Cold ion-atom chemistry driven by spontaneous radiative relaxation: a case study for the formation of the $\text{YbCa}^+$ molecular ion

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Received 29 August 2013, revised 5 November 2013
Accepted for publication 25 November 2014
Published 17 December 2013

Abstract
Using both quantum and semi-classical methods, we calculate the rates for radiative association and charge transfer in cold collisions of $\text{Yb}^+$ with $\text{Ca}$. We demonstrate the fidelity of the local optical potential method in predictions for the total radiative relaxation rates. We find a large variation in the isotope dependence of the cross sections at ultra-cold gas temperatures. However, at cold temperatures, $1 \text{ mK} < T < 1 \text{ K}$, the effective spontaneous radiative rates for the different isotopes share a common value of about $1.5 \times 10^{-15} \text{ cm}^3 \text{ s}^{-1}$. It is about five orders of magnitude smaller than the chemical reaction rate measured in Rellergert et al (2011 Phys. Rev. Lett. 107 243201).

Keywords: charge transfer, ion-atom collision, cold and ultra-cold collisions, isotope effects, radiative recombination

((Some figures may appear in colour only in the online journal)

1. Introduction

Advances in the cooling, trapping, and manipulation of ultra-cold atoms have opened new vistas in our understanding of quantum degenerate matter. In recent years, laboratory techniques have advanced so that it is now feasible to cool ions and explore their interactions with neutral matter in the sub- to milli-Kelvin temperature range. The development of hybrid, ion–atom traps (Schmid et al 2010, Zipkes et al 2010, Ratschbacher et al 2012, Rellergert et al 2011, Hall et al 2011, 2013) has allowed researchers to explore competing pathways for reaction between cold atoms and ions, including non-radiative and radiative charge transfer as well as radiative association, in which ions and atoms combine to form a molecule at cold temperatures. Among possible applications for cold ion–atom chemistry are quantum-limited control of chemical reactions and buffer gas cooling of single ion clocks (Ratschbacher et al 2012). The aforementioned reactions are also important in astrophysical applications (Rellergert et al 2011, Stancil and Zygelman 1996, Zygelman et al 1998). Laboratory efforts in measuring accurate rate coefficients of the latter enhances the atomic data base employed in astrophysical models.

In a recent laboratory study (Rellergert et al 2011), Rellergert et al used a hybrid trap to investigate the interactions between cold $^{134}\text{Yb}^+$ ions with $^{40}\text{Ca}$ atoms. They observed a large, on the order of $2 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$, chemical reaction rate coefficient and, based on a preliminary theoretical estimate, suggested that radiative charge transfer was the dominant process behind this rate. However, their theoretical estimate also predicted that nearly half of the chemical reactions should produce $\text{YbCa}^+$ molecules through radiative association, which disagreed with the experimental observation that the fraction of reactions leading to molecule formation was $\leq 0.02$. They suggested this discrepancy could be due to systematic effects, but called for more investigation into the system. The measured rate was several orders of magnitude larger than is typically observed in radiative quenching calculations.
force in the entrance channel leading to a potential minimum at undulating line. The oscillations are due to the strong polarization thick lines) participating in the radiative association process. In the Figure 1. (see illustration in figure1). The efficiency of a transition at the internuclear separation in which a transition occurs that theory a photon is emitted with energy equal to the energy by Kramers and Ter-Haar (Kramers and Ter Haar 1946). In

distance. The total rate is then estimated by a, classical, time average over all localized transitions as the quasi-molecule evolves. This picture was later refined, e.g. see West et al (1982), so that the quantum nature of the entrance channel is fully taken into account. The relationships between the various semi-classical theories and those obtained using the FGR methods (Sando 1971) was explored in Zygelman and Dalgarno (1988).

In our discussion below we briefly summarize the various theoretical approaches and use them to calculate the rates for processes (1,2). We provide a rigorous upper bound for the sum of rates for processes (1,2). Atomic units are used throughout, unless otherwise indicated.

