Statistics of Particle Penetration

By PETER SIGMUND

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Synopsis

An attempt is made to formulate a statistical theory of particle penetration phenomena in real, i.e. nonrandom stopping media. The treatment concentrates on individual collision events that are well localized in space, but correlated via the trajectory of the penetrating particle. Since Poisson statistics does not govern a sequence of collision events in an ordered structure, the mean-free-path concept fails, and penetration theory has to be developed from the bottom. As it turns out, the cross section keeps to be the leading concept when defined as an atomic parameter without recourse to the mean free path. However, a sequence of competing correlation terms occurs which do not influence the average behavior of the beam but enter fluctuations and higher-order averages. These correlation terms depend on the structure of the stopping medium. The leading one contains the pair correlation function of the structure, and can be expressed in terms of the structure factor as measured in x-ray or neutron diffraction.

As a main result, one obtains an extension of Bothe's formula—which governs the statistics of particle penetration through thin layers—to ordered matter. The more specific discussion refers to energy loss and multiple scattering of heavy charged particles, as well as inner-shell processes, in ideal molecular gases, real gases, and amorphous solids or liquids. Previous results derived for the energy loss and multiple scattering of ions in molecular gases are contained in the present description. In that work it has already been documented that correlation effects may become pronounced when the cross sections are large. The present work shows that correlations may be positive or negative, dependent on whether the attractive or repulsive interaction between target particles dominates in the range of impact parameters governing the collision events in question.

PETER SIGMUND
Physical Laboratory II
H. C. Ørsted Institute
DK-2100 Copenhagen Ø

present address:
Physics Institute
Odense University
DK-5230 Odense

1. Introduction

In studies of the interaction of particles with matter, it is convenient to distinguish between collision and penetration phenomena. Collisions are distinct events on a microscopic level, such as the excitation of an electron by an alpha particle or the dissociation of a molecule by a photon. Penetration phenomena may be observed on a larger scale, under conditions where the chance for one particle to undergo several collisions is appreciable. Typical examples of penetration phenomena are the stopping and multiple scattering of charged particles in dense matter, and various ionization phenomena.

From an experimental point of view, the transition between collision and penetration phenomena is gradual. Collision phenomena are investigated by means of thin targets to ensure single events. The requirements to be imposed on a target in order to be called "thin" depend on the desired accuracy of experimental results. Even in a dilute gas target, there is a non-zero probability for more than one collision in an individual passage.

From a theoretical point of view, one may consider collision events to be governed by the appropriate equations of motion, such as Schrödinger's equation, while statistical considerations are essential in the treatment of penetration phenomena.

The statistical theory of particle penetration was developed early in this century[1-4]. This theory is generally based on the assumption of statistical independence of different collision events. Therefore, quantitative predictions originate in Poisson's distribution, Boltzmann's equation, or equivalent first principles.

According to standard penetration theory[5], the probability P for a projectile to initiate a certain event while penetrating a small path length Δx is

$$P = \mathcal{N}\Delta x \sigma, \tag{1}$$

where $\mathcal{N}$ is the number of target particles (atoms, molecules, electrons, etc) per volume, and σ the cross section for the event in question. A necessary condition for sucessive events to be statistically independent is the possibility to make Δx small enough so that $P \ll 1$. A lower limit of Δx is set by the duration of a collision. For an order-of-magnitude estimate, take an interaction radius a, and set $\Delta x \gtrsim 2a$, $\sigma \sim \pi a^2$, and $\mathcal{N} \sim (3/4\pi)(2/d)^3$, where d is a mean interparticle spacing in the target. Then, successive collisions can be statistically independent only if

$$P \sim 12(a/d)^3 \ll 1 \; ; \tag{2}$$

For processes on an atomic scale, this condition is easily fulfilled when the target is a gas. In solid or other dense media, eq. (2) is obeyed for processes on a nuclear scale, while for atomic processes, it is more or less violated except for very rare events.

Once eq. (2) is not fulfilled, the concept of a free flight path becomes meaningless, and hence Poisson statistics does not readily apply. Conversely, the very fact of an appreciable probability for a projectile to interact with every target particle along its path suggests an influence of target structure on the statistics of collision processes. Another important consequence is the possibility of collective processes that has been pointed out long ago[6,7].

The present paper deals with the statistics of penetration phenomena in situations where the basic collision events are well-localized but not statistically independent. There is a wide range of penetration phenomena—in particular in solid targets—where correlations between individual collision events are nonnegligible but not dominating. In the opposite case of strong correlation, a redefinition of the basic collision event may be possible such as to make correlations weak. An attempt is made in this work to provide a description that is flexible enough to allow for both spatial and other types of correlations.

The relation between such a treatment and conventional penetration theory is similar in several ways to that between the kinetic theory of real and ideal gases, and some of the statistical methods applied here are indeed common in gas theory.

Despite the desired flexibility, the guiding principles have been chosen to aim at a treatment of heavy-charged-particle penetration phenomena, where the importance of correlation effects has been documented recently[8-12].

At present, the treatment has been limited to the case of negligible feedback of collisions on the motion of the projectile particle. This implies small energy loss and deflection and, more important, neglect of possible changes of projectile state during penetration. A more general treatment incorporating the latter class of phenomena is in preparation[13].

In sects. 2 & 3, general expressions are derived for the frequency spectrum of individual collision events and the cumulative effect of collisions in the presence of correlations. The main result of this treatment is a generalization of Bothe's formula[2] to correlated systems. In sections 4 and 5, spatial correlations are considered by means of an impact-parameter treatment of individual collisions. In section 6, comments will be made on how to select suitable statistical variables, and in section 7, various methods of evaluating cumulative effects are extended to correlated systems. Finally, section 8 contains more explicit results for energy loss and multiple scattering. In particular, this section provides the link between the general considerations made in the present work and the more special situations treated earlier[9,10,12].

2. Statics of a Single Type of Event

Take a target with a number of independent degrees of freedom, or modes of excitation, and consider a specific event, called A in the following. As an example, take a solid target, and denote by A the process of K-shell excitation in any of the constituent atoms of the solid which may be assumed monoatomic for the moment.

Let a projectile interact with the target, but disregard all action of the target on the projectile. Within the above example, take an energetic ion moving on a straight line through the solid with negligible energy loss, and disregard electron capture and loss as well as excitation and deexcitation of the ion.

In the following, it will be convenient to speak of an "ion" and "target atoms" as well as the "passage" of an ion, even though the projectile need not be an ion and the target particles might be electrons, nucleons, molecules, plasmons, phonons, etc. It will, however, be assumed that the target has macroscopic dimensions, i.e., that the number of target atoms is large.

Even though we deal with independent modes of excitation in the target, there may exist a more or less pronounced correlation of events A in any individual passage. Within the above example, only those atoms have nonzero probability for K-shell excitation that are located within some microscopically small distance from the trajectory of the ion. Thus, excitation processes in different target atoms are correlated via the trajectory of the ion.

Suppose there are z different ways of initiating event A, i.e. z different target atoms. Let P_i $(i = 1, \ldots z)$ be the probability for initiating A in the i-th atom in a particular passage. Then, the probability F_n for n events A in that passage is given by

$$F_n = \sum_{1 \le i_1 < \ldots < i_n \le z} P_{i_1} \ldots P_{i_n} \cdot \prod_{j(\neq i_1, \ldots i_n)} (1 - P_j) ; \quad n = 0, 1, \ldots z ; \quad (3)$$

each term in the sum representing the probability for event A to occur in a given selection of n atoms, and not to occur in the remaining $(z-n)$ atoms. The sum includes all $\binom{z}{n}$ different selections of n atoms.

The F_n are readily seen to be the coefficients of a power series expansion of the generating function (or partition function)

$$f(s) = \prod_{i=1}^{z} (1 - P_i + sP_i) \equiv \sum_{n=0}^{z} F_n s^n \quad (4)$$

where s is a dimensionless variable with no physical significance at this point.

From (4) one obtains

$$f(1) = 1 = \sum_{n=0}^{z} F_n , \quad (5a)$$

i.e., the individual probabilities add up to 1. Moreover, taking derivatives one obtains average values,

$$f'(1) = \sum_n n F_n = \sum_i P_i \; ; \tag{5b}$$

$$f''(1) = \sum_n n(n-1) F_n = \sum_{i \neq j} P_i P_j \; ; \tag{5c}$$

etc. This results in the Taylor series

$$f(s) = 1 + (s-1) \sum_i P_i + \tfrac{1}{2}(s-1)^2 \sum_{i \neq j} P_i P_j \ldots \tag{6}$$

Consider now the average $\overline{F}_n$ over many different passages, i.e. the frequency spectrum for event A when a macroscopic beam of projectiles interacts with the target.

Let us make the important assumption at this stage that the beam is homogeneous, i.e. equal *a priori* probability P per beam particle to undergo event A for all target atoms,

$$\overline{P}_i = P \;\; \text{for } i = 1, \ldots z \tag{7}$$

Eq. (7) requires that no target atoms are "shadowed" systematically by others. The relation is readily fulfilled in case of an ion beam spread uniformly over a macroscopic target area, since it has already been assumed that individual trajectories are not governed by target structure, and that energy loss and scattering are inappreciable. An example of a situation where (7) is not generally fulfilled is that of an internal source of projectiles, e.g. from radioactive decay[12], and, especially, channeling phenomena in crystals[14].

In case of a homogeneous beam, (5b) and (7) yield[14]

$$\bar{n} = \sum_n n \overline{F}_n = \sum_i \overline{P}_i = zP \; ; \tag{8}$$

Similarly, from (5c),

$$\overline{n^2} - \bar{n}^2 = zP(1-P) + \sum_{i \neq j} Q_{ij} \tag{9}$$

where

$$Q_{ij} = \overline{(P_i - P)(P_j - P)} \tag{9a}$$

Obviously, Q_{ij} is a measure of the correlation between the events initiated in two given atoms i and j.

