Electron Capture into Excited States in Collisions of Highly Charged Ions with Atoms: A Theoretical and Experimental Challenge

The current status of the studies on creation of excited states by charge transfer collisions of multiply charged ions with atoms is discussed. Both theoretical and experimental problems involved in these studies are considered and suggestions are made for resolving some of them.

INTRODUCTION

The recognition of the extremely important role that charge transfer collisions between multiply charged ions and atoms play in many high-temperature laboratory and astrophysical plasmas has led in the last six or seven years to a dramatic increase in their investigation. Particularly strong stimulus for these studies has come from thermonuclear fusion research, the search for powerful VUV and x-ray lasers and from astrophysics. Until quite recently, the main directions of the charge exchange studies involving multiply charged ions have been oriented primarily toward understanding the basic physical mechanisms governing the process, and development of adequate theoretical and experimental methods for obtaining quantitative information on the total cross sections. The progress achieved thus far along these lines has already been
reflected in several review articles.\textsuperscript{1-8} The present-day situation of the research in this field is characterized by efforts to extend the experimental studies of the process in the region of high ionic charges $q$ to gain a better understanding of the collision dynamics for complex (many-electron) target atoms and to obtain accurate information about the final state distributions of captured electrons. The last issue has particularly important practical implications (see below).

Creation of excited states is one of the major features of the charge exchange process involving highly charged ions with atoms (except at very high collision energies). This feature of the process results from two circumstances. First, the ionic potential well is much deeper than the atomic one, and consequently the initial electronic energy level is in quasiresonance with the excited levels of the highly charged ion. Second, the electron capture probability is greatest when the energy resonance condition is satisfied. Coupled together, these two circumstances lead to preferential population of a finite group of excited ionic states. Under certain conditions one can create population inversion on this basis and possibly a lasing effect.\textsuperscript{9,10} In present-day magnetic fusion devices, the radiation from the excited impurity-ion states, created by charge transfer, may constitute a substantial fraction of the total energy loss.\textsuperscript{11,12} It can also be used for plasma diagnostic purposes.\textsuperscript{13} Modeling and diagnostics of fusion plasmas require fairly accurate knowledge of the populations of excited ionic states created by electron capture.

The first experimental evidence for capture into excited states in atom–highly-charged ion collisions has come from the spectroscopic\textsuperscript{13,14} and corpuscular\textsuperscript{15} tokamak plasma diagnostics. Anomalously intense radiation from the O$^{7+}$ and C$^{5+}$ excited ionic states respectively has been observed in these experiments after injection of neutral hydrogen beams into the plasma.

In direct ion–atom collision studies, first evidence about the formation of excited states has been obtained by the groups of de Heer\textsuperscript{16,17} and Afrosimov\textsuperscript{18} on many-electron targets. Theoretical investigations of the problem started soon after, and were limited to specific collision energies and colliding systems.\textsuperscript{19-23} Systematic investigations, both experimental and theoretical, of the electron capture into excited states in atom–multiply-charged ion (MCI) collisions have begun quite recently and this paper aims to analyze their first results and the problems encountered in the studies.

**THE CONCEPT OF RESONANCE IN THE CHARGE TRANSFER PROCESS**

Within the problem of electron capture into excited states in atom–MCI collisions,

$$A(n_0,l_0,m_0) + B^{q+} \rightarrow A^+ + B^{q-11+}(n,l,m),$$

(1)

one is mainly concerned with the question of the distribution of captured electrons over the final state quantum numbers $n,l,m$. In Eq. (1) $n_0,l_0,m_0$ are the initial state electron quantum numbers and $q$ is the ionic charge. The answer to this question depends on the collision dynamics which in turn depend on the collision energy region and the electronic structure of the colliding particles. However, there is always one particular final ionic level which in a given collision energy range is dominantly populated. This level is determined by the energy resonance condition for the process. The energy resonance condition has a dynamic character and its role in the process can be demonstrated even within the perturbation theory framework. The first-order perturbational result for the $n_0 \rightarrow n$ capture probability in an atom–fully stripped ion system is proportional to\textsuperscript{22} (hereafter we use atomic units)

$$P_n \sim n^{-3} \left[ \frac{\omega_n}{v} + \frac{v}{2} + \frac{q^2}{n^2} + Q^2 \right]^{-6},$$

