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EFFECTS OF THE DENSITY OF GASEOUS TARGETS ON ELECTRON CAPTURE AND LOSS BY FAST HEAVY IONS*

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Recent experimental charge-exchange studies of heavy ions have been utilized to modify the theory of Bohr and Lindhard for electron capture and loss by heavy ions passing through dense gases. New results are presented for the difference between the average ion charges obtained in dense and dilute gases, as well as for the residual ion excitation after charge-changing collisions. Implications are indicated for the measurement of radiative lifetimes of excited heavy ions.

In early studies with fission fragments, Lassen[1] showed that the distribution of charge states in a heavy-ion beam penetrating through gaseous media may be strongly influenced by the density of the target (see Fig. 1 for recent illustrative data). This dependence was attributed by Bohr and Lindhard[5] (BL) to the effect of stripping from excited states of the ions. Later experiments[6-8] confirmed the gross predictions of the BL theory, but direct measurements of charge-changing cross sections in dense gases[9] reveal the necessity of refining modifications.

The purpose of this Letter is to interpret recent experimental results in order to achieve an improved description of the density effect.[10] In particular, we obtain a more reliable estimate for the difference between the mean ionic equilibrium charges measured in dense and dilute gases, and also useful information about the states of ionic excitation in which fast heavy ions are left after charge-changing collisions. These refinements are also essential for the experimental investigation of effective radiative lifetimes for excited states of even highly ionized heavy projectiles. It will be shown that the density effect may already occur at much lower target densities than hitherto believed; in general, the effect cannot be disregarded in gas targets intended to produce charge exchange of heavy ions at energies below approximately 40 MeV.

The basic assumption of the BL theory is that an excited electron can be stripped from a heavy ion more easily than an electron which is bound in the ground state. In a dense gas target, the most loosely bound electron in colliding ions is considered to have an average residual excitation $\bar{\epsilon}I^*$, where I^* denotes the ground-state ionization potential for that electron. It is expected that the cross section for loss of such an excited electron increases substantially when $\bar{\epsilon}$ increases. In addition, the BL model takes into account that capture by an excited ion may lead to a state in which the total excitation energy exceeds the binding energy of the most weakly bound electron. In these cases, an electron will be ejected by a rapid autoionization process, so that the capture cross section appears to be reduced. In the BL theory, the cross sections per atom for loss and capture of a single electron by excited heavy ions of charge q are given by the linear approximations

$$\sigma_l^*(q)=\sigma_0[1+\alpha_l(\bar{q}-q)]+\sigma_0\beta_l\bar{\epsilon}, \tag{1a}$$

$$\sigma_c^*(q)=\sigma_0[1-\alpha_c(\bar{q}-q)]-\sigma_0\beta_c\bar{\epsilon}, \tag{1b}$$

where the first terms on the right-hand side denote the cross sections for the ground state (σ_l and σ_c), and σ_0 is the cross section when $\sigma_l(\bar{q})$

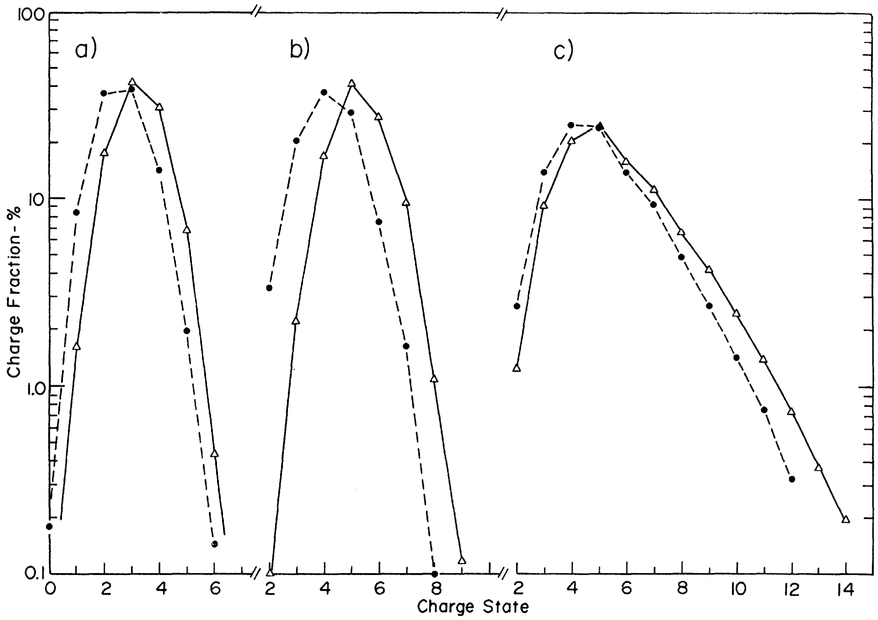

FIG. 1. Equilibrium charge-state fractions in dilute and dense gases for (a) 4-MeV chlorine ions in H_2, (b) 14-MeV bromine ions in H_2, and (c) 12-MeV iodine ions in Ar. The three distributions in dense gases (triangles) have been measured by Ryding (Ref. 2) at target densities of approximately 3×10^{16} molecules/cm^3. The distributions for low target densities [full circles in (a) and (b)] have been computed (Ref. 3) from experimental ground-state cross sections. In (c) the low target densities (full circles) have been measured by Moak (Ref. 4) at a target density of approximately 6×10^{14} molecules/cm^3. In the case of iodine in argon, the low and high densities still do not differ sufficiently to produce the theoretical maximum difference in the charge distributions.

