Electron Capture and Loss by Moving Ions in Dense Media

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Several experiments have shown an anomaly in the stopping power which appears to be greater for incident heavy particles in condensed media than in gases. It is suggested that this anomaly is characteristic of incident ions, i.e., of particles that are not completely stripped of orbital electrons, and it seems to be due to the reaction of the perturbed medium upon the moving ion having charge Ze, which reaction results in a field $F=(1/Ze)(dW/dz)$ acting against the motion of the ion (dW/dz is the energy loss per cm). Although F is relatively weak in gases, it is substantial in condensed media and may cause the spontaneous emission of electrons carried by the ion (autoionization), thus increasing the ions' effective charge. The higher effective charge accounts for the higher stopping power in condensed media. It has been determined that when an incident helium atom passing through a liquid undergoes a collision, thus emerging in a singly ionized and excited state, it cannot exist in such a state since it becomes instantly doubly ionized. However, the same type of collision occurring in a gas leaves the atom singly ionized. The effective charge of the incident particle depends upon the values ϕ_0, ϕ_1, and ϕ_2, representing the probability of the particle's being neutral, singly ionized, and doubly ionized, respectively, where $\phi_0:\phi_1:\phi_2=\sigma_{10}\sigma_{21}:\sigma_{01}\sigma_{21}:\sigma_{01}\sigma_{12}$ (σ_{if} represents the cross section for electron loss or capture; i and f represent the charge before and after the event, respectively). It is shown that σ_{if} is higher for a condensed medium than for a gas when $i<f$ and lower when $i>f$. σ_{if} has been calculated for He passing through argon gas at 400 kev, and gives roughly $\phi_0:\phi_1:\phi_2=0.05:0.75:0.20$; thus, an average square charge is $\langle e^2\rangle_{\text{Av}}=1.55e^2$. If autoionization is taken into account, σ_{if} for He in liquid argon gives roughly $\phi_0:\phi_1:\phi_2=0.04:0.70:0.26$, and thus $\langle e^2\rangle_{\text{Av}}=1.74e^2$.

I. INTRODUCTION

In the past several years attention has been called to an apparent anomaly in the behavior of an alpha particle passing through water. Early experiments performed by Michl[1] and Philipp[2] and confirmed more recently by Appleyard[3] have established that water in the liquid state has a stopping power for alpha particles higher by 10–20 percent than that of water vapor. Consequently, the Bragg rule which claims additivity of atomic stopping power for all media is questioned.

In contrast to the aforementioned work, two experiments appear to throw a cloud of uncertainty on the situation. For example, de Carvalho[4] reaches the conclusion that the stopping power for alpha particles is the same for vapors and condensed media, and similar results have been obtained by Ellis, Rossi, and Failla.[5,6] Platzman[7] has given a critical review of the experiments and discussed the influence of electronic binding on the stopping power.

It is difficult on the basis of the few and discordant experimental studies that have been made to reach a conclusion about the existence and the possible extent of the anomaly. The purpose of this paper is to point out a factor that may account in part for the larger stopping power for ions moving in condensed media.

II. EFFECT OF THE MEDIUM

A. General

It is suggested that the anomaly in the stopping power is due to a "density effect" of the surrounding medium upon a moving particle. The "density effect" referred to here is of an entirely different nature than the "density effect" suggested by Swann,[8] the theory of which has been given by Fermi.[9] The latter density effect accounts for a decreased stopping power in condensed media, while we are concerned with an increased stopping power. Furthermore, the Swann-Fermi density effect is characteristic of particles having relativistic velocities, while we are interested in particles moving rather slowly.

We are concerned here with particles that are not completely stripped of orbital electrons and which successively lose and capture electrons while colliding with the surrounding atoms. Because of the "density effect," an ion of a given velocity has a higher effective charge when traversing a medium in a condensed state (solid or liquid) than when traversing a medium of the same composition but in the gaseous state. The higher effective charge may account for the higher stopping power of ions traversing condensed media and thus give a plausible explanation of the anomaly.

The dependence of the charge of a moving ion upon the character of the surrounding medium has been studied experimentally by Lassen,[10] who has shown that a fission fragment in various gases has a much lower average charge than a fission fragment of the same velocity passing through a solid. The results of Lassen are significant because they confirm the effect of the density of the medium on the charge of the particle. Furthermore, they corroborate the existence of the anomaly in the stopping power, at least insofar as they relate to fission fragments.

Further confirmation of the effect of the density of the medium on a moving ion may be obtained from the

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[2] K. Philipp, Z. Physik **17**, 23 (1923).
[3] R. K. Appleyard, Proc. Cambridge Phil. Soc. **47**, 443 (1950).
[4] H. G. de Carvalho, Phys. Rev. **78**, 330 (1950).
[5] Ellis, Rossi, and Failla, Phys. Rev. **86**, 562 (1952).
[6] R. H. Ellis, Jr., Rev. Sci. Instr. **25**, 336 (1954).
[7] R. L. Platzman, *Symposium on Radiobiology*, edited by J. J. Nickson (John Wiley & Sons, New York, 1952), p. 139.
[8] W. F. G. Swann, J. Franklin Inst. **226**, 598 (1938).
[9] E. Fermi, Phys. Rev. **57**, 485 (1940).
[10] N. O. Lassen, Kgl. Danske Videnskab. Selskab, mat.-fys. Medd. **26**, 5 (1951).

experiments of Allison, Casson, and Weyl[11] on helium particles that attained charge equilibrium in metal foils and from the experiments of Snitzer[12] on helium particles passing through gases. Allison and Warshaw[13] have compared the results of these experiments and have shown that the ratio He^{++}/He^{+} in ions which have been equilibrated by scattering from a metal surface is considerably greater than in those in which equilibrium has been produced by passage through air. These experiments cover an energy range 175 kev–400 kev for the incident helium ion, and they appear to indicate that the effective charge, and consequently the stopping power of alpha particles, is higher in metal foils than in air.

