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Collisions between linear polar molecules trapped in a microwave field

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Abstract. The collisions between linear polar molecules, trapped in a microwave field with circular polarization, are theoretically analyzed. We demonstrate that the collisional dynamics is mostly controlled by two ratios ν/B and $x = \mu_0 E_0/\hbar B$ (ν is the microwave frequency, B is the molecular rotational constant, μ_0 is the dipole moment, and E_0 is the electric field amplitude). We discuss the dependence of collision cross sections on these ratios in order to find an advantageous condition for evaporative cooling.

PACS. 34.50.Pi State-to-state scattering analyses – 33.80.Ps Optical cooling of molecules; trapping – 33.55.Be Zeeman and Stark effects

1 Introduction

The cooling and trapping of polar molecules has recently been an attractive subject (see the review in [1]) on the road to bringing intriguing realizations of novel dilute quantum gases of fermions [2–6] or bosons [7–12] and even to using them as the qubits of a scalable quantum computer [13]. Improving the precision of molecular spectra using cold polar molecules is also worthwhile. The improved accuracy of the inversion frequency of NH_3 or ND_3 molecules makes it possible to measure the time dependence of (m_e/m_p), where m_p is the proton mass and m_e is the electron mass [14,15]. The electrons of polar molecules with heavy nuclei (e.g. YbF molecules) are under a high electric field and the spectra of polar molecules are an advantageous tool for detecting violations in time-reversal symmetry.

Cold molecules have been obtained by combining laser-cooled atoms, by cooling molecules through collisions with cold gas, or by obtaining a slow molecular beam. The first of these uses photoassociation or Feshbach resonance. Kerman et al. produced ultracold RbCs* molecules through photoassociation [16]. Using Feshbach resonance, Inouye et al. [17] produced cold KRb molecules and Stan et al. [18] produced cold LiNa molecules. Molecules constructed with photoassociation or Feshbach resonance are mostly in excited states, although RbCs molecules in an absolute ground state have also been produced [19,20]. Molecular cooling through collisions with cold gas has also been developed since 1997. Using a static magnetic field, a Harvard group trapped CaH molecules, pre-cooled through collisions with helium vapor [21,22]. There has also been an idea to cool trapped molecules (with the methods discussed below) through collisions with laser-cooled atoms [23]. Several methods have been developed to obtain slow molecular beams. A deceleration method using a time-varying electric field has been elaborated to load polar molecules into a trap electrode [24–27]. Bethlem et al. loaded decelerated ND_3 molecules into a space enclosed by quadrupole electrodes [28] and Crompvoets et al. loaded them into a space enclosed by ring electrodes [29]. Van de Meerakker et al. trapped OH molecules in a quadrupole electrode using the same method [30]. Rieger et al. loaded ND_3, CH_3Cl, and CH_2O molecules selected by a quadrupole guide into an electric trap [31]. A counter-rotating beam source [32,33] or billiard-like collisions in crossed beams [34] can also be used to obtain a slow molecular beam.

The trap loss is caused by the inelastic collision or by the Majorana effect. It is preferable to trap molecules in the absolute ground state, where inelastic collisions are not possible at an ultra-low temperature. There is no Majorana effect on molecules in the $J = 0$ state either (J is the quantum number of the total rotational angular momentum). Using an AC electric field inside an octopole electrodes system, van Veldhoven et al. succeeded in trapping para-ND_3 molecules in the absolute ground state [35]. The optical field also traps absolute ground state molecules. Both these methods have problems where the trap size is very small and the trap potential is shallow.

Recently DeMille et al. proposed to trap polar molecules using a standing wave of electromagnetic field inside a microwave resonant cavity [36]. The trap size is much larger than that of the electrode and optical trap, which makes loading easy. Trapped molecules can be cooled with different methods inside the trapping area, i.e., with buffer-gas cooling or sympathetic cooling with

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laser-cooled atoms. It is also possible to construct cold molecules from laser-cooled atoms in the trapping area. The trap potential is much deeper than that in an electrode system or in an optical field.

