Energy-loss scaling in 0.5–3.5-keV Ne^+ and Ne collisions with H_2 and D_2

N. Andersen,* M. Vedder, A. Russek, and E. Pollack
Department of Physics, University of Connecticut, Storrs, Connecticut 06268
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Energy losses are measured in $Ne^+ + D_2$, $Ne^+ + H_2$, and $Ne + D_2$ collisions for beam energies $0.5 \leq E \leq 3.5$ keV and scattering angles $\theta \leq 5$ deg. Rotational and vibrational excitation of the target molecule is found, but the probability of direct electronic excitation is seen to be extremely small. The results indicate that the most probable laboratory energy loss T_0 for a projectile with mass M_p scattered by a homonuclear binary molecule with atomic masses M scales so that the quantity $f = T_0 M/(M_p E\theta^2)$ is a function of the reduced scattering angle $\tau = E\theta$ only, as recently predicted theoretically by Sigmund. The function $f(\tau)$ is found to be the same for the $Ne^+ + D_2$ and $Ne^+ + H_2$ systems, but is strongly dependent on the charge state of the projectile.

I. INTRODUCTION

The last two decades have shown rapid progress in our understanding of electronic excitation in ion-atom and atom-atom collisions in the lower keV energy range. Mainly due to the development of the molecular orbital (MO) model for the binary projectile-target system,[1] detailed insight has been obtained into the various excitation mechanisms that are active at different stages of the collision. For a large number of simple and for several complicated systems quantitative agreement has been obtained between cross section measurements for the dominating inelastic processes and theoretical estimates, based on calculations of the potential curves for the quasimolecule.

The situation with respect to ion-molecule and atom-molecule collisions is considerably different. These collisions are much more difficult to study because of the additional degrees of freedom inherent in a triatomic system. Even electronically adiabatic collisions can be inelastic in the rotational and vibrational degrees of freedom. Until very recently, this field was exclusively the domain of chemists, whose concern with chemical reactions at realistic temperatures concentrates their interest on collisions at very low energy. A recent review by Toennies[2] discusses both the theoretical and experimental status of cross sections for rotational and vibrational excitation at low collision energies (generally less than 10 eV). There has however been some work involving H_2 at higher collision energies. Herrerro and Doering[3] investigated H^+ on H_2 collisions and reported total cross sections for vibrational excitation for laboratory energies up to 1.5 keV (1.0 keV in the center-of-mass reference frame). Gillen, Mahan, and Winn[4,5] have studied $O^+ + H_2$ and $O^+ + D_2$ collisions at energies up to 50 eV in the center-of-mass reference frame and Mahan and Winn[6] have investigated $Ne^+ + H_2$ and $Ne^+ + D_2$ collisions at center of mass energies up to 23 eV. In addition Fernandez *et al.* reported on the small-angle charge exchange in $He^+ + H_2$,[7] and Bray *et al.*[8] studied the small-angle quasielastic, inelastic, and charge exchange scattering at low keV energies in this system.

In principle differential cross sections for vibrational excitation as well as elastic scattering in collisions of atoms or ions with diatomic molecules can provide a valuable experimental check of a substantial region of the adiabatic potential surface for the triatomic molecule or molecular ion.[2] The desired information is, however, extremely difficult to obtain because of the very large number of coupled rotational channels involved. Consequently, significant advantages are anticipated if the differential scattering experiments are carried out at the highest possible collision energies for which the electronic state remains adiabatic, namely the low keV range. At such collision energies, the nuclear motion in the collision should for all practical purposes be completely classical and completely impulsive. During these very short collision times ($\approx 10^{-15}$ sec), there is no time for vibrational or rotational motion or relaxation.

At the present stage quantitative results for even the simplest processes in an ion-molecule collision may be useful to distinguish the quantum-mechanical excitation aspects from the classical effects. In this context scaling properties, which turned out to be very powerful tools for the analysis of ion-atom collisions,[9] are of particular interest. Collisions where electronic excitation is weak or absent seem here to provide the appropriate test systems to guide our understanding of the purely classical aspects of three- (or several-) body interactions. The insight gained may greatly facilitate the analysis of more complicated systems where elec-

tronic excitations are important.

