Collisions of bosonic ultracold polar molecules in microwave traps

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Abstract. The collisions between linear polar molecules, trapped in a microwave field with circular polarization, are theoretically analyzed. The microwave trap suggested by DeMille et al (2004 Eur. Phys. J. D 31 375) seems to be rather advantageous in comparison with other traps. Here we have demonstrated that the microwave trap can provide successful evaporative cooling for polar molecules in a rather broad range of frequencies of the ac field. We suggest that not only ground-state polar molecules but also molecules in some other states can be safely trapped. But the state in which molecules can be safely loaded and trapped depends on the frequency of the ac field.

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1. Introduction

In the last few years, considerable progress has been made in the development and investigation of various types of electromagnetic traps for different molecules and especially polar molecules. The building of such traps should have made it possible to perform many interesting experimental studies and applications involving the manipulation of cooled and trapped molecules with kinetic temperatures near zero kelvin \[2\]–\[9\]. Schemes for creating samples of cold molecules are being developed now in several laboratories around the world where simple molecules at kinetic temperatures in the mK to \(\mu\)K range are being created and studied.

One of the main purposes in the perfection of traps is to keep samples of molecules dense and cold enough, and the microwave trap seems to be very advantageous in restraining polar molecules \[1\]. In this article, we study the collisional dynamics of linear polar molecules in the \(^1\Sigma\) state trapped in a microwave field with circular polarization at cold temperatures. The choice of linear polarization does not seem very appealing as has been shown in \[1\], as ac Stark levels have a lot of avoiding crossings, which enhances the collisional loss. Polar molecules have strong permanent electric dipole moments and therefore the collisional dynamics are mostly ruled by the dipole–dipole interaction:

\[
V_{\mu\mu}(\vec{R}) = \frac{\vec{\mu}_1 \cdot \vec{\mu}_2 - 3 (\vec{e}_R \vec{\mu}_1)(\vec{e}_R \vec{\mu}_2)}{R^3}. \tag{1}
\]

Here, \(\vec{\mu}_i\) is the electric dipole moment of the molecule \(i\), \(\vec{R}\) is the intermolecular separation and \(\vec{e}_R = \vec{R}/R\). Thus, in our model, collisions are controlled by two ratios \(\nu/B\) and \(x = \mu \mathcal{E}/hB\) (\(\nu\) is the microwave frequency, \(B\) is the molecular rotational constant and \(\mathcal{E}\) is the electric field strength). We are using the coupled-channels method to calculate elastic and inelastic cross sections depending on the frequency and strength of an ac electric field. It has to be noted that the collisional dynamics of cold polar molecules in the dc electric field were rather intensively theoretically explored in the literature, and this has revealed quite rich physics \[10\]–\[17\].

One of the main goals is to understand whether polar molecules in their absolute ground state can have successful evaporative cooling. At first sight it seems rather an inappropriate question as molecules are in their absolute ground state. Naturally, if the energy of the first excited rotational state is smaller than the microwave frequency \(\nu < B\) it seems that the system is safe against two-body inelastic collisions and collision losses are excluded. This is true for bare particles. But it might be not completely true for particles dressed with a microwave field, especially for parameters that provide a large and deep trap \(\nu \sim B, \mu \mathcal{E} \gtrsim B\). As we have seen, the reason is that the ac field and the dipole–dipole interaction are mixed states with different angular moments and ‘dressing’. We have found that even for the most favorable case \(\nu < B\), there are inelastic processes. But these processes are causing mostly the ‘undressing’ or ‘dressing’ of the colliding particles without changing their internal structure. Here we have to be careful about the term ‘mostly’ as dressed states are the superposition of a lot of pure states. But what we can be sure of is that these ‘dressing’–‘undressing’ processes do not cause the loss of molecules from the trap.

