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## Electron Capture by 40-, 155-, and 600-MeV Protons in Thin Foils of Mylar, Al, Ni, and Ta

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The charge-exchange process has been studied at much higher energies than previously examined. Equilibrium charge distributions have been measured in foils of Mylar, Al, Ni, and Ta for protons of 155 and 600 MeV, and deuterons of 80 MeV. Estimates have been made for the loss cross sections  $\sigma_l$ , and the resulting capture cross sections  $\sigma_c$  are compared with theoretical expectations. Up to 155 MeV,  $\sigma_c$  can be reasonably well accounted for by the non-radiative capture process. Above this energy there are significant departures from this simple behavior which are consistent with the predicted onset of radiative capture. However, because of possible relativistic effects, such an interpretation is not certain. The present data suggest several interesting areas for future theoretical and experimental study.

### INTRODUCTION

There have been many studies made on the equilibrium charge distributions of ions at low energies. By low energies, we mean here energies where there are significant amounts of more than

one charge state (this means a range of less than 1 MeV for protons to several MeV/nucleon for heavy ions). Reviews of the experimental results are given by Allison<sup>1</sup> for protons and  $\alpha$  particles, and by Northcliffe<sup>2</sup> and Nikolaev<sup>3</sup> for heavier ions.

Although there is no completely satisfactory

theory, several semiempirical relationships have been put forth which adequately describe the data, at least under certain limiting conditions.

Phenomenologically, the processes of electron capture and loss have been nicely summarized in a semiclassical way by Bohr.<sup>4</sup> The loss of an electron is closely related to the process of ionization of atoms by charged particles and has been given a quite successful theoretical treatment. The probability of pickup of an electron is more complicated, but involves imparting to the electron being captured a velocity approximately equal to that of the projectile ion. For high energies, this probability decreases rapidly, the stripping process dominates, and one is dealing essentially with a beam of fully charged projectiles.

Nevertheless, there are several reasons why the study of the electron-capture process in these high-energy regions is interesting from a theoretical point of view. For one thing, many of the calculational approaches used, such as the Born approximation, impulse approximation, etc., become more justified and in the limit provide an asymptotic value which can be tested. Also, at high velocities one can restrict consideration essentially to the pickup of 1s electrons from the target atom to the 1s state of a hydrogen-like projectile, thus simplifying considerably the calculations involved. The experimental data in this high-energy region are, on the other hand, much more sparse and, to our knowledge, completely restricted to  $Z=1$  projectiles (i. e., protons and deuterons).

Early theoretical models of the electron-capture process were given by Thomas<sup>5</sup> (classical mechanics) and by Oppenheimer<sup>6</sup> and Brinkman and Kramers<sup>7</sup> (quantum mechanics). The latter is essentially a first Born approximation, considering only the interaction between projectile nucleus and the electron being captured, and we shall conform to the general literature practice of referring to it as OBK. More sophisticated approaches, including a more complete first Born and higher approximations, distorted-wave approximations, impulse approximations, etc., have been treated by a number of authors. A recent review of the various approaches and their degree of success, especially in the high-energy limit, is given by McDowell and Coleman.<sup>8</sup>

There is at present no general consensus as to the correct asymptotic relationship. However, because of its mathematical tractability, the OBK results offer an attractive basis for comparison with other procedures. There appears to be some evidence that most of these other approaches will give values that are proportional, if not equal to, the OBK results in the high-energy limit.

Although there are varying degrees of minor discrepancy, the OBK treatment does appear to

provide a reasonably good description of the experimental cross sections at the highest energies previously tested.<sup>9-11</sup>

There is a second possibility by which an energetic ion may pick up an electron, which is generally ignored in discussing electron capture. This is called radiative capture and is the inverse reaction of the photoelectric effect. In principle, this process can only take place with free electrons. However, at sufficiently high projectile energies, the ion velocity will be considerably greater than the bound electron velocity, and it should be possible to consider the electrons as being free. In effect, the reason given for ignoring radiative capture is that it is calculated to have a much smaller cross section than the normal nonradiative capture process.<sup>12,13</sup>

However, if the energy dependence of this cross section is examined, it is seen to be significantly less steep than the OBK process. It should therefore be expected that at sufficiently high energies this process will dominate the nonradiative one. Although this reaction has never been experimentally seen (except in plasmas<sup>12,13</sup>) the cross sections based on the inverse reaction and assumption of free electrons have sometimes been used in astrophysical calculations.<sup>14-16</sup> Therefore, we thought it would be very important to try to verify this aspect of electron-capture probability.

We have become interested in the electron-capture process in connection with our studies on the production and interaction of cosmic rays.<sup>17</sup> In particular, there are some components of the cosmic rays—those that decay by pure electron capture in the nuclear sense—which, in the absence of orbital electrons, will be completely stable in the interstellar medium. It was our interest to find out to what degree this absence of orbital electrons could be expected to hold.

To this end we have undertaken a systematic program to study the electron capture of various ions in a range of target mediums and at much higher energies than have been looked at before. We present here the results of our studies using protons of 155 MeV (Orsay synchrocyclotron) and 600 MeV (CERN synchrocyclotron). In order to extend the energy range on the low end to a region where previous calculations and experiments have been made, we also made use of the 80-MeV deuteron beam at Orsay. We assume, on the basis of previous evidence,<sup>10</sup> that these are equivalent to 40-MeV protons as far as the charge-exchange process is concerned.

Although we have measured only the equilibrium distributions, we have been able to make estimates of the loss cross section and therefore of the capture cross section.

