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Scattering and Stopping of Swift Diatomic Molecules under Coulomb Explosion

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The scattering and stopping of the fragments of a fast diatomic molecule under Coulomb explosion has been analysed theoretically. The central assumption in the scheme is the dominance of Coulomb explosion, while electronic stopping (including wake forces) and elastic scattering are treated as perturbations. Charge exchange has been neglected.

Coulomb images of penetration phenomena are heavily distorted. For small penetrated layer thicknesses, images appear contracted in the direction of the molecular axis, and expanded perpendicular to it. This distortion is described quantitatively by a linear transformation.

General expressions have been derived for the effect of continuous and stochastic forces on the distribution of fragment velocities from Coulomb explosion (the "ring pattern"). Moreover, relations have been found that allow to scale velocity distributions valid in the absence of Coulomb explosion into distributions allowing for Coulomb explosion.

Applications concern the shift in ring pattern due to electronic stopping, the lateral broadening due to multiple scattering, and the effect of zero-point motion on the Coulomb image of a molecule.

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Introduction

This work addresses the penetration of fast molecules at projectile speeds in the electronic-stopping region, i.e., typically in the upper keV or MeV energy range. Such molecules dissociate when entering the target region and get stripped of all or part of their electrons. Therefore, the fragment ions repel each other via their mutual Coulomb forces. They can be detected, and their velocities can be measured [1] and may be utilized for imaging purposes. If the structure of the incoming molecular ion is known one may image penetration phenomena like wake forces and multiple scattering [2,3]. If the structure is unknown, the Coulomb image can be utilized to reveal it in considerable detail [4].

Coulomb explosion images can have excellent resolution but the imaging is far from uniform. This has been pointed out repeatedly in the literature [5,3,6], but a general quantitative treatment is missing. It is the purpose of the present work to discuss the problem in fairly general terms, to outline a quantitative description, and to derive pertinent scaling laws for the specific case of diatomic molecules.

What is the Problem?

Figure 1a shows a particular geometry discussed by Plesser [3]. A diatomic molecule AB is oriented parallel to its direction of motion, i.e., the direction of the beam. Let one of the atoms, say A, receive a little impulse perpendicular to the beam. This could be caused by small-angle scattering on a target atom. If the atoms A and B were noninteracting, A would appear scattered laterally while B would appear undeflected. In the presence of a repulsive Coulomb force between A and B, both atoms

receive velocity increments in opposite directions along the molecular axis. That axis, however, rotates since the initial impulse transfers angular momentum to the molecule. The angle of rotation will be finite because angular momentum is conserved while the distance grows. Ultimately, the molecular axis will make some nonvanishing angle with the beam and hence, the velocity change due to Coulomb explosion will have a component perpendicular to the beam. As a result, atom B will appear deflected opposite to the initial kick, and atom A will be scattered at a *larger* angle than expected.

The important feature is that neither Coulomb explosion nor the scattering event alone would lead to lateral deflection of atom B. It is the interference of the two effects that generates the peculiar behavior.

Is this a special feature for a particular geometry? Well, look at the same situation again but with the molecule aligned perpendicular to the beam (fig. 1b). For noninteracting atoms A and B, the signal observed at a detector would not differ from the one in figure 1a. In the presence of Coulomb explosion, the primary effect will be the lateral deflection of the particles in opposite directions due to the Coulomb force. The impulse given to atom A will in part go to center-of-mass motion, i.e., it will be shared by A and B in accordance with their masses. The portion going into relative motion raises the internal energy and increases the terminal velocity of Coulomb explosion. Thus, atom A will receive an additional deflection *opposite* to the initial impulse, while B is deflected even more. Again an interference between impulse and Coulomb explosion, but now with the sign switched.

The *cause* of the impulse is immaterial for the above considerations. The point to be made is that the effect of *any* force on the motion of a molecule is strongly affected by the presence of Coulomb explosion.

Dominating Coulomb Explosion

In typical molecular-ion experiments involving thin penetrated foils, Coulomb explosion is by far the dominating feature. This shows up in the characteristic ring patterns [1,2] of the distribution of fragment velocities relative to the beam. For diatomic molecules with a definite internuclear distance, oriented at random, the velocities would lie on two concentric spheres in the absence of disturbing effects.

In practice, the initial internuclear distance obeys a statistical distribution, molecules experience scattering, stopping, and wake forces, and their velocities due to zero-point motion may be significant. These effects distort the ring pattern but they do not wipe it out, except for long penetrated pathlengths. Therefore, the approach to be outlined here is to treat the Coulomb explosion rigorously and to consider other ongoing effects as perturbations.