(2)

$$\omega_n = (2n_0)^{-2} - (q/2n)^2,$$

(3)

where $Q$ is the transferred momentum and $v$ is the relative collision velocity. For small values of $v$ (when $Q$ is also small), the transition probability $P_n$ attains its maximum at $n_m = q/n_0$, which expresses the "static" energy resonance condition, $\omega_n = 0$. At high collision velocities, the momentum transfer term in Eq. (2) starts to play a more important role and the maximum of $P_n$ is attained when the denominator in Eq. (2) is minimal. The corresponding value $n_m$ of the dominantly populated level by the charge exchange process becomes velocity dependent (dynamic resonance condition).

While $n_m$ is determined by the energy resonance condition only, the
population of neighboring energy levels (the n distribution) is significantly affected by the dynamic mechanism of the process operating in the considered energy range.

In the case of low-energy collisions of many-electron atoms with incompletely stripped ions, the resonance (or quasiresonance) condition is closely associated with the strong coupling of one or more pairs of molecular electronic states at certain internuclear distances. In this case the specificity of the electronic structure plays a decisive role in the collision dynamics and no general conclusions can be ruled out about the final state distributions of captured electrons.

EXPERIMENTAL SITUATIONS AND PROBLEMS

Two experimental techniques have so far been employed for the analysis of electron capture into specific final states: photon emission spectroscopy and kinetic energy (or translational) spectroscopy. A detailed discussion of these techniques can be found elsewhere. 4,23

Photon emission spectroscopy is based on detection of photons emitted after electron capture into excited projectile states by absolutely calibrated monochromators. The spectrum of the emitted radiation may cover a broad range of wavelengths from the visible to the x-ray region. The spectral range is especially large when the ionic charge and the collision energy are high. This circumstance imposes serious technical problems in the experiment. The determination of the absolute cross sections for capture into particular final states from the corresponding emission cross sections requires, among other things, correction for the cascade contribution to the population of the considered level. In its turn, the account of cascade transitions presupposes knowledge of the population of all upper levels (with respect to the one considered), which makes the problem "self-coupled." The problem of cascading can be resolved if reliable theoretical calculations for the level population cross sections exist. Apart from cascading, the derivation of the electron capture cross section from the emission cross section is obscured by the lifetime effects 24 and the fact that other nonradiative processes may contribute to the level decay. It is also obvious that the optical method cannot be used for measuring electron capture into a metastable or ground ionic state.

Kinetic energy spectroscopy is based on the equality of the change of kinetic energy of colliding partners with the change of potential energy of the bound electrons after the collision. 25 Since in the charge transfer process the projectile ions are scattered predominantly into the forward direction (and thereby the change of kinetic energy in the collision is little affected by the recoil-nucleus energy), the postcollision projectile energy spectra are directly related to the energy defects of the electron capture reaction channels. The determination of the partial capture cross sections is possible by comparison with the corresponding total capture cross section. 25 For capture into fully stripped ions this method cannot provide information about the distribution of captured electrons over the angular momentum quantum numbers. It is evident that the selectivity of the translational spectroscopy method is determined by the energy spread of the projectile ion beam. This method requires coincident measurements of at least three kinematic variables of the collision process. 4

