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Molecular effects in beam-foil spectroscopy

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The character of the ionic beam downstream from the foil in a beam-foil experiment depends on whether the incident particle entering the foil is atomic or molecular. In this paper it is demonstrated how incident molecules affect charge-state distributions, beam-particle scattering, charge exchange, excitation functions, mean lives and transition probabilities, alignment, and linear straggling after the foil. The possibility of exploiting certain molecular effects for further study is also discussed.

I. INTRODUCTION

Collision studies of energetic ions and molecules with gases and solid targets is a large important field of atomic physics. Basically, an ion or molecule is accelerated to the desired energy and directed to a target which can be an atom, molecule, or solid. The result of the collision is studied by measuring the emitted radiation (photons), emitted particles (electrons, atoms, molecules, and solid pieces), and the kinematic parameters (masses, energies, and velocities) of the collision fragments.

The beam-foil technique,[1] a particular area of beam target physics, is used to produce spectra of highly ionized species and measure mean lives of ions via time-of-flight procedures. The role of molecules in beam-foil experiments has been minimal because it is so well established that the foil completely dissociates the incident molecule, soon after it enters. No beam molecules have been detected after an amorphous[2] foil and no radiation from beam molecules has been seen.[3] Incident molecules have been used to extend the emergent particle energy range to lower energies. For example, if the minimum available accelerator energy is 200 keV then one gets 100-keV O^+ after the foil by accelerating 200-keV O_2^+. This technique is analogous to extending the emergent particle range to higher energies than available by accelerating multiply charged species. These methods succeed and have been used because, *within experimental uncertainties*, radiation emitted after the foil is independent of the incident particle condition whether it be ion, molecule, neutral, excited, or charged. This independence insures that the parameters in a carefully constructed beam-foil experiment will be strictly *independent* of, or *proportional* to, beam current (number of particles per sec).

The degree to which beam-foil parameters are strictly characterized by just two groups is surprisingly high considering the wide range of conditions used in various experiments. However, "nonlinear" effects do occur after the foil when incident particles are molecules. To show why, we will compare the atom-foil interaction with the molecule-foil interaction.[4]

II. ATOMS

When a monatomic ion enters a solid foil, collisions scatter it and destroy its initial charge and excitation state. The atom finally emerges and goes off with velocity v_f at angle θ from its initial path [see Fig. 1(a)]. It is in charge state Q_f and excitation state E*. After leaving the last surface and foil electron cloud, it continues downstream unperturbed by the foil and other beam particles.[5,6] It is a fact that for a 1-cm^2-diam 1-μA beam of 1.0-MeV protons going through a 200-Å-thick foil, the average distance between

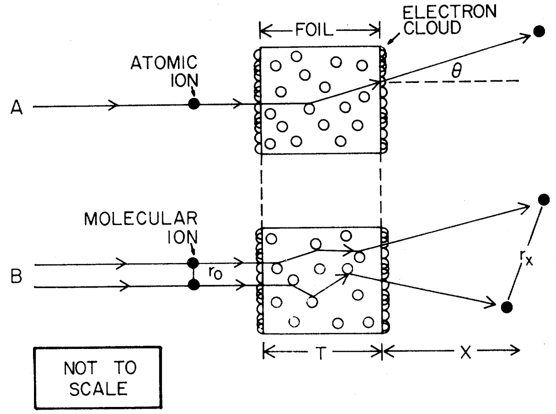

FIG. 1. (a) An atomic ion enters a thin foil and is scattered through angle θ. When $X \geq T$ downstream the ion is unperturbed. (b) A molecular ion with internuclear distance r_0 enters a thin foil where the molecular bond is broken and each ion is independently scattered. After foil interaction the ion-ion pair has internuclear separation r_x.

two protons is 6×10^{-2} cm, yielding an essentially zero probability for their being in the foil at the same time—*let alone at the same place*. Exactly the opposite is true for incident molecular particles where each projectile-foil collision *absolutely ensures* the presence of a near neighbor in the foil and after it.