Fragment ions may experience charge exchange. Since charge exchange affects the repulsive Coulomb force, it is not in general a weak perturbation. It will be excluded here.

For clarity, the treatment will presently be restricted to diatomic ions.

Effect of an Impulse, Seen from the Molecule

Undisturbed Coulomb explosion is governed by energy conservation,

$$v = dr/dt = v_{\infty} [1 - V(r)/V(D)]^{1/2} , \quad (1)$$

where v is the relative velocity, $r = r(t)$ the internuclear distance, D the initial distance (at $t = 0$), and $V(r)$ a repulsive central-force potential allowing for screening. v_∞ is the terminal speed determined by $V(D) = M_0 v_\infty^2 / 2$, with M_0 being the reduced mass.

Let the molecule experience a sudden change in the velocity of relative motion by an increment Δv at some time $t \geq 0$. Let Δv first be directed parallel to the molecular axis. From energy conservation, the terminal velocity changes to $v_\infty + \Delta v(\infty)$ with

$$\Delta v(\infty) = \Delta v [1 - V(r)/V(D)]^{1/2} \equiv P(t) \Delta v \quad (2)$$

where r is the internuclear distance at the time of the impulse. Eq. (2) is the leading term in a Taylor expansion in Δv . Terms of higher order will always be ignored in the following. The dimensionless parameter $P(t)$, as defined by eq. (2), is a direct measure of the distortion of the Coulomb image of Δv for this particular geometry.

Consider now an impulse Δv given perpendicular to the molecular axis at some time $t \geq 0$. It is easily verified that up to first order in Δv , the Coulomb explosion is unaffected. However, the molecular axis rotates, with the angle of rotation determined by angular momentum conservation,

$$\phi(\infty) = r \Delta v \int_t^\infty \frac{1}{r(t')^2} dt' \quad (3)$$

By means of eq. (1), the lateral velocity at $t = \infty$ reads

$$\Delta v(\infty) = v_\infty \phi(\infty) = \Delta v r \int_t^\infty \frac{dr'}{r'^2 [1 - V(r')/V(D)]^{1/2}} \equiv Q(t) \Delta v, \quad (4)$$

with a dimensionless quantity $Q(t)$ defined by eq. (4). Again, (4) is the leading term in a Taylor expansion in Δv .

Fig. 2 shows P and Q for unscreened Coulomb interaction. It is seen that both P and Q approach 1 for $r \gg D$, i.e., if Δv is imparted at a time when the Coulomb explosion is essentially finished. Pronounced distortions appear when $t \approx 0$, i.e., $r/D = 1$: An impulse parallel to the axis given at $t = 0$ will not show up in the Coulomb image to first order. Conversely, a lateral impulse will show up twice as large as "expected". Clearly, "the Coulomb force acts like a strongly astigmatic lens" [6].

Application: Zero-Point Motion of a Diatomic Molecule

Consider "pure Coulomb explosion" of a diatomic molecule, i.e., neglect penetration phenomena but take full account of the zero-point motion of the molecule in its initial state. Does the Coulomb image provide information about that state, i.e., the spatial configuration and the distribution of velocities?

The internuclear distance D affects the Coulomb explosion to zero'th order, cf. eq. (1). Evidently, its distribution is one of the prime parameters measured by the technique [4]. The initial motion can be envisaged as a velocity increment given to the molecule at $t = 0$. It follows from fig. 2 that the vibrational motion will not be visible because $P(t=0) = 0$, while the rotational motion will be imaged. However, an observer who is unaware of the content of fig. 2 is likely to extract rotational velocities from experimental data that are too high by a factor of 2, i.e., to overestimate rotational energies by a factor of four.

Including rotational motion in simulating Coulomb images was empirically found important long ago [3]. No argument was given why vibrational motion should be less important. Figure 2 provides the reason. A generalization to polyatomic molecules would evidently be of interest.

Effect of an Impulse, Seen from the Beam

Consider a diatomic molecule with its axis oriented along $\underline{\Omega} = (\cos\Phi, \sin\Phi\cos\chi, \sin\Phi\sin\chi)$ so that Φ is the angle between the molecule and the beam, the latter defining the x-axis.