Most of the experimental results have so far been obtained by using the optical method. This method has been extensively used by the FOM (Amsterdam) group to determine state-selective electron capture in collisions of Ne\(^{q+}\) \((q = 1–4)\) and Ar\(^{q+}\) \((q = 1–6)\) ions with inert gas atoms and H\(_2\), in the energy region 20–1200 keV and the spectral range 200–5000 Å. 16,17,26–28 Using the same method, measurements of capture into excited states have been performed for Li in collisions with He\(^{2+}\) 29,30 and Ne\(^{q+}\) \((q = 1,2)\), 31 and Xe with Ne\(^{2+}\). 32 A modification of the method recently proposed by Matsumoto et al. 33 has been applied for determination of state-selective capture cross sections in Na + Ar\(^{2+}\) 33 and Xe + Ne\(^{2+}\) 34 collisions. An example of the data provided by this method is presented in Figure 1 for the Li + He\(^{2+}\) → Li + He\(^{+}\) \((nl)\) reaction 30 and compared with theoretical predictions. 35 Capture into excited states in H\(_2\) + C\(^{6+}\), O\(^{8+}\) collisions in the energy range 0.6–8 keV/amu has been measured by Afrosimov et al. 36 and in H\(_2\) + Au\(^{8+}\) \((q = 12–18)\) collisions at an energy of 100 keV/amu by Hvelplund et al. 37

The kinetic energy spectroscopy method for electron capture into excited states has so far been applied by the group of Afrosimov for He + He\(^{2+}\) 38 and He + Ar\(^{5+}\) 39 collisions in the low energy range. The results for the He + Ar\(^{6+}\) → He\(^{+}\) + Ar\(^{5+}\) \((nl)\) reaction are shown in Figure 2 (from Ref. 38). Cross section measurements of state-selective electron capture by the translational spectroscopy method are currently underway in several laboratories. 40-42

Despite the significant experimental activity on state-selective ele-
tron capture in atom–multicharged ion collisions, the results obtained thus far are still fairly fragmentary and cannot provide a consistent picture of the process. In most of the experiments, either the energy region or the spectral range investigated have been rather too restricted to allow detailed information about the final state distribution of captured electrons and its energy dependence. For the $E = 100$ keV/amu collisions of Au$^+(q = 12, 14, 18)$ ion on H$_2$ only a small part of the $n$ distributions (their high $n$-value part) has been reported recently. Angular momentum distributions of captured electrons have also been provided for the 200 keV collisions of Ar$^{6+}$ ions with inert gas atoms (for capture into $n = 4$ and 5 principal shells of Ar$^{6+}$). These distri-

![Graph](image1)

**FIGURE 1** Cross sections for capture into excited states in the Li + He$^{2+}$ → Li$^+$ + He$^+$ ($n$) reaction. Symbols connected by solid lines are the experimental data, and dashed curves are the theoretical predictions.

![Graph](image2)

**FIGURE 2** Experimental cross section data for capture into excited states in the He + Ar$^{5+}$ → He$^+$ + Ar$^{5+}$ (nl) reaction.

butions, however, cannot be put even in qualitative agreement with the results of the existing theories developed for one-electron systems (see below). On the other hand, theoretical predictions for many-electron systems do not exist.

Experimental efforts are now oriented towards measuring state-selective capture in one-electron or quasi-one-electron systems which will make possible a direct comparison between theory and experiment.

THEORETICAL DESCRIPTIONS OF THE STATE-SELECTIVE ELECTRON CAPTURE

The process of electron capture into specific final states reflects the collision dynamics in a direct manner and therefore can provide a sensitive test for the theoretical models. In relation to the underlying physical mechanisms, a detailed analysis of the theoretical methods for treating the state-selective electron capture has recently been done. In this section we shall give a summary of that analysis and emphasize the problems which still have to be resolved in the theory.

Low-Energy Region ($v \ll 1$)

Three groups of theoretical methods have been developed for treating the charge transfer problem in atom–MCI collisions at low energies.
decay models (DM), multichannel Landau–Zener models (M-LZ), and different versions of the close-coupling (CC) method.\textsuperscript{46}

Due to the very concept of the decay models (i.e., the assumption of a quasicontinuum of final ionic states), they are not able to provide information about the final state \((n, l, m)\) distributions of captured electrons. Nevertheless, the energy resonance condition and the fact that the charge transfer process dominantly takes place at an internuclear distance \(R_0 = 2n_q^2(2q)^{1/2}\) lead to the following value of the principal quantum number of the preferentially populated final state energy level:\textsuperscript{47-49}

\[
n_m^{\text{DM}} = n_{\text{off}} \left[ 1 + \frac{q - 1}{\sqrt{2q}} \right]^{1/2} \frac{1}{\sqrt{q}} \approx 2^{1/4} n_q^{1/4}.
\]  

