Electronic States of Ions Recoiling into Vacuum and Gases¹

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¹Research sponsored by the U. S. Atomic Energy Commission under contract with the
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Abstract

Electronic states of ions recoiling into vacuum and gases, Sheldon Datz
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Physica Scripta (Sweden).

In the study of hyperfine interactions of nuclear states created by nuclear reaction or Coulomb excitation the recoiling ion undergoes a series of collisions in which electrons are captured and lost and in which electronic excitation and deexcitation take place. A knowledge of the electronic states of the ion on a time scale commensurate with the lifetime of the nuclear states (1-100 psec) is vital to the understanding of the field at the nucleus.

Atomic collision processes in dilute and dense media are discussed in terms of charge changing and excitation of both inner and outer shell electrons and the application to some perturbed angular correlation measurements are given.

1. Introduction

The subject of hyperfine interactions in highly stripped ions as studied by recoil into vacuum and gases was discussed in an excellent review by Professor Goldring at the Oak Ridge Heavy Ion Summer Study in 1972 [1]. In this lecture he posed the questions which we will attempt to answer in this talk. These answers or more accurately partial answers will also bear on the problems of recoiling multielectron systems. But first consider the general experiment as described by Goldring. He points out that heavy ion reactions make it possible to study hyperfine interactions of nuclei with electrons in the $1s$ state.

"These specific hyperfine interactions have some very attractive features: the fields generated by $1s$ electrons are very large, producing measurable effects on even exceedingly short-lived nuclear levels, and the fields are very well known and understood, in contrast to most other hyperfine fields which have to be calibrated experimentally and which can serve in effect only to compare nuclear magnetic moments of different states and not to measure them directly.

"In passing high-velocity reaction products through a foil (usually the target foil), it is quite straightforward, at least with low- Z atoms, to strip these atoms down to essentially any desired level. It is much harder to control the degree of excitation of the ions and, in particular, to ensure that a large fraction of them are in the ground state. In fact, previous measurements of hyperfine interactions in isolated ions which were carried out with heavy atoms indicated quite the opposite, namely, that those ions were produced in general in highly excited states which subsequently decayed in a long and complex cascade of electromagnetic transitions.

"In the case of s electrons, the motion induced by the hyperfine interaction can be described by a single Larmor frequency ω given by

$$\omega = \frac{\mu_N g H(0)}{h} (2l + 1),$$

where μ_N is the nuclear magneton, g is the gyromagnetic ratio of the nucleus, $H(0)$ is given for hydrogen-like states with quantum numbers n, l, j .

$$H(0) = \frac{Z^3}{n^3} \frac{1}{(l+1/2)(j+1)} K,$$

where $K = hcR_\infty a^2 / \mu_B = 0.126$ MG. In atoms with one electron outside of closed shells, the fields are very similar to the one-electron fields quoted above, provided the total number of electrons is small. Characteristic fields for

Insert Table I. some few-electron configurations are listed in Table I.

"It is evident that for any given atom the fields are ordered in a regular manner of magnitude as the energy of excitation increases.

Insert Fig. 1.

"A frequently used experimental arrangement is shown in Fig. 1, where the scattered particles or reaction products are detected in a ring counter in the forward direction. This system has an added advantage for the type of experiment considered here, in that it tends to produce high recoil velocities.

"For hyperfine interactions in s levels, the time-integrated attenuation coefficients are given by

$$G_k = 1 - \frac{k(k+1)}{(2l+1)^2} \frac{(\omega\tau)^2}{1 + (\omega\tau)^2},$$

or introducing the attenuation parameter η ,

$$G_k = 1 - k(k+1)\eta,$$

$$\text{where } \eta = \frac{1}{(2l+1)^2} \frac{(\omega\tau)^2}{1 + (\omega\tau)^2}.$$

Insert Fig. 2. The quantity $(\omega\tau)^2 / [1 + (\omega\tau)^2]$ is shown in Fig. 2 for the lowest configurations. When this quantity is zero, there is no perturbation, and the saturation of unity corresponds to hard-core perturbation. The hyperfine perturbation can also be computed for more complex configurations, but the calculations are considerably more involved.

$$\omega = \frac{\mu_N g H(0)}{h} (2l + 1),$$

where μ_N is the nuclear magneton, g is the gyromagnetic ratio of the nuclear level, and $H(0)$ is given for hydrogen-like states with quantum numbers n, l, j, b

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"It is evident that for any given atom the fields are ordered in a hierarchy of magnitude as the energy of excitation increases.