Let the molecule receive a velocity increment $\underline{\Delta v}$ at some time $t = 0$. Splitting $\underline{\Delta v}$ into components parallel and perpendicular to the molecular axis we may summarize the effect on the Coulomb image in the form

$$\underline{\Delta v}(\infty) = P (\underline{\Delta v} \cdot \underline{\Omega}) \underline{\Omega} + Q [\underline{\Delta v} - (\underline{\Delta v} \cdot \underline{\Omega}) \underline{\Omega}] , \quad (5)$$

or

$$\underline{\Delta v}(\infty) = \underline{T}(r) \cdot \underline{\Delta v} , \quad (6)$$

by introduction of a tensor $\underline{T}$ with the elements

$$T_{ij} = Q \delta_{ij} + (P-Q) \Omega_i \Omega_j \quad \text{for } i, j = 1, 2, 3. \quad (7)$$

Eq. (6) is a fundamental relationship for Coulomb imaging of diatomic molecules. It provides a linear link between the object and the image, but the reconstruction of the image is complicated firstly since $\underline{T}$ is a tensor, and secondly through the dependence on the Coulomb-exploded distance r .

Continuous Forces

Let a diatomic molecule be acted upon by some continuous force such as the electronic stopping power or the interaction with a target surface. Over a time interval $(t, \Delta t)$, this force imparts a

velocity increment $\Delta v = (\underline{F}_A/M_A - \underline{F}_B/M_B)\Delta t$ to the relative motion of the molecule if the force can be separated into individual forces on atoms A and B. If these forces are weak compared to the Coulomb force, their effect on the Coulomb image will be given by

$$\Delta v(\infty) = \int_0^{\tau} dt \underline{T}(t) \cdot (\underline{F}_A(t)/M_A - \underline{F}_B(t)/M_B) \quad (8)$$

where $t=0$ specifies the start of Coulomb explosion and τ the time when the forces cease to be acting, i.e., the dwell time in the target.

If the forces are independent of time, the integration in (8) affects only $P(t)$ and $Q(t)$. After definition of effective values P' and Q' by the relations

$$P' = \frac{1}{\tau} \int_0^{\tau} P(t) dt ; \quad Q' = \frac{1}{\tau} \int_0^{\tau} Q(t) dt , \quad (9)$$

eq. (8) reduces to

$$\Delta v(\infty) = \tau \underline{T}' \cdot (\underline{F}_A/M_A - \underline{F}_B/M_B) , \quad (10)$$

where $\underline{T}' = \underline{T}(P', Q')$ in eq. (7).

P' and Q' for unscreened Coulomb interaction are shown in fig. 3. It is seen that the transverse effect, expressed by $\int Q dt$, dominates heavily over a wide range of values of r/D .

Application: Shift in Ring Pattern due to Stopping

Consider a heterogeneous diatomic molecule AB with atomic stopping powers $(dE/dx)_A$ and $(dE/dx)_B$, respectively. In the absence of other disturbances, the relative velocity will be given by

$$\underline{v}(\infty) = v_{\infty} \underline{\Omega} + \underline{\Delta v}(\infty) = v_{\infty} \underline{\Omega} - \Delta v' \underline{T}' \cdot \underline{e}_x, \quad (11)$$

where $v_{\infty} \underline{\Omega}$ is the velocity vector for undisturbed Coulomb explosion, $\underline{e}_x$ a unit vector in the beam direction and

$$\Delta v' = \tau \left[\frac{1}{M_A} \frac{dE}{dx} \Big|_A - \frac{1}{M_B} \frac{dE}{dx} \Big|_B \right]. \quad (12)$$

The observable ring pattern is found by averaging over the distribution of initial orientations which is assumed to be uniform,

$$F(\underline{v}) = \frac{1}{4\pi} \int_0^{2\pi} \sin\Phi \, d\Phi \int_0^{\pi} d\chi \, \delta(\underline{v} - \underline{v}(\infty)) \quad (13)$$

and reads

$$F(\underline{v}) = \frac{1}{4\pi v_{\infty}^2} \delta(v - v_{\infty} + P' \Delta v' \cos\theta) \quad (14)$$

where θ is the angle of observation against the x-axis.

Eq. (14) represents a spherical distribution shifted along the beam axis (fig. 4). The interesting feature is the occurrence of the factor P' . It implies that this shift, real as it must be, will appear suppressed in the Coulomb image if the target is thin.

