Universal Four-Boson States in Ultracold Molecular Gases: Resonant Effects in Dimer-Dimer Collisions

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We study the manifestations of universal four-body physics in ultracold dimer-dimer collisions. We show that resonant features associated with three-body Efimov physics and dimer-dimer scattering lengths are universally related. The emergence of universal four-boson states allows for the tunability of the dimer-dimer interaction, thus enabling the future study of ultracold molecular gases with both attractive and repulsive interactions. Moreover, our study of the interconversion between dimers and Efimov trimers shows that $B_2 + B_2 \rightarrow B_3 + B$ rearrangement reactions can provide an efficient trimer formation mechanism. Our analysis of the temperature dependence of this reaction provides an interpretation of the available experimental data and sheds light on the possible experimental realization of rearrangement processes in ultracold gases.

PACS numbers: 31.15.xj, 21.45.-v, 34.50.-s, 34.50.Cx, 67.85.-d

In both nuclear and atomic physics, the simplest examples of quantum halo states are weakly bound dimers, with large radii extending well into classically forbidden regions. A remarkable consequence of their large size is that quantum-halo dimers obey universal scaling laws, i.e., many of their properties are independent of the details of their short-range interaction, typically characterized by a short length scale $r_0$. The notion of a halo state extends in a nontrivial way to more complex few-body systems, of which three-body Efimov states are the most prominent example. Recently, the notion of universality has been extended to four-boson systems and a new universal picture has emerged. For each Efimov trimer precisely two four-boson states have recently been shown to exist, whose energies are universally related to the Efimov trimer energy. Thus, the four-body system inherits many of the characteristics of three-body Efimov physics, as the geometric scaling of energies and length scales. More recently, a first experimental evidence of such universal four-body states has been found in an ultracold gas of Cs atoms, through the observation of resonant losses due to four-body recombination satisfying the universal predictions of Ref. [6]. These findings, along with the rapid experimental advances in controlling few-body correlations in ultracold quantum gases, magnify the importance of four-body scattering processes. They offer a path for the observation of universal physics and can potentially enrich the range of experimentally accessible phenomena.

In this Letter, we explore dimer-dimer scattering processes and analyze the consequences of the recently-discovered universal properties of four-boson systems. Our interest concentrates on such processes near a Feshbach resonance, where, through application of an external magnetic field, the $s$-wave two-body scattering length $a$ can be tuned from $-\infty$ to $+\infty$. Much of the recent progress in understanding dimer-dimer correlations in ultracold physics has been devoted to dimers formed from fermionic atoms. For bosons, the relatively simple fermionic few-body physics is replaced by a much more complex structure involving multiple few-body halo states that can strongly affect dimer-dimer correlations and the collisional behavior of entire gas. Here we advocate that ultracold molecular gases are perhaps the best candidates for exploring universal four-body physics at its full complexity. In fact, the search has already begun. In Ref. [11], Ferlaino et al. formed an ultracold sample of Cs$_2$ dimers and found by varying $a$ that the loss coefficient exhibits a pronounced minimum which allows for longer lifetimes. The temperature dependence of the loss rate was also found to display an intriguing behavior, deviating from the expected Wigner threshold law.

In the present exploration of four-boson universal physics we find that, in contrast to the fermionic case where dimers interact repulsively, bosonic dimers display resonance effects caused by four-body states. Near such resonances, the dimer-dimer scattering length $a_{dd}$ can vary from $-\infty$ to $+\infty$, thereby resulting effectively in either attractive or repulsive interactions. Our studies of dimer-trimer conversion, similar in spirit with the one in Ref. [13], further indicate that a molecular gas can be used to form one of Efimov trimers. We find that when the atom-trimer collision threshold becomes nearly degenerate with the dimer-dimer threshold, most of the molecular losses are due to $B_2 + B_2 \rightarrow B_3 + B$ rearrangement scattering. Because of the vanishingly small kinetic energy released in such process, it allows trimers to remain trapped. Here we show that the temperature dependence of this rearrangement reaction rate can partially explain some recent experimental observations by Ref. [10], which indicate the existence of a trimer state just above the dimer-dimer collision threshold.