There is, therefore, a fundamental difference in the beam-foil (particle-solid) condition depending on whether monatomic or molecular ions are used.

III. MOLECULES

Whatever the molecular weight, charge, or structure of the incident molecule, it is completely dissociated by the foil. Collisional energy exchanges are in the keV range whereas the molecular bond strengths are in the eV range. The first few collisions [see Fig. 1(b)] break the bond and give the ions large relative velocities inconsistent with a stable molecular configuration while foil nuclei and electrons completely perturb the electronic levels of the dissociated molecule. As the molecular ion proceeds through the foil, collisions continue to increase the velocity randomness of each atomic ion until they emerge in free space. After this the ions, separated by only a few angstroms, continue downstream unhampered by further foil collisions. However, for small r_x the ions can still interact and possibly share molecular electron orbits as a true molecule. As they continue to separate, repelling each other through Coulomb forces which also affect the local E fields of each ion, r_x increases until the original and captured electrons adiabatically relax into field-free eigenstates of the individual monatomic system.

Beam-foil experiments generally deal with the condition of beam particles in the downstream region, far (greater than a few micrometers) from the last foil surface and small nonequilibrium region just beyond. For incident monatomic ions the nonequilibrium region after the foil is no larger than a few hundred angstroms, after which the beam particles continue unperturbed by the foil and other very distant beam atoms. For incident molecular ions, however, the nonequilibrium region can extend many millimeters downstream from the foil, essentially extending the foil interaction region.

We investigated the molecular problem by seeing how near neighbors, in and after the foil, affected beam parameters after the foil. For example, if the combination of 1 μA of 1-MeV N_2^+ and 2 μA of 0.5-MeV N^+ gives identical results for all beam parameters after the foil no molecular effect exists.

IV. EXPERIMENTS

The molecular effect after the foil was studied for seven cases: (a) charge-state distributions, (b) beam-particle scattering, (c) charge exchange, (d) excitation functions, (e) mean lives and transition probabilities, (f) alignment and fine-structure interference, and (g) linear straggling.

A. Charge-state distributions

1.0-MeV O^+ and 2.0 MeV O_2^+ ions were directed through carbon foils and the equilibrium charge-state distributions 30 cm downstream were measured using electrostatic deflection and a foil-covered Faraday cup (see Fig. 2). The signals displayed in Fig. 2 show that the O_2^+ charge-state distribution centroid is more neutral than the one for O^+. The emergent atomic ions in both cases have equal velocities. Evidently electron pickup "at the foil" by the O-O molecular system is more efficient than for the atomic case. When a double foil (two foils separated by 1 mm) is used, the O-O charge-state distribution is identical to the atomic case.

B. Beam-particle scattering

The half-width of each charge-state peak of Fig. 2 for the O-O case is about 4% broader than the width of the corresponding peak for the O case. This increased broadening is caused by ionic repulsion in the O-O system after the foil, which increases the rms scattering angle $\theta_{\rm rms}$. The broadening remains when double foils are used even though the charge-state distribution is identical to the O case.

C. Charge exchange

H^+, H_2^+, and H_3^+ ions at energies between 0.2 and 1.5 MeV were directed through a carbon foil

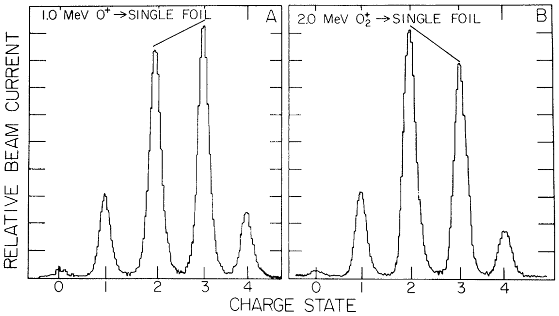

FIG. 2. Relative abundance of oxygen charge states after single foil interaction. (a) Charge state distribution from 1.0-MeV O^+. Charge state +3 is *more* abundant than +2. (b) Charge state distribution from 2.0-MeV O_2^+. Charge state +3 is *less* abundant than +2.