Stochastic Forces

Consider now the cumulative effect of a multitude of actions coming at random, i.e., a sequence of velocity increments $\underline{\Delta v}_i$ given at times t_i ,

$$\underline{\Delta v}(\infty) = \sum_v \underline{T}(t_v) \cdot \underline{\Delta v}_v . \quad (15)$$

The statistical distribution of terminal velocities is then given by

$$F(\underline{v}) = \frac{1}{(2\pi)^3} \int d^3k e^{i\underline{k} \cdot (\underline{v} - \underline{v}_\infty \underline{\Omega})} \exp[-N v_0 \int_0^{\tau} dt \sigma(\underline{k}, t)] \quad (16)$$

with

$$\sigma(\underline{k}, t) = \int d\sigma(\underline{\Delta v}, t) [1 - e^{-i\underline{k} \cdot \underline{T}(t) \cdot \underline{\Delta v}}] . \quad (17)$$

In the absence of Coulomb explosion ($v_\infty = 0$ and $\underline{T}$ reducing to the unit tensor), (16) and (17) reduce to the conventional Bothe-Landau formula in the theory of energy loss and multiple scattering [7], with v_0 being the beam velocity, N the density of scatterers, and $d\sigma(\underline{\Delta v})$ the differential cross section. The underlying physical assumption is the statistical independence of different scattering events. When Coulomb explosion is added, the zero-point in the velocity distribution becomes the undistorted Coulomb velocity $v_\infty \underline{\Omega}$, and the velocity increments $\underline{\Delta v}$ in the individual events need to be replaced by their Coulomb images $\underline{T}(t) \cdot \underline{\Delta v}$. The time dependence in the cross section in (17) originates in the time dependence of the internuclear distance $r(t)$. It is of very minor importance except for thick targets.

Fundamental Scaling Relation

Eqs. (16) and (17) can be evaluated in a variety of ways [8]. Here, attention will be given to the question of how the velocity distribution in the presence of Coulomb explosion relates to what would be measured in the absence of Coulomb explosion.

Following the step leading from (8) to (10), the exponent in (16) will be approximated according to

$$\int_0^{\tau} dt \sigma(\underline{k}, t) \approx \tau \sigma(\underline{k}, P', Q') . \quad (18)$$

Eq. (18) is asymptotically exact at low thicknesses, and its accuracy at moderate values of τ [8] hinges on the fact that P is small and Q varies slowly according to figs. 2 and 3.

With this, the integration over $\underline{k}$ in eq. (16) can be transformed into an integration over the variable $\underline{K} = \underline{k} \cdot \underline{T}'$. The resulting integral can be expressed by $F_0(\underline{v})$, the distribution in relative velocity in the absence of Coulomb explosion,

$$F(\underline{v}) = \frac{1}{P' Q'^2} F_0 \left(\underline{T}'^{-1} \cdot (\underline{v} - v_{\infty} \underline{\Omega}) \right) \quad (19)$$

where $\underline{T}'^{-1}$ is the inverse matrix,

$$\underline{T}'^{-1} = \frac{1}{Q'} \delta_{ij} + \left(\frac{1}{P'} - \frac{1}{Q'} \right) \Omega_i \Omega_j . \quad (20)$$

Projected Distributions

In the evaluation of eq. (19), care need to be taken of the anisotropy of scattering processes experienced by penetrating particles: Scattering on target nuclei causes lateral deflection while scattering on target electrons causes stopping. Either of these processes alone affects the ring pattern in all three dimensions.

Amongst several projected distributions, the most prominent one is the lateral velocity distribution caused by lateral scattering. It is found by first separating eq. (19) into components,

$$F(\underline{v}) = \frac{1}{P'Q'^2} \delta \left[\underline{T}'^{-1} \cdot (\underline{v} - v_{\infty} \underline{\Omega})_x \right] f_0 \left[\underline{T}'^{-1} \cdot (\underline{v} - v_{\infty} \underline{\Omega})_{\rho} \right], \quad (21)$$

where the subscript ρ indicates the component lateral to the beam, i.e., a vector in the (y,z) plane. The Dirac function expresses the neglect of stopping and straggling, and $f_0(v_{\rho})$ is the distribution in lateral relative velocity in the absence of Coulomb explosion.

