

Losses, Many-Body Correlations, and Universality in Ultracold Molecules

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Recent experimental developments have allowed physicists to freeze molecules' motion down to an ultracold temperature regime where quantum effects become profound. Furthermore, each molecule can be precisely prepared at chosen internal states and the mutual interactions between molecules are also highly tunable. As such, ultracold molecules have emerged as a powerful platform in multiple disciplines across physics and chemistry. Meanwhile, a grand challenge exists as to how losses of molecules depend on a quantum many-body environment. In this article, the recent experimental and theoretical progress of exploring losses of ultracold molecules is reviewed. Since the conventional theoretical scheme of treating isolated pairs of molecules is no longer applicable to the quantum degenerate regime that has been reached in recent experiments, an alternative framework of universal relations between two-body losses and many-body correlations has been established. Regardless of microscopic parameters ranging from the temperature and the particle number to the interaction strength, these universal relations always hold. This approach unfolds a simple universality behind complex loss processes of many-body systems and provides physicists and chemists with a new tool to explore ultracold molecules.

of ultracold molecules has emerged. Soon after the realization of degenerate Fermi gases, the Feshbach resonance has been used to associate two atoms to a molecule to access molecular clouds at low temperatures. A wide range of important phenomena, such as BEC-BCS crossover, vortices in fermionic superfluids, and the strongly interacting fermionic superfluids with the highest record of the ratio of the transition temperature to the Fermi temperature, have been extensively explored.^[7–11] Furthermore, the creation of molecules at well-controlled internal states opened the door for physicists to manipulate degrees of freedom of molecules that are inaccessible in atoms, ranging from the much richer nuclear spins to rotational and electronic states.^[13–49] It is thus expected that ultracold molecules will allow physicists to study a broad spectrum of important problems in multiple disciplines, including but not limited to novel quantum matter,^[50–53] fast quantum information processing,^[54–59] and quantum chemistry.^[60–68]

1. Overview

The realization of Bose–Einstein condensates in laboratories has brought physicists to an ultracold world in which intriguing quantum phenomena arise in a temperature regime down to a few nano-Kelvin.^[1,2] In the past many years, a vast range of important quantum states and quantum phenomena have been accessed and explored in the field of quantum gases.^[3–12] While the study of ultracold atoms continues to prosper, a new platform

New opportunities are accompanied by new challenges. Molecules often suffer losses. For instance, collisions between two KRb molecules lead to a chemical reaction that results in the production of two other molecules K_2 and Rb_2 , $2KRb \rightarrow K_2 + Rb_2$.^[60] As such, the number of KRb molecules in the sample decays as time goes by, an intrinsic challenge for utilizing KRb molecules for whatever purposes in quantum science and technology. Other molecules, such as LiNa,^[17] LiK,^[18] LiRb, and LiCs,^[21] are also known to be chemically unstable.^[69,70] Whereas some species of molecules, including NaK,^[22] NaRb,^[27] and RbCs,^[28–36] are chemically stable, another loss mechanism exists. Such molecules could form long-lived complexes. For instance, in the process of $2NaRb \rightarrow Na_2 + Rb_2$, a variety of intermediate complexes denoted by $Na_2Rb_2^*$ could form.^[61] Despite the fact that it is typically difficult to detect long-lived complexes, recent experimental progress has made it possible to image the complexes formed by KRb molecules.^[64,65] Nevertheless, as far as the molecular system is concerned, the formation of long-lived complexes also provides a source of losses.

A critical question in the study of ultracold molecules then arises. How to understand losses of ultracold molecules in the temperature regime where quantum effects are profound? A well-established framework in the literature is to consider the collisions of two molecules. Though it is a sophisticated problem

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to take into account short-range physics that gives rise to losses, such a two-body quantum mechanical problem is still tractable and theoretical results obtained from this approach have well explained some key observations in experiments.^[71–77] However, a recent experimental breakthrough has imposed an even bigger challenge to theorists. The long-expected degenerate fermionic molecules have been realized for the first time in laboratories by Jun Ye's group at JILA.^[63] At such a low-temperature regime down to a fraction of the Fermi temperature, many-body correlations become drastic and it is no longer sufficient to consider only collisions of an isolated pair of molecules. Though theoretical predictions work well in the high-temperature regime, the measured decay rate of molecules below the Fermi temperature deviates significantly from theoretical results. Some new theoretical frameworks are thus desired to capture how quantum many-body correlations affect losses of ultracold molecules.

A new theoretical approach has been recently established to correlate two-body losses and many-body correlations through universal relations governed by contacts, the central quantities in dilute quantum systems.^[78] The concept of contacts was originally invented for studying two-component fermionic atoms with *s*-wave scatterings.^[79–81] As of now, contacts have been well accepted as the most fundamental quantities that govern quantum gases and other related dilute systems.^[82–121] Implementing contacts in ultracold molecules, we are able to unfold exact relations between the loss rate and other quantities, including but not limited to the momentum distribution and the density-density correlation function, which are valid at any temperatures, any interaction strength, and any particle numbers. On the one hand, these universal relations show that universality exists in chemical reactions and other two-body losses in a simple form despite the fact that the quantum many-body environment could be very complicated. On the other hand, this framework provides both experimentalists and theorists with a powerful tool to study losses in quantum many-body systems, since such universal relations always hold even in the parameter regimes where theoretical calculations of many physical quantities become difficult.

In this article, we will review some recent experimental and theoretical studies of ultracold molecules. In Section 2, we will start from a brief survey of applications of ultracold molecules in a variety of disciplines, loss mechanisms in experiments, the conventional theoretical schemes to study losses, and the recent JILA experiment realizing degenerate fermionic molecules. We will then discuss universal relations in lossy systems in Section 3. Both *s*-wave and high partial wave scatterings will be covered. As two prototypical examples, the momentum distribution and the density–density correlation function will be discussed. Last but not the least, in Section 4, we will address some interesting future directions, such as low dimensions and finite electric fields that produce dipole–dipole interactions in molecules.

2. Ultracold Molecules: A Powerful Apparatus

2.1. A Broad Range of Applications

In the ultracold regime, both the internal degrees of freedom of a single molecule and the mutual interactions between molecules are highly controllable. Ultracold molecules thus can be implemented for multiple purposes. Here, we briefly survey applica-

tions of ultracold molecules in condensed matter physics, quantum information processing, and quantum chemistry.

2.1.1. Exotic Quantum Phases

One of the central tasks in condensed matter physics is to search for new quantum phases. In the past decade, a variety of interesting quantum states have been predicted in ultracold molecules. For instance, by applying static and microwave fields to dress rotational states of molecules, it has been found that both the shape and the strength of interactions between molecules can be tuned, and consequently, a crystalline phase could form.^[50] Changing the interaction strength further gives rise to melting to a superfluid phase. Chiral topological superfluids could arise when the time-averaged interaction induced by microwaves becomes attractive at large separations between molecules.^[51] Applying a tilted electric field to a 2D array of molecules, whose positions are pinned, the effective Hamiltonian describing the rotational degree of freedom may support topological flat bands and fractional Chern insulators have been predicted to exist.^[52] An effective spin-orbit coupling can also be produced from dipole–dipole interactions in 2D polar molecules and leads to chiral excitations, which inevitably affect spin dynamics and a variety of other properties of the molecular system.^[53]

2.1.2. Quantum Information Processing

Long coherence times in internal degrees of freedom of ultracold molecules, such as nuclear spins and rotational states, make them a promising platform for quantum information processing. It has been suggested that electric dipole moments of ultracold molecules could be implemented as qubits in the sense that the dipole moments oriented along (against) the electric field represent $|0\rangle$ ($|1\rangle$).^[54] Whereas each qubit can be individually controlled by an electric field gradient, dipole–dipole interaction between molecules could further couple two qubits and deliver controlled NOT gates with a long decoherence time. By selectively coupling either $|0\rangle$ or $|1\rangle$ to excited states, the dipolar interactions can be switched on and off to deliver phase gates.^[55] Recently, it has also been suggested that rotational states of ultracold molecules can be implemented for error-correcting codes, robust storage, and coherent processing of quantum information.^[59]

2.1.3. Quantum Chemistry

It is of fundamental interest to explore how chemical reactions occur at low temperatures when quantum effects are profound. In a pioneering experimental work, it has been found that quantum statistics become critical in determining the reaction rate.^[60] Current experiments have found that the reaction rates of *s*-wave scatterings are typically one or two orders of magnitude larger than those of *p*-wave scatterings in the low-temperature regime due to the absence of the barrier, whereas *p*-wave scatterings naturally experience an angular momentum barrier. The temperature of the molecular sample has been further cooled down to a fraction of the Fermi temperature, opening the door

to study chemical reactions in quantum degenerate gases.^[63] Internal states of molecules have been precisely controlled to study loss rates with and without chemical reactions.^[61] The formation of long-lived complexes sometimes is also referred to as a sticky collision.^[62] Though it is difficult to observe intermediates of reactions, a recent experiment has extended lifetimes of intermediate complexes by constraining the number of exit channels and has used ionization spectroscopy to achieve a direct observation of intermediates.^[64]

2.2. A Key Issue: Losses

As previously mentioned, both chemical reactions and the formation of long-lived complexes could cause losses in the molecular samples. In this section, we briefly review conventional theoretical treatments of losses and a grand challenge raised by a recent experiment.