"A frequently used experimental arrangement is shown in Fig. 1, where scattered particles or reaction products are detected in a ring counter in the backward direction. This system has an added advantage for the type of experiment considered here, in that it tends to produce high recoil velocities.

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"If the atoms are distributed among a variety of configurations i with probabilities $p(i)$, then the overall attenuation coefficients will be given by

$$G_k = \sum_i p(i) G_k(i)$$

or

$$\eta = \sum_i p(i) \eta(i).$$

Evidently in any given case there is only a limited number of configurations that are "turned on."

"The basic atomic problem in such measurements is the knowledge or evaluation of the coefficients $p(i)$. In general there is very little a priori information on these parameters, and one can estimate them only in cases where there is reason to believe that most of the ions are in their ground states and where, therefore, the distribution of atomic levels is linked to the rather well-known charge distribution of the ions."

This excerpt from Professor Goldring's paper serves to outline the method and poses the principal problem. What are the states of the recoiling atoms on a time scale comensurate with the nuclear lifetimes in question, i.e., 1 - 100 psec? In the case of recoil vacuum excited states with lifetimes < 1 psec are unimportant in the perturbation and those greater than 100 psec last "forever." On the other hand for recoil into gases and solids the collisional cross sections for excitation and deexcitation become important and the steady state of the ion over the period may become important.

In this talk we shall discuss the atomic states of recoiling ions as they penetrate solids, as they emerge into vacuum, and as they make collisions in gases.

2. Charge Exchange and Excitation of Recoiling Ions

In work on recoil into vacuum and gases the excited nuclear state is generally prepared by nuclear reaction or nuclear Coulomb excitation. This is, on an atomic scale, an extremely violent collision, and if heavy ions are involved

will generally lead to multiple ionization and excitation of the colliding partners. However, this event is not "typical" on an atomic scale and, except for outer shell remnants of excitation, will be relaxed in a period of $\sim 10^{-14}$ sec.

The recoiling ion will undergo a series of collisions. In the first collision that it makes it will in all probability lose an outer electron, in the second collision it may lose a second one, and so on. The cross section for electron loss σ_L will, in general, decrease with increasing charge state q . In the meantime the probability of electron capture σ_C by the penetrating ion increases with increasing charge state and ultimately a steady state between capture and loss is attained where the most probable "equilibrium" charge is attained, and it is unaffected by additional collisions, i.e., after traversing a given target thickness ρ the fractional population ϕ_q of charge state q depends on relative cross sections for production and loss. Considering only single electron capture and loss

$$d\phi_q/d\rho = \phi_{q-1}\phi_L(q-1) + \phi_{q+1}\sigma_C(q+1) - \phi_q[\sigma_L(q) + \sigma_C(q)].$$

At equilibrium the derivative is zero, and if σ_C increases with q at about the same rate that σ_L decreases the most probable charge state $\bar{q}$ is that for which $\sigma_C \sim \sigma_L$.

For dealing with the charge changing events occurring with a great variety of ions in many materials we will use as a reference frame the simple theories developed by Bohr and Lindhard [2]. (This and similar theories are discussed in a recent review by Betz [3].) They treated the problem of electron capture and loss in a highly simplistic manner. However, their treatment gives a good physical representation of the problem. The fundamental units here are the Bohr radius and velocity: $a_0 = h^2/me^2$ and $v_0 = e^2/h$. An electron in an ion or atom of nuclear charge Z has an orbital velocity v_L defined in terms of the electron binding energy and in reduced units has a radius and velocity

$$a = a_0(v^2/n), \quad v_L = v_0(n/v)$$

where $(Z - n)$ is the number of electrons with orbital radius smaller than $\underline{a}$

(i.e., the screened nuclear charge). The quantity v is an "effective quantum number" based on a Thomas Fermi statistical model.

