Statistical fluctuations in the yield of ionization due to protons or α particles

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An analytic method is presented for the calculation of various indices that characterize statistical fluctuations in ionization yields, specifically for the incidence of an ion such as a proton or α particle at a high speed. A general expression for the Fano factor for ion incidence is derived, and the relation to the Fano factor for electron incidence is clarified. For an illustration, the theory is applied to protons incident on argon. The resulting Fano factor for protons of energies between 0.1 and 2 MeV is nearly the same as that for electrons at the same speeds, and it is considerably lower than the values measured with α particles.

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I. INTRODUCTION

Statistical fluctuations in the total ionization caused by the absorption of ionizing radiation in matter were first discussed by Fano [1]. He showed that the variance D in the number of ions produced is given as

$$D = FN , \tag{1}$$

where N is the mean number of the ions and F is a constant, generally less than unity, for radiation of sufficiently high energies. The quantity F, called the Fano factor, represents the theoretical limit of precision in the energy determination through ionization measurements.

Knipp *et al.* [2] gave an analytic method for evaluating the statistical fluctuations in the ionization yield resulting from *the incidence of an electron* of a fixed energy. Rau, Inokuti, and Douthat [3] showed that the Fano factor and other indices characterizing the fluctuations can be expressed more transparently as an integral involving the electron degradation spectrum. An application of the method to molecular hydrogen was presented by Inokuti, Douthat, and Rau [4].

The method is now extended to treat *the incidence of an ion without electronic structure* such as a proton or an α particle. For such an event, the ionization is produced either directly by the incident ion or indirectly by secondary electrons. Therefore the yield and the fluctuations receive contributions from these two sources.

The present work was stimulated by recent results on argon, both experimental [5–7] and theoretical [8–10], which are shown in Table I. In summary, the Fano factor is about 0.15 for electron incidence and about 0.2 for α-particle incidence. As a step toward understanding the difference between electron incidence and α-particle incidence, we developed the analysis described below.

II. THEORY

A. Basic equation for the probability distribution of ionization

Let us first consider the incidence of a simple ion, such as a proton, an alpha particle, or any charged particle that carries no electron, at a high speed. Let the medium be composed of a single species of molecules having a single ionization threshold I, for simplicity of discussion. (This assumption is nonessential; it can be readily removed without causing serious complications. We use the term molecule for the simplicity of discussion; when the medium is condensed matter, we refer to a structural unit appropriate for a specific discussion.)

TABLE I. Summary of current data on the Fano factor of argon.

Incident radiation	Energy	The Fano factor	Authors
		Experiment	
α particles	5.3 MeV	$0.20 \begin{cases} +0.01 \\ -0.02 \end{cases}$	Kase *et al.* [5]
α particles	5.68 MeV	0.19 ± 0.01	Alkhazov *et al.* [6]
Electrons	0.26 keV and 2.82 keV	0.14 ± 0.02	Neumann [7]
		Theory	
Electrons	≤ 2.5 keV	0.16	Alkhazov [8]
Electrons	≤ 5 keV	0.15	Grosswendt [9]
Electrons	≤ 2 keV	0.16	Kowari *et al.* [10]

Let $\tilde{P}(T,j)$ be the probability that the incidence of an ion of kinetic energy T produces precisely j ion pairs in the medium. (Throughout the present paper, we put a tilde over every symbol that refers to the ion as opposed to an electron.) Following Knipp *et al.* [2], we will write an equation for $\tilde{P}(T,j)$. To do so, we must start with cross sections for individual collisions of the ion with molecules. Several possibilities exist. First, the ion may collide with a molecule, transfer energy E to it, and ionize it, and cause the production of a secondary electron having kinetic energy $E-I$. Let the differential cross section for this ionization process be $d\tilde{\sigma}_i(T,E)/dE$. Second, the ion may collide with a molecule and transfer energy E_s to it, without causing ionization; most often, the molecule is excited to a discrete level allowing dissociation. Another kind of nonionizing collision leads to energy transfer to the translational, vibrational, or rotational motion of the molecule; such collisions are important at low particle energies [11,12]. Let the cross section for the energy transfer E_s be $\tilde{\sigma}_s(T)$. (We use the discrete index s again for simplicity of discussion. Energy transfer to the translational motion is certainly not discrete but depends continuously on the scattering angle.) We call the sum of all the cross sections the total cross section $\tilde{\sigma}_{\text{tot}}(T)$,

$$\tilde{\sigma}_{\text{tot}}(T)=\sum_s \tilde{\sigma}_s(T)+\tilde{\sigma}_i(T) , \tag{2}$$

where $\tilde{\sigma}_i(T)$ is the total ionization cross section,

$$\tilde{\sigma}_i(T)=\int dE \frac{d\tilde{\sigma}_i(T,E)}{dE} . \tag{3}$$

For simplicity we restrict our treatment to the incidence of an ion of sufficiently high energies so that most of the energy loss is due to electronic excitation or ionization [13] as opposed to nuclear motion of atoms or molecules in the medium. Therefore we neglect ionization by recoil ions, which is important at lower incident energies and has indeed been discussed by Lindhard and coworkers [11,12].

