Overbarrier model with electron backcapture

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We present an extension of the classical overbarrier model [F. Sattin, Phys. Rev. A **62**, 042711 (2000)] to include the effect of electron back-capture. Back-capture is the process by which an electron that has already been captured by the projectile ion is re-captured by the target atom. Back-capture reduces the electron capture cross section at low impact velocities when the projectile ionization energy is less than that of the target. This creates a cross section peak. We alter the location of this peak to correspond to that predicted by an adiabatic criterion by using a free parameter of the model. These extensions bring the overbarrier model more in line with experimental data, especially at low impact velocity.

Keywords:

I. INTRODUCTION

The classical overbarrier model is a simple analytic model that describes the charge exchange between a target atom or ion and a projectile ion [1–6]. In the literature, the model and all of its variations predict that the electron capture cross section is a monotonically decreasing function of velocity [5–10]. In fact, the analytic formula shows that the model predicts close to a $1/v$ velocity dependence when ionization is neglected. With ionization, the falloff with velocity is stronger.

Experiments, however, reveal that the cross section as a function of velocity contains a peak at non-zero velocity for most reactions. Massey attributed the low velocity fall-off to the fact that the collision proceeds slowly enough (adiabatically) that the system is able to adjust itself to perturbations without a transition taking place, thus making the cross section small [11]. The cross section should be low when the system obeys the adiabatic criterion:

$$\frac{a|\Delta E|}{hv} \gg 1 \tag{1}$$

where a is the adiabatic parameter which should be on the order of atomic dimensions, ΔE is the difference in the target and projectile binding energies and is called the energy defect, and h is Planck's constant.

The overbarrier model fails to correctly predict the velocity dependence below this Massey peak because it doesn't take into account the projectile's binding energy, and therefore the energy defect. The projectile's binding energy can be included in a natural way in the overbarrier model by including electron back-capture, the process in which a target electron is captured back by the target atom/ion after originally being captured by the projectile. In the context of the overbarrier model, the back-capture process is similar to the capture process, but with the roles of the projectile and target reversed. Thus, the probability for back-capture is inversely proportional to the binding energy of the projectile ion.

At low collision velocity, where there is sufficient time for many captures and back-captures, the charge exchange cross section is controlled by the energy defect. Essentially, the electron will preferentially end up bound to the object (target or projectile) with the greatest binding energy. So, when the binding energy of the projectile is lower than that of the target, it is reasonable to expect a decrease in the electron capture cross section at low velocity, giving the Massey peak. This is generally true in experiment [12, 13], and we show it is also true in the overbarrier model with back-capture included.

II. REVIEW OF THE OVERBARRIER MODEL

The overbarrier model has several versions which vary in the details. We use the version that can be found in [5, 6, 14]. These references provide particularly nice derivations of the model. To review, consider an electron bound in the potential well of a target where the core of the target has effective charge Z_t. As a projectile ion with charge Z_p approaches to an internuclear distance R, its potential well overlaps with that of the target creating a saddle-shaped potential field. In the plane of the three particles, we use the cylindrical coordinate system of [6] in which the internuclear axis is labeled with coordinate z and the electron's distance from this axis is labeled with coordinate ρ. Then, the total (kinetic plus potential) energy of the electron (in atomic units) is

$$E = \frac{p^2}{2} - \frac{Z_t}{\sqrt{\rho^2 + z^2}} - \frac{Z_p}{\sqrt{\rho^2 + (R-z)^2}}. \tag{2}$$

When the electron is bound to the target, and in the limit $R \to \infty$, the energy of the electron is approximated as

$$E(R) = -E_t - \frac{Z_p}{R} \tag{3}$$

where E_t is the binding energy of the target electron in the absence of the projectile.