We have previously considered the elastic and inelastic collisions [37–43] between molecules trapped by dc electric field. In this paper we discuss collisional dynamics between the cold linear polar molecules in the $^1\Sigma$ state trapped by a microwave field with a circular polarization (σ^-), that is more advantageous to obtain a deep potential than the linearly polarized microwave [36]. The collisional dynamics is mostly controlled by two ratios ν/B and $x = \mu_0 E_0/hB$ (ν is the microwave frequency, B is the molecular rotational constant, μ_0 is the dipole moment, and E_0 is the electric field amplitude). We focus on collisions at ultra-low energies and use the Born approximation for an analysis of elastic and inelastic cross sections at different regimes of a microwave field. We have found that the collisional dynamics does not look so perspective for evaporative cooling in most considered here cases. Even at small red detunings ($\nu/B \lesssim 2$) the collisional loss for bosons is large enough for a successful evaporation. This is true for any x. Only the case $\nu/B < 1$ and with a small x looks favourable for evaporative cooling. Meanwhile more deliberate calculations within the coupled-channels model may reveal some other intriguing features and will be done elsewhere.

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

Here we have analyzed the AC Stark shift, which determines the trap depth. The energy levels of $^1\Sigma$-type molecules can be described by the rotation J, total spin F (i.e., including nuclear spin), and vibration υ quantum numbers. For simplicity we will neglect hyperfine splitting and consider molecules only in the $\upsilon = 0$ vibrational ground state. So we treat polar molecules as rigid rotors with a permanent dipole moment. The AC-Stark splitting is characterized by $|J, M, n\rangle$, where M is the projection of J on the direction of the external electric field and n is the deviation of the photon number (see [36] for details). Thus the Hamiltonian for a polar $^1\Sigma$ molecule in a field is $H = H_{rot} + H_{field}$. Considering circularly polarized microwave radiation (σ^-), the non-zero Hamiltonian matrix elements (normalized by $\hbar B$) are given by

$$\langle J, M, n\,|H|\,J, M, n\rangle = J\,(J+1) + n\frac{\nu}{B} \qquad (1)$$

$$\langle J+1, M+1, n+1\,|H|\,J, M, n\rangle = \frac{x}{2}\frac{\sqrt{J+M+1}\sqrt{J+M+2}}{2\sqrt{2J+1}\sqrt{2J+3}}$$

$$\langle J+1, M-1, n-1\,|H|\,J, M, n\rangle = \frac{x}{2}\frac{\sqrt{J-M+1}\sqrt{J-M+2}}{2\sqrt{2J+1}\sqrt{2J+3}}. \qquad (2)$$

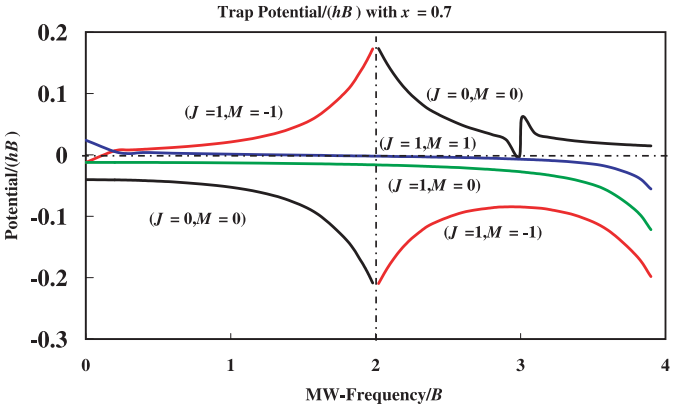

Fig. 1. (Color online) The trap potentials of the linear polar molecules in the $J = 0$ and 1 states are shown as functions of the microwave frequency for $x = \mu_0 E_0/\hbar B = 0.7$.

