Dynamics of ultracold polar molecules in a microwave field

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Abstract
We analyze the temporal evolution of the population of ultracold polar molecules in a microwave field with a circular polarization. The molecules are in their ground $^1\Sigma_1$ state and treated as rigid rotors with a permanent dipole moment which interact with each other via the dipole–dipole (DD) interaction $V_{dd}$. The mw field mixes states with different quantum and photon numbers and the collisional dynamics in the mw field is mostly controlled by the ratios of the mw field frequency versus the rotational constant, and mw field Rabi frequency versus the rotational constant. There exists a special scattering process which is elastic by nature and due to a rotational energy exchange between the ground and the first excited rotational states. To analyze dynamics of polar molecules system in the mw field the equation of motion for the bare and dressed states is solved under different mw field parameters and molecular gas characteristics. Depending on the ratio of the Rabi frequency of a mw field and the magnitude of the DD interaction, beatings and oscillations occur in the bare and dressed states time-development. At a certain relation between the magnitudes of the mw detuning $\delta$ and the DD interaction $\delta = \pm V_{dd}$, peak structures appear in the population of the excited bare state. Each peak is associated with an avoided crossing between the dressed states adiabatic curves at the same position of mw detuning.

1. Introduction
The cooling and trapping of atoms, which is now easily achieved in numerous labs around the world, has moved in recent years in the direction of cooling and trapping of polar molecules. Possessing a permanent dipole moment and interacting via long range dipole–dipole (DD) forces, ultracold polar molecules offer new prospects of exciting physics in the fields of controlled ultracold chemistry [1], quantum computing [2] and fundamental symmetries tests [3]. The temperature regimes under study are cold (with $1 \text{ mK} < T < 1 \text{ K}$) and ultracold (with $T < 1 \text{ mK}$). For different kinds of useful applications the aim is to produce different order high densities of cold and ultracold molecular ensembles. For example, the test of fundamental forces requires $10^{-8}$–$10^{-6}$ cm$^{-3}$ and $T < 1 \text{ K}$, while for molecules in optical lattices, the number density is $>10^{13}$ cm$^{-3}$ and $T < 100 \text{ nK}$ is needed [4]. In comparison with atoms, the molecular structure is more complex because molecules have additional internal degrees of freedom and an elaborate energy levels structure. Homo-nuclear molecules are non-polar, while hetero-nuclear molecules possess a permanent dipole moment about $0.01$–$1$ au.

In the presence of an external electric field, an induced dipole moment occurs, such that it has a different magnitude and sign for different energy levels. For the electric field values easily achieved in lab conditions, the induced dipole moment reaches its maximum, equal to the permanent dipole moment which means that a full electrical polarization is achieved.

Creating high space densities of ultracold polar molecules can not be achieved by a simple extension of the atomic laser cooling techniques because of the existence of multiple decay channels within the molecular multi-level system. However direct [5], or cavity enhanced, laser cooling [6] has been proposed. A modern review on...
manipulation of molecules with electromagnetic fields can be found in [7] where a recent progress on the physics of polar molecules in the microwave field is discussed as well.

A microwave (mw) trap for polar molecules was suggested in [8]. It has the advantages of being able to trap molecules in their absolute ground state, as well as having a large enough depth (>1 K) and volume (>1 cm³) to allow easy loading. In addition, the alignment of molecules by the trapping field, as well as the strong DD interactions between them, lead to a large elastic collisions cross-section which facilitates evaporative cooling, as shown in [9]. DeMille et al also show that it is advantageous to use circularly polarized light in the mw trap. In this case the ground state energies as a function of field strength have no avoided crossings and a deep trap can be created with a red detuned field. In the case of linear polarization the avoided crossings in the energy-level diagram prevent the meaningful use of the trap. In order to use the mw trap to efficiently trap and further evaporatively or sympathetically cool polar molecules, one needs to know the processes occurring in the trap due to the interaction of the molecules with the mw field. In [10] we considered collisions at ultracold temperatures between fermionic $^1\Sigma$-state diatomic polar molecules in a circularly polarized microwave field. We demonstrated that elastic and inelastic collisions of polar molecules at ultracold temperatures are affected by an external microwave field and that they are enhanced at resonance frequencies. We show that the collision dynamics of polar molecules in a microwave field is largely determined by the long-range DD interaction and through this interaction, two polar molecules can resonantly exchange internal energy by undergoing a transition between neighboring rotational levels. This process may stabilize the system and enable the successive evaporative cooling of molecules.

