paper were formulated by the writers in 1940 for a review article, the publication of which was postponed because of the war. It will appear shortly in another journal.