Here, it is assumed that n is much smaller than the mean photon number N. In the AC-electric field the $J, M, N+n$ are not good quantum numbers and the dressed state formalism should be applied. For example, the lowest energy state with a given mean photon number N (trapped molecules are mostly in this state) is described as $\Phi_i\,(N) = \sum p\,(J, M, n)\,|J, M, n\rangle$. For the ($\nu/B < 2$, $x \ll 1$) case $p\,(J = 0) \approx 1$ and this state is an almost pure $J = 0$ state. For the $2 < \nu/B < 4$ case the lowest state becomes almost pure $(J = 1, M = -1)$ one. At $\nu/B \approx 2$, $\Phi_i\,(N)$ is almost a half and half mixing of $|0, 0, 0\rangle$ and $|1, -1, 1\rangle$ states because of the crossing of these two levels. By changing the microwave frequency from ν_i ($\nu_i < 2B$) to ν_f ($2B < \nu_f < 4B$) , trapped molecules transform from the $|0, 0, 0\rangle$ state to $|1, -1, -1\rangle$ state. Figure 1 shows the dependence of the trap potentials of the molecules in the AC electric field for the $J = 0$ and 1 states on the microwave frequency at $x = 0.7$. The trap potentials are defined as the difference of eigenvalues of the Hamiltonian H and diagonal Hamiltonian matrix elements. The calculations were done taking $0 \le J \le 5$ ($-J \le M, n \le J$) states into account. At $\nu/B < 2$, the $|0, 0, 0\rangle$ state is dominant at $\Phi_i\,(N)$. For $2 < \nu/B < 4$ case $\Phi_i\,(N)$ is dominated by the $|1, -1, -1\rangle$ state. As ν/B gets close to four, the mixing between $|1, -1, -1\rangle$ and $|2, -2, -2\rangle$ states becomes significant and for $\nu/B > 4$ the $\Phi_i\,(N)$ is dominated by the $|2, -2, -2\rangle$ state. So the larger frequency of the AC electric field, the larger angular moment J of the state $\Phi_i\,(N)$ will be dominated. The depth of the potential can be also changed by altering the amplitude of the AC-field for a fixed frequency as well. Figures 2a and 2b show the AC Stark energy shifts as functions of x for $\nu/B = 0.3$ and 2.3 cases respectively. The potential depth of the ground state can be larger than hB for $x > 3$.

3 Collisions of cold trapped molecules

We will consider the scattering of two polar molecules in a microwave field. A theoretical description of molecule-molecule scattering is complicated by the complexity of

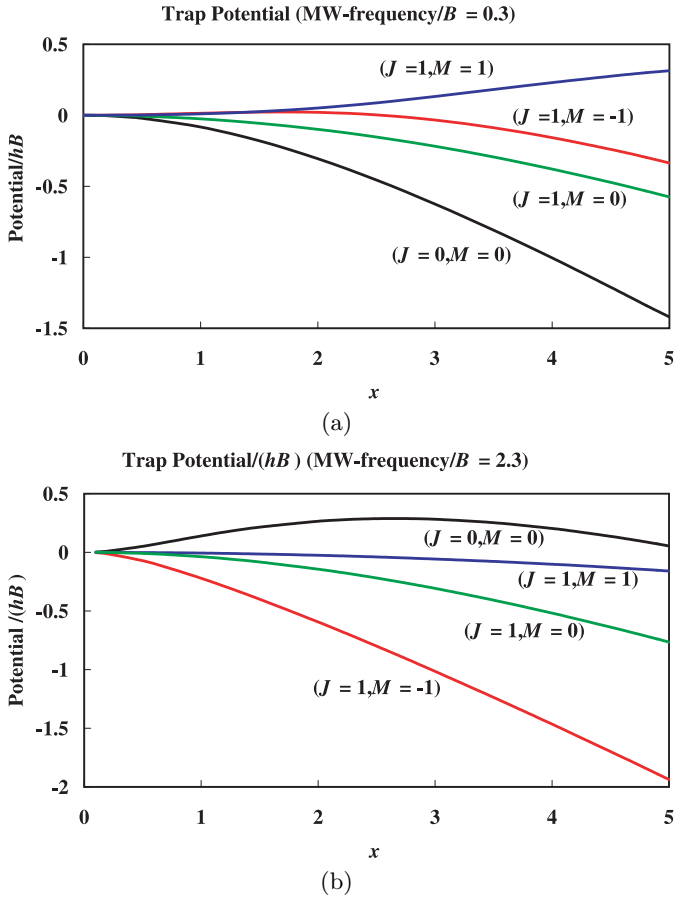

Fig. 2. (Color online) The trap potentials of linear polar molecules in the $J = 0$ and 1 states versus x $(=\mu_0 E_0/\hbar B)$ at a microwave frequency of (a) $0.3B$ and (b) $2.3B$ respectively.

