REVIEW OF STRIPPING OF HEAVY IONS*

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Electron capture and loss phenomena are reviewed which occur when heavy ions penetrate through matter. Stripper targets consisting of dilute and dense gases, large molecules and solids are examined with regard to their effects on heavy ion charge state distributions. The discussion includes semiempirical predictions for average equilibrium charge states, density effects in gases and solids, tails of charge distributions, and charge changing cross sections.

Introduction

The acceleration of heavy ions is being pursued with increasing efforts and especially during the last years acceleration techniques have been studied in great detail. In reaching the goal to produce intense beams of ions as heavy as uranium and energetic enough to overcome the Coulomb barrier even for the heaviest targets, many new problems must be solved which were not important for the design of conventional light particle accelerators. One of these problems concerns the ionic charge of heavy ions which is an influential new parameter. In this paper, the variation of ionic charge due to collisions with matter ("stripping") will be discussed, as well as some associated phenomena of practical interest.

The effects of charge stripping on heavy ion acceleration are twofold. On the one hand, the passage of heavy ions through specially designed strippers can be exploited to produce a substantial increase of the ion charge which reduces the effective potential required for further acceleration. In order to find the most suitable stripper and to utilize the highest possible charge states, it is necessary to investigate the effects of strippers on heavy ion beams in great detail. On the other hand, random stripping in the residual gas of an accelerator may lead to beam losses. In order to calculate the vacuum which guarantees a satisfactory particle transmission, it is necessary to know charge changing cross sections. These cross sections are very complex quantities and they can hardly be estimated without extensive knowledge about fundamentals of charge changing processes.

The Average Equilibrium Charge

When a monoenergetic ion beam penetrates through matter, the charge of the ions fluctuates due to electron capture and loss processes. The resulting charge distribution depends initially on the charges present in the incident beam and on the target thickness, but it changes rapidly towards an equilibrium distribution, F(q), which depends only on the nuclear charge and the velocity of the ions, Z and V, and on the target species. The required equilibrium thickness of the target increases slowly with V and is typically between 1 and 50 μg/cm^2 for beam energies below 200 MeV. The energy loss in these targets usually does not exceed a few percent of the initial ion energy.

From a given equilibrium charge distribution it is possible to derive a well defined average charge, $\bar{q} = \sum_q F(q)\, q \,/ \sum_q F(q)$. Since 1940, the functional dependence of $\bar{q}$ on Z and V has been the subject of theoretical and experimental studies. For many practical purposes it is convenient to use explicit expressions for $\bar{q}(Z,V)$ which hold over large ranges of Z and V. In the following, the most important ones among such formulas are discussed.

Gaseous Strippers

As early as in 1941 Bohr has given a theoretical estimate for gaseous targets,[1]

$$\bar{q}/Z = V/(V_0\, Z^{2/3}), \qquad (\bar{q}/Z < 0.5) \tag{1}$$

where $V_0 = e^2/\hbar$. This represents a remarkable first order approximation though later experiments showed that Eq.(1) overestimates $\bar{q}$ systematically. An improved formula is obtained by the generalized exponential[2,3]

$$\bar{q}/Z = 1 - \exp\left[-V/(V_0 Z^{2/3}) \right]. \tag{2}$$

This expression overestimates $\bar{q}$ only at low energies where it may give values which deviate from experimental data obtained in N_2, O_2 or Ar strippers by as much as $\Delta\bar{q} \approx 2$, but for higher energies where $\bar{q}/Z > 0.3$ it seems that Eq.(2) predicts $\bar{q}$ within the experimental errors.

Closer agreement with experimental results may be obtained by fitting the data separately at low and high ionizations:[4]

$$\bar{q}/Z = \begin{cases} A\ V/Z^{1/2} & , \quad (\bar{q}/Z < 0.3) \qquad (3a) \\ \lg(V/mZ^{\alpha_1})/\lg(n/Z^{\alpha_2}) & , \quad (\bar{q}/Z \gtrsim 0.3) \qquad (3b) \end{cases}$$

where the five parameters are determined empirically and amount for targets of N_2 or Ar to A = 0.18; $\alpha_1 = 0.4$; $\alpha_2 = 0.3$; m = 0.9; n = 7. The linear approximation Eq.(3a) predicts $\bar{q}$ generally within one charge state. However, identical parameters are given for N_2 and Ar targets though $\bar{q}$ differs in these gases often by $\Delta\bar{q} \approx 0.5$. Also, the parameters do not allow a smooth transition between Eqs.(3a) and (3b); in the case of 90 MeV iodine ions where $\bar{q}/Z$ is close to 0.3, the two formulas give charge states which differ by $\Delta\bar{q} \approx 1$.