Let us now make use of the assumption that z is a large number, i.e., that we deal with a macroscopic target. With target thickness x and target (or beam) area S, we have

$$z = NSx, \tag{10}$$

so z can be made as large as desired by a mere increase of S. So long as the projectiles interact over a microscopic distance, this does not affect the essential physical properties of the system. (We keep in mind that bounds exist on the target thickness x because of the requirement of negligible beam attenuation).

By definition of P, we must have

$$P = \frac{\sigma}{S}, \tag{11}$$

where σ is the cross section for event A. Obviously, P is a very small quantity, unlike the statistical variables P_i which may take on values between 0 and 1.

From (8), (10), and (11), one obtains the standard prescription for determining cross sections[5],

$$\bar{n} = Nx\sigma ; \tag{12}$$

Note, however, that $\bar{n}$ need not be small in order that (12) be valid, nor has it been assumed that collision events be statistically independent.

Consider now the fluctuation, eq. (9), in case of a macroscopic target. We have $zP(1-P) \cong zP$, since $P \ll 1$. The correlation coefficients Q_{ij} differ from zero only if atoms i and j lie within some welldefined microscopic volume. Hence, if the target area S is made very large, only one summation in the double sum extends over all atoms while the other is limited. In other words, the double sum increases as $\propto z$ rather than $\propto z^2$, so we can write

$$\overline{n^2} - \bar{n}^2 = z(P + \Delta P_2) \tag{13}$$

with

$$\Delta P_2 = \frac{1}{z} \sum_{i \neq j} Q_{ij} , \tag{13a}$$

and ΔP_2 being finite for $z \to \infty$.

Consider now the beam average over the partition function (6),

$$\bar{f}(s) = 1 + (s-1)\bar{n} + \tfrac{1}{2}(s-1)^2(\overline{n^2}-\bar{n}) + \ldots \equiv \sum_n \overline{F}_n s^n ; \tag{14}$$

After insertion of (12) and (13) one finds

$$\bar{f}(s) = 1 + (s-1)zP + \tfrac{1}{2}(s-1)^2[(zP)^2 + z\Delta P_2] + \ldots \tag{14'}$$

or, rearranging according to powers of z,

$$\bar{f}(s) = 1 + z[(s-1)P + \tfrac{1}{2}(s-1)^2\Delta P_2 + \ldots] + \tfrac{1}{2}z^2[(s-1)^2P^2 + \ldots] + \ldots \tag{14''}$$

where the dots indicate terms containing correlations.

It is evident (and well-known[15]) that in the absence of correlations, $\bar{f}(s)$ becomes exponential,

$$\bar{f}(s) = e^{(s-1)zP}; \tag{15}$$

This results in Poisson's distribution[1.5]

$$\overline{F}_n = e^{-Nx\sigma}\,\frac{(Nx\sigma)^n}{n!} \tag{16}$$

for the frequency spectrum of an individual event A governed by a cross section σ.

It is tempting to generalize eq. (15) in accordance with (14''), i.e. to write

$$\bar{f}(s) = e^{z[(s-1)P + \frac{1}{2}(s-1)^2\Delta P_2 + \ldots]}; \tag{17}$$

The proof is central. It is carried out, therefore, in some detail.

Write the beam average of eq. (6) in the form

$$\bar{f}(s) = \sum_{n=0}^{z}\frac{a_n}{n!}(s-1)^n, \tag{18}$$

where

$$a_n = \sum_{i \neq \ldots \neq i_n} \overline{P_{i_1}\ldots P_{i_n}} \quad \text{for} \quad \begin{array}{l} n \geqslant 1 \\ \\ n = 0 \end{array}$$

Introducing correlation coefficients

$$b_n = \sum_{i_1 \neq \ldots \neq i_n} Q_{i_1 \ldots i_n}; \qquad n \geqslant 1 \tag{19}$$

with

$$Q_{i_1 \ldots i_n} = \overline{(P_{i_1} - P) \ldots (P_{i_n} - P)} \tag{20}$$

we find the following identities,

$$a_n = b_n - \sum_{m=1}^{n} (-)^m \binom{n}{m} (z-n+m) \ldots (z-n+1) P^m a_{n-m}$$

$$\cong b_n - \sum_{m=1}^{n} (-)^m \binom{n}{m} (zP)^m a_{n-m} \quad \text{for } 1 \leqslant n < z;$$

From this follows

$$a_n = \sum_{m=0}^{n} \binom{n}{m} b_{n-m} (zP)^m \tag{21}$$

with $b_0 = 1$. Insertion of (21) into (18), and rearrangement of summations yields

$$\bar{f}(s) \cong e^{(s-1)zP} \sum_{n \geqslant 0} \frac{b_n}{n!} (s-1)^n ; \tag{22}$$

Errors in (22) arise from those terms where n becomes comparable with z. (Note that $z \propto S$ while $z \cdot P$ is independent of S).

Consider first the case where all P_i are uncorrelated. Then, $b_n = 0$ for $n \geqslant 1$, and (22) becomes identical with eq. (15).

Consider next the case of pair correlations only, which may be realized by a diatomic molecular gas. Then, higher-order correlations are nonzero only to the extent that they can be decomposed into pair correlations. By means of simple combinatorics, (19) yields

$$b_{2m} = (2m-1)(2m-3) \ldots 3 \cdot 1 \, (b_2)^m$$

$$b_{2m-1} = 0$$

for $m = 1, 2 \ldots$. Therefore,

$$\sum_0 \frac{b_n}{n!}(s-1)^n = e^{\frac{1}{2}(s-1)^2 b_2};$$

With this, and (13a) and (19), (22) becomes identical with (17).

In case where both second and third order correlations are present, while higher correlations decompose, one finds

$b_4 = 3(b_2)^2$;
$b_5 = 10 b_2 b_3$;
$b_6 = 15(b_2)^3 + 20(b_3)^2$;
$b_7 = 105(b_2)^2 b_3$;
etc.

so

$$\sum_0 \frac{b_n}{n!}(s-1)^n = e^{\frac{1}{2}(s-1)^2 b_2 + \frac{1}{6}(s-1)^3 b_3};$$

Similarly, in case of higher correlations, one may write

$$\bar{f}(s) = e^{z\{(s-1)P + \sum_{n \geqslant 2} \frac{\Delta P_n}{n!}(s-1)^n\}} \tag{23}$$

where

$$\Delta P_n = \frac{1}{z} \sum_{i_1 \neq \ldots \neq i_n}^{(i)} Q_{i_1 \ldots i_n}, \tag{24}$$

and the index (i) indicates that only irreducible n-th order correlations are to be included in the sum.

In the above treatment, z was essentially considered infinite. This requires the functional behavior of $\bar{f}(s)$ to be determined by a number n_0 of terms in the Taylor series (18) with $n_0 \ll z$. If s is restricted to the range $|s| \leqslant 1$, and the ΔP_n bounded according to $|\Delta P_n| \lesssim P$, one must require that the relation

$$\frac{(zP)^n}{n!} \ll 1$$

be obeyed for all $n \geqslant n_0$ with some value $n_0 \ll z$. By means of Stirling's formula and eq. (12), this becomes equivalent with

$$\frac{Nx\sigma}{\log\frac{n_0}{e}} < 1 , \tag{25}$$

where $e = 2.718$. By comparison with eqs. (1) and (2) one finds that (25) is a rather weak requirement.

In most applications of the present treatment, only pair correlations are important, i.e. all ΔP_n for $n \geqslant 3$ can be ignored. In that case, $\bar{f}(s)$ according to (23) reduces to a generating function for Hermite polynomials[16]. By term-by-term comparison with (14) one finds

$$\overline{F}_n = e^{-z(P-\Delta P_2/2)} \cdot \sum_{m=0}^{[n/2]} \frac{(z\Delta P_2/2)^m \, [z(P-\Delta P_2/2)]^{n-2m}}{m! \, (n-2m)!} ; \tag{26}$$

This is a generalization of Poisson's formula (16) to systems with pair correlation.

In subsequent applications, the partition function $\bar{f}(s)$ itself will actually become more useful than the frequency spectrum $\overline{F}_n$.

3. Cumulative Effect of a Multitude of Events

Consider now two mutually exclusive events A_1 and A_2 such that one ion can initiate A_1 or A_2 in a given target atom, or none but not both. Let P_{i1} and P_{i2} be the respective probabilities $(i = 1, \ldots z)$, and let $F_{n_1 n_2}$ be the joint frequency spectrum for n_1 events A_1 and n_2 events A_2 in an individual passage. This quantity follows readily from the partition function

$$f(s_1 , s_2) = \prod_{i=1}^{z} ((1 - P_{i1} - P_{i2}) + s_1 P_{i1} + s_2 P_{i2}) = \sum_{0 \leqslant n_1 + n_2 \leqslant z} F_{n_1 n_2} s_1^{n_1} s_2^{n_2} , \tag{27}$$

where s_1 and s_2 are independent dimensionless variables. Eq. (27) is a straight generalization of eq. (4).