The equation for $\tilde{P}(T,j)$ is then

$$\tilde{\sigma}_{\text{tot}}(T)\tilde{P}(T,j)=\sum_s \tilde{\sigma}_s(T)\tilde{P}(T-E_s,j)+\int dE \frac{d\tilde{\sigma}_i(T,E)}{dE}\sum_{k=0}^{j-1}\tilde{P}(T-E,k)P(E-I,j-k-1) . \tag{4}$$

The right-hand side of Eq. (4) enumerates all possible contributions to $\tilde{\sigma}_{\text{tot}}(T)\tilde{P}(T,j)$ classified by alternative results of a collision of an ion at energy T. If the collision is nonionizing and results in energy loss E_s of the ion, the contribution is $\tilde{\sigma}_s(T)\tilde{P}(T-E_s,j)$. The sum of all such contributions is the first term. If the collision is ionizing and results in the energy loss E of the ion and in the production of a secondary electron of energy $E-I$, the ion (now with energy $T-E$) may lead to k ion pairs, and the secondary electron may lead to $j-k-1$ ion pairs. The contribution of this chain of events is

$$[d\tilde{\sigma}_i(T,E)/dE]\tilde{P}(T-E,k)P(E-I,j-k-1) .$$

The last factor has no tilde, because it refers to an electron. More precisely, $P(E-I,j-k-1)$ represents the probability that *a secondary electron* of kinetic energy $E-I$ produces $j-k-1$ ions. This quantity has been fully treated in Refs. [3,4]. We here regard it as known. The total of all such contributions is the second term on the right-hand side of Eq. (4).

The function satisfies not only Eq. (4) but also the condition that

$$\tilde{P}(T,j)=\delta_{j0} \tag{5}$$

for $T<I$, which means that no ion pair is produced by an ion at energies lower than the ionization threshold. Beginning with Eq. (5) and ascending in T, one solves Eq. (4) to determine $\tilde{P}(T,j)$. This procedure is straightforward in principle, and the result is unique.

The integral over E in the second term on the right-hand side of Eq. (3) or (4) extends over all possible values of E, i.e., $I<E<T$. However, the upper limit is in practice much smaller than T, about $2mv^2=4(m/M)T$, where v is the speed of the ion, M is the mass of the ion, and m is the electron mass. This limit implies when the binding energy of the electron to be struck out is negligible; energy transfer E greater than the above value indeed occurs upon inner-shell ionization but at a small probability. (See Sec. 2.2 of Inokuti [13] for a fuller discussion of this point.) In the following discussion, we will use the abbreviated notation $d\tilde{\sigma}_T(E)$ for $dE[d\tilde{\sigma}_i(T,E)/dE]$.

Equation (4) is effectively the same as Eq. (3.8) of Lindhard *et al.*, [12] who also gave an extensive discussion of the yield of ionization and other related quantities. The main contribution of Rau, Inokuti, and Douthat [3] and of the present work is to relate the treatment with the Spencer-Fano theory [14] of slowing-down spectra and thus to make the analytic and numerical procedure more effective. Recent generalizations of the Spencer-Fano theory, including time-dependent cases [15–17], show the effectiveness of our approach.

B. Moments

Let us consider the moment $\tilde{M}(T,\mu)$ of the probability distribution $\tilde{P}(T,j)$, viz.,

$$\tilde{M}(T,\mu)=\sum_{j=0}^{\infty} j^{\mu}\tilde{P}(T,j) . \tag{6}$$

According to Eq. (5), the moment is subject to the boundary condition that

$$\tilde{M}(T,\mu)=\delta_{\mu 0} \tag{7}$$

for $T<I$. Multiplying both sides of Eq. (4) by j^{μ} and summing the result over j gives the following equation for the moment:

$$\tilde{\sigma}_{\text{tot}}(T)\tilde{M}(T,\mu)=\sum_s \tilde{\sigma}_s(T)\tilde{M}(T-E_s,\mu)+\sum_{\nu=0}^{\mu}\sum_{\lambda=0}^{\nu}\begin{bmatrix}\mu\\ \nu\end{bmatrix}\begin{bmatrix}\nu\\ \lambda\end{bmatrix}\int d\tilde{\sigma}_T(E)\tilde{M}(T-E,\nu-\lambda)M(E-I,\lambda)\ , \tag{8}$$

where the last factor without tilde, $M(E-I,\lambda)$, represents the moment of the ionization probability distribution due to an electron of kinetic energy $E-I$,

$$M(E-I,\lambda)=\sum_{j=0}^{\infty} j^{\lambda}P(E-I,j)\ . \tag{9}$$

The derivation of Eq. (8) is similar to the treatment of Sec. IV of Rau, Inokuti and Douthat [3], concerning the electron incidence. In the present work, we presume that the electron problem has been solved; thus we treat $M(E-I,\lambda)$ as known.