The process of electron loss is treated as simple ionization in which a bound electron is removed to the continuum. Starting with the cross section for transfer of energy greater than T by collision between a free electron at rest and a heavy particle with charge $z^* e$ and velocity v

$$\sigma = 2\pi a_0^2 z^{*2} \left(\frac{v_0}{v}\right)^2 \left(\frac{mv_0^2}{T} - \frac{mv_0^2}{T_{\max}}\right)$$

where the maximum transferred energy is $T_{\max} = 2mv^2$. Introducing for each ion electron $T = mv^2/2$ and summing over all ion electrons they obtained

$$\sigma_L = \pi a_0^2 z^{*2} z^{1/3} (v_0/v^*)^3$$

where v^* , the binding energy of the most loosely bound electron, is in the order of V and z^* is the nuclear charge for light gases or the core charge for heavy ones. In order to estimate the dependence of the loss cross section on ion charge q , v^* is approximated by $v_0 q Z^{-1/3}$ and

$$\sigma_L = \pi a_0^2 z^{2/3} z^{4/3} q^{-3} (v/v_0)^2.$$

The process of electron capture is much more delicate since in this case we are moving into a well-defined bound state and energy conservation restrictions on the motion become more stringent. Bohr and Lindhard considered capture in the following manner. During the approach of the highly charged ion, the electron will be exposed to a strong field of force, giving rise to an increasing polarization of the binding, which may subsequently lead to its rupture. At a distance R between the two systems given by

$$qe^2/R^2 = mv_e^2/a$$

the force from the ion and the atomic binding force are approximately equal. The possibility for electron release is determined not only by a comparison between the forces, but also the process must be completed in a time of the order a/v . Therefore, the ion may have travelled a distance comparable with

R before the electron is liberated from the atomic field. After the release from the atom, the electron will be captured if its total energy relative to the ion has a negative value. The condition for capture is that the process of electron release is effectively completed at a distance from the ion smaller than R' , determined by

$$qe^2/R' = 1/2 mv^2.$$

Assuming that the release takes place at the distance R, capture occurs with a cross section of πR^2 if $R < R'$, but for $R > R'$ there will be no capture. Thus on this assumption only strongly bound atomic electrons can contribute to capture. In a heavy atom, the contribution will arise mainly from a comparatively narrow region of orbital velocities around $v/2$. Summing over the electrons in the atom,

$$\sigma_c = \pi a_0^2 q^2 z^{1/3} (v_0/v)^3$$

for the total capture cross section for atoms in which a considerable part of the electrons have velocities comparable with v .

The result of this reasoning is that at equilibrium, the bound electron with orbital velocity equal to the ion velocity will be the most frequently captured and lost and that the average charge on a heavy ion is attained when the ion velocity matches the velocity of the most weakly bound remaining electron. Using the Thomas-Fermi atomic model this leads to the result

$$\bar{q}/z = v/(v_0 z^{2/3}).$$

This extremely simple approach has proven remarkably valid for ions emerging from dilute gas targets (O_2 and N_2) as shown in Fig. 3 for numerous energetic ions ranging from sulphur to uranium.

Using instead the extrapolated ionization potentials Lamb [4] calculated curve 1 in Fig. 3 while Brunnings, Knipp and Teller [5] using another approach to the Thomas-Fermi model arrived at curve 3. Betz [3] using the same criteria

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and not the kinetic energy. Here iodine charge states from O_2 and N_2 targets are plotted versus ion velocity.

2.1. Excited States

In the discussion above we have been considering only ground state ion configurations. In practice we must also consider the effect of excited states on both capture and loss processes, especially in condensed media. For example, the excitation cross section for a given (outer shell) electron on an ion is of the same order as the ionization cross section and the ionization cross section for the excited state is, in general, higher than that for the ground state. In a dilute gas radiative decay will occur between collisions but as the target becomes more dense the effective loss cross section will be increased by the presence of excited states.

Capture into excited states for light ions (e.g., protons and alpha particles) has been extensively studied both theoretically and experimentally. Oppenheimer showed that alpha particles capture electrons from hydrogen atoms mainly into s states with cross sections which, for sufficiently high velocity, are simply given by $\sigma_v = \sigma_1/v^3$ - ($v \gg v_0$) where v is the principal quantum number and σ_1 is the cross section for capture into $1s$. Hence in these cases only ~20% will be captured into higher v states. For higher z single electron ions we shall see later that this is not the case. For heavy ions at velocities $v \approx v_0$ it is anticipated that capture into excited states will predominate since these involve lower and more closely spaced bound states for the captured electron. Here again in a dilute gas target excitation will be relaxed but the presence of an excited electron in a subsequent collision will reduce the effective capture cross section for two reasons: first, for the same reason given in the case of excitation from the ground state, the ionization cross section for the excited electron is higher and; second, the presence of one excited electron at the time of capture of a second electron can lead to an auto-ionizing state of the ion in which the total excitation energy of the ion goes into removal of one of the electrons.