In this paper we analyze the temporal evolution of the population of ultracold polar molecules interacting with a circularly polarized mw field. In section 2 we compose the Hamiltonian of a ground state polar molecule in the presence of the mw field. Section 3 presents a summary of the dynamics of dipolar collisions in the mw field, based on reference [10]. We construct the basis set of two-molecule bare states and derive the equation of motion for the expansion coefficients of the wavefunction in sections 4 and 5. In section 6 a dressing transformation is performed to give the form of the equation of motion for the two-molecule dressed states. Section 7 gives the main numerical results for the populations of bare and dressed states under different mw trap parameters, followed by Conclusions and Acknowledgements.

2. Diatomic polar molecules in a microwave field

We consider the interaction of diatomic polar molecules with a circularly polarized microwave (mw) field. For simplicity we assume that the molecules are in their ground $^1\Sigma$ state and can be regarded as rigid rotors with a permanent dipole moment. Their rotational energy is given by $E_J = BJ (J + 1)$, where $B$ is the rotational constant and $J$ is the rotational quantum number. In other words, in our model we will neglect the molecular vibrational motion as well as the molecular hyperfine structure. The total Hamiltonian of the molecule + field system can then be written as [8]

$$\dot{H}^{\text{mol}} = BJ^2 + \hbar \omega (\hat{N} - \bar{N}) + H_{\text{Stark}},$$

(1)

where the first term is the field-free molecular Hamiltonian, the second one is the Hamiltonian of the free field and the third one describes the interaction of the molecular dipole moment $\vec{\mu}$ with the mw electric field $\vec{E}$:

$$H_{\text{Stark}} = -\mu \hat{n} \cdot \vec{E} = -\mu \hbar \omega \left[ \hat{n} \cdot \hat{a}^\dagger + \hat{n} \cdot \hat{a} \right].$$

(2)

Here $\vec{n}$ denotes the unit vector along the molecular axis, $\hat{a}$ represents the unit vector of the field polarization, while $\hat{a}^\dagger$ and $\hat{a}$ are the creation and annihilation operators. $\hat{N}$ is the photon number operator and $\bar{N} = \left( E_0^2/2 \right) \left( \varepsilon_0 V / \hbar \omega \right)$ is the average number of photons, $E_0$ is an amplitude of the mw field, $V$ is the volume of a fictitious box used to define the boundary conditions of the quantized electromagnetic field. Since we assume a strong field, the average number of photons is much bigger than the photon number deviation $n$, e.g. $\bar{N} \gg n$ which implies that the dipole-field interaction is much smaller then the field energy. It is worth noting that the use of a circularly polarized field directly diminishes the terms in the interaction Hamiltonian to two instead of the usual four [11].

In order to describe the molecule-field interaction it is convenient to work in the so-called dressed-state picture [12]. The molecule + field bare basis states consist of the products of the free molecular eigenfunctions and the wavefunctions of the free field:

$$|JMn\rangle = |JM\rangle |\bar{N} + n\rangle,$$

(3)

where $M$ is the projection of the angular momentum $J$ on the space-fixed axis and $\bar{N} + n$ is the photon number. The mw field mixes states with different $J$, $M$ and $n$ numbers, thus creating the field-dressed basis given by
where \( p(JMn) \) denotes the mixing coefficients. Due to the mixing, \( J, M \) and \( n \) are not good quantum numbers anymore and in the above notation they stand for the respective quantum numbers at zero field. The dressed-state approach to study cold atomic collisions in an external field was first used by Julienne [13]. Later, Moerdijk et al [14] constructed a two-atom dressed-state basis of ground and excited atomic states in a radiation field. With this basis, they calculate collisional rate constants between atomic states within the dressed-degenerate internal state approximation and show that the results quite agree with their full dressed-state coupled-channel approach. For completeness we have compared the two-atom dressed-states basis sets used in both works [13, 14] and proved that they are equivalent.