2.2.1. Theoretical Treatments: Conventional Schemes

A well-established model in the literature was proposed by Idziaszek and Julienne for studying threshold collision rates of a pair of ultracold reactive molecules.^[71] This model considers two molecules interacting via the van der Waals interaction at large distances and a complex potential at short distances that leads to the chemical reaction. The long-range part of the interaction potential in the entrance channel of the two colliding molecules is written as

$$U_l(\mathbf{r}) = -\frac{A}{r^6} + \frac{\hbar^2 l(l+1)}{Mr^2} \quad (1)$$

where M is the mass of each molecule. The first term is the van der Waals interaction potential characterized by the length scale $\tilde{r} = (MA/\hbar^2)^{1/4}$, and the second term is the centrifugal barrier of the l -th partial wave. Whereas there are many scattering channels for the molecular collisions, the near-unity probability of chemical reactions simplifies the treatments. The idea is that once two molecules in the entrance channel manage to overcome the long-range barrier shown in Equation (1) and collide in the short-range, there will be barely flux reflected back to the entrance channel and the complex reaction process can be well approximated by a nearly unit probability of reaction that occurs in the short-range. Combining this approach and the generalized multi-channel quantum-defect theory (MQDT), nice agreements between the theory and experiments have been accessed above the Fermi temperature.

In the weakly interacting regime, it has been found that the loss rate is determined by the imaginary part of the scattering length. In this regime, $q_\epsilon^{2l+1}|a_l| \ll 1$ and $q_\epsilon \tilde{r} \ll 1$, where $q_\epsilon = (M\epsilon/\hbar^2)^{1/2}$ and ϵ is the collision energy. The scattering length a_l of the l -th partial wave is defined by $q_\epsilon^{2l+1} \cot \eta_l = -1/a_l$, where η_l is the phase shift of l -th partial wave scatterings. When the reaction probability reaches unity, a_l approaches a universal constant, $a_0 = [2\pi\tilde{r}/\Gamma^2(1/4)](1-i)$ and $a_1 = [(\pi/18)\tilde{r}^3/\Gamma^2(3/4)](-1-i)$, the universal inelastic (or reactive) loss rate is determined solely by the quantum transmission of the long-range potential. For instance,

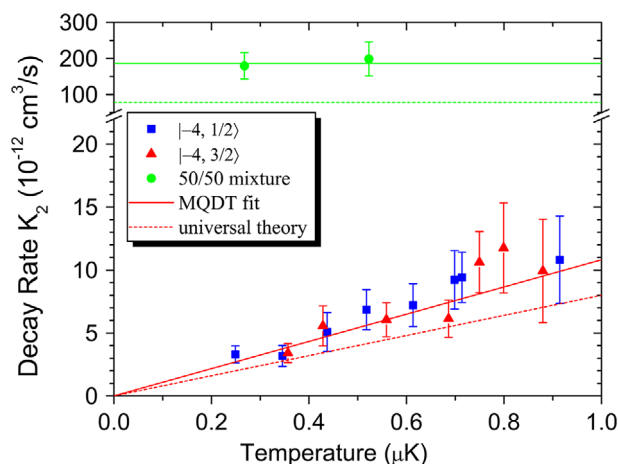


Figure 1. The temperature dependence of loss rate \mathcal{K} of $^{40}\text{K}^{87}\text{Rb}$ molecules above the Fermi temperature. $^{40}\text{K}^{87}\text{Rb}$ molecules are prepared in their rovibronic ground state. The blue squares, red triangles, and green dots are the experimental data^[60] for a single component gas of molecules in spin states $|F = -4, M_F = 1/2\rangle$, $|F = -4, M_F = 3/2\rangle$, and for 50/50 mixture of these two spin states, respectively. The dashed lines and solid lines are the prediction of MQDT in the universal regime and in the non-universal regime, respectively. Reproduced with permission.^[71] Copyright 2010, The American Physical Society.

the loss rates of the s -wave ($l = 0$) and the p -wave ($l = 1$) are written as

$$\mathcal{K}_{l=0} = -4g \frac{\hbar}{M} \text{Im}(a_0) \quad (2)$$

$$\mathcal{K}_{l=1} = -12g \frac{\hbar}{M} q_\epsilon^2 \text{Im}(a_1) \quad (3)$$

The factor $g = 1$ for distinguishable particles and $g = 2$ for identical particles, respectively. To implement the above two-body results in a many-body system, a thermal average has been considered, $\mathcal{K}_i(T) = \int \mathcal{K}_i f(\epsilon) d\epsilon$, where $f(\epsilon) d\epsilon = (2/\sqrt{\pi})(k_B T)^{-3/2} \exp[-\epsilon/(k_B T)] \epsilon^{1/2} d\epsilon$ is the Boltzmann distribution. If the temperature is well above the Fermi temperature, such an average is allowed since correlations between different pairs of molecules are negligible. The resultant temperature-dependent universal inelastic loss rates for the identical molecules are given by

$$\mathcal{K}_{l=0}(T) = -8 \frac{\hbar}{M} \text{Im}(a_0) \quad (4)$$

$$\mathcal{K}_{l=1}(T) = -\frac{144\pi^2}{h} \text{Im}(a_1) k_B T \quad (5)$$

These results well explained the experimental data that were collected above the Fermi temperature, as shown in **Figure 1**.

Whereas some molecules such as KRb ^[60,63] have exothermic chemical reactions, molecules including NaK ,^[22] NaRb ,^[27] and RbCs ^[31] have chemical reactions $\text{AB} + \text{AB} \rightarrow \text{A}_2 + \text{B}_2$ that are endothermic. These molecules are considered as chemically sta-

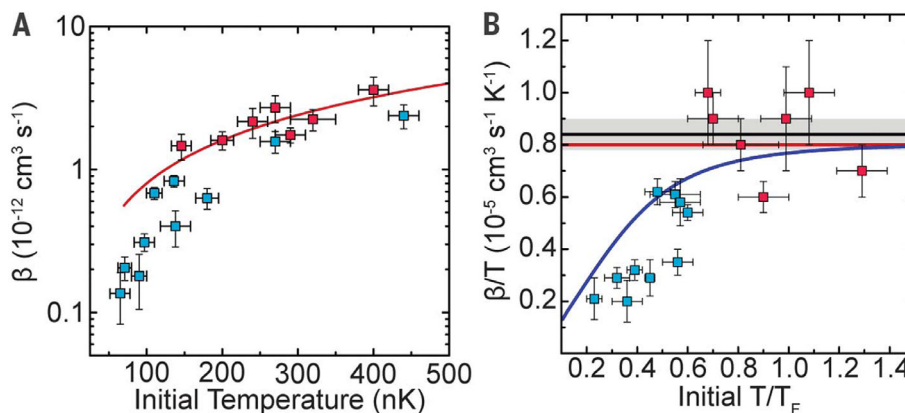


Figure 2. The temperature dependence of reaction rate of $^{40}\text{K}^{87}\text{Rb}$ molecules above and below the Fermi temperature. A) The decay rate β for initial temperatures ranging from $T = 70$ to 450 nK . $dn/dt = -\beta n^2$ and n is the average classical molecular density of the bulk gas. Solid blue points and solid red points correspond to $T/T_F \leq 0.6$ and $T/T_F > 0.6$, respectively. The red curve is the theoretical prediction from MQDT. B) The scaled decay rate from (A) as a function of T/T_F . The solid black line is the average β/T for $T/T_F > 0.6$ and the gray bar is its corresponding error range. The red line and the blue curve are the MQDT prediction and the average relative density fluctuations, respectively. Reproduced with permission.^[63] Copyright 2019, American Association for the Advancement of Science.

