Universal rates for reactive ultracold polar molecules in reduced dimensions

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The successful production of relatively dense gases or lattices of ultracold molecules in their ro-vibrational ground states opens up a number of new opportunities in physics and chemistry. Reaction rates of ultracold molecules can be quite large, as measured and calculated for the fermionic species $^{40}$K$^{87}$Rb. This highly reactive species belongs to the class of molecules that have universal reaction rates that can be calculated analytically from a knowledge of the long-range van der Waals (vdW) potential alone, given unit probability of reaction at short range. When an electric field is used to polarize a gas of this polar species, reaction rates become even larger and significantly more likely, even for quasi-two-dimensional (quasi-2D) geometry. This is essential, for example, to achieve evaporative cooling in order to reach quantum degeneracy and control such loss processes. This is possible only if the dipolar gas is in its rotational ground state, with a vdW length of the order of 1 nm, where strong chemical forces permit the reaction to occur.

Universal collision rates are completely determined by quantum threshold dynamics associated with the long range potential. Consequently, there are two distinct classes of mixed alkali-metal diatomic species. Those with energetically allowed reaction channels, KRb, LiNa, LiK, LiRb, and LiCs, are expected to be universal. By contrast, the species NaK, NaRb, NaCs, KCs, and RbCs have no reactive channels, and are expected to be non-universal in their ground rotational, vibrational, and spin state. However, even these species, as well as reactive homonuclear dimers, can have universal inelastic relaxation rates when vibrationally or rotationally excited. Universal species do not have scattering resonances, since threshold bound states decay too fast to make one, whereas non-universal species can have numerous scattering resonances. This paper treats the broad class of universal collisions.

Figure 1 shows the long-range potential $V(r, z)$ for two dipoles in quasi-2D geometry and illustrates essential features of reduced dimensional collisions, where

$$V = \frac{\mu \Omega^2 z^2}{2} + \frac{\hbar^2 (m^2 - 1/4)}{2 \mu \rho^2} - \frac{C_6}{r^6} + \frac{d^2}{r^3} \left(1 - \frac{3 z^2}{r^2}\right). \quad (1)$$

Here $r = (\rho, \varphi, z)$ represents the distance between the two molecules in cylindrical coordinates, and $r \equiv |r|$. The molecules are confined in the $z$-direction by a harmonic trap of frequency $\Omega$ and characteristic length $a_0 = \sqrt{\hbar/\mu \Omega}$, where $\mu$ is the reduced mass of the pair. The dipoles are assumed to be aligned along $z$, so the projection $m$ of their relative angular momentum is conserved. The second term represents the $m$-dependent centrifugal potential in 2D. The third term is the isotropic vdW potential, assuming the molecules are in their rotational ground state, with a vdW length $\bar{a} = [2\pi/\Gamma(1/4)^2] (2\mu C_6/\hbar^2)^{1/4}$. The last term describes universal elastic and reactive rates of quasi-two-dimensional and quasi-one-dimensional collisions of highly reactive ultracold molecules interacting by a van der Waals potential. Exact and approximate calculations for the example species of KRb show that stability and evaporative cooling can be realized for spin-polarized fermions at moderate dipole and trapping strength, whereas bosons or unlike fermions require significantly higher dipole or trapping strengths.
is the anisotropic dipole potential, with induced dipole moment \(d\) and dipolar length \(a_d = \mu d^2/\hbar^2\).

Our model relies on the separation of length scales: \(a_s \gg a_h \gg \bar{a} \gg a_c\), where \(a_c = 2\pi/\kappa\) is the DeBroglie wave length for a collision with relative kinetic energy \(E_k = \hbar^2\kappa^2/2\mu\). These inequalities are readily satisfied for experiments with \(^{40}\text{K}^{87}\text{Rb}\), for which \(a_c\), \(a_h\) and \(\bar{a}\) can be on the order of hundreds of nm, tens of nm, and less than 10 nm, respectively. Under these conditions, collisions are essentially quasi-2D [20, 21], or if additional confinement at frequency \(\Omega\) is provided along \(x\), quasi-1D [22]. Figure 1(a) illustrates the quasi-2D vdW case with \(d = 0\) for \(|m| = 1\), where the minimal action path followed by the two colliding particles lies in the plane \(z = 0\) of the contour diagram of \(V(\rho, z)\).