2. Theory

2.1. Radiative association

The cross section for the radiative association process

\[ \text{Yb}^+ + \text{Ca} \rightarrow \text{YbCa}^+ + h\omega \]

where \( h\omega \) is the energy of the emitted photon is given by the expression (Zygelman et al 1998)

\[
\sigma_{RA} = \sum_j \frac{8 \pi^3 \omega_{\alpha J}^2}{3c^2k^2} [J(J+1)M_{J+1}^2(k, n) + JM_{J-1}^2(k, n)]
\]

\[
M_{J,k}(n, k) = \int_0^{\infty} dR f_j(kR) D(R) \phi_{f\alpha}(R) \tag{3}
\]

where \( D(R) \) is the transition dipole moment between the \( \Sigma^+ \) states of the \( \text{YbCa}^+ \) molecular ion. \( \phi_\alpha \) is a rotational-vibrational eigenstate of the \( \Sigma^+ \) ground state, with binding energy \( \epsilon_\alpha \), and is characterized by the angular momentum and vibrational quantum numbers \( J, n \) respectively.

\[
f_j(kR) \text{ is the wavefunction that satisfies the radial Schrödinger equation}
\]

\[
f''_j(kR) - \frac{J(J+1)}{R^2} f_j(kR) - 2\mu V_A(kR) f_j(kR)
\]

\[
+ k^2 f_j(kR) = 0
\]

where \( V_A(kR) \) is the BO energy of the excited \( \Sigma^+ \) state, \( \mu \) is the reduced mass of the collision system and \( k \) is the wavenumber for the incident collision partners in that channel. It has the asymptotic form

\[
f_j(kR) \rightarrow \sqrt{\frac{2\mu}{\pi k}} \sin \left( kR - \frac{J\pi}{2} + \delta_j \right),
\]

where \( \delta_j \) is a phaseshift, as \( R \rightarrow \infty \). The energy of the emitted photon is given by

\[
h\omega_{fJ} = \frac{k^2}{2\mu} + V_A(\infty) + |\epsilon_{fJ}| - V_X(\infty). \tag{6}
\]

2.2. Radiative charge transfer

The cross section for the radiative charge transfer process,

\[ \text{Yb}^+ + \text{Ca} \rightarrow \text{Yb} + \text{Ca}^+ + h\omega \]

is given by Zygelman et al (1989)

\[
\sigma_{CT} = \int_0^{\omega_{\alpha J}} d\omega \frac{d\sigma}{d\omega},
\]

\[
\frac{d\sigma}{d\omega} = \sum_J \frac{8 \pi^2 \omega^3}{3 c^2k^2} [JM_{J-1}^2(k, n') + (J+1)M_{J+1}^2(k, n')]
\]

\[ \tag{7} \]
where
\[ M_{J,J'}(k, k') = \int_0^\infty dR f_J(kR) D(R) f_{J'}(k'R). \] (8)

Here \( f_J(kR) \) is a solution to (4) and \( f_{J'}(k'R) \) obeys the corresponding equation for the, \( X^2\Sigma^+ \), exit channel with wavenumber and partial wave \( k', J' \) respectively. The radial wavefunctions are normalized as in (5) and
\[ h\omega = \frac{k^2}{2\mu} - \frac{k^2}{2\mu} + \Delta E \]
\[ \Delta E \equiv V_\chi(\infty) - V_\chi(\infty). \] (9)

According to (9) the maximum angular frequency \( \omega_{\text{max}} \) is given by
\[ h\omega_{\text{max}} = \frac{h k^2}{2\mu} + \Delta E. \] (10)

The sums given by (7) can be evaluated as in Stancil and Zygelman (1996), but here we use a simplified expression, derived in the appendix, in which \( \sigma_{\text{CT}} \) is replaced by its upper bound, i.e.
\[ \sigma_{\text{CT}} < \sigma_{\text{CT}} = \frac{8\pi^2\omega_{\text{max}}^3}{3c^4k^2} \sum_j (2J + 1) \int_0^\infty dR f_J^2(kR) D^2(R). \] (11)