The choice of our target materials was largely

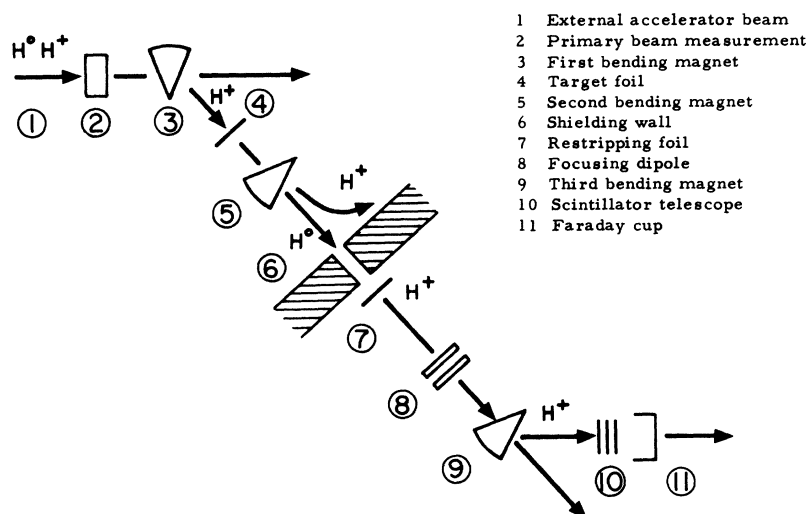


FIG. 1. Schematic representation of experimental setup.

dictated by experimental conditions and consisted of foils of Mylar plastic, Al, Ni, and Ta. The comparison of solids with gases is, also of course, of interest in its own right, especially since there have been several recent proposals as to the source of density effects in the target medium.<sup>18</sup> Some preliminary data that we have obtained on gases suggest there are no large differences for  $Z = 1$  projectile systems, and therefore the present data should be representative in at least giving the gross features of the electron-capture process in this very high-energy region.

It had been our hope that, with these studies, we would be able to make a rough test of existing theoretical predictions and thus permit more accurate predictions for experimentally difficult systems in this higher-energy range. Unfortunately, in extending the present study into the relativistic energy region we have also gotten into a theoretical "no-man's land." Although the additional difficulties thus presented have not yet been satisfactorily accounted for, we believe the experimental data contained here are of sufficient interest to justify publication at the present time. We hope that it will help stimulate further calculations, especially as to relativistic and radiative capture effects.

Coincident with this work, we have made similar (though less complete) studies using helium ions. These results will be published elsewhere.

#### EXPERIMENTAL PROCEDURE

Because the results reported here were obtained during several runs and at two accelerators, there are a number of differences in details of the experimental setup. However, there are basic features in common which are shown schematically in Fig. 1.

The extracted beam from the accelerator was

first focused and passed through a bending magnet to ensure a beam of pure protons. The protons then passed through a thin target foil where equilibrium between  $H^0$  and  $H^+$  was established. The primary beam of  $H^+$  was then deviated by a second magnet, with the neutrals, of course, being unaffected. It was necessary to accomplish this separation as far from the final detection system as possible in order to reduce background problems. After passing through a shielding wall, the  $H^0$  particles were restripped back to  $H^+$  (the equilibrium being such that this is accomplished with virtually 100% efficiency) and analyzed with a third magnet. This last step was needed to further reduce the background, especially from neutrons produced in the target and walls of the second magnet. The particles were finally refocused and passed to the detection system, which was a telescope of plastic scintillators. During initial runs at Orsay, a single scintillator was used, the energy spectrum of the protons giving adequate separation from background. However, for the 600-MeV experiment at CERN, it was necessary to have the lowest possible background and a telescope of three scintillators, about 30 cm apart, was utilized. For real events, we required that these three counters give a fast coincidence signal which was then used to open a linear gate, and the slow energy signal from one of the detectors was stored in a multichannel analyzer. For the smallest cross sections, both the energy and coincidence information were found to be necessary, since even with the triple coincidence requirement there was a background of about 0.5 counts/sec. Most of these were low-energy particles, and by considering only the events under the 600-MeV proton peak, the background was 0.1 counts/sec. (Stated in another way, this background was about

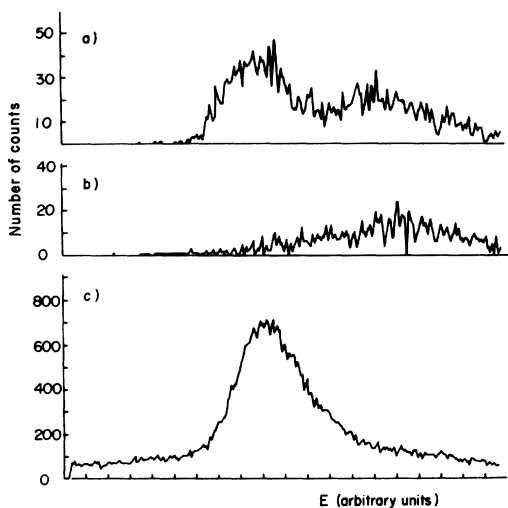


FIG. 2. Coincident gated scintillator spectra at 600 MeV. Peak at right is due to low-energy background events. (a)  $H^0$  component from Ta foil corresponding to integrated primary flux of  $1 \times 10^{15}$  protons. (b) Blank target foil for same flux. (c) Proton calibration taken in ion-source-off mode with second magnet off.

$2 \times 10^{-13}$  of the primary  $H^+$  beam.) Even this background was about equal to the count rate for our light targets, but it was very reproducible and we took sufficiently good statistics to subtract it with confidence. In Fig. 2, we show examples of the spectra for a Ta foil, a background (no target foil), and a calibration peak with 600-MeV protons.