Sigmund[10] recently proposed a simple scaling law for energy-loss distributions in vibrationally and rotationally inelastic, but electronically elastic collisions. The scaling law is derived in the *sudden approximation*, i.e., the neglect of motion of the atoms of the molecule *during* the collisions, or

$$\omega L/v \ll 1 . \tag{1}$$

Here ω is the frequency of vibration, L is the interaction length, and v the velocity of the projectile. For a D_2 molecule $\omega = 5.8 \times 10^{14}$ sec^{-1}. If L is assumed to be the D-D internuclear distance of 1.4 a.u. then for a Ne-D_2 collision $\omega L/v = 0.44 \times E^{-1/2}$, with E in keV. The condition (1) therefore puts a lower limit of about 1 keV on the energy range of interest here. The approximation (1) was also assumed by Baudon[11] in his theoretical treatment of 0.5–3.0 keV N_2^+-He collisions. The aim of the scattering experiments reported in the present work is to test the validity of the new scaling law, and study the effects of varying the target mass or changing the charge state of the projectile. For this purpose the $Ne^+ + D_2$, $Ne^+ + H_2$ and $Ne + D_2$ systems are particularly appropriate, since inelastic processes are known to be weak for these collisions.[6,12,13] Furthermore, the large projectile to target mass ratio introduces some additional advantages which will be evident from the kinematical equations discussed below. A brief report on some of the findings in this paper has been given previously.[14]

II. EXPERIMENTAL

A. Apparatus

The experimental setup has been described in detail elsewhere.[8,15] Briefly, a well collimated beam of Ne^+ ions is directed into a scattering cell containing the target gas at a pressure in the 10^{-4}–10^{-3} Torr range. Beam energies range from 0.5–3.5 keV, with beam currents of 5–100 pA. The energy spread of the incident beam is 1–2 eV and the angular spread about 0.25° (full width at half-maximum). Particles scattered through an angle θ are collimated by small apertures. Scattering angles are in the range 0.6° to 5°. The position of the scattered beam is known to within 0.05°. The energy-loss spectra (ELS) for the scattered ions are measured with a parallel plate electrostatic analyzer having an energy resolution of 0.5 eV at 1.0 keV. In addition to the broadening introduced by the analyzer the lines are also broadened by the acceptance angle of the detector [see Eq. (6) in Ref. 8]. The spectra for the neutral particles are recorded using a time-of-flight (TOF) technique. For neutral projectiles, the ion beam is first chopped with a 0.33-MHz pulse, and then neutralized by resonant change exchange. The remaining ions are removed from the beam by a set of electrostatic deflector plates before entering the collision chamber. The energy losses are determined from the measured flight time between the collision cell and the particle detector situated 63 cm away. This distance gives an energy resolution of 0.4–3.2 eV for beam energies in the 0.5–2 keV range.

B. Basic parameters

Here we shall present the basic equations of the scattering processes that are relevant to this experimental situation and of use in the interpretation. Let E be the energy of the incident beam of particles with mass M_p, which collide with a homonuclear diatomic molecule with atomic masses $M_1 = M_2 = M$. For small scattering angles

$$\theta \simeq \Delta p/p , \tag{2}$$

where Δp denotes the momentum transfer and p the momentum of the incident particle.

The energy loss T of a beam particle elastically scattered by an *atomic* target with mass M_T is simply

$$T = \Delta p^2/2M_T = (M_p/M_T)E\theta^2 . \tag{3}$$

However, a *molecular* target will yield an energy-loss distribution corresponding to the *distribution* of impact parameters, molecular orientations and internuclear distances leading to the selected scattering angle θ. The measured distribution will of course be further broadened due to experimental effects such as the angular divergence of the beam and the finite acceptance angle of the analyzer.