ble. However, two-body losses were still discovered in these molecules.^[61,62] Mayle et al. proposed a “sticky collision” hypothesis in 2013.^[72] The collisions of two AB molecules can form transient complexes, A_2B_2^* . They proposed that the formed A_2B_2^* complex can live long enough such that it can have inelastic collisions with a third molecule, which lead to the loss of all three molecules from the system. The lifetime (or sticking time), τ_{stick} , was obtained by the Rice–Ramsperger–Kassel–Markus (RRKM) theory^[122] and MQDT.^[72,74] $\tau_{\text{stick}} = 2\pi\hbar\rho$, where ρ is the density of states. Christianen et al. estimated lifetimes for similar alkali complexes in 2019 and found that the lifetimes were about three orders of magnitude shorter, which makes a secondary collision nearly impossible.^[75] Instead, they proposed that the observed losses are due to the interaction between the complexes and the light of the optical dipole trap.^[76] Nowadays, long-lived complexes have attracted considerable interest from physicists due to their potential role in dealing with losses of ultracold molecules, especially those which were chemically stable. More discussions of long-lived complexes in the ultracold regime can be found from a review by Liu and Ni.^[66]

2.2.2. A Recent Experimental Breakthrough and a Challenging Question

While the aforementioned theoretical predictions for loss rates of ultracold reactive molecules work well above the Fermi temperature, a recent experiment of Jun Ye’s group in JILA has cooled the fermionic polar molecules to the long-expected degenerate regime, down to $T/T_F = 0.3$, where T_F is the Fermi temperature.^[63] This is the first realization of degenerate Fermi gases of polar molecules in the laboratory, representing a breakthrough in atomic, molecular, and optical (AMO) physics. When the temperature is below the Fermi temperature, the previously mentioned theoretical results no longer agree well with experimental data. When $T/T_F < 0.5$, the detected loss rate shows a

large suppression, deviating significantly from the theoretical results, as shown in **Figure 2**.

As seen from previous discussions, the universal rate constants are obtained solely from a two-body problem, that is, collisions of an isolated pair of molecules. The Boltzmann distribution of the scattering energies used in the thermal average does not take into account correlations between different pairs. When the temperature is well above T_F , this is certainly a good approximation. However, this treatment becomes less accurate with decreasing the temperature. In particular, in the quantum degenerate regime, quantum many-body effects become profound and correlations between different pairs of molecules cannot be ignored anymore. A new theoretical scheme is thus desired to include both the two-body physics and the many-body correlations to fully capture the losses of ultracold molecules. Though this is, in general, an extremely challenging task, the length scale separation that naturally exists in typical experiments on ultracold atoms and molecules allows us to fulfill this lofty goal.

3. Universal Relations between Two-Body Loss Rate and Other Quantities

It has been well-known that dilute quantum systems are governed by the so-called contacts through universal relations between different physical quantities. By “dilute”, it means that the range of interactions between any two particles, which is denoted by r_0 , is much smaller than the average inter-particle distance, which is characterized by the inverse of the Fermi momentum k_F , that is, $r_0 \ll k_F^{-1}$, as shown in **Figure 3**. Such a length scale separation leads to a universal asymptotic form of the many-body wavefunction, whose short-distance is only determined by two-body physics no matter how complex the many-body correlations might be. Such an asymptotic form of the many-body wavefunction thus establishes contacts as the central quantity to fully capture the many-body correlations of the many-body system. As $a_l \rightarrow \infty$, three-body recombination may become important and a three-body length scale needs to

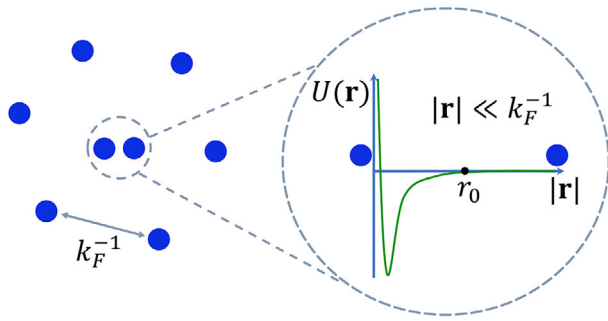


Figure 3. A length scale separation in dilute quantum systems. The blue solid spheres represent identical particles. Inside the dashed circle are two particles with the relative distance much smaller than the average inter-particle distance. The enlarged plot of the dashed circle is a schematic of the two-body collisions. The green solid curve depicts the short-range interaction $U(r)$ between these two particles.

be taken into account. In order to incorporate such three-body effects, three-body contacts have been theoretically studied and also measured in experiments.^[90,91,123,124] In this section, we will first introduce the concept of contacts and universal relations in lossless quantum systems and then generalize it to the system with two-body losses. A universal relation between two-body loss rate and contacts is first given for s -wave scatterings and then for high partial wave scatterings subsequently. Moreover, we present a number of universal relations to directly connect two-body losses to other physical quantities, such as the momentum distribution and density correlation functions. The temperature dependence of the loss rate will be presented as well.

3.1. Contacts in Lossless Systems

3.1.1. s -Wave Contacts

s -wave contact and universal relations (also known as Tan relations) were first discovered by Shina Tan in 2005.^[79–81] In his original papers, a two-component Fermi gas was considered. At low temperatures, the s -wave scattering dominates. The many-body wavefunction must behave like $\Psi \rightarrow G(1/r - 1/a_0)$ when two particles with different spins approach each other, that is, r , the distance between these two particles, is much smaller than k_F^{-1} . G , as a function of the center of mass of these two chosen particles and all other particles, captures all many-body effects. Based on this consideration, Tan derived a number of universal relations such as

$$n_\sigma(\mathbf{k}) \rightarrow \frac{C}{k^4}, \quad k \rightarrow \infty \quad (6)$$

$$\frac{dE}{d(-1/a_0)} = \frac{\hbar^2 V}{4\pi M} C \quad (7)$$

$$E = \sum_{\mathbf{k}\sigma} \frac{\hbar^2 k^2}{2M} \left(n_\sigma(\mathbf{k}) - \frac{C}{k^4} \right) + \frac{\hbar^2 VC}{4\pi a_0 M} \quad (8)$$

where $\sigma = \uparrow, \downarrow$ represents the pseudospin of two-component fermions, V is the volume of the system, M is the mass of each

fermion, E is the total energy of the system, \mathbf{k} is the momentum, $k = |\mathbf{k}|$, C is the s -wave contact density, and $C = VC$ defines the total contact. As seen from these universal relations, the momentum distribution of each component has a power-law asymptotic behavior at large k , with its amplitude as the contact density. Equation (7) is called the adiabatic relation and shows that the first derivative of the total energy with respect to the inverse of scattering length is also determined by contacts. Though the total kinetic energy appears divergent, that is, $\sum_{\mathbf{k}\sigma} [\hbar^2 k^2 n_\sigma / (2M)] \rightarrow \infty$ based on Equation (6), by subtracting C/k^4 in the summation and adding a constant proportional to the contact outside the summation, magically, one obtains the total energy of the system, as shown by the energy functional in Equation (8). Without loss of generality, we have considered a sum of discrete \mathbf{k} in Equation (8) for a finite system. When the size of the system approaches infinity, the sum will be replaced by an integral over \mathbf{k} . The energy E on the right-hand side of Equation (8) is the total internal energy of the system, regardless of whether bound states of molecules exist, since the universal asymptotic behavior of the many-body wave function $\Psi \rightarrow G(1/r - 1/a_0)$ applies to a generic case, in which there may or may not be bound states of molecules. Whereas Tan relations were derived originally by a novel mathematical method, they were obtained using some other methods and generalized to other universal relations,^[82,83] and were verified in experiments.^[84,85,91,92] Contacts and Tan relations have also been generalized to low dimensions and multi-component systems and have been widely used in ultracold atomic physics^[86–90,93–98] and related fields.^[99–101]