(4)

For the hydrogen atom–fully stripped ion system \((n_0 = 1)\), the unitarized distorted wave approximation (UDWA)\textsuperscript{50} gives the following expression for \(n_m\) (in the region \(n < 2\)):

\[
n_m^{\text{UDWA}} = q^{0.758}.
\]  

(5)

Up to \(q = 50\), expressions (4) (with \(n_0 = 1\) and without taking the limit \(q^{1/2} \gg 1\)) and (5) coincide.

The multichannel Landau–Zener model for the atom–MCI charge exchange problem\textsuperscript{51} has recently been extensively used for cross section calculation of the state-selective electron capture in hydrogen atom–fully stripped ion systems.\textsuperscript{52-54} Rotational coupling between the Stark states in the ionic channels has been taken into account (M-LZ-RC) and a large number of channels have been included in the calculations. Partial cross sections \(\sigma_n\) for \(n\)-selective capture have been calculated at collision energies of 0.5, 1, 10 and 25 keV/amu for all \(n\) giving considerable contribution to the total cross section and for ionic charges \(5 < q < 36.\textsuperscript{55}\) Energy dependent cross sections \(\sigma_n\) (all important \(n\), \(E = 3 \times 10^2 - 80\) keV/amu) have been calculated for \(q = 6, 8, 10, 13, 22, 24, 26, 28, 42,\textsuperscript{53}\) 13,\textsuperscript{52} and 18,36,54.\textsuperscript{54} Some illustrative examples of these calculations are shown in Figures 3 and 4.

The cross section \(\sigma_{nl}\) for electron capture into a specific \(n, l\) substate is connected with \(\sigma_n\) by the relation

\[
\sigma_{nl} = W_{nl} \sigma_n.
\]  

(6)

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure3.png}
\caption{\(n\) distributions in the \(E^+ + q \rightarrow H^+ + (q - e)\) reaction for \(E = 10\) keV/amu, obtained from the M-LZ-RC calculations.\textsuperscript{55}}
\end{figure}

Within the M-LZ-RC model explicit expressions for the relative populations \(W_{nl}\) can be derived in the weak—and strong—mixing regimes of Stark states.\textsuperscript{21,52} In the first case \(W_{nl}\) has the form

\[
W_{nl} = (2l + 1) \frac{(n - 1)!^2}{(n + l)! (n - 1 - l)!}.
\]  

(7)
whereas in the strong-mixing regime, \( W_{nl} \) is equivalent to the statistical distribution:

\[
W_{nl} = \frac{1}{n^2} (2l + 1).
\]

(8)

The mixing rate of Stark states depends on the angular velocity of the rotation of internuclear axis, \( \Omega_n \sim v/R_n \) (where \( R_n \) is the crossing point for the \( n \)th final energy level). Thus, for a given \( q \) the form of the \( l \) distribution of captured electrons depends on both the collision velocity and the level considered (via \( R_n \) in \( \Omega_n \)). Generally speaking, for energies below 1 keV/amu, \( W_{nl} \) is given by Eq. (7), provided \( n \) is not too high. For \( n < 8 \) the \( l \) distribution then has a maximum at \( l = 1 \) which shifts toward higher \( l \) with increasing \( n \). For energies above 10 keV/amu, according to the M-LZ-RC model, a statistical (or close to \( n^2 \)) distribution of captured electrons is expected, with \( l = n - 1 \) as the maximum populated level. However, with increasing \( n \) the mixing parameter \( \Omega_n \sim v/R_n \sim n^{2.4} \) \((e > 0)\) decreases and \( W_{nl} \) reaches a maximum at \( l = l_m < (n - 1) \).