Sigmund[10] has recently derived a scaling law for this distribution in the case of electronically elastic collisions. In particular, he predicted that the position T_0 of the maximum of the energy-loss distribution scales as

$$T_0 = (M_p/M)E\theta^2 f(E\theta) = (M_p/M)(\tau^2/E)f(\tau) . \tag{4}$$

Here $\tau = E\theta$ is the reduced scattering angle and f is a function which depends on the scattering potential between the incoming particle and an atom of the target molecule. For small τ values $f(\tau) = \frac{1}{2}$, while $f(\tau) = 1$ for large values. Comparison of Eqs. (3) and (4) shows that these two limiting situations correspond to elastic scattering from the entire molecule ($M_T = 2M$, the elastic limit), and scattering from only one of the atoms of the molecule ($M_T = M$, the binary limit), respectively. For reasonably narrow distributions it may be heuristically useful to think of M/f as an effective molecular scattering mass [compare Eqs. (3) and (4)]. In this

language the Sigmund theorem, Eq. (4), may be restated as: The effective molecular mass is a function of the reduced angle only.

The following assumptions are made in the derivation of Eq. (4): (i) the collision energy is sufficiently high to justify neglect of vibrational and rotational motion during the collision; (ii) there is a negligible displacement during the collision [assumptions (i) and (ii) constitute the "sudden approximation" which is valid in the keV energy range]; (iii) small laboratory scattering angles θ; (iv) the molecular scattering potential can be approximated as a sum of two spherically symmetric potentials one centered on each atom. This makes it possible to express the total deflection of the projectile as a sum of two individual deflections from the atoms of the molecule.

Several reasons for the choice of the $Ne^+ + D_2$, $Ne^+ + H_2$, and $Ne + D_2$ collision systems to test this theory now become evident from these equations, apart from the virtue of small electronic excitation mentioned above. The large M_p/M ratio yields a large energy loss which facilitates a relatively precise determination of this quantity. It also assures fulfillment of assumption (iii) of small laboratory scattering angles, while at the same time the center-of-mass scattering angle and the energy loss may be substantial. This situation corresponds to violent interactions approaching a binary projectile-atom encounter. Both H_2 and D_2 targets are used, since the only essential difference between these cases is the mass difference, whereas the $Ne^+ + H_2$ and $Ne^+ + D_2$ interaction potentials should be similar, leading to the same f function for these interactions. Finally, a comparison of data using Ne^+ and Ne beams will reveal how sensitive the f function is to the charge state of the projectile.

III. RESULTS AND DISCUSSION

Sections III A and III B will present data for the $Ne^+ + D_2$ collisions only. Section III C will also include results for $Ne^+ + H_2$. Section III D compares the results with those obtained for the $Ne + D_2$ system.

A. Energy-loss distributions for Ne^+

Figures 1(a)–1(e) display typical ELS for 3.5-keV $Ne^+ + D_2$ collisions, at $E\theta^2$ values of "0", 1, 5, 10, 15, and 20 keV deg^2 [(a) shows the energy spread of the incident beam]. The spectra all show nearly symmetric distributions with a single well-defined maximum. The measured distributions broaden with increasing scattering angle. At small angles the broadening is dominated by the effect due to the finite acceptance angle of the detector. The experimentally determined T_0 values agree with the predictions of Eq. (4) using $f = 0.5$ for small angles ($E\theta^2 < 5$ keV deg^2), corresponding to the elastic limit. For larger scattering angle, the energy losses gradually exceed the value for elastic scattering. The detailed behavior of this break away from the elastic limit is displayed in Fig. 2, where T_0 is plotted versus $E\theta^2$ for incident energies of 1.5, 2.5, and 3.5 keV. It is particular-