In order to make a calibration with the primary beam, it was necessary, of course, to have a large ( $> 10^6$ ) reduction in the intensity of the primary beam. We accomplished this by turning off the accelerator ion source and operating on the residual beam. In this way we were able to conveniently operate at  $10^3$  counts/sec while retaining all other primary beam characteristics.<sup>19</sup> We also used such an "ion-source-off" mode to align and profile the beam, check coincidence timing, etc.

The method of determining the primary beam current differed at the two accelerators. At Orsay an ionization chamber was installed directly at the output of the accelerator and a Faraday cup behind our scintillator counters. Both before and after a run, the scintillators were taken out of position, the second bending magnet turned off, and the full beam at the detection position measured with the Faraday cup. The ionization chamber was calibrated against this reading, and, thus, at any point during the run we had a measure of the primary beam intensity. Typical beam fluxes were of the order of  $10^{12}$  particles/sec.

At CERN, in addition to finding it impractical to measure the 600-MeV beam with a Faraday cup, we were not, because of radiation safety require-

ments, allowed to have the full external beam at the position of our scintillators. We, therefore, made our primary beam measurement just before the first bending magnet using the standard induction counter of the machine. This is estimated to have an absolute accuracy better than 20% and a relative precision of less than 5%. In order to measure the transmission from that point to our scintillators, we profiled the beam at the output of the shielding wall, using the ion-source-off mode. These measurements indicated that we were getting essentially 100% transmission, and the beam characteristics were very close to those predicted using theoretical calculations.

Since we are dealing with an extremely small component of the beam, it is of course important to ensure that what we measure are really  $H^0$  particles. To this end we have made several tests to try to eliminate any other possibilities. First, our physical arrangement is such that, in principle at least, only a particle originally charged, becoming neutral in the target, and finally becoming charged again will be able to successfully negotiate the three magnetic fields. We have replaced the target foil with a blank target holder, and used this as a background correction. Such a correction was entirely negligible except at 600 MeV, in which case it amounted to almost 50% for the lightest target. We also scanned with the third magnet in order to ensure that there was a peak at the field corresponding to the full-energy protons. In fact, the background at other values of the field was essentially identical to a "target-out" background. Probably the most likely source of this background is due to primary beam protons which are scattered in such a way as to pass through the second magnet. (We have in fact seen such a phenomenon when the gas pressure in the region of the second magnet becomes too high.) The most dramatic way we found for showing that we had truly neutral particles was by removing the stripping foil after the second magnet and observing that our count rate was again essentially equal to background. Unfortunately, such a test was not available to us at CERN, because the vacuum line did not extend past the third magnet and thus the vacuum window was the stripping foil. There a further check against possible scattering in the target foil was made by using two Al foils differing by a factor of 50 in thickness. If scattering or any other type of reaction in the foil were responsible, one would expect to see this reflected proportionally in the count rate. We observed the same count rate with both foils.

For the results reported here, the targets foils were sufficiently thick for complete equilibrium to be obtained. In order to try to measure the absolute capture cross section, we attempted to

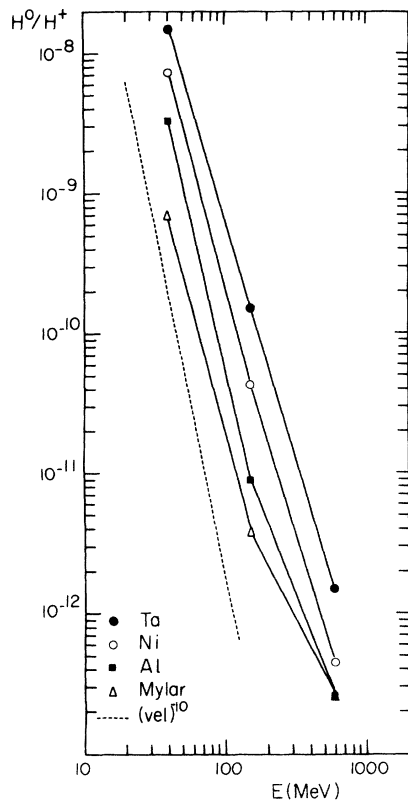


FIG. 3. Measured equilibrium ratios ( $H^0/H^+$ ) as function of energy. Lowest energy points are from deuteron run plotted at equivalent proton velocity.

utilize thinner foils, and therefore measure a ratio before equilibrium was reached. We were severely limited in the thickness of foil we could use due to experimental difficulties. Because of the necessity of having maximum intensity, we could not collimate the primary beam. To avoid contributions due to the target holder we thus required a target foil of approximately  $10 \times 10$  cm. The thinnest foil we were successfully able to use under these conditions was an aluminum one  $0.77 \mu$  ( $4.6 \times 10^{18}$  atoms/cm<sup>2</sup>) thick. Even with such a foil we were unable to see any variation in the  $H^0/H^+$  ratio to within our experimental precision (10%). This is consistent with estimates of the cross sections made by other means, as described further on.

### RESULTS

The results of our measured equilibrium distributions for the three energies are shown in Fig. 3. The lowest energy points are those measured with 80-MeV deuterons and are plotted at the equivalent proton velocity. The straight lines joining the points are given only as a guide to the eye and are not meant to represent the actual slopes of the curves.