Equation (8) is solvable starting with $\mu=0$ and ascending in μ. To see this clearly, one may recast Eq. (8) as

$$\tilde{\sigma}_{\text{tot}}(T)\tilde{M}(T,\mu)-\sum_s\tilde{\sigma}_s(T)\tilde{M}(T-E_s,\mu)-\int d\tilde{\sigma}_T(E)\tilde{M}(T-E,\mu)$$

$$=\sum_{\nu}{}'\sum_{\lambda}{}'\begin{bmatrix}\mu\\ \nu\end{bmatrix}\begin{bmatrix}\nu\\ \lambda\end{bmatrix}\int d\tilde{\sigma}_T(E)\tilde{M}(T-E,\nu-\lambda)M(E-I,\lambda)\ , \tag{10}$$

where $\sum'_{\nu}\sum'_{\lambda}$ denotes the summation as in Eq. (8), but the single term with $\nu=\mu$, $\lambda=0$ is subtracted. (This term appears as the last term on the left-hand side.)

The right-hand side of Eq. (10) contains the moments $\tilde{M}(T,\mu)$ of orders lower than μ and the electron moments $M(T,\mu)$, all of which are presumed to be known. Thus Eq. (10) is an inhomogeneous linear equation for $\tilde{M}(T,\mu)$. It is convenient to write the equation in the compact form

$$\tilde{\Omega}_T\tilde{M}(T,\mu)=\tilde{R}(T,\mu)\ . \tag{11}$$

Here $\tilde{\Omega}_T\tilde{M}(T,\mu)$ represents the left-hand side of Eq. (10) and $\tilde{R}(T,\mu)$ the right-hand side. We may call $\tilde{\Omega}_T$ the Fowler operator for the ion. It is similar to the Fowler operator Ω_T for the electron treated by Rau, Inokuti, and Douthat [3], but $\tilde{\Omega}_T\tilde{M}(T,\mu)$ lacks the secondary-electron term involving $M(E-I,\lambda)$, which is now incorporated into the inhomogeneous term $\tilde{R}(T,\mu)$.

Let us examine the first three moments. For $\mu=0$, we have $\tilde{R}(T,0)=0$. Starting with Eq. (7) and ascending in T, we obtain

$$\tilde{M}(T,0)=1 \tag{12}$$

for any T, which means that the total probability is conserved, as it should be. For $\mu=1$, we have

$$\tilde{R}(T,1)=\tilde{\sigma}_i(T)+\int d\tilde{\sigma}_T(E)M(E-I,1)\ . \tag{13}$$

Here $\tilde{M}(T,1)$ is the *mean* number of ionization, which we will denote simply by $\tilde{N}(T)$. Equation (11) with $\mu=1$ is the Fowler equation for the mean ionization yield due to the ion. The inhomogeneous term $\tilde{R}(T,1)$ consists of two terms, the first representing direct ionization by the ion and the second representing ionization by secondary electrons.

For $\mu=2$, we have

$$\tilde{R}(T,2)=\int d\tilde{\sigma}_T(E)[2\tilde{M}(T-I,1)M(E-I,1)+M(E-I,2)+2\tilde{M}(T-E,1)+2M(E-I,1)+1]\ . \tag{14}$$

For simplicity, we will write

$$\tilde{M}(T-E,1)=\tilde{N}(T-E)\ , \tag{15}$$

which means the mean number of ionization caused by an ion of energy $T-E$. Likewise,

$$M(E-I,1)=N(E-I) \tag{16}$$

is the mean number of ionization caused by an electron of energy $E-I$.

The Fano factor $F(E-I)$ for an electron of energy $E-I$ is defined as

$$F(E-I)N(E-I)=M(E-I,2)-[N(E-I)]^2\ . \tag{17}$$

By use of Eqs. (15)–(17), one may recast Eq. (14) as

$$\tilde{R}(T,2)=\int d\tilde{\sigma}_T(E)\{2\tilde{N}(T-E)N(E-I)+2\tilde{N}(T-E)+[N(E-I)+I]^2+F(E-I)N(E-I)\}\ . \tag{18}$$

C. Variance

The variance of the ionization yield is the most important of many indices characterizing the statistical fluctuations. Perhaps the most effective indices are the cumulants treated by Inokuti, Douthat, and Rau [4]. In the present article we limit our treatment to the variance,

$$\tilde{D}(T)=\sum_j[j-\tilde{N}(T)]^2\tilde{P}(T,j)=\tilde{M}(T,2)-[\tilde{N}(T)]^2\ . \tag{19}$$

This variance also satisfies the Fowler-type equation

$$\tilde{\Omega}_T\tilde{D}(T)=\tilde{\rho}_i(T)\ , \tag{20}$$

where

$$\tilde{\rho}_i(T)=\tilde{R}(T,2)-\tilde{\Omega}_T[N(T)]^2\ , \tag{21}$$

as we readily see by combining Eq. (11) with Eq. (19).