We study dimer-dimer processes using the adiabatic
hyperspherical representation for the four-body problem [14], which offers a simple and conceptually clear description of the bound and scattering properties. Here, the Schrödinger equation reduces to a simple system of ordinary differential equations given by,
\[ \left[ -\frac{\hbar^2}{2\mu} \frac{d^2}{dR^2} + W_\nu \right] F_\nu + \sum_{\nu' \neq \nu} W_{\nu\nu'} F_{\nu'} = EF_\nu, \] (1)
where the hyperradius \( R \) describes the overall size of the system, and \( \nu \) is a collective index that represents all quantum numbers necessary to label each channel. In Eq. (1), \( \mu \) is the four-body reduced mass, \( E \) is the total energy, and \( F_\nu \) is the hyperradial wave function. \( W_{\nu\nu'}(R) \) are the nonadiabatic couplings that drive inelastic transitions between channels and \( W_\nu(R) \) are effective potentials that support bound and quasi-bound states dictating many of the scattering properties of the system.

Figure 1(b) illustrates the resonant effects possible in dimer-dimer collisions (\( a > 0 \)). In general, one expects pronounced features in ultracold scattering observables whenever a bound state crosses, or emerges from, the collision threshold, or when an additional decay channel becomes energetically available. In Fig. 1(b), for instance, as \( a \) increases and approach \( a_{dd}^* \), where an Efimov trimer is created and causes a divergence in the atom-dimer scattering length \( a_{dd} \), an infinite number of four-boson states emerge potentially causing resonant effects in ultracold atom-molecule gas mixtures [8]. For a pure molecular gas, however, it is primarily collisions of the type \( B_2 + B_2 \) that provide the pathway to detailed study of four-body physics. As Fig. 1(b) shows, there exist precisely two four-boson states emerging from the \( B_2 + B_2 \) threshold at two specific values of \( a \), which we denote as \( a_{dd,i}^* \) (where \( i = 1 \) and \( 2 \)), causing \( a_{dd} \) to diverge at values [13]. In the vicinity of \( a_{dd,i}^* \), therefore, the dimer-dimer interaction can be tuned from attractive, \( a_{dd} < 0 \), to repulsive, \( a_{dd} > 0 \), irrespective of the effectively repulsive character (\( a > 0 \)) of the interatomic interactions. At \( a = a_{dd}^* \), however, a different process takes place. Here, the \( B_2 + B_2 \) and \( B_3 + B_3 \) channels become degenerate [see Fig. 1(b)] and one can expect an enhancement of the \( B_2 + B_2 \rightarrow B_3 + B \) rearrangement reaction rate over other dimer loss processes.

A key property resulting from the universality in the four-boson problem is that the aforementioned dimer-dimer resonances and rearrangement reaction are universally related to the three-body Efimov physics. The values for \( a_{dd,i}^* \) and \( a_{dd}^* \) are controlled by a single three-body parameter, \( a_{ad}^* \). We, therefore, express our results in terms of the universal ratios \( a_{dd,i}^*/a_{ad}^* \) and \( a_{dd}^*/a_{ad}^* \), which determine the positions of the dimer-dimer resonances, \( a_{dd,i}^* \), and the critical scattering length \( a_{ad}^* \), at which the \( B_2 + B_2 \rightarrow B_3 + B \) rearrangement reaction is enhanced. In practice, our calculations extract the values for such ratios from the energies in the region near the third Efimov state [see Fig. 1(b)] where the states are largely unaffected by nonuniversal short-range physics. We obtain
\[ \frac{a_{dd,1}^*}{a_{ad}^*} \approx 2.37, \quad \frac{a_{dd,2}^*}{a_{ad}^*} \approx 6.6, \quad \text{and} \quad \frac{a_{dd}^*}{a_{ad}^*} \approx 6.73. \] (2)

We expect that such ratios are universal within an error of less than 2%, as determined by comparison of our value for \( a_{dd}^*/a_{ad}^* \) to the value obtained using the semi-analytical results from Ref. [7].