Extensive experimental data on S, As, I and U ions stripped in air at energies between 5 and 80 MeV has been used to modify Eq.(2) as follows:[2,3,5]

$$\bar{q}/Z = 1 - C \exp\left[-V/(V_0 Z^{\gamma}) \right]. \qquad (V \gtrsim V_0) \tag{4a}$$

With empirically determined parameters C and γ which depend slightly on Z (see Table I), the data could be fitted in practically all cases within the experimental errors of ± 0.5 charge states. Substitution of the dependence $\gamma(Z)$ yields[3,5]

$$\bar{q}/Z = 1 - C(bZ^a)^{V/V_0}, \quad (10 \lesssim Z \lesssim 92;\ V \gtrsim V_0) \tag{4b}$$

where a = 0.0667 and b = 0.710. It should be kept in mind, however, that all these semiempirical estimates are useful mainly for interpolation purposes and that extrapolations beyond the investigated ranges of both Z and V may be risky. For example, due to the simplifying substitution $\gamma(Z)$ Eq.(4b) gives too small values for $Z \gtrsim 92$ in gaseous targets. Fig.1 shows a comparison between Eqs.(1)-(4) for the particular case of iodine ions, stripped to equilibrium in targets of N_2, O_2, and air. The experimental data points which are plotted in Fig.1 have been taken from the original tables[3,5-9] and represent most, if not all of the data which has been measured up to date for this case. In general, there is satisfactory agreement between the

Table I. Parameters C and γ (Eq. (4a), ref. 2,3,5).

	Air-Stripper		Formvar-Foilstripper	
Ion	C	γ	C	γ
S	1.135	0.663	1.083	0.604
As	1.117	0.628	1.098	0.538
I	1.065	0.641	1.030	0.518
U	(1.01)	(0.70)	1.030	0.510

experimental data and the semiempirical estimates from Eq.(3) and (4), and, for higher energies, from Eq.(2). The data measured by Ryding et al.[8] lies systematically above the estimates from Eqs.(3) and (4), but it has been found that this shift to higher charge states is due to the density effect which occurs in gases[10] and which is described later on.

Solid Strippers

Several analytical approximations for $\bar{q}$ in solids are available. Eq.(3b) can be used with the modified parameters[4] $\alpha_1 = 0.1$; $\alpha_2 = 0.6$; $m = 1.2$; $n = 5$. No value for A in Eq.(3a) has been given in ref.4, but it is estimated that $A = 0.33$ is a useful approximation for heavy ions stripped in carbon foils in the range $\bar{q}/Z < 0.3$. The semiempirical relations Eqs.(4a) and (4b) hold also for Formvar or carbon strippers when the parameters C and γ are taken from Table I, and when $a = 0.0527$ and $b = 0.714$. Taking into account experimental data at energies above 100 MeV, Nikolaev and Dmitriev developed the following expression:[11]

$$\bar{q}/Z = \left[1 + (V/V'Z^{\alpha})^{-1/k} \right]^{-k} , \quad (Z \gtrsim 20) \qquad (5)$$

where $V' = 3.6 \times 10^8$ cm/sec, $\alpha = 0.45$ and $k = 0.6$. The two formulas for solids, Eqs.(4b) and (5) are also displayed in Fig.1, together with the data available for iodine ions stripped in C and Formvar foils. Eq.(3b) is not shown since it differs from Eq.(5) by less than one unit of charge in the entire range of Fig.1.

It is interesting to point out a shell effect which occurs obviously at $\bar{q} \approx 25$. For that charge, all electrons are stripped off from the N-shell and further stripping requires that those M-electrons be removed which are more tightly bound. The sudden increase in the ionization potential is reflected in a less steep increase of $\bar{q}(V)$. An extrapolation of Eq.(4b) to 100 MeV, for example, overestimates $\bar{q}$ by $\Delta\bar{q} \approx 1.5$. For iodine velocities where $\bar{q} \gtrsim 25$, Eqs.(3b) and (5) are more accurate than Eq.(4b), whereas the opposite is the case in the range where $\bar{q} < 25$.

In summarizing the above discussion, one can state that it is possible to predict mean equilibrium charge states for heavy ions with an average uncertainty of approximately ± 1 units of charge for both gaseous and solid targets. However, the effects of the nuclear charge of the target are often pronounced, and excitation and shell effects may produce noticeable changes which are difficult to predict.

