Field Induced Long-lived Super-Molecules

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We demonstrate that the long-lived bound states (super-molecules) can exist in the dilute limit when we tune the shape of effective potential between polar molecules by an external microwave field. Binding energies, average sizes, and phase diagrams for both s-orbital (bosons) and p-orbital (fermions) dimers are studied, together with bosonic trimer states. We explicitly show that the non-adiabatic transition rate can be easily tuned small for such ground state super-molecules, so that the system can be stable from collapse even near the associated potential resonance. Our results, therefore, suggest a feasible cold molecule system to investigate both novel few-body and many-body physics (for example, the p-wave BCS-BEC crossover for fermions and the paired condensate for bosons) that can not be easily accessed in single species atomic gases.

Since the successful realization of a high-phase-space density of molecules at JILA [1], systems of ultracold polar molecules become one of the most exciting developments in strongly correlated physics. However, the strong dipolar interaction between polar molecules may render the system unstable to collapse in the high density regime (also observed in atomic gases with negative scattering length). Recently there have been several methods proposed to overcome such an unwanted situation by reducing the system dimensions in an optical lattice [2–7]. On the other hand, an effective repulsive core can also be generated to stabilize the system if a proper detuned AC field is applied [8]. These mechanism open new possibilities to investigate interesting many-body physics of polar molecules, including the p-wave superfluid between fermionic molecules [4], the itinerant ferro-electricity [9], and dimerized superfluidity [10] etc.

In addition to many-body physics, it is also interesting to study the few-body physics of polar molecules in the dilute limit. In Ref. [11], the authors proposed a DC-field linked bound state by pumping the electrons to a meta-stable state, which, however, has a very short life time and is therefore not easily accessible. DC field induced inter-layer(-tube) bound states in a multi-layer(-tube) system were also proposed [6], but the mutual long-ranged repulsion makes the critical density for such bound state formation much lower than the density available in current experiments [12].

In this paper, we investigate a different few-body bound state (super-molecule) in the presence of an external AC field [13]. The effective potential between polar molecules can be tuned to have an attractive and shallow well (see Fig. 1(a) and (b)), which supports only a few or even no bound state (dimers and trimers) between molecules. Their average sizes are of sub-micrometer scale, strongly suppressing the few-body inelastic scattering rate and stabilizing the whole system from collapse. As a result, our work suggests a new area for novel few-body physics with a long-lived ground state. Some important many-body physics not accessible in atomic gases, for example the p-wave BCS-BEC crossover between single species fermions [14] and the Z2 transition [15] between the condensate and the pair condensate of single species bosons near the Feshbach resonance, may become feasible without particle loss.

In the low temperature and dilute limit, the only important internal degrees of freedom is the rotational states, if considering close-shell molecules (SrO as an example) without hyperfine structure. As a result,
the general two-body Hamiltonian with dipolar interaction becomes
\[ H = \sum_{i=1,2} \left( \frac{p_i^2}{2m} + Bj_i \cdot E(t) \right) + \frac{d_i^2}{|r_i - r_{12}|^6}, \]
where \( m, \mathbf{J}_i, \) and \( B \) are the molecular mass, angular momentum operator and the rotational constant respectively. \( E(t) \) is the external field, and \( \mathbf{d}_i \) is the electric dipole moment operator. \( \mathbf{r}_i \) and \( \mathbf{p}_i \) are the position and momentum operators. \( \mathbf{e}_{12} \equiv \mathbf{r}_{12}/|\mathbf{r}_{12}| \)
and \( \mathbf{r}_{12} \equiv \mathbf{r}_1 - \mathbf{r}_2 \). Here we will consider only the case when a circularly-polarized microwave field is tuned to be close to the transition energy between \([0, 0] \) and \([1, 1] \) (see Fig. 1(a)). Here \( |J, M\rangle \) is the rotational state labeled by the angular momentum quantum number.

