Universal Rates for Reactive Ultracold Polar Molecules in Reduced Dimensions

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Analytic expressions describe 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 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.

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The successful production of relatively dense gases [1] or lattices [2] of ultracold molecules in their rovibrational ground states opens up a number of new opportunities in physics and chemistry [3,4]. Reaction rates of ultracold molecules can be quite large, as measured and calculated for the fermionic species $^{40}K^{87}Rb$ [5–7]. 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 the unit probability of reaction at short range [6]. When an electric field is used to polarize a gas of this polar species, reaction rates become even larger and significantly limit the lifetime of the dipolar gas [7,8]. Achieving stable dense samples of such reactive ultracold molecules requires understanding and controlling such loss processes. This is essential, for example, to achieve evaporative cooling in order to reach quantum degeneracy and thus utilize the molecules for new and exotic condensed matter phenomena that have been proposed [9–11]. Reference [12] predicted that molecular dipoles can be stabilized by tightly confining the molecules in a single plane of quasi-two-dimensional (quasi-2D) geometry. This is because the molecules can be arranged so as to experience repulsive dipolar forces at long range and thus never come together within chemical interaction distances in order to react. First steps have been taken towards understanding these effects by recent quasi-2D calculations with adiabatic potentials [13] or quantum dynamics for fermionic $^{40}K^{87}Rb$ [14].

Here we characterize quasi-2D and quasi-1D elastic and reactive collision rates of a broad class of bosonic and fermionic molecules that have universal rate constants. Analytic expressions apply in the case of vdW interactions. Quantum dynamical and approximate calculations for quasi-2D collisions of polar molecules show how collision rates scale with confinement length, dipole strength, and collision energy. Our analysis connects the critically important experimental domain between the vdW and strong dipole limits and provides a general framework for future research on molecular cooling, chemical reactivity, and condensed matter or quantum information applications for species with universal collision rates.

Universal collision rates occur for chemical species that have a near unit probability of reaction or inelastic relaxation when they are close enough together, on the order of typical chemical interaction distances $a_c \lesssim 1$ nm, where strong chemical forces permit the reaction to occur [6]. 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 [15], are expected to be universal. By contrast, the species NaK, NaRb, NaCs, KCs, and RbCs have no reactive channels [15] and are expected to be nonuniversal in their ground rotational, vibrational, and spin state. However, even these species, as well as nonreactive homonuclear dimers, can have universal inelastic relaxation rates when vibrationally or rotationally excited [16,17]. Universal species do not have scattering resonances, since threshold bound states decay too fast, whereas nonuniversal species can have prominent scattering resonances [18]. This Letter treats the broad class of universal collisions.

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

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

Here $\mathbf{r} = (\rho, \phi, z)$ represents the distance between the two molecules in cylindrical coordinates, and $r \equiv |\mathbf{r}|$. The molecules are confined in the z direction by a harmonic trap of frequency Ω and characteristic length $a_h = \sqrt{\hbar/\mu\Omega}$, where μ 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 poten-

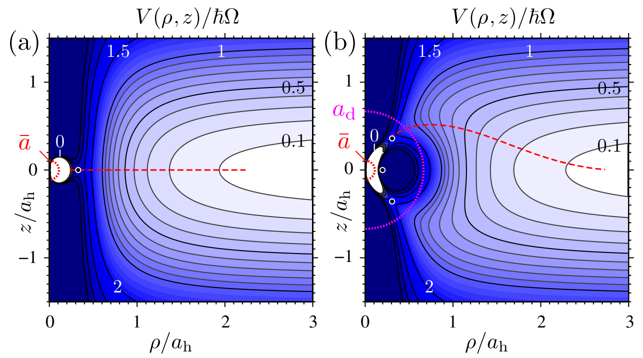

FIG. 1 (color online). Potential $V(\rho, z)/\hbar\Omega$ versus ρ/a_h and z/a_h for $|m| = 1$ for ^{40}K^{87}Rb in a $\Omega/2\pi = 50$ kHz trap, where $a_h = 56.4$ nm, $\bar{a} = 6.25$ nm with $C_6 = 16130$ a.u. [6,28]. (a) vdW only case, $d = 0$; (b) dipolar case for $d = 0.2$ D (with 1 D = 1 Debye $= 3.336 \times 10^{-30}$ C m). Saddle points (white circles), (a) in-plane and (b) out-of-plane minimum action paths (dashed lines). Dotted half-circles indicate $\bar{a}$ and a_d.