Equilibrium Charge State Distributions

Apart from the large differences in the mean charge which is produced by gaseous and solid strippers, the actual equilibrium charge distributions in both of these target groups depend significantly on the nuclear charge of the target. Fig.2 shows distributions for 12 MeV iodine ions in two gaseous targets (H_2, O_2) and in two solids (C, Au). This particular example is typical in that light targets like H_2 and He produce distributions which are much narrower and more symmetrical than the ones obtained in heavier targets. The differencss in the distribution widths are due to multiple electron loss processes which are much less important in light targets than in heavier ones. A useful approximation for the full e^{-1} width, Γ, of distributions in most targets, except in light gases, is $\Gamma \approx 0.77\,Z^{1/2}$. From this, the corresponding intensity of the most probable charge state can be derived, $F_{max} \approx 1.47\,Z^{-1/2}$. For I (0) ions, Γ and F_{max} amount to 5.6 (2.2) and 20% (50%). The asymmetries which can be found especially at lower ion velocities are not yet understood, and it is interesting to point out that they are not a direct consequence of the presence of cross sections for multiple electron loss.

The average charge in a C target is usually higher than the one in a Au target ($\Delta\bar{q} \lesssim 2$ for I), but the shape of the charge distributions does not differ much and is quite symmetrical for charge states with intensities above ~1%. Other solids generally give mean charges which lie between or close to the values for C and Au. It should be noted that the symmetry of charge distributions can be greatly distorted by shell effects. A good example for such a case has been given by Moak et al.[12]; the equilibrium distribution for 140MeV Br ions stripped in C shows an unusual decrease of charge fractions F_q with $q > 25$. It is believed that this distortion is caused by the difficulty of removing electrons from the L-shell of the ions.

Obviously, for a given ion velocity the highest charge states with intensities close to the possible maximum F_{max} are obtained in light solid media (C,Be). These low Z foils have the additional advantage over heavy targets of scattering the ions to a smaller extent and, thus, of better preserving beam quality. However, the advantages of foil strippers cannot always be exploited when high intensity beams are desired. For heavy ion beam currents of ≳ 1 μA, lifetimes of foils are often only a few minutes. In many practical cases where high beam intensities are present, the only alternative is to use gaseous strippers, either the usual mono- or diatomic gases, vapor jets or very large molecules with atomic weights above ~350 (see below). These gaseous targets are reliable, but they may introduce vacuum problems and require technically a more sophisticated stripping apparatus.

Density Effects

Density Effect in Solids

It has been known from early studies with fission fragments that average equilibrium charges of heavy ions are markedly higher in solid targets as compared with gaseous strippers. A generally accepted qualitative explanation for this density effect has been given by Bohr and Lindhard.[13] They argue that the increase of the mean charge obtained from a solid takes place already inside the solid, though the excitation of the ions in solids may result in a subsequent emission of electrons from the ions immediately after their escape into vacuum, which increases the mean charge to a certain extent.

In a recent publication, however, a quantitatively opposite result has been obtained.[14] It has been argued that the average equilibrium charges of heavy ions inside solids are not much larger than those in gases, and that any observed large difference is mainly due to the emission of Auger electrons after the ions leave the solid. Details of that conjecture are given in ref. 14. Though this new model has not yet been verified experimentally, it is in accord with recent observations concerning ionic excitation.[10] In addition, it is now easier to understand that the stopping power for heavy ions is nearly independent of whether the medium is a gas or a solid.[15]

Density Effect in Gases

It has been known for about 20 years that the equilibrium distribution of charge states in a heavy ion beam penetrating through gaseous media may shift to higher charge states when the pressure of the target gas is increased. The question arises whether this effect which is different from the one in solids, may be utilized to obtain higher charge states for practical purposes. Bohr and Lindhard presented a detailed explanation[13] which included quantitative estimates. The basic assumption in this model is that for high enough densities of the target gas the lifetimes of excited ionic states become comparable with the average free path between two collisions. Since excited electrons can be stripped off more easily, and since electron capture by excited ions may result in subsequent emission of an Auger electron, (i) the effective electron loss cross section increases and (ii) the effective electron capture cross section decreases. Both effects shift the charge distribution to higher charge states with a total maximum increase of $\Delta\bar{q} \approx 0.2\,\bar{q}$.

