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CHARGE STATES AND EXCITATION OF FAST HEAVY IONS PASSING THROUGH SOLIDS: A NEW MODEL FOR THE DENSITY EFFECT*

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The theory of Bohr and Lindhard for electron capture and loss by heavy ions penetrating through solids has been modified. It is concluded that the difference in the average equilibrium charge of fast ions traversing solids and dilute gases is mainly due to Auger processes which occur after the ions leave the solid. The charge states inside solids are—in disagreement with the deductions of Bohr and Lindhard—not much larger than those in gases.

The mean ionization $\bar{q}$ of energetic heavy ions which penetrate through matter reaches an equilibrium value determined by the competition between capture and loss of electrons. It has been known for many years that $\bar{q}$ is greater when the ions traverse solids rather than gases. Lassen¹ found this effect in a study of fission fragments, and many further experiments²⁻⁶ revealed even more pronounced differences. An example is given in Fig. 1, which shows the equilibrium charge spectrum of 12-MeV iodine ions following passage through oxygen gas and a carbon foil.⁶ The resulting mean charges are 6 and 12, respectively.^{7,8} It has always been assumed that such investigations measure essentially the equilibrium charge distribution inside the stripper.

The difference $\Delta\bar{q}$ between the mean charge in a solid compared with a gas stripper was explained by Bohr and Lindhard⁹ (BL) in a pioneering paper of 1954, from which we quote below and to which we refer hereafter as the BL theory. In this generally accepted model, those collisions of the ions with the target atoms which do not produce ionization result in excitation of the ion's most loosely bound electron. In a dilute gas stripper, de-excitation back to the ion's

ground state takes place before the next collision occurs. In a solid, however, the collision rate is too high for such radiative de-excitation to be important. Rather the collisions constantly increase the excitation of the outermost electron until that electron is lost. This reasoning applies consecutively to inner electrons and, consequently, the charge of the ion will steadily increase until the capture cross section becomes large enough to match the probability for loss of the excited electron. A new equilibrium value of the mean ionization is finally maintained by direct competition between capture and loss. Mainly one or, at most, a very few electrons will be in states of fairly high excitation and "an electron captured into an excited state will be lost from the same state." Thus, "suppression of readjustment of ion excitation" is expected to be the principal mechanism for increasing the charge inside solids to the values observed after the ions passed through the target, though Bohr and Lindhard also state that "the high excitation of the ions in solids may result in a subsequent emission of electrons from the ions immediately after their escape into vacuum, which increases the mean charge to a certain extent."

Several recent investigations have shown that

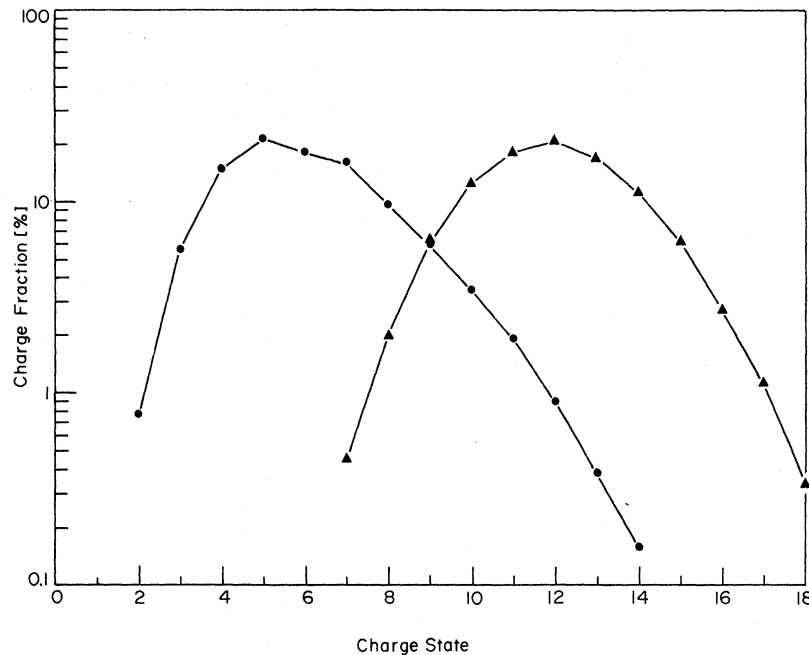


FIG. 1. Equilibrium charge spectrum of 12-MeV iodine ions following passage through oxygen gas (circles) and a carbon foil (triangles). See also Ref. 7.

the stopping power for heavy ions is nearly independent of whether the medium is a gas or a solid.^{10,11} These results are difficult to understand if the mean-square charge inside a solid differs markedly from that in a gas, as predicted by the BL theory.