### 2.3. Optical potential approach

An alternative approach for the calculation of the total radiative loss cross section is given by the local optical potential method (Zygelman and Dalgarno 1988). In it, the collision system in the incoming \( A^1\Sigma^+ \) state experiences, in addition to the BO energy \( V_\chi(R) \), a complex absorptive potential that has the form
\[ V_{\text{opt}} = -\frac{i\Lambda(t)}{2}, \]
\[ A(R) \equiv \frac{4}{3c^3} D^2(R)(V_\chi(R) - V_\chi(R))^3 \] (12)

where \( \Lambda(R) \) is an \( R \)-dependent Einstein-A coefficient that is illustrated in figure 2. The cross section for radiative quenching is given by
\[ \sigma = \frac{\pi}{k^2} \sum_j (2J + 1)(1 - \exp(-4\eta_j)) \] (13)

where \( \eta_j \) is the imaginary part of the Jth partial wave phase shift \( \delta_j \) for the radial wave \( f_J(kR) \) that satisfies
\[ f''_J(kR) - \frac{J(J + 1)}{k^2} f_J(kR) - 2\mu(V_\chi(R) + V_{\text{opt}}(R))f_J(kR) \]
\[ + k^2 f_J(kR) = 0. \] (14)

### 3. Ultra-cold limit

In the limit of ultra-cold temperatures in which only the \( s \)-wave of the entrance channel participates, the total radiative association cross section takes the form (Zygelman et al 2001)
\[ \sigma = \sum_n \frac{16\mu\pi\omega_{\text{opt}}}{3c^3k} \int_0^{R_0} dR f(R) D(R)\phi_{nJ=1}(R)^2 \] (15)

Figure 2. Einstein-A coefficient as a function of internuclear distance. The minimum near \( R = 8a_0 \) is a consequence of the transition dipole moment (Rellergert et al 2011) undergoing a sign change at that internuclear separation.

where \( \phi(R) \) is the \( s \)-wave solution to (4) subject to the boundary condition Zygelman et al (2001)
\[ \frac{d\phi(R)}{dR}|_{R_0} = 1 \]
at some, sufficiently large radius \( R_0 \) and \( \phi_{nJ=1}(R) \) are \( J = 1 \) rotational-vibrational states of the \( X^2\Sigma^+ \) potential. Because the overlap integral in (15) is independent of the incoming wavenumber \( k \), (15) predicts that the association cross section, in the ultra-cold regime, scales as the inverse of the collision velocity and, therefore, the rate tends to a constant.

In calculating \( \phi(R) \) one typically matches the numerical solution for \( f_J(kR) \) with the asymptotic form given by expression (5). Because of the polarization potential \( C_4/R^2 \), one must typically integrate far into the asymptotic region to achieve convergence. Exact solutions for the \( C_4/R^2 \) potential are given by radial Mathieu functions and better convergence can be achieved by employing the latter in the evaluations for the phasishifts e.g Spector (1964), Holzwarth (1973).

### 4. Results

Figure 1 illustrates the mechanism for radiative association for the Yb\(^+\) ion and Ca atom that approach in the \( A^1\Sigma^+ \) electronic BO state. The BO energies where taken from the data of the \textit{ab initio} calculations reported in Rellergert et al (2011). At large internuclear distances this potential has the form
\[ V_\chi(R) \rightarrow -\frac{C_4}{R^4}, \quad C_4^4 = 78.5. \] (16)