2.2. Experimental Measurements of Charge Changing Cross Sections

Considering the crudeness of the model discussed in the above the match to experimental results is qualitatively good. But there are differences which are observed which significantly affect the ion state and in many cases are not completely understood. As an example consider the cross sections obtained

Insert Fig. 5.

for electron capture by 13.9 MeV Br ions in H_2 , He and Ar shown in Fig. 5 [6]. As expected, single-capture probabilities $\sigma_c(q)$ increases with the charge state q , but in all cases characteristic anomalies were found. Compared with a smooth trend in $\sigma_c(q)$, the cross sections $\sigma_c(6)$ and to some extent $\sigma_c(8)$ are above the trend whereas $\sigma_c(7)$ is clearly below. In fact, in all cases, $\sigma_c(7)$ is smaller than $\sigma_c(6)$.

In bromine ions with charge $q = 5$, all five 4p electrons are removed. A slight inflection from $q = 5$ to $q = 7$ corresponds to removal of the two 4s electrons and the jump at $q = 8$ corresponds to removal of the first electron from the 3d shell. On the basis of energetics and of density states, one might expect capture to occur principally to highly excited levels of the lower-charge state ion. This effect would tend to blur any clear-cut shell boundaries in electron capture. An alternative explanation can be made if one proposes the necessity for small excitation of a bound electron to stabilize electron capture. In this case the availability of a 4s electron enhances the $\sigma_c(6)$, while the necessity for exciting an electron out of a closed shell depresses $\sigma_c(7)$. In any case, a more detailed knowledge of the actual capture process is required in order to fully understand the shell effects observed in the cross sections.

An important effect which occurs in heavy ion collisions with heavy targets

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is multiple ionization. In Fig. 6 we have plotted the charge changing cross sections for 110 MeV I^{12+} ions in H_2 , He and Ar [7]. The point at 13+ is the single electron loss cross section; the point at 14+ the double loss cross section, etc. Here it can be seen that in the Ar target the cross section for loss of as many as 12 electrons in a single collision is still measurable and

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that the sum of the multiple loss cross sections is actually greater than single loss cross section. The cross section for ionization to states higher than 25+ drops sharply since it requires removal of an electron by the cross sections given for 162 MeV I^{17+} in O_2 . Note also that multiple ionization in light target gases is much less significant. The implication here is that multiple ionization involves multiple electron interaction. We will discuss this more fully later.

2.3. Effects of Dense Media on Charge Changing Collisions

At this point it would be well to consider again the role of excitation in charge changing collisions. Although, as we shall later see, inner shell ionization is also of importance, let us confine our attention to the outer remaining shell of the ion. If only a single electron in the outer shell is excited, only radiative deexcitation is possible with lifetimes in the order of 10^{-10} sec. If multiple excitation takes place with much shorter lifetimes, $\sim 10^{-12}$ sec, associated with autoionization (and hence electron loss) are also possible.

The probability that an excitation caused by one collision will still be present in a subsequent collision will depend on the mean time between collisions ($\tau = 1/n\sigma v$). If we assume a cross section of 10^{-16} cm² for a given process and consider a gas target at ~ 1 torr (5×10^{16} atoms/cm³) we can see that with a velocity of say $v_0 = 2 \times 10^8$ cm/sec the time between collisions is 10^{-9} sec, that even in this relatively dilute target excited states can be of importance

in determining the equilibrium charge. This can be seen in Fig. 7. The difference between the two experiments is that in one case a long cell (150 cm) was used which assured relaxation between collisions whereas in the short cell (3 cm) pressures in the order of 1 torr were necessary to attain the same target thickness. (Note here that the depression at charge 6+ is due to the previously noted depression in capture cross section.)

If one now considers the atom density in solid targets ($\sim 5 \times 10^{22}$ atoms/cm³)

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The probability that an excitation caused by one collision will still be present in a subsequent collision will depend on the mean time between collisions ($\tau = 1/\nu$). If we assume a cross section of 10^{-16} for a given process and consider a gas target at ~ 1 torr (5×10^{16} atoms/cm³) we can see that for a velocity of say $v_0 = 2 \times 10^8$ cm/sec the time between collisions is 10^{-9} sec. So even in this relatively dilute target excited states can be of importance in determining the equilibrium charge. This can be seen in Fig. 7. The difference between the two experiments is that in one case a long cell (150 cm) was used which assured relaxation between collisions whereas in the short cell (3 cm) pressures in the order of 1 torr were necessary to attain the same target thickness. (Note here that the depression at charge $6+$ is due to the previously mentioned depression in capture cross section.)