Using the adiabatic and rotating wave approximations \( \mathbb{R} \), we obtain the full effective interaction \( \mathbb{H}_\text{eff} \), which has the leading order terms in the long distance:
\[
V_{\text{eff}}(\mathbf{r}) = -\frac{d_{\text{eff}}^2}{|\mathbf{r}|^6}(1 - 3 \cos^2 \theta) \quad (1)
\]
\[ + \frac{\Delta}{20\Omega^2 |\mathbf{r}|^6} \left[ C_6^{(1)}(\theta) + C_6^{(2)}(\theta) \right] + \mathcal{O}(|\mathbf{r}|^{-9}), \]
where \( d_{\text{eff}} \equiv \frac{3\Omega^2/|\mathbf{r}|^6}{4\sqrt{4+4(\Omega/\Delta)^2}} \)
with \( \mathbf{d} \) being the bare electric dipole moment. \( C_6^{(1)}(\theta) \equiv \frac{(1 + 3\Delta)(1 + (\Omega/\Delta)^2)}{3 + 12 + 6\Omega^2/|\mathbf{r}|^6} \)
and \( C_6^{(2)}(\theta) \equiv \frac{30\Omega^2}{3 + 12 + 6\Omega^2/|\mathbf{r}|^6} \sin^4 \theta \) are two coefficients with \( \xi \equiv \sqrt{1 + 4(\Omega/\Delta)^2} \), \( \theta \) is the angle between the microwave propagation and the relative position of the two molecules (see the inset of Fig. 1(b)). We let \( \hbar = 1 \).

We can see that \( V_{\text{eff}}(\mathbf{r}) \) is dominated by the dipolar-like term with the effective dipole moment, \( d_{\text{eff}} \), as \( |\mathbf{r}| \to \infty \), but its next order correction is like a hard-core potential to prevent possible chemical reactions \( \mathbb{R} \mathbb{R} \mathbb{R} \). Therefore, one can always expect a potential minimum at \( |\mathbf{r}| = r_m \) in the \( x - y \) plane.

In Fig. 1(b), we show the full numerical \( V_{\text{eff}}(\mathbf{r}) \) for different \( \theta \) with the detuning \( \Delta/B = 10^{-4} \) and the Rabi frequency \( \Omega/B = 10^{-3} \). Note that, by using \( r_B \equiv \left( \frac{\hbar}{2\mu} \right)^{1/3} \) and \( B \) as the unit of length and energy, \( V_{\text{eff}}(\mathbf{r}) \) is universal for all molecules, once the external field parameters (\( \Omega/B \) and \( \Delta/B \)) are specified. Using SrO as an example (\( d = 8.9 \text{ Debye} \ B \sim 10^{10} \text{Hz} \)), we find \( r_B \sim 11 \text{ nm} \) and hence the super-molecules can be as long as 100 nm as shown in Fig. 1(c) and (d). The dimer super-molecule has a donut-type wave function for bosons (s-orbital state) and has two knots for fermions in the p$_z$-orbital state [13].

Since all the important and non-trivial properties of the effective potential lies on the \( x - y \) plane, from now on, we will consider 2D molecular motion for simplicity and use \( V_{\text{eff}}(\mathbf{r}) \equiv V_{\text{eff}}(|\mathbf{r}|, \theta = \pi/2) \) as the effective interaction. Such system can be physically realized by adding a strong confinement in the \( z \) direction. The finite width correction can be neglected because the donut type super-molecule has a very small thickness (less than \( 8r_B \sim 88 \text{ nm} \) even for SrO).

In Fig. 2(a) and (b), we show the phase diagrams for the number of 2D dimer states in terms of the detuning \( \Delta/B \) and Rabi frequencies, including both s- and p-orbitals. In the limit of small detuning (say \( \Delta/B < 10^{-4} \)), the bound state is solely determined by \( \Omega \), while in the limit of large detuning, it depends on \( \Delta \) only. In the intermediate regime, there is a triangular window (0.001 < \( \Delta/B < 0.01 \)), where no dimer bound state exists in both orbital.