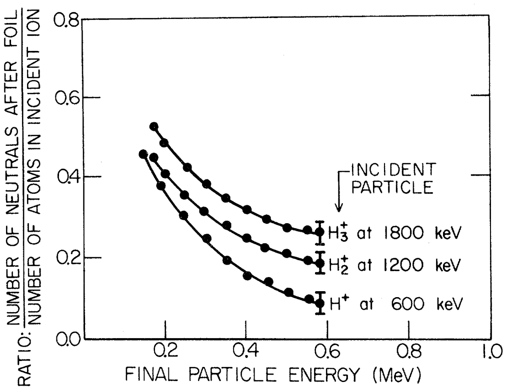

FIG. 3. Ratio of number of neutral hydrogens after a single foil interaction to the number of ions in the incident molecule. At 600 keV, a beam of hydrogen atoms from dissociated H_3^+ is about three times more neutral than the beam that was initially H^+.

and the charge exchange was measured for each case. We then determined the fraction of neutral particles after the foil and normalized this fraction to the total number of protons per molecule in the incident beam. The results shown in Fig. 3 indicate that the beam originating from an incident molecular ion before the foil is more neutral after the foil than the beam originating from atomic ions. The most neutral fraction occurs for the H_3^+ beam. This result is consistent with the oxygen results of experiment A and with those of Meggitt *et al.*[7]

D. Excitation functions

We measured the relative intensity of Ly α $(2p \rightarrow 1s)\lambda 1216$ Å. From the intensity $I = A(2p-1s) \times nh\nu$ we calculated the ratio n_{2p}/n of the $2p$ population to the neutral population n after the foil. $A(2p-1s)$ is the total transition probability per sec for the $2p-1s$ transition to occur and $h\nu$ is the energy of the Ly α photon. This showed a molecular effect on the $2p$ excitation function. The results given in Fig. 4 show that the relative intensity of Ly α from the beam for molecules incident is less than for protons incident, and that the molecular curves H_2^+ and H_3^+ almost coincide to form a group distinct from the H^+ case.

The Ly α radiation in this experiment can be affected by two different mechanisms: (i) A H^+ close to a H^* $(2s)$ can quench the $2s$ state and increase the Ly α radiation by E-field mixing of the $2s$-$2p$ levels. (ii) A H^+ (or H) close to a H^* can be considered a hydrogen quasimolecular ion emitting radiation different from Ly α wavelength. This would decrease the Ly α radiation for molecules incident and most likely occurs in this case.

In experiments like this we must distinguish between a change in transition probability caused

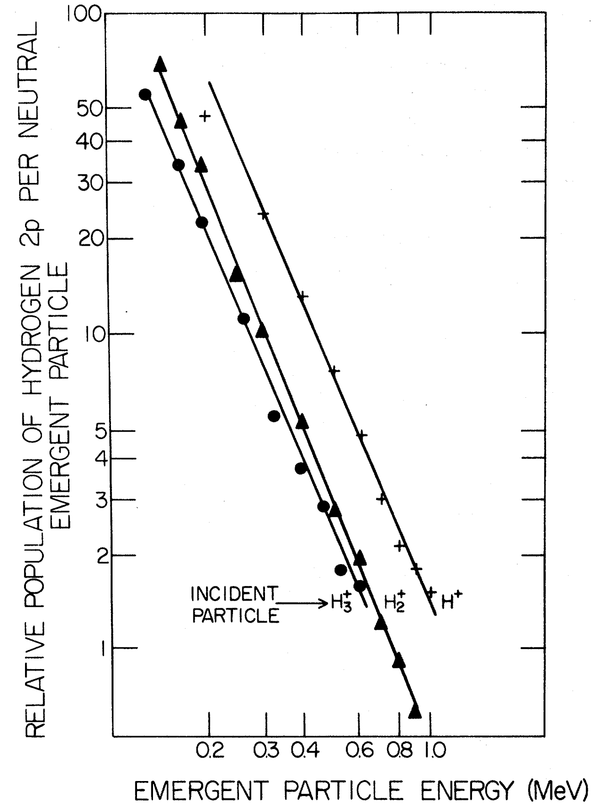

FIG. 4. Ratio of number of $2p$ states after a single foil to the number of ions in the incident molecule. At 600 keV a beam of hydrogen atoms from dissociated H_3^+ contains fewer $2p$ states per beam particle than the beam that was initially H^+.