It is possible to rewrite the expression for $\tilde{\rho}_i(T)$ in a much more transparent form, viz.,

$$\tilde{\rho}_i(T)=\sum_s \tilde{\sigma}_s(T)[\tilde{N}(T)-\tilde{N}(T-E_s)]^2+\int d\tilde{\sigma}_T(E)[\tilde{N}(T)-\tilde{N}(T-E)-N(E-I)-1]^2 + \int d\tilde{\sigma}_T(E)F(E-I)N(E-I) \; . \tag{22}$$

In this equation, the first term represents the mean-squared deviation due to nonionizing collisions of the ion, the second term the mean-squared deviation due to ionizing collisions, and the last term the contributions due to succeeding collisions of secondary electrons, where $F(E-I)$ represents the Fano factor for an electron of kinetic energy $E-I$. The derivation of Eq. (22) from Eq. (21) is in the Appendix.

D. Evaluation of the moments and variance by use of the slowing-down spectrum

Equation (11) for the moment and Eq. (20) for the variance can be solved by ascending in T from the initial condition, Eq. (7). However, it is much more effective to solve the equation by use of the slowing-down spectrum, as first shown by Rau, Inokuti, and Douthat [3]. The key concept is the adjoint. We multiply Eq. (11) by a function $\tilde{y}(T)$, integrate over T, and obtain

$$\int dT\, \tilde{y}(T)\tilde{\Omega}_T\tilde{M}(T,\mu)=\int dT\, \tilde{y}(T)\tilde{R}(T,\mu) \; . \tag{23}$$

We then rewrite the left-hand side as

$$\int dT\, \tilde{y}(T)\tilde{\Omega}_T\tilde{M}(T,\mu)=\int dT[\tilde{\Omega}_T^{\dagger}\tilde{y}(T)]\tilde{M}(T,\mu) \; . \tag{24}$$

We have thus introduced the adjoint operator $\tilde{\Omega}_T^{\dagger}$, which acts on $\tilde{y}(T)$ and leaves the integral unchanged. The existence of an adjoint operator is well established for a wide class of operators in a wide class of function spaces. As Rau, Inokuti and Douthat [3] showed, the function $\tilde{y}(T)$ appropriate for use here is the slowing-down spectrum. In the present problem of the ion incidence, $\tilde{y}(T)$ is the slowing-down spectrum of the ion. If the ion has the initial energy T_0, we may write the slowing-down spectrum as $\tilde{y}(T_0,T)$. Physically, $\tilde{y}(T_0,T)dT$ represents the total path length of the ion during the slowing from $T+dT$ to T, when the ion had kinetic energy T_0. According to the Spencer-Fano theory, $\tilde{y}(T_0,T)$ obeys the equation

$$\tilde{\Omega}_T^{\dagger}\tilde{y}(T_0,T)=\delta(T-T_0)/n \; , \tag{25}$$

where n is the density of molecules in the medium. Combining Eqs. (23)–(25), we obtain

$$\tilde{M}(T_0,\mu)=n\int_I^{T_0} dT\, \tilde{y}(T_0,T)\tilde{R}(T,\mu) \; . \tag{26}$$

This represents the solution of Eq. (11); in other words, when $\tilde{y}(T_0,T)$ is known, $\tilde{M}(T_0,\mu)$ can be calculated as the integral on the right-hand side of Eq. (26). Likewise, the solution of Eq. (20) is written as

$$\tilde{D}(T_0)=n\int_I^{T_0} dT\tilde{y}(T_0,T)\tilde{\rho}_i(T) \; . \tag{27}$$

In other words, the variance is the average of $\tilde{\rho}_i(T)$ with the weight $\tilde{y}(T_0,T)$.

Parenthetically, $\tilde{y}(T_0,T)$ is most often readily evaluated. When T_0 and T greatly exceed most of the possible values of energy transfer (E_s and E), it is sensible to use the continuous-slowing-down approximation; then $\tilde{y}(T_0,T)$ is the reciprocal of the stopping power $n\tilde{\sigma}_{\rm st}(T)$, where $\tilde{\sigma}_{\rm st}(T)$ is the stopping cross section

$$\tilde{\sigma}_{\rm st}(T)=\sum_s E_s\tilde{\sigma}_s(T)+\int d\tilde{\sigma}_T(E)E \; . \tag{28}$$

Consequently, one arrives at the following expression for the Fano factor for the incidence of an ion of energy T_0:

$$\tilde{F}(T_0)\equiv\frac{\tilde{D}(T_0)}{\tilde{N}(T_0)}=\frac{\int_I^{T_0} dT\, \tilde{y}(T_0,T)\tilde{\rho}_i(T)}{\int_I^{T_0} dT\, \tilde{y}(T_0,T)\tilde{R}(T,1)} \; , \tag{29}$$

where $\tilde{R}(T,1)$ is given by Eq. (13) and $\tilde{\rho}_i(T)$ by Eq. (22). A consequence of this result is noteworthy. According to the second mean-value theorem of integral calculus [18], the right-hand side of Eq. (29) is equal to the value $\tilde{\phi}(T_m)$ of the function

$$\tilde{\phi}(T)=\tilde{\rho}_i(T)/\tilde{R}(T,1) \tag{30}$$

evaluated at T_m, a value of the ion kinetic energy T somewhere in the interval of integration. In other words, for some value T_m, $\tilde{F}(T_0)=\tilde{\phi}(T_m)$ and $I<T_m<T_0$. If $\tilde{\phi}(T)$ varies slowly with T, the precise value of T_m is immaterial, and we immediately obtain an estimate of $\tilde{F}(T_0)$ from $\tilde{\phi}(T)$ alone. Only if $\tilde{\phi}(T)$ is sensitive to T over a large interval of T, we need the slowing-down spectrum to determine $\tilde{F}(T_0)$.