In Fig. 2 we show our numerical calculations for \( a_{dd} \) and for the dimer-dimer relaxation rate, \( V_{rel}^{dd} \), calculated using:
\[ a_{dd} = - \frac{\text{Re}[\tan \delta_{dd}]}{k} \quad \text{and} \quad V_{rel}^{dd} = \frac{8\hbar}{mk} \sum_f |S_{fplane}^f|^2, \] (3)
where \( k^2 = 2mE_{col}/\hbar \), with \( E_{col} = E - 2E_b \) being the collision energy, and \( \exp(2i\delta_{dd}) = S_{dd,dd} \). The S-matrix

FIG. 1: (color online). (a) Spectrum of the four-boson system from our numerical calculations [8] and (b) a schematic representation of the region important for dimer-dimer collisions (see text). The four-boson states are represented by black solid lines, while the collision thresholds are represented by solid red lines for the atom-atom-dimer and dimer-dimer collisions, and by green dashed lines for atom-trimer collisions.

Figure 1(a) shows the spectrum for the two-, three- and four-boson systems from our numerical study [8], and Fig. 1(b) shows a schematic representation of the region marked by a circle in Fig. 1(a). [Note that we use the function \( F(x) \equiv \text{sgn}(x) \ln(1 + |x|) \) to visualize the whole energy landscape.] The black solid lines are the energies of the four-boson states, the red-solid lines represent the dimer-dimer (lower) and dimer-atom-atom (upper) collision thresholds, and the green-dashed lines are the Efimov trimer energies, or more precisely, the atom-trimer collision thresholds. The structure shown in Fig. 1(b) repeats every time \( a \) increases by the Efimov geometric factor \( e^{7/50} \approx 22.7 \), reflecting the pervasive influence of Efimov physics on the four-boson properties.
elements are obtained from the solutions of Eq. (11). In our calculations, the only decay channel is the one associated with the lowest (relatively deep) Efimov state. We, therefore, do not take into account the decay into deeply bound two-body states [12], but we still expect the general aspects of our results to remain valid.

Figure 2 shows our numerical results, with the atomic mass chosen to be that of Cs, and \( a_{\text{ad}} = 400 \) a.u., accordingly with the observations of Ref. [9], where \( a_{\text{ad}} = 400 \) a.u. Figure 2(a) then shows the dimer-dimer resonances caused by the four-boson states shown in Fig. 1(b). We note, however, that from Fig. 2(a) we obtain \( a_{\text{ad}}^{*}/a_{\text{ad},1} \approx 3.57 \), which deviates from our predicted values from Eq. (2), \( a_{\text{ad}}^{*}/a_{\text{ad},1} \approx 2.84 \), in about 20%. These deviations are produced by finite-range effects that introduce nonuniversal corrections [18] to the energies of the two-, three- and four-body systems in Fig. 1(b). Figure 2(b) shows our results for the thermally averaged \( V_{\text{rel}}^{dd} \). It clearly exhibits an enhancement of the inelastic loss at \( a_{\text{ad},1}^{*} \), corresponding to the dimer-dimer resonance discussed above. Near \( a_{\text{ad},2,3}^{*} \) (\( \approx a_{\text{ad}}^{*} \)), however, the resonance effect is masked by an enhanced rate for the rearrangement reaction \( B_2 + B_2 \rightarrow B_3 + B \). In fact, our calculations show an efficiency of about 98% for the \( B_2 + B_2 \rightarrow B_3 + B \) reaction. In the presence of deeply bound two-body states, we still expect a high dimer-trimer conversion efficiency because of the stronger overlap between these states than with other deeply bound channels. Therefore, the dominance of the \( B_2 + B_2 \rightarrow B_3 + B \) reaction, and its negligible energy released, makes this process promising for the formation of Efimov trimers which can (under certain conditions) remain trapped. In a pure molecular sample, a clear experimental signature of such rearrangement reactions, and therefore of trimer formation, would simply be achieved by detecting the reappearance of atoms for \( a > a_{\text{ad}}^{*} \). The lifetime of such trimer states can, however, be a critical issue, since it depends on its intrinsic decay rate [13, 16] and on collisions with other atoms.