Recent investigation of this density effect have not confirmed the above model.[10,16] In particular, conjecture (i) turned out to be less significant than anticipated, and the shift of the mean charge was found to be almost constant in the velocity range investigated and amounted typically to $\Delta\bar{q} \approx 1$ (see Fig.3).

At present, charge changing cross sections and atomic lifetimes are not known well enough to allow reliable predictions to be made about the gas densities required to produce the maximum density effect. There is evidence that the theory[13] predicts too short lifetimes for highly ionized excited atoms. For heavy ions stripped at energies below 40 MeV, for example, the density effect has been expected to occur at pressures above ~ 10 torr, but it has been found at much lower pressures of the order of 10^{-2} to 10^{-1} torr.[16] These findings enhance the usefulness of the effect for stripping heavy ions at energies below approximately 40 MeV. However, the effect is not very large and on the basis of our present understanding, one cannot expect a significant increase of $\bar{q}$ when heavy ions are stripped at much higher energies in differentially pumped chambers or in transverse vapor jets which operate usually at pressures up to ~ 1 torr.

Density Effect in Fluorocarbon Strippers

A very promising alternative for increasing the mean charge of heavy ions to values above the ones obtained from dilute or dense gases has been recently described.[17] It was found that gaseous fluorocarbon targets to some extent show the density effect known from solids. This is possible because of the large number of atoms in such a molecule; a typical example is C_8F_{16} with a molecular weight of 400. The effect of that stripper may be illustrated for 12 MeV iodine ions: the average equilibrium charge in dilute O_2, dense O_2 (~ 0.1 torr), C_8F_{16} and in a formvar foil amounts to 4.8, 6.2, 7.6, and 10.5, respectively (see also Fig.2). The fluorocarbon stripper is not as effective as a solid because the molecule is probably not large enough. Still, this new kind of stripper is superior to the usual gases in its stripping efficiency. In addition, very little vapor is required to produce approximate charge state equilibrium. Therefore, a good vacuum can be easily maintained in the stripping region, and beam losses due to scattering are no worse than with the more standard stripping gases and are appreciably smaller than in foils. It is also worth noting that due to the similarity in the mechanisms for the density effect in these large molecules and in foils, one should expect that the advantage of fluorocarbon targets can be exploited at all ion velocities.

High Charge State Tails in Equilibrium Distributions

Most investigators have studied equilibrium distributions for those charge states which showed a relative intensity of more than $\sim 0.1\%$. Only the most intense charge fractions are generally of practical importance; it may be useful, however, to know the abundance of charge states far above the mean charge. In recent experiments, these smaller fractions in the intensity range 10^{-1} to $10^{-6}\%$ have been systematically investigated for Br, Se, and I ions between 6 and 18 MeV,[18] stripped in gases and solids. It was found that the slow decrease of charge fractions for increasing charge states above the mean extends also to intensities below 0.1%. For example, I^{25+} ions have been detected with a relative intensity of $2 \times 10^{-5}\%$, stripped in xenon at 12 MeV, where the average charge is only 5+ (see Fig. 4). It is not fully understood why these high charge states are formed with comparatively high intensities. One observes that these charge fractions emerge from already very thin targets and are slightly scattered off from the forward direction. This points to the importance of close collisions which produce inner shell vacancies followed by Auger cascades.

Charge Changing Cross Sections

Cross sections for electron capture and loss by fast heavy ions in collisions with target atoms are being investigated for almost 40 years. Most of the approaches are contained in the treatments by Bohr,[19] Bohr and Lindhard,[13] and in the review article by Nikolaev.[20] In many particular cases useful results have been obtained, but a satisfactory fundamental understanding has not yet been achieved. Nevertheless, the existing information is complete enough to allow rough estimates to be made in most cases of interest. For example, a semiempirical model has been developed[21] which predicts cross sections for all ions. The accuracy of the calculated values is, of course, limited because various crude approximations have been applied. In particular, cross sections for the loss of several electrons in a single collision have been neglected. Fig. 5 illustrates that especially in heavy targets these events contribute significantly. Fig. 6 shows cross sections calculated for iodine ions,[21] and Fig. 7 gives a comparison of total charge changing cross sections with experimental data.[22]