Moreover, this is supposedly true not only for molecules in their absolute ground state \(|J = 0, M = 0\rangle\), but for example in the \(|J = 1, M = -1\rangle\) state at frequencies around \(\nu/B = 3\), for molecules \(|J = 2, M = -2\rangle\) at frequencies around \(\nu/B = 5\) and so on \((J\) is rotational quantum number). Actually how far this ‘around’ goes depends on the strength of the ac field.
We have found that during the collisions, the parity of the projection of the total momentum of two colliding dressed molecules is conserved. Our calculations demonstrate that the cross sections for such positive and negative projection parities are different by several orders of magnitude.

2. Polar $^1\Sigma$-type molecules in a microwave field

Here we analyze the ac-Stark shift, which determines the trap depth. The energy levels of $^1\Sigma$-type molecules can be described by rotation $J$, total spin $F$ (i.e. including nuclear spin) and vibration $\nu$ quantum numbers. For simplicity, we will neglect hyperfine splitting and consider molecules only in the $\nu = 0$ vibrational ground state. So we treat polar molecules as rigid rotors with a permanent dipole moment. The dressed-state formalism is particularly convenient for describing the photon–atom interaction in the strong field limit \[19, 20\] where the dressed states are eigenstates of the Hamiltonian of the total system: particle plus photon. The basis states of the rigid rotor plus field Hamiltonian, $H = H_{\text{rot}} + H_{\text{field}}$, are $|J, M, \tilde{N} + n\rangle$, where $M$ is the projection of $J$ on the space-fixed axis, which is conveniently chosen parallel to the wave vector of the microwave field. $\tilde{N} + n$ is the dressed state photon number and $n \ll \tilde{N}$ is the deviation of the photon number. The ac-Stark splitting is caused by the molecule–field interaction $H_{\text{Stark}} = -\vec{\mu} \vec{E}$. Considering the circularly polarized microwave field ($\sigma^-$), the nonzero Hamiltonian matrix elements (normalized by $\hbar B$) are given by

$$
(J, M, n | H_{\text{rot}} + H_{\text{field}}| J, M, n) = J (J + 1) + n \frac{\nu}{B},
$$

$$
(J + 1, M + 1, n + 1 | H_{\text{Stark}}| J, M, n) = \frac{x \sqrt{J + M + 1} \sqrt{J + M + 2}}{2 \sqrt{2J + 1} \sqrt{2J + 3}},
$$

$$
(J + 1, M - 1, n - 1 | H_{\text{Stark}}| J, M, n) = \frac{x \sqrt{J - M + 1} \sqrt{J - M + 2}}{2 \sqrt{2J + 1} \sqrt{2J + 3}}.
$$

Here, it is assumed that $n$ is much smaller than the mean photon number $N$. In the ac electric field, the $J, M$ and $N + n$ are not good quantum numbers and the dressed state formalism should be applied.

2.1. Dressed states

The ac electric field mixes states with different $J, M$ and $n$ and hence none of them is a good quantum number. Thus, we can only mark our states by their origin at zero field (figure 1) where they can be assigned the $J$ and $M$ quantum numbers. Figure 1 shows the energies of dressed states versus the applied electric field frequency for the $J = 0$ and 1 states at $x = 0.7$ within $\nu/B = [0, 4]$ frequency range.

First we would like to consider the ground and so the lowest-field seeking state $|J = 0, M = 0\rangle$, though the term ‘lowest-field seeking state’ does not seem appropriate in dressed state formalism. In practice, we transform the molecular state to a field-dressed basis for performing scattering calculations and the state with a given mean photon number $\tilde{N}$ is described as

$$
|\tilde{N}\rangle \equiv \left| J(M)\tilde{N}; \vec{E} \right\rangle = \sum_{JMn} p_{JMn} |JMn\rangle,
$$

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where \( p_{(J,Mn)} \) stands for the eigenfunctions of the \( H_{\text{rot}} + H_{\text{field}} + H_{\text{Stark}} \) Hamiltonian determined numerically at each value of the field. We will continue to refer to the molecular states by the quantum numbers \( J, M \) and \( n \), with the understanding that they are only approximately good in a field and that equation (4) is the appropriate molecular state. Note that ac-Stark splitting should be irrelevant to the value of \( |n| \) as \( n \ll \tilde{N} \) so we choose small values of \( n \). Further we are mostly interested in cases in which the strength of the ac field is not large (\( \mu E \lesssim B \)), which is found to be more practical for a microwave trap [1]. In this case, the above mentioned mixing is not that strong and allows us to analyze different cases.