3.1.2. High Partial Wave Contacts

It was not until 2015 that the concept of contact and universal relations were generalized to high partial wave scatterings.^[102–104,106,107,110,114] At low temperatures, s -wave scatterings are typically more important than high partial wave scatterings because of the absence of the centrifugal barrier. Nevertheless, near the Feshbach resonance of a certain high partial wave scattering, contacts of this partial wave will be dominant. For simplicity, we consider single component bosons (or fermions) in a single partial wave scattering channel here. The Hamiltonian of N bosons (or fermions) in lossless systems is written as

$$H = \sum_{i=1}^N \left[-\frac{\hbar^2}{2M} \nabla_i^2 + V_{\text{ext}}(\mathbf{r}_i) \right] + \sum_{i>j} U(\mathbf{r}_i - \mathbf{r}_j) \quad (9)$$

where \mathbf{r}_i is spatial coordinate of the i -th particle, $V_{\text{ext}}(\mathbf{r}_i)$ is the external potential, and $U(\mathbf{r}_i - \mathbf{r}_j)$ is a two-body interaction that vanishes beyond a characteristic length scale r_0 , as shown in Figure 3. When the distances between any two particles are much smaller than k_F^{-1} , as the probability of finding more than two particles at short distance is negligible, the many-body wavefunction has the asymptotic form

$$\Psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N) \xrightarrow{r_{ij} \ll k_F^{-1}} \sum_{\epsilon} \psi_{lm}(\mathbf{r}_{ij}; \epsilon) G_{lm}(\mathbf{R}_{ij}; E - \epsilon) \quad (10)$$

where $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ and $r_{ij} = |\mathbf{r}_{ij}|$ denotes the distance between the i -th and the j -th particles. As a short-hand notation, $\mathbf{R}_{ij} = \{(\mathbf{r}_i + \mathbf{r}_j)/2, \mathbf{r}_{k \neq i,j}\}$ is a set of parameters including the center of mass coordinate of the i -th and the j -th particles and the coordinates of all the other $N - 2$ particles. ϵ is the two-body collision energy. l and m are the angular momentum quantum numbers. For instance, $l = 1, m = 0, \pm 1$ for p -wave scatterings. $\psi_{lm}(\mathbf{r}_{ij}; \epsilon) = \varphi_{lm}(r_{ij}; \epsilon) Y_{lm}(\hat{\mathbf{r}}_{ij})$ is the two-body wavefunction determined solely by the two-body Hamiltonian, $H_2 = -(\hbar^2/M)\nabla^2 + U(\mathbf{r}_{ij})$. All many-body effects are included in $G_{lm}(\mathbf{R}_{ij}; E - \epsilon)$, which depends on the center of mass motion of the i -th and the j -th particles and the motion of all other $N - 2$ particles. G_{lm} can be viewed as a “normalization factor” of the two-body wavefunction $\psi_{lm}(\mathbf{r}_{ij}; \epsilon)$ in a many-body environment. In the most generic case, where multiple partial wave scatterings exist, a sum over l and m needs to be included in Equation (10). Near the Feshbach resonance of a particular partial wave, it is sufficient to keep only the dominant channel so as to keep notations simple. Though the full expression of $\psi_{lm}(\mathbf{r}_{ij}; \epsilon)$ requires the detailed information of $U(\mathbf{r}_{ij})$, due to a finite range beyond which $U(\mathbf{r}_{ij})$ vanishes, it has the universal form

$$\varphi_{lm}(r_{ij}; \epsilon) \xrightarrow{r_0 < r_{ij} \ll k_F^{-1}} \frac{q_\epsilon^{l+1}}{\tan \eta_l} [j_l(q_\epsilon r_{ij}) - \tan \eta_l n_l(q_\epsilon r_{ij})] \quad (11)$$

where $q_\epsilon = (M\epsilon/\hbar^2)^{1/2}$, $j_l(x)$ ($n_l(x)$) is the spherical Bessel function of the first (second) kind, η_l is the phase shift that can be expanded in the low energy limit as $q_\epsilon^{2l+1} \cot \eta_l = -1/a_l + r_l^e q_\epsilon^2 + O(q_\epsilon^4)$, where a_l and r_l^e are the scattering length and the effective range, respectively. As a result, $\varphi_{lm}(r_{ij}; \epsilon) = \varphi_{lm}^{(0)}(r_{ij}) + \varphi_{lm}^{(1)}(r_{ij})q_\epsilon^2 + O(q_\epsilon^4)$, where

$$\varphi_{lm}^{(0)}(r_{ij}) \xrightarrow{r_0 < r_{ij} \ll k_F^{-1}} \frac{(2l-1)!!}{r_{ij}^{l+1}} - \frac{1}{a_l} \frac{r_{ij}^l}{(2l+1)!!} \quad (12)$$

$$\varphi_{lm}^{(1)}(r_{ij}) \xrightarrow{r_0 < r_{ij} \ll k_F^{-1}} r_l^e \frac{r_{ij}^l}{(2l+1)!!} + \frac{1}{a_l} \frac{r_{ij}^{l+2}}{2(2l+3)!!} + \frac{(2l-3)!!}{2r_{ij}^{l-1}} \quad (13)$$

Taking Equations (10), (12), and (13) as the starting point, a number of universal relations can be derived. Some of them are listed as follows.

Momentum Distribution: Based on the definition of the momentum distribution $n(\mathbf{k}) = \sum_{i=1}^N \int \prod_{j \neq i} d\mathbf{r}_j \int d\mathbf{r}_i \Psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N) \exp(-i\mathbf{k} \cdot \mathbf{r}_i)^2$, one obtains

$$n(\mathbf{k}) \xrightarrow{k_F \ll k \ll r_0^{-1}} \left[C_{lm} k^{2l-4} + \sum_{s=1}^l (C_{lm}^s + D_{lm}^s) k^{2l-4-2s} \right] |Y_{lm}(\hat{\mathbf{k}})|^2 \quad (14)$$

The contacts $\{C_{lm}, C_{lm}^s\}$ are defined as

$$C_{lm} = (4\pi)^2 N(N-1) \int d\mathbf{R}_{ij} |g_{lm}^{(0)}|^2 \quad (15)$$

$$C_{lm}^s = (4\pi)^2 N(N-1) \sum_{i=0}^s \int d\mathbf{R}_{ij} g_{lm}^{(i)*} g_{lm}^{(s-i)} \quad (16)$$

where $d\mathbf{R}_{ij} = d[(\mathbf{r}_i + \mathbf{r}_j)/2] \prod_{k \neq i,j} d\mathbf{r}_k$ and $g_{lm}^{(s)} = \sum_\epsilon q_\epsilon^{2s} G_{lm}(\mathbf{R}_{ij}; E - \epsilon)$. D_{lm}^s , on the other hand, are contacts originated from the center of mass of two particles and vanish when the center of mass momentum of the two particles approaches zero. More detailed discussions of D_{lm}^s can be found elsewhere.^[103,105,112,114]

Adiabatic Relations: The adiabatic relations for the l -th partial wave scatterings are expressed as^[102,103]:

$$\frac{dE}{d(-1/a_l)} = \frac{\hbar^2 C_{lm}}{(4\pi)^2 2M} \quad (17)$$

$$\frac{dE}{dr_l^e} = \frac{\hbar^2 C_{lm}^1}{(4\pi)^2 4M} \quad (18)$$

Compared to s -wave scatterings, there is an additional adiabatic relation related to the effective range, which cannot be ignored for any finite l .

Radiofrequency Spectroscopy: A radiofrequency (RF) field can be used to transfer atoms to another empty hyperfine spin state. While the RF spectroscopy can also be applied to molecules, studies of universal relations about RF spectroscopy have been focusing on atoms. Here, it is useful to briefly survey the main results of such universal relations about RF spectroscopy of atoms. When the RF Rabi frequency Ω_{RF} is small such that the coupling can be regarded as a perturbation, the transition rate has an asymptotic form for large RF frequency ω ,

$$\Gamma_{\text{RF}}(\omega) \rightarrow 2\pi \hbar \Omega_{\text{RF}}^2 \sum_{\mathbf{k}} n(\mathbf{k}) \delta(\hbar\omega - \hbar^2 k^2/M) \quad (19)$$

according to Fermi's Golden rule. Thus, when ω is much larger than all the other relevant frequencies in the many-body systems, the transition rate has a power-law asymptotic behavior stated as^[103,107]

$$\Gamma_{\text{RF}} \rightarrow \frac{\Omega_{\text{RF}}^2 MV}{8\pi^2 \hbar} [C_{lm} (M\omega/\hbar)^{l-3/2} + (C_{lm}^s + D_{lm}^s) (M\omega/\hbar)^{l-5/2} + \dots] \quad (20)$$

where V is the volume of the system.