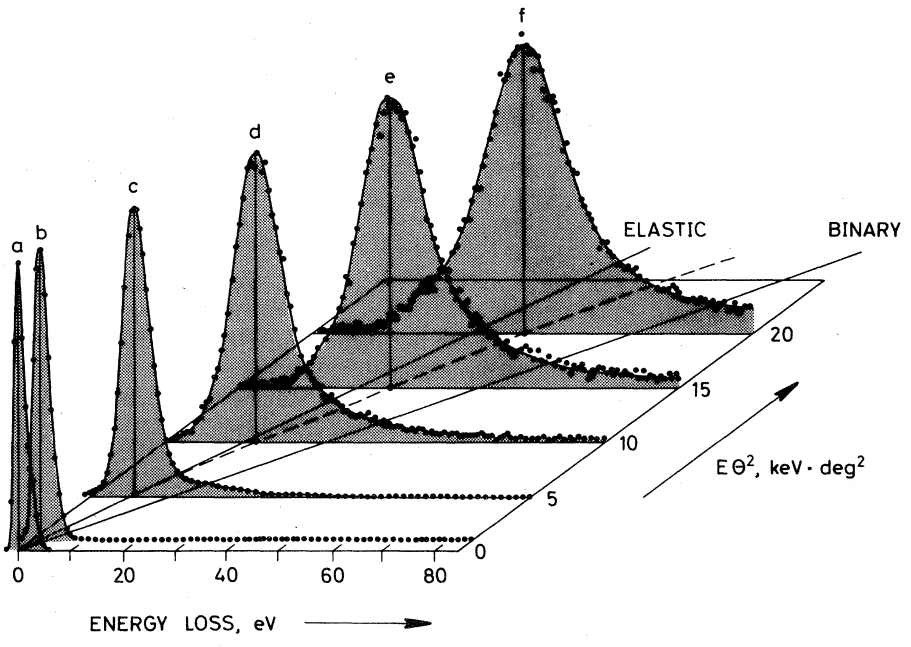

FIG. 1. ELS for 3.5-keV $Ne^+ + D_2$ collisions, measured at $E\theta^2 = 0, 1, 5, 10, 15$, and 20 keV deg^2.

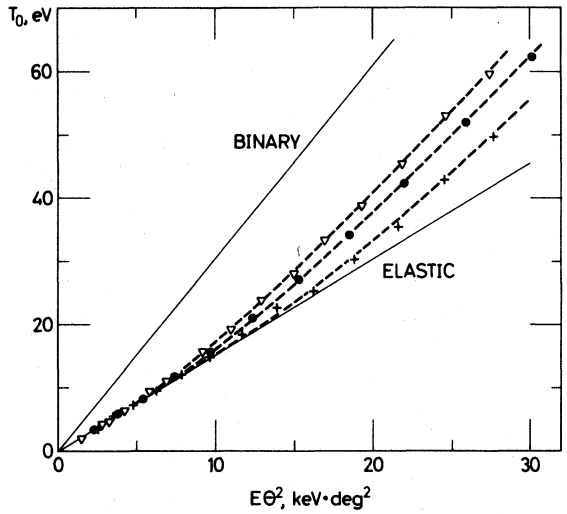

FIG. 2. Most probably energy loss T_0 plotted vs $E\theta^2$ for $E=1.5$(×), 2.5(●), and 3.5(▽) keV. The internal energy acquired by the molecule at any $E\theta^2$ is the difference between the experimental and elastic limit curves.

ly simple to identify the internal energy acquired by the molecule since at any $E\theta^2$ it is the difference between the experimental and elastic limit curves.[8] A slight dependence (to be analyzed further in Sec. III B) of the T_0 vs $E\theta^2$ dependence on incident energy is seen.

In all spectra the direct electronic excitation is found to be very weak, and negligible for the present purpose. Figure 3 shows this inelastic electronic excitation at an angle $\theta=0.53°$ (where it is found to be at a maximum). About a 1% excitation of D_2 is seen, while no excitation of Ne^+ is apparent. Hodge *et al.*[12] have investigated the details of electronic excitation and capture for this collision in the same energy and angular range, and found extremely small cross sections. Gusev *et al.*[13] measured the total cross sections for H_α and H_β emission due to dissociation of the excited H_2 molecule and found very small cross sections in the whole range from threshold up to 30 keV. It may thus be concluded that electronic excitation is indeed very small for this collision, as required by the analysis of Sigmund.