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Insert Fig. 8.

2.4. Inner Shell Ionization

Thus far all we have considered is the exchange of outermost electrons, whereas for high velocity heavy ions inner-shell ionization can contribute measurably to the charge state distribution and in the case of collision in dense media can lead to a steady state inner-shell excitation which will be relaxed only when the particle emerges into vacuum.

Two distinct mechanisms for inner-shell ionization in heavy particle collisions have been investigated: Coulomb ionization and quasi-molecular electron promotion.

Consider first inner-shell ionization which occurs by the collision of fast light particles in which the Coulomb interaction between particles and electrons is predominant and provides the perturbation necessary to cause the excitation of the inner bound electrons to outer states. Clearly this mechanism is only strictly valid if the probability for ionization is small enough and the speed large enough. This condition is met if the scattered wave amplitude from the collision between particle of atomic number Z with velocity v and the

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atomic electron have small amplitude; i.e., if $Ze^2/h < 1$. The theory for this type of ionization has been worked out using the plane wave Born approximation (PWBA) and the classical binary encounter approximation (BEA).

For K-shell ionization with protons a great mass of data exists at energies ranging from 100 keV to 160 MeV on many targets. The results are in fact quite good agreement with the predictions. In Fig. 9 a plot of some of the data in terms of reduced units of energy is compared with theory [9]. New data and some modifications in theory to take into account non-linear polarization and binding effects have further reduced the discrepancies. Qualitatively speaking the cross section for Coulomb ionization first increases with velocity ($\sigma \sim E^4$ at lower velocities), reaches a maximum at a velocity equal to the orbital electron velocity and then decreases. Thus as the projectile velocity increases maxima in the ionization cross sections for the outer shells occurs first and then the next inner shell, etc.

As the Z of the ion increases the ionization cross section naturally increases as Z^2 but other effects also come into play. Recent work with heavier ions, for example, carbon, oxygen, and nitrogen, in the MeV per nucleon region, carried out in a number of laboratories has shown that, in addition to normal Coulomb ionization of, for example, K electrons in a target, multiple ionization of adjacent shells (L, M, ...) occurs during the same collision. In Fig. 10 the normal K line from aluminum produced by proton bombardment is compared with the spectrum observed with 14-MeV nitrogen-ion bombardment. In the latter case a whole set of lines is produced in the $K\alpha$ region. Each of the lines in this complex set comes from transitions which take place while the excited atom has in addition to the K vacancy, L vacancies; for example, line No. 2 has a single L vacancy, No. 3 a double L vacancy, etc. Thus the shift in the line positions in resolution measurements of the K x-ray lines can be used to estimate the number of simultaneous excitations in the L shell. A great body of experimental work on this subject has come into existence in the last two years, and the theory is based essentially on a binomial distribution for independent events applied to the ionization of the various shells.

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As the Z of the ion increases the ionization cross section naturally increases as Z^2 but other effects also come into play. Recent work with heavier ions, for example, carbon, oxygen, and nitrogen, in the MeV per nucleon region, carried out in a number of laboratories has shown that, in addition to normal Coulomb ionization of, for example, K electrons in a target, multiple ionization in adjacent shells (L, M, ...) occurs during the same collision. In Fig. 10 the normal K line from aluminum produced by proton bombardment is compared with the spectrum observed with 14-MeV nitrogen-ion bombardment. In the latter case a whole set of lines is produced in the $K\alpha$ region. Each of the lines in this complex set comes from transitions which take place while the excited atom has, in addition to the K vacancy, L vacancies; for example, line No. 2 has a single L vacancy, No. 3 a double L vacancy, etc. Thus the shift in the low-resolution measurements of the K x-ray lines can be used to estimate the degree of simultaneous excitation in the L shell. A great body of experimental data on this subject has come into existence in the last two years, and the theory based essentially on a binomial distribution for independent events appears

to describe most of the results.

Even more recent work by Sellin and co-workers at ORNL has revealed dramatic effect of particle charge state on Coulomb ionization cross section. An example is shown in Fig. 11 where the cross section for Ne K x-ray produced in a single collision with 80 MeV Ar ions is plotted as a function of the Ar ion charge state [11]. It is seen to vary exponentially and increase by a factor of ~ 60 with increasing charge state. We will utilize this observation later when we discuss excitation states in solids.