We have not indicated any errors in Fig. 3 because they include two types which must be considered separately. The only truly random errors are those due to counting statistics. In all cases these were less than 10%. In addition, there are those systematic errors such as absolute primary beam measurement, transmission efficiency, and absolute energy which contribute to all measurements of a given run in the same way. These must be considered in an absolute sense and in making a comparison between energies, but not in comparing different targets at the same energy. We estimate these errors to be 30% for the  $H^0/H^+$  ratio and 1% in the energy determination. We have not made direct energy measurements during our experiments, but have relied upon previously established determinations of the extraction energies of the respective machines, with small corrections made for losses in thin window foils where necessary.

One should note at first that the fraction  $H^0/H^+$  is extremely small and falls rapidly with energy. A curve with  $v^{-10}$  dependence is shown for comparison. Such a dependence is in accord with the OBK predictions in the asymptotic limit, although various other models have suggested anywhere from a  $v^{-2}$  to  $v^{-10}$  dependence. It can also be noted that, with the exception of the 600-MeV points for Mylar and Al, the lines are essentially parallel. That is, the energy dependence is roughly the same for all four target materials. The importance of the exceptions will be discussed later.

In order to simplify the comparison of these results with various theoretical predictions, we have transformed the measured equilibrium distributions to capture cross sections ( $\sigma_c$ ). For essentially single-component systems, the equilibrium ratio ( $R$ ) is given by

$$R = \frac{[H^0]}{[H^+]} = \frac{\sigma_c}{\sigma_i} \quad (1)$$

By knowing or estimating the loss cross section ( $\sigma_i$ ), we are thus able to find  $\sigma_c$ . The difficulty is that the highest energy for which  $\sigma_i$  has been measured is for 17.9-MeV protons and does not include any solid targets or the range of  $Z$  that we consider in the present paper.<sup>20,21</sup> We are therefore forced to make a number of interpolations and extrapolations. It should be stressed that while these estimations introduce some uncertainty, the situation is actually not as bad as might first appear. There are two reasons for this. First, the process of electron loss is rather well accounted for by theoretical calculations and thus extrapolated estimates are expected to be fairly reliable.<sup>20,21</sup>

More important is the fact that  $\sigma_i$  is much less sensitive than  $\sigma_c$  to both energy and the  $Z$  of the

TABLE I. Experimental and theoretical capture cross sections.

Target	Energy (MeV)	$\sigma_c$ (OBK) <sup>a</sup> (cm <sup>2</sup> /atom)	$\sigma_c$ (rad) <sup>b</sup> (cm <sup>2</sup> /atom)	$\sigma_c/\sigma_i$ <sup>c</sup>	$\sigma_i$ <sup>d</sup> (cm <sup>2</sup> /atom)	$\sigma_c$ <sup>e</sup> (cm <sup>2</sup> /atom)
Mylar	40	$6.2 \times 10^{-27}$	$8.4 \times 10^{-29}$	$6.8 \times 10^{-10}$	$3.5 \times 10^{-18}$	$2.4 \times 10^{-27}$
	155	$2.9 \times 10^{-30}$	$3.1 \times 10^{-30}$	$3.8 \times 10^{-12}$	$9.0 \times 10^{-19}$	$3.4 \times 10^{-30}$
	600	$7.7 \times 10^{-34}$	$1.0 \times 10^{-31}$	$2.7 \times 10^{-13}$	$2.3 \times 10^{-19}$	$6.1 \times 10^{-32}$
Al	40	$7.5 \times 10^{-26}$	$1.8 \times 10^{-28}$	$3.3 \times 10^{-9}$	$9.0 \times 10^{-18}$	$3.0 \times 10^{-26}$
	155	$5.6 \times 10^{-29}$	$6.6 \times 10^{-30}$	$8.8 \times 10^{-12}$	$2.5 \times 10^{-18}$	$2.2 \times 10^{-29}$
	600	$1.7 \times 10^{-32}$	$2.2 \times 10^{-31}$	$2.6 \times 10^{-13}$	$6.0 \times 10^{-19}$	$1.6 \times 10^{-31}$
Ni	40	$1.8 \times 10^{-25}$	$3.9 \times 10^{-28}$	$7.4 \times 10^{-9}$	$1.9 \times 10^{-17}$	$1.4 \times 10^{-25}$
	155	$1.0 \times 10^{-27}$	$1.4 \times 10^{-29}$	$4.3 \times 10^{-11}$	$5.4 \times 10^{-18}$	$2.3 \times 10^{-28}$
	600	$6.1 \times 10^{-31}$	$4.8 \times 10^{-31}$	$4.5 \times 10^{-13}$	$1.4 \times 10^{-18}$	$6.3 \times 10^{-31}$
Ta	40	$4.9 \times 10^{-28}$	$1.0 \times 10^{-27}$	$1.5 \times 10^{-8}$	$3.6 \times 10^{-17}$	$5.4 \times 10^{-25}$
	155	$7.1 \times 10^{-28}$	$3.7 \times 10^{-29}$	$1.4 \times 10^{-10}$	$1.0 \times 10^{-17}$	$1.4 \times 10^{-27}$
	600	$1.4 \times 10^{-29}$	$1.2 \times 10^{-30}$	$1.5 \times 10^{-12}$	$2.5 \times 10^{-18}$	$3.8 \times 10^{-30}$
N <sub>2</sub>	37.5					$2.7 \times 10^{-27}$ (Ref. 11)
Ar	37.5					$8.7 \times 10^{-26}$ (Ref. 11)

<sup>a</sup>OBK cross sections for capture of 1s electrons from target atom as given by Eq. (2).

<sup>b</sup>Radiative capture cross sections as given by Eq. (3) assuming all electrons in target are free.