The modification to the BL model is suggested by experiments with Cl, Br, and I ions in dense gases which show that the total cross section for electron loss does not increase significantly with the density of the target gas.^{7,8} These results lead to the conclusion that the electron loss per atom is hardly affected by the excitation of a single electron. As a consequence, we argue, substantial excitation due to successive collisions is probable not only for a single particular electron but for all of the many outer electrons which also contribute to the loss process. Thus, under equilibrium conditions, a large number n of outer electrons will have an average excitation $\epsilon\bar{I}$, where $\bar{I}$ is an average ionization potential and $0 \leq \epsilon < 1$. In a shell-model description, one expects that ϵ has almost the same value for all electrons in a subshell, and decreases rapidly for inner shells. Within the solid, the equilibrium charge will be determined by competition between capture, on the one hand, and loss due to direct collisions and Auger processes on the

other hand. We conjecture that the Auger effect serves as a readjustment process which, in competition with further excitation, contributes to maintaining the equilibrium level of excitation. Although the probability for occurrence of an Auger process per ion is difficult to estimate, one expects on general grounds that it is proportional to $n(n-1)$ and will increase with the average excitation ϵ .

For a quantitative discussion of the consequences of our modification, it is necessary to consider the cross sections which govern the charge-exchange processes between ions of various charge states q . We illustrate the calculation by using the linear BL approximation for dilute gases:

$$\begin{aligned}\sigma_i(q) &= \sigma_0[1 + \alpha(\bar{q}_g - q)], \\ \sigma_c(q) &= \sigma_0[1 - \beta(\bar{q}_g - q)],\end{aligned}\tag{1}$$

where $\bar{q}_g$ is the mean equilibrium charge, for which loss and capture cross sections are of equal magnitude σ_0 . A very small residual excitation ϵ is expected to increase the loss cross sections in solids to $\sigma_i(q) + \epsilon\sigma_0$. Equating the increased loss with the capture cross section, Eq. (1), one obtains a mean charge, which is shifted from $\bar{q}_g$ by $\Delta\bar{q}_i = \epsilon/(\alpha + \beta)$. Equation (1) leads

also to an expression for the full half-width Γ of the equilibrium charge distribution,⁹ obtained in the absence of excitation, $\Gamma = [8 \ln 2 / (\alpha + \beta)]^{1/2}$. Elimination of α and β then yields:

$$\Delta \bar{q}_i = (\Gamma^2 / 8 \ln 2) \epsilon \quad (\epsilon \ll 1). \quad (2)$$

Equation (2) is deficient in several aspects. First, the dependence of the cross sections on q is generally stronger than estimated in Eq. (1). Experimental results on heavy ions suggest relations of the form $\sigma(q) \propto q^a$. Second, higher equilibrium excitation should be included. We take this into account and describe the related increase in the probability for removing one or more electrons in single collisions by a factor $f_i(\epsilon)$. As f_i will depend on q , we define $\bar{f}$ as the average of f_i over all relevant charge states. $\bar{f}$ may also contain a small decrease of the capture cross section, as well as the part of the loss due to the Auger ionization inside the solid. (For small excitation, the BL theory suggests the approximation $\bar{f} = 1 + \epsilon$). Finally, the distribution width is broadened by a factor γ , due to the influence of collisions with target atoms in which k electrons are simultaneously lost or captured ($k > 1$). We denote the average probabilities of these events compared with single loss and capture by γ_k^l and γ_k^c . With these additions, Eq. (2) changes to the closer approximation

$$\Delta \bar{q}_i = [\Gamma^2 / (8 \gamma^2 \ln 2)] \ln \bar{f}(\epsilon), \quad (3)$$

where

$$\gamma^2 = \sum_k k^2 \gamma_k^l / (2 \sum_k \gamma_k^l) + \sum_k k^2 \gamma_k^c / (2 \sum_k \gamma_k^c).$$

In light targets, where multiple loss and capture is small, γ^2 is not much larger than 1, but in heavy targets we must reckon with $\gamma^2 \approx 2$ or more. Numerous experiments are in accord that, for a given ion species Z , Γ does not vary too much, provided that $\bar{q}_e$ is not too close to 1 or Z . For example, Γ is approximately 3.5 for Br and 4.5 for I ions, stripped in argon gas, over a wide range of $\bar{q}_e$. In these examples, the factor in brackets in Eq. (3) is then of the order of 1.