The appearance of a two-body bound state implies a potential resonance in the low energy scattering amplitude. For our 2D effective potential with a \( r^{-3} \) tail in large distance, the low energy pseudo-potential (\( \tilde{V}_{\text{ps}} \)) can be obtained to be (see Ref. [16] for details):
\[
\tilde{V}_{\text{ps}}(\psi(\mathbf{r})) = \delta(\mathbf{r}) \frac{2\pi}{2\mu \ln(k\alpha_3/2\beta_0)} \left[ \ln \left( \frac{kr}{2\beta_0} \right) \right]^2 \times \frac{\partial}{\partial r} \left[ \frac{\psi(r)}{\ln(kr/2\beta_0)} \right],
\]
where \( \psi(r) \) is the relative wavefunction, \( \beta_0 \equiv e^{-\gamma} \) and \( \gamma \approx 0.57722 \) is Euler’s constant. Here \( \alpha_3 \equiv m\beta_0^{3} \) is the effective scattering length [16] with \( P_0 \) being the only parameter determined from the short-range effective interaction (see Refs. [16] for details). The incident momentum \( k \) is related to the system density. The 2D pseudo-potential is valid for \( k\alpha_3 < 1 \).

In Fig. 2(c), we show the calculated s-wave scattering length, \( a_3 \), as a function of the Rabi frequency at \( \Delta/B = 0.003 \). The corresponding binding energies are also shown in Fig. 2(d). As expected, the divergence of the scattering length (similar to Feshbach resonance) indicates the appearance of an s-orbital bound state with
a binding energy typically smaller than one $\mu$K (taking $B \sim 10$ GHz). The tunability of the scattering length will certainly affect the many-body properties, and this result remains true for the 3D system.

In addition to dimer states, we also investigate three-body bound states (trimers). We note that the Efimov related physics in 3D has recently been investigated for dipolar atoms [17] or 2D systems [18] with short-ranged interaction. However, the effective interactions of our system are not simple dipolar-like, but with several intrinsic length scales. Therefore one should not expect universal results in our current system. Here we apply the stochastic variational method (SVM) with Gaussian function basis to study trimer states of bosonic molecules [19]. The uncertainty of our calculation is about three or-four parameters.

In Fig. 2(d), we also show the binding energy of a typical trimer super-molecule as a function of Rabi frequency. If we stay close to the threshold of bound state formation, there is a finite parameter range ($2 \times 10^{-4} \leq \Omega/B \lesssim 4 \times 10^{-4}$) in which the energy differences from dimer’s are less than $10^{-6}B \sim 10k$ Hz for SrO, smaller than the typical trapping frequency. In other words, even though two super-molecules (four particles) may have inelastic scattering, leading to the formation of one trimer and one free particle, the energy gain of the later should not be high enough to escape the trap. As a result, we do not expect a significant loss rate from such four-body (two super-molecule) scattering, and the resulting ground state should be a mixture of dimer and trimer super-molecules. This is a novel few-body system composed of field-dressed polar molecules. For molecules with smaller dipole moment, $r_B$ is smaller. The stable regime of mixture would be enlarged. When we move away from the threshold, the excited states start to show up (at $\Omega/B \sim 5 \times 10^{-4}$ with $\Delta/B = 0.003$) with energy lower than a dimer.

Now we consider the loss rate of a super-molecule due to the breakdown of the adiabatic approximation. The effective potential is made up of the avoided crossing between two eigenstates in the weak field limit (see Fig. 1(a)). They can be denoted by $|1\rangle$ (effective potential) and $|2\rangle$, merge to $|g,g,N\rangle$ and $|g,e_+;N-1\rangle$ respectively as $r \to \infty$. Here $|g,g,N\rangle$ denotes the two molecules in the rotational ground state with the photon number $N$, while $|g,e_+;N-1\rangle$ denotes the symmetric superposition of one molecule in the ground state and the other one in the excited state with one photon absorption. The nonadiabatic loss thus mainly from the inter-state transition from $|1\rangle$ to $|2\rangle$ due to the relative kinetic energy between two particles neglected before (see Fig. 1(a)). The transition to other states is certainly much smaller and therefore neglected here. Using Landau-Zener formula [20], we can estimate the transition rate to be $\gamma_2 = f_{ave}e^{-2\omega t}$ with the transition parameter,