the short-range interaction between molecules. It is therefore worthwhile to seek situations in which the influence of short-range physics is minimal and the bulk effects are ruled by the dipolar part of interaction [41–43]. Here we assume that the intermolecular interaction is only dominated by the dipole-dipole interaction as well and discuss the use of simplified methods, i.e., the Born approximation with ultra-low kinetic energy. Though in some cases these simplified methods might give rather rough approaches they are useful to understand the dependence of cross sections on the molecular constants (e.g., permanent dipole moment, mass, and rotational constant) quite transparently.

The cross-sections of processes $|\Phi_{i1}, \Phi_{i2}\rangle \rightarrow |\Phi_{f1}, \Phi_{f2}\rangle$ caused by collisions between the same kinds of molecules are obtained from

$$\sigma\left[|\Phi_{i1}, \Phi_{i2}\rangle \rightarrow |\Phi_{f1}, \Phi_{f2}\rangle\right] = \sum_{L,M_L}\sum_{L',M'_L} \frac{8}{\left[1+\delta\left(\Phi_{i1}, \Phi_{i2}\right)\right]\left[1+\delta\left(\Phi_{f1}, \Phi_{f2}\right)\right]} \times \frac{\pi}{k^2} P\left[|\Phi_{i1}, \Phi_{i2}\rangle \rightarrow |\Phi_{f1}, \Phi_{f2}\rangle, (L, M_L) \rightarrow (L', M'_L)\right] \quad (3)$$

where k is the incident wave number and L (L') are even for bosons and odd for fermions. $M_L(M'_L)$ are the quantum numbers for the total angular momentum of relative motion and its trajectory parallel to the electric field before (and after) the collision, respectively. P is the opacity function [40]. In the Born approximation this function is given by

$$\frac{\pi}{k^2} P\left[|\Phi_{i1}, \Phi_{i2}\rangle \rightarrow |\Phi_{f1}, \Phi_{f2}\rangle, (L, M_L) \rightarrow (L', M'_L)\right] = \frac{m^2}{16\pi\varepsilon_0^2\hbar^4} G_{L,L'}\left(\frac{k'}{k}\right) F\left(\Delta M_{J1}, \Delta M_{J2}, L, M_L, L', M'_L\right) \quad (4)$$

$$G_{L,L'}\left(\frac{k'}{k}\right) = \frac{k'}{k}\left[\int_0^\infty j_L^*(kr)\,\frac{1}{r}\,j_{L'}(k'r)\,dr\right]^2$$

$$\frac{k'}{k} = \sqrt{1+\frac{\Delta E}{K}}, K = \frac{(\hbar k)^2}{m}$$

where k' denotes the wave number of the scattered wave, m is the molecular mass, and ΔE is the change of the total internal energies, including the AC Stark shift. F is an operator, proportional to $|\langle\Phi_{i1}|\vec{\mu}|\Phi_{f1}\rangle|^2 |\langle\Phi_{i2}|\vec{\mu}|\Phi_{f2}\rangle|^2$ [39]. As the different values of the molecular spin J are mixed in a field, in practice we transform the molecular state into a field-dressed basis for performing scattering calculations, $|\Phi_\alpha\rangle = \sum_\alpha c_\alpha |\Phi_\alpha\rangle_0$ where $|\Phi_\alpha\rangle_0$ is the wave function without an electric field and c_α where α stands for the eigenfunctions of the Hamiltonian $H = H_{rot} + H_{field}$ determined numerically at each value of the field and the frequency. We will continue to refer to molecular states by the quantum numbers J, and M with the understanding that they are only approximately good in a field, and that $|\Phi_\alpha\rangle$ is the appropriate molecular state. The eigenfunctions only depend on the values of x and ν/B. Following the $m^2\mu_0^4$ dependence of the cross-section (4) it is appropriate to define the reduced cross section as $\rho = \sigma/m^2\mu_0^4$. At ultracold energies the Born approximation yields the following behavior [39]

$$\rho \propto \left(1+\frac{\Delta E}{K}\right)^{1/2} \quad \text{for bosons}$$
$$\rho \propto \left(1+\frac{\Delta E}{K}\right)^{-1/2} \quad \text{for fermions.} \quad (5)$$