Density Correlation Function: Another physical quantity that is important in AMO and condensed matter physics is the density correlation function, $S(\mathbf{r}_{ij}) = \int d\mathbf{R}_{ij} \langle n(\mathbf{r}_i) n(\mathbf{r}_j) \rangle$. Based on Equations (10), (12), and (13), $S(\mathbf{r})$ has the explicit form in the regime, $r_0 < |\mathbf{r}_{ij}| \ll k_F^{-1}$. One obtains

$$\begin{aligned} S(\mathbf{r}_{ij}) \rightarrow \frac{|Y_{lm}(\hat{\mathbf{r}}_{ij})|^2}{(4\pi)^2} & \left\{ C_{lm} \frac{[(2l-1)!!]^2}{r_{ij}^{2l+2}} + C_{lm}^1 \frac{(2l-3)!!(2l-1)!!}{2r_{ij}^{2l}} \right. \\ & + (r_l^e C_{lm}^1 - \frac{2}{a_l} C_{lm}) \frac{1}{(2l+1)r_{ij}} - \frac{2}{a_l} C_{lm}^1 \frac{(2l-3)!!}{(2l+3)!!} r_{ij} \\ & \left. + (\frac{1}{a_l^2} C_{lm} - \frac{r_l^e}{a_l} C_{lm}^1) \frac{r_{ij}^{2l}}{[(2l+1)!!]^2} - \frac{1}{a_l^2} C_{lm}^1 \frac{r_{ij}^{2l+2}}{2(2l+1)!!(2l+3)!!} \right\} \quad (21) \end{aligned}$$

Discussions about other universal relations such as the energy functional, the pressure relation, the virial theorem, and the

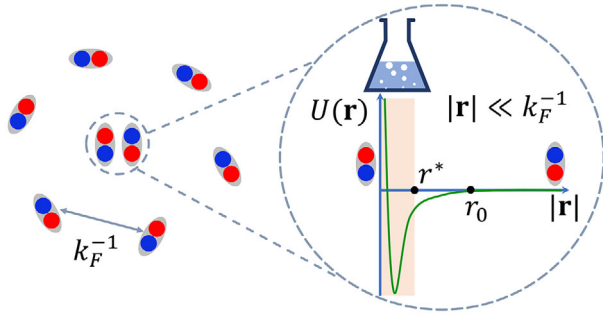


Figure 4. A length scale separation in dilute reactive molecules. The blue and red solid spheres represent two species of atoms. The distance between the circled two molecules is much smaller than the average interparticle distance, k_F^{-1} . The enlarged plot of the regime inside the dashed circle depicts the chemical reaction. The real part of the interaction, $U_R(r)$, is presented by the green solid curve. The imaginary part of the interaction, $U_I(r)$, is finite only in the shaded area, where the reaction takes place. Reproduced with permission.^[78] Copyright 2020, American Association for the Advancement of Science.

photoassociation rate can be found elsewhere.^[102,103,106,107] Since these universal relations apply to any many-body eigenstates, a thermal average does not change the format of these relations. For systems with mixed partial wave scatterings, to fully characterize the pairwise correlation of the system, contacts should be defined as a matrix.^[100,107,110] In superfluids, such a contact matrix is directly connected to the multiple order parameters when a superfluid includes mixed partial waves.^[107,110]

3.2. Contacts in Lossy Systems

The length scale separation in dilute systems also applies when losses exist. Contacts and universal relations can therefore be generalized to ultracold atoms and molecules with two-body inelastic losses.^[78,125–127] As an example, here, we consider the single component ultracold molecules in a single partial wave scattering channel denoted by (l, m) . To characterize losses, both non-Hermitian Hamiltonians with complex interactions and master equations can be implemented.

3.2.1. Complex Interactions

The dynamics of the reactive molecules can be described by Hamiltonian with complex short-range interactions. The many-body Hamiltonian can be written as

$$H = \sum_{i=1}^N \left[-\frac{\hbar^2}{2M} \nabla_i^2 + V_{\text{ext}}(\mathbf{r}_i) \right] + \sum_{i>j} U(\mathbf{r}_{ij}) \quad (22)$$

where $U(\mathbf{r}_{ij}) = U_R(\mathbf{r}_{ij}) + iU_I(\mathbf{r}_{ij})$ is a complex short-range interaction with a characteristic length scale, r_0 , as shown in **Figure 4**. $U(\mathbf{r}_{ij})$ is finite only when $|\mathbf{r}_{ij}| \leq r_0$. The inelastic collisions occur on an even shorter length scale, $r^* < r_0$. $U_I(\mathbf{r}_{ij}) \leq 0$ when $|\mathbf{r}_{ij}| < r^*$ and zero elsewhere. The many-body wavefunction satisfies the time-dependent Schrödinger equation

$$i\hbar \partial_t \Psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N) = H \Psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N) \quad (23)$$

For any systems that have finite sizes, there is no net current across the boundary. We obtain

$$\partial_t N = \frac{4}{\hbar} \sum_{i>j} \int d\mathbf{R}_{ij} d\mathbf{r}_{ij} U_I(\mathbf{r}_{ij}) |\Psi(\mathbf{R}_{ij}, \mathbf{r}_{ij})|^2 \quad (24)$$

which can be rewritten in the second quantization using bosonic (or fermionic) operators as

$$\partial_t N = \frac{2}{\hbar} \int dx dx' U_I(|\mathbf{x}' - \mathbf{x}|) \langle \Psi^\dagger(\mathbf{x}) \Psi^\dagger(\mathbf{x}') \Psi(\mathbf{x}') \Psi(\mathbf{x}) \rangle \quad (25)$$

3.2.2. Master Equations

Another well-established approach to capture particle losses is the quantum master equation. As we will show, it can be used to derive the non-Hermitian Hamiltonian with complex interactions. In an open quantum system, effects of the environment become critical. When the time that the environment takes to forget information acquired from the system is much shorter than characteristic time scales of the quantum system, the Markov approximation is applicable. The time evolution of ρ , the density matrix of the quantum system, obeys the Lindblad master equation

$$\hbar \frac{d\rho}{dt} = -i[H_0, \rho] + \mathcal{D}[\rho] \quad (26)$$

where H_0 is the Hamiltonian that describes the unitary time evolution of the system and the dissipator \mathcal{D} describes the dissipation due to couplings to the environment. Here, we consider losses due to inelastic collisions,

$$\begin{aligned} \mathcal{D}[\rho] = & -\frac{1}{2} \int dx_1 dx_2 \Gamma(|\mathbf{x}_1 - \mathbf{x}_2|) \\ & \times (2\Psi(\mathbf{x}_2)\Psi(\mathbf{x}_1)\rho\Psi^\dagger(\mathbf{x}_1)\Psi^\dagger(\mathbf{x}_2) - \{\Psi^\dagger(\mathbf{x}_1)\Psi^\dagger(\mathbf{x}_2)\Psi(\mathbf{x}_2)\Psi(\mathbf{x}_1), \rho\}) \end{aligned} \quad (27)$$

where $\Psi(\mathbf{x})$ is the fermionic field operator satisfying $\{\Psi(\mathbf{x}), \Psi^\dagger(\mathbf{x}')\} = \delta(\mathbf{x} - \mathbf{x}')$. $\Gamma(|\mathbf{x}_1 - \mathbf{x}_2|)$ describes a finite range dissipation that is finite only at short distances. The loss rate of the total particle number, $dN/dt = \int dx (d/dt) \text{Tr}(n(\mathbf{x})\rho)$ and $n(\mathbf{x}) = \Psi^\dagger(\mathbf{x})\Psi(\mathbf{x})$, is written as

$$\frac{dN}{dt} = \frac{2}{\hbar} \int dx dx' \Gamma(|\mathbf{x}' - \mathbf{x}|) \langle \Psi^\dagger(\mathbf{x}) \Psi^\dagger(\mathbf{x}') \Psi(\mathbf{x}') \Psi(\mathbf{x}) \rangle \quad (28)$$

This equation works for any finite range dissipators.