The above discussion represents the major general conclusions of the present theory.

III. APPLICATION TO ARGON

A. Method of calculation

Calculations for proton incidence on argon were performed according to the theory described in Sec. II D. The following data were used as input.

The stopping cross section $\tilde{\sigma}_{\rm st}(T)$ was evaluated from the analytic expressions that Andersen and Ziegler [19] determined from a survey of experimental data. Using the continuous-slowing-down approximation, we set $\tilde{y}(T_0,T)=[n\tilde{\sigma}_{\rm st}(T)]^{-1}$.

The ionization cross sections of many gases for proton impact have been extensively reviewed by Rudd and coworkers [20–22]. Rudd [21] gave an especially detailed analysis for argon including the contribution of each shell to both the differential and total ionization cross sections. We used the analytic expressions given by Rudd [21].

No comparably thorough knowledge of the discrete-excitation cross section $\tilde{\sigma}_s(T)$ of argon for proton impact appears to be available. For the present work, we adopted an estimate, based on the assumption that the stopping cross section of Ref. [19] is reliable. The estimate is simplified because discrete excitation accounts for a minor part of the stopping cross section $\tilde{\sigma}_{st}(T)$ as given by Eq. (28). Thus we can replace the sum over s in Eq. (28) with a single term and write

$$\tilde{\sigma}_{st}(T)=E_{ex}\tilde{\sigma}_{ex}(T)+\int d\tilde{\sigma}_T(E)E \ , \qquad (31)$$

where $\tilde{\sigma}_{ex}(T)$ is an effective discrete-excitation cross section and E_{ex} is an effective excitation energy. The second term on the right-hand side of Eq. (31) is calculable from the differential ionization cross section given by Rudd [21]. Using the stopping cross section $\tilde{\sigma}_{st}(T)$ given by Andersen and Ziegler [19], we determined the first term, $E_{ex}\tilde{\sigma}_{ex}(T)$, by subtraction. In numerical calculations of the Fano factor, we also replace the first term on the right-hand of Eq. (22) with $\tilde{\sigma}_{ex}(T)[\tilde{N}(T)-\tilde{N}(T-E_{ex})]^2$. We chose $E_{ex}=13.4$ eV so that $E_{ex}/I=0.86$, which is reasonable on the basis of electron-collision and oscillator-strength data [23]. (We also examined the sensitivity of our results to the value of E_{ex}. Variations of E_{ex} of a few tenths of an eV cause no appreciable changes in our results.)

As for the degradation of secondary electrons, we used the results of Kowari, Kimura, and Inoktui [10]. for $N(E-I)$ and $F(E-I)$ to evaluate $\tilde{\rho}_i(T)$ of Eq. (22) and $\tilde{R}(T,1)$ of Eq. (13).

To evaluate the mean yield of ionization $\tilde{N}(T_0)$ for proton incidence, we basically used Eq. (26) for $\mu=1$. However, we extended Eq. (26) to account for shellwise contributions and Auger effect contributions. For the latter we used the schematization of Kowari, Kimura, and Inokuti [10], in which a 200-eV electron is ejected after L-shell ionization.

B. Results on the W value

The mean yield of total ionization is customarily expressed in terms of the W value, $\tilde{W}(T_0)=T_0/\tilde{N}(T_0)$ in our present notation. With the use of the ionization cross sections of Ref. [21], we obtained 31.1 eV for protons of 1 MeV, for instance, as shown in Table II. This result is considerably higher than the literature values of 26.66 eV for protons and 26.40 eV for α particles of high energies [24]. It is generally believed [24] that the mean yield of ionization is nearly the same for electrons, protons, and α particles at comparable high speeds.

A probable reason for the high W value we obtained is that the ionization cross sections of Ref. [21] might be too small. We also note that Ref. [21] cites uncertainties of about 10%. We have therefore repeated the calculations by using values of the ionization cross sections that are enhanced by 5% and 10% over the values of Ref. [21]. (Results are shown in Table II.) However, the lowest W value of 28.3 eV we thus obtained is still higher than the literature values by 6%. There is a limit to the enhancement of the ionization cross sections: values of the ionization cross section that are too large would render the excitation cross section $\tilde{\sigma}_{ex}(T)$ negative, for the fixed value of the stopping cross section.

TABLE II. Results of calculations for protons of 1 MeV.