A quantitative study of the density effect is difficult, since there is no exact method for calculating the effective charge of ions moving in various media. Some estimates have been made by Bohr,[14–16] Lamb,[17] Knipp and Teller,[18] and Brunings, Knipp, and Teller,[19] who assumed that the effective charge of an ion is a unique function of its velocity. They considered a relationship between the orbital velocity of the most easily removed electron and the velocity of the ion and assumed that this relationship is independent of the surrounding medium. They have neglected, therefore, the effect of the medium upon the moving ion, and such an effect is the subject of our discussion.

We take the specific case of an incident particle having atomic number $Z_1=2$ (helium), velocity $v=2v_0(v_0=e^2/\hbar)$, and surrounded by a medium having atomic number $Z_2=18$ (argon). Because of the density effect, the effective charge of the particle is higher in liquid argon than in argon gas. The effective charge is expressed in terms of cross sections (σ_{if}) for electron capture and loss; i and f represent the charge of the moving particle before the event and after the event, respectively. It will be shown that for electron loss, i.e., when $i<f$,

$$\sigma_{if}^{(d)} \geqslant \sigma_{if}^{(g)}; \tag{1}$$

and conversely, for electron capture, i.e., when $i>f$,

$$\sigma_{if}^{(d)} \leqslant \sigma_{if}^{(g)}. \tag{2}$$

In the above inequalities, $\sigma_{if}^{(d)}$ and $\sigma_{if}^{(g)}$ refer to condensed media and to gases, respectively. We shall use the superscripts (d) and (g) to designate quantities that relate to condensed media and gases, respectively.

[11] Allison, Casson, and Weyl (to be published). Some of the results of these experiments are briefly stated in reference 13.
[12] E. Snitzer, Phys. Rev. **89**, 1237 (1953).
[13] S. K. Allison and S. D. Warshaw, Revs. Modern Phys. **25**, 779 (1953).
[14] N. Bohr, Rev. **58**, 654 (1940).
[15] N. Bohr, Phys. Rev. **59**, 270 (1941).
[16] N. Bohr, Kgl. Danske Videnskab. Selskab, mat.-fys. Medd. **18**, 8 (1948).
[17] W. E. Lamb, Phys. Rev. **58**, 696 (1940).
[18] J. K. Knipp and E. Teller, Phys. Rev. **59**, 659 (1941).
[19] Bruning, Knipp, and Teller, Phys. Rev. **60**, 657 (1941).

B. Autoionization

(1) *Formulation of the Problem*

In an investigation of the interaction of charged particles with matter, attention is usually centered on the effect of the incident particle on the surrounding medium. Here we take the opposite point of view and consider the reaction of the perturbed medium on the particle. As shown by Bohr,[16] the perturbed medium is polarized and the polarization is localized in the wake of the incident particle. Since the polarization is not uniformly distributed, we obtain an electric field which is directed against the motion of the particle. The magnitude of this field at the position of the particle is

$$F=(1/Ze)(dW/dz), \tag{3}$$

where dW/dz is the energy loss per unit of length of the particle track (aligned along z axis), and Ze is the charge of the incident particle.

The value dW/dz depends on the number of atoms per unit volume in the stopping medium and if the medium is condensed the values dW/dz and F are relatively large. If, however, the medium is a gas, both dW/dz and F are relatively small.

The field F acts as a brake on the incident particle, and if the particle is an ion, its structure is distorted by polarization along the direction of motion. In a gas, F is small, and the resulting polarization is weak, having negligible effect on the structure of the moving ion. However, in the case of a solid or liquid, we have a strong polarizing field which may cause emission of electrons carried by the ion (autoionization). This process would increase the effective charge on the ion, and the higher effective charge would account, at least in part, for the higher energy loss due to ionization and excitation of the medium.

Autoionization under the effect of a strong electric field has been discussed by Oppenheimer[20] and more in detail by Lanczos[21] and by Bethe[22] in connection with the Stark effect in hydrogen. We shall consider this effect as applied to a nucleus of charge number Z_1 carrying a single bound electron.

The wave function of an electron in the Coulomb field due to the nucleus and in the potential field eFz due to the surrounding medium can be expressed as follows:

$$\frac{\chi_1(\xi)\chi_2(\eta)}{\xi^{\frac{1}{2}}\eta^{\frac{1}{2}}}e^{im\varphi}, \tag{4}$$

where ξ, η, φ are parabolic coordinates, m is the magnetic quantum number, and $\chi_1(\xi)$, $\chi_2(\eta)$ satisfy the

[20] J. R. Oppenheimer, Phys. Rev. **31**, 66 (1928).
[21] C. Lanczos, Z. Physik **62**, 518 (1930); **65**, 431 (1930); **68**, 204 (1931).
[22] H. A. Bethe, *Handbuch der Physik* (Springer, Berlin. 1933), Vol. 24, No. 1, p. 410.

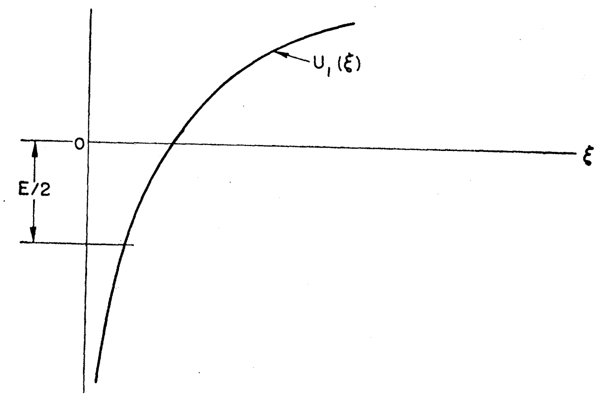

FIG. 1. Plot of $U_1(\xi)$.

following equations:

$$\frac{d^2\chi_1}{d\xi^2}+\left[\frac{E}{2}+\frac{(2n_1+m+1)Z_1}{2n\xi}-\frac{m^2-1}{4\xi^2}-\frac{F\xi}{4}\right]=0, \quad (5)$$

$$\frac{d^2\chi_2}{d\eta^2}+\left[\frac{E}{2}+\frac{(2n_2+m+1)Z_1}{2n\eta}-\frac{m^2-1}{4\eta^2}+\frac{F\eta}{4}\right]=0. \quad (6)$$

Here n is the "effective" principal quantum number, and n_1, n_2 are the "effective" parabolic quantum numbers; the electron energy E is expressed in atomic units of $me^4/\hbar^2$, and the field F is expressed in atomic units of $m^2e^5/\hbar^4$.