The competition between the centrifugal barrier and the vdW attraction determines a single saddle point at a distance \(r \gtrsim \bar{a}\), separating the long-distance 2D-scattering region from the short-range “core,” \(r < \bar{a}\), where the molecules are accelerated towards one another by the attractive potential and experience 3D scattering. Two additional out-of-plane saddle points appear when \(d\) increases so \(a_d/\bar{a} \gtrsim 2.71(\bar{a}/a_h)^{3/2}\), as in Fig. 1(b), indicating a crossover to dipolar-dominated scattering, which is fully reached for \(a_d > a_h \gg \bar{a}\). We show below that a strong enhancement of the ratio of elastic to reactive collisions in this regime will allow for an efficient cooling of the molecular gas.

Reference [6] characterized elastic and reactive collision rates for 3D collisions by a complex scattering length \(\tilde{a}_j(k)\), where \(\hbar k\) is the 3D momentum. For the special universal class of highly reactive molecules with unit short-range reaction probability, \(\tilde{a}_0(k) = (1 - i)\bar{a}\) for \(s\)-wave collisions of like bosons or unlike fermions, and \(\tilde{a}_1(k) = (1 - i)(k\bar{a})^2\bar{a}\) for \(p\)-wave collisions of like fermions, with \(\bar{a} = 1.064\bar{a}\) [6]. These explain the measured rates of 3D collisions of ultracold \(^{40}\text{K}^{87}\text{Rb}\) [5, 9]. Universal species have only incoming scattering current in the entrance channel in the vdW “core” of the collision, \(r < \bar{a}\), and this provides a universal boundary condition for both the vdW and dipolar cases illustrated in Fig. 1.

While the general model can be extended to nonreactive species like RbCs, where one expects a rich resonance structure, we here focus on universal species like KRb.

Applying universal theory to quasi-1D and quasi-2D collisions in a vdW potential is straightforward for the \(d = 0\) case by combining the methods and notation of Refs. [6, 23]. We give here only the resultant formulas, valid for \(\kappa\bar{a} \ll 1\). Assume the molecule is prepared in its vibrational, rotational, and spin ground state and in the ground state of confined motion. Only the first channel \(j\) of a coupled channels expansion is needed near threshold for small \(d\), and we set the index \(j = 0\) for like bosons or unlike fermions and \(j = 1\) for like fermions. Elastic and reactive collisions in \(N\) dimensions, \(N = 1, 2, 3\), are described by an \(S\)-matrix element \(S_{jj} = \exp(i\theta_j)\) written in terms of a complex phase \(\theta_j(\kappa)\) with

\[
\tan \theta_j(\kappa) = \frac{1 - S_{jj}(\kappa) - \bar{a}_{j}(\kappa)\kappa^{N-2}}{1 + S_{jj}(\kappa)} = -\tilde{a}_{j}(\kappa)\kappa^{N-2},
\]

where \(\kappa = p, q, k\) represents the momentum in 1D, 2D, and 3D respectively [23]. The quantity \(\tilde{a}_{j}(\kappa)\) on the right hand side defines the complex scattering phase through

\[
\tilde{a}_{j}(\kappa) = \frac{L_j(k)}{\kappa a_h^{3-N}} \frac{(1 - i)j^2(1 + r_j) - i}{1 + sr_j + r_j^2/2},
\]

Table I gives the lengths \(L_j(\kappa)\) and ratios \(r_j(\kappa)\) for \(N = 1, 2, 3\), where \(\xi_0 = \bar{a}/a_h\) and \(\xi_1 = \bar{a}_1\bar{a}/a_h^3\). For the case in Fig. 1, \(\xi_0 = 0.111, \xi_1 = 0.00145, q = (1/126) \text{nm}^{-1}\), \(q^2a_h^2 = 0.200\) for \(E_k = k_B \text{240 mK}\), where \(k_B\) is Boltzmann’s constant. Assuming \(a_h/a_h \ll 1\), we find \(r_j \ll 1\) and the right-hand factor in Eq. (3) can be approximated by \((-1)^j - i\), as in 3D [6]. The lengths \(L_j\) change only weakly across dimensions \(N\), so the scaling is mainly given by the factor \(a_h^{3-N}\).