A question of great practical importance concerns the vacuum requirements for heavy ion accelerators. When ions collide with the residual gas atoms in a vacuum chamber of an accelerator, abrupt changes of the ionic charge lead to beam losses or at least to reduced beam quality. The transmission of beam particles which do not undergo a charge changing collision can be expressed by

$$T = N/N_o = \exp\left(-3.35 \times 10^{16} \int_0^L \sigma_{tot} P \, d\ell\right),$$

where L, P, and σ_{tot} denote the total path length in cm, the residual gas pressure in torr, and the total charge changing cross section in cm^2/molecule. With the semiempirical cross section model[21] mentioned above, it is possible to estimate sufficiently correct pressures P which are required to accelerate heavy ions with a desired transmission. For example, in order to accelerate I or U ions to a final energy of 7 MeV/amu with a transmission of 90%, a vacuum of 10^{-6} torr is required in a linear accelerator with a typical length of 100 m. In a synchrotron, where the ions travel a greater distance for reaching the same high energy, a value $T \sim 0.9$ can be attained only for $P \lesssim 10^{-9}$ torr. However, since effects of accumulated beam losses on the performance of accelerating systems are not yet completely predictable, it may be disputed how much particle loss one can tolerate due to charge changing processes. It should also be noted that σ_{tot} is largest at low ion velocities (see Fig. 7) so that the initial stages of acceleration may be particularly critical.

The study of charge changing cross sections, as well as of charge state distributions for heavy ions is a continuing program in many laboratories. There is reason to believe that our fundamental understanding of collisions between energetic heavy ions and atoms will rapidly improve, providing the answers to many vital questions in this field of growing importance.

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* This work was supported in part by the U.S. Atomic Energy Commission, Contract No. (30-1)-2098.

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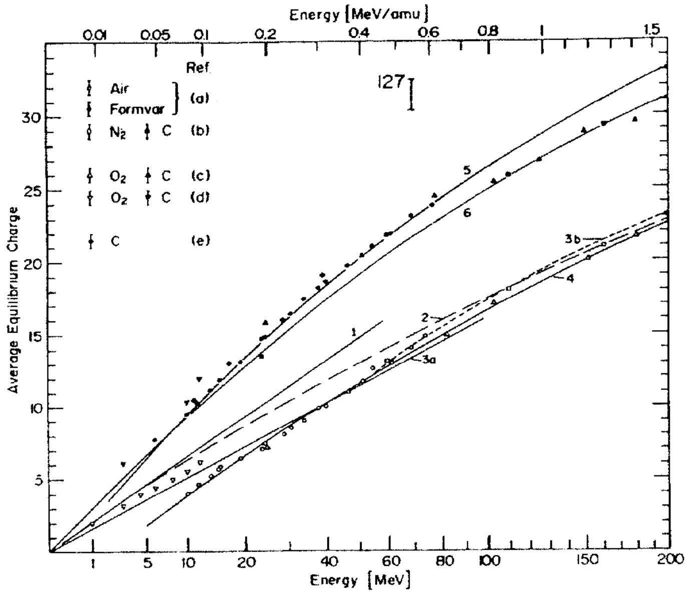

FIG. 1. Average equilibrium charge for iodine ions stripped in gases and solids. Experimental data from ref. a -[3,5]; b -[9]; c -[7]; d -[8]; e -[6]. Theoretical results from Eqs. 1 - (1); 2 - (2); 3a,b - (3a,b); 4,5 - (4b); 6 - (5).

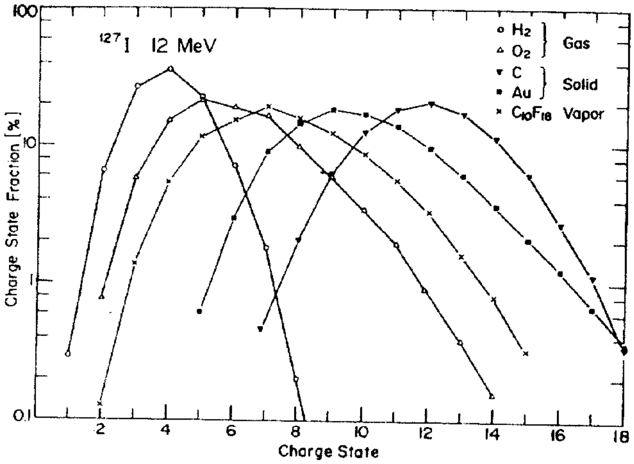

FIG. 2. Equilibrium charge distributions for 12-MeV iodine ions, from ref.[8,17].