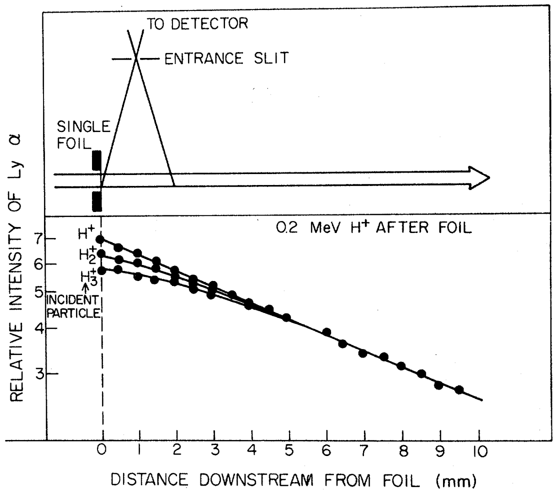

FIG. 5. Top. The geometrical arrangement of detector, slit, foil, and beam used in decay experiments. Bottom. Lyman α decay curves after the foil as a function of incident molecular ion.

by electric fields and a change in atomic level population caused by quasimolecules.

E. Mean lives and transition probabilities

We directed H^+, H_2^+, and H_3^+ particles at energies of 200, 400, and 600 keV, respectively, through a thin foil and measured the intensity of Ly α as a function of distance downstream from the foil in each case. The results are shown in Fig. 5 Close to the foil the radiation from the beam initially H^+ yields a pure exponential decay. The radiation from the beam initially H_2^+ and H_3^+ deviates successively more from the true intensity at $x = 0$, indicating a molecular perturbation immediately after the foil caused by the presence of near-neighbor hydrogens which inhibit Ly α radiation as in experiment C. When double foils are used all three curves coincide exactly. This result is consistent with the two-foil result of experiment A.

F. Alignment and interference effects

The first undisputed effect of molecules after a single foil was demonstrated in the early experiments dealing with hydrogen fine-structure levels.[8,9] The interference effect, which appeared as a periodic intensity variation of the Balmer lines after the foil, occurred for H_2^+ and H_3^+ incident particles *only when* double foils were used. In those experiments, H_3^+ was routinely accelerated to 200 keV to get 67-keV H after the foil, a higher fraction of neutrals and, therefore, higher yields of hydrogen radiation. The mystery concerning double foils for molecules persisted until the same interference effect occurred in H-like helium with just a single foil.[10,11]

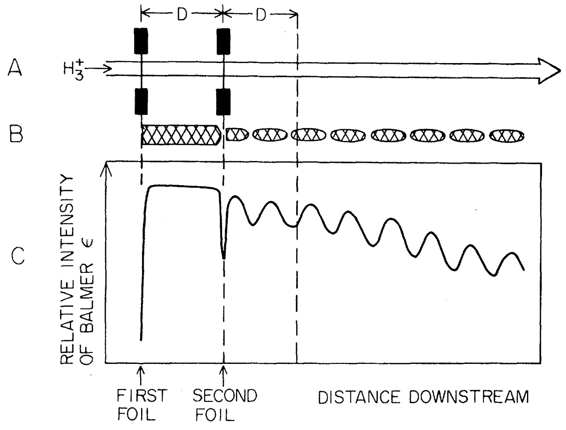

FIG. 6. (a) Arrangement of a molecular hydrogen beam and two foils used in a coherence-interference experiment. After the first foil the atoms enter an external magnetic field applied perpendicular to the beam and line of sight. (b) Image of the beam in Balmer δ(λ4102 Å) radiation. (c) Densitometer trace of image shown in b. Oscillations appear only after the second foil.