Comparing Equations (28) and (25) in the previous section, we conclude that these two approaches provide us with the same result of the decay rate if we identify U_I and Γ , that is, $U_I(|\mathbf{x}' - \mathbf{x}|) = \Gamma(|\mathbf{x}' - \mathbf{x}|)$. Thus, universal relations derived from the Lindblad equation are exactly the same as those derived from the non-Hermitian Hamiltonian with a complex interaction. It is worth mentioning that a non-Hermitian Hamiltonian is, in general, only an approximation of the Lindblad master equation at short times.^[128] Nevertheless, in dilute quantum systems, the probability of having more than two particles within a distance smaller than r_0 is negligible. The non-Hermitian Hamiltonian can still be

Table 1. The two-body inelastic loss rate in different dimensions. $\tilde{\varphi}_s^{(0)}(r)$ is a wave function obtained from extending the actual wave function $\varphi_s^{(0)}(r)$ outside the potential ($r > r_0$) into the regime $r < r_0$. $\mathbf{R}_{ij} = (\mathbf{R}_{ij}^x, \mathbf{R}_{ij}^z)$.

	One dimension ($s = l$)	Two dimensions ($s = l$)	Three dimensions ($s = lm$)
$\partial_t N$	$-\frac{\hbar}{M} \sum_{v=1}^3 \kappa_v C_v^{(s)}$	$-\frac{\hbar}{2\pi^2 M} \sum_{v=1}^3 \kappa_v C_v^{(s)}$	$-\frac{\hbar}{8\pi^2 M} \sum_{v=1}^3 \kappa_v C_v^{(s)}$
$C_1^{(s)}$	$4N(N-1) \int d\mathbf{R}_{ij}^z g_s^{(0)} ^2$	$(2\pi)^2 N(N-1) \int d\mathbf{R}_{ij}^z g_s^{(0)} ^2$	$(4\pi)^2 N(N-1) \int d\mathbf{R}_{ij} g_s^{(0)} ^2$
$C_2^{(s)}$	$8N(N-1) \int d\mathbf{R}_{ij}^z \text{Re}(g_s^{(0)*} g_s^{(1)})$	$2(2\pi)^2 N(N-1) \int d\mathbf{R}_{ij}^z \text{Re}(g_s^{(0)*} g_s^{(1)})$	$2(4\pi)^2 N(N-1) \int d\mathbf{R}_{ij} \text{Re}(g_s^{(0)*} g_s^{(1)})$
$C_3^{(s)}$	$8N(N-1) \int d\mathbf{R}_{ij}^z \text{Im}(g_s^{(0)*} g_s^{(1)})$	$2(2\pi)^2 N(N-1) \int d\mathbf{R}_{ij}^z \text{Im}(g_s^{(0)*} g_s^{(1)})$	$2(4\pi)^2 N(N-1) \int d\mathbf{R}_{ij} \text{Im}(g_s^{(0)*} g_s^{(1)})$
κ_1	$-\frac{M}{\hbar^2} \int_0^\infty \varphi_s^{(0)}(r) ^2 U_l(r) dr$	$-\frac{M}{\hbar^2} \int_0^\infty \varphi_s^{(0)}(r) ^2 U_l(r) r dr$	$-\frac{M}{\hbar^2} \int_0^\infty \varphi_s^{(0)}(r) ^2 U_l(r) r^2 dr$
κ_2	$-\frac{M}{\hbar^2} \text{Re}(\int_0^\infty \varphi_s^{(0)*}(r) \varphi_s^{(1)}(r) U_l(r) dr)$	$-\frac{M}{\hbar^2} \text{Re}(\int_0^\infty \varphi_s^{(0)*}(r) \varphi_s^{(1)}(r) U_l(r) r dr)$	$-\frac{M}{\hbar^2} \text{Re}(\int_0^\infty \varphi_s^{(0)*}(r) \varphi_s^{(1)}(r) U_l(r) r^2 dr)$
κ_3	$\frac{M}{\hbar^2} \text{Im}(\int_0^\infty \varphi_s^{(0)*}(r) \varphi_s^{(1)}(r) U_l(r) dr)$	$\frac{M}{\hbar^2} \text{Im}(\int_0^\infty \varphi_s^{(0)*}(r) \varphi_s^{(1)}(r) U_l(r) r dr)$	$\frac{M}{\hbar^2} \text{Im}(\int_0^\infty \varphi_s^{(0)*}(r) \varphi_s^{(1)}(r) U_l(r) r^2 dr)$
κ_1	$\text{Im}(-a_0)$	$\text{Im}(\ln(1/a_0))$	$\text{Im}(1/a_0)$
κ_1	$\text{Im}(1/a_{>0})$	$\text{Im}(1/a_{ >0})$	$\text{Im}(1/a_l)$
κ_2	$\text{Im}(-r_l^e/2)$	$\text{Im}(-r_l^e/2)$	$\text{Im}(-r_l^e/2)$
κ_3	$\int_0^{r_0} \{[\text{Im}\tilde{\varphi}_s^{(0)}(r)]^2 - [\text{Im}\varphi_s^{(0)}(r)]^2\} dr$	$\int_0^{r_0} \{[\text{Im}\tilde{\varphi}_s^{(0)}(r)]^2 - [\text{Im}\varphi_s^{(0)}(r)]^2\} r dr$	$\int_0^{r_0} \{[\text{Im}\tilde{\varphi}_s^{(0)}(r)]^2 - [\text{Im}\varphi_s^{(0)}(r)]^2\} r^2 dr$

used to evaluate the two-body decay rate in dilute systems satisfying $r_0 \ll k_F^{-1}$.

3.2.3. s-Wave Contacts

Though we focus on decay rates of a single component molecule here, it is useful to briefly survey the results of two-component fermionic gases with inelastic s-wave interactions. Braaten presented the explicit form of the universal relation of such two-body inelastic loss rate in 2010.^[125] Later on, he and his collaborators derived these results using the operator product expansion and the Lindblad equation in 2013 and 2017, respectively.^[126,127] The total numbers of these two components are denoted by N_\uparrow and N_\downarrow , respectively. Under the zero-range approximation of dissipators, $\Gamma = g\delta(\mathbf{x} - \mathbf{x}')$, Equation (28) reduces to^[127–129]

$$\frac{dN_\sigma}{dt} = \frac{2}{\hbar} g \int d\mathbf{x} \langle \Psi_\uparrow^\dagger(\mathbf{x}) \Psi_\downarrow^\dagger(\mathbf{x}) \Psi_\downarrow(\mathbf{x}) \Psi_\uparrow(\mathbf{x}) \rangle \quad (29)$$

The two-body loss rate for the spin- σ component Fermi gases is then written as

$$\frac{dN_\sigma}{dt} = -\frac{\hbar}{2\pi M} \text{Im}\left(\frac{1}{a_0}\right) C \quad (30)$$

where $C = C_{00}/(4\pi)$ is the Tan's definition of s-wave contact.

3.2.4. High Partial Wave Contacts

Universal relations for ultracold reactive molecules with inelastic high partial wave scatterings were pointed out by us in 2020.^[78] Starting from a non-Hermitian Hamiltonian in Equation (22), discussions were parallel to Section 3.1.2. As the interaction $U(\mathbf{r})$ becomes a complex potential, a_l , r_l^e , and q_e in Equations (10)–(13) become complex quantities. Using Equations (10), (22), and (23), we found that the decay of the total particle number is written as

$$\partial_t N = -\frac{\hbar}{8\pi^2 M} \sum_{v=1}^3 \kappa_v C_v^{(lm)} \quad (31)$$

where the three contacts $C_v^{(lm)}$ are defined in Table 1. As shown later, $C_1^{(lm)}$ is the same as the contact determining the leading term of the momentum distribution, which is similar to that of systems without losses.^[102–104,106] $C_{2,3}^{(lm)}$, however, are new quantities in systems that possess two-body losses. κ_v in Equation (31) are microscopic parameters that are dependent purely on the two-body physics and thus are independent of the temperature and the total particle number of the system. Their explicit expressions are given in Table 1. In contrast to κ_1 and κ_2 that can be rewritten as familiar parameters like the scattering length and the effective range, κ_3 is a new parameter emerged in the systems with a complex short-range interaction. As such, Equation (31) can be rewritten as

$$\partial_t N = -\frac{\hbar}{8\pi^2 M} \left[\text{Im}\left(\frac{1}{a_l}\right) C_1^{(lm)} - \text{Im}\left(\frac{r_l^e}{2}\right) C_2^{(lm)} + \kappa_3 C_3^{(lm)} \right] \quad (32)$$

Equations (31) and (32) apply to arbitrary partial wave scatterings and are universal in the sense that they are valid for any particle numbers, any temperatures, and any short-range interactions with arbitrary interaction strengths, as well as any real external potentials. In these expressions, $C_v^{(lm)}$ fully capture the many-body effects. While two-body physics are included in κ_v . These two equations also hold for any many-body eigenstates. In particular, a thermal average does not change their forms. Therefore, they apply to any finite temperatures, provided that the reaction rate is slower than the characteristic time scales to establish quasi-equilibrium in the many-body system. In other words, when the many-body system has a well-defined temperature at any time, $C_v^{(lm)}$ should be taken as their thermal averages.