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The saddle-shaped potential is a maximum along the internuclear axis at the distance

$$z_0 = \frac{\sqrt{Z_t}}{\sqrt{Z_t}+\sqrt{Z_p}}R \tag{4}$$

The electron is able to reach the saddle point if its maximum excursion is equal to z_0. This occurs when the internuclear spacing R is less than some maximum

$$R_m = \frac{\alpha\sqrt{Z_pZ_t}+Z_t}{E_t}. \tag{5}$$

This expression is derived from Eq. 2 with $z = z_0$ and $\rho = 0$. When E in Eq. 2 is set to $E(R)$ in Eq. 3, $\alpha = 2$; however, if $E = -E_t$, $\alpha = 1$. Sattin showed that $\alpha = 1$ gave better agreement with certain experiments [6, 8]. However, $\alpha = 2$ is consistent with the rest of the model, which always uses the fully perturbed binding energy ($E(R)$ in Eq. 3), so we use $\alpha = 2$ in all calculations.

When $R < R_m$, the electron can cross over the potential barrier at time t and be captured by the projectile if its orbit intersects the potential opening and if the electron is in the part of the orbit that crosses the barrier. The fraction of electron orbits that intersect the opening at time t is given by

$$N_\Omega = \frac{\sqrt{Z_p}}{2\sqrt{Z_t}(\sqrt{Z_t}+\sqrt{Z_p})^2}\left[(\sqrt{Z_t}+\sqrt{Z_p})^2 - Z_p - E_tR\right]. \tag{6}$$

The fraction of electrons that cross any surface perpendicular to their motion within the time interval dt is dt/T, where T is the period of the electron orbit:

$$\begin{aligned} T &= 2\int_0^{r_{turn}} \frac{dr}{p} = \sqrt{2}\int_0^{r_{turn}} \frac{dr}{\sqrt{\frac{Z_t}{r}+\frac{Z_p}{|\mathbf{R}-\mathbf{r}|}-E_t-\frac{Z_p}{R}}} \\ &\approx \sqrt{2}\int_0^{1/E_t} \frac{dr}{\sqrt{\frac{Z_t}{r}-E_t}} = \frac{\pi}{\sqrt{2}}Z_tE_t^{-3/2} \end{aligned} \tag{7}$$

where $r = \sqrt{\rho^2+z^2}$ is the distance of the electron from the target and r_{turn} is the maximum radial excursion of the electron, which occurs when $p = 0$. Note that p is obtained from equating Eqs. 2 and 3. The approximation on the second line of Eq. 7 is then obtained by applying the large R limit (to zeroth order in r^2/R^2), which is consistent with Eq. 3. This is also equivalent to using the momentum without considering the perturbation as was done in [6]. We note that in [8], Sattin mistakenly left out the $Z_p/|\mathbf{R}-\mathbf{r}|$ term in the momentum [15].

Next, let $W(t)$ represent the probability for the electron to still be bound to the target at time t. Its time rate of change is then given by

$$\frac{dW(t)}{dt} = -N_\Omega\frac{f_T}{T}W(t). \tag{8}$$

Note that like Sattin [6], we multiply the rate equation by a constant corrective factor f_T. Sattin justifies this as a correction to the period, which should be less than is calculated in Eq. 7 due to finite angular momentum, which we have neglected. Other justifications for this correction include our neglect of quantum tunneling and partial electron screening in multi-electron atoms and ions. We prescribe a method for setting this free parameter in Section IV, so that the model can better reproduce known experimental results.

The probability for the electron to leak from the target and be captured by the projectile is

$$P_l(b) = 1 - W(\infty) = 1 - \exp\left(-\frac{f_T}{T}\int_{-t_m}^{t_m} N_\Omega\, dt\right). \tag{9}$$

Assuming a straight line collision trajectory with impact parameter b and velocity v,

$$R = \sqrt{b^2+(vt)^2}, \tag{10}$$

the limits of the integral ($\pm t_m$) are derived with $R = R_m$, meaning $-t_m < t < t_m$ is the time range in which charge exchange can occur. The integral in Eq. 9 can be performed analytically due to this known dependence of R on time, meaning the leakage probability can be calculated analytically. The total cross section

$$\sigma = 2\pi\int bP_l(b)db \tag{11}$$

involves an integral which is not normally analytically tractable, so numerical integration is required.