$=\sigma_c(\bar{q})$. The values of α_l and α_c are approximated by $3/\bar{q}$ and $2/\bar{q}$. Equating $\sigma_l{}^*$ with $\sigma_c{}^*$, Eq. (1) yields the shift of the mean charge $\bar{q}$ in the equilibrium charge distribution, $\Delta\bar{q}=\bar{\epsilon}(\beta_l+\beta_c)/(\alpha_l+\alpha_c)$. The residual ion excitation after electron loss is expected to amount on the average to about $\frac{1}{2}I^*$, whereas the average excitation of an electron after capture by an ion in the ground state is believed to be about $\frac{2}{3}I^*$ in heavy targets, but higher in light targets such as H^2 or He. On those grounds, Bohr and Lindhard approximate $\bar{\epsilon}\approx\frac{1}{2}$ and $\beta_l\approx\beta_c\approx1$ for heavy target gases, which results in their estimate for the maximum of the density effect,

$$\Delta\bar{q}=\tfrac{1}{5}\bar{q}. \tag{2}$$

In light targets, and especially for very fast ions, BL predict larger values of $\Delta\bar{q}$ than one would deduce from Eq. (2).

The modifications of the BL theory for the density effect in gases which we present below are based on a variety of charge-exchange measurements for heavy ions. For the present purpose, the relevant results are these:

(1) Direct measurements of electron-loss cross sections for heavy ions do not show the predicted increase [Eq. (1a)] with the density of the target gas.[9] This leads to a substantial modification of the BL estimates for electron loss from excited states: In Eq. (1a), the parameter β_l will be close to zero so that

$$\sigma_l{}^*(q)\approx\sigma_l(q). \tag{1a$'$}$$

(2) Electron-capture cross sections for excited states have been measured for Cl, Br, and I ions with charge states between 2+ and 8+ and velocities between $1.2v_0$ and $4.6v_0$ ($v_0=e^2/\hbar$), stripped in targets of mostly He, but also H_2 and Ar.[3,9] In all cases studied, the effect of the excited states was to decrease the capture cross sections by a factor g_q generally between 2 and 3. Since no systematic variation of g_q was observed and since the decrease of σ_c is too large to make Eq. (1b) practical, it is advantageous to replace the BL estimate for σ_c [Eq. (1b)] by

$$\sigma_c{}^*(q)=\sigma_c(q)/g_q. \tag{1b$'$}$$

(3) The width Γ_0 of the equilibrium charge distribution[11] can be derived from the cross sections for ground-state ions,[5,12] $\Gamma_0=(\alpha_l+\alpha_c)^{-1/2}$; with the specific BL estimates for α_l and α_c one obtains $\Gamma_0=(\bar{q}/5)^{1/2}$. However, this increase of Γ_0 with $\bar{q}$ is not observed experimentally. In-

stead, in the energy ranges investigated, Γ_0 seems to be rather independent of $\bar{q}$, provided that $\bar{q}$ is not too close to zero. Thus, it is convenient to use the experimental expressions for Γ_0 in the derivation of $\Delta\bar{q}$ rather than the uncertain parameters α_l and α_c.

(4) Cross sections for loss and capture of more than one electron in a single collision should be taken into account. The addition of these probabilities leads to a broadening of the width Γ_0 by a factor γ; especially in heavy targets, γ may be as large as 1.5.[12]

In addition to points 1-4, we take into account that the ground-state cross sections for electron capture and loss are approximated better by $\sigma \propto q^a$ with appropriate values of a. Combining all of the above, we obtain a new estimate for the increase of the mean charge at high densities:

$$\Delta\bar{q} = (\Gamma_0/\gamma)^2 \ln\bar{g}(\bar{\epsilon}), \qquad (2')$$

where $\bar{g}$ is the average g_q over all relevant charge states. The factor $(\Gamma_0/\gamma)^2$ is quite insensitive to the target material, depends mainly on the ion species, and has a value of less than about 1.2 and 2 for Br and I ions, respectively. Using the experimental values for g_q noted above, $\Delta\bar{q}$ will generally be less than about 2, and will typically be about 1. These values are mostly smaller than anticipated in the BL model [Eq. (2)]. Despite the rather small increases in the mean charge, the charge fractions obtained in the equilibrium charge distributions with and without the density effect may well differ by more than a factor of 2, provided that q is not too close to $\bar{q}$.

The empirical result $\sigma_l^* \approx \sigma_l$ may be understood if one argues that in a heavy ion a large number of electrons contribute significantly to the loss process, and that the total residual excitation of an ion passing through a dense gas does not exceed I^*. In that case, even though the loss cross section for a particular excited electron will increase as anticipated in the BL theory, the total loss cross section will change very little as all the other contributing electrons are still in (or close to) the ground state. Moreover, the effects of loss from excited states are diluted by ions which are in the ground state before they undergo an ionizing collision.