The energy level diagram of the manifold of molecular states coupled by the circularly polarized mw field is shown in figure 1. Since the selection rules for molecular transitions caused by circularly polarized light are \( \Delta M = \mp 1 \) for a left/right polarization, the \( \bar{N} + 0 \) manifold is a superposition of the \( |000\rangle + |111\rangle + \cdots \) states, the \( \bar{N} + 1 \) manifold consists of the \( |001\rangle + |221\rangle + |112\rangle + \cdots \) states, and so on. The circularly polarized mw field can not mix states which belong to different manifolds, however, the DD interaction can cause such transitions, leading to unwanted inelastic processes.

Being high-field seeking, the ground state molecules can not be trapped by the use of static electromagnetic fields and switched or radiative fields need to be used. The microwave trap for polar molecules [8] has the advantage of being able to trap molecules in their absolute ground state, as well as having a large enough depth and volume for easy loading. The energies of the dressed states versus the applied mw field strength [10] show that ground state molecules are attracted (and therefore can be trapped) to regions of strong field at negative detunings from the \( J = 0 \rightarrow J = 1 \) resonance. In this case the ground dressed state is in the form

\[
\ket{\bar{N}0} \approx \frac{1}{\sqrt{2}} \left( \ket{000} + \ket{111} \right).
\]

While out of resonance the above dressed state is almost a pure \( |000\rangle \) rotational state, at exact resonance position (e.g. zero detuning) it represents a 50/50 mixture of the bare ground and first excited states as shown in [10].

### 3. Dipolar collisions in a microwave field

In our study we only consider bi-molecular interaction where the molecules are treated as two rigid rotors with permanent dipole moments interacting via the DD interaction. In this case the collisional dynamics is mostly controlled by the ratios of the mw field linear frequency versus the rotational constant, e.g. \( v/B \) and mw field Rabi frequency versus the rotational constant, e.g. \( x = \mu E/hB \). Though in general it is not a good idea to use...
partial wave representation for a system with a possibly strong DD interaction we may still safely apply it to ultracold systems as only a few partial waves contribute considerably to the problem. Further, we want to simplify our methodical consideration as much as possible and so we consider fermionic only species with the only \( l = 1 \) partial wave. Thus we use the ‘channel states’ of the scattering problem given by

\[
\{JMn\}, \quad \{JMn'\} \equiv \{JMn\} \otimes \{JMn'\} \otimes |l = 1 M_l⟩,
\]

(6)

where \( l \) is the angular momentum quantum number of the scattering complex and \( M_l \) is the projection of the partial wave \( l \) on the laboratory-fixed axis. For fermionic species the above wavefunction should always be antisymmetric. For our purposes it is sufficient to account only for \( p \)-wave scattering via the \( l = 1 \) anti-symmetric state of the scattering complex and so the rest of the wave function should be symmetric. Setting \( l \) to 1 corresponds to the case where the angles between the colliding molecules are fixed and the DD interaction is always attractive.

The lowest-energy strong-field-seeking state for the \( N = 0 \) manifold is the \(|000⟩\) state. Since the DD interaction can cause transitions between different manifolds, leading to inelastic relaxation, the inelastic channels for the incident channel \(|000⟩\), \(|000'⟩\) are as follows

\[
\{00 - 1\}, \quad \{00 - 1\}', \quad \{00 - 1\}, \quad \{11 - 2\}', \quad \{11 - 2\}, \quad \{11 - 2\}', \quad \{22 - 3\}, \quad \{22 - 3\}'.
\]

(7)

\[
\{00 - 1\}, \quad \{22 - 3\}', \quad \{11 - 2\}, \quad \{22 - 3\}', \quad \{22 - 3\}.'\ldots.
\]

(8)

All of these final states belong to the \(|N - 1, N - 1⟩\) manifold. However, there exists a special final channel \(|00 - 1\⟩, \quad |001⟩\) of the \(|N - 1, N + 1⟩\) manifold which is elastic by nature. The expression for the DD interaction between the incident and final channels

\[
\langle \{000\}, \quad \{000\} |V_{DD} |\{00 - 1\}, \quad \{001⟩\rangle \propto \langle \{00\}, \quad (1 - 1) |V_{DD} |\{1 - 1\}, \quad (00⟩\rangle
\]

(9)

shows that this scattering process is due to rotational energy exchange between the ground and first excited rotational states \(|000⟩\) and \(|1 - 1 \rangle \) \[10\].