Each of the Eqs. (5) and (6) represents formally the one-dimensional Schrödinger equation of a particle having energy $E/2$ moving in potential fields $U_1(\xi)$, $U_2(\eta)$, where

$$U_1(\xi)=-\frac{(2n_1+m+1)Z_1}{2n\xi}+\frac{m^2-1}{4\xi^2}+\frac{F\xi}{4}, \quad (7)$$

$$U_2(\eta)=-\frac{(2n_2+m+1)Z_1}{2n\eta}+\frac{m^2-1}{4\eta^2}-\frac{F\eta}{4}. \quad (8)$$

Following Lanczos, we neglect the terms $(m^2-1)/4\xi^2$ and $(m^2-1)/4\eta^2$, and we take $m=0$. Then,

$$U_1(\xi)=-\frac{(2n_1+1)Z_1}{2n\xi}+\frac{F\xi}{4}, \quad (9)$$

$$U_2(\eta)=-\frac{(2n_2+1)Z_1}{2n\eta}-\frac{F\eta}{4}. \quad (10)$$

(2) *Delayed Ionization*

The potentials $U_1(\xi)$ and $U_2(\eta)$ are of the form illustrated in Figs. 1 and 2. The form of $U_1(\xi)$ gives discrete energy levels. The form of $U_2(\eta)$ exhibits a potential barrier separating two regions within which the electron motion is classically possible for any negative energy E: region I in the immediate neighborhood of the nucleus and region II at sufficiently large distances from the nucleus. Consequently, a bound electron has a finite probability of penetrating through the barrier and escaping from the neighborhood of the nucleus into the distant region. The probability[21] of such an escape per unit of time is

$$S=e^{-2I_2}/4I_1, \text{ atomic units of } (me^4/\hbar^3), \quad (11)$$

where

$$\alpha=F/Z_1^3, \quad (12)$$

$$I_1=\frac{\cos(\varphi_2/2)}{\alpha n}[K(k_2)-E(k_2)], \quad (13)$$

$$I_2=\frac{\sin\varphi_2\sin(\varphi_2/2)}{6\alpha n^3}\left[\frac{E'(k_2)}{\sin^2(\varphi_2/2)}-2K'(k_2)\right], \quad (14)$$

$$\sin^2\varphi_2=16n^3(n_2+\tfrac{1}{2})\alpha, \quad (15)$$

$$k_2=\tan(\varphi_2/2). \quad (16)$$

In the above formulas, K, K', E, and E' are complete elliptic integrals of the parameter k_2.[23]

The "effective" parabolic quantum numbers that appear above vary with F and are not integers. For $F=0$, they become integers and identical with the parabolic quantum numbers; i.e., $n_1=\nu_1$, $n_2=\nu_2$. The principal quantum number $\nu=\nu_1+\nu_2+1$.

For a given applied field F, the effective quantum numbers n, n_1, and n_2 may be determined from (15) and (16) given above and (17), (18), (19), (20), and (21) which follow[22]:

$$n=n_1+n_2+1, \quad (17)$$

$$\frac{n_1+\frac{1}{2}}{\nu_1+\frac{1}{2}}=\frac{3\pi}{8}\frac{\sin^2\varphi_1}{(\cos\varphi_1)^{\frac{1}{2}}[(1+\cos\varphi_1)K(k_1)-2\cos\varphi_1E(k_1)]}\equiv F_1(\varphi_1), \quad (18)$$

$$\frac{n_2+\frac{1}{2}}{\nu_2+\frac{1}{2}}=\frac{3\pi}{8}\frac{\sin^2\varphi_2}{2\cos(\varphi_2/2)[E(k_2)-\cos\varphi_2K(k_2)]}\equiv F_2(\varphi_2), \quad (19)$$

$$\tan^2\varphi_1=16n^3(n_1+\tfrac{1}{2}), \quad (20)$$

$$k_1=\sin(\varphi_1/2). \quad (21)$$

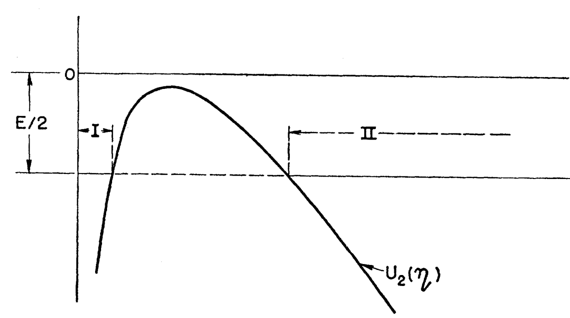

FIG. 2. Plot of $U_2(\eta)$.

[23] For a table of functions K, K', E, and E', see for instance L. M. Milne-Thomson, *Jacobian Elliptic Function Tables* (Dover Publications, Inc., New York, 1950), p. 106. In these tables, the parameter $m=k_2^2$.

We shall now determine the effective parabolic quantum numbers corresponding to $\nu=1$.

We put $\nu_1=0$ and $\nu_2=0$ in (18) and (19), respectively. Then, taking into account (18), (19), (15), and (20), we obtain

$$\sin^2\varphi_2/F_2(\varphi_2)=\tan^2\varphi_1/F_1(\varphi_1). \tag{22}$$

This yields a relation between φ_1 and φ_2 which is shown graphically in Fig. 3. Taking into account (18), (19), and (17), we obtain for $\nu_1=\nu_2=0$:

$$n=[F_1(\varphi_1)+F_2(\varphi_2)]/2. \tag{23}$$

By means of the graph of Fig. 3, we replace φ_1 by φ_2 in (23). This gives n as a function of φ_2, which is represented in Fig. 4 by curve A.