Taylor expansion of (27) similar to (6) yields

$$f(s_1 , s_2) = 1 + \sum_{\alpha} (s_\alpha - 1) \sum_i P_{i\alpha} + \tfrac{1}{2} \sum_{\alpha\beta} (s_\alpha - 1)(s_\beta - 1) \sum_{i \neq j} P_{i\alpha} P_{j\beta} + \ldots \tag{28}$$

or, by analogy with (17) or (23),

$$\bar{f}(s_1, s_2) = \exp z\{\sum_\alpha (s_\alpha - 1) P_\alpha + \tfrac{1}{2}\sum_{\alpha\beta} (s_\alpha - 1)(s_\beta - 1)\Delta P_{2.\alpha\beta} + \dots\} \tag{29}$$

where

$$P_\alpha = \overline{P_{i\alpha}} \tag{29a}$$

$$\Delta P_{2.\alpha\beta} = \frac{1}{z}\sum_{i \neq j} \overline{(P_{i\alpha} - P_\alpha)(P_{j\beta} - P_\beta)} \tag{29b}$$

etc., and $\alpha, \beta = 1, 2$.

Consider now some cumulative effect U,

$$U = n_1 u_1 + n_2 u_2 = \sum_\alpha n_\alpha u_\alpha,$$

where u_α is the contribution of an individual event of type A_α to U. Then, the probability density $G(U)$ is given by

$$G(U) = \sum_{n_1, n_2} \overline{F_{n_1 n_2}} \; \delta(U - \sum_\alpha n_\alpha u_\alpha)$$

or, after Fourier transformation,

$$G(U) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt \, e^{itU} \cdot \sum_{n_1 n_2} \overline{F}_{n_1 n_2} \cdot (e^{-itu_1})^{n_1} \cdot (e^{-itu_2})^{n_2}; \tag{30}$$

Comparison with (27) shows that the integrand can be expressed by means of the partition function $\bar{f}(s_1, s_2)$. Hence,

$$G(U) = \frac{1}{2\pi}\int dt e^{itU} \cdot \bar{f}(e^{-itu_1}, e^{-itu_2})$$

or, after insertion of (28),

$$G(U) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt e^{itu - z\Delta} \tag{31}$$

with

$$\Delta = \sum_\alpha (1 - e^{-itu_\alpha})P_\alpha - \tfrac{1}{2}\sum_{\alpha\beta} (1 - e^{-itu_\alpha})(1 - e^{-itu_\beta})\Delta P_{2.\alpha\beta} {}^{+-} \dots \tag{32}$$

The notation has been chosen so that eq. (32) remains true for any number of events A_α, $\alpha = 1,2,3,\ldots$ Moreover, if α is a continuous variable, the quantities $P_\alpha, \Delta P_{2,\alpha\beta}$ go over into corresponding probability densities and correlation functions. Finally, if u_α (and U) is a vectorial rather than a scalar quantity, eqs. (31) and (32) have to be read correspondingly, with t being a vector variable, too.

In the absence of correlations, (31) reduces to Bothe's formula[2]

$$G(U) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt\, e^{itU - Nx\sum_\alpha (1 - e^{-itu_\alpha})\sigma_\alpha} \tag{33}$$

which is the conventional starting point for the theory of small-angle multiple scattering[2,12] and energy-loss spectra[4], when specified suitably. Standard derivations[4,17] of eq. (33) are based on Boltzmann's equation.

For completeness one may wish to generalize eq. (32) to the case of a polyatomic target. Let there be $z_{(I)}$ atoms of species $I\ (= 1, 2, \ldots)$, and allow each species to undergo a set of events $A_{(I)\alpha}$ with $\alpha = 1, 2, \ldots$ Then, eq. (32) amplifies to

$$\Delta = \sum_I c_{(I)} \sum_\alpha (1 - e^{-itu_{(I)\alpha}}) P_{(I)\alpha} -$$

$$-\frac{1}{2} \sum_{I,J} c_{(I)} c_{(J)} \sum_{\alpha,\beta} (1 - e^{-itu_{(I)\alpha}}) \cdot (1 - e^{-itu_{(J)\beta}}) \Delta P_{2(IJ)\alpha\beta} + - \ldots, \tag{34}$$

where $P_{(I)\alpha}$ is the probability per beam particle to initiate an event $A_{(I)\alpha}$ in a given target atom of type I, i.e.

$$P_{(I)\alpha} = \frac{\sigma_{(I)\alpha}}{S}, \tag{35}$$

and $\sigma_{(I)\alpha}$ the respective cross section. $c_{(I)}$ is the concentration of species I,

$$c_{(I)} = z_{(I)}/z, \tag{36}$$

and $u_{(I)\alpha}$ the contribution of one event $A_{(I)\alpha}$ to the cumulative effect U. Finally,

$$\Delta P_{2(IJ)\alpha\beta} = \frac{z}{z_{(I)} z_{(J)}} \sum_{i \in I} \sum_{j \in J}' \overline{(P_{i\alpha} - P_{(I)\alpha})(P_{j\beta} - P_{(J)\beta})} \tag{37}$$

where the prime indicates omission of the term $i \neq j$ (for $I = J$).

4. Spatial Correlation. Impact Parameter Treatment

In this section, correlations will be evaluated for the case where the target particles (preferably atoms or molecules) can be taken as well localized in space for a certain amount of time, and where the essential parameter determining the collision events is the initial location of a target particle relative to the trajectory of a beam particle.

Let $\boldsymbol{p}$ be the vectorial impact parameter with respect to some specified point 0 within the target particle, e.g. the nucleus of an atom or the center-of-mass of a molecule. If the probability to initiate an event A in a given target particle depends on $\boldsymbol{p}$ only, we have

$$\sigma = \int P(\boldsymbol{p}) d^2p \tag{38}$$

according to (11). If $P(\boldsymbol{p})$ is essentially different from zero only for $\boldsymbol{p}$ covering some microscopic area $\ll S$, the integration in (38) can in practice be extended over the entire $\boldsymbol{p}$-plane.

Consider now the correlation coefficient Q_{ij}, eq. (9a) for two target particles i and j, located in $\boldsymbol{r}_i = (x_i, \varrho_i)$ and $\boldsymbol{r}_j = (x_j, \varrho_j)$, respectively. Here, the x-axis is parallel to the trajectory, and ϱ_i and ϱ_j are two-dimensional vectors specifying lateral positions. Let the trajectory have the lateral coordinate ϱ.

Introducing Fourier transforms

$$\sigma(\boldsymbol{k}) = \int_S d^2p \, P(\boldsymbol{p}) e^{i\boldsymbol{k}\cdot\boldsymbol{p}} ; \tag{38a}$$

$$P(\boldsymbol{p}) = \frac{1}{S}\sum_{\boldsymbol{k}} \sigma(\boldsymbol{k}) e^{-i\boldsymbol{k}\cdot\boldsymbol{p}} ; \tag{38b}$$

we can write

$$P_i - P = P(\varrho - \varrho_i) - P = \frac{1}{S}\sum_{\boldsymbol{k}}' \sigma(\boldsymbol{k}) e^{-i\boldsymbol{k}\cdot(\varrho - \varrho_i)}, \tag{39}$$

where the prime indicates ommission of the term $\boldsymbol{k} = 0$. Then, averaging over the beam yields

$$Q_{ij} = \frac{1}{S}\int d^2\varrho \, (P_i - P)(P_j - P) = \frac{1}{S^2}\sum_{\boldsymbol{k}}' |\sigma(\boldsymbol{k})|^2 e^{i\boldsymbol{k}\cdot(\varrho_i - \varrho_j)} ; \tag{40}$$

Similarly, the higher correlation coefficients, eq. (20), read

$$Q_{i_1 \ldots i_n} = \sideset{}{'}\sum_{\boldsymbol{k}_1} \cdots \sideset{}{'}\sum_{\boldsymbol{k}_n} \delta_{\boldsymbol{k}_1 + \ldots + \boldsymbol{k}_n, 0} \cdot \prod_{m=1}^{n} \left(\frac{\sigma(\boldsymbol{k}_m)}{S} e^{i\boldsymbol{k}_m \cdot \boldsymbol{\varrho}_{i_m}} \right)$$

where $\delta_{\boldsymbol{k}, \boldsymbol{k}'}$ is Kronecker's symbol.

From (13a) and (40) we obtain the leading correlation term in eq. (17),

$$z\Delta P_2 = \frac{1}{S^2} \sideset{}{'}\sum_{\boldsymbol{k}} |\sigma(\boldsymbol{k})|^2 < \sum_{i \neq j} e^{i\boldsymbol{k} \cdot (\boldsymbol{\varrho}_i - \boldsymbol{\varrho}_j)} > ; \tag{41}$$

Here, the sum over individual target particles has been put into brackets that symbolize their distribution. Indeed, in an experiment, projectiles hit the target at different times. Therefore, a time average is taken over the set of instantaneous positions $\{\boldsymbol{r}_1, \ldots \boldsymbol{r}_z\}$.

As it stands, eq. (41) implies that all motion of target atoms during the time interval of an individual passage can be ignored. In practice, target motion is required only to be inappreciable for the amount of time needed by the projectile to penetrate the correlation volume, i.e., the volume within which atomic positions are strongly correlated. In the absence of long-range order, this does not mean a severe restriction even for only moderately fast projectiles. (Otherwise time-dependent correlations have to be introduced[18]). Systems with long-range order such as crystals need special care for a different reason (cf. below).