B. f function for Ne^+

Figure 4 shows the f values at $E=3.5$ keV, calculated from the most probable energy loss T_0 by means of Eq. (4), as a function of $\tau=E\theta$. It is seen that the elastic limit is followed up to $\tau\simeq 5$ keV deg. The curve then breaks away rather abruptly from the elastic limit and approaches the binary limit, $f=1$, near $\tau=20$ keV deg. A similar effect was

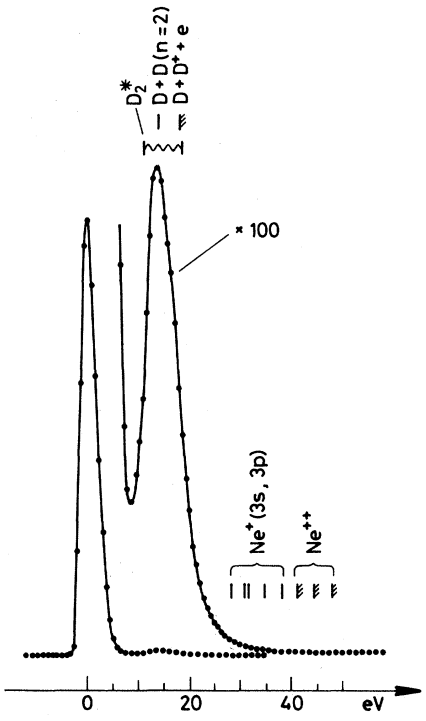

FIG. 3. ELS measured at $E=3.5$ keV and $\theta=0.53°$. The electronic excitation is seen to be very weak and due mainly to excitation of the D_2 molecule. Within the limits of the detection efficiency, no excitation of Ne^+ is observed.

found in O^++HD collisions[5] which showed peaks corresponding to impulsive O^++H and O^++D scattering. The region $\tau>17.5$ keV deg could not be studied in the present experiment. An important feature of presenting data on a τ plot is derived from the fact that to a good approximation τ has the same value in the laboratory and center of mass frames.[9,16,17]

C. Scaling of f for Ne^+

Figures 5(a)–5(d) show the f functions, calculated from the measured T_0 values for incident energies in the 1.5–3 keV range. The similar τ behavior is striking. Indeed, when plotted in this way, all experimental data follow (within the experimental uncertainties) a common curve indicated by the heavy lines. The scaling property, Eq. (3), is thus very well fulfilled for the Ne^++D_2 collision.

A further check of the scaling law is obtained by a determination of the f function for the Ne^++H_2 collision, as shown in Fig. 5(e). For a given beam energy and scattering angle the measured energy loss is found to be *twice* the value obtained for Ne^+ +D_2, yielding the same f function for Ne^++H_2 as

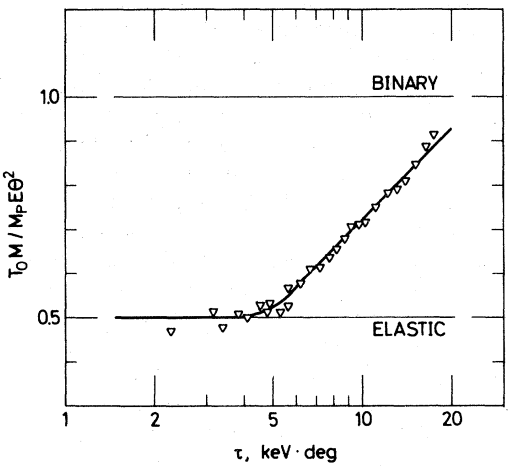

FIG. 4. $f = T_0M/M_pE\theta^2$ evaluated from the measured T_0 value at $E = 3.5$ keV and scattering angles θ out to 5°.

for $Ne^+ + D_2$. This clearly shows that the τ variation of f is determined by the projectile-target atom potential which is of course the same in $Ne^+ + D$ and in $Ne^+ + H$.