The elastic \(K_{j}^{el}\) and reactive \(K_{j}^{re}\) scattering rate constants follow from the formulas in Ref. [23]:

\[
K_{j}^{el} = \frac{\pi \hbar}{\mu} g_j \frac{1 - |S_{jj}|^2}{\kappa^{N-2}}, \quad K_{j}^{re} = \frac{\pi \hbar}{\mu} g_j \frac{1 - |S_{jj}|^2}{\kappa^{N-2}},
\]
where $g_0 = 1/\pi, 2/\pi, 2$ and $g_1 = 1/\pi, 4/\pi, 6$ for molecules colliding in like spin states in $N = 1, 2, 3$ dimensions respectively, which take into account the $N$-fold degeneracy in $j = 1$. Unlike bosons or fermions have rate constants $(\langle K + K_1 \rangle)/2$. The upper bound, or unitarity limit, on the rate constants follows immediately upon replacing the $S$-matrix expressions by their upper bounds, $|1 - S_{jj}|^2 \leq 4$ and $|1 - |S_{jj}|^2 \leq 1$, respectively. The elastic $\Gamma_{j}^e$ and reactive $\Gamma_{j}^r$ collision rates per particle are

$$
\Gamma_j^e(\kappa) = \frac{4\pi \hbar}{\mu} q_j L_j(\kappa) \frac{n}{a_h^{3-N}} f_j(\kappa) \eta_j(\kappa), \quad (5a)
$$

$$
\Gamma_j^r(\kappa) = \frac{4\pi \hbar}{\mu} q_j L_j(\kappa) \frac{n}{a_h^{3-N}} f_j(\kappa), \quad (5b)
$$

with $n$ the density of the collision partner in $N$ dimensions (units of cm$^{-N}$), $n/a_h^{3-N}$ an equivalent 3D density (units of cm$^{-3}$) for $N = 1$ or 2 dimensions and $\eta_j(\kappa) = 2 L_j(\kappa) \kappa^{N-2}/a_h^{N}$ gives the ratio of elastic to reactive collision rates. The factor $f_j(\kappa) = (1 + r_j(\kappa) + r_j(\kappa)^2/2 + \eta_j(\kappa) + \eta_j(\kappa)^2/2)^{-1}$ approaches unity as $\kappa \to 0$, except for $j = 0$ for $N = 1, 2$, where $f_0(q) \to |p_{an}/\pi_0|^2/8$ and $f_0(q) \to \pi/2i\ln^2[2B/\pi a_0^{-2}]$, respectively. However, for realistic traps and energies in the nK-regime the full expression for $f_0(q)$ is required in 2D, since the logarithmic term only becomes dominant at much lower energies. The expressions in Eqs. (5a) and (5b) give explicitly the scaling with $\kappa$ and $a_h$, as well as all the known threshold laws for collision rates in reduced dimensions [20, 22, 25].

We use both coupled-channels (CC) methods and analytic/semiclassical approximations to show the effect of the dipole moment on 2D collision rates. The former uses a spherical harmonic basis set and a renormalized Numerov method to propagate the wave function $\Psi(r)$ with universal incoming wave boundary conditions in the vdW core [6] out to distances $r \gg a_h$. Then $\Psi(r)$ is matched onto a cylindrical basis and propagated to larger $r$ to yield the 2D $S$-matrix. The numerical results agree with the vdW limits from Eqs. (5), (4), and Table I and the dipolar results in Ref. [11].

Elastic collisions are well-described by a unitarized Born approximation (UBA), $S_{ij}^{2D} = (1 - iK_{jj}^{2D})(1 + iK_{jj}^{2D})^{-1}$, where the $K$-matrix element in 2D includes the vdW term from Eq. (4) plus the dipolar term,

$$
K_{jj}^{2D}(q) = -\bar{a}_{j}^{2D}(q) + 2\sqrt{\pi} \frac{a_h}{a_h} \phi_j(q a_h), \quad (6)
$$

with $\phi_0(x) = -0.65471 + 0.94146x - 0.39010x^2 + O(x^3)$ and $\phi_1(x) = -0.35555x + 0.36042x^2 - 0.13417x^3 + O(x^4)$. Equation (6) shows how the elastic constant scales with $q$, $a_h$, and $a_h$. Figures 2(a) and 3(a) show that the UBA is an excellent approximation for dipolar bosons and fermions for a wide range of realistic $E_q$, $\Omega$ and $d$.