One final consideration of the model is that of ionization. During the collision process, it is possible that the electron is ionized and not captured by the projectile. There are different models within the overbarrier model that have been used to account for ionization [6, 16]. We follow Sattin's, which calculates the electron's binding energy to the projectile while in the saddle point of the potential [6]. This binding energy to the projectile is

$$E_p' = E_t + \frac{Z_p - Z_t}{R} - \frac{v^2}{2} + \frac{v_{et}v^2t}{R}, \tag{12}$$

$$\text{where} \quad \frac{v_{et}^2}{2} = \frac{(\sqrt{Z_t}+\sqrt{Z_p})^2 - Z_p}{R} - E_t. \tag{13}$$

Note that the binding energy E_p' is not generally equal to the projectile's ground state binding energy E_p. The condition for ionization is that $E_p'(t) < 0$, which one can see from Eq. 12 primarily occurs for large v and small or negative t. If $E_p'(t) > 0$ for the time interval $t_i < t < t_f$ which is within $\pm t_m$, the capture probability is

$$P_c(b) = 1 - \exp\left(-\frac{f_T}{T}\int_{-t_i}^{t_f} N_\Omega\, dt\right). \tag{14}$$

III. EXTENSION TO INCLUDE BACK-CAPTURE

Accounting for the return of the electron to the target once it has been captured by the projectile is relatively straight-forward in the case when the projectile is the same element as the target and $Z_p = Z_t$ [17]. It is trickier when the capture and back-capture are not identical processes. To do this calculation, we first define three time-dependent probabilities, w_t, w_p, and w_i, which represent the probability for the electron to be bound to the target, bound to the projectile, and ionized, respectively. They are constrained in that $w_t + w_p + w_i = 1$. We can then write evolution equations for each of these probabilities,

$$\begin{aligned} \frac{dw_t}{dt} &= -j_{tp}w_t - j_{ti}w_t + j_{pt}w_p \\ \frac{dw_p}{dt} &= -j_{pt}w_p - j_{pi}w_p + j_{tp}w_t \\ \frac{dw_i}{dt} &= j_{ti}w_t + j_{pi}w_p \end{aligned} \tag{15}$$

where j_{tp} and j_{pt} are the rates of charge exchange from the target to the projectile and from the projectile to the target, respectively, while j_{ti} and j_{pi} are the rates of ionization for an electron bound to the target and to the projectile. Given the initial conditions $w_t = 1, w_p = w_i = 0$ at $t \to -\infty$, the summation constraint ($w_t + w_p + w_i = 1$) is automatically satisfied. The charge exchange and ionization rates are

$$\begin{aligned} j_{tp} &= \frac{f_T}{T_t} N_{\Omega,t}\ \Theta(E'_p)\ \Theta(t_{m,t} - |t|) \\ j_{ti} &= \frac{f_T}{T_t} N_{\Omega,t}\ \Theta(-E'_p)\ \Theta(t_{m,t} - |t|) \\ j_{pt} &= \frac{f_T}{T_p} N_{\Omega,p}\ \Theta(E'_t)\ \Theta(t_{m,p} - |t|) \\ j_{pi} &= \frac{f_T}{T_p} N_{\Omega,p}\ \Theta(-E'_t)\ \Theta(t_{m,p} - |t|) \end{aligned} \tag{16}$$

where Θ is the Heaviside step function. $T_t, N_{\Omega,t}$, and $t_{m,t}$ are equivalent to T, N_Ω, and t_m from the previous section. The added subscript t simply denotes that they apply to the electron while it is bound to the target. The new variables $T_p, N_{\Omega,p}$, and $t_{m,p}$, denote quantities for the electron bound to the projectile, while E'_t is the counterpart of E'_p – it is the binding energy to the target while in the saddle point, and is not generally equal to E_t. To get the expressions for these new variables, simply take all of the expressions in the previous section and exchange the subscripts, $t \to p, p \to t$.

The differential equations (Eqs. 15) must be solved numerically. One point to keep in mind is that the time limits during which the equations must be solved are $-t_{m,t} < t < \max(t_{m,t}, t_{m,p})$. The back-capture may proceed even after the original capture shuts off, or it may shut off before the original capture does. This depends on the relative values of Z_t, Z_p, E_t, and E_p.