<sup>c</sup>Measured ratio H<sup>0</sup>/H<sup>+</sup>.

<sup>d</sup>Extrapolated loss cross sections as described in text.

<sup>e</sup>Calculated capture cross sections using columns 5 and 6.

target atom. Thus, the values of  $R$  reflect, for the most part, differences due to  $\sigma_c$  and only weakly depend on  $\sigma_i$ . In particular, the energy dependence will be overwhelmingly dominated by that of  $\sigma_c$ . The values we have used for  $\sigma_i$  are given in Table I. In each case we have taken the highest-energy experimental values available (interpolated or extrapolated to the appropriate  $Z$  using the  $Z^{2/3}$  dependence suggested by Bohr<sup>4</sup>) and extrapolated to the desired energy by assuming a  $v^{-2}$  dependence.

Such a situation is, of course, far from being completely satisfactory and the resulting uncertainties in the absolute values of  $\sigma_c$  are significant. Nevertheless, the relative values as a function of energy should be much less affected.

The values of  $\sigma_c$  obtained in this way are tabulated in Table I and plotted in Figs. 4 and 5. It is interesting to note that, although we are considering an atomic process, the cross sections involved are considerably smaller than typical nuclear cross sections and are four orders of magnitude smaller than any previously measured electron-capture cross sections.

For the reasons mentioned above, we have again not shown errors in Figs. 4 and 5. These will include the statistical and systematic errors already noted but, much more importantly, they will also include the error in estimating  $\sigma_i$ . It is difficult to assign a value to the latter, but we believe that the errors are probably no worse than a factor of 2 on an absolute basis and considerably

better on a relative one.

The only available experimental results that can be compared with those in the present paper are those of Acerbi *et al.*<sup>11</sup> for 37.5-MeV protons in N<sub>2</sub> and Ar gases which are also given in Table I. If we compare our results for deuterons in Mylar at the same velocity, we find surprisingly good agreement. This is quite gratifying considering the many possible sources of discrepancy.

Similarly, the value for Ar gas falls rather nicely between our values for Al and Ni. Such agreement gives us some confidence in the method we have used to obtain  $\sigma_c$  from  $R$ .

#### THEORETICAL COMPARISON AND DISCUSSION

The probability for nonradiative electron capture is given by<sup>7</sup>

$$\sigma_c(\text{OBK}) = \pi a_0^2 (2^{18}/5) Z^5 Z'^5 S^8 [S^2 + (Z + Z')^2]^{-5} \times [S^2 + (Z - Z')^2]^{-5}, \quad (2)$$

where  $Z$  and  $Z'$  equal projectile and target charge, respectively,  $S$  equals projectile velocity in terms of the electron velocity of hydrogen, and  $a_0^2 = 2.8 \times 10^{-17}$ .

The basis for this expression is essentially a first-Born-approximation calculation neglecting all but the projectile-electron interaction and should be most accurate in the energy region where the projectile velocity is much greater than that of the electron being captured. As written, the

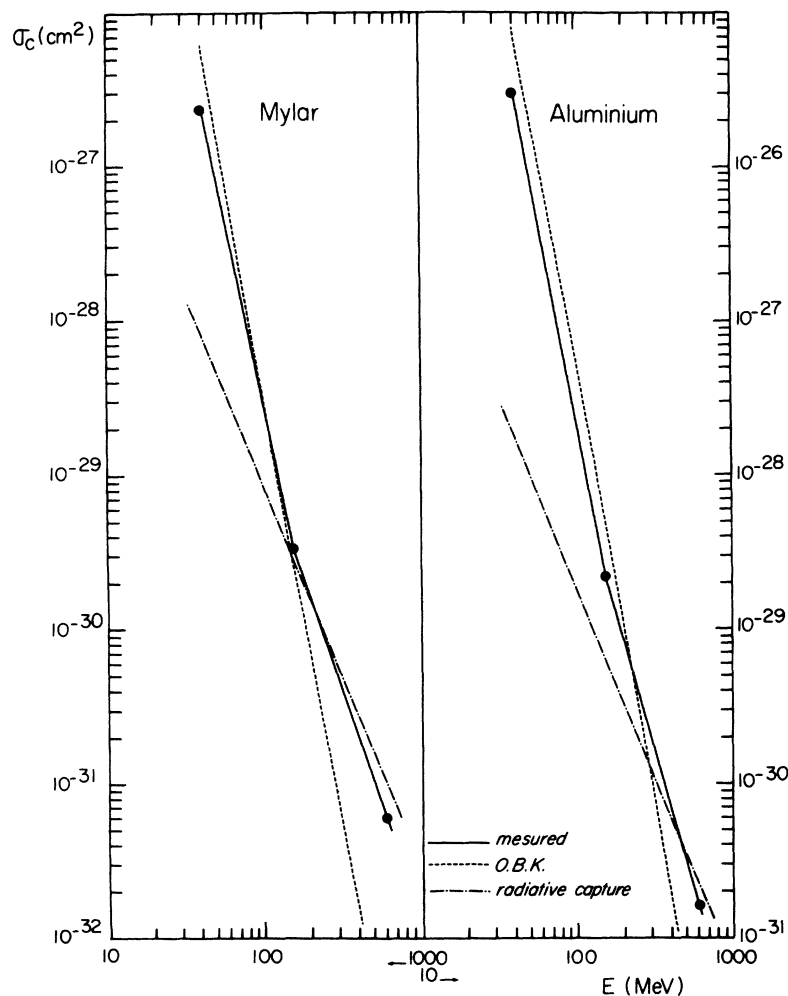


FIG. 4. Experimental and theoretical capture cross sections for Mylar and Al, calculated as described in text.

probability is only for capture into the  $1s$  state of the projectile and should be multiplied by 1.23 to take into account capture into higher  $s$  states.<sup>6</sup> States of higher angular momentum are neglected.