The quantity in brackets in eq. (41) is quite closely related to the structure factor that is measured in conventional diffraction experiments. Indeed, take a situation where the target thickness x is much larger than the range of the correlation of the structure in question, i.e., disregard surface effects. By means of the pair distribution[19] $g_2(\boldsymbol{r})$ --- $\mathcal{N} \cdot g_2(\boldsymbol{r}) d^3 r$ is the probability to find a target particle in $(\boldsymbol{r}, d^3 r)$ if there is one in $\boldsymbol{r} = 0$ --- we have

$$< \sum_{i \atop j} e^{i\boldsymbol{K} \cdot (\boldsymbol{r}_i - \boldsymbol{r}_j)} > = z < \sideset{}{'}\sum_{i} e^{i\boldsymbol{K} \cdot \boldsymbol{r}_i} > = z \cdot \mathcal{N} \int d^3 r \, g_2(\boldsymbol{r}) e^{i\boldsymbol{K} \cdot \boldsymbol{r}} = \tag{42}$$

$$\equiv z \cdot \mathcal{N} \cdot \int d^3 r (g_2(\boldsymbol{r}) - 1) e^{i\boldsymbol{K} \cdot \boldsymbol{r}} \text{ for } \boldsymbol{K} \neq 0$$

where $\boldsymbol{K} = (0, \boldsymbol{k})$. With the common definition [20] of the structure factor

$$\hat{S}(\boldsymbol{K}) - 1 = \mathcal{N} \int d^3 r (g_2(\boldsymbol{r}) - 1) e^{i\boldsymbol{K} \cdot \boldsymbol{r}}, \tag{43}$$

(41) reads

$$z\Delta P_2 = Nx\frac{1}{S}\sum_{\boldsymbol{k}}{}'|\sigma(\boldsymbol{k})|^2(\hat{S}(\boldsymbol{k})-1) = Nx\frac{1}{(2\pi)^2}\int d^2k\,|\sigma(\boldsymbol{k})|^2(\hat{S}(\boldsymbol{k})-1)\,, \quad (44)$$

where $\hat{S}(\boldsymbol{k})$ stands for $\hat{S}((0,\boldsymbol{k}))$.

For third-order correlations, eqs. (24) and (41) yield

$$z\Delta P_3 = Nx\frac{1}{(2\pi)^4}\int d^2k \int d^2k' \cdot \sigma(\boldsymbol{k})\sigma(\boldsymbol{k}')\sigma^*(\boldsymbol{k}+\boldsymbol{k}')\cdot(\hat{S}(\boldsymbol{k},\boldsymbol{k}')-1)\,, \qquad (45)$$

where

$$\hat{S}(\boldsymbol{K},\boldsymbol{K}')-1 = N^2\int d^3r\,e^{i\boldsymbol{K}\cdot\boldsymbol{r}}\cdot\int d^3r'\,e^{i\boldsymbol{K}'\cdot\boldsymbol{r}'}\cdot[g_3(\boldsymbol{r};\boldsymbol{r}')-1]\,, \qquad (45a)$$

and $N^2g_3(\boldsymbol{r};\boldsymbol{r}')d^3r\,d^3r'$ is probability to find atoms in $(\boldsymbol{r},d^3r)$ and $(\boldsymbol{r}',d^3r')$ if there is one in $\boldsymbol{r}=0$. Higher-order correlations form correspondingly.

These results are readily generalized to the multitude of events discussed in the foregoing section. By comparison with (9a) and (13a) we find instead of (44)

$$z\Delta P_{2(IJ)\alpha\beta} = Nx\frac{1}{(2\pi)^2}\int d^2k\,\sigma_{(I)\alpha}(\boldsymbol{k})\cdot\sigma^*_{(J)\beta}(\boldsymbol{k})(\hat{S}_{(IJ)}(\boldsymbol{k})-1)\,, \qquad (46)$$

where

$$\hat{S}_{(IJ)}(\boldsymbol{K})-1 = N\int d^3r\,(g_{2(IJ)}-1)\cdot e^{i\boldsymbol{K}\cdot\boldsymbol{r}} \qquad (46a)$$

and $Nc_{(J)}g_{2(IJ)}(\boldsymbol{r})d^3r$ is the probability to find an atom of type J in $(\boldsymbol{r},d^3r)$ if there is an atom of type I in $\boldsymbol{r}=0$.

Formal evaluation of eq. (42) for an ideal crystal yields

$$\sum_{i\neq j} e^{i\boldsymbol{K}\cdot(\boldsymbol{r}_i-\boldsymbol{r}_j)} = z^2\sum_{\boldsymbol{Q}}\delta_{\boldsymbol{K},\boldsymbol{Q}}\,, \qquad (47)$$

where $\boldsymbol{Q}$ runs over the reciprocal lattice[21] of the structure in question. Consequently, one would find from (41)

$$z\Delta P_2 = N^2x^2\sum_{\boldsymbol{q}}{}'|\sigma(\boldsymbol{q})|^2 \qquad (47')$$

where $Q = (0, q)$. Thus, long-range order causes an x^2-dependence of $z \cdot \Delta P_2$ on target thickness which is essentially different from what was found in eq. (45). At the same time, the validity of the various assumptions entering (47′) is by no means proven. Indeed, ignoring governed motion in crystals in the discussion of the effects of long-range order is hardly justified. Therefore, the following discussion will concern systems with short-range order only.

For very thin targets, surface effects become important in the structural correlations, so eq. (44) is no longer obvious. As an extreme example, take a planar layer characterized by a correlation $\hat{g}_2(\varrho) - 1$. Then, (41) yields

$$z \Delta P_2 = n \frac{1}{(2\pi)^2} \int d^2 k \, |\sigma(\boldsymbol{k})|^2 \cdot n \int d^2 \varrho \, (\hat{g}_2(\varrho) - 1) \, e^{i\boldsymbol{k} \cdot \varrho}, \tag{48}$$

where $n = z/S$. The only noticeable difference between (48) and (44) lies in the quantitative behavior of the two pair distribution functions.

It may be worthwhile to stress that the description put forward in this section is based on the assumption that individual events are correlated *only* through the trajectory of the projectile. In particular, the characterization of the elementary event by a probability $P(\boldsymbol{p})$, as expressed by eqs. (38) and (40), rather than a quantal transition amplitude ignores quantal interference between different events. Thus, the treatment excludes coherent scattering processes (in the language of diffraction theory[18]). Also, it is implied that the basic event is defined in a manner so that individual interaction regions do not overlap substantially.

5. Examples: Molecular Gas, Hard-Sphere Gas, and Amorphous Solid

A very simple case of a medium with short-range order is an ideal gas consisting of randomly oriented diatomic molecules. When viewed as an assembly of atoms, such a system has pair correlations only, and the correlation function can be approximated by

$$g_2(\boldsymbol{r}) - 1 = \frac{\delta(r - d)}{4\pi N d^2} \tag{49}$$

in the simplest case of a homonuclear molecule with individual atoms assumed spherically symmetric, and vibrational motion around the equilibrium distance d ignored. (N is the number of *atoms* per volume). Then, (43) and (44) yield

$$\hat{S}(\boldsymbol{K}) - 1 = \frac{\sin Kd}{Kd} \tag{49a}$$

and

$$z\Delta P_2 = Nx\frac{1}{2\pi d} \int_0^\infty dk\,|\sigma(k)|^2\sin kd\,; \tag{50}$$

Expressions of this type have been investigated previously[9,10]. A very important limiting case is that of short-range interaction, where the cross section (38) is made up predominantly from contributions within the range $p \ll d$. Setting

$$P(\boldsymbol{p}) \sim \sigma\cdot\delta(\boldsymbol{p}) \tag{51}$$

in (38a), one obtains from (50)

$$z\Delta P_2 \sim Nx\cdot\frac{\sigma^2}{2\pi d^2} \tag{52}$$

It is easily shown[9,10] that (52) is the leading term in an asymptotic expansion of (50) in terms of $\sigma/2\pi d^2$. Thus, the short-range approximation applies for $\sigma \ll 2\pi d^2$; this means that correlation effects are supposed not to be dominating.

It may be worthwhile to stress that correlations in the ideal molecular gas originate in the (deliberately taken) view of the system as an assembly of atoms. If, instead, the description is based on projectile-molecule collisions as the basic event, Poisson statistics applies. Indeed, previous derivations[9,10] of the present results were found along that line which is, in fact, more direct in case of the molecular gas, but much less general. Thus, with regard to the present description, the molecular gas constitutes mainly a useful test case.

As another illustrative example, consider a real gas, e.g. a noble gas with a hard-sphere-like repulsive interaction, so that

$$g_2(\boldsymbol{r}) \cong \begin{cases} 0 \\ 1 \end{cases} \text{for} \begin{array}{l} r < b \\ r > b \end{array} \tag{53}$$

where b is twice the hard-sphere radius. Then, (43) and (44) yield

$$\hat{S}(\boldsymbol{k}) - 1 = -\frac{4\pi N}{k^3}(\sin kb - kb\cos kb) \tag{53a}$$

and

$$z\Delta P_2 = -Nx \cdot 2N \int_0^b r\, dr \int_0^\infty dk\, |\sigma(k)|^2 \sin kr \; ; \tag{54}$$

In the short-range limit, (54) reads

$$z\Delta P_2 \sim -Nx\sigma^2 \cdot 2Nb \, , \tag{55}$$

i.e., ΔP_2 becomes negative. Unlike (52), (55) is a density-dependent correction to $z \cdot P$, as is obvious from the underlying physical model.