Ionization by quasi-molecular promotion is dominant at ion velocities below electron orbital velocities and was first observed in relatively low energy collisions a little more than 10 years ago. Multiple ionization and large angle scattering of Ar at energies as low as 25 KeV was observed.

of the results of Everhart and Kessel [12] is shown in Fig. 12. At a distance of $0.24 \text{ \AA}$ corresponding to overlap of the atomic L shells the degree of ionization is seen to increase from 1 to 3 and corresponds to the liberation of 2 electrons from the Ar L shell.

A qualitative explanation of this data was proposed by Fano and Lieberman. They proposed a model involving transitions between individual electron orbitals in a transient diatomic structure. Strong coupling of deep lying levels consistent with the exclusion principle promotes certain electrons to ionization as the ions depart from each other after collision. The quasi-molecule formed during the collision is describable in terms of the united atom model for electron orbitals.

Such a correlation diagram for the Ar-Ar system is shown in Fig. 13 (the united atom (i.e., $r = 0$) in this case is Kr). This shows that when two Ar atoms are brought together two of the 2p electrons are put into the $4s$ molecular orbital. This orbital is highly repulsive and makes many crossings with other orbitals. The probability of these electrons returning to their initial orbitals after a violent collision is practically zero. These levels start to interact at distances corresponding to L shell radii. Hence the observation of an effective geometric cross section for ionization of two L electron electrons in such

describe most of the results.

Even more recent work by Sellin and co-workers at ORNL has revealed a systematic effect of particle charge state on Coulomb ionization cross section. An example is shown in Fig. 11 where the cross section for Ne K x-ray production created in a single collision with 80 MeV Ar ions is plotted as a function of Ar ion charge state [11]. It is seen to vary exponentially and increase by a factor of ~ 60 with increasing charge state. We will utilize this observation later when we discuss excitation states in solids.

Ionization by quasi-molecular promotion is dominant at ion velocities much lower than electron orbital velocities and was first observed in relatively low energy collisions a little more than 10 years ago. Multiple ionization in large angle scattering of Ar at energies as low as 25 KeV was observed. A plot of the results of Everhart and Kessel [12] is shown in Fig. 12. At a distance of $0.24 \text{ \AA}$ corresponding to overlap of the atomic L shells the degree of ionization was seen to increase from 1 to 3 and corresponds to the liberation of 2 electrons from the Ar L shell.

A qualitative explanation of this data was proposed by Fano and Lichten [13]. They proposed a model involving transitions between individual electronic orbitals to a transient diatomic structure. Strong coupling of deep lying levels consistent with the exclusion principle promotes certain electrons to ionization when the ions depart from each other after collision. The quasi-molecule formed in collision is describable in terms of the united atom model for electron orbitals. Such a correlation diagram for the Ar-Ar system is shown in Fig. 13. The united atom (i.e., $r = 0$) in this case is Kr). This shows that when two atoms are brought together two of the 2p electrons are put into the 4f molecular orbital. This orbital is highly repulsive and makes many crossings. The probability of these electrons returning to their initial orbitals after a violent collision is practically zero. These levels start to interact at distances corresponding to L shell radii. Hence the observation of an essentially symmetric cross section for ionization of two L electron electrons in such collisions.

Barat and Lichten [14] have shown that it is really quite simple to predict which electrons will be ionized in a collision involving two dissimilar atoms. The 1s electrons of the lower Z atom correlate along the 2s orbital to the 2s orbital of the united atom, etc. When we get to the 2p electrons of the lower Z atom, they are placed into the 4f orbital which leads to ionization. There are many other possibilities including rotational coupling of non-crossing levels which we will not discuss here. The main point I wish to make is that large cross sections for ionization exist which moreover will depend strongly on the individual collision pair.

The yield and nature of the x-ray spectrum can thus be a strong function of the atomic state of the penetrating ion and in the last two years a number of studies have been made utilizing this observation. Here I will just mention two. The first involves x-ray production in solid and gas targets with iodine ions in Se, Br₂ and Kr targets [15]. These are adjacent elements in the periodic table and the principal difference is that Se is a solid while Br₂ and Kr are gases. From observations of the I L x-ray spectra (energy spectra) it could be determined that the state of the I ion following an L shell ionization was the same for all targets. This is not unexpected because an atypically violent collision is necessary to create such a vacancy. Nonetheless, the cross section for L shell ionization was shown to be larger by a factor

Insert Fig. 14.

in Se as compared to Kr as shown in Fig. 14 which shows absolute yields of x rays observed with a proportional counter. The principal difference in x-ray production in solid and gaseous targets appears to arise from the state of the ion prior to the collision. The high collision frequency in a solid lead to considerable steady-state ionic excitation in the M shell of I. For example, in the ion energy range investigated, the I M-vacancy production cross section in the system I-Se is approximately 10^{-17} cm^2 . The lifetime of an M vacancy will be about $5 \times 10^{-14} \text{ sec}$, while the time between exciting collisions in the solid will be about $5 \times 10^{-15} \text{ sec}$. Thus, in a solid the ion would be expected to have, on the average, several M-shell vacancies.