The first term on the right-hand side of Equation (32) is consistent with the result presented in the previous section for s-wave scatterings, which considered zero-range interactions. When the range of the interaction is finite, that is, $r_0 > 0$, it is well-known that some other length scales in addition to the scattering length are required to characterize the scatterings. For instance, even for elastic high partial wave scatterings, the effective range needs to be taken into account. Here, for a generic complex short-range interaction, all three microscopic parameters are required, as

shown in Equations (31) and (32). Consequently, all three contacts are presented in the aforementioned universal relations. These contacts also show up in other universal relations including other physical observables. Therefore, Equations (31) and (32) allow us to directly correlate the two-body loss rate to a wide range of physical quantities.

Momentum Distribution: Equation (14) still applies in lossy systems. It is clear that the contact in the leading term of the momentum distribution is exactly $C_1^{(lm)}$ in Equation (31). We obtain

$$\bar{n}(k) \equiv \int d\Omega n(\mathbf{k}) \xrightarrow{k_F \ll k \ll r_0^{-1}} C_1^{(lm)} k^{2l-4} \quad (33)$$

where Ω is the solid angle.

Density Correlation Function: To enhance the signal-to-noise ratio, we define an integrated density correlation by $P(x, D) = \int_x^{x+D} d\mathbf{r} S(\mathbf{r})$, which is $S(\mathbf{r})$ integrated over a shell with inner and outer radii, x and $x + D$, respectively. We obtain

$$\begin{aligned} \frac{\partial P(x, D)}{\partial D} \Big|_{D \rightarrow 0} &\rightarrow \frac{x^2}{16\pi^2} \left\{ C_1^{(lm)} \frac{[(2l-1)!!]^2}{x^{2l+2}} + C_2^{(lm)} \frac{(2l-3)!!(2l-1)!!}{2x^{2l}} \right. \\ &+ \left[\text{Re}(r_l^e) C_2^{(lm)} - \text{Re}\left(\frac{2}{a_1}\right) C_1^{(lm)} - \text{Im}(r_l^e) C_3^{(lm)} \right] \frac{1}{(2l+1)x} \\ &- \left[\frac{2}{2l+1} \text{Re}\left(\frac{1}{a_1}\right) C_2^{(lm)} + \text{Im}\left(\frac{1}{a_1}\right) C_3^{(lm)} \right] \frac{x}{(2l-1)(2l+3)} \\ &+ \left[\frac{1}{|a_1|^2} C_1^{(lm)} - \text{Re}\left(\frac{r_l^e}{a_1^*}\right) C_2^{(lm)} + \text{Im}\left(\frac{r_l^e}{a_1^*}\right) C_3^{(lm)} \right] \frac{x^{2l}}{[(2l+1)!!]^2} \\ &\left. - \frac{1}{|a_1|^2} C_2^{(lm)} \frac{x^{2l+2}}{2(2l+1)!!(2l+3)!!} \right\} \end{aligned} \quad (34)$$

Equation (34) can be used to fit experimental data of the density correlation function, from which all three contacts, $C_{1,2,3}^{(lm)}$, as well as microscopic parameters, a_1 and r_l^e , can be obtained. Equation (34) can also be regarded as a generalization of the well-known result of using contacts to extract density correlation function in the absence of losses to systems with losses.^[79,88,103]

A unique advantage of the aforementioned universal relations is that, no matter whether the loss rate or any other physical observables, such as the momentum distribution and the density correlation functions, can be computed accurately in theories, Equations (31)–(34) are always valid. As such, they allow experimentalists to correlate the loss rate to many-body correlations at any temperatures and any interaction strength and to explore the central role of contacts in determining chemical reactions in many-body systems.

3.2.5. Temperature Dependence of The Loss Rate

As an application of the universal relations, here, we discuss the same temperature regime as that considered by Idziaszek and Julienne.^[71] For a two-body system in free space such that the center of mass and the relative motion are decoupled, ϵ in $C_v^{(lm)}$

Table 2. Analytical expressions for contacts $C_v^{(l)}$ of two particles in different limits. Line 1 is the results in the weakly interacting regime. Line 2 shows the results at resonance. Line 3 shows the outcomes for bound states. $\delta_{l,l'}$ is the Kronecker delta. Reproduced with permission.^[78] Copyright 2020, American Association for the Advancement of Science.

a_l	$C_1^{(l)}$	$C_2^{(l)}$	$C_3^{(l)}$
$a_l \rightarrow 0_{\pm}$	$4(2l+1)(4\pi)^3 q_c^{2l} a_l ^2 / V$	$2C_1^{(l)} \text{Re}(q_c^2)$	0
$a_l \rightarrow \infty$	$4(2l+1)(4\pi)^3 q_c^{2l-4} 1/r_l^e ^2 / V$	$2C_1^{(l)} \text{Re}(q_c^2)$	0
Bound	$2(4\pi)^2 \text{Re}(2\delta_{l,0}/a_0 - (1 - \delta_{l,0})/r_l^e)$	$2C_1^{(l)} \text{Re}(q_c^2)$	$2C_3^{(l)} \text{Im}(q_c^2)$

becomes a good quantum number and $G(\mathbf{R}_{ij}; E - \epsilon)$ becomes a delta function in the energy space. For scattering states with $\epsilon > 0$, the wavefunction is written as $\Psi^{(2)}(\mathbf{r}_1, \mathbf{r}_2) = \phi_c(\mathbf{R}_{12})\psi_l(\mathbf{r}_{12})$, where $\phi_c(\mathbf{R}_{12})$ is a normalized wavefunction of the center of mass and $\psi_l(\mathbf{r}_{12}) = \sqrt{8\pi/V} [i/(\cot \eta_l - i)] [\cot \eta_{jl}(q_c |\mathbf{r}_{12}|) - n_l(q_c |\mathbf{r}_{12}|)] \sum_m Y_{lm}(\hat{\mathbf{r}}_{12})$. To simplify notations, we define $C_v^{(l)} = \sum_m C_v^{(lm)}$, which is the sum of contributions from $m = -l$ to $m = l$. If we consider a scattering state, $C_3^{(l)} = 0$. In contrast, if a bound state is considered, $C_3^{(l)} = 2C_1^{(l)} \text{Im}(q_c^2)$. In both the weakly and strongly interacting regime, analytical results can be obtained, as shown in Table 2. Using the second order virial expansion, thermal averaged contacts can be derived and the p -wave results are shown in Table 3 as an example.

Using Table 3, Idziaszek and Julienne's results can be recovered. If the system is prepared at the upper branch, the bound state is not occupied and $C_3^{(1)} = 0$. In a homogeneous system, the decay rate can be written as

$$\partial_t N = \frac{144\pi^2}{h} \text{Im}(a_1) N n k_B T + \frac{360\pi^2}{h} \text{Im}\left(\frac{a_1}{a_1^*} r_1^e\right) \frac{M|a_1|^2}{\hbar^2} N n k_B^2 T^2 \quad (35)$$

If we only consider the contribution from the scattering length, the first term on the right-hand side of Equation (35) readily recovers the result obtained by Idziaszek and Julienne.^[71] The second term on the right-hand side of Equation (35) takes into account the contribution from r_1^e . This result shows that the effective range gives rise to a temperature dependence of the loss rate different from that determined by the scattering length.