In the incident \( A^1\Sigma^+ \) channel the system can relax via the emission of a photon, and in the case of association, the final state is a bound rotational-vibrational level of the \( X^2\Sigma^+ \) channel. In radiative charge transfer the collision partners can exit in that channel, as a re-arranged Yb–Ca \(^+\) pair. In the exit channel,
\[ V_\chi(R) \rightarrow -\frac{C_4}{R^4} + \Delta E, \quad C_4^4 = 71.5, \quad \Delta E = -0.0052 \] (17)
as \( R \rightarrow \infty \). In calculating the radiative association cross sections given by (3) we need to itemize all bound states supported by the \( X^2\Sigma^+ \) channel. The total number of bound
states can be approximated using the Jeffreys–Wentzel–Kramers–Brillouin (JWKB) expression

\[ n = \sum_{J} \text{Floor} \left[ \int_{R_c}^{\infty} dR \sqrt{ -2 \mu (V_X(R) - \Delta E) - \frac{(J + 1/2)^2}{R^2} } \right] \approx 54 803 \]

(18)

where \( R_c \) is a classical turning point and \( \text{Floor}[x] \) is the integer lower bound of \( x \). Because of the large reduced mass \( \mu \), the number of bound states contributing is much larger than that for association of lighter species in which typically several hundred rotational-vibrational levels are supported (see Zygelman et al. 1998). However, at cold temperatures the centrifugal repulsion in the entrance channel limits the number of partial waves that participate and so limits, because of the \( J \pm 1 \) selection rules, the rotational-vibrational levels accessed. For example, at a collision energy corresponding to a temperature of 1 mK, only levels with \( J \) up to the value \( \approx 15 \) contribute to the association rate. In figure 3 we present the results of our calculations for a collision temperature of 1 mK. In that figure the circles represent the partial wave association cross sections obtained using the FGR expression (7), the symbol \( X \) in that figure represents the upper limit for total radiative relaxation, which is obtained by adding the association cross sections (7) with those given by expression (11). The square icons represent the cross sections predicted by expression (13). It is evident, from this figure, that for \( J < 10 \) the optical potential method provides an excellent approximation for the total cross sections, and for \( J > J_{\text{max}} \) (Zygelman and Hunt 2012),

\[ J_{\text{max}} = \sqrt{8 \mu k^2 C_a^2} = \sqrt{24 \mu^2 k_B T C_a^2} \approx 12 \]

(19)

the optical potential method is somewhat less reliable, though still gives reasonable order of magnitude estimates. \( J_{\text{max}} \) is the critical angular momentum for which the collision system, approaching in the incident channel at a given energy, has sufficient collision energy (here given by \( 3/2k_B T \), where \( k_B \) is the Boltzmann constant and \( T \) is the temperature in Kelvin) to overcome the centrifugal potential barrier (Zygelman and Hunt 2012). For larger \( J \) tunneling resonances can access the inner region where the transition dipole moment is non-negligible and induce a radiative transition. In table 1 we tabulate the various cross sections at several representative collision temperatures. In the second column we itemize the association cross section obtained using the FGR method described above. For the radiative charge transfer cross sections, itemized in the third column, we use expression (11). Thus the upper bound for the total radiative relaxation cross sections are given in column 4. The last column gives the results obtained using the local optical potential method. The table shows that, over the temperature range considered, the local optical potential method predicts cross sections that are less than the upper bound itemized in column 4. Secondly, the differences between the predictions of the two theories are small. The optical potential cross section differs by less than 4% from the upper limit values over the entire temperature range, including the ultra-cold region. We also note that the optical potential method predicts cross sections that are larger than the radiative association cross sections which underscores an observation cited made in Zygelman et al. (1989), that the optical potential method provides a reliable upper bound for the total (RR) cross section. In figure 4 we plot the total radiative relaxation cross section, obtained using the optical potential method, for the gas temperature range 1 mK \( \leq T \leq 1K \). Though the optical potential method provides a good approximation for the total radiative relaxation rate, calculation of the photon emission spectrum requires the use of the FGR method. In figure 5 we
illustrate the association cross sections $\sigma_{Jn}$ at $T = 1$ mK, for the individual rotational-vibrational levels as function of the frequency of the emitted photon. The structure in the emission pattern, which shows regions of suppressed and enhanced emission is a result of the oscillations in the incoming wave illustrated in figure 1. In the limit as $T \to 0$ we define a complex scattering length (Zygelman et al 2001) for the $s$-wave solution to (14),

$$a = -\frac{1}{k} \tan \delta(k) = 1.64 \times 10^5 - 1.8591i \quad k \to 0.$$ \hfill (20)