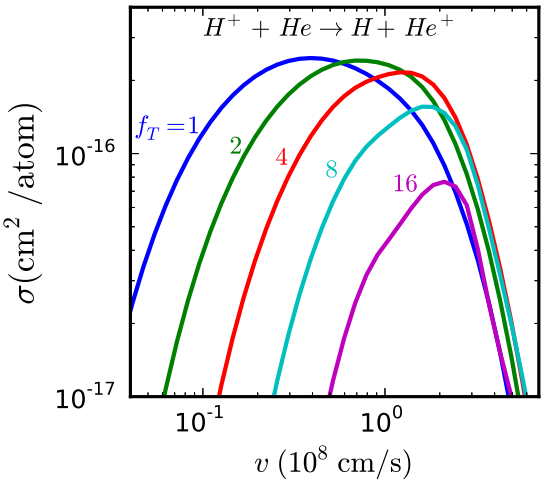

FIG. 1: Charge exchange cross sections between H and He calculated with the overbarrier model with back-capture using various values of the free parameter f_T.

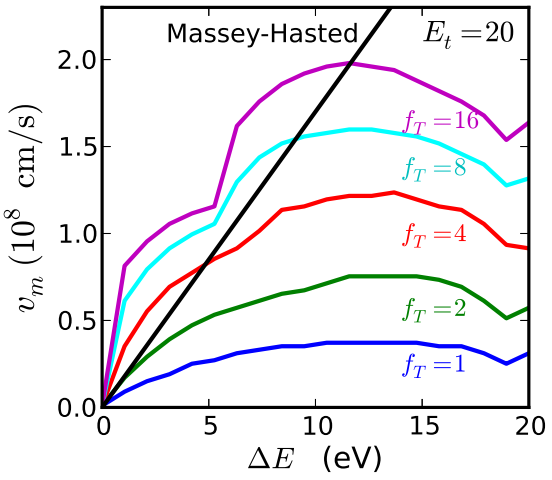

FIG. 2: The velocity v_m at which the cross section peaks as a function of the energy defect ΔE calculated from the overbarrier model with back-capture. The straight Massey-Hasted line is the prediction of v_m from the adiabatic criterion.

IV. RESULTS

Before calculating cross sections, we must choose how to set the model's free parameter, f_T. Recall, f_T multiplies the rates in Eq. 16. To set this, we appeal to past theoretical and experimental studies that have attempted to characterize the shapes of charge exchange cross sections. In particular, there have been several attempts

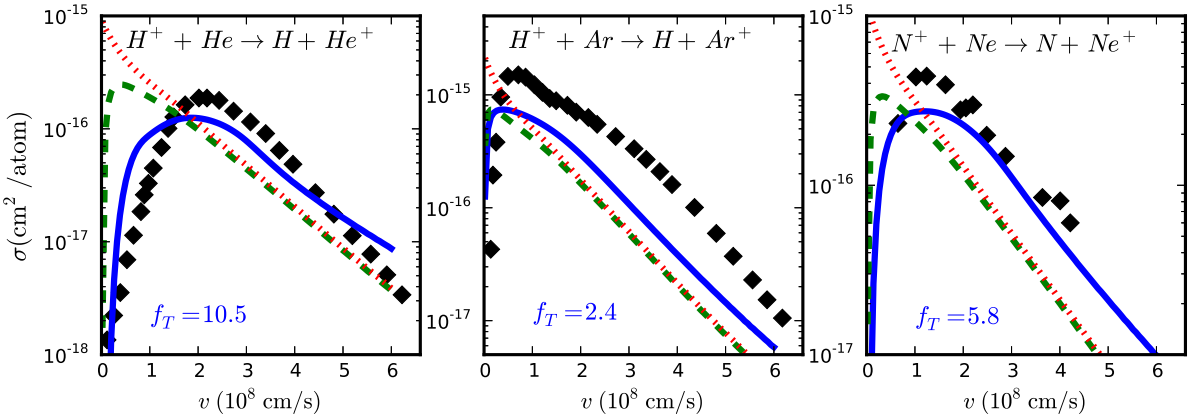

FIG. 3: Charge exchange cross sections. The black diamonds are experimental data from [12, 13]. The solid blue lines are overbarrier model with back-capture calculations that use the value of f_T indicated on the bottom of each figure, which is calculated from the procedure using the adiabatic criterion. The green dashed lines are overbarrier model with back-capture calculations using $f_T = 1$, and the short-dashed red lines are overbarrier model without back-capture calculations using $f_T = 1$.