tial in 2D. The third term is the isotropic vdW potential, assuming the molecules are in their rotational ground state, with a vdW length $\bar{a} = [2\pi/\Gamma(1/4)^2](2\mu C_6/\hbar^2)^{1/4}$ [16,19]. The last term is the anisotropic dipolar 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: $a_\kappa \gg a_h \gg \bar{a} \gg a_c$, where $a_\kappa = 2\pi/\kappa$ is the de Broglie wavelength for a collision with relative kinetic energy $E_\kappa = \hbar^2\kappa^2/2\mu$. These inequalities are readily satisfied for experiments with ^{40}K^{87}Rb, for which a_κ, a_h, and $\bar{a}$ can be on the order of hundreds of nanometers, tens of nanometers, and less than 10 nm, respectively. Under these conditions, collisions are essentially quasi-2D [20–22] or, if additional confinement at frequency Ω is provided along x, quasi-1D [23]. 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 $\tilde{a}_1(k) = (-1 - i)(k\bar{a})^2\bar{a}_1$ for p-wave collisions of like fermions, with $\bar{a}_1 \approx 1.064\bar{a}$ [6]. These explain the measured rates of 3D collisions of ultracold ^{40}K^{87}Rb [5,6]. 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.

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. [6,24]. 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 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

$$\tan\theta_j(\kappa) = i\frac{1 - S_{jj}(\kappa)}{1 + S_{jj}(\kappa)} = -\tilde{a}_j(\kappa)\kappa^{N-2}, \qquad (2)$$

where $\kappa = p$, q, and k represents the momentum in 1D, 2D, and 3D, respectively [24]. The quantity $\tilde{a}_j(\kappa)$ on the right-hand side defines the complex scattering phase through

$$\tilde{a}_j(\kappa) = \frac{L_j(\kappa)}{a_h^{3-N}}\frac{(-1)^j(1 + r_j) - i}{1 + r_j + r_j^2/2}. \qquad (3)$$

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 $\xi_1 = \bar{a}_1\bar{a}^2/a_h^3$. For the case in Fig. 1, $\xi_0 = 0.111$, $\xi_1 = 0.001\,45$, $q = (1/126)$ nm^{-1}, and $q^2a_h^2 = 0.200$ for $E_\kappa = k_B 240$ nK, where k_B is Boltzmann's constant. Assuming $\bar{a}/a_h \ll 1$, 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 $\mathcal{K}_j^{\rm el}$ and reactive $\mathcal{K}_j^{\rm re}$ scattering rate constants follow from the formulas in Ref. [24]:

$$\mathcal{K}_j^{\rm el} = \frac{\pi\hbar}{\mu}g_j\frac{|1 - S_{jj}|^2}{\kappa^{N-2}}, \quad \mathcal{K}_j^{\rm re} = \frac{\pi\hbar}{\mu}g_j\frac{1 - |S_{jj}|^2}{\kappa^{N-2}}, \qquad (4)$$

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 and fermions have rate constants $(\mathcal{K}_0 + \mathcal{K}_1)/2$. The upper bound, or unitarity limit, on the rate constants follows immediately upon replacing the S-matrix expressions by their upper bounds $|1 - S_{jj}|^2 \le 4$ and $1 - |S_{jj}|^2 \le 1$, respectively. The elastic $\Gamma_j^{\rm el} = \mathcal{K}_j^{\rm el} n$ and reactive $\Gamma_j^{\rm re} = \mathcal{K}_j^{\rm re} n$ collision rates per particle are

$$\Gamma_j^{\rm el}(\kappa) = \frac{4\pi\hbar}{\mu} g_j L_j(\kappa) \frac{n}{a_h^{3-N}} f_j(\kappa)\eta_j(\kappa), \tag{5a}$$

$$\Gamma_j^{\rm re}(\kappa) = \frac{4\pi\hbar}{\mu} g_j L_j(\kappa) \frac{n}{a_h^{3-N}} f_j(\kappa), \tag{5b}$$

with n the density of the collision partner in N dimensions (units of cm^{-N}) and n/a_h^{3-N} an equivalent 3D density (units of cm^{-3}) for $N = 1$ or 2 dimensions, and $\eta_j(\kappa) = 2L_j(\kappa)\kappa^{N-2}/a_h^{3-N}$ 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 $f_0(p) \to [pa_h/\xi_0]^2/8$ and $f_0(q) \to \pi/2\ln^2[2B/\pi q^2 a_h^2]$, 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 becomes dominant only at much lower energies. The expressions in Eqs. (5a) and (5b) give the known threshold law limits for collision rates in reduced dimensions [20,21,23,26]. The $\Gamma_j^{\rm el}(\kappa)/\Gamma_j^{\rm re}(\kappa)$ ratio is $\sqrt{\pi}(\kappa a_h)^{-1}$ times larger for quasi-2D than 3D, as found in Ref. [22]. Also, $\Gamma_0^{\rm el}(\kappa)$ for bosons agrees with the result of Ref. [20] if the complex scattering length $\bar{a}(1 - i)$ is introduced into their derivation.