There have been a number of more detailed approaches to the problem including higher Born approximations, impulse approximations, and distorted-wave calculations. Although there is still considerable controversy on the subject, there is as yet no complete agreement on the correct high-energy asymptotic value. Probably the most extensive investigations in this area have been made by Mapleton, using various forms of the Born approximation.<sup>22</sup> He concludes that the asymptotic value of this approximation will become proportional to the OBK values, although only at quite high energies. Other authors argue that the second Born term is significant, and will lead eventually in the asymptotic limit to a  $v^{-11}$  dependence.<sup>23-25</sup>

From a practical point of view, the OBK type of treatment appears to give reasonable agree-

ment with experiment at the highest energies previously tested. The model has the merit of being relatively simple mathematically, and for our purposes we are most interested in seeing if the gross features of the predicted behavior are correct.

In this context the most important features are the velocity dependence and the dependence on target charge. At sufficiently high energies where the projectile velocity is much greater than the electron velocity of the target atom (i. e.,  $S \gg Z'$ ), Eq. (2) shows that these dependences become  $v^{-12}$  and  $Z'^5$ , respectively.

In Table I and Figs. 4 and 5 we have plotted the OBK cross sections calculated using Eq. (2) and considering only capture from the  $1s$  state of the target atom. This approximation is only valid when the projectile ion velocity is greater than the  $1s$  electron velocity. In Table II the velocity of the  $K$  electrons in each of the target materials is given, as well as that of the proton at the various energies. It can be noted that, except for Ta, the

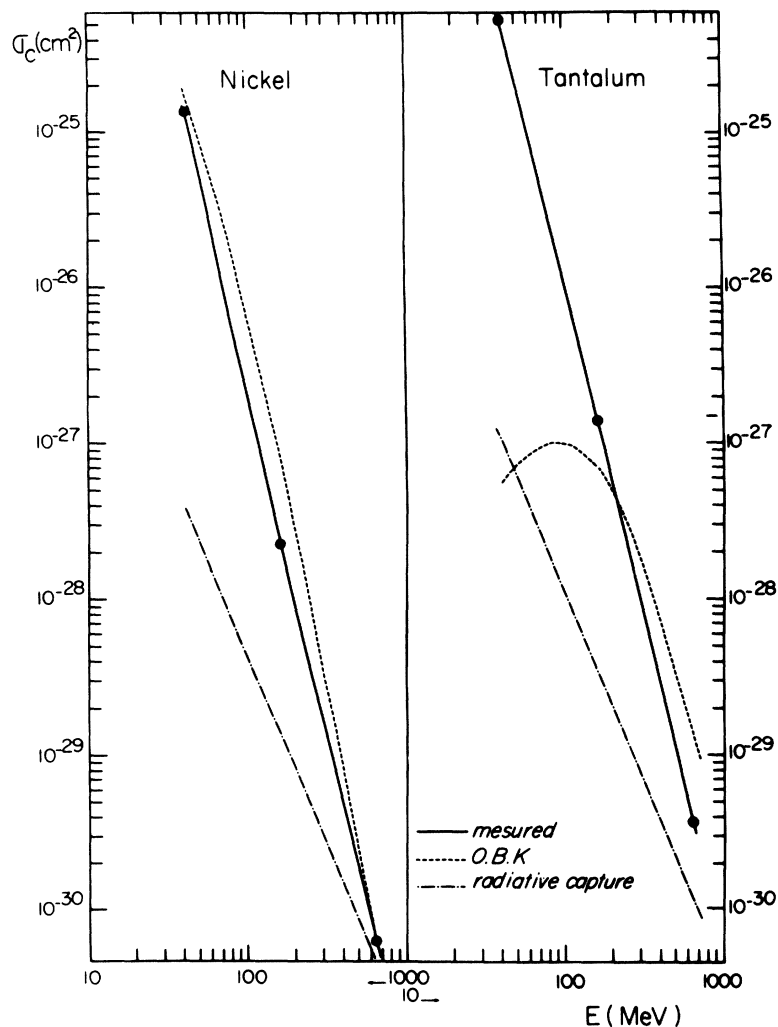


FIG. 5. Experimental and theoretical capture cross sections for Ni and Ta, calculated as described in text.

proton velocity is greater than that of the  $K$  electron at all energies in question. For Ta, the proton velocity falls below the  $1s$  electron velocity at about 125 MeV and is reflected by the rapid decrease in the calculated cross section shown in Fig. 5. This is not intended to imply that the total cross section should be expected to fall also, since, of course, at that point one begins to pick up  $2s$  and  $2p$  electrons. The calculation for these processes becomes more involved and we have not made them here. The turnover in Fig. 5 is shown merely to illustrate that the departure from the calculated  $1s$  capture, as measured, comes at about the velocity that one would predict (i. e., where the projectile velocity falls below the  $1s$  electron velocity).

The calculations for the Mylar target have been made on the assumption that it consists essentially of carbon ( $\frac{2}{3}$ ) and oxygen ( $\frac{1}{3}$ ) atoms. (Because both  $\sigma_e$  and  $\sigma_p$  for hydrogen are much smaller in comparison, it can be neglected.) The carbon-oxygen

composite turns out to give values very close to a  $Z = 7$  target (i. e., nitrogen).