For a more general system, eq. (44) reads in the short-range limit

$$z\Delta P_2 \sim Nx \frac{\sigma^2}{(2\pi)^2} \int d^2k\, (\hat{S}(\boldsymbol{k}) - 1) \; ;$$

Let the medium be isotropic, i.e., $g_2(\boldsymbol{r}) \equiv g_2(\boldsymbol{r})$. Then, (43) reads

$$\hat{S}(\boldsymbol{K}) - 1 = N \int_0^\infty 4\pi r^2 dr\, (g_2(r) - 1) \frac{\sin Kr}{Kr} \, , \tag{56}$$

so

$$z\Delta P_2 \sim Nx\sigma^2 \cdot 2N \int_0^\infty dr\, (g_2(r) - 1) \; ; \tag{57}$$

Also this result has been derived previously for a special case[12]. It shows that in the case of short-range interaction, the sign of the correlation term ΔP_2 is determined by the sign of the structural pair correlation function $g_2(r) - 1$. In case of a positive correlation, like (49), ΔP_2 becomes positive, cf. (52), and vice versa for eqs. (53) and (55).
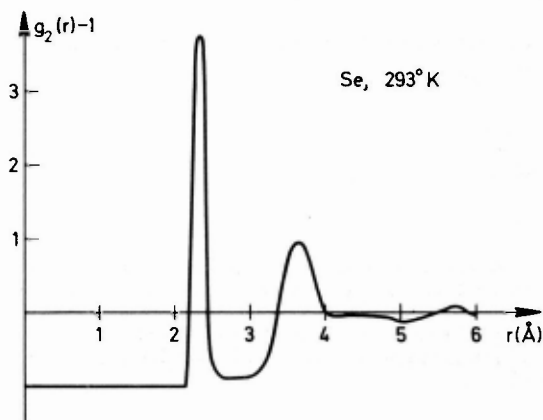

Fig. 1. Pair Correlation for amorphous selenium at room temperature, measured by means of neutron diffraction[22]. In the region $r \lesssim 2\mathring{A}, g_2(r)$ has been set equal to zero, while the function tabulated in ref. 22 oscillates, and takes on negative values in certain intervals. (In the evaluation of eq. (57), however, the tabulated function has been employed).

In a liquid or amorphous solid, the pair correlation function looks generally like in Fig. 1, i.e., both positive and negative values are represented. The most predominant effects are the repulsive interaction at small r, giving rise to a contribution of the type of eq. (54) or (55), and a nearest-neighbor correlation of the type of eq. (50) or (52). However, while in a real gas, $\mathcal{N}$ is so small that (55) is a minute correction, the high density of amorphous matter makes the repulsive region dominate, as is evident from the figure. For amorphous selenium, integration of a measured correlation function[22] yields

$$4\pi\mathcal{N}\int_0^\infty dr\,(g_2(r)-1) = -C\mathring{A}^{-2} \; ;$$

with* $C \simeq 1.3$.

Thus, the resulting correlation term in the short-range limit

$$z\Delta P_2 = -\mathcal{N}x\frac{\sigma^2}{2\pi}\cdot\frac{C}{\mathring{A}^2}$$

is very similar in magnitude but opposite in sign to the one for the diatomic gas, eq. (52).

It is instructive to study the behavior of eqs. (50) and (54) in case the interaction is not of extreme short-range nature like (51). For analytic convenience, take the following model,

$$\sigma(k) = \sigma\cdot e^{-ka} \qquad\qquad (58)$$

corresponding to

$$P(\boldsymbol{p}) = \frac{\sigma a}{2\pi}(p^2+a^2)^{-3/2} \qquad\qquad (58a)$$

according to eq. (38b). Here, a is an arbitrary interaction radius. When (58) is inserted into (50) and (54) one obtains

$$z\Delta P_2 = \frac{\mathcal{N}x\sigma^2}{2\pi(d^2+4a^2)} \qquad\qquad (59a)$$

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* The numerical value of the constant C has been determined by straight summation of the tabulated values of structure factor and pair correlation in ref. 22. Both procedures lead to the same value $C \simeq 1.3$ within their apparent accuracy of $\pm$ 20 pct. The actual error in C may be greater: Because of experimental limitations on the range of k-values covered, the pair distribution $g_2(r)$ as deduced in ref. 22 takes on negative values in certain intervals. This unphysical behavior has not been corrected for in the present evaluation of C.

for the diatomic gas, and

$$z\Delta P_2 = -Nx\sigma^2 \cdot 2\mathcal{N}\left[b - 2a\,\text{arctg}\,\frac{b}{2a}\right] \tag{59b}$$

for the hard-sphere gas.

Eqs. (52) and (55) are limiting cases of (59a) and (59b) for small values of a, as expected. In the opposite limit of $a \gg b, d$, one obtains, instead,

$$z\Delta P_2 \sim \frac{Nx\sigma^2}{8\pi a^2} \tag{60a}$$

for the molecular gas, and

$$z\Delta P_2 \sim -Nx\sigma^2 \cdot \frac{Nb^3}{6a^2} \tag{60b}$$

for the hard-sphere gas.

One may recognize that ΔP_2 *decreases* monotonically when a increases from zero to infinity, i.e., correlations are seemingly most pronounced in the limit of short-range interaction. In interpreting this result, the reader ought to keep in mind that it only holds to the extent that the cross section is kept constant. Most often, σ itself will increase approximately as $\propto a^2$.

It is also obvious that (59b) decreases more rapidly with increasing a than does (59a). Therefore, one may anticipate that in case of an amorphous medium, the relative significance of repulsive and attractive correlations is shifted in the direction of increased importance of positive correlations for long-range interaction. Whether or not this leads to positive values of ΔP_2 depends on the details of the interaction and the structure.

The results derived so far—when suitably extended to more general situations as prescribed in sect. 3—are of immediate significance when fluctuations are to be determined, cf. eq. (13). Higher than pair correlations do not enter here. When interest ist directed toward a frequency spectrum like (26) or the probability density of some cumulative effect, like (31), the significance of higher-order correlations, ΔP_n for $n \geqslant 3$ needs to be discussed.

Let us consider the limit of short-range interaction. Insert (45a) into (45), and apply (51). Then,

$$z\Delta P_3 \sim Nx\sigma^3 \cdot \mathcal{N}^2\int d^3r\int d^3r' \cdot (g_3(\boldsymbol{r}\,;\boldsymbol{r}') - 1)\,\delta(\varrho)\,\delta(\varrho') \tag{61}$$

where $\boldsymbol{r} = (x,\varrho)$ and $\boldsymbol{r}' = (x',\varrho')$.

If the medium is isotropic, the correlation function $g_3(\boldsymbol{r};\boldsymbol{r}') - 1$ is invariant toward rotation of the coordinate system, i.e. it can only depend on $\boldsymbol{r}, \boldsymbol{r}'$, and $|\boldsymbol{r}-\boldsymbol{r}'|$. Therefore, the angular integrations can be carried out, and

$$z\Delta P_3 \sim Nx\sigma^3 \cdot (2N)^2 \int_0^\infty dr \int_0^\infty dr' [\hat{g}_3(r;r') - 1], \tag{62}$$

where

$$\hat{g}_3(r;r') = \frac{1}{2}\left[g_3(r,r',|r-r'|) + g_3(r,r',r+r')\right]; \tag{62a}$$

Thus, we obtain the important result that in the short-range limit, ΔP_3 picks up only correlations between *collinear* atomic positions.

This means that in a polyatomic molecular gas, triple (and higher) correlations are significant only provided that are collinear atoms. The magnitude of these correlations has been investigated in considerable detail in the special case of multiple scattering off triatomic molecules[10]. These estimates will not be repeated here. However, comparison of eq. (62) with eq. (57) shows that the ratio of $\Delta P_3/\Delta P_2$ is of higher order small in terms of $\sigma/2\pi d^2$, in addition to the fact that its numerical value must be small in the absence of any correlation that forces three or more atoms on straight lines.

Thus, it appears that within the range of validity of the short-range approximation, i.e., so long as correlation effects are moderate, pair correlations are responsible for the dominant correction to Poisson statistics, and higher correlations can be ignored in the extended Bothe formula.

In the opposite limit of long-range interaction, such a simple picture can hardly be expected to be true. Indeed, collective effects, and hence higher correlations tend to be important in such situations. It is, therefore, advisable to redefine the basic event and thus to create a description where correlations are moderate or even weak. Such a procedure will be sketched in the following section.

6. Selection of Statistical Variables

In the two previous sections, attention was restricted to collisions between well-localized particles, e.g. projectile ions and target atoms. The (more or less pronounced) deviations from Poisson statistics are in this case directly related with spatial correlations between target atoms. It is worth noting that such spatial correlations only exist when the target atoms interact. Indeed, Poisson statistics does apply in case of an ideal gas, while deviations were found in case of molecular binding, the real gas, and the amorphous solid.

This is a rather general result. Indeed, it appears that at least in case of large cross sections, correlation effects are the rule rather than the exception in particle penetration through non-

random media. However, one has the option of selecting generalized coordinates describing the initiated events in a way so correlations are minimized. Rather than trying to establish general criteria for a wide variety of phenomena, let us consider two simple examples that illustrate this point.

First, take the process of K-shell excitation by an ion beam in an amorphous solid, and ask for the probability G_ν for ν target-K-shell vacancies per ion.

Consider two types of correlation. First, every target atom has two K-electrons, and the respective excitation probabilities may be strongly correlated. Only in the limit of very-high-speed ions would it be feasible to treat this type of correlation within the short-range approximation. Second, consider the spatial correlation between different target atoms. Since, for ions with moderate velocities (comparable with the velocities of K- or L-shell electrons) cross sections for K-excitation can become appreciable on an atomic scale[23], the latter class of correlation may well be sizable. It is, however, weak enough to be well described by the short-range approximation. A third (and often important[23]) correlation effect associated with the projectile state is outside the scope of this paper and therefore ignored.