For fermionic molecules the elastic, exchange and total inelastic cross sections \( \sigma \) versus collisional energy \( E \) show the typical Wigner threshold laws at \( E \rightarrow 0 \), namely \( \sigma_{el} \propto E^3 \), which is characteristic of the short-range interaction, \( \sigma_{exc} \propto \text{const} \) and \( \sigma_{inel} \propto E^{1/2} \). A plot of the elastic, exchange and total inelastic cross sections versus the mw field linear frequency \( \nu / B \) for different mw field strengths shows that at weak fields, e.g. values of the parameter \( x \ll 1 \), the rotational excitation exchange dominates over the inelastic and other elastic processes \[10\]. However, strong mw fields \( x = 1 \) cause mixing of states from all different manifolds and thus inelastic collisions are strong. Since the aim of our work is to study the time-dependent behavior of polar molecules in a mw field under the influence of rotational exchange collisions only, we restrict ourselves to the region of weak fields.

4. Time-dependent effects

In the case of weak fields \( x \ll 1 \), the excitation scheme in figure 1 can be truncated to a two-level one, e.g. we take into account only excitations between the rotational ground state \( J = 0 \) and the first rotationally excited state \( J = 1 \).

As we consider bi-molecular processes, the two-molecule states consist of products of two single-molecule eigenstates. For example, the two-molecule state \(|000, 000⟩ \equiv |000⟩|000⟩\) means that both molecules are in the ground state \(|000⟩\). The state \(|000, 1 - 1 - 1⟩ \equiv |000⟩|1 - 1 - 1⟩\) refers to the first molecule being in the ground state and the second one being in the excited state \(|1 - 1 - 1⟩\). The state \(|1 - 1 - 1, 1 - 1 - 1⟩ \equiv |1 - 1 - 1⟩|1 - 1 - 1⟩\) implies that both molecules are in the excited state \(|1 - 1 - 1⟩\), etc. The number of all possible two-state combinations in figure 1 is 36, so the two-molecule wavefunction is expanded in products of single-molecule states as follows

\[
Ψ(t) = \sum_{ab} p_{ab}(t) |a⟩ \otimes |b⟩,
\]

(10)

where \( |a⟩ = |JMn⟩ \), with \( p_{ab}(t) \) being the time-dependent expansion coefficients. Since most of the matrix elements involving the DD interaction are zero, we truncate the basis to seven states only, namely:
For convenience we have chosen to work in a symmetrized basis, where the degenerate states \(|00, 00\rangle\) and \(|00, 11\rangle\) are replaced by their symmetric combination. The states formed by the antisymmetric combinations are uncoupled from the system and therefore excluded.

5. Equations of motion of the bare states

With the two-molecule wave function of the form (10) we write down the Schrödinger’s equation (SE) for the total two-molecule Hamiltonian

\[
\hat{H} = \hat{H}_1^\text{mol} + \hat{H}_2^\text{mol} + \hat{V}_{DD}.
\]

(12)

Here \(\hat{H}_1^\text{mol}\) and \(\hat{H}_2^\text{mol}\) are the Hamiltonians of each molecule given by (1) and \(\hat{V}_{DD}\) denotes the DD interaction between the permanent dipole moments of the two polar molecules.

The Stark interaction matrix element is given in [9]:

\[
\langle J + 1, M - 1, n - 1 | \hat{H}_\text{stark} | J, M, n \rangle = \frac{\mu E \sqrt{J - M + 1} \sqrt{J - M + 2}}{2 \hbar} \frac{\sqrt{2J + 1}}{2 \hbar}.
\]

(13)

The reduced angular matrix element for the DD interaction can be found, for example, in [15].