By means of (15) and (19), we represent n as a function of φ_2 and obtain

$$n^3=\sin^2\varphi_2/8\alpha F_2(\varphi_2). \tag{24}$$

In accordance with (24), n has been plotted as a function of φ_2 for various values of α. (See curves B). The intersection of each of the curves B with the curve A gives n and φ_2 corresponding to a given α. By means of (15), (20), and Fig. 3, the values n and φ_2 determine the corresponding values of n_1 and n_2. Figure 4 shows n_1 and n_2 as functions of α for $\nu_1=\nu_2=0$.

(3) *Instantaneous Ionization*

The width of the barrier separating the region I from the region II decreases with increase in F, and for sufficiently large values of F, the barrier disappears entirely, causing instantaneous ionization.

The quantity α expresses the effectiveness of the medium in producing ionization, and we shall determine for what values of α the potential barrier disappears, i.e., the instantaneous ionization takes place. We shall assume that the bound electron was initially in one of the following orbits: (1) $\nu_1=0$, $\nu_2=0$; (2) $\nu_1=1$, $\nu_2=0$; and (3) $\nu_1=0$, $\nu_2=1$.

The potential barrier disappears for $\varphi_2=90°$. Inserting this value in Eqs. (15) (16), (17), and (19), we obtain

$$n=[6.664(2\nu_2+1)\alpha]^{-\frac{1}{3}}, \tag{25}$$

$$n_1=[6.664(2\nu_2+1)\alpha]^{-\frac{1}{3}}-0.833\nu_2-1.0835. \tag{26}$$

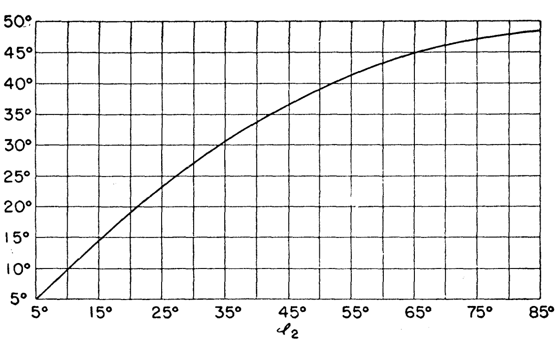

FIG. 3. Plot of φ_2 as a function of φ_1.

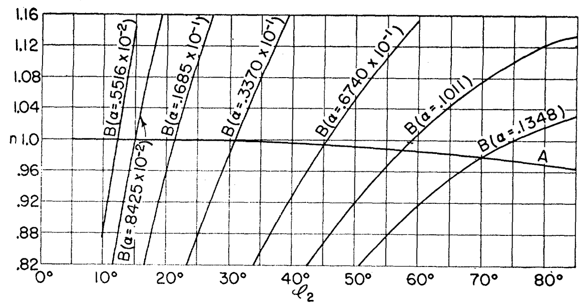

FIG. 4. Plot of n as a function of φ_2.

We have thus determined n and n_1 corresponding to a given α and ν_2. By means of (20) we determine φ_1 from n, n_1, and by means of (21) we determine k_1 from φ_1. The values φ_1 and k_1 are thus functions of α and ν_2. Substituting these values in (18), we obtain a relationship between ν_1, ν_2, and α. If this relationship is satisfied, the potential barrier disappears and instantaneous ionization takes place. The results can be stated as follows:

For $\nu_1=0$, $\nu_2=0$, the instantaneous ionization takes place if

$$\alpha>0.1666 \text{ atomic unit.} \tag{27}$$

For $\nu_1=1$, $\nu_2=0$, the instantaneous ionization takes place if

$$\alpha>0.0138 \text{ atomic unit,} \tag{28}$$

and for $\nu_1=0$, $\nu_2=1$, we have instantaneous ionization if

$$\alpha>0.0088 \text{ atomic unit.} \tag{29}$$

C. Numerical Example

(1) *Magnitude of the Applied Field*

We shall now apply our results to a helium particle moving with the velocity $v=2v_0$ in argon which may be either in the gaseous or the liquid state. The incident particle has three possible charge states, and we designate by ϕ_0, ϕ_1, ϕ_2 the probability of the particle's being neutral, singly ionized, and doubly ionized, respectively.

A doubly ionized helium atom (He^{++}) has no bound electrons; thus, even though the surrounding medium exerts on it a field F, its structure does not change whatever the magnitude of the field may be. Consequently, the stopping power of He^{++} is substantially independent of the state of condensation of the medium.

The energy loss of a neutral helium atom (He^0) is due entirely to close collisions with the surrounding atoms. Therefore, we shall neglect the effect of the medium and assume that $F=0$, both for condensed media and gases. Thus, the stopping power of He^0 is substantially independent of the state of condensation of the medium.

We shall consider now the reaction of the medium on a singly ionized helium atom (He^+). In accordance with (3), the value F is determined by the stopping

power dW/dz of He^+ in argon. There seems to be no direct measurement of this value. However, we can derive it from the measurements made by Reynolds, Dunbar, Wenzel, and Whaling[24] on incident hydrogen atoms in argon. These measurements gave the value 32×10^{-15} ev cm^2 for the cross section for a hydrogen atom having $v=2v_0$. It has been shown by Hall[25] that a hydrogen atom at $v=2v_0$ is completely stripped of its orbital electron. Therefore, the above cross section applies to a proton in argon gas, as well as to a proton in liquid argon, and it can be used to determine the corresponding values of dW/dz for both media. We obtain for an incident proton in liquid and gas, respectively,

$$\begin{aligned}(dW/dz)^{(d)}&=6.92\times10^8 \text{ ev/cm},\\ (dW/dz)^{(g)}&=8.75\times10^5 \text{ ev/cm}.\end{aligned} \tag{30}$$