Regarding the \( m \) distribution of captured electrons, the DM and M-LZ-RC models predict that for a given \( l \) the \( m = 0 \) state will be predominantly populated. Arguments for this statement are given elsewhere.\(^{45}\)

In the low-energy region, the collision dynamics in the charge transfer process are best described by the molecular-orbital close-coupling (MOCC) method. In order to get an accurate prediction of the \( n \) and \( l \) distributions of captured electrons, the number of states included in the basis should be very large. With increasing ionic charge \( q \) and collision energy the number of significantly populated principal shells also increases, each involving a large number of angular and magnetic substates. The MO-CC calculations, recently carried out for the \( H + C^5^+ \) system,\(^{52}\) have shown that a 33-state MO basis is necessary in order to get a 10% accuracy of the results. Some results on the \( n \) and \( l \) distributions obtained for this system are shown in Figure 5. In all MO-CC calculations in the velocity region \( 0.5 < v < 1 \), special attention should be paid to the problem of an adequate description of the electron momentum transfer effects (the problem of translational factors).\(^{46, 55}\) If this problem is not treated adequately then only the total capture cross section can be calculated, but not the partial ones.\(^{56}\)

As seen from Figures 3 and 5a, the large basis MO-CC calculations confirm qualitatively the conclusions of the M-LZ-RC model about the \( n \) distributions of captured electrons. However, the MO-CC \( l \) distributions in Figure 5b (points connected by full lines) confirm only partly the results of the M-LZ-RC model (points connected by dashed lines). Thus, at present no clear theoretical predictions can be given regarding the general form of the \( l \) distributions of captured electrons in the low energy region.
selective electron capture (especially the $n$ and $l$ distributions) the size of the basis should be rather large. Thus, for the state-selective capture reactions $H + He^{2+} \rightarrow H^+ + He^+$ ($n,l,m$), where $n$ is taken equal to 1 and 2, and $H + Li^{3+} \rightarrow H^+ + Li^{2+}$ ($n,l,m$) with $n = 1$, 2 and 3, eight$^{37}$ and 20$^{58}$ states have been included in the basis. With increase of the ionic charge $q$ the size of the basis increases tremendously, since a large number of levels with high $n$ participate in the collision dynamics. Very extensive 35-state AO-CC calculations have recently been performed for the $H + C^6+$ system by Fritsch and Lin.$^{59}$ The limited information about the state-selective electron capture gained so far from the AO-CC calculations does not allow any general conclusions about the $n$ and $l$ distributions of captured electrons. In the case of $H + Li^{3+}$ collisions, at energies of 25 and 100 keV/amu and $n = 2$ and 3, it has been found that the sublevel $l = 1$ is predominantly populated.

The system of AO-CC equations can be simplified if only one target AO is retained in the basis, together with a complete set of orbitals representing the final (ionic) states. If, further, only the coupling between the initial and final states is retained, the approximated system of coupled equations can be treated either within a multichannel Vainshtein–Presnyakov–Sobel’man (M-VPS) approximation$^{60}$ or within a unitarized distorted wave approximation (UDWA).$^{50}$ While the M-VPS method has not yet been employed for state-selective electron capture cross section calculations, the UDWA has recently been used$^{61}$ to produce systematic partial cross section data, $\sigma_n$ and $\sigma_{nl}$, for the hydrogen atom–fully stripped ion systems with ionic charges $q = 1−6,8,10,14$ and at selected collision energies between $E = 10$ keV/amu and $E = 2−5$ MeV/amu (see also Ref. 20). It should be mentioned that for energies above $\sim 500$ keV/amu, UDWA may overestimate the total capture cross section by a factor of 1.5–2. The second order terms, omitted in UDWA, may introduce significant errors in the computed $\sigma_n$ and $\sigma_{nl}$ even at much lower energies.