We use both coupled-channels (CC) methods and analytical or 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 $\Psi(\mathbf{r})$ with universal incoming wave boundary conditions in the vdW core [6] out to distances $r \gg a_h$. Then $\Psi(\mathbf{r})$ is matched onto a cylindrical basis and propagated to larger ρ to yield the 2D S matrix. The numerical results agree with the vdW limits from Eqs. (3) and (4), Table I, and the dipolar results in Ref. [14].

Elastic collisions are well-described by a unitarized Born approximation (UBA) $S_{jj}^{\rm 2D} = (1 - iK_{jj}^{\rm 2D}) \times (1 + iK_{jj}^{\rm 2D})^{-1}$, where the K-matrix element in 2D includes the vdW term from Eq. (3) plus the dipolar term:

$$K_{jj}^{\rm 2D}(q) = -\tilde{a}_j^{\rm 2D}(q) + 2\sqrt{\pi}\frac{a_d}{a_h}\phi_j(qa_h), \tag{6}$$

with $\phi_0(x) = -0.65471 + 0.94146x - 0.39010x^2 + \mathcal{O}(x^3)$ and $\phi_1(x) = -0.35555x + 0.36042x^2 - 0.13417x^3 + \mathcal{O}(x^4)$. Equation (6) shows how the elastic rate constant scales with q, a_d, 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, Ω, and d.

TABLE I. Lengths $L_j(\kappa)$ and ratios $r_j(\kappa)$ defining the universal complex scattering phase in N dimensions; ζ is the Riemann ζ function, $B = 0.905$ [20], and $W(0) = 0.328$ [25].

N	3D	2D	1D
L_0	$\bar{a}$	$\sqrt{\pi}\bar{a}$	$2\bar{a}$
L_1	$(k\bar{a})^2\bar{a}_1$	$(3\sqrt{\pi}/2)(q\bar{a})^2\bar{a}_1$	$6(p\bar{a})^2\bar{a}_1$
r_0	0	$(2/\sqrt{\pi})\xi_0\ln[2B/\pi q^2 a_h^2]$	$2\xi_0\zeta(1/2)$
r_1	0	$(2/\sqrt{\pi})\xi_1 W(0)$	$24\xi_1\zeta(-1/2)$

Reaction rates can be estimated by using an instanton technique [12,27] with $V(\rho, z)$ to get the transmission probability $P_j^{\rm 2D}$ for tunneling through the barrier separating the long-distance 2D-scattering region $r \gg \bar{a}$ from the short-range vdW core $r \lesssim \bar{a}$ (see Fig. 1):

$$P_j^{\rm 2D}(q) = A_j e^{-S_j^{\rm cl}/\hbar}, \qquad S_j^{\rm cl} = 2\int_{\mathbf{r}_1}^{\mathbf{r}_2} ds\sqrt{2\mu[\tilde{V}_j(\mathbf{r}_j^{\rm cl}) - E_q]}.$$

Here, $\mathbf{r}_j^{\rm cl}$ is the path of minimal action given by the classical trajectory of a particle in the inverted potential with inner and outer turning points $\mathbf{r}_1$ and $\mathbf{r}_2$, respectively, and $S_j^{\rm cl}$ the associated Euclidian action. We use $\tilde{V}_j = V_j + \hbar^2/8\mu\rho^2$ to take into account the semiclassical Langer correction to the centrifugal term in the potential ($m^2 - 1/4 \to m^2$), ensuring correct threshold laws for $P_j^{\rm 2D}(q)$. We find $A_1 = 1.0297(\bar{a}/a_h)$ by equating the analytic expression for $P_1^{\rm 2D}(q)$ to the analytic $1 - |S_{11}|^2$ in Eq. (4) in the vdW limit $d \to 0$, assuming A_j is independent of d for $a_d \lesssim a_h$. Since $S_0^{\rm cl} \to 0$ 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 $S_0^{\rm cl}$ to estimate $\mathcal{K}_0^{\rm el}$ at finite d where a barrier exists and $a_d \lesssim a_h$.