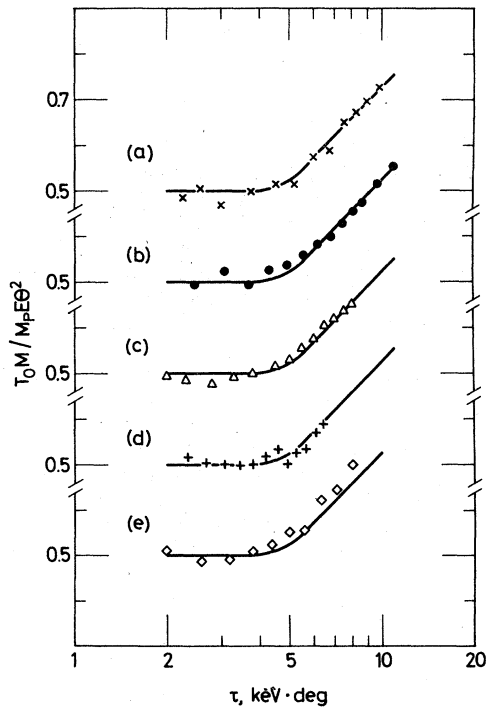

FIG. 5. f values for (a) × 3.0 keV, (b) ● 2.5 keV, (c) △ 2.0 keV, and (d) + 1.5 keV $Ne^+ - D_2$ collisions, (e) □ 3.0 keV $Ne^+ - H_2$. To a good approximation τ has the same value in the laboratory and center-of-mass frames.

D. Charge state effects

Sections III A–III C above considered f as a function of τ for Ne^+ collisions. The f function for the neutral $Ne + D_2$ collision is also determined in order to analyze the effect of the charge state of the projectile. Due to the relatively modest energy resolution in the TOF spectra (see Sec. II A) these measurements are less accurate than those from the ELS technique, as indicated by the error bars on Fig. 6. Even with this reservation it may however by concluded that the scaling law is also valid for this collision. Of greater interest is the dramatic difference seen between the f function for the neutral collision as compared to the f function for Ne^+ impact. For the $Ne + D_2$ collision, the break away from the elastic limit takes place at $\tau \simeq 2$ keV deg and the binary limit is reached near $\tau \simeq 10$ keV deg, while for the $Ne^+ + D_2$ collision the break away from the elastic limit is seen to take place at 5 keV deg and the binary limit reached at 25 keV deg. This difference was quite unexpected. At the substantial scattering angles involved in this experiment, the scattering was heretofore believed to be almost entirely due to the repulsive screened Coulomb core-core interaction, negligibly perturbed by valence (chemical bond) forces. Consequently, no significant difference was anticipated between $Ne + D_2$ and $Ne^+ + D_2$ collisions, because these systems differ only in the valence forces. However, the remarkable difference observed between these two collision systems clearly indicates that valence forces play a significant role in the scattering process in this τ range.

A second unexpected experimental result is the small value of the most probable energy loss due to vibrational-rotational excitation at small τ. The distance of closest approach between Ne^+ and D

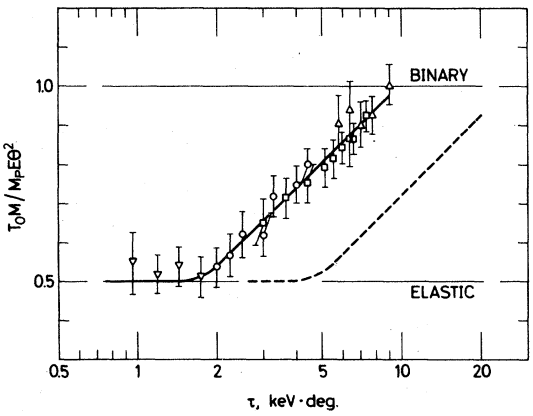

FIG. 6. f values for (▽) 0.5 keV, (○) 1.0 keV, (□) 1.5 keV, and (△) 2.0 keV $Ne + D_2$ collisions, determined by TOF techniques. The dashed line shows the f function for the $Ne^+ + D_2$, H_2 systems from Figs. 4 and 5.