In general, the agreement between the measured and OBK cross sections can be considered as quite satisfactory, considering the many assumptions made and the large experimental uncertainties. The slopes of the curves are fairly accurately reproduced except for two notable exceptions which we discuss below. The fact that the OBK predictions generally appear to be somewhat higher than the experimental points may lend support to Mapleton's argument<sup>22</sup> that the correct Born approximation approaches the OBK limit only very slowly, even at quite high energies.

The OBK cross sections, as given, have been made without any consideration of relativistic effects being taken into account. Since the  $\beta$  of 600-MeV protons is 0.79, this is clearly a serious omission. We know of only one paper where the possible relativistic effects on electron capture have been considered. Mittleman<sup>26</sup> has made cal-



TABLE II. Projectile and *K*-electron velocities.

Projectile	Target	<i>K</i> -electron velocity ( $\beta$ )	Projectile velocity ( $\beta$ )	Projectile velocity <i>K</i> -electron velocity
80-MeV $^2\text{H}$	Mylar <sup>a</sup>	0.040	0.283	7.1
	Al	0.078		3.6
	Ni	0.179		1.6
	Ta	0.469		0.6
155-MeV $^1\text{H}$	Mylar		0.513	12.8
	Al			6.6
	Ni			2.9
	Ta			1.1
600-MeV $^1\text{H}$	Mylar		0.792	19.8
	Al			10.0
	Ni			4.4
	Ta			1.7

<sup>a</sup>An average *K*-electron velocity for  $\text{N}_2$  has been assumed for Mylar.

culations for 10-MeV protons in  $\text{H}_2$  and concluded that the relativistic effect at this energy was only about 3%. He has also proposed, however, that in the ultrarelativistic energy region, the energy dependence will become equal to  $E^{-1}$ .

Mapleton has given calculations up to 100-MeV protons in various gases without making any corrections.<sup>22</sup> The highest previous experiments at 37.5 MeV<sup>11</sup> indicate rather good agreement with calculations without any large correction apparently needed.

In making relativistic corrections, there are several factors to consider. The most obvious is that one must use a correct relativistic velocity for the projectile ion. Since the capture probability is so strongly sensitive to velocity, this may be an important factor even for small corrections. In addition, one must use a correct relativistic velocity for the electron being captured. One would not normally consider this an important effect except for the very heaviest elements. However, as has been pointed out by Mittleman,<sup>26</sup> it is precisely the highest-energy part of the electron distribution that one is probing during capture at high projectile velocities. This, in turn, means that one needs to substitute the Dirac equation for the Schrödinger equation in the development of the OBK formalism in order to have a correct description of the high-energy portion of the electron distribution. Finally, the interaction should properly be treated using relativistic quantum electrodynamics.

Although we are examining these problems, we cannot at present give any quantitative estimate as to their effect on the calculated values. It appears, however, that at 600 MeV such corrections will be substantial and the nonrelativistic values given here must be taken with this in mind. For example, by simply inserting the correct relativistic velocity for the projectile ion at 600 MeV, one changes the value calculated from Eq. (2) by as much as two orders of magnitude.

It is quite conceivable that the corrections mentioned could account for the rather dramatic change in slope measured for  $\sigma_c$  between 150 and 600 MeV in Mylar and, to a lesser extent, aluminum. Similar effects are not seen for Ni and Ta but it is not at all obvious that the effects will be of equal importance for different *Z* targets. Further calculations in this area would be quite interesting.

A second possible explanation for the change in slope at high energies is that radiative capture has become important. This process involves the simultaneous capture of an electron and the emission of a high-energy photon. (It should not be confused with the sequential capture into an excited state of the projectile ion followed by radiative decay. That process can be roughly taken into account by the  $n^{-3}$  rule of Oppenheimer,<sup>6</sup> and leads only to about a 20% increase in the nonradiative capture probability.) The cross section for radiative capture can be calculated by considering the inverse reaction, the photoelectric effect, and doing detailed balance with the appropriate density of states. Momentum conservation in such a reaction is preserved by the photon, and thus this process can occur with "free" electrons. When the projectile ion has a velocity much larger than the bound electron, it should be adequate to consider them as free. At lower projectile velocities, it is not known what effect the binding will have on the cross section for such a process.

The possibility of radiative capture has generally been ignored by those authors who calculate electron-capture probabilities. Presumably this is because the cross sections, for all energies previously considered, are calculated to be much smaller than the corresponding nonradiative capture cross sections. Nevertheless, the cross section for radiative capture has a weaker energy dependence, becoming  $E^{-1}$  in the relativistic limit, so it is possible that at sufficiently high energies it could become larger than the nonradiative capture. In fact, for some problems of astrophysical interest, several authors have made calculations on the basis outlined above, with the assumption that the nonradiative capture would be negligible in comparison to the radiative<sup>14-16</sup> (without—it might be noted, however—taking into consideration the possible relativistic effects on the OBK cross sections, as suggested above).