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it could be determined that the state of the I ion following an L shell event was the same for all targets. This is not unexpected because an atypically violent collision is necessary to create such a vacancy. Nonetheless, the cross section for L shell ionization was shown to be larger by a factor of 10 for Se as compared to Kr as shown in Fig. 14 which shows absolute yields of x-rays observed with a proportional counter. The principal difference in the x-ray production in solid and gaseous targets appears to arise from the state of the ion prior to the collision. The high collision frequency in a solid can lead to considerable steady-state ionic excitation in the M shell of I. For example, in the ion energy range investigated, the I M-vacancy production cross section in the system I-Se is approximately 10^{-17} cm^2 . The lifetime of an M-vacancy will be about 5×10^{-14} sec, while the time between exciting collisions in the solid will be about 5×10^{-15} sec. Thus, in a solid the I ion should be expected to have, on the average, several M-shell vacancies. This

2

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In a recently completed work we studied yields of Si-K x rays by
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The case of argon ions presents a different picture. The x-ray yield
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$\bar{q} = 14$. The same prefoil caused no difference in yield when the solid target was used. These observations imply that the argon ions emerging from the solid into vacuum lose ~ 3 electrons by auto-ionizing and/or Auger events. (This observation is in accord with the proposed model of Betz and Grodzins [17] relating to excitation of heavy ions in solids.)

3. Application to Ions Recoiling Into Vacuum and Gases

The presence of excited states could greatly disturb the determination of σ_{eff} . Insert Table II. σ_{eff} as can be seen from a perusal of Table II which lists the lifetimes of various allowed transitions for some 1, 2, and 3 electron systems. Of course, states present in metastable configurations will not contribute at all to the atomic factor.

From all of the preceding sections it is clear that excitation of ions emerging from solids is ubiquitous especially in the case of heavy ions where inner shell excitation is also present. At least in the case of inner shell excitation the states are relaxed in times short compared to 10^{-12} seconds. The degree of outer shell excitation which results from this relaxation has not yet been determined. In fact, very little has been determined concerning the population of excited states of emergent ions. In the great amount of work that has been done on optical beam foil spectroscopy there have been no quantitative intensity measurements so that no population information has been obtained.

Only a few experiments bring information directly to bear on the problem. Sellin et al. [18] in their work on metastable auto-ionizing $1s2s2p$ quartet states found that, rather than being a truly rare species, about 1% of the 3 electron ions are in these states. Although admittedly this represents the end product of many cascades it implies that many more ions leave the foil in shorter lived radiative states which can interfere with PAC measurements.

The most direct experiment to date, albeit not a solid foil experiment, has been carried out by Macdonald and co-workers [19]. They measured the electron capture cross section for F^{9+} in Ar and simultaneously measured

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The presence of excited states could greatly disturb the determination of yield. This can be seen from a perusal of Table II which lists the lifetimes of various allowed transitions for some 1, 2, and 3 electron systems. Of course, ions in metastable configurations will not contribute at all to the attenuation.

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the absolute cross sections for K x rays emitted following capture into a given excited state. They found that the total x-ray emission cross section was ca. 70% of the total charge capture cross section, i.e., 70% of the capture took place into optically allowed excited states (emission from metastable states would not have been observed on the time (distance) scale of the experiment). The relative amounts of capture into a given state followed the expectations of the Brinkman-Kramers expression as formulated by Dimitriev. In this experiment it can be shown that for capture from the Ar K shell the expected dependence is $1/n^3$ or stronger, i.e., the bulk of the capture takes place into the 1s shell of F^{8+} . However, the total capture from Ar K is quite small compared to capture from Ar L. For capture from the Ar L shell into the $n = 1, 2$ and 3 levels of F^{8+} the calculated ratios are 1:5.7:2.9. Thus if electron capture is the last event which gives the one electron configuration a large fraction of the system will not be in the 1s state during the nuclear decay period. This observation is not in contradiction to PAC measurements on recoil into vacuum where the attenuation factor is seen to grow with increasing population of one electron configurations. It simply means that the g factor determined from these measurements might be too small. In the one reported experiment where PAC measurements were carried out in coincidence [20] with a given emergent

Insert Fig. 18. charge state (See Fig. 18). The attenuation for O^{7+} is probably considerably smaller than the true attenuation that would obtain with a pure O^{7+} 1s ion. The presence of excited states is strongly evidenced by the attenuation in coincidence with charge 6+ where the ground state, $1s^2 2s^2 2p^2 S_0$, should lead to zero attenuation.