that will result in scattering at these angles is approximately equal to the D_2 intermolecular separation. Thus, it was expected that these collisions would be closer to the binary limit of Fig. 2 than to the elastic limit. The experimental results imply either that strong scattering forces exist at anomalously large distances (where the two Ne-D forces would be nearly parallel), or that the scattering forces are nonadditive. Either alternative is suggestive of a substantial scattering contribution from valence forces. It is interesting to note, in this connection, that the Ne-D_2 collision system, which is a closed shell system with only weak van der Waals forces between projectile and target (in addition to the core-core screened Coulomb repulsion between each D and the Ne projectile) exhibits more vibrational-rotational excitation and a more rapid approach to the binary limit than does the Ne^+-D_2 collision system.

IV. CONCLUSIONS

The results of the measurements presented above strongly confirm the Sigmund scaling law, Eq. (4), for the $Ne^+ + D_2$, $Ne^+ + H_2$, and $Ne + D_2$ collisions in the 0.5–3.5 keV energy range.

Theoretical calculations of the f function, which turned out to depend drastically on the charge state of the projectile, are now needed to clarify the physics underlying its variation with τ, and in particular, to display the position and width of the corresponding impact parameter distributions.

Experiments on other collisions systems are required in order to explore how generally applicable the scaling law is. A crucial point in this connection is whether the assumption of no electronic excitation is indeed necessary. The similarity in τ dependence between the elastic and the inelastic peaks in various collisions may indicate that the scaling principle could be generalized, but the experimental evidence at present is too limited to draw any definite conclusions on this aspect.

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*Permanent address: Physics Laboratory II, H. C. Orsted Institute, DK-2100 Copenhagen, Denmark.

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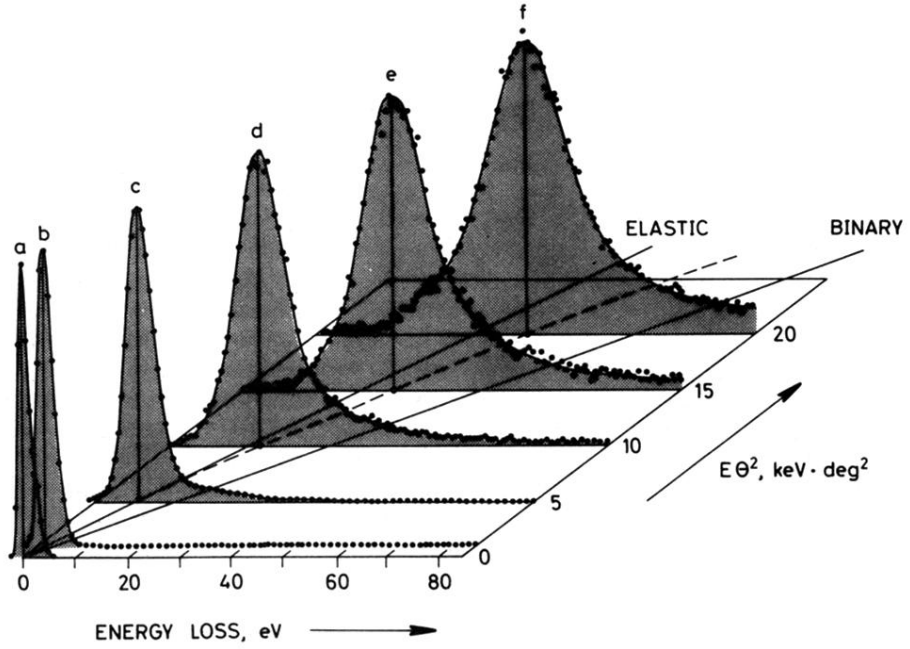

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