Therefore, the elastic collision cross section, for which $\Delta E = 0$, does not depend on K. At $\Delta E \gg K$, the inelastic collision cross sections are proportional to $K^{-1/2}$ for boson molecules and proportional to $K^{1/2}$ for fermion molecules. The validity of the Born approximation was discussed in reference [42]. The same arguments hold for the present paper. Following the criteria derived there and estimating the dipole-dipole interaction as μ_0^2/r^3, assuming that the condition is satisfied when

$$K < \frac{32}{27}\frac{16\pi^2\varepsilon_0^2\hbar^6}{m^3\mu_0^4}. \quad (6)$$

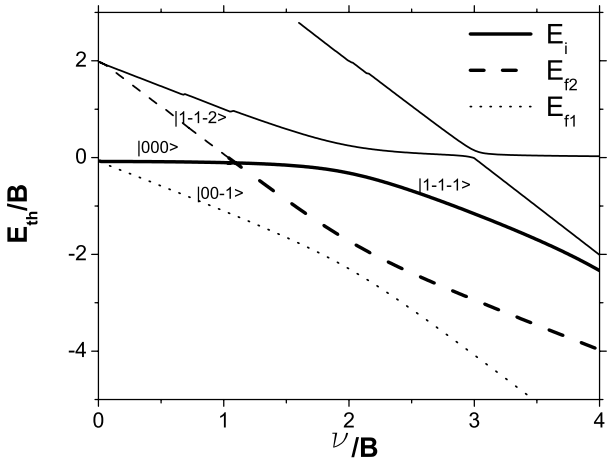

Fig. 3. The threshold energies of the Φ_{i,f_1,f_2} states.

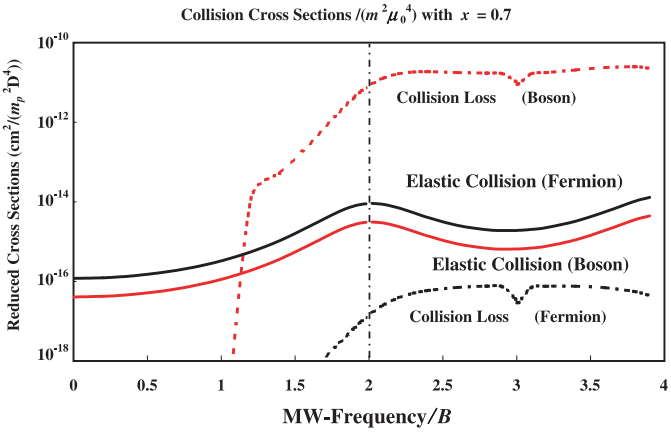

Fig. 4. (Color online) The reduced collision cross sections between trapped molecules versus ν/B at $x = \mu_0 E_0/\hbar B = 0.7$. The collision kinetic energy is taken as $10^{-6}\hbar B$. Solid lines indicate the reduced elastic collision cross sections and dotted lines indicate the reduced cross section of the $\Phi_i \to \Phi_f$ transition. The thick and thin lines indicate the collision between the fermion and boson molecules, respectively.

This criteria is valid when $K/k_B < 2$ nK for ClCN molecules ($\mu_0 = 2.8$ D, $m = 61m_p$). However, the dipole-dipole interaction is actually much weaker than μ_0^2/r^3 as the effective potential C_{eff}/r^3 is defined by the mixing of different partial waves [41] and the Born approximation is also valid within the region of higher kinetic energy. The above criterion is more evident for fermionic species as they have centrifugal barriers for any odd partial waves. But for bosonic species we have to keep in mind that the $L = 0 \to L' = 0$ contribution to a realistic dipole-dipole interaction rigorously vanishes by symmetry. Moreover as the threshold behavior of elastic collision is similar for both bosons and fermions (4), we may assume that their behavior would not different at ultracold energies even when taking into account the short range interaction. This still leaves open the possibility that some accidental resonance arises a situation not treated in this work.