The results on electron capture allow conclusions to be drawn on the residual ion excitation. First, we consider the decrease of the capture cross sections [Eq. (1b′)], which depends on the probability $W_c(\epsilon_c)$ for finding an electron in a state with excitation energy $\epsilon_c I^*$ after being captured by a ground-state ion. Useful information can be obtained from experiments in which the capturing ion of charge q was formed and excited itself by electron capture. For sufficiently long radiative lifetimes, the total ion excitation after the second capture process then amounts to about $I_t = \bar{\epsilon}_c(I_q^* + I_{q-1}^*)$, on the average, provided that $W_c(\epsilon_c)$ is not affected too strongly by charge and excitation of the capturing ion. Independent of the distribution of excitation over the electrons in the ion, autoionization can occur for $I_t \geq I_{q-1}^*$, observable in a reduced capture cross section. Then, the corresponding experimental values for σ_c/σ_c^* (point 2) lead to the conclusion that electrons are captured into states of low ($\epsilon_c < \frac{1}{2}$) and high ($\epsilon_c > \frac{1}{2}$) excitation with comparable probabilities. If capture from light targets took place preferentially into states of extremely high excitation, as is assumed in the BL theory, one should expect capture cross sections σ_c^* which are much smaller than those found experimentally. Further estimates for $W_c(\epsilon_c)$ can be derived if assumptions are made about the distribution of excitation levels. For example, if only a very few excited states exist below $\epsilon \approx \frac{1}{2}$, one may infer from the values of g_q (point 2) that electron capture into the ground state occurs with a relative probability of approximately 30% (and 70% of the capture processes form excited states with $\epsilon_c > \frac{1}{2}$).

It is essential to bear in mind that $\bar{\epsilon}_c$ generally differs from $\bar{\epsilon}$ for a dense target [Eq. (1)] where the charge distribution is assumed to be close to charge equilibrium. Then, the capturing ions are not only formed by electron capture but also by a variety of other collision processes, such as single and multiple loss, excitation without a change of charge, autoionization, and simultaneous and successive occurence of these effects. Under such conditions, the residual excitation in a dense target will be the result of a statistical competition between these processes and should, therefore, reflect to a larger extent the density distribution of the excitation levels.

The measurement of cross sections for excited states and the effective lifetimes of those states are closely connected. In targets where the time Δt_c between two successive collisions is sufficiently short, experimental nonequilibrium distributions allow the determination not only of cross sections for ground and excited states, but also of the related effective lifetimes τ_q. Such an analysis is strongly affected by the influence of excitation on the cross sections [see Eqs. (1a′) and (1b′)], and will be dealt with in detail in a later report. Preliminary results indicate that lifetimes of excited heavy ions can be much longer than anticipated in the BL theory. For example, iodine ions of charge 7+ were found to have excited states which live as long as 10^{-7} sec.[3]

As a consequence of large values τ_q, the density effect cannot be neglected in many charge-exchange measurements. For example, when I^{7+} ions are among the charge states under investigation, gases at pressures as low as 10^{-3} Torr are not necessarily sufficiently diluted to exclude the occurrence of the density effect. This sensitivity to density satisfactorily explains the apparent discrepancies in experimental equilibrium distributions which have been measured under conditions similar except for use of different ranges of target densities [see Fig. 1(c)].

Finally, it is interesting to compare the density effects in gases and solids. Although some arguments and formulas for the description of these two effects appear quite similar, basic differences exist in the mechanisms. In solids, the observed large increase in the mean charge can be explained from a model of many-electron excitation which affects only the loss cross sections and gives rise to substantial Auger processes after the ions emerge from the solid.[12] The suppression of readjustment of ion excitation between successive collisions, which leads to the buildup of excitation, can be attained only in targets with the high density of solids. The effect in gases is based both on residual excitation of essentially a single outer electron and capture of electrons into excited states followed by autoionization between collisions. This primarily influences the capture cross sections and, compared with solids, occurs at densities lower by many orders of magnitude. As far as the mean equilibrium charge states are concerned, the density effect in gases is not only much smaller than in solids, but is also more difficult to produce for higher ion velocities. Then, higher charge states with much shorter lifetimes become dominant and the gas pressures required to fulfill the condition $\tau_q \gtrsim \Delta t_c$ are outside the generally useful range.

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[9] G. Ryding, H. D. Betz, and A. Wittkower, Phys. Rev. Lett. 24, 123 (1970).

[10] The density effect in gases must not be confused with the density effect in solids, which is based on a different mechanism and has been discussed previously by H. D. Betz and L. Grodzins, Phys. Rev. Lett. 25, 211 (1970).

[11] We use the definition $\Gamma_0^2 = \sum_q F_q(\bar{q}-q)^2$, where Fq are the relative charge fractions of the equilibrium charge distribution. In case of a Gaussian distribution, Γ_0 is related to the full half-width Γ by $\Gamma^2 = \Gamma_0^2 \ln 2$.

[12] Betz and Grodzins, Ref. 10.