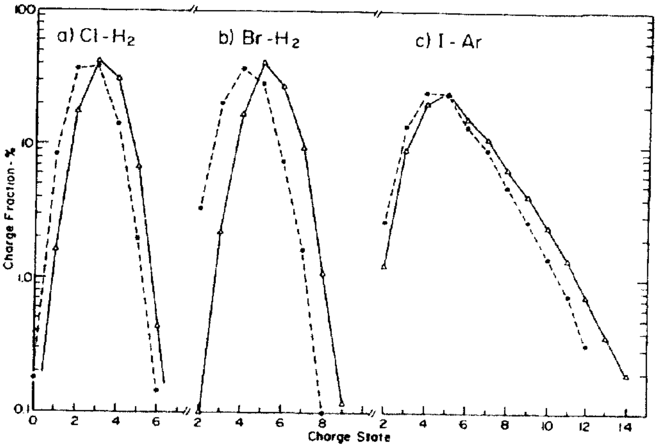

FIG. 3. Equilibrium charge distributions in dilute (●) and dense (Δ) gases for (a) 4-MeV chlorine in H_2, (b) 14-MeV bromine in H_2, and (c) 12-MeV iodine ions in Ar, from ref.[8,10].

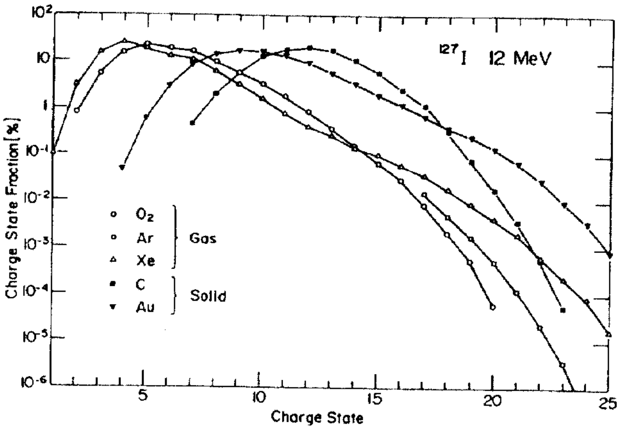

FIG. 4. Equilibrium charge distributions and high charge state tails for 12-MeV iodine ions from ref.[18].

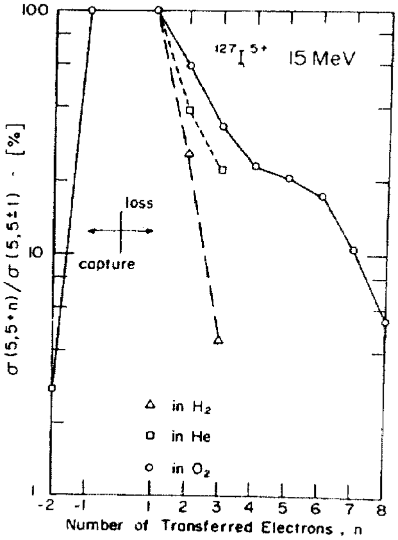

FIG. 5. Relative multiple capture and loss cross sections for 15-MeV iodine 5+ ions.

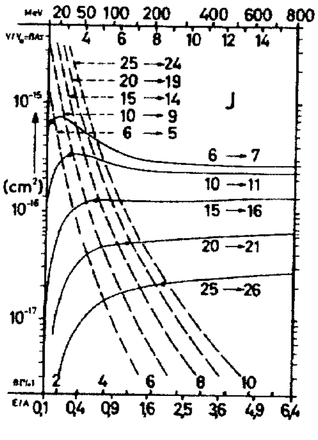

FIG. 6. Calculated charge changing cross sections for iodine ions in N_2, from ref.[21].

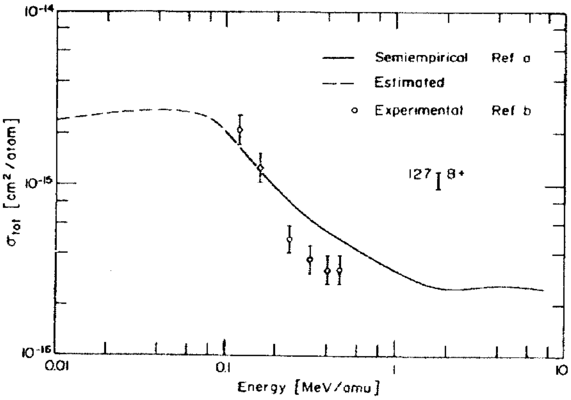

FIG. 7. Experimental and calculated total charge changing cross sections for iodine 8+ ions in N_2, from ref. a -[21]; b -[22].