Therefore, the total RR cross section, according to the optical potential method, has the limiting value

$$\sigma = \frac{4\pi}{k} \left| \text{Im}[a] \right| = \frac{4\pi\delta}{k} \times 1.859.$$ \hfill (21)

Defining the rate coefficient

$$k_{RR} \equiv \langle v \sigma \rangle \quad T \to 0$$ \hfill (22)

we obtain $k_{RR} \approx 2.2 \times 10^{-12}$ cm$^3$ s$^{-1}$ and is about three orders of magnitude larger than the corresponding rate in the temperature range $1 \text{ mK} < T < 1 \text{ K}$. In figure 6 we plot the effective rates $k \equiv \langle v \sigma \rangle$ for different isotopes of the Yb$^+$ ion. At temperatures $T > 1 \mu$K the three rates, corresponding to the isotopes labelled in that figure, merge to a common value of about $1.5 \times 10^{-15}$ cm$^3$ s$^{-1}$. This feature can be attributed to Langevin behaviour (Vogt and Wannier 1954) which predicts that at low temperatures, but high enough that many partial waves contribute, ion-atom cross sections scale as the inverse of the collision velocity and therefore the rate coefficient tends to a constant. The value of that constant is only weakly dependent on the reduced mass of the collision system (e.g. see (2) in Zygelman and Hunt (2012)) and that behaviour is evident in figure 6. In the ultra-cold temperature regime, where $s$-wave scattering dominates, the $1/\nu$ behaviour in the cross sections is also operative, e.g. see equations (20) and (21), but for a different reason. Whereas in the Langevin regime the cross sections are governed largely by the $C_4$ coefficient, the ultra-cold $s$-wave phaseshift is also sensitive, as required by Wigner-threshold theory (Joachim 1984), to short-range parameters. So the presence of a real, or virtual, bound state near threshold, in the incoming $A^2\Sigma^+$ state of the molecular ions, can strongly influence that cross section. As a consequence, radiative quenching rates which are nearly constant in both the Langevin and ultra-cold regions, can suffer rapid variations in the temperature range that adjoins the two territories. This behaviour is illustrated in figure 6 by the rate for the $^{174}$Yb isotope. For this isotope, a bound state near the threshold leads to significant enhancement in the $s$-wave cross section at ultra-cold temperatures, the corresponding rate differs significantly from that in the Langevin region.

5. Summary and discussion

We have presented a computational study of the collision induced radiative processes (1,2) at gas temperatures that range from the cold to ultra-cold regimes. We found that at cold temperatures the total effective rates for these radiative processes is no larger than about $10^{-15}$ cm$^3$ s$^{-1}$. We evaluated the fidelity of the local optical potential method (West et al 1982, Zygelman and Dalgarno 1988) in its ability to predict radiative quenching rates, and found that it provides very accurate estimates for the latter even in the ultra-cold regime. We validated Langevin behaviour (which predicts nearly constant rates as a function of temperature) at higher temperatures but found dramatic departures, and a strong isotope dependence, in the transition from the cold to ultra-cold regimes. At ultra-cold temperatures the association rate also tends to a constant (Zygelman et al 2001) but for a different reason. Wigner threshold behaviour for the $s$-wave is strongly influenced by short-range parameters. Thus, there is a collision energy range in which there could be a rapid change in the effective rate, as illustrated in figure 6. This study demonstrates that is unlikely that the large rate reported in Rellergert et al (2011) is solely due to processes (1,2).