2.2. The \( \nu \lesssim 2B \) case

Let us consider the \( |\tilde{000}\rangle \) state, which is a pure \( |000\rangle \) if we neglect the molecule–field interaction \( H_{\text{Stark}} \) and describe molecules in their absolute ground state. In [1], it was pointed out that the most relevant trap parameters are at \( x \gtrsim 1 \) and the ratio \( \nu/B \sim 1 \). In this case, the state of interest is approximately described as

\[
|\tilde{000}\rangle \approx p_{(000)} |000\rangle + p_{(1-1-1)} |1-1-1\rangle.
\]

It should be noted that at \( x \gtrsim 1 \), \( p_{(000)} \approx p_{(1-1-1)} \approx 1/\sqrt{2} \). At larger \( x \), admixtures of other states, like \( |2-2-2\rangle \) and so on, are not small. Here we would like to point out that if trapped molecules are in this state \( |\tilde{000}\rangle \), it is equal to saying that trapped molecules are in \( |00|n\rangle \) states as the ac-Stark shift is the same for all of them. Moreover, the \( H_{\text{rot}} + H_{\text{field}} + H_{\text{Stark}} \) Hamiltonian does not mix, for example, states like \( |\tilde{000}\rangle \) and \( |\tilde{00} \pm \rangle \). But the dipole–dipole interaction can mix them as the

\[
|\tilde{000}\rangle |\tilde{000}\rangle \Rightarrow |\tilde{00} \pm \rangle |\tilde{00} \pm \rangle
\]
scattering processes are possible. To reveal the possibility of such processes, one has to see that at $x \gtrsim 1$ and the ratio $\nu/B \sim 1$, these states are mostly described as
\begin{equation}
\left| \tilde{00} \right\rangle \approx p_{(00-1)} \left| 00 \right\rangle - 1 + p_{(1-1-2)} \left| 1 -1 -2 \right\rangle ,
\end{equation}
\begin{equation}
\left| \tilde{00} \right\rangle \approx p_{(001)} \left| 001 \right\rangle + p_{(1-10)} \left| 1 -10 \right\rangle .
\end{equation}
Thus the
\begin{equation}
\left| \tilde{00} \right\rangle \left\langle \tilde{00} \right| \sim p_{(1-1-1)} \left| 1 -1 -1 \right\rangle \left\langle 1 -1 -1 \right| \mu \left| 00 -1 \right\rangle ,
\end{equation}
\begin{equation}
\left| \tilde{00} \right\rangle \left\langle \tilde{00} \right| \sim p_{(000)} \left| 000 \right\rangle \left\langle 000 \right| \mu \left| 1 -10 \right\rangle
\end{equation}
matrix elements provide for the above mentioned process and their values depend on $p_{(J\mu\nu)}$ eigenstates.

At $\nu \lesssim B$, only two inelastic processes may have considerable cross sections:
\begin{equation}
\left| \tilde{00} \right\rangle \left\langle \tilde{00} \right| \Rightarrow \left| \tilde{00} \right\rangle \left\langle 00 \right| \Rightarrow \left| \tilde{00} \right\rangle \left\langle 00 \right| \Rightarrow \left| \tilde{00} \right\rangle \left\langle 00 \right| .
\end{equation}
Note that the threshold energies of $\left| \tilde{00} \right\rangle$ and $\left| \tilde{00} \right\rangle$ channels are degenerated.

At $\nu \gtrsim B$, the $\left| (1 -1 -2) \right\rangle$ state becomes energetically accessible (see figure 1) so that the
\begin{equation}
\left| \tilde{00} \right\rangle \left\langle \tilde{00} \right| \Rightarrow \left| \tilde{00} \right\rangle \left\langle \tilde{00} \right| \Rightarrow \left| \tilde{00} \right\rangle \left\langle \tilde{00} \right| \Rightarrow \left| \tilde{00} \right\rangle \left\langle \tilde{00} \right| .
\end{equation}
inelastic process is possible.