In predicting total electron capture cross sections in the intermediate energy region (in particular for $E = 25−200$ keV/amu), the classical trajectory Monte Carlo (CTMC) method$^{62}$ has had considerable success. This method has also been used for systematic investigation of the state-selective electron capture in hydrogen atom–fully stripped ion systems,$^{19,63}$ with $q$ ranging between 6 and 20 and for collision energies $E = 25,50$ and 100 keV/amu. Illustrative examples about the $n$ and $l$ distributions of captured electrons, provided by the CTMC method, are given in Figures 6 and 7 (data taken from Ref. 63) and compared with

FIGURE 5 The partial cross section $\sigma_n$ (part a) and the $l$ distributions $W_{nl} = \sigma_{nl}/\sigma_n$ (part b) of captured electrons in the $C^6^+ + H$ collisions obtained by 33-state MO-CC calculations.$^{57}$ The points in part b connected with dashed lines represent the $l$ distributions from Eq. (7) ($E = 1.32$ keV/amu) and Eq. (8) ($E = 21$ keV/amu).

Intermediate Energy Region ($\nu \sim 1$)

An atomic orbital expansion close-coupling (AO-CC) method, which also takes into account translational effects, can provide an adequate description of the charge transfer dynamics at intermediate collision energies. Like in the MO-CC method, in order to describe the state-
the UDWA results. In both approximations, the $n$ distributions are similar, although with increasing $q$ the disagreement becomes significant. The maxima of CTMC $n$ distributions are given by $n_m = q^{3/2}$. The CTMC $l$ distributions show the following features. For $n < n_m$, the states with the highest value of $l$ are predominantly populated. For $n > n_m$, the CTMC $l$ distributions exhibit maxima at $l = l_m = q - 1$. More specifically, for $n > q - 2$, $l_m = q - 2$. The UDWA $l$ distributions have analogous behavior, but for somewhat higher energies. Although an analysis of the approximations involved in UDWA has brought some doubts about the reliability of the UDWA $l$ distributions, this question still needs more investigation.

Regarding the $m$ distributions of captured electrons, UDWA predicts
that for $E > 25$ keV/amu the state $m = 0$ is predominantly populated, with a rapid decrease of $\sigma_{nlm}$ with increasing $m$. For $E < 10$ keV/amu, no generalizations about the $m$ distributions can be made from the UDWA results.

High Energy Region ($v \gg 1$)

We shall start our discussion of the final state distributions of captured electrons in the high energy region with the predictions of the Brinkman–Kramers (BK) approximation. Many of the BK results regarding these distributions remain valid also in the higher-order theories, at least for capture from the ground target state. In connection with the questions discussed here, one should stress that the applicability of the BK approximation presumes fulfillment of the condition $v \gg q/n$, which for highly charged ions appears to be rather restrictive. For a one-electron system, explicit expressions for the cross sections $\sigma_{nlm}^{BK}$, $\sigma_{nl}^{BK}$ and $\sigma_{nlm}^{BK}$ can be derived. From the expression for $\sigma_{nl}^{BK}$ one obtains that the $n$ distribution has a maximum at

$$n_{m}^{BK} = q 17^{1/2} \left[ 2(25v^3 + v^2v_0^2) + 25v_0^4 \right]^{1/2} - 7(v^2 - v_0^2) \right]^{1/2}, \quad (9)$$

where $v_0 = (q_0/n_0)$ is the initial state electron velocity.

For $v \ll v_0$ (formally), $n_{m}^{BK} = \frac{q}{v_0} = (q/q_0) n_0$, i.e., the dominantly populated level is the isoeenergetic one (resonance). For $v \to \infty$, $n_{m}^{BK} \to (17/3)^{1/2} q/v$. This means that independently of the initial state parameters, the electron capture maximizes at the $n = 1$ final state. Thus, the $n$ distribution in the BK approximation is peaked at $n_{m}^{BK}$, and with increasing velocity its maximum shifts toward smaller $n$.