In Table I and Figs. 4 and 5, we show the radiative capture cross sections, calculated in the following manner. All the electrons of the target atom have been assumed to be "free." As was shown before, the proton velocity is almost always larger than even the inner-shell electrons, although not always *much* larger, as may be required. Using the equation for the relativistic photo effect,<sup>27</sup> the cross section per electron is given by

$$\sigma_c(\text{rad}) = \frac{3}{2} \frac{Z^5}{137^4} \phi_0 \frac{k^2}{(k+\mu)^2 - \mu^2} \left(\frac{u}{k}\right)^5 (\gamma^2 - 1)^{3/2} \times \left[ \frac{4}{3} + \frac{\gamma(\gamma-2)}{\gamma+1} \left( 1 - \frac{1}{2^\gamma(\gamma^2-1)} \ln \frac{\gamma+(\gamma^2+1)}{\gamma-(\gamma^2-1)} \right) \right], \quad (3)$$

where  $\phi_0 = 6.65 \times 10^{-3} \text{ cm}^2$ ,  $\mu$  is the electron rest mass,  $k$  is the electron kinetic energy in the rest frame of the projectile, and  $\gamma = (k + \mu)/\mu$ .

One notes immediately in Figs. 4 and 5 the importance of  $\sigma_c(\text{rad})$ . For Mylar and aluminum,  $\sigma_c(\text{rad})$  becomes larger than  $\sigma_c(\text{OBK})$  for our highest energy points. For Ni and Ta, it is never as large. Of course, this difference is easily understood when one realizes that  $\sigma_c(\text{rad})$  is dependent only on the number of electrons in the target atom (i. e.,  $Z'$ ), while  $\sigma_c(\text{OBK})$  approaches a  $Z'^5$  dependence in the high-energy limit. The measured cross sections, in fact, appear to reflect very nicely the predicted changes in slope due to the onset of radiative capture. Thus, while because of the difficulties mentioned previously with regard to making relativistic corrections, we cannot say definitely that what we observe is radiative capture, the observations are at least not inconsistent with such a hypothesis.

#### CONCLUSIONS

We believe we have demonstrated in the present paper that although there are formidable experimental problems, it is possible to measure electron-capture cross sections well into the relativistic energy region. Such an extension presents both new difficulties and some interesting features.

It has been our goal to explore the gross features of the process in this energy range, and to try initially to make a rough comparison with existing theory. Although there are minor discrepancies, it appears that these features can be adequately accounted for by the OBK approach up to 150 MeV. Above this energy we do not feel that meaningful comparisons can be made until the appropriate relativistic corrections are made on the nonradiative capture expression. Our data are consistent with (but not conclusive of) the radiative capture process becoming dominant at the high energies.

Several additional pieces of experimental information would certainly be desirable. It would obviously be preferable to be able to make a direct measurement of  $\sigma_c$ , rather than just the equilibrium distribution. This probably will require a measurement in gases. Such measurements necessitate tight collimation and present significant

experimental difficulties with high-energy beams and very small cross sections which we are considering. Nevertheless, we have recently begun a series of measurements of this type.

Second, it would be interesting to have measurements at several energies in the relativistic region in order to establish the energy dependence more accurately. It is interesting to note that the relativistic limits in both the nonradiative and radiative cases predict a  $E^{-1}$  dependence. Because of the different target- $Z$  dependences, however, an experiment with several different targets should be able to decide which is the dominating process.

Last, it would be very desirable to have similar information on heavier projectile systems. Both  $\sigma_c(\text{OBK})$  (in the high-energy limit) and  $\sigma_c(\text{rad})$  have a  $Z^5$  dependence on projectile charge. To date however, there are virtually no experimental data in the energy region where such a dependence could be tested. We have completed some experiments on He ions at 100 MeV.<sup>28</sup> Unfortunately, the prospects for data on heavier ions is not immediately promising, since even the presently anticipated heavy-ion accelerators give energies considerably lower than the region where the approximations used are expected to be valid.

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## Range-Energy Relations for Helium Ions and Protons in Ar, N<sub>2</sub>, O<sub>2</sub>, and Air (0.2-2.0 MeV)\*

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The ionization extrapolated ranges of H<sup>+</sup> and He<sup>+</sup> in Ar, N<sub>2</sub>, O<sub>2</sub>, and air have been measured from 200 keV to 2.0 MeV with an accuracy of  $\pm 1\%$ . A magnetically analyzed beam of ions was brought through a differential pumping system into a gas stopping cell. A single-grid ionization chamber, movable parallel to the beam axis, was used to obtain the data necessary to plot a Bragg curve. The observed range-energy relations are compared with previous data where possible.

### INTRODUCTION

Range-energy relations of protons and  $\alpha$  particles in gases were studied extensively prior to 1954,<sup>1-5</sup> although since that time the main thrust of range-energy-relation studies has been directed toward the study of energetic particles penetrating solids.

Range-energy relations are of importance in various fields of science, e.g., nuclear safety, nuclear medicine, and atmospheric explosions. The purpose of this investigation was to measure the range-energy relations for protons and  $\alpha$  particles in Ar, N<sub>2</sub>, O<sub>2</sub>, and air. This paper describes the experimental apparatus and procedure, presents the collected data, and discusses and compares these results with previously measured and calculated results.

An extensive discussion of the various ranges and associated terms is given by Holloway and Living-

ston.<sup>3</sup> The ionization extrapolated range was determined by impinging a monoenergetic beam of ions on a gas target and obtaining a Bragg curve. The Bragg curve was determined by plotting the ionization produced by the beam in a collection chamber vs the distance of the collection chamber from the entrance aperture of the gas cell. By drawing the steepest tangent to this curve and extrapolating to zero ionization current, the ionization extrapolated range is obtained. Unless otherwise noted, "range" means the ionization extrapolated range in centimeters at 15 °C temperature and 760 Torr. To obtain the range in units of mg/cm<sup>2</sup>, multiply the range in centimeters by 0.0424 times the molecular weight of the target.

### EXPERIMENTAL APPARATUS AND PROCEDURE

The ion beam was produced by a Van de Graaff