The values (30) should closely approximate $(dW/dz)^{(d)}$ and $(dW/dz)^{(g)}$ for He^+ ions having the same velocity as protons. Therefore, substituting these values in (3), the field exerted by the surrounding liquid argon on an incident He^+ ion is

$$F^{(d)}=0.1348 \text{ atomic unit,} \tag{31a}$$

and the corresponding field exerted by argon gas is

$$F^{(g)}=1.7\times10^{-4} \text{ atomic unit.} \tag{31b}$$

(2) *Autoionization*

From (12) and (31a,b), we obtain for an incident He^+ ion

$$\alpha^{(d)}=0.0168 \text{ atomic unit,} \tag{32a}$$

$$\alpha^{(g)}=0.21\times10^{-4} \text{ atomic unit.} \tag{32b}$$

Comparison of the values (32a,b) with inequalities (27), (28), (29) leads to the following conclusions: (a) He^+ cannot be brought into an excited state in liquid argon since such a state would lead to an instantaneous ionization. (b) The instantaneous ionization does not occur in argon gas or liquid if He^+ is in the ground state, and it does not occur in argon gas if He^+ is in its first excited state.

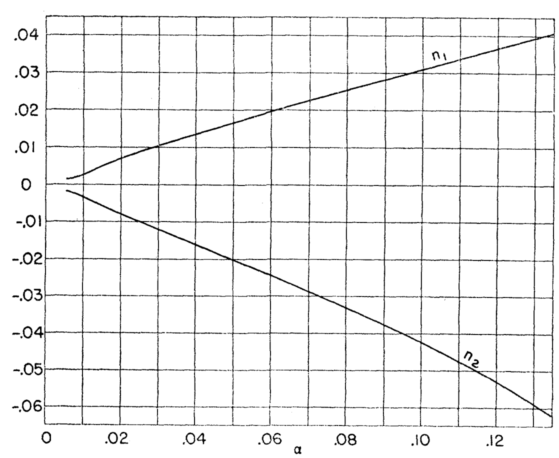

FIG. 5. Plot of n_1 and n_2 as a function of α.

We shall now determine the probability of autoionization per unit of time for He^+ in the ground state, both in argon gas and liquid.

It is apparent from Fig. 5 that for $\nu_1=\nu_2=0$ the effective quantum numbers do not vary significantly with F. For the value (32a), we have $n_1=0.0058$, $n_2=-0.0066$, and for the value (32b), we may assume both n_1 and n_2 equal to zero. These values when substituted in (11) give the following:

$$S^{(d)}\sim0.41\times10^3 \text{ sec}^{-1}, \tag{33a}$$

$$S^{(g)}\ll0.01 \text{ sec}^{-1}. \tag{33b}$$

Designating by $\Sigma_a^{(d)}$ and $\Sigma_a^{(g)}$ the probability of autoionization per cm of the particle track in liquid and gas, respectively, we obtain

$$\Sigma_a^{(d)}=S^{(d)}/v=9.56\times10^{-7} \text{ cm}^{-1}, \tag{34a}$$

$$\Sigma_a^{(g)}=S^{(g)}/v=0.25\times10^{-11} \text{ cm}^{-1}. \tag{34b}$$

In order to determine the effectiveness of autoionization, we shall compare the values (34a,b) with the probability of ionization $\Sigma_c^{(d)}$ and $\Sigma_c^{(g)}$ per cm of the particle track due to the collision of the particle with the surrounding atoms. Taking into account that the cross section for electron loss is of the order of magnitude of πa_0^2 (where $a_0=\hbar^2/me^2$), we obtain the following for the liquids and the gas, respectively:

$$\Sigma_c^{(d)}\sim N^{(d)}\pi a_0^2=1.86\times10^6 \text{ cm}^{-1}, \tag{35a}$$

$$\Sigma_c^{(g)}\sim N^{(g)}\pi a_0^2=2.36\times10^3 \text{ cm}^{-1}, \tag{35b}$$

where $N^{(d)}$ and $N^{(g)}$ designate the number of atoms per cm^3 in liquid argon and argon gas, respectively.

The values (34*a*,*b*) are very small when compared with (35*a*,*b*) and, therefore, the autoionization of He^+ in the ground state may be neglected.

We may summarize our results as follows: If an incident helium atom (whether ionized or not) passing through argon in liquid or gaseous state undergoes a collision as a result of which it emerges as a He^+ ion having its electron in the orbit $\nu=1$, no autoionization takes place. If, however, the incident atom emerges as a He^+ ion in an excited state, it cannot exist in such state in liquid argon because it becomes instantly doubly ionized. However, this situation does not occur if the incident atom emerges as an excited He^+ ion in argon gas. In gas, the autoionization can occur only if the excited level is very close to the continuum, and this case can be, for our purposes, ignored.

[24] Reynolds, Dunbar, Wenzel, and Whaling, Phys. Rev. **92**, 742 (1953).
[25] T. Hall, Phys. Rev. **79**, 504 (1950).

III. ELECTRON LOSS AND CAPTURE

A. General

So far the results obtained are based on a relatively exact theory and an effect of the medium on the autoionization of an incident particle has been established. The particle while passing through the medium undergoes continual collisions during which it captures and loses orbital electrons. We shall now investigate the effect of the medium on the cross section for the electron loss and capture. In that connection, we shall take into account only those events that result in a loss or capture of a single electron and neglect the cross section involving double electron transfers.

The problem is difficult since there is no exact method for computing the cross section for helium ions moving in argon. Some investigations[26,27] dealing with the problem were concerned with the passage of hydrogen ions through hydrogen gas and, therefore, are of little interest for our purposes.

Therefore, we shall rely on the methods established by Bohr[16] which, in order of magnitude, agree with the experiments. Our purpose is not to calculate exactly the cross section for electron loss and capture, but rather to give a general discussion of various factors that influence the magnitude of these cross sections in gases and condensed media. By applying uniformly the same general considerations to all media and by taking into account the characteristic effects caused by the condensed media, we can make some estimates as to the extent to which the cross sections in condensed media depart from the corresponding cross sections in gases. In view of the cursory nature of Bohr's theory, our results should be interpreted as merely indicating orders of magnitude.