Numerous $\sigma_{nl}$ calculations have shown that for a given $n$ such that $n > q/v$, $W_{nl}^{0} = \sigma_{nl}^{BK}/n_{m}^{BK}$ is maximum at $l = n - 1$. If the condition $q/nv > 1$ is satisfied, then $W_{nl}^{0}$ has a maximum for $l = l_{m} < n - 1$, and in the limit $q/nv \gg 1$, the BK approximation predicts that the $l = 0$ substate is predominantly populated for any $n$. Another famous result of the BK approximation is Oppenheimer's $n^3$ law:

$$\frac{\sigma_{nl}}{\sigma_{n}^{0}} = \left( \frac{n_l}{n} \right)^3, \quad n > n_l, \quad (10)$$

The meaning of Eq. (10) is that starting from a sufficiently high $n$, the relative $l$ distribution of captured electrons remains unchanged with further increase of $n$. Recent numerical investigations of Dubé and Briggs have confirmed this conclusion and suggested that the condition for applying relation (10) is $n, v/q > 6$ (within a 10% accuracy).

The question arises how the higher-order theories for the charge transfer process modify the first-order BK results. The simplest of them is the eikonal-BK approximation of Chan and Eichler. This approximation allows derivation of explicit expressions for the partial cross sections $\sigma_{m}, \sigma_{nl}$ and $\sigma_{nlm}$ for one-electron systems. The numerical calculations for $H + C^6+$ and $H + O^8+$ systems have shown that apart from a reduction of the absolute cross section values, the form of the $n$ and $l$ distributions remains essentially the same as in the BK approximation.

The state-selective electron capture has recently been investigated in the second Born approximation as well as in the continuum distorted wave (CDW) approximation. The investigations of the reaction $H(1s) + C^6+ \to H^+ + C^5^+ (nl)$ in an approximate second Born approximation have led to the following conclusions. (1) The collision velocity $v_c$, at which the second Born term begins to dominate the first one strongly, depends on the final state angular quantum number $l$ (as well as on $m$). For the investigated final states ($1 < n < 3$), the following typical values for $v_c$ have been found: $v_c \sim 80$, for $l = 0$; $v_c \sim 30-20$ for $l = 1$ ($m = 0, 1$) and $v_c \sim 15-5$ for $l = 2$ ($m = 0-2$). (2) The relative contribution of the second Born term in the partial cross section for different final principal shells increases with the velocity, being always mostly pronounced for the group of levels around $n_{m}^{BK}$(iv). Thus, for $v \sim 1-2$, this contribution amounts to $\sim 10\%$ for the levels $n = 6-7$, whereas for $v \sim 15-20$ it amounts to $\sim 50\%$ and is concentrated on the $n = 1-2$ levels. (3) The relative contribution of the second Born term in $\sigma_{nl}$ increases rapidly with increasing $l$ (and for $n > n_l$ is independent of $n$).

Some results on the $n$ and $l$ distributions in the approximate second Born approximation (approximate because the second Born amplitude is calculated approximately) are shown in Figures 8 and 9. They are compared with the corresponding results of the CDW approximation. Only the relative populations $P_n = \sigma_{nl}/\sigma_{n}^{0}$ and $W_{nl} = \sigma_{nl}/\sigma_{n}$ are shown. While the $n$ distributions (Figure 8) exhibit similar form
(close to the one in the BK approximation), the \( l \) distributions are considerably different. The disagreement between \( P_n^{B2} \) and \( P_n^{CDW} \) increases with increasing projectile charge \( q \) (which is equivalent to decreasing the collision velocity). The reason might lie in the approximations made in the calculation of \( \sigma_n^{B2} \). The same argument may apply also for the disagreement between \( W_n^{B2} \) and \( W_n^{CDW} \).

In Ref. 72, CDW calculations of \( W_n \) have been carried out also for the \( n = 5 \) shell at \( v = 1.04 \), showing a maximum at \( l = n - 1 \). These results are in close agreement with the 33-state MO-CC calculations of Green et al.\(^{55} \) The preferential population of the \( l = n - 1 \) state in the velocity range \( v < 10 \) and for final state principal shells \( n = 1, 2 \) and 3 has also been found in the CDW calculations of the state-selective capture in the \( \text{H} + \text{F}^+ \) system.\(^{73} \) Generalizing these results, one can infer that for \( n \) shells satisfying the inequality \( nv/q < 1 \), the \( l \) distribution is peaked at \( l = n - 1 \). When \( nv/q > 1 \), the \( l \) distribution gets a maximum at some \( l_m < n - 1 \), which gradually shifts towards \( l_m = 0 \) when the value of the parameter \( nv/q \) increases. This conjecture, following also from the BK approximation, needs more rigorous justification.