2.3. The $2B \lesssim \nu \lesssim 4B$ case

In this region of frequencies, our state of interest $\left| \tilde{00} \right\rangle$ is approximately described as
\begin{equation}
\left| \tilde{00} \right\rangle \approx p_{(000)} \left| 000 \right\rangle + p_{(1-1-1)} \left| 1 -1 -1 \right\rangle + p_{(2-2-2)} \left| 2 -2 -2 \right\rangle .
\end{equation}
If the strength of the field is not large (see figure 1), then in the middle of this region this state is an almost pure $\left| 1 -1 -1 \right\rangle$ state. As $\nu/B$ gets close to 5, the state is dominated by the $\left| 2 -2 -2 \right\rangle$ state. So the larger the frequency of the ac electric field, the more a particular angular moment $J$ will dominate. But this is only true if the strength of a field is not large ($x \lesssim 1$). Otherwise, at a large $x$, the dressed state will be a superposition of a lot of states.

Compared with the $\nu \lesssim 2B$ case, there is one extra open channel for inelastic collisions:
\begin{equation}
\left| \tilde{00} \right\rangle \left\langle \tilde{00} \right| \Rightarrow \left| \tilde{00} \right\rangle \left\langle \tilde{00} \right| \Rightarrow \left| \tilde{00} \right\rangle \left\langle \tilde{00} \right| .
\end{equation}
Moreover, our state in this region of frequencies becomes a weak-field seeking one, while the state $\left| (1 -1 -2) \right\rangle$ becomes a strong-field seeking state.
The main difference from the dc electric field case is that, in general, inelastic processes are always allowed for any nonzero field and for any state. Of course an inelastic cross section will be very small at small fields but not exactly zero. In the following, we focus only on bosonic species. The case of fermionic species will be considered elsewhere.

We have solved the coupled-channel equations using the logarithmic derivative propagator method [21] to calculate total state-to-state cross sections. The scattering calculations quickly become computationally expensive as more rotational states, partial waves and dressed-state photon numbers are included. We have found that it is enough to take \( J = 0, 1; \: l = 0, 2 \) for bosons and \( l = 1, 3 \) for fermions and \( \Delta n = 0, \pm 1, 2 \) for changing photon numbers to get reasonably converged cross sections. The only two-body interaction taken into account here is the dipole–dipole interaction, which is sufficient to describe the qualitative behavior of cross sections.

We have not chosen any particular polar molecule for calculation but a rigid rotor with the rotational constant \( B = 6 \, \text{GHz} \) and dipole moment \( \mu = 3 \, \text{D} \). For this molecule, we are mostly interested in the lowest energy strong-field seeking state of the ground vibrational state. The cross sections were calculated for dressed molecules, which means we show them for molecules in \(|(J, M)\rangle\) basis. The trap depth for this particular molecule is 12 mK at \( x = 0.5 \) and 50 mK at \( x = 1 \) if \( \nu/B = 1/3 \). It is 60 mK at \( x = 1 \) and \( \nu/B = 1 \). For the high-frequency case \( \nu/B = 2.5 \), it is only 3.5 mK at \( x = 1 \).

We have found that if one takes into account only the dipole–dipole interaction in the presence of an ac field, the only conserved value for two molecules in \(|(J_1, M_1, n_1); (J_2, M_2, n_2)\rangle\) states is \((-1)^{M_1 + M_2 + m_1 + n_1 + n_2}\), which we call a projection parity. Thus, the system of the coupled-channel equations is split into two blocks with positive and negative projection parity. The nature of this symmetry will be described elsewhere.