Figure 2 shows that the variation of $\mathcal{K}_1^{\rm el}/(E_q/k_B)$ and $\mathcal{K}_1^{\rm re}/(E_q/k_B)$ with energy is relatively weak. As in 3D, $\mathcal{K}_1^{\rm re}/(E_q/k_B)$ is independent of E_q at low d, but quite unlike in 3D [7,8], 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 $\mathcal{K}_1^{\rm re}$. As d increases, the existence of out-of-plane saddle points gives alternative paths with a lower barrier, so $\mathcal{K}_1^{\rm re}$ starts to increase. As d increases more, the increasing out-of-plane barrier strength

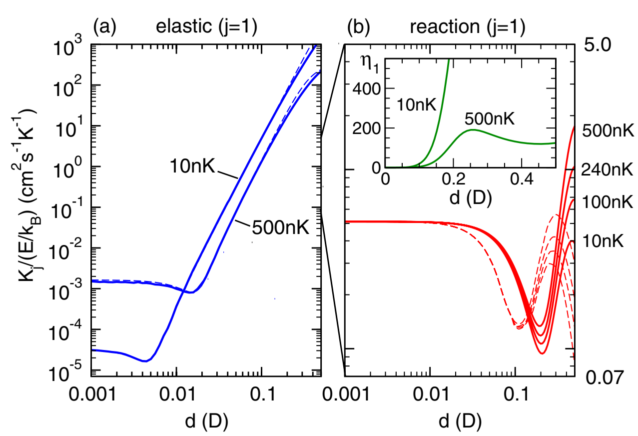

FIG. 2 (color online). Quasi-2D rate constants per energy, $K_1/(E_q/k_B)$, for elastic (a) and reactive (b) collisions of identical KRb fermions ($j = 1$) versus dipole strength d for an $\Omega/2\pi = 50$ kHz trap ($a_d = a_h$ for $d = 0.25$ D) and different E_q/k_B; CC (solid lines) and UBA/instanton (dashed lines). The inset in (b) shows the ratio $\eta_1 = \Gamma_1^{\rm el}(\kappa)/\Gamma_1^{\rm re}(\kappa)$.

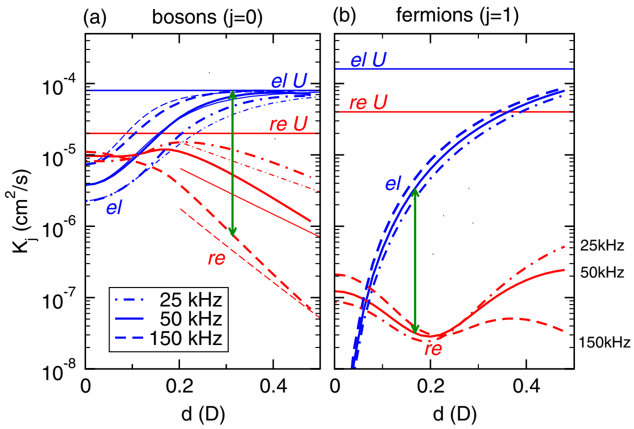

FIG. 3 (color online). Quasi-2D elastic (el) and reaction (re) rate constants K_j for identical KRb (a) bosons ($j = 0$) and (b) fermions ($j = 1$) at a collision energy of $E_q = k_B 240$ nK $= h5$ kHz for three different trap frequencies $\Omega/2\pi = 25$, 50, and 150 kHz. CC (thick lines), UBA/instanton (thin lines), unitarity limits (horizontal lines). Vertical arrows indicate where $\eta_j = 100$.

eventually will cause $P_j^{2D}(q)$ to again decrease with an increasing dipole, evident in the CC and instanton 150 kHz strong trap case in Fig. 3(b). Comparing Fig. 3(a) shows the instanton approximation also gives the qualitative trends for the boson case.

Figure 3 illustrates the stability and cooling properties expected for universal polar bosons and fermions. In both cases, $\mathcal{K}_j^{el}$ changes from the vdW limit by rapidly increasing with d until it approaches the 2D unitarity limit at large d. Stability requires that $\Gamma_j^{re}(q)$ remain small enough that the lifetime $1/\Gamma_j^{re}(q)$ is sufficiently long, of the order of 1 s or longer, as achieved for $^{40}K^{87}Rb$ fermions in 3D [5]. Equation (5b) predicts that $\Gamma_1^{re}(\kappa)$ 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_h. The initial decrease in $\mathcal{K}_1^{re}$ indicates not only increased fermionic stability in 2D, but also the possibility of evaporative cooling, which requires the ratio $\eta_j = \mathcal{K}_j^{el}/\mathcal{K}_j^{re} \gg 1$. Figure 3(b) shows that η_1 reaches a magnitude near 100 for $d = 0.18$ D, nearly independent of the trap Ω. Figure 2(b) shows that the η_1 ratio increases with lower E_q, indicating that the evaporation improves as the 2D gas cools.

Figure 3(a) shows that there is less room to improve the bosonic elastic $\Gamma_0^{el}(q)$ with increasing d, since it is only an order of magnitude below unitarity in the vdW limit. The reactive $\Gamma_0^{re}(q)$ 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 Ω. 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.

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