$$\Gamma \equiv \frac{|V_{12}|^2}{|dE_{12}(r)/dt|_{r_m}}. \quad (3)$$

Here $f_{ave}$ is the frequency for two molecules to collide with each other, $E_{12} \equiv E_{12}^{(1)} - E_{12}^{(2)}$ is the energy difference between the two unperturbed energy eigenstates, and $V_{12} = \Omega$ is the coupling between them via microwave absorption. Within the semi-classical approximation, we have $dE_{12}(r)/dt = E_{12}^{(2)}(r)v(r)$, where $v(r) = dr/dt = \sqrt{2(E_0 - V_{2D}(r))/\mu}$ is the local velocity. Here $E_0$ is the bound state energy and $\mu = m/2$ is the reduced mass. By estimating $f_{ave} = \int f_1^2 \frac{dr}{v(r)}$ for the two turning points at $r_1$ and $r_2$, we can calculate the lifetime $\tau_2 \sim \gamma_2^{-1}$ directly. In Fig. 3(a) we show the full calculation of the transition parameter, $\Gamma$, as a function of $\Omega$ for SrO ($B \sim 10^{10}$Hz and $r_B \sim 11$nm). The typical value of $\Gamma$ is always larger than 5 for SrO, and therefore the obtained life time ($\tau_2$) is very long. We also get $\tau_2 \sim 0.1$ sec for KRb molecule ($B \sim 10^9$Hz and $r_B \sim 3.5$nm, if omitting its fine structure).

In order to understand the life time in more details, we can qualitatively estimate its order of magnitude when the bound state is just formed ($E_B \to 0$). First we have $f_{ave} \sim \delta v/2\delta r \sim m(\delta v)^2/2 \sim E_K$, where $\delta v(\delta r \sim 1)/m\delta r$ is the uncertainty of radial position/velocity and $E_K \sim V_m$ is the kinetic energy. Since $E_{12}(r) \to \Delta$ as $r \to \infty$ and changes within the length scale of $r_m \sim \Delta r/C$ with a field-dependent constant $C$, we then have $E_{12}(r) \sim \Delta/r_m$ and therefore $\Gamma \sim C |\Omega|^2/\Delta E_K$. For example, for the parameter of SrO with $\Omega/B \sim 10^{-4}$ and $\Delta/B \sim 10^{-3}$, we have $C \sim 1$ (see Fig. 1(a)) and hence a very long life time as calculated above ($E_K/B \sim 4 \times 10^{-7}$ and $\Gamma \sim 23$). We note that, different from the situation in the continuous state, the life time of a super-molecule is longer for stronger dipole moment because the length
scale, \( r_B = (d^2/B)^{1/3} \), as well as the radial size of the super-molecules, also becomes larger, reducing the relative collision velocity between two molecules. This is consistent with the fact that the adiabatic approximation is more accurate for particles with larger mass. Similar analysis can be also applied to the trimer case.

The results presented here open a new area for studying few-body physics as well as many-body physics with ultracold molecules. In usual atomic gases, Feshbach molecules is long-lived almost without two- and three-body loss near the threshold of dimer formation, suggesting a much more feasible scenario to study important many-body physics such as the phases mentioned above. Although larger clusters may exist when the density is higher, above statement and prediction should still apply near the threshold of dimer-trimer formation. Much more complicated analysis will be needed to fully address this issue.

In summary, we show that the shapes of the 2D effective potentials between polar molecules can be designed to make long-lived super-molecules. This creates a feasible system for studying novel few-body physics as well as many-body problems.

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