B. Electron Capture

We consider a particle of charge Z_1e penetrating with velocity v into the electron cloud surrounding a nucleus of charge Z_2e and subsequently capturing an electron into its orbit ν. By means of statistical arguments, Bohr expressed the cross section for capture as follows:

$$\sigma_c=\sigma f N, \tag{36}$$

where $N\sim Z_2^{\frac{1}{3}}(v/v_0)$ is the number of electrons that participate in the capturing process (i.e., those electrons that have orbital velocities comparable with the particle velocity). $\sigma\sim 4\pi a_0^2 Z_1^2(v_0/v)^4$ is the cross section corresponding to the energy transfer of the order of $mv^2/2$ to an electron, and f is the fraction of the velocity space corresponding to those electrons that have relative velocities, with respect to the incident particle, comparable to the velocity of the electron after capture.

If the particle captures an electron into an orbit ν, then the electron velocity is Z_1v/ν, and, therefore,

$$f\sim(Z_1v_0^3/\nu v)^3. \tag{37}$$

Substituting the above values of N, σ, and f in (36), we obtain:

$$\sigma_c^{(\nu)}\sim\frac{4\pi a_0^2 Z_1^5 Z_2^{\frac{1}{3}}}{\nu^3}\left(\frac{v_0}{v}\right)^6. \tag{38}$$

Bohr's formula has been derived under the assumption that $\nu=1$, i.e.,

$$\sigma_c^{(1)}\sim 4\pi a_0^2 Z_1^5 Z_2^{\frac{1}{3}}(v_0/v)^6. \tag{39}$$

The total capture cross section can be expressed as

$$\sigma_c=\sum_1^\infty \sigma_c^{(\nu)}\sim 4.8\pi a_0^2 Z_1^5. \tag{40}$$

C. Electron Loss and Excitation

Bohr considered this problem in the system of coordinates moving with the particle and neglected the binding energy of the orbital electron attached to the incident particle. He dealt, therefore, with a stationary electron perturbed by a screened Coulomb field moving with a velocity v. The character of the perturbation depends upon the screening parameter defined by Bohr as

$$\zeta=b/a, \tag{41}$$

where b is the collision diameter, i.e.,

$$b=2Z_2e^2/mv^2, \tag{42}$$

and a is the radius of the deflecting atom, i.e.,

$$a=a_0/Z_2^{\frac{1}{3}}. \tag{43}$$

Substituting (42) and (43) in (41), we obtain

$$\zeta=2Z_2^{4/3}(v_0/v)^2. \tag{44}$$

Bohr took as an example an alpha particle moving in air with a velocity $v=6v_0$, for which (44) gives $\zeta\sim 1$. He dealt, therefore, with the case of "intermediate screening" for which the deflecting field may be approximated by a potential of the type k/r^2 (k is a suitable constant). By assuming such a potential, Bohr obtained for the cross section for electron loss

$$\sigma_l\sim\pi a_0^2 Z_2^{\frac{2}{3}} Z_1^{-1}(v_0/v). \tag{45}$$

In our case, however, the surrounding medium is argon, and the velocity of the particle is $v=2v_0$. Consequently, $\zeta\sim 20$, and the problem arises whether we have "intermediate screening" or "excessive screening."

In the case of "excessive screening," the scattering of the electron by the screened Coulomb field is substantially isotropic, and the cross section for ionization is

$$\sigma_l\sim\sigma_0(T_m-I)/T_m, \tag{46}$$

where $\sigma_0=\pi a_0^2/Z_2^{\frac{2}{3}}$ is the geometrical cross section, $T_m=2mv^2$ is the maximum energy transferred, and

[26] D. R. Bates and A. Dalgarno, Proc. Phys. Soc. (London) A55, 919 (1952).
[27] J. D. Jackson and H. Schiff, Phys. Rev. 89, 359 (1953).

$I = mZ_1^2 v_0^2/2$ is the ionization energy. Consequently,

$$\sigma_I \sim \frac{\pi a_0^2}{Z_2^{\frac{2}{3}}} \left[1 - \frac{Z_1^2}{4}\left(\frac{v_0}{v}\right)^2\right]. \tag{47}$$

In order to determine whether the case of $\zeta \sim 20$ is better approximated by "intermediate screening" or "excessive screening," we have applied both (45) and (47) to two cases for which experimental data are available, i.e., the measurement of the charge distribution of incident hydrogen atoms in air[28] for $v = 1.26v_0$ and in neon[29] for $v = 1.46v_0$. The expression (45) gives values that differ from the measured values by less than 15 percent, while expression (47) gives values about 40 times too small. We shall, therefore, use (45) and base our considerations on "intermediate screening."

According to Bohr, $\sigma_I \sim \pi i^2$, where i is the impact parameter corresponding to the angular deflection $\vartheta = \vartheta_I$, for which the energy transfer is equal to the ionization energy I. Consequently, the expression (45) represents the cross section for processes for which the energy transfer to the electron $T > I$.

We shall determine the cross section σ_{exc} for processes for which the energy transfer to the electron is $T > T_{\text{exc}'}$, where T_{exc} is the energy required to raise an electron from the ground state to an excited state characterized by the quantum number ν:

$$T_{\text{exc}} = \tfrac{1}{2} m Z_1^2 v_0^2 \left(1 - \frac{1}{\nu^2}\right). \tag{48}$$

Consequently, the angular deflection is $\vartheta > \vartheta_{\text{exc}}$, where

$$\vartheta_{\text{exc}} = \frac{Z_1 v_0}{v}\left(1 - \frac{1}{\nu^2}\right)^{\frac{1}{2}}, \tag{49}$$

and

$$\sigma_{\text{exc}} \sim \frac{\pi a_0^2 Z_2^{\frac{2}{3}}}{Z_1 (1 - \nu^{-2})^{\frac{1}{2}}}. \tag{50}$$

D. Charge Exchange in Argon Gas

Previously, we have found that in argon gas F is negligibly small. Therefore, the incident particle is undisturbed by the surrounding medium before and after the collision, and no autoionization takes place. We shall calculate the cross section for electron capture and loss for an incident helium particle having $v = 2v_0$.