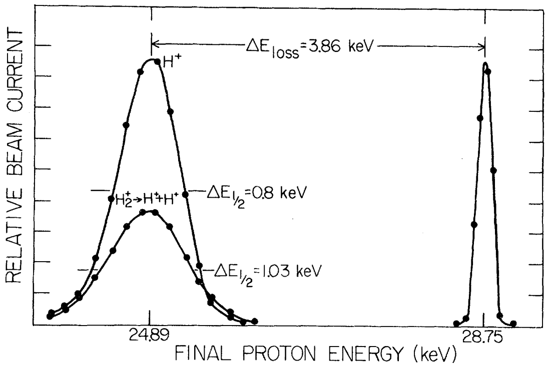

FIG. 7. Energy loss and linear straggling of protons after foil interaction from H^+ and H_2^+ incident ions. The half-width of the profile for protons from dissociated H_2^+ is larger than that for H^+ incident.

The interference effect as it appeared in the first experiments is shown in Fig. 6. H_3^+ molecular ions enter the first foil and are dissociated. Between the two foils the dissociated molecular parts (H^+ and H^*) repel, separate, and radiate light and then enter the second foil separated enough to permit the coherent foil excitation of each atom to remain unaffected by near-neighbor H^+ and H^*. Consequently, the *s-p* level interference modulates the radiation after the foil. Between the foils the near-neighbor perturbation destroys the coherence and/or phase of each excited state and the subsequent possibility of an interference effect. Although the atoms separate far enough in this region, the H^* excited states that finally arise do *not* have definite phase with respect to the first foil. Therefore the interference effect never occurs *regardless* of how large r_x becomes.

The molecular effect and two-foil role has been verified for H^+, H_2^+, and H_3^+; D_2^+, and D_3^+; as well as $^4He^+$ and $^3He^+$ particles. It is the single known instance where a molecular interaction *prohibits* a phenomenon from occurring.

G. Energy loss and linear straggling

We accelerated H^+ and H_2^+ particles to energies of 28.75 keV and 47.50 keV, respectively, and sent them through a thin foil. The energy loss of the *undeflected* H^+ in each case was measured with an 18-cm electrostatic analyzer. Figure 7 shows the energy profiles of protons *after* the foil for the case where the incident beam was atomic and molecular. Although no difference in energy loss

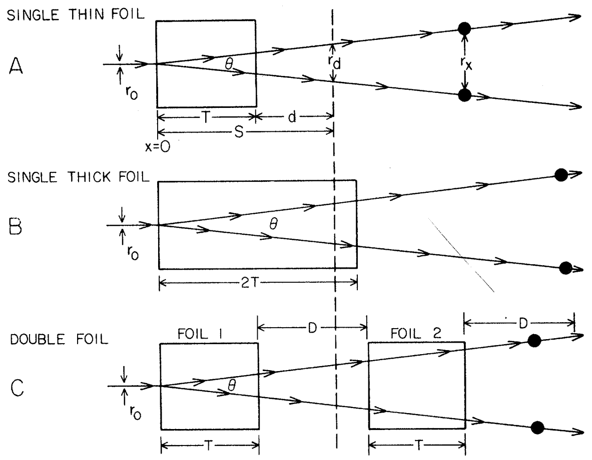

FIG. 8. (a) A single *thin* foil interaction with a molecular ion. The molecule with internuclear distance r_0 enters the foil at $x=0$ and dissociates. The ions emerge from the foil along paths defined by θ. At $X > S\,(r_x > r_d)$ the interionic perturbation is zero. Since for $S > T$, r_d occurs *after* the last foil surface, the beam will display molecular effects downstream. (b) A single *thick* foil interaction with a molecular ion. For this case $S > T$, therefore r_d occurs *before* the last foil surface. The beam will be free from molecular effects downstream. (c) A double thin-foil interaction with a molecular ion. For this case $S > T$ for the first foil but $S < 2T + D$ for the second. Therefore r_d occurs *before* the last surface as in b and no molecular effects occur downstream.

between the H^+ and H_2^+ particles is detected, the width of the H_2^+ energy loss peak is larger than the one for H^+. This increased width indicates linear straggling caused by a repulsion of the H^+-H^+ system components after the foil which is not present in the H^+ case. This result complements the increased scattering results for the O-O case of experiment B.