Writing down the SE for the two-molecule wavefunction (10) with the total two-molecule Hamiltonian given by (12) we then obtain the equations of motion for the expansion coefficients \(p(t)\):

\[
i\hbar \frac{\partial}{\partial t} \vec{p} = \hat{H} \vec{p},
\]

(14)

where the matrix form of the Hamiltonian \(\hat{H}\) is given by

\[
\begin{pmatrix}
0 & \sqrt{2} \Omega & 0 & V_{dd} \\
\sqrt{2} \Omega & \delta & \sqrt{2} \Omega & 0 \\
0 & \sqrt{2} \Omega & 2\delta & 0 \\
V_{dd} & 0 & \Omega & 0 \\
0 & \Omega & \delta & \Omega \\
0 & \Omega & \Omega & 2\delta
\end{pmatrix}
\]

(15)

Here \(\delta = \omega - 2B_2/\hbar\) is the detuning of the microwave field from the \(|00\rangle \rightarrow |1 - 1\rangle\) transition, while \(\Omega\) is the Rabi frequency between the ground and first excited states, as defined by (13). The Hamiltonian (15) is block-diagonal with the 3x3 block belonging to the \([\bar{N} + 0, \bar{N} + 0]\) manifold and the 4x4 block belonging to the \([\bar{N} - 1, \bar{N} + 1]\) manifold. The two manifolds are connected by the exchange DD interaction

\[
V_{dd} = (000, 1 - 1 - 1) V_{DD} |1 - 10, 00 - 1\rangle,
\]

(16)

which leads to exchange of rotational energy between the ground \(|00\rangle\) and first excited \(|1 - 1\rangle\) states. Equation (14) for the expansion coefficients \(p(t)\) describes the time evolution of all the two-molecule bare states in the system.

It is worth noting that in reality the Hamiltonian in the above equation is still time-dependent due to the \(1/r(t)^3\) term in the DD interaction matrix elements. The intermolecular distance is given by \(r(t) \equiv p^2 + v^2 t^2\) [18], where \(p\) is the impact parameter for the collision and \(v\) is the molecular velocity. In our calculations, however, we apply the adiabatical approximation and assume that the molecules are at fixed locations with a constant separation of \(r\). This approximation is justified due to the slow molecular motion. Here we have to
point out that in the limit of the strong DD interaction ($V_{DD} > 2B_e$) the adiabatical approximation will not work and the spatial uncertainty leads to decoherence and washes out the Rabi-like oscillation as well as the other oscillations we found (see figure 2(a)). Such observations were clearly demonstrated for the DD-interacting Rydberg atoms at Forster resonance [19, 20]. In the case of polar molecules it might be quite difficult to reach the strong DD interaction as they have to be placed in a very close proximity which is far below the optical resolution of optical dipole traps while it is quite possible with Rydberg atoms due to their huge dipole moments [21].

6. Equations of motion of the dressed states

In each molecular level excitation scheme (see figure 1) the mw field mixes the states $|000\rangle$ and $|1-1-1\rangle$, $|001\rangle$ and $|1-1-2\rangle$, and, $|00-1\rangle$ and $|1-1-2\rangle$ respectively, via the Stark interaction [22], creating the following single-molecule dressed states [12]

$$
|000\rangle^+ = S |000\rangle + C |1-1-1\rangle,
|000\rangle^- = C |000\rangle - S |1-1-1\rangle,
|00-1\rangle^+ = S |00-1\rangle + C |1-1-2\rangle,
|00-1\rangle^- = C |00-1\rangle - S |1-1-2\rangle,
|001\rangle^+ = S |001\rangle + C |1-10\rangle,
|001\rangle^- = C |001\rangle - S |1-10\rangle.
$$

In the above we have defined the notation

$$C \equiv \cos \left(\frac{\theta}{2}\right), \quad S \equiv \sin \left(\frac{\theta}{2}\right),$$

Figure 2. Population of bare (a) and dressed (b) states as a function of time for different values of intermolecular distance $r$ at a value of $x = 0.01$ and $\delta \approx 0$. 