Any collision resulting from an electron capture into an orbit $\nu \geqslant 2$ is followed by the emission of a photon as a result of which the electron falls into the ground state. Therefore, the capture cross section is determined by (40), and we obtain

$$\sigma_{10}^{(g)} \sim 0.19\pi a_0^2, \tag{51}$$

$$\sigma_{21}^{(g)} \sim 6.28\pi a_0^2. \tag{52}$$

[28] H. Kanner, Phys. Rev. **84**, 1211 (1951).
[29] P. M. Stier (private communication).

For the electron loss cross section, we obtain by means of (45)

$$\sigma_{01}^{(g)} \sim 3.43\pi a_0^2, \tag{53}$$

$$\sigma_{12}^{(g)} \sim 1.71\pi a_0^2. \tag{54}$$

E. Charge Exchange in Liquid Argon

The mechanism of collisions of helium particles with the surrounding atoms depends on the applied field F. Bohr's theory of electron capture and loss is applicable when $F = 0$, but in view of the cursory nature of Bohr's arguments, we shall apply them also when $F \neq 0$. However, account will be taken of the effect of the field F on the incident particle prior to and after the collision. Collisions leading to the following transitions will be considered: $He^+ \to He^0$; $He^0 \to He^+$; $He^{++} \to He^+$; and $He^+ \to He^{++}$.

(1) *Transition* $He^+ \to He^0$

Prior to this transition, the incident particle was a singly ionized helium atom having its bound electron in the orbit $\nu = 1$. It has been shown previously that such a particle is "stable," i.e., its autoionization is negligible both in liquids and gases. As a result of a collision with an argon atom, the incident particle emerged as a neutral helium atom, and as such, it remained stable since the reaction of the medium to a neutral particle may be neglected ($F \sim 0$). Therefore, the state of condensation of the medium does not affect the charge of the particle before or after the transition. However, the collision process is different in gases and in liquids because of a polarizing field in liquids. On the other hand, the general assumptions underlying Bohr's theory are not substantially modified by the presence of a polarizing force. Therefore, we obtain

$$\sigma_{10}^{(d)} = \sigma_{10}^{(g)}. \tag{55}$$

(2) *Transition* $He^0 \to He^+$

We assume that the process is the same in gases and liquids since, as indicated above, He^0 and He^+ are stable in both media. Thus,

$$\sigma_{01}^{(d)} = \sigma_{01}^{(g)}. \tag{56}$$

(3) *Transition* $He^{++} \to He^+$

In gases an incident He^{++} particle may capture an electron either into its lowest orbit or into an excited state. In the latter case, the electron capture is followed by the emission of a photon and thus does not change the character of the transition. In liquid, however, the electron can be captured only into its lowest orbit. A capture into an orbit $\nu \geqslant 2$ is not possible since an excited He^+ ion becomes instantly ionized. We shall apply, therefore, the formula (50) and obtain

$$\sigma_{21}^{(d)} \sim 5.24\pi a_0^2. \tag{57}$$

(4) *Transition* $He^+ \rightarrow He^{++}$

In gases, this transition occurs only if He^+ receives energy at least equal to its ionization energy. In liquids, however, a smaller energy transfer is required to produce this transition since any collision that would excite He^+ will be followed by an immediate ionization.

Therefore, we apply the formula (50) by taking $\nu=2$ and obtain

$$\sigma_{12}{}^{(d)} \sim 1.98\pi a_0{}^2. \qquad (58)$$

IV. EFFECTIVE CHARGE

A. Charge Equilibrium

If an incident helium particle undergoes many charge exchange collisions without substantially altering its velocity, a state of equilibrium is reached in which the charge composition of the particle remains constant as long as its velocity does not substantially change.

We shall determine now whether there is a state of equilibrium for a helium particle moving through argon with velocity $v=2v_0$. In that connection, we shall consider a charge exchange cycle, the cross section for which can be either $\sigma_i{}^{(1)}$ or $\sigma_i{}^{(2)}$, where

$$\sigma_i{}^{(1)} = \sigma_{if} + \sigma_{fi}, \qquad (59)$$

$$\sigma_i{}^{(2)} = \sigma_{ik} + \sigma_{kl} + \sigma_{lk} + \sigma_{ki}. \qquad (60)$$

In the first case, the particle undergoes transition of the type $i \rightarrow f \rightarrow i$, involving one intermediate state, and in the second case, the transition is of the type $i \rightarrow k \rightarrow l \rightarrow k \rightarrow i$, involving two intermediate states. We obtain from (51), (52), (53), and (54)

$$\sigma_0{}^{(1)} = \sigma_1{}^{(1)} \sim 3.6\pi a_0{}^2; \quad \sigma_2{}^{(1)} \sim 8\pi a_0{}^2,$$

and $\sigma_0{}^{(2)} = \sigma_2{}^{(2)} \sim 11.6\pi a_0{}^2$. Consequently, the mean free path for the charge exchange cycle satisfies the following inequality:

$$\lambda \leqslant (N\sigma_0{}^{(1)})^{-1} \text{ cm}, \qquad (61)$$

where N is the number of argon atoms per cm^3. The atomic stopping power of an incident helium particle having $v=2v_0$ measured by Weyl[30] is 79×10^{-15} ev cm^2. Consequently, the energy loss per cm of path due to ionization, excitation, and charge exchange collision is

$$dW/dz \sim N\times 79\times 10^{-15} \text{ ev/cm}, \qquad (62)$$

and the maximum value of the energy loss over the distance λ is

$$\Delta W_{\max} = 79/\sigma_0{}^{(1)} \sim 250 \text{ ev}. \qquad (63)$$

It is apparent from (63) that it should take many mean free paths to change substantially the velocity of the particle, and, therefore, we are justified in considering the particle in the state of charge equilibrium.