In a realistic experiment, a trapping potential always exists. It has been found that the temperature dependence of the loss rate of the total particle number is different from that of a homogeneous system. The two terms become proportional to $1/\sqrt{T}$ and \sqrt{T} , respectively. Nevertheless, if one considers the decay rate of the density at the center of the trap, a temperature dependence similar to Equation (35) can be recovered.^[78] It is worth mentioning that though the NIST experiment has observed the deviation of the decay rate from the linear dependence on T with decreasing the temperature,^[63] it is unclear at the moment whether such deviation is indeed due to the correction from the effective range or some other effects, such as high order virial expansions and inhomogeneity of the dissipations in the traps.^[77]

Table 3. Analytical expressions for thermal averaged contacts $\langle C_v^{(l)} \rangle_T$ in different limits. Line 1 and 2 are the results in the weakly interacting regime. Bound states exist when a_1 is positive and their contributions are taken into consideration in Line 1 as well. Line 3 shows the results at resonance. N_D represents the number of dimers. $\lambda_T = [(2\pi\hbar^2)/(Mk_B T)]^{1/2}$ is the thermal wave length. Reproduced with permission.^[78] Copyright 2020, American Association for the Advancement of Science.

a_1	$\langle C_1^{(l)} \rangle_T$	$\langle C_2^{(l)} \rangle_T$	$\langle C_3^{(l)} \rangle_T$
$a_1 \rightarrow 0_+$	$72(2\pi)^4 a_1 ^2 \lambda_T^{-2} nN$ $+2(4\pi)^2 \text{Re}(-1/r_1^e) N_D$	$360(2\pi)^5 a_1 ^2 \lambda_T^{-4} nN$ $+4(4\pi)^2 \text{Re}(-1/r_1^e) \text{Re}(1/(a_1 r_1^e)) N_D$	$4(4\pi)^2 \text{Re}(-1/r_1^e) \text{Im}(1/(a_1 r_1^e)) N_D$
$a_1 \rightarrow 0_-$	$72(2\pi)^4 a_1 ^2 \lambda_T^{-2} nN$	$360(2\pi)^5 a_1 ^2 \lambda_T^{-4} nN$	0
$a_1 \rightarrow \infty$	$24(4\pi)^2 1/r_1^e ^2 \lambda_T^2 nN$	$12(4\pi)^3 1/r_1^e ^2 nN$	0

Table 4. The low energy expansion of phase shift η_l in different dimensions.^[131]

	One dimension	Two dimensions	Three dimensions
$l = 0$	$q_e \tan \eta_0 = \frac{1}{a_0}$	$\frac{\pi}{2} \cot \eta_0 = \ln(\frac{q_e a_0}{2} e^\gamma)$	$q_e \cot \eta_0 = -\frac{1}{a_0} + r_0^e q_e^2$
$l = 1$	$q_e \cot \eta_1 = -\frac{1}{a_1} + r_1^e q_e^2$	$\frac{\pi}{2} [q_e^2 \cot \eta_1 - q_e^2 \ln(\frac{q_e r_0}{2} e^{\gamma-1/2})] = -\frac{1}{a_1} + r_1^e q_e^2$	$q_e^3 \cot \eta_1 = -\frac{1}{a_1} + r_1^e q_e^2$
$l > 1$		$\frac{\pi}{2} q_e^{2l} \cot \eta_l = -\frac{1}{a_l} + r_l^e q_e^2$	$q_e^{2l+1} \cot \eta_l = -\frac{1}{a_l} + r_l^e q_e^2$

4. Conclusions and Outlook

We have seen that ultracold molecules in the quantum degenerate regime provide physicists and chemists an ideal platform to explore how two-body losses are influenced by a quantum many-body environment. Universal relations established by contacts lead to a new perspective and a unique tool to answer this challenging question. It is expected that very rich results can be obtained along this direction that bridges AMO physics, condensed matter physics, and quantum chemistry. Here, we provide a brief outlook from our own perspectives.

In experiments, a strong external confinement may exist along the transverse directions. Molecules are then prepared in quasi-1D or quasi-2D traps. It will be interesting to study the two-body inelastic loss rate in low dimensions. Discussions in Section 3.2.4 can be straightforwardly generalized to strictly one or two dimensions. Since Equations (10) and (24) still apply in low dimensions and the only difference comes from the expressions of the two-body wavefunctions, we obtain

$$\psi_l(\rho_{ij}; \epsilon) \xrightarrow{r_0 < \rho_{ij} \ll k_F^{-1}} \frac{\pi}{2} \frac{q_e^l}{\tan \eta_l} [J_l(q_e \rho_{ij}) - \tan \eta_l N_l(q_e \rho_{ij})] Y_l(\hat{\rho}) \quad (36)$$

in two dimensions (2D) and

$$\psi_0(z_{ij}; \epsilon) \xrightarrow{r_0 < z_{ij} \ll k_F^{-1}} \frac{\cot \eta_0}{q_e} [\cos(q_e |z_{ij}|) - \tan \eta_0 \sin(q_e |z_{ij}|)] \quad (37)$$

$$\psi_1(z_{ij}; \epsilon) \xrightarrow{r_0 < z_{ij} \ll k_F^{-1}} [\cot \eta_1 \sin(q_e |z_{ij}|) + \cos(q_e |z_{ij}|)] \frac{z_{ij}}{|z_{ij}|} \quad (38)$$

in one dimension (1D), where $\mathbf{r}_{ij} = (\rho_{ij}, z_{ij})$. $Y_l(\hat{\rho}) = [(x + iy)/\rho]^l / \sqrt{2\pi}$ is the generalized spherical harmonics in 2D. J_l (N_l) is the Bessel function of the first (second) kind. The low energy expansion of η_l is presented in Table 4. The decay of the total particle number in 2D is captured by

$$\partial_t N = -\frac{\hbar}{2\pi^2 M} \text{Im}(\ln \frac{1}{a_0}) C_1^{(0)} \quad (39)$$

for s-wave scatterings and

$$\partial_t N = -\frac{\hbar}{2\pi^2 M} \left[\text{Im}(\frac{1}{a_1}) C_1^{(l)} - \text{Im}(\frac{r_1^e}{2}) C_2^{(l)} + \kappa_3 C_3^{(l)} \right] \quad (40)$$

for high partial wave scatterings. The explicit expressions of $C_v^{(l)}$ and κ_3 are given in Table 1. Similarly, the decay of the total particle number in 1D is captured by

$$\partial_t N = \frac{\hbar}{M} \text{Im}(a_0) C_1^{(0)} \quad (41)$$

for even wave scatterings and

$$\partial_t N = -\frac{\hbar}{M} \left[\text{Im}(\frac{1}{a_1}) C_1^{(1)} - \text{Im}(\frac{r_1^e}{2}) C_2^{(1)} + \kappa_3 C_3^{(1)} \right] \quad (42)$$

for odd wave scatterings. The explicit expressions of $C_v^{(l)}$ and κ_3 are given in Table 1.

The strength of the transverse confinement is actually a tuning parameter in experiments. With gradually increasing the transverse confinement, the system can be tuned from three dimensions (3D) to 2D or 1D. Whereas the dependence of the loss rate on the transverse confinement has been observed in experiments,^[68] such a dimensional crossover problem has not been studied in theory yet. It will be interesting to explore the full expressions of the universal relations in strongly anisotropic 3D traps that are beyond the scope of expressions in strictly 1D and 2D. Such universal relations in dimensional crossover are expected to provide us with new aspects of chemical reactions across different dimensions.

The electric field can be turned on or off in experiments.^[63,67] If the electric field is absent, molecules are unpolarized and interact with each other by the van der Waals potential. When a finite electric field polarizes molecules, the dipole-dipole interaction $\sim A/|\mathbf{r}|^3$ needs to be considered. Such a power-law potential

has a characteristic length, $\tilde{r} = M|A|/\hbar^2$ and the scattering theory applies.^[130] As $k_F\tilde{r} \ll 1$ can be used to define a dilute system, universal relations still arise in lossless systems. When losses exist, it will be interesting to study universal relations in the presence of the electric field induced dipole–dipole interactions, from which new insights about how the electric field may control the decay rate of molecules can be obtained.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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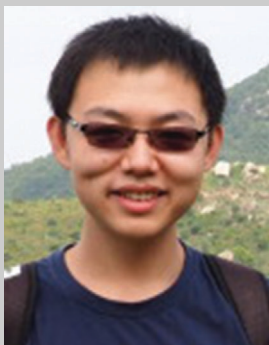
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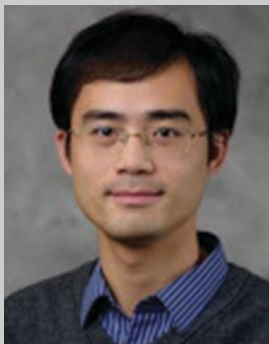
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