These seven experiments show that "nonlinear" effects occur after the foil when the incident particles are molecules and suggest that they will turn up in virtually every experiment where sufficient sensitivity exists.

We therefore modify the statement that molecules do not survive the foil interaction to say: Molecules are definitely dissociated by the foil but the individual ions are not scattered enough to prevent ion-ion perturbation, formation of quasimolecules, and quasimolecular effects after the foil. The ions can still be close enough to share electrons as an adiabatic molecule even though they *diverge, converge, or travel downstream in parallel paths.* Dissociation is considered complete when ions are separated enough to be unperturbed.

It is instructive to examine the role of (a) thin foils, (b) thick foils—folded and sandwich, (c) double foils, multiple foils, and (d) first layers of solid targets.

The first multiple-foil experiments[8,10] showed that the main role of the double foil is to afford a large enough interaction region for molecular parts to separate before leaving the *last* foil surface to enter the downstream free-space region. The same role can be achieved with *very* thick foils but the effects are complicated by increased straggling, scattering, and energy loss. The relationship between the various parameters describing thin foil, thick foil, and double foil interaction is given in Fig. 8. S represents the distance the molecular parts must travel after entering the foil before they are free of near-neighbor perturbations, d is the distance between this point and the downstream surface of the first foil. D is the spacing between the double foil surfaces. The observation point X is far downstream ($X \ggg T$ and d). In order to prevent molecular effects one must choose conditions so that r_x at the *last* downstream surface is large enough to prevent interference between the emergent ions. Fig. 8(c) shows how this can be achieved with thick and multiple foils.

There is a fundamental difference between the beam condition at $X = T + D$ after the first foil [Fig. 8(c)] and at $X = 2T + 2D$ after the second foil. In both places the atoms are separated enough to be unperturbed. However, at $X = T + D$ the beam condition developed out of the foil interaction *plus* near-neighbor perturbations occurring over the distance *d*. The second foil destroys all such beam

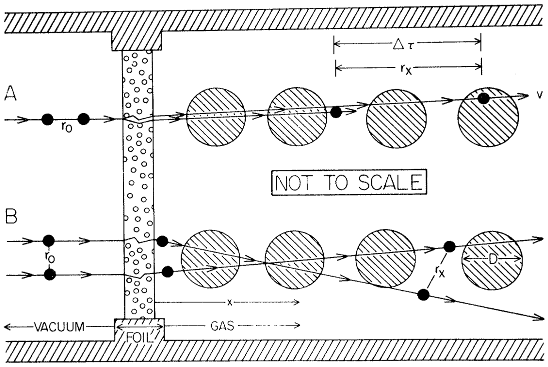

FIG. 9. Geometrical detail of a beam-foil-gas collision where individual ions of a foil-dissociated molecule collide with gas atoms (shaded circles) after the foil. (a) The linear molecule is oriented parallel to the beam (perpendicular to foil). The collision will be the result of two successive impacts separated by a time $\Delta t = r_x/v$, where v is the emergent ion velocity. The time Δt can be as short as 10^{-17} sec. (b) The orientation is perpendicular to the beam (parallel to foil). For small x the emergent ion pair acts as a molecule (or two close ions). As x increases until $r_x > D$ after which all collisions are between gas atoms and single isolated ions.

states and recreates them—identical to those created by the first foil. Therefore the states at $X=2T+2D$ have developed *only* from the foil interaction. This result occurs in experiment E where the interference effect does not develop at *any* point downstream from the first foil even though r_x^i becomes larger than r_a. For incident monatomic particles, d is probably the order of angstroms as compared to micrometers and mm for molecular particles. No deviation from the expected exponential decay of radiation close to the foil for monatomic ions incident has been reported,[12] although several searches have been made. However, the region close to the foil is a challenge to investigate because of complicated geometrical problems associated with the foil-beam-optical system arrangement.