Ionization cross sections	$\tilde{W}$ (eV)	$\tilde{F}$
Values in Ref. [21]	31.1	0.236
5% greater	29.6	0.184
10% greater	28.3	0.157

Thus we consider that the comparison of the present results with experimental data is less than conclusive. This finding is in contrast with our work on the W value for electron incidence, for which our earlier results [10] were reasonably consistent with experiment.

C. Results on the Fano factor

Table II shows also the values of the Fano factor for the three calculations. As the value of the ionization cross section increases, both the W value and the Fano factor decrease monotonically and smoothly. The present finding is reminiscent of the correlations between the Fano factor and the W value seen in other contexts. Kimura *et al.* [25] evaluated the Fano factor for Ar-H_2 mixtures over the entire range of composition for electron incidence and found that the Fano factor and the W value not only change together with varying composition but even show an almost linear relation. Krajcar-Bronić [26] surveyed experimental data for various gases and also found remarkable correlations between the Fano factors and the W values.

As we stated in Sec. II B, the 10% enhancement of the ionization cross sections led to a W value close to experiment. Thus it is probably appropriate to consider the values of the Fano factor resulting from the same ionization cross-section values.

Table III shows the energy dependence of the results. The third column gives the Fano factor as defined by Eq. (29), and the fourth column gives a new quantity $\tilde{F}_p(T_0)$, which represents the direct contribution of the proton. The quantity is defined by

$$\tilde{F}_p(T)=\frac{\int_I^{T_0} dT\,\tilde{y}(T_0,T)[\tilde{\rho}_i(T)]_p}{\int_I^{T_0} dT\,\tilde{y}(T_0,T)\tilde{R}(T,1)} \ , \qquad (32)$$

TABLE III. Results of calculations for various incident energies.

Incident proton energy T_0 (MeV)	$\tilde{W}(T_0)$ (eV)	$\tilde{F}(T_0)$	$\tilde{F}_p(T_0)$
2.00	28.5	0.163	0.096
1.50	28.4	0.157	0.089
1.000	28.3	0.157	0.086
0.750	28.4	0.160	0.088
0.500	29.0	0.172	0.098
0.250	31.7	0.216	0.144
0.125	36.1	0.291	0.225

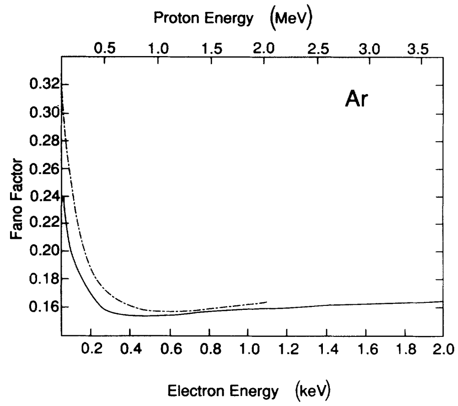

FIG. 1. Comparison of the Fano factors for proton incidence and electron incidence at the same speed. The chained curve refers to the proton, whose kinetic energy in MeV is shown on the upper horizontal axis. The solid curve refers to the electron, whose kinetic energy in keV is shown on the lower horizontal axis. The data for the electron are taken from Kowari, Kimura, and Inokuti [10]. However, they are truncated in this figure at the electron energy of 50 eV, and therefore the steep rise at lower energies is not shown here. Figure 8 and Table II of Kowari, Kimura, and Inokuti [10] show the data completely.

where

$$[\tilde{\rho}_i(T)]_p = \sum_s \tilde{\sigma}_s(T)[\tilde{N}(T) - \tilde{N}(T - E_s)]^2 + \int d\tilde{\sigma}_T(E)[\tilde{N}(T) - \tilde{N}(T - E) - N(E - I) - 1]^2 . \quad (33)$$

This equation differs from Eq. (22) for $\tilde{\rho}_i(T)$ in that the last term is dropped. Comparison of $\tilde{F}(T)$ with $\tilde{F}_p(T)$ indicates that the role of secondary electrons is clearly important at high proton energies.

To show the difference between proton incidence and electron incidence at the same speed, we present Fig. 1. The Fano factors for both particles show similar increase with decreasing speed and very gradually approach a plateau value at high energies. Throughout the region of our study, the Fano factor for proton incidence has a higher value and a smoother energy dependence than the Fano factor for electron incidence, as expected from our theory.

Finally, Fig. 2 shows $\tilde{\phi}(T)$, defined by Eq. (30) as a function of T. As we expected on general grounds, $\tilde{\phi}(T)$ is mildly dependent on T at high T, and its mean value is close to the Fano factor at high T. The narrow region $T < 0.2$ MeV, where $\tilde{\sigma}_{\rm st}(T)$ increases sharply with decreasing T, contributes little to the integrals in Eq. (29) that determine the Fano factor, if the incident energy T_0 is much higher.

The calculated Fano factor, 0.157, for the 1.0- to 1.5-MeV proton should apply to α particles of the same speeds, i.e., at energies of 4–6 MeV. However, our results are considerably lower than the results of measurements with α particles [5,6] as quoted in Table I.