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where $\Theta$ is the mixing angle, such that $\tan \Theta = -\Omega/\delta$. The two-molecule dressed states are then constructed as the products of the single-molecule ones and similarly to (11) we choose seven of them to represent the two-molecule dressed-state basis as follows:

$$\begin{align*}
00^+, 00^+ &\rightarrow |1\rangle \\
\frac{\sqrt{3}}{2} [00^+, 00^-] + \frac{\sqrt{3}}{2} [00^-, 00^+] &\rightarrow |2\rangle \\
00^-, 00^- &\rightarrow |3\rangle \\
\frac{\sqrt{3}}{2} [00^-, 00^+] + \frac{\sqrt{3}}{2} [00^+, 00^-] &\rightarrow |4\rangle \\
\frac{\sqrt{3}}{2} [00^+, 00^+] + \frac{\sqrt{3}}{2} [00^-, 00^-] &\rightarrow |5\rangle .
\end{align*}$$

As in the bare states case (10), we expand the two-molecule dressed states wavefunction in the two-molecule dressed states basis:

$$\Psi(t) = \sum_{\langle \alpha \beta \rangle} p_{\alpha \beta}(t) |\alpha\rangle \otimes |\beta\rangle, \quad |\alpha\rangle = |JM\rangle,$n(19)

where $p_{\alpha \beta}(t)$ are the time-dependent expansion coefficients.

Based on the definition of the two-molecule dressed (19) and bare (11) states bases, together with the decomposition (17), the following dressing transformation can be performed:

$$P_{\alpha \beta} = \begin{pmatrix}
S^2 & \sqrt{2} CS & C^2 & 0 & 0 & 0 & 0 \\
\sqrt{2} CS & C^2 - S^2 & -\sqrt{2} CS & 0 & 0 & 0 & 0 \\
C^2 & -\sqrt{2} CS & S^2 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & S^2 & CS & CS & C^2 \\
0 & 0 & 0 & CS & -S^2 & C^2 & -CS \\
0 & 0 & 0 & CS & C^2 & -S^2 & -CS \\
0 & 0 & 0 & C^2 & -CS & -CS & S^2
\end{pmatrix} P_{a \beta}.$$

The equations of motion for the expansion coefficients either for $p_{\alpha \beta}(t)$ or $p_{a \beta}(t)$ can be solved numerically only.

7. Populations dynamics

Solving the equations of motion of the bare and dressed states yields the time evolution of the populations of these states for a sudden turning on of the mw field. Our aim is to study the effect of different mw field parameters and molecular gas characteristics on the dynamics of all the states. The results were computed numerically using standard numerical methods for solving complex differential equations. We tested several ranges of the intermolecular distance $r$, which correspond to various densities of the molecular gas. For reference we denote the intermolecular distance at which the magnitude of the DD interaction becomes equal to the energy of the $J = 0 \rightarrow J = 1$ transition, e.g. $V_{dd} = 2B_o$, as $r_o$.

Figure 2(a) shows the populations of all the bare states which contain the $|00, 00\rangle, |1 - 1, 1 - 1\rangle$ and $|00, 1 - 1\rangle$ states (11) as a function of time for different values of $r$, while the populations of all the dressed states containing the $++$, $--$ and $+-$ ones (19) are plotted in figure 2(b). Our calculations show that the behavior of the populations dynamics is defined by the ratio of the Rabi frequency $\Omega$ and the magnitude of the DD interaction $V_{dd}$. At large enough distances, e.g. $r/r_o = 100$, the DD interaction is too weak to affect the system. We have the case of two independent molecules dressed by the mw field, therefore we see the typical Rabi oscillations in the populations of the bare states (figure 2(a)) and no change in time in the populations of the dressed states (figure 2(b)). As the density of the molecular gas increases, $(r/r_o = 10)$, the intermolecular interaction becomes significant and causes beatings in the evolution of the bare states populations, while the dressed states start to slightly oscillate. At a critical distance of about $(r/r_o = 5)$ the strength of the DD interaction
becomes comparable to the magnitude of the Rabi frequency and the dressed states start to rapidly oscillate, while beatings and oscillations occur in the bare states time-development.

Next, we investigate the influence of initial conditions on the bare states population as a function of mw detuning. For this purpose we take the last case in figure 2, when the DD interaction $V_{dd}$ is much stronger than the Rabi frequency. The different initial conditions correspond to different photon distributions.

When all ground states regardless of photon number are equally populated, e.g. $p_{000}(t) = p_{000}(0)$, the photon distribution corresponds to a flat line. The population of all the states containing the $|00, 1\rangle$ state as a function of mw detuning (figure 3(a)) exhibits a peak at the position at which the detuning of the mw field becomes equal but opposite in sign to the magnitude of the DD interaction, $\delta = -V_{dd}$.