The role of sandwich foils, i.e., foils for which the empty intermediate space D in Fig. 8(c) is replaced with a different material, such as Be to make a C-Be-C foil, is the same as that for thick foils with thickness $T+D+T$ Significant differences in x-ray production due to inner-shell matching might occur[13] but this effect is not pertinent to the mechanism we are discussing.

Increasing the thickness of foils already thick enough to ensure charge and excitation state equilibrium will only increase straggling, scattering, and energy loss and perhaps broaden some distribution. Basically the beam character after thin and thick foils is similar. The first layer of a thick foil or solid targets acts essentially like the thin foil in the beam-foil-gas[14,15] case where the foil changes the incident beam character and "prepares it" for further interaction.

There are two experiments in which the effect of molecules after the foil can be exploited. One can study (i) quasimolecules with varying impact parameter r_x after the foil and (ii) multiple-(simultaneous) particle impact of gas atoms (or molecules) after the foil.

H. Quasimolecules after the foil

When the ions of a dissociated molecule leave the foil they can have velocities such that r_x *decreases* with x before increasing. At the "crossing point" two effects might occur: (a) the appearance of quasimolecular radiation. Because r_x is not constant, the radiation could be continuous or atomic lines might be broadened. No specific search for molecular radiation has been carried out although continuous radiation "at the foil" has been seen.[16] (b) the second effect is the appearance of characteristic x rays and Auger electrons far downstream from the foil. The emission rate will not be related by the exponential decay of excited states formed at the foil. In some respects, the converging ion-ion pairs after the foil resemble a crossed-beam situation.

J. Multiple-particle impact, beam-gas experiments

A variation of a beam-foil-gas experiment, where the foil breaks up the molecule and prepares it for further collisions in a gas cell[14] is illustrated in Fig. 9. Molecular ions pass through the foil covering the entrance of the cell, dissociate, and enter the case cell. Downstream they collide with gas atoms simultaneously and with varying impact parameter. As x increases, r_x increases, causing collisions which are initially "molecule"-atom close to the foil to become finally ion-atom at large x. Since the excitation function depends on the nature of the bombarding system, the radiation from the gas will vary with x also.

For example, high-energy (>2-MeV) hydrogen molecular ions H_2^+ or H_3^+ would dissociate and totally strip at the foil giving just protons after the foil and in the gas where electron pickup is negligible. Therefore radiation at the foil can be related to H^+-H^+-atom collisions; at large x to H^+-atom collisions and at intermediate x to (H^+-H^+)-atom system with impact parameters r_x.

VII. CONCLUSION

We introduced the general problem of molecules in beam-foil spectroscopy experiments and showed how molecular effects appear in some beam-foil data. We presented a *geometrical* description of the molecule-foil interaction to bring into perspective the parameters and ideas involved and to serve as a guide for explaining certain effects and developing certain experiments. Finally, we described experiments which could exploit the molecular effect.

The molecular effects should be pursued for the following reasons: The beam-foil-gas case with molecules gives a new set of initial conditions for interaction studies. Information about molecular bonds and ion-ion perturbations might be inferred from studies of the interaction products if the kinematics of the interaction can be defined and controlled. Quasimolecular effects could lead to the x-ray emission, molecular radiation, or broadened atomic lines from the interaction region. Beam-foil-gas collisions using particle pairs with varying impact parameters will give results which can be compared to pure beam-gas (atom-atom or molecule-atom) results. Documentation of molecular effects and current-dependent effects after the foil is necessary to get a complete description of the beam and to insure good beam-foil data.

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