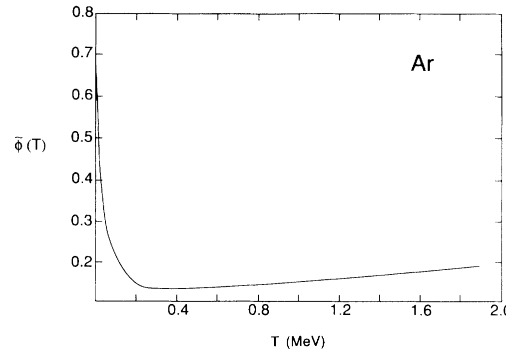

FIG. 2. The function $\tilde{\phi}(T)$, defined by Eq. (30).

IV. CONCLUDING REMARKS

We have presented a theory for calculating the W value and the Fano factor for the incidence of protons and α particles. The theory presumes a knowledge of the cross sections for all of the major processes, and results of its application are subject to the uncertainties in the current cross-section data. In our example of the first application, argon, several issues remain to be resolved.

First, it is difficult to obtain a W value in good agreement with the literature values. The discrepancy of our result from the literature values suggests that the ionization cross sections given by Rudd [21] are probably too small by at least 10%. The discrepancy might even suggest that the shape of the secondary-electron spectra needs to be reconsidered.

Second, our calculations led to a tight correlation between the Fano factor and the W value as the ionization cross sections were varied. The Fano factors we obtained for protons are almost the same as those for electrons of the same speeds given by both theory and experiment (as seen in Table I). In contrast, measurements [6,7] with α particles gave considerably higher values of the Fano factor, while the W values in the literature [23] are nearly the same for α particles, protons, and electrons at high speeds. This situation is remarkable in view of the correlation between the W value and the Fano factor that is often observed in other contexts [25,26].

Kase *et al.* [5] and Doke, Ishida, and Kase [27] proposed to explain the difference between the Fano factors for α particles and electrons as being attributable to effects of elastic nuclear collisions, following the idea of Lindhard and Nielsen [11]. To clarify this issue, we can include the effects of nuclear collisions in our formalism and repeat our calculations. A first step toward a definitive result in this effort seems to be reevaluation of ionization and other cross sections so that a satisfactory W value is obtained. The energy losses to nuclear translational motion should be important at lower incident energies and in gases of lighter molecules. (At such lower energies, the influence of electron capture and loss by the incident ion may also have to be considered.) In this connection, the new measurements by Ishida, Kikuchi, and Doke [28], on helium are extremely intriguing. We intend to study helium in the future.

In conclusion, we present a suggestion to experimenters. Measurements of the Fano factors of molecular hydrogen for protons or α particles will be valuable for clarifying the role of nuclear elastic collisions and resulting recoil particles. In particular, comparison between H_2 and D_2 will be most informative because of the mass ratio of about 2.

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APPENDIX: THE DERIVATION OF EQ. (22)

First, it is necessary to rewrite $\tilde{\Omega}_T[\tilde{N}(T)]^2$ as follows:

$$\begin{aligned}\tilde{\Omega}_T[\tilde{N}(T)]^2 &\equiv \tilde{\sigma}_{\mathrm{tot}}(T)[\tilde{N}(T)]^2-\sum_n \tilde{\sigma}_s(T)[\tilde{N}(T-E_s)]^2-\int d\tilde{\sigma}_E[\tilde{N}(T-E)]^2 \\ &= \tilde{\sigma}_{\mathrm{tot}}(T)[\tilde{N}(T)]^2-\sum_s \tilde{\sigma}_s(T)[\tilde{N}(T)-\tilde{N}(T-E_s)]^2-2\tilde{N}(T)\sum_s \tilde{\sigma}_s(T)\tilde{N}(T-E_s)+[\tilde{N}(T)]^2\sum_s \tilde{\sigma}_s(T) \\ &\quad -\int d\tilde{\sigma}_T(E)[\tilde{N}(T)-\tilde{N}(T-E)]^2-2\tilde{N}(T)\int d\tilde{\sigma}_T(E)\tilde{N}(T-E)+[\tilde{N}(T)]^2\int d\tilde{\sigma}_T(E) \\ &= -\sum_s \tilde{\sigma}_s(T)[\tilde{N}(T)-\tilde{N}(T-E_s)]^2-\int d\tilde{\sigma}_T(E)[\tilde{N}(T)-\tilde{N}(T-E)]^2 \\ &\quad +2\tilde{N}(T)\left[-\sum_s \tilde{\sigma}_s(T)\tilde{N}(T-E_s)-\int d\tilde{\sigma}_E(T)\tilde{N}(T-E)+\tilde{\sigma}_{\mathrm{tot}}(T)\tilde{N}(T)\right] . \end{aligned} \tag{A1}$$