Reaction rates can be estimated using an instanton technique [12, 22] with $V(\rho, z)$ to get the transmission probability $P^{2D}$ for tunneling through the barrier separating the long-distance 2D-scattering region $r \gg a$ from the short-range vdW core $r \approx a$ (see Fig. 1):

$$
P_{j}^{2D}(q) = A_j e^{-S_j^{2D}/s_h}, \quad S_j^{2D} = 2 \int_{r_1}^{r_2} ds \sqrt{2\mu(V_j(r_s^2) - E_q)}. 
$$

Here, $r_s^2$ is the path of minimal action given by the classical trajectory of a particle in the inverted potential with inner and outer turning points, $r_1$ and $r_2$, and $S_j^{2D}$ the associated Euclidian action. We use $V_j = V_j + h^2/2\mu r^2$ to take into account the semiclassical Langer-correction to the centrifugal term in the potential $(m^2 - 1/4 \to m^2)$, insuring correct threshold laws for $P_{j}^{2D}(q)$. We find $A_j = 1.0297(\bar{a}/a_h)$ by equating the analytic expression for $P_{j}^{2D}(q)$ to the analytic $1 - |S_1|^2$ in Eq. (1) in the vdW limit $d \to 0$, assuming $A_j$ is independent of $d$ for $a_d \approx a_h$. Since $S_1^0 \to 0$ as $d \to 0$ due to the disappearance of a centrifugal barrier for $j = 0$, we set $A_0 = 1$ to ensure unitarity is satisfied and only use $S_0^e$ to estimate $\kappa_0^e$ at finite $d$ where a barrier exists and $a_d \leq a_h$.

Figure 2 shows that the variation of $\kappa_{j}^e(E_q/k_B)$ and $\kappa_{j}^r(E_q/k_B)$ with energy is relatively weak. As in 3D, $\kappa_{j}^e(E_q/k_B)$ is independent of $E_q$ at low $d$, but quite unlike in 3D [17, 5], it decreases relative to the vdW limit when $d$ increases. The instanton method gives the qualitative explanation. At small $d$ the barrier to the in-plane path increases with $d$, thus decreasing $\kappa_{j}^e$. As $d$ increases, the existence of out-of-plane saddle points gives alternative paths with a lower barrier, so $\kappa_{j}^e$ starts to increase. As $d$ increases more, the increasing out-of-plane barrier strength eventually will cause $P_{j}^{2D}(q)$ to again decrease with increasing dipole, evident in the CC and instanton 150 kHz strong trap case in Fig. 3(b). Com-
Comparing Figure 3(a) and 3(b) shows the instanton approximation gives the qualitative trends for the boson case even better than for the fermion case at finite $d$.

Figure 3 illustrates the stability and cooling properties expected for universal polar bosons and fermions. In both cases $\mathcal{K}_{el}^j$ changes from the vdW limit by rapidly increasing with $d$ until it approaches the 2D unitarity limit at large $d$. Stability requires that $\Gamma_j^q(q)$ remain small enough that the lifetime $1/\Gamma_j^q(q)$ is sufficiently long, order of 1 s or longer, as achieved for $^{40}$K$^{87}$Rb fermions in 3D [9]. Equation (5b) predicts that the reaction rate per particle for identical fermions in the vdW limit is only $1/\sqrt{\pi}$ times lower in 2D than 3D, if the 2D system has the same equivalent 3D density $n^{2D}$/$a_0$. The initial decrease in $\mathcal{K}_{re}^j$ indicates not only increased fermionic stability in 2D, but also the possibility of evaporative cooling, which requires the ratio $\eta_j = K_{el}^j/K_{re}^j \gg 1$. Figure 3(b) shows that $\eta_1$ reaches a magnitude near 100 for $d = 0.18$ D, nearly independent of the trap $\Omega$. Figure 3 shows that the $\eta_j$ ratio increases with lower $E_j$, indicating that the evaporation improves as the 2D gas cools.

Figure 3(a) shows that there is less room to improve the bosonic elastic $\Gamma_j^q(q)$ with increasing $d$, since it is only an order of magnitude below unitarity in the vdW limit. The reactive $\Gamma_j^q(q)$ at low dipole strength is much larger than for fermions, so universal polar bosons have shorter lifetimes and poorer stability than fermions at the same dipole and trap strength. Furthermore, getting $\eta_0 \gtrsim 100$ requires either large $d$ or large $\Omega$. Figure 3 shows that $\eta_0 = 100$ in a 50 kHz trap near $d = 0.5$ D or in a 150 kHz trap for $d = 0.3$ D. Thus, stability and evaporation for universal polar bosons may be achievable.

In conclusion, we have developed universal analytic expressions for quasi-2D or quasi-1D collisions of highly reactive ultracold bosonic or fermionic molecules in the absence of an electric field and have calculated quasi-2D collisions of universal polar molecules with a dipole moment. While prospects for stability and evaporative cooling in 2D are much better for universal polar fermions than bosons, either species would benefit from larger dipole moments or tighter confinement. Non-reactive, non-universal species are expected to be very different from universal ones and to show numerous shape or Feshbach resonances as electric or magnetic fields are tuned.

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