In addition to reactions (1,2), the system may also undergo the reaction

$$\text{Yb}^+ + \text{Ca} \to \text{YbCa}^+ (A^2\Sigma^+) + h\nu.$$ \hfill (23)

where association proceeds into the weakly bound ($A^2\Sigma^+$) molecular state in which the collision partners initially approach. Because process (23) is driven by a dipole moment.
that, at large $R$, is proportional to the internuclear distance, one might anticipate a significant rate for it. However, because the well is shallow leading to small photon energies, the contribution of the $\omega^2$ factor in RA is suppressed. Therefore, we found that its rate is negligible at the temperatures operative in this experiment.

We conclude that both additional experimental and theoretical studies are necessary in order to understand and reveal cold chemistry of the Ca$^+$ Yb system. On the experimental side, revisiting this reaction with new methods recently developed to better probe both the products of the reactions (Schowalter et al 2012) and the role, if any, of excited electronic states (Sullivan et al 2012) may help elucidate the relevant pathways. While, on the theoretical front, improved ab initio molecular potentials might help better understand the potential role of non-adiabatic effects in this reaction (Zygelman et al 1989).

Acknowledgments

We thank Professor Svetlana Kotochigova for use of the ab initio molecular data.

Appendix. Bound on RCT cross sections

According to (7)–(9) the frequency of the emitted photon, during an RCT transition, is

$$h \omega = \frac{k^2}{2\mu} - \frac{k^2}{2\mu} + V_A(\infty) - V_X(\infty)$$

$$= \frac{k^2}{2\mu} - E' + V_A(\infty) - V_X(\infty).$$  \hspace{1cm} (A.1)

Now $d(h \omega) = -dE'$ and $h \omega_{\text{max}} = \frac{k^2}{2\mu} - E' + V_A(\infty) - V_X(\infty)$, which corresponds to $E' = 0$, and $h \omega = 0$ for $E_{\text{max}}' = \frac{k^2}{2\mu} + V_A(\infty) - V_X(\infty)$. Therefore (7) can be written as

$$\sigma = \frac{8 \pi^2}{3} \alpha_0^2 \int_{E_{\text{max}}'}^{E'} dE' J M_{J,J-1}(k, E') [J M^2_{J,J-1}(k, E')]$$

$$+ (J + 1) M^2_{J,J-1}(k, E').$$  \hspace{1cm} (A.2)

We have the inequality

$$\sigma < \frac{8 \pi^2}{3} \alpha_0^2 (E'_{\text{max}})^2 \int_{E_{\text{max}}'}^{E'} dE' [J M^2_{J,J-1}(k, E')]$$

$$+ (J + 1) M^2_{J,J-1}(k, E').$$  \hspace{1cm} (A.3)

Consider the integral

$$\int_0^{\infty} dE' J M^2_{J,J-1}(k, E')$$

$$= \int_0^{\infty} dE' \int_0^{\infty} dR f_j(kR) D(R) f_{J-1}(k'R)$$

$$\times \int_0^{\infty} dR' f_j(k'R') D(R') f_{J-1}(k'R').$$  \hspace{1cm} (A.4)

Thus

$$\int_0^{\infty} dE' J M^2_{J,J-1}(k, E') \leq \sum_{E'} J M^2_{J,J-1}(k, E')$$

$$= J \int_0^{\infty} dR f_j(kR) D(R) f_{J-1}(kR).$$  \hspace{1cm} (A.5)

where the sum $\sum_{E'}$ includes all, bound and continuum states of the exit channel, and the second inequality follows from closure properties for the final states for a given value of $J$. Therefore we obtain the inequality

$$\sigma < \frac{8 \pi^2}{3} \alpha_0^2 (E'_{\text{max}})^2 \int_{J}^{\infty} dR f_j(kR) D(R) f_{J-1}(kR).$$  \hspace{1cm} (A.6)

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