Two Comparisons of Theoretical and Experimental Results
As discussed in the preceding subsections, almost all of the theoretical cross section calculations of state-selective electron capture have been done for one-electron systems. No experimental data on \( \sigma_n \) or \( \sigma_m \) for these systems exist. Only in one case, namely for the reaction \( \text{Li} + \text{He}^{2+} \rightarrow \text{Li}^+ + \text{He}^+ \) (\( n,l \)) the experimental data\(^{30,74} \) can be directly compared with the theoretical predictions.\(^{35} \) This is done in Figure 1 for the partial cross sections \( \sigma_n \). The agreement can be considered fairly satisfactory (except for the \( n = 2 \) state), keeping in mind that the calculations have been made in a two-state AO-CC approximation. This approximation is evidently inadequate to describe the process at higher
Partial cross sections $\sigma_{3p}$, $\sigma_{4p}$ and the summed cross sections $\sigma_{3s} + \sigma_{3p}$ and $\sigma_{4s} + \sigma_{4p}$ have also been measured for this system\textsuperscript{74} and the first two of them compare favorably with the two-state AO-CC calculations of Bransden and Ermolaev.\textsuperscript{35}

The upper part of the $n$ distributions for the state-selective capture $\text{Au}^{\ast +} + \text{H}_2 \rightarrow \text{Au}^{(q - 1)\ast +} \left( n \right) + \text{H}_2^\ast \left( q = 12, 14, 18 \right)$ has recently been measured\textsuperscript{43} for an energy of 100 keV/amu. For this energy and the range of ion charges involved, the ratio of the total cross sections for capture from $\text{H}_2$ and $\text{H}$ is close to one. On the other hand, since in this experiment capture to highly excited levels ($9 < n < 14$) was measured, the one-electron approximation for the final electronic state can be adopted as reasonable. On the basis of such considerations, Hvelplund et al.\textsuperscript{43} compared their data with the theoretical results for the system hydrogen atom–fully stripped ion (with corresponding ionic charge). The comparison for $q = 12$ and $q = 14$ is shown in Figure 10. The sources of the evident disagreement may be several: the projectile-product ion $\text{Au}^{(q - 1)\ast +}$ has a core with some effective charge $q' \neq q$; a Franck–Condon factor for the $\text{H}_2\,(O) \rightarrow \text{H}_2^\ast \left( \nu' \right)$ transition might have to be introduced in the theoretical cross sections before they are compared with the experimental ones; the cascading correction, based on the CTMC theoretical data,\textsuperscript{61} may also introduce significant errors in the experimental data, etc. However, perhaps even more striking is the disagreement between the different theoretical results, shown in Figure 10.

CONCLUDING COMMENTS

The reviewed (albeit briefly) present-day experimental and theoretical situation in the problem of state-selective electron capture in atom–MCI collisions has revealed that our understanding of the general features of this process is still incomplete. Even in the best (theoretically) studied hydrogen atom–fully stripped ion system, no conclusive statements can be made about the details of the $l$ distributions of captured electrons, in particular regarding their velocity dependence. Large basis close-coupling calculations would be able to produce accurate data for the $n$ and $l$ distributions in any particular collision system, but the size of the necessary basis (especially for high $q$) would be intolerably large. Significant refinements of the existing models for the charge exchange process are needed in order to achieve a better description of the state-selective electron capture. Finally, there is substantial lack of theoretical efforts for describing the capture into excited states in many-electron collision systems, to which all of the existing experimental data are pertinent.

One of the ways to achieve a convergence of the theoretical and
experimental efforts is to study the alkali atom–fully (or highly) stripped ion systems, since the one-electron approximation for them may be considered acceptable (at least at low collision energies). However, from the point of view of having a sensitive test for the theoretical models, one needs experimental cross section data for state-selective capture in the proper one-electron collision systems.

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