Figure 2 shows cross sections for bosonic and fermionic species for the \(|(000)\rangle\) state at \( x = 1 \) and \( \nu/B = 1 \) parameters, which are proposed to be more relevant for the future experiment [1]. The black and red curves are for positive and negative projection parities, respectively. The largest inelastic cross section is for \(|(000)\rangle|(000)\rangle \Rightarrow |(00-1)\rangle|(001)\rangle \) transitions and, moreover, it is not small and the threshold behavior is similar to that of an elastic process. The latter is clear as these states are degenerate. Such an inelastic transition can be described as the ‘dressing’ or ‘undressing’ of molecules by one photon each. Will this ‘undressing’ cause the loss of molecules from the trap? We believe that it will not as in the limit of a strong field the Stark shift as well as the trap depth will be the same for dressed states with \( \tilde{N} \) and \( \tilde{N} \pm 1 \) photons.

The only inelastic process that causes trap loss is \(|(000)\rangle|(000)\rangle \Rightarrow |(00-1)\rangle|(1-1-2)\rangle\). But the cross section for it is relatively small in the region from about 0.1–100 \( \mu \text{K} \) and maybe even at lower temperatures. One can see that there exists some resonance structure. We do not take any notice of these resonances as they are defined by an unknown short-range part of the interaction, which in our case is simply modeled by the inner wall of the interaction. Thus, after all, our ‘absolute ground state’ molecules undergo inelastic transitions. However, at \( x \approx 1 \) and \( \nu/B \approx 1 \), trapped molecules will feel pretty safe.

It is worth noting that the elastic cross sections are much larger for positive parities at ultracold energies (about 1 \( \mu \text{K} \)) especially, while they are more or less of the same order at cold energies (about 1 mK). We have found that the threshold behavior of the inelastic cross section
Figure 2. Elastic (solid lines) and inelastic (non-solid lines) cross sections versus the collisional energy for $x \approx 1$, $\nu/B \approx 1$. The black and red curves are for the positive and negative projection parities respectively. The dashed curves are for $|\tilde{00}\rangle|\tilde{00}\rangle \rightarrow |\tilde{00}^{-1}\rangle|\tilde{00}^{-1}\rangle$, the dotted curve is for $|\tilde{00}\rangle|\tilde{00}\rangle \rightarrow |\tilde{00}^{-1}\rangle|\tilde{00}^{-1}\rangle$ and the dot–dot–dashed curve is for $|\tilde{00}\rangle|\tilde{00}\rangle \rightarrow |\tilde{00}^{-1}\rangle|\tilde{00}^{-1}\rangle$ transitions.

Figure 3 shows cross sections for $x = 0.5$ and different $\nu/B$ ratios. Though the trap depth is not that deep for this $x$, it is still several mK. We would like to pay attention to the relatively small value of the field strength at least. The only reason we can think of is that, for a given projection parity, not all projections $m_l$ of the partial wave $l$ are involved. This phenomenon will be studied elsewhere.

For bosonic molecules with a negative projection parity does not follow the Wigner law at the shown collisional energies at least. The only reason we can think of is that, for a given projection parity, not all projections $m_l$ of the partial wave $l$ are involved. This phenomenon will be studied elsewhere.

Figure 3 shows cross sections for $x = 0.5$ and different $\nu/B$ ratios. Though the trap depth is not that deep for this $x$, it is still several mK. We would like to pay attention to the relatively small value of the field strength as, in this case, dressed states are mostly superpositions of only two pure states or even sometimes one pure state (see discussions in section 2). In this case, it is easy to analyze hidden details. At $\nu/B \approx 0.3$, the field is rather weak, so dressed states are almost pure states (for example, it means that, say, $|\tilde{00}\rangle \approx |(00\rangle$, see equations (5)–(8)) and all inelastic cross sections are small. At $\nu/B \approx 1$, the dressed states are mostly superpositions of two states (equations (5)–(8)), which considerably enhance ‘dressing’–‘undressing’ inelastic processes involving considerable transitions to $|J = 1, M = -1\rangle$ states by means of the dipole–dipole interaction (equation (8)). Nevertheless, such transitions are between equally trapped states $|(00\rangle|n\rangle)$. At $\nu/B = 2.5$, the dressed state of interest is by the largest part the $|(1-1-1)\rangle$ state and it is already a weak-field seeking state. Moreover, one can see that inelastic cross sections which may cause the loss from a trap are really small. We may suppose that it is possible to prepare polar molecules in their $|J = 1, M = -1\rangle$ states and then load them into a microwave field at a frequency $\nu$ around $3B$. It is probable that the same trick can be done with, say, $J = 2, M = -2$ molecules by loading them into a microwave field at a frequency $\nu$ of about $5B$.