From the point of view of simplicity of the general statistical description, one might initially attempt to describe the system in terms of one type of target particle (K-electron) and one type of event (K-excitation). Although the two K-electrons are strongly correlated, such a description may be feasible for high-speed ions where Poisson's law is almost fulfilled since spatial correlations are negligible and binding effects small. However, at moderate velocities, in the presence of both types of correlation, one would have to cope with correlations of both third and fourth order. Instead, it is more attractive to apply the (conventional) description in terms of the *atom* as the basic target particle and two types of event, A_1 and A_2, representing single and double K-excitation, respectively. Then, eq. (31) yields

$$G_\nu = \frac{1}{2\pi} \int_0^{2\pi} dt\, e^{it\nu - z\Delta} \tag{63}$$

with

$$\Delta = \frac{1}{S}\left\{\sigma(t) - \sigma^2(t)\cdot\mathcal{N}\int_0^\infty dr(g_2(r) - 1)\right\} \tag{64}$$

and

$$\sigma(t) = (1 - e^{-it})\sigma_1 + (1 - e^{-2it})\sigma_2 \; ; \tag{65}$$

Here, σ_1 and σ_2 are the cross sections for single and double excitation of the K-shell of an individual atom, and $g_2(r) - 1$ is the pair correlation characterizing the structure of the amorphous target. The short-range approximation has been applied to correlations between different target atoms, and the integration over t in (63) has been restricted to an interval size 2π, unlike in (31), in order to produce a probability G_ν instead of a density $G(\nu)d\nu$.

Further evaluation of eq. (63) in case where σ_1 and σ_2 are well-known parameters could be done by way of the procedure discussed in the subsequent section.

Determination of the atomic parameters σ_1 and σ_2 is a problem of atomic physics that has been investigated in specific cases[23-25]. The description of correlations in terms of impact parameters appears too qualitative to be applied in cases where proper quantal calculations are available, but may well be helpful in situations where only empirical cross sections exist. Since the present discussion serves to illustrate a rather specific point, this line is not followed presently.

The second example concerns the energy loss of an ion penetrating a molecular gas target. This problem has been treated previously as a rather simple case where correlations play a role, both from a particle-penetration[9] and a molecular-collisions[26] point of view. The present discussion concentrates on the question of finding the most suitable variables to minimize correlation effects in energy-loss straggling.

It is convenient, for that purpose, to switch to the notation used in previous work[9,26]. We consider the fluctuation in *nuclear* energy loss,

$$\overline{(\Delta E - \overline{\Delta E})^2} = N'xW = N'x \; < \; d^2p(T(p_1) + T(p_2))^2 > \tag{66}$$

of an ion to an ideal gas of homonuclear diatomic molecules. Here, p_1 and p_2 are impact parameters relative to target nuclei, $T(p)$ is the energy transferred to an atom in an individual ion-atom collision at impact parameter p (calculated from classical scattering theory), and N' the number of molecules per volume. W is called the straggling parameter of a molecule. It is readily seen to have the form

$$W = W_1 + W_2 + 2\Delta W_{12} , \tag{67}$$

where

$$W_1 = W_2 = \int d^2p(T(p))^2 \tag{67a}$$

is the straggling parameter of an individual atom, and

$$\Delta W_{12} = < \int d^2p \, T(p_1) \, T(p_2) \; > \tag{68}$$

a correlation term.

It has been shown[9] that ΔW_{12} becomes

$$\Delta W_{12} \sim \frac{S_1 S_2}{2\pi d^2} \tag{69}$$

in the short-range limit, i.e., for large internuclear distance d. Here,

$$S_1 = S_2 = \int d^2p \, T(p) \tag{69a}$$

is the atomic stopping cross section. Conversely, in the long-range limit, i.e., for small d, one finds[9]

$$\Delta W_{12} \sim W_1 - \frac{d^2}{6} \int d^2p \left(\frac{dT}{dp}\right)^2 ; \tag{70}$$

Eq. (69) shows that in the short-range limit, the correlation term ΔW_{12} is small, while according to (70), it is large in the long-range limit. In other words, the statistical variables $T_1 (= T(p_1))$ and T_2 are appropriate to describe the interaction in the short-range limit, but are inconvenient in the opposite case. In a triatomic molecule, or a dense medium, higher correlations would enter essentially.

It is more attractive, for long-range interaction, to split the energy loss according to

$$T_1 + T_2 \equiv T_e + T_i \tag{71}$$

into an elastic and an inelastic part T_e and T_i, respectively, where

$$\mathrm{T}_e = \frac{1}{4M} (\boldsymbol{P}_1 + \boldsymbol{P}_2)^2 , \quad T_i = \frac{1}{4M} (\boldsymbol{P}_1 - \boldsymbol{P}_2)^2 , \tag{71a}$$

and $\boldsymbol{P}_1 = \boldsymbol{P}(p_1)$ and $\boldsymbol{P}_2 = \boldsymbol{P}(p_2)$ are momentum transfers to individual atoms. p_1 and p_2 are vectorial impact parameters, and M is the atomic mass.

The straggling parameter splits correspondingly into

$$W = W_e + W_i + 2\Delta W_{ei} , \tag{72}$$

where W_e and W_i represent the fluctuation in elastic and inelastic energy loss, respectively, and ΔW_{ei} a correlation term.

In appendix A, the following expressions are derived for the three terms,

$$W_e = \left< \int d^2p \, T_e^2 \right> \sim 4W_1 - \frac{d^2}{6} \int d^2p \left(\frac{dT}{dp}\right)^2 , \tag{73a}$$

$$W_i = \left< \int d^2p \, T_i^2 \right> \sim \frac{d^2}{6} \int d^2p \left(\frac{dT}{dp}\right)^2 , \tag{73b}$$

$$\Delta W_{ei} = \left< \int d^2p \, T_e T_i \right> \sim 0 \tag{73c}$$

in the long-range approximation, and

$$W_e \sim W_i \sim \tfrac{1}{2} W_1 + \frac{S_1 S_2}{2\pi d^2} \tag{74a}$$

$$\Delta W_{ei} \sim \tfrac{1}{2} W_1 \tag{74b}$$

in the short-range approximation.

Thus, the correlation is weak in the long-range limit but strong in the opposite case. In other words, the statistical variables T_e and T_i are complementary to T_1 and T_2.

As is indicated in appendix A, the evaluation hinges on the simplifying assumption of small-angle scattering. The present results should, therefore, not be utilized uncritically in molecular-collision physics. The example does, however, illustrate in a very simple way the usefulness of introducing "collective" modes to reduce correlation effects in the long-range approximation.

There is another, essentially independent reason to favor collective coordinates T_e and T_i in case of long-range interaction. The use of the energy-loss functions $T(p_1)$ and $T(p_2)$ in eq. (66) implies that the constituent atoms in the molecule act as free atoms with regard to scattering of the projectile. This approximation, while justified for close collisions, must in general break down for distant collisions because of the distortion of the projectile-atom interaction by valence effects. This does not affect the conclusion that T_e and T_i are appropriate variables for collisions at large impact parameters, but the relation with atomic scattering parameters becomes more complex than indicated in eq. (71a).

7. Evaluation of Cumulative Effects

Before going into applications, let us look at various ways of how to evaluate some cumulative effect U on the basis of eqs. (31) and (32), or (34).

Quite often, one will be satisfied with the average value $\overline{U}$ and the fluctuation $\overline{\Delta U^2} = \overline{(U - \overline{U})^2}$. According to (31) we have

$$\overline{U^n} = \int dU\, U^n G(U) = i^n \frac{\partial^n}{\partial t^n} e^{-z\Delta(t)} \Big|_{t=0}, \tag{75}$$

i.e.,

$$\int dU\, G(U) = 1 ; \tag{76a}$$

$$\overline{U} = z \sum_\alpha u_\alpha P_\alpha ; \tag{76b}$$

$$\overline{\Delta U^2} = z \left(\sum_\alpha u_\alpha^2 P_\alpha + \sum_{\alpha\beta} u_\alpha u_\beta \Delta P_{2,\alpha\beta} \right) ; \tag{76c}$$

These relationships could also have been derived directly from eqs. (8) and (13). Eq. (76b) and the first part of (76c) are the usual relations for mean value and fluctuation[5]. The rest of (76c) is not present in standard penetration theory for random media.

In particular, in case of short-range interaction, eqs. (57) and (76c) yield

$$\overline{\Delta U^2} \cong \mathcal{N}x \left\{ \sum_\alpha u_\alpha^2 \sigma_\alpha + \left(\sum_\alpha u_\alpha \sigma_\alpha \right)^2 \cdot 2\mathcal{N} \int_0^\infty dr (g_2(r) - 1) \right\} \tag{77}$$

for an isotropic medium.

In case of a polyatomic target, we have

$$\overline{U} = z \sum_I c_{(I)} \sum_\alpha u_{(I)\alpha} P_{(I)\alpha} ; \tag{76b'}$$

$$\overline{\Delta U^2} = z \left\{ \sum_I c_{(I)} \sum_\alpha u_{(I)\alpha}^2 P_{(I)\alpha} + \sum_{IJ} c_{(I)} c_{(J)} \sum_{\alpha\beta} u_{(I)\alpha} u_{(J)\beta} \Delta P_{2(IJ)\alpha\beta} \right\} \tag{76c'}$$

according to (34), and

$$\overline{\Delta U^2} \cong \mathcal{N}x \left\{ \sum_I c_{(I)} \sum_\alpha u_{(I)\alpha}^2 \sigma_\alpha + \right. \tag{77'}$$

$$\left. + \sum_{IJ} c_{(I)} c_{(J)} \left(\sum_\alpha u_{(I)\alpha} \sigma_\alpha \right) \cdot \left(\sum_\beta u_{(J)\beta} \sigma_\beta \right) \cdot 2\mathcal{N} \int_0^\infty dr (g_{2(IJ)}(r) - 1) \right\}$$

by means of (46) and (46a) if the short-range approximation is valid. (77') approaches a similar form as (77) only if the medium is disordered, i.e., when $g_{2(IJ)}(r) - 1$ is independent of I and J.

Extension of these relationships to vectorial quantities and to the continuum case is straightforward.