When the photon distribution changes to a step-function ($p_{000}(t) > p_{000}(0)$), a second peak appears at the symmetrical position $\delta = V_{dd}$ (figure 3(b)), and becomes of the same intensity as the first one when the photon distribution is that of a delta-function, e.g. when all the initial population is in the $|000\rangle$ state (figure 3(c)).

It is worth noting that the case of a flat-line photon distribution corresponds to the semi-classical case described by Shah et al [23]. There, the magnitude of the DD interaction $V_{dd}$ enters one of the diagonal elements of the Hamiltonian as $-\delta - V_{dd}$, giving rise to only one peak in the states population at negative detuning, when $\delta = -V_{dd}$ and $V_{dd} > 0$ is attractive.

In order to understand the origin of the peaks in bare states population shown in figure 3, we investigate the behavior of the two-molecule dressed states eigenvalues as a function of mw detuning. As it can be seen from figure 4, each peak in the population of bare states at $\delta = \pm V_{dd}$ is associated with an avoided crossing between the dressed states adiabatic curves at the same position of the mw detuning. The gap between the adiabatic curves of the $|000, 1\rangle$ and $|000, 1\rangle$ states is proportional to the magnitude of the DD interaction. Therefore, in the case of $V_{dd} = 0$ both curves approach their asymptote and no avoided crossings are present, leading to no peak structures in the bare states population. From this we can conclude that the occurrence of the observed peaks is entirely due to the intramolecular DD interactions. Unlike the peak structures, the avoided crossings exist for both positive and negative detunings regardless of the initial conditions (and photon distributions respectively), since the adiabatic curves reflect the behavior of the dressed states which are time independent, while the initial conditions determine the time-dependent dynamics of the bare states.

Finally, we investigate the effect of the ratio between the magnitude of the DD interaction and the Rabi frequency of the mw field on the shape of the bare states population as a function of mw detuning. At a fixed value of the Rabi frequency $\Omega/2B_c \approx 0.001$, we increase the intermolecular distance till the value of $V_{dd}$ becomes much smaller compared to $\Omega$. Figure 5 shows that with diminishing the DD interaction the distance between the
two peaks in the bare $|00, 1 - 1\rangle$ state population, placed at $\delta = \pm V_{dd}$, decreases until they become indistinguishable.

The same effect is observed in the case when the value of the DD interaction (e.g. the distance between molecules) is kept constant while the Rabi frequency of the field is gradually increased from about a hundred times smaller to a couple of times bigger than $V_{dd}$, as shown in figure 6.

8. Conclusions

In this paper we investigated the population dynamics of ultracold polar molecules in a circularly polarized mw field. The molecules are assumed to be in their ground $^1\Sigma$ state and only rotational motion is accounted for,
neglecting molecular vibrational and hyperfine structure effects. We restricted ourselves to the region of weak mw fields, where the elastic exchange collisions, due to the rotational exchange DD interaction, dominate over the inelastic and other elastic processes. Under these conditions, the excitation scheme is truncated to a two-level one, involving excitations between the rotational ground and first excited states only. We worked in a field-dressed basis, created by the mw field which mixes states with different quantum and photon numbers. In this basis, the SE for the two-molecule bare and dressed states were solved numerically using standard numerical methods. Our simulations show that the population dynamics is mostly controlled by the ratio of the Rabi frequency of a mw field and the magnitude of the DD interaction. With the decrease of an intermolecular distance, the DD interaction becomes comparable to the Rabi frequency and causes beatings and oscillations in the evolution of bare and dressed states populations. Studies of the effect of initial conditions on the bare states population show that when the photon distribution is that of a delta-function, two symmetric peaks appear in the population of the excited bare states at values of mw detuning $\delta = \pm V_{dd}$. The occurrence of the peak structures is explained with the existence of two avoided crossings between the dressed states adiabatic curves at the same positions of the mw field detuning. As the magnitude of the DD interaction becomes smaller than the mw field strength, the distance between the two peaks decreases until they become indistinguishable.

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**Figure 6.** Population of bare states as a function of mw detuning for different ratios of $V_{dd}$ and $\Omega$ at a value of $V_{dd}/2B_s \approx 0.01$ (corresponding to $r/s = 5$) and initial condition $\rho_{00}(t = 0) = 1$. 
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