The distribution in lateral velocity v_{ρ} is found by integrating (21) over v_x . The Dirac function allows separation of v_x and v_{ρ} with the result that

$$f(v_{\rho}) = \frac{1}{Q' [Q' \cos^2 \Phi + P' \sin^2 \Phi]} f_0 \left[\sqrt{\frac{v_{\rho}^2 \sin^2 \chi}{Q'^2} + \frac{(v_{\infty} \sin \Phi - v_{\rho} \cos \chi)^2}{(Q' \cos^2 \Phi + P' \sin^2 \Phi)^2}} \right]. \quad (22)$$

The geometry has been chosen such that χ is the angle between the lateral velocity and the plane defined by the molecule axis and the beam direction. Hence, f is normalized according to

$$\int_0^{\infty} p dp \int_0^{2\pi} d\chi f(v_{\rho}) = 1 \quad (23)$$

for all Φ , provided that $\int 2\pi v_{\rho} dv_{\rho} f_0(v_{\rho}) = 1$.

Consider three limiting cases of (22).

For $\Phi = 0$, i.e., a molecule aligned with the beam, (22) reduces to

$$f(v_{\rho}) = \frac{1}{Q'^2} f_0 \left[\frac{v_{\rho}}{Q'} \right] \quad (24a)$$

i.e., the distribution without Coulomb explosion scaled by Q' . Note that the atoms have experienced deflection perpendicular to the molecule axis.

For $\Phi = \pi/2$ and $\chi = 0$, the molecule is aligned normal to the beam, and the scattering distribution is considered parallel to the molecular axis. Eq. (22) yields

$$f(v_\rho) = \frac{1}{P'Q'} f_0 \left[\frac{v_\rho - v_\infty}{P'} \right] \quad (24b)$$

i.e., a distribution centered around the Coulomb velocity v_∞ with a width scaled by P' .

Finally, for $\Phi = \pi/2$ and $\chi = \pi/2$, the molecule is still aligned normal to the beam, but the scattering distribution is observed out-of-plane. Then, (22) yields

$$f(v_\rho) = \frac{1}{P'Q'} f_0 \left[\left(\frac{v_\infty^2}{P'^2} + \frac{v_\rho^2}{Q'^2} \right)^{1/2} \right]. \quad (24c)$$

Discussion

Figs. 2 and 3 demonstrate a dramatic difference between the Coulomb images parallel and perpendicular to the molecular axis. The existence of an effect of this kind has been recognized repeatedly, first in connection with the analysis of wake phenomena [5], and subsequently in multiple scattering [3,6,9,10]. The present work provides a general scheme for a theoretical treatment.

The reduction of the shift in the ring pattern due to different stopping powers, as expressed by eq. (14), appears particularly striking since it affects an average behavior rather than a fluctuation.

Similar effects in the average behavior must be expected in experiments on the scattering of fast molecules on surfaces [11,12], and need to be taken care of in the analysis.

While there is little doubt that wake forces acting on molecules can be studied by their Coulomb images [2,3], it is less clear to what extent image distortion due to the dynamic effects considered here has been incorporated explicitly or implicitly in common theoretical or numerical treatments.

While this paper mainly presents a general framework for further study, the case of multiple angular scattering has been treated more explicitly because of its importance in Coulomb explosion imaging [3,6,9,10]. A point need to be made about the scattering distribution f_0 characterizing multiple scattering in the absence of Coulomb explosion, eq. (22). In view of the dominance of small-angle events, there is a chance for correlated scattering of *both* fragment ions by one and the same target atom (fig.5). This becomes significant for molecules aligned with the beam, and at least for homonuclear molecules, scattering effects in the relative motion will be suppressed for such aligned molecules. Within the present framework, this feature is unaffected by Coulomb explosion, but it needs to be taken into account in the analysis of the velocity distributions of molecular fragments, whether Coulomb-exploded or not. A treatment based on earlier, similar work with atomic ions scattered on molecular targets [13,14] is in progress [8].

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Figure Captions

- Fig.1 Illustrating interference between Coulomb explosion and scattering (see text). v_0 is the beam velocity and Δv a velocity increment received by scattering. a) Molecule oriented parallel to the beam; b) Molecule oriented normal to the beam.
- Fig.2 The quantities P and Q defined by eqs. (2) and (4), giving the image distortion parallel and perpendicular to the molecule axis, respectively. $r(t)$ is the internuclear distance at the time the molecule receives a velocity increment Δv . D is the initial internuclear distance.
- Fig.3 The quantities P' and Q' defined by eq. (9). r is the internuclear distance at the time of emergence from the target. D is the initial internuclear distance.
- Fig.4 Ring pattern due to Coulomb explosion perturbed by stopping. Straggling, multiple scattering and wake force ignored.
- Fig.5 Correlated molecular scattering: The impact parameters s_A , s_B to a target atom C are related.

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FIG. 1

FIG. 2

FIG. 3

FIG. 4

FIG. 5

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