Consider next the case of a discrete probability distribution, as discussed briefly in the foregoing section. For simplicity, let u_α be given by

$$u_\alpha = \nu ; \; \nu = 0, \pm 1, +2, \ldots \tag{78}$$

so

$$U = \sum n_\nu \nu$$

and

$$G_U = \frac{1}{2\pi} \int_0^{2\pi} dt e^{itU - z\Delta(t)} \tag{79}$$

or

$$G_U = \frac{1}{2\pi i} \oint d\zeta \zeta^{-1-U} e^{-z\Delta(\zeta)}$$

with

$$\Delta(\zeta) = \sum_\nu (1 - \zeta^\nu) P_\nu - \tfrac{1}{2} \sum_{\nu\mu} (1 - \zeta^\nu)(1 - \zeta^\mu) \Delta P_{2,\nu\mu} ; \tag{80}$$

The integration over ζ goes along a closed path including the point $\zeta = 0$. Hence,

$$G_U = \frac{1}{U!} \frac{\partial^U}{\partial \zeta^U} e^{-z\Delta(\zeta)} \Big|_{\zeta = 0} ; \tag{81}$$

Thus, exact evaluation of the generalized Bothe formula is possible in this case.

Eq. (81) may become useful in the analysis of ionization phenomena.

Let us return to the more general case of a probability density (31), but apply the diffusion approximation[27], i.e., assume that the bits u_α are "small". Then, (32) reads

$$\Delta \cong it \sum_\alpha u_\alpha P_\alpha + \tfrac{1}{2} t^2 \Big(\sum_\alpha u_\alpha^2 P_\alpha + \sum_{\alpha\beta} u_\alpha u_\beta \Delta P_{2,\alpha\beta} \Big) \tag{82}$$

up to second order in u_α, and (31) yields

$$G(U) = \frac{1}{\sqrt{2\pi \overline{\Delta U^2}}} e^{\frac{-(U - \bar{U})^2}{2 \overline{\Delta U^2}}} \tag{83}$$

with $\bar{U}$ and $\overline{\Delta U^2}$ given by (76b) and (76c). The limitations of the gaussian approximation have been discussed extensively[2,5,17,27].

Finally, let us write eq. (32) in the case of short-range interaction,

$$z\Delta \cong Nx \Big\{ \sum_\alpha (1 - e^{-itu_\alpha}) \sigma_\alpha - \tfrac{1}{2} \Big(\sum_\alpha (1 - e^{-itu_\alpha}) \sigma_\alpha \Big)^2 \cdot 2N \int_0^\infty dr (g_2(r) - 1) \Big\} ; \tag{84}$$

Eq. (84) offers itself for a perturbation treatment[8,10] in case of weak correlation. Indeed, take some representative value of t in the Bothe integral (33), such as the median value $t_m(U)$ defined by

$$\int_{-\infty}^\infty dt e^{itU - Nx\sigma(t)} = 2 \cdot \int_{t_m}^\infty dt e^{itU - Nx\sigma(t)}, \tag{85}$$

and write (84) in the form

$$z\Delta \cong Nx \cdot \sigma(t) \Big\{ 1 - \frac{\sigma(t_m)}{2} \cdot \frac{1}{2\pi d_{eff}^2} \Big\} = Nx_{eff}\sigma(t) \tag{86}$$

with

$$\sigma(t) = \sum_\alpha (1 - e^{-itu_\alpha}) \sigma_\alpha \tag{87a}$$

$$\frac{1}{2\pi d_{eff}} = 2\mathcal{N}\int_0^\infty dr\,(g_2(r) - 1) \tag{87b}$$

and

$$x_{eff} = x\left\{1 - \tfrac{1}{2}\frac{\sigma(t_m)}{2\pi d_{eff}^2}\right\}; \tag{87c}$$

In this manner, for weak correlation, results derived on the basis of Poisson statistics can be modified such as to incorporate correlations.

8. Applications

Stopping of Charged Particles.

Let us consider a standard problem of the theory of particle penetration, the energy-loss spectrum of a charged particle after penetration of a path length x in a stopping medium. According to common notation[1,5,28,29], the individual event is characterized by a spectrum σ_α of energy loss T_α or, in continuum notation, $d\sigma(T)$, so the mean energy loss $\overline{\Delta E}$ and fluctuation $\Omega^2 = (\Delta E - \overline{\Delta E})^2$ are given by (76b) and (76c),

$$\overline{\Delta E} = \mathcal{N}xS; \tag{88}$$

$$\Omega^2 = \mathcal{N}x(W + \Delta W); \tag{89}$$

where

$$S = \sum_\alpha T_\alpha \sigma_\alpha = \int T\,d\sigma(T); \tag{88a}$$

$$W = \sum_\alpha T_\alpha^2 \sigma_\alpha = \int T^2\,d\sigma(T); \tag{89a}$$

$$\Delta W = \frac{1}{(2\pi)^2}\int d^2k\,|S(\boldsymbol{k})|^2(\hat{S}(\boldsymbol{k}) - 1); \tag{89b}$$

and

$$S(\boldsymbol{k}) = \sum_\alpha T_\alpha \sigma_\alpha(\boldsymbol{k}) \tag{90}$$

according to eq. (44). The quantity $S(\boldsymbol{k})$ can also be written in the form

$$S(\boldsymbol{k}) = \int d^2p\,e^{i\boldsymbol{k}\cdot\boldsymbol{p}}\overline{T}(\boldsymbol{p}) \tag{90'}$$

by means of (38a), where

$$\overline{T}(\boldsymbol{p}) = \sum_{\alpha} T_{\alpha} P_{\alpha}(\boldsymbol{p}) = \int dT\, T P(\boldsymbol{p}, T)\,, \tag{90a}$$

and $P(\boldsymbol{p}, T)\, dT$ is the probability for energy loss (T, dT) in a collision at impact parameter $\boldsymbol{p}$. By comparison of (90) with (88a), as well as (38a) with (38), it is evident that

$$S(\boldsymbol{k} = 0) = S\,, \tag{91}$$

and therefore, in the short-range limit,

$$\Delta W \cong \frac{S^2}{(2\pi)^2} \int d^2k\, (\hat{S}(\boldsymbol{k}) - 1) = S^2 \cdot 2N \int_0^{\infty} dr\, (g_2(r) - 1)\,, \tag{92}$$

where the second identity assumes the medium to be isotropic. The latter result has been mentioned in ref. 12.

Specifically, for a diatomic gas, insertion of (49a) into (89b) yields

$$\Delta W = \frac{1}{2\pi d} \int_0^{\infty} dk\, |S(k)|^2 \sin kd\,, \tag{93}$$

a result that has been derived previously by a rather different procedure[9]. The consequences of eq. (93) have been discussed in considerable detail[9,12]. It is, however, worthwhile to repeat that in the short-range approximation, ΔW becomes positive for the molecular gas. Experimental checks[11] have confirmed both the sign and the magnitude of the predicted values of ΔW.

According to the discussion following eq. (57), the value of ΔW in case of an amorphous stopping medium is roughly equal in magnitude, but opposite in sign, to the one for the diatomic gas. For example, for amorphous selenium (cf. footnote on p. 20),

$$\Delta W \cong -\frac{S^2}{2\pi} \cdot \frac{1.3}{\text{Å}^2}\,;$$

For the full energy-loss spectrum $G(\Delta E)\, d(\Delta E)$, eq. (32) yields

$$G(\Delta E) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt\, e^{it\Delta E - z\Delta(t)} \equiv \frac{1}{2\pi i} \int_{c - i\infty}^{c + i\infty} ds\, e^{s\Delta E - z\Delta(-is)} \tag{94}$$

where

$$z\Delta(t) = Nx \left\{ \sigma_{tr}(t) - \tfrac{1}{2} \cdot \frac{1}{(2\pi)^2} \cdot \int d^2k\, |\sigma_{tr}(t, \boldsymbol{k})|^2 (\hat{S}(\boldsymbol{k}) - 1) \right\} \tag{95}$$

and

$$\sigma_{tr}(t, \boldsymbol{k}) = \sum_\alpha (1 - e^{-itT_\alpha}) \sigma_\alpha(\boldsymbol{k}) \; ; \tag{95a}$$

$$\sigma_{tr}(t) \equiv \sigma_{tr}(t, \boldsymbol{k} = 0) \; ; \tag{95b}$$

The second version of eq. (94) is a generalization of Landau's formula[4] to a nonrandom stopping medium. In the short-range limit, for an isotropic medium, eq. (95) simplifies to

$$z\Delta(t) = Nx \left\{ \sigma_{tr}(t) - [\sigma_{tr}(t)]^2 \cdot \mathcal{N} \int_0^\infty dr(g_2(r) - 1) \right\} \; ; \tag{96}$$

When (95) is evaluated in the diffusion approximation, eq. (82), (94) takes the form of a gaussian with the width Ω^2, eq. (89), centered around $\overline{\Delta E}$, eq. (88). Expansion up to third order in t yields an extension of Vavilov's formula[30],

$$z\Delta(t) \cong Nx \{ itS + \tfrac{1}{2} t^2 (W + \Delta W) - \tfrac{1}{6} i t^3 (Q + \Delta Q) \} \tag{97}$$

where

$$Q = \sum T_\alpha^3 \sigma_\alpha = \int T^3 d\sigma(T) \; ; \tag{98}$$

and

$$\Delta Q = 3 \cdot \frac{1}{(2\pi)^2} \int d^2 k (\hat{S}(\boldsymbol{k}) - 1) \cdot \mathrm{Re}\, S(\boldsymbol{k}) W^*(\boldsymbol{k}) \; ; \tag{99}$$

In particular, in the short-range approximation, we have

$$\Delta Q \cong 3SW \cdot 2\mathcal{N} \int_0^\infty dr(g_2(r) - 1) \tag{99'}$$

and therefore

$$\frac{\Delta Q/Q}{\Delta W/W} \cong 3 \frac{W^2}{QS} \; , \tag{100}$$

independent of the actual correlation function. The dimensionless ratio (100) is of the order of ~ 1, cf. eqs. (88a), (89a), and (98). Therefore, the skewness correction $\Delta Q/Q$ must normally be suspected to be of the same order of magnitude as the straggling correction $\Delta W/W$.