These conclusions are based on the assumption that the excitation state of the ion emerging from a solid is the same as that obtained from a single ionization event from Ar gas. Although in general it is found from beam foil measurements that higher excitation states are observed after emergence from solid targets, it cannot be determined without further experiment what the effects of excitation will be on hyperfine interactions of ions recoiling into vacuum.

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Table I

The field $H(0)$ generated by some few-electron configurations.

ATOMIC SPECIES	$H(0)$	$[MG]$		
	$1^2S_{1/2}$	$2^2S_{1/2}$	$2^2P_{1/2}$	
	$(1s)^1$	$(1s)^2 (2s)^1$	$(1s)^2 (2s)^2$	$(2p)^1$
OXYGEN	85.5	7	1	
FLUORINE	122	10	2	
NEON	167	15	3	
MAGNESIUM	289	30	6	

Table II

<u>Ground State</u>	<u>Ion</u>	<u>Transition</u>	<u>Lifetime τ(psec)</u>
$1s^2 2s_{1/2}$	O^{7+}	$1s - 2p$	0.4
		$1s - 3p$	1.3
		$1s - 4p$	3.0
$1s^2 1s_o$	O^{6+}	$1s^2 - 1s2p$	0.3
		$1s^2 - 1s3p$	1.0
$1s^2 2s^2 2s_{1/2}$	O^{5+}	$2s - 2p$	2500
		$2s - 3p$	40
$1s^2 2s^2 2s_{1/2}$	P^{12+}	$2s - 2p$	1000
		$2s - 3p$	1.6

Figure Captions

- Fig. 1. A frequently used experimental arrangement with cylindrical symmetry around the beam, yielding high recoil velocities.
- Fig. 2. The quantity $(\omega\tau)^2/[1 + (\omega\tau)^2]$ as a function of the nuclear parameter $g\tau (Z/8)^3$.
- Fig. 3. Mean relative charge at equilibrium $\bar{q}/Z$ for various ions emerging from O_2 and N_2 targets as a function of reduced velocity.
- Fig. 4. Mean charge for ^{127}I ions at equilibrium in O_2 and N_2 targets.
- Fig. 5. Charge capture cross sections for 13.9 MeV Br in H_2 (o), He ($\square$) and Ar (Δ).
- Fig. 6. Charge capture and loss cross sections for 110 and 162 MeV I ions.
- Fig. 7. Equilibrium charge distributions for 12 MeV Br at two different gas pressures.
- Fig. 8. Most probable charge states of bromine ions emerging from solid and gaseous targets.
- Fig. 9. K shell ionization cross sections versus proton energy. The solid curve is the PWBA calculation.
- Fig. 10. Al K x-ray spectra induced by proton and 14 MeV nitrogen ion bombardment showing increase in multiple L ionization.
- Fig. 11. Intensities of Ne K and Ar K x rays as a function of Ar charge state for 80 MeV Ar on Ne.
- Fig. 12. Most probable charge state of Ar ions from single collision with Ar as a function of distance of closest approach.
- Fig. 13. Diabatic energy level diagram for the Ar-Ar system as a function of internuclear distance.
- Fig. 14. Proportional counter x-ray spectra from collisions of 42.5 MeV I in Se (solid) and Kr and Br_2 .
- Fig. 15. Diabatic correlation diagram for I - Se system.

Fig. 16. Si K ionization cross section obtained from 40 MeV O ion bombardment of SiH₄ gas and Si as a function of incident charge state.

Fig. 17. Si K ionization cross section obtained from 86 MeV Ar ion bombardment as a function of incident charge state.

12 MeV I Ions in Ar Gas.

Fig. 18. Composite graph of all K-shell ionization cross sections from proton bombardment at the Duke University VAN DE GRAAFF accelerator. See text for explanation of coordinates and curves.

ORNL-DWG. 74-5147