The terms within the large parentheses are equal to $\tilde{\sigma}_i(T)+\int d\tilde{\sigma}_T(E)\tilde{N}(E-I)$, according to the Fowler equation for $\tilde{N}(T)$, i.e., Eq. (11) with $\mu=1$. Thus one may write

$$\begin{aligned}\tilde{\Omega}_T[\tilde{N}(T)]^2 &= -\sum_s \tilde{\sigma}_s(T)[\tilde{N}(T)-\tilde{N}(T-E_s)]^2-\int d\tilde{\sigma}_T(E)[\tilde{N}(T)-\tilde{N}(T-E)]^2+2\tilde{\sigma}_i(T)\tilde{N}(T) \\ &\quad +2\tilde{N}(T)\int d\tilde{\sigma}_T(E)N(E-I) . \end{aligned} \tag{A2}$$

Inserting this into Eq. (21), one obtains

$$\begin{aligned}\tilde{\rho}_i(T) &= \sum_s \tilde{\sigma}_s[\tilde{N}(T)-\tilde{N}(T-E_s)]^2+\int d\tilde{\sigma}_T(E)[\tilde{N}(T)-\tilde{N}(T-E)]^2 \\ &\quad +\int d\tilde{\sigma}_T(E)\{-2\tilde{N}(T)-2\tilde{N}(T)N(E-I)+2\tilde{N}(T-E)N(E-I)+2\tilde{N}(T-E)+[N(E-I)+1]^2 \\ &\qquad +F(E-I)N(E-I)\} . \end{aligned} \tag{A3}$$

It is elementary to see that this is equivalent to Eq. (22).

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[1] U. Fano, Phys. Rev. **72**, 26 (1947).
[2] J. K. Knipp, T. Eguchi, M. Ohta, and S. Nagata, Prog. Theor. Phys. **10**, 24 (1953).
[3] A. R. P. Rau, M. Inokuti, and D. A. Douthat, Phys. Rev. A **18**, 971 (1978).
[4] M. Inokuti, D. A. Douthat, and A. R. P. Rau, Phys. Rev. A **22**, 445 (1980).
[5] M. Kase, T. Akioka, H. Mamyoda, J. Kikuchi, and T. Doke, Nucl. Instrum. Methods **227**, 311 (1984).
[6] G. D. Alkhazov, A. P. Komar, and A. A. Vorob'eV, Nucl. Instrum. Methods **48**, 1 (1967).
[7] W. Neumann, in *The Seventh Symposium on Microdosimetry, 8–12 September 1980, Oxford, U.K.*, edited by J. Booz, H. G. Ebert, and H. D. Hartfield (Harwood Academic, London, 1981), p. 1067.
[8] G. D. Alkhazov, Zh. Tekh. Fiz. **41**, 1949 (1971) [Sov. Phys.—Tech. Phys. **16**, 1540 (1972)].
[9] B. Grosswendt, J. Phys. B **17**, 1391 (1984).
[10] K. Kowari, M. Kimura, and M. Inokuti, Phys. Rev. A **39**, 5545 (1989).
[11] J. Lindhard and V. Nielsen, Phys. Lett. **2**, 209 (1962).
[12] J. Lindhard, V. Nielsen, M. Scharff, and P. V. Thomsen, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **33**, No. 10 (1963).
[13] M. Inokuti, Rev. Mod. Phys. **43**, 297 (1971).
[14] L. V. Spencer and U. Fano, Phys. Rev. **93**, 1172 (1954).
[15] M. Inokuti, M. A. Dillon, and M. Kimura, Int. J. Quantum Chem. Symp. **21**, 251 (1987).
[16] M. Inokuti, M. A. Dillon, and M. Kimura, Phys. Rev. A **38**, 1217 (1988).
[17] K. Kowari, M. Inokuti, and M. Kimura, Phys. Rev. A **42**, 795 (1990).
[18] J. H. Hardy, *A Course of Pure Mathematics* (Cambridge University Press, Cambridge, England, 1955), p. 244.
[19] H. H. Andersen and J. F. Ziegler, *Hydrogen Stopping*

Powers and Ranges in All Elements (Pergamon, New York, 1977).
[20] M. E. Rudd, Y.-K. Kim, D. H. Madison, and J. W. Gallagher, Rev. Mod. Phys. **57**, 965 (1985).
[21] M. E. Rudd, Phys. Rev. A **38**, 6129 (1988).
[22] M. E. Rudd, Nucl. Tracks Radiat. Meas. **16**, 213 (1989).
[23] E. Eggarter, J. Chem. Phys. **62**, 833 (1975).
[24] International Commission on Radiation Units and Measurements, *Average Energy to Produce an Ion Pair* (International Commission on Radiation Units and Measurements, Bethesda, MD, 1979), Report No. 31.
[25] M. Kimura, I. Krajcar-Bronić, M. Dillon, and M. Inokuti, Phys. Rev. A (to be published).
[26] I. Krajcar-Bronić and M. Kase (unpublished).
[27] T. Doke, N. Ishida, and M. Kase, Nucl. Instrum. Methods (to be published).
[28] N. Ishida, J. Kikuchi, and T. Doke, Jpn. J. Appl. Phys. (to be published).