After insertion of (97) into (94) one obtains

$$G(\Delta E) \cong \frac{1}{2\pi} \int_{-\infty}^\infty dt\, e^{-\frac{t^2}{2} Nx(W + \Delta W)} \cdot \cos\{t(\Delta E - NxS) + \tfrac{1}{6} t^3 Nx(Q + \Delta Q)\} \tag{101}$$

or, in terms of Airy functions[16], the usual expression[29,30] with W replaced by $W + \Delta W$ and Q by $Q + \Delta Q$.

With regard to experimental checks of correlation effects on energy loss, straggling measurements on amorphous solids are desirable, preferably in targets where the pair correlation or the structure factor are known with sufficient accuracy, so that at least one of the two integrals in eq. (92) can be evaluated. As was the case in molecular targets, one might first wish to trace the energy region where $|\Delta W/W|$ is largest, i.e., where the stopping power has its maximum. In this region, the significant contributions to the stopping power originate from collisions at impact parameters $\lesssim a_0 v/v_0$ with $v \sim v_0$ for light ions,[5] where a_0 and v_0 are the Bohr radius and Bohr velocity. Therefore, the short-range approximation appears acceptable.

As a word of caution, one may add that the straggling and skewness parameters occurring in this discussion are to be understood strictly as defined through the second and third moment over an energy-loss distribution integrated over all scattering angles at a well-defined penetration depth. They are not necessarily related in a simple manner with the half-width of an energy-loss distribution integrated over a limited range of scattering angles. However, the problem of finding such a relationship[5] is only loosely connected with the effect of correlations on straggling, and therefore not discussed in more detail in this context.

Multiple Scattering

The problem of small-angle multiple scattering is a two-dimensional analog of the problem of energy loss, except for the fact that energy loss is a one-way process[31]. For a recent summary, the reader is referred to ref. 12.

In accordance with the notation applied in ref. 12, consider a series of scattering processes at small angles $\vec{\varphi}$, and take $\vec{\varphi}$ as a vector in a plane perpendicular to the beam. Then, the angular distribution after path length x reads, according to (31),

$$G(\alpha)\, d^2\alpha = \frac{d^2\alpha}{(2\pi)^2} \int d^2\varkappa\, e^{i\varkappa \cdot \alpha - z\Delta(\varkappa)}, \tag{102}$$

where, by means of (32) and (44)

$$z\Delta(\varkappa) = Nx\left\{ \sigma_0(\varkappa) - \tfrac{1}{2} \cdot \frac{1}{(2\pi)^2} \cdot \int d^2k\, (\hat{S}(\boldsymbol{k}) - 1)\, \sigma(\varkappa, \boldsymbol{k})\, \sigma(\varkappa, -\boldsymbol{k}) \right\}, \tag{103}$$

and

$$\sigma(\mathbf{x},\mathbf{k}) = \sum_{\vec{\varphi}} (1 - e^{-\mathbf{x}\cdot\vec{\varphi}}) \sigma_{\vec{\varphi}}(\mathbf{k}) \equiv \sum_{\vec{\varphi}} \int d^2 p\, P_{\vec{\varphi}}(\mathbf{p}) e^{i\mathbf{k}\cdot\mathbf{p}}(1 - e^{-i\mathbf{x}\cdot\vec{\varphi}}) ; \tag{104a}$$

$$\sigma_0(\mathbf{x}) \equiv \sigma(\mathbf{x},0) ; \tag{104b}$$

Multiple scattering is normally dominated by nuclear collisions[5]. Therefore, application of the short-range approximation appears well justified[10], and

$$z\Delta(x) \cong Nx\left\{\sigma_0(\mathbf{x}) - [\sigma_0(\mathbf{x})]^2 \cdot N \int_0^\infty dr(g_2(r)-1)\right\} ; \tag{105}$$

The transport cross section $\sigma_0(\mathbf{x})$ simplifies to

$$\sigma_0(\mathbf{x}) = \int d\sigma(\varphi)(1 - \mathcal{J}_0(x\varphi)) \equiv \sigma_0(x) \tag{104b'}$$

in case of azimuthally symmetric scattering. Similarly, (102) reads

$$G(\mathbf{\alpha}) \equiv G(\alpha) = \frac{1}{2\pi} \int_0^\infty x dx \mathcal{J}_0(x\alpha) e^{-z\Delta(x)} ; \tag{106}$$

Eq. (106) is Bothe's formula in the standard form applying to small-angle scattering. $\mathcal{J}_0$ is the zero-order Bessel function of the first kind.

Eq. (105) can be discussed along the same lines as eq. (96). In the particular case of a diatomic molecule, (105), (106), and (49) yield

$$G(\mathbf{\alpha}) \cong \frac{1}{2\pi} \int_0^\infty x d x \mathcal{J}_0(x\alpha) \cdot e^{-Nx\{\sigma_0(x) - \frac{1}{2}(\sigma_0(x))^2/2\pi d^2\}} , \tag{107}$$

a result that has been derived previously by a different procedure; it has also been evaluated explicitly, and confirmed experimentally[8,10].

It is an interesting feature of eq. (107) that it predicts a *smaller* half-width of the multiple-scattering profile for a diatomic gas, in comparison with the equivalent random medium. Since, in the short-range limit, the sign of the correction depends on the factor $2N\int_0^\infty dr(g_2(r)-1)$, cf. eq. (57), it may readily be concluded that for an amorphous target, the multiple scattering profile is *broader* than the profile for an otherwise equivalent random medium.

However, correlation effects on the multiple-scattering half-width in molecular gases have been found to be pronounced only at extremely small target thicknesses which are hard to achieve experimentally in solids. Therefore, these effects are important only in case of very thin amorphous layers ($\ll 100\,\mathring{A}$).

Appendix A. Derivation of eqs. (73) and (74)

According to (71a) we have

$$T_{\{ {}^e_i \}} = \tfrac{1}{2}(T_1 + T_2) \pm \frac{\boldsymbol{P}_1 \cdot \boldsymbol{P}_2}{2M} \tag{A1}$$

and hence

$$W_{\{ {}^e_i \}} = \; <\!\int d^2 p \, T_{\{ {}^e_i \}}^{\;2}\!> \; = \tfrac{1}{4} \; <\!\int d^2 p \,(T_1 + T_2)^2\!> \; + X_1 \pm X_2 \tag{A2}$$

$$\Delta W_{ei} = \; <\!\int d^2 p \, T_e \, T_i\!> \; = \tfrac{1}{4} \; <\!\int d^2 p \,(T_1 + T_2)^2\!> \; - X_1 \tag{A3}$$

where

$$X_1 = \frac{1}{4M^2} \; <\!\int d^2 p \,(\boldsymbol{P}_1 \cdot \boldsymbol{P}_2)^2\!> \tag{A4}$$

and

$$X_2 = \frac{1}{4M^2} \; <\!\int d^2 p \,(P_1^{\;2} + P_2^{\;2})(\boldsymbol{P}_1 \cdot \boldsymbol{P}_2)\!> \; ; \tag{A5}$$

Let us assume small-angle scattering to be dominating. Then, the momentum transfer is approximately perpendicular to the ion trajectory, or

$$\boldsymbol{P}_1 \equiv \boldsymbol{P}(\boldsymbol{p}_1) \cong \frac{\boldsymbol{p}_1}{p_1} P(p_1), \tag{A6}$$

and correspondingly for $\boldsymbol{P}_2$.

Intergrals are evaluated within the following scheme[9,10],

$$<\!\int d^2 p f(\boldsymbol{p}_1, \boldsymbol{p}_2)\!> \; = \int d^2 p_1 \int d^2 p_2 f(\boldsymbol{p}_1, \boldsymbol{p}_2) \; <\!\delta(\boldsymbol{p}_1 - \boldsymbol{p}_2 - \boldsymbol{b})\!> \; ,$$

where $\boldsymbol{b}$ is the projection of the molecular axis on a plane perpendicular to the ionic trajectory. Moreover,

$$<\!\delta(\boldsymbol{p}_1 - \boldsymbol{p}_2 - \boldsymbol{b})\!> \; \sim \; \frac{1}{2\pi d^2} + \mathrm{O}\!\left\{ \frac{1}{d^4} \right\} \tag{A7a}$$

for short-range interaction, and

$$<\!\delta(\boldsymbol{p}_1 - \boldsymbol{p}_2 - \boldsymbol{b})\!> \; \sim \; \delta(\boldsymbol{p}_1 - \boldsymbol{p}_2) + \frac{d^2}{6} \vec{\nabla}^2_{\boldsymbol{p}_1} \delta(\boldsymbol{p}_1 - \boldsymbol{p}_2) + \mathrm{O}\{d^4\} \tag{A7b}$$

for long-range interaction. After carrying out the integrations one finds

$$X_1 \sim \begin{cases} \dfrac{S_1^{\,2}}{4\pi d^2} & \text{(short-range)} \\[2em] W_1 - \dfrac{d^2}{12}\int d^2p\left(\dfrac{dT}{dp}\right)^2 & \text{(long-range)} \end{cases} \qquad\qquad\text{(A8)}$$

and

$$X_2 \sim \begin{cases} 0 & \text{(short-range)} \\[2em] 2W_1 & \text{(long-range)} \end{cases} \qquad\qquad\text{(A9)}$$

apart from terms that are small of order d^{-4} or d^4, respectively. After inserting X_1 and X_2 into (A2) and (A3), and applying (69) and (70), one finds (73) and (74).

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