We have found that the cross sections have rather a weak dependence on the frequency of an ac field (figures 4 and 5) for large values of the strength of the field ($x \approx 1$ at $E \approx 4 \text{ kV cm}^{-1}$).
Figure 3. Elastic (solid lines) and inelastic (non-solid lines) cross sections versus the collisional energy at $x \approx 0.5$ for (a) $v/B = 0.3$, (b) 1 and (c) 2.5. The dashed curves are for $|\langle 000 \rangle \rangle\langle 000 \rangle \rangle \Rightarrow |\langle 00 -1 \rangle \rangle\langle 001 \rangle \rangle$, the dotted curves are for $|\langle 000 \rangle \rangle\langle 000 \rangle \rangle \Rightarrow |\langle 00 -1 \rangle \rangle\langle 00 -1 \rangle \rangle$, the dot–dot–dashed curves are for $|\langle 000 \rangle \rangle\langle 000 \rangle \rangle \Rightarrow |\langle 00 -1 \rangle \rangle\langle 1 -1 -2 \rangle \rangle$, and the dot–dashed curve is for $|\langle 000 \rangle \rangle\langle 000 \rangle \rangle \Rightarrow |\langle 1 -1 -2 \rangle \rangle\langle 1 -1 -2 \rangle \rangle$ transitions.
Figure 4. Elastic (solid line) and total inelastic (dotted line) cross sections versus the microwave electric field strength at the collision energy $1 \mu K$ for $\nu/B = 0.3$. The value $x \approx 1$ is attained at $E = 4 \text{kV cm}^{-1}$.

Figure 5. Elastic (solid line) and inelastic (non-solid lines) cross sections versus the microwave electric field strength at the collision energy $1 \mu K$ for $\nu/B = 2.5$. The inelastic processes are marked as on figure 3.

Both elastic and inelastic cross sections are more or less of the same order. Of course there is rather a rich resonance structure of cross sections, which reflects the fact of existence of a lot of bound states in closed channels. We do not pay attention to this fact as the short-range potential is unknown and simply parameterized by the inner wall. But we would like to note that the positions of resonances will depend not only on the strength of a field (as it does for the dc field [22]) but also on the frequency. Thus, by simply changing the frequency of a field, one can catch a resonance. This issue will be investigated by us elsewhere. Figure 4 shows that at large
values of field, the inelastic ‘dressing’—‘undressing’ cross sections become comparable with the elastic one, but the low frequency \( \nu/B = 0.3 \) does not allow other trap loss processes. Thus, the evaporative cooling may be successful for rather large strengths of ac field as well. But it has to be checked as for a larger field we have to take a larger Hilbert space of basis states \( |J Mn \rangle \). This means that some other inelastic channels with larger \( J, M \) and \( n \) may appear at a larger field and ‘destroy’ such a pleasant picture. But it will be very costly numerically as we will have to handle up to several thousands of coupled-channel equations. Figure 5 shows the same pattern of behavior for a higher frequency. The only difference is that there are already some inelastic channels at large fields that definitely cause trap losses.

To recapitulate, we have computed scattering cross sections for cold collisions of the bosonic polar \( ^1\Sigma \)-type molecules as functions of both collision energy and ac electric field parameters. We found that even ground-state molecules have inelastic transitions. However, most of them are harmless and allow molecules to be kept in a trap. We suggested that polar molecules in \( |J, M = -J \rangle \) states may be successfully evaporatively cooled in the appropriate region of frequencies of a microwave field and will be restrained and shielded by this field against collision losses.

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