Our results can be compared to the experimental data for Cs\(_2\) of Ref. [10] in order to understand the observed scattering length and temperature dependences. In Ref. [10] it was found that \( V_{\text{rel}}^{dd} \) exhibits a minimum for relatively large values of \( a \), and that for the largest values of \( a \) it was found that \( V_{\text{rel}}^{dd} \) increases with the temperature, in contrast to the constant behavior predicted by the Wigner threshold law. Based on the temperature dependence, which we discuss in details below, we believe that the minimum observed in Ref [10] in order to understand the observed scattering length and temperature dependences. In Ref. [10] it was found that \( V_{\text{rel}}^{dd} \) exhibits a minimum for relatively large values of \( a \), and that for the largest values of \( a \) it was found that \( V_{\text{rel}}^{dd} \) increases with the temperature, in contrast to the constant behavior predicted by the Wigner threshold law. Based on the temperature dependence, which we discuss in details below, we believe that the minimum observed in Ref [10] in order to understand the observed scattering length and temperature dependences.
edge of the spectrum for Cs$_2$ [20], this new energy scale can be associated with a trimer state, allowing for the enhancement of the $B_1 + B_2 \rightarrow B_2 + B$ reaction as the temperature is increased [Fig. 2(b)].

In our model, therefore, the important energy scale in the system will be $\Delta = |E_{3b} - 2E_b| < E_b$, associated with the energy of the trimer state. For $E_{col} < \Delta$, our simple model assumes a Wigner threshold law behavior, that $V_{rel}^{dd} = \hbar c_\eta/m$, where $c_\eta$ has dimensions of length. For $E_{col} > \Delta$, however, we assume that the rate is unitarity-limited, $V_{rel}^{dd} = f_\eta \hbar 4\pi/(2\frac{1}{2}m^2) E_{col}^{-1/2}$, with $f_\eta \leq 1$. Its thermal average is:

$$V_{rel}^{dd}(T) = \left[ \text{Erf} \left( \frac{\Delta}{k_b T} \sqrt{\frac{J}{T}} \right) - \frac{\Delta}{k_b T} \frac{2}{\pi} e^{-\Delta/k_b T} \right] \frac{\hbar c_\eta}{m}$$

$$+ \frac{8\hbar \sqrt{2\pi}}{m^{3/2}(k_b T)^{1/2}} f_\eta,$$  (4)

where $k_b$ is the Boltzmann’s constant. The inset of Fig. 3 shows a comparison between Eq. (4) and our numerical calculations for $a < a^{\ast}_dd$ and establishes the validity of our analytical model. We then use Eq. (4) to fit the experimental data shown in Fig. 3 and find excellent agreement. However, since our model does not specify the nature of the trimer state, we can not determine if the temperature dependence in Ref. [10] is caused by an Efimov state.

In summary, we have demonstrated the universal properties of ultracold dimer-dimer collisions and determined their connection with three-body Efimov physics. The existence of universal resonances allows for the tunability of the dimer-dimer interaction and opens up the possibility of study molecular gases with attractive and repulsive interactions. Our numerical results show that ultracold molecular gases might offer an efficient path for the creation of a gas of Efimov trimers via $B_2 + B_2 \rightarrow B_3 + B$ rearrangement reactions. In particular, the observation of resonant effects in dimer-dimer collisions obeying the threshold law where $a_{dd}/a^{\ast}_{dd}$ and $a^{\ast}_{dd}/a^{\ast}_{dd}$ within the limits imposed by nonuniversal corrections we discussed here, is hereby shown to provide a simultaneous verification of both three- and four-body universal physics.

This work was supported in part by the National Science Foundation. We thank the group of R. Grimm for sharing their experimental data and for stimulating discussions. The Keck Foundation provided computational resources.

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In fact, for Cs the corresponding values for $E_b$ are much higher than the temperatures in Ref. [10] and therefore the threshold law should apply.

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