[30] P. K. Weyl, Phys. Rev. **91**, 289 (1953).

B. Charge Composition in Liquid Argon and Argon Gas

Under these conditions, the distribution of charges of the particle is described by

$$\phi_0 : \phi_1 : \phi_2 = \sigma_{10}\sigma_{21} : \sigma_{01}\sigma_{21} : \sigma_{01}\sigma_{12}, \qquad (64)$$

where ϕ_0, ϕ_1, and ϕ_2 represent the probability of the particle's being neutral, singly ionized, and doubly ionized, respectively.[12]

We shall now proceed to determine the distribution of charges of the particle in argon gas and in liquid argon. Substituting (51), (52), (53), and (54) in (64), we obtain for the incident particle having $v=2v_0$ in the gas

$$\phi_0{}^{(g)} : \phi_1{}^{(g)} : \phi_2{}^{(g)} = 0.05 : 0.75 : 0.20, \qquad (65)$$

and, consequently, the effective charge of the alpha particle is

$$\langle \epsilon^2 \rangle_{\text{Av}}{}^{(g)} = \phi_1{}^{(g)} e^2 + 4\phi_2{}^{(g)} e^2 = 1.55 e^2. \qquad (66)$$

Similarly, substituting (55), (56), (57), and (58) in (64), we obtain for the liquid

$$\phi_0{}^{(d)} : \phi_1{}^{(d)} : \phi_2{}^{(d)} = 0.04 : 0.70 : 0.26, \qquad (67)$$

and the effective charge is

$$\langle \epsilon^2 \rangle_{\text{Av}}{}^{(d)} = 1.74 e^2. \qquad (68)$$

It may be of interest to note that the measured values for the alpha particle in argon gas are as follows[12]:

$$\phi_0{}^{(g)} : \phi_1{}^{(g)} : \phi_2{}^{(g)} = 0.07 : 0.71 : 0.22, \qquad (69)$$

and

$$\langle \epsilon^2 \rangle_{\text{Av}}{}^{(g)} = 1.59 e^2. \qquad (70)$$

By comparison of (66) and (68), it is apparent that the average square charge of the alpha particle in liquid exceeds the one in gas by about 13 percent.

V. CONCLUSION

Our results may be summarized as follows:

1. An ion moving in a condensed medium is subjected to a polarizing force that causes autoionization. Therefore, the effect of the medium should account for a higher effective charge of the ion. This effect, insofar as it relates to fission fragments, is supported by experiments of Lassen,[10] who reported a large difference between the charges of fission fragments in gaseous and solid stopping media. Furthermore, experiments of Allison, Casson, and Weyl[11] and of Snitzer[12] support the existence of this effect for alpha particles in the energy range 175 kev–400 kev.

2. An alpha particle having velocity $v=2v_0$ has a larger effective charge in liquid argon than in argon gas, since it has been established that any collision in the liquid leading to an excited He^+ is followed by instantaneous ionization, and such a collision, when occurring in gas, does not cause ionization.

3. An alpha particle having velocity $v=2v_0$ has an effective charge higher by 13 percent in liquid argon than in argon gas. To derive this value, we used an approximate theory of electron capture and loss, and, therefore, our result indicates merely an order of magnitude. It is not possible to evaluate this since the reliability of the experimental data is questioned. The fact that only five experiments have been reported and results are contradictory indicates the formidable difficulties in obtaining exact data. By assuming that the anomaly in the stopping power exists, it is questioned whether those measurements that reported the anomaly are sufficiently precise. According to Appleyard,[3] the average stopping power of an alpha particle in the energy range from zero to 4.5 Mev is 15 percent higher in water than the theoretical one for water vapor. Qualitatively, this agrees with out results. However, according to our interpretation, the anomaly exists only for alpha particles in the energy range below 2 Mev. For energies above this range, the alpha particle is completely stripped of its orbital electrons, and, therefore, its stopping power should be the same for liquid media and gases. However, Appleyard found that the stopping power at 4.5 Mev is higher in water than in water vapor, and these results cannot be explained on the basis of our assumptions.

TABLE I. Comparison of experimental data with calculated values.

	Calculated	Experimental
$(\phi_2/\phi_1)^{(g)}$	0.27 (argon gas)	0.31 (argon gas) Snitzer[a]
$(\phi_2/\phi_1)^{d}$	0.37 (liquid argon)	0.50 (gold) Allison, Casson, Weyl[b]
Density effect P	37%	61%

[a] See reference 12.
[b] See reference 11.

4. According to Michl,[1] the range of a Po alpha particle (5.298 Mev) is 20 percent smaller than that calculated for water vapor and according to Philipp,[2] the range of RaC alpha particles (7.680 Mev) is 16 percent smaller. If Michl's and Philipp's measurements are exact, then our approximate values of $\langle\epsilon^2\rangle_{\mathrm{Av}}$ in liquid argon and gas represent an underestimate. Also, this appears from the experimental data of Snitzer[12] and of Allison, Casson, and Weyl[11] shown in Table I. The density effect measured as P designates the increase in percent in the value ϕ_2/ϕ_1 due to the state of condensation of the medium. We obtain from (65) and (67) $P=37$ percent. However, the experiments give $P=61$ percent, which indicates a higher effective charge than the one calculated.

As stated above, the scarcity of data is partly caused by experimental difficulties. Possibly, as suggested by Platzman,[7] this energy region was considered of minor importance in experiments of nuclear physics. However, if such a situation existed in the past, it certainly has undergone a fundamental change in the last several years. The density effect in low-energy regions has become of definite practical interest in problems dealing with the interaction of charged particles and living tissue and in any processes involving charge exchange between the incident ions and the surrounding medium.

The computations have been carried out by Mary Todd of the ORNL Mathematics Panel.