After leaving the solid, the total excitation of the ion may be sufficient for electron emission. The upper limit of the number of possible Auger electrons is approximated by

$$q_A = n\epsilon. \quad (4)$$

In the BL model, the small value of n limits q_A and a substantial increase of the mean equilibrium charge results from $\Delta \bar{q}_i$. In the present model, however, n is large and an evaluation of

Eqs. (3) and (4) shows that $\Delta \bar{q}_i$ is never much greater than 2 and is generally about 1 or less. A pronounced effect ($\Delta \bar{q} \gg 1$) must be attributed to large values of q_A . This conclusion holds quite independently of ϵ and, thus, a detailed calculation of ϵ and $\bar{f}(\epsilon)$ is not required.

As an example, we consider iodine ions at 12 MeV (see Fig. 1). Under equilibrium conditions the most probable charge in dilute oxygen gas is about 5 so that at most a few electrons remain on the average in the O shell. A major contribution to excitation and loss processes will thus come from the 18 electrons in the N shell. In a solid, with ϵ as high as 0.5, the increase in the loss cross section gives only a shift [Eq. (3)] of $\Delta \bar{q}_i \approx 1$; even with $\epsilon = 0.85$, $\Delta \bar{q}_i$ is only 2. On the other hand, with values of ϵ larger than 0.3 the Auger process [Eq. (4)] readily accounts for the remaining (and principal) difference of about 6 electrons.

The present model, which is not in contradiction with any experimental data, offers simple explanations of several experimental phenomena, and leads to some predictions which could be tested.

(1) With the approximate equality of charge states inside gases and solids, no dramatic discrepancy remains in the evaluation of energy-loss measurements for heavy ions in gases and solids, though 20% effects may still persist [Eq. (3)] in some energy range where $\bar{q}_e$ is not too large. We emphasize that the detailed mechanism of energy loss in solids is still not fully understood. For example, 20% effects may well result from the increased orbital dimensions of the outer electrons on the highly excited atoms.

(2) Equilibrium charge distributions in heavy gases are found to show greater asymmetries than those obtained from solids. This effect is clearly visible in Fig. 1 and we attribute parts of it to the final Auger process, which—itsself statistical—tends to symmetrize the initial asymmetries to an extent proportional to the final charge increase. Incidentally, the distribution should be somewhat broadened at the maximum. In addition, the total excitation energy, $\epsilon n \bar{l}$, may diminish with increasing charge state inside the solid, which further enhances a steeper decrease of the final charge distribution for charges above the mean.

(3) In a solid, equilibrium in the excitation will be attained only after all relevant n electrons have experienced sufficient collisions. One therefore expects that the equilibrium thickness

d_{∞} in solids is noticeably larger than in gases. There is some evidence that d_{∞} in a carbon foil is 10 times as much as in argon gas for Br ions at 140 MeV.⁵

(4) In dilute gaseous targets, charge-changing cross sections can be derived from the measurement of nonequilibrium charge distributions. A similar technique is applicable to solids only if one also includes the complex excitation cross sections and Auger probabilities.

(5) We conjecture that inside a solid the equilibrium value of $n\epsilon$ will be reached when the Auger process which results in de-excitation and electron loss balances collision excitation and loss. In that case, the mean time for de-excitation by the Auger effect is of the order of

$$\Delta t \approx n/(\sigma^* g V), \quad (5)$$

where σ^* , g , and V are an effective excitation cross section, the density of the solid, and the ion velocity, respectively. In the above example for iodine ions passing through carbon, we estimate $\sigma^* \approx 4 \times 10^{-16} \text{ cm}^2/\text{atom}$ and Eq. (5) leads to $\Delta t \approx 10^{-15} \text{ sec}$, a not unreasonable lifetime for the Auger effect. This mean time is then also the time for the first electrons to de-excite after the ions leave the solid.

Possibilities for experimental tests of the present model are evident. We also note that the properties of the emerging ions—the many states of excitation and angular momentum—have implications for several fields such as beam foil spectroscopy or perturbed angular correlations

of nuclear states following recoil from solids into vacuum.

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DETECTION OF A VORTEX-FREE REGION IN ROTATING LIQUID HELIUM II†

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We observe a nonlinear dependence of the number of vortex lines in a rotating container on the rotation frequency. The data are consistent with a model in which there are about two missing rows of vortices near the walls.

The question of the distribution of quantized vortex lines in rotating superfluid helium has received considerable attention, both theoretical^{1,2} and experimental,³⁻⁷ in the past decade. Most experimental studies have been carried out either in situations in which the distribution is indistinguishable from a uniform one, or at the other extreme, near the threshold for production of vortices. In the latter case the distribution may be determined more by the dynamics of vortex nu-

cleation than by considerations of thermodynamic equilibrium.

We will present here the results of an experiment concerning an intermediate region—one far enough from threshold so that nucleation should not interfere with the achievement of thermodynamic equilibrium, yet close enough so that certain nonuniformities in the distribution predicted by the equilibrium theory become apparent.

In the absence of boundaries the free energy of