in the literature at predicting the collision velocity v_m at which the charge exchange cross section peaks (the Massey peak) [18–22]. Hasted applied Massey's adiabatic criterion (Eq. 1), asserting that

$$v_m = \frac{a|\Delta E|}{h} \tag{17}$$

and found that $a \approx 7 \times 10^{-8}$cm by fitting Eq. 17 to experimental data [18]. The significance of this expression is the proportionality between v_m and the energy defect $\Delta E = E_t - E_p$. Drukarev later showed that $v_m \sim |\Delta E|$ only in the limit $|\Delta E| \ll E_t$. For $|\Delta E| \ll E_t, v_m \sim |\Delta E|^{1/2}$ [19]. Others used this result to derive their own relations with $v_m \sim |\Delta E|^{1/2}$ [20–22]. We choose to use the Massey-Hasted result to set the free parameter f_T, but one can use the other relations in a similar way.

To do this, we note that the overbarrier model with back-capture produces a charge exchange cross section peak as long as $E_p < E_t$ regardless of the value f_T. We show one example of this with the reaction $\mathrm{H}^+ + \mathrm{He} \rightarrow \mathrm{H} + \mathrm{He}^+$ in Fig. 1 ($E_{\mathrm{bind}} = E_t = 24.6$ for He and $E_{\mathrm{bind}} = E_p = 13.6$ for H). It is apparent that f_T controls both the magnitude of the cross section maximum, σ_m, as well as the velocity at which it occurs, v_m. Since the Massey-Hasted relation predicts v_m for a given reaction, we can set f_T so that our model predicts a cross section peak equal to this Massey-Hasted prediction.

To illustrate this further, we show the overbarrier model's functional dependence of v_m on ΔE and f_T in Fig. 2 for $E_t = 20$. Alongside the $v_m(\Delta E, f_T)$ curves, we plot the Massey-Hasted prediction for the peak velocity. The intersection of the $v_{m,OBM}(\Delta E, f_T)$ overbarrier model (OBM) curves with the Massey-Hasted curve at the particular ΔE of the collision:

$$v_{m,M-H}(\Delta E) = v_{m,OBM}(\Delta E, f_T), \tag{18}$$

provides a solution for f_T. For example, taking a collision in which $E_t = 20$ and $E_p = 15$ so that $\Delta E = 5$, the Massey-Hasted prediction is $v_m = 8.5 \times 10^7$, which looking at Fig. 2 is what the overbarrier model predicts for $f_T \approx 4$. So we can use the overbarrier model with back-capture with $f_T = 4$ to calculate the charge exchange cross section for this reaction.

With this procedure, we calculate the charge exchange cross sections for a few collision reactions for which experimental data exists [12, 13]. All of these reactions have $E_p < E_t$. We show these calculations along with the experimental data in Fig. 3. In the figure, we also include calculations using the overbarrier model with $f_T = 1$ with and without back-capture. $f_T = 1$ corresponds to the uncorrected case. It is clear that without back-capture, the overbarrier model does not contain any peak. Furthermore, it is necessary to use the f_T corrective factor to obtain the peak at the correct velocity and to obtain better overall agreement with experiment, which is generally within a factor of two or so.

Overall, while the inclusion of back-capture makes the overbarrier model more calculation intensive, it allows for the reproduction of the well-known Massey peak. This, combined with our use of the Massey-Hasted adiabatic criterion and a reaction rate multiplier, we have greatly improved the model's accuracy, especially at low velocity.

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