In this paper, we discuss the results of calculations taking $L \to L' = 0,2 \to 0,2$ and $1 \to 1,3$ into account for boson and fermion molecules respectively. The trapped molecules are considered at $x = 0.7$ and within $\nu/B = [0,4]$ frequency range. In this case the state of interest is approximately described as a mixture of $|000\rangle$ and $|1-1-1\rangle$ states:

$$\Phi_i \approx p_1|000\rangle + p_2|1-1-1\rangle; \quad p_1^2 + p_2^2 \approx 1.$$

The contribution of other states is small. The main difference from the DC electric field case is that in general inelastic processes are always allowed for any a non-zero field. Of course an inelastic cross section will be very small at small fields but not exactly zero. In order to understand it let us consider the energies of the dressed states near our state versus ν/B Figure 3. They are

$$\Phi_{f1} \approx q_1|00-1\rangle + q_2|1-1-2\rangle$$
$$\Phi_{f2} \approx q_2|00-1\rangle + q_1|1-1-2\rangle$$
$$q_1^2 + q_2^2 \approx 1.$$

The AC-electric field does not mix Φ_i with $\Phi_{f_{1,2}}$. But the dipole-dipole interaction can cause a $\Phi_i \to \Phi_{f_{1,2}}$ transition. The $\Phi_i - \Phi_{f_1}$ transition is always possible as $E_i - E_{f1} \approx \hbar\nu$. But the matrix element $\langle\Phi_i|\mu|\Phi_{f_1}\rangle = p_2q_1\langle 1-1-1|\mu|00-1\rangle$ is not negligible only near the adiabatic crossing of $\Phi_{f_{1,2}}$ states near $\nu/B = 2$ where $p_2q_1 \approx 1/2$. The $\Phi_i \to \Phi_{f_2}$ transition is only possible for $\nu/B > 1$ (see Fig. 3) and the collisional kinematics $E_i - E_{f2}$ is approximately defined as

$$\begin{array}{ll} 2\hbar\,(\nu - B) & \text{for } \nu/B < 2 \\ 2\hbar\,(B) & \text{for } 2 < \nu/B < 4. \end{array}$$

Of course these expressions should be corrected near adiabatic crossings but it is not important for not very large fields ($x < 1$). The matrix element $\langle\Phi_i|\mu|\Phi_{f_2}\rangle \approx p_2q_2\langle 1-1-1|\mu|00-1\rangle$ near $\nu/B = 2$ where $p_2q_2 \approx 1/2$ and it is very small at $1\langle\nu/B \lesssim 2$ where $p_2q_2 \ll 1$. But in the $2\langle\nu/B\langle 4$ case this matrix element is almost pure $\langle 1-1-1|\mu|00-1\rangle$ and an inelastic transition is only defined by this matrix element. Figure 4 shows the reduced collision cross sections between trapped molecules as functions of ν/B for $x = 0.7$. When $\nu/B \approx 1$ (trapped molecules are mostly in the $|J = 0, M = 0\rangle$ state), inelastic collisions are negligible at the kinetic energy $K \lesssim 2\hbar B$. Then the inelastic collision cross section is growing drastically after this point up to $\nu/B \approx 2$ (where the above described adiabatic crossing appears) and is staying rather steady further. The reduced elastic collision cross section for fermionic molecules is larger than that for boson molecules with a factor of 2.7, when only dipole-dipole interaction is considered. The inelastic cross section for fermions is considerably smaller than for bosons simply reflecting the threshold behavior [42]. At larger fields (larger x) the general behavior of the reduced cross section is qualitatively similar.

Figure 5 shows the reduced elastic cross section for molecules in the Φ_i state as functions of x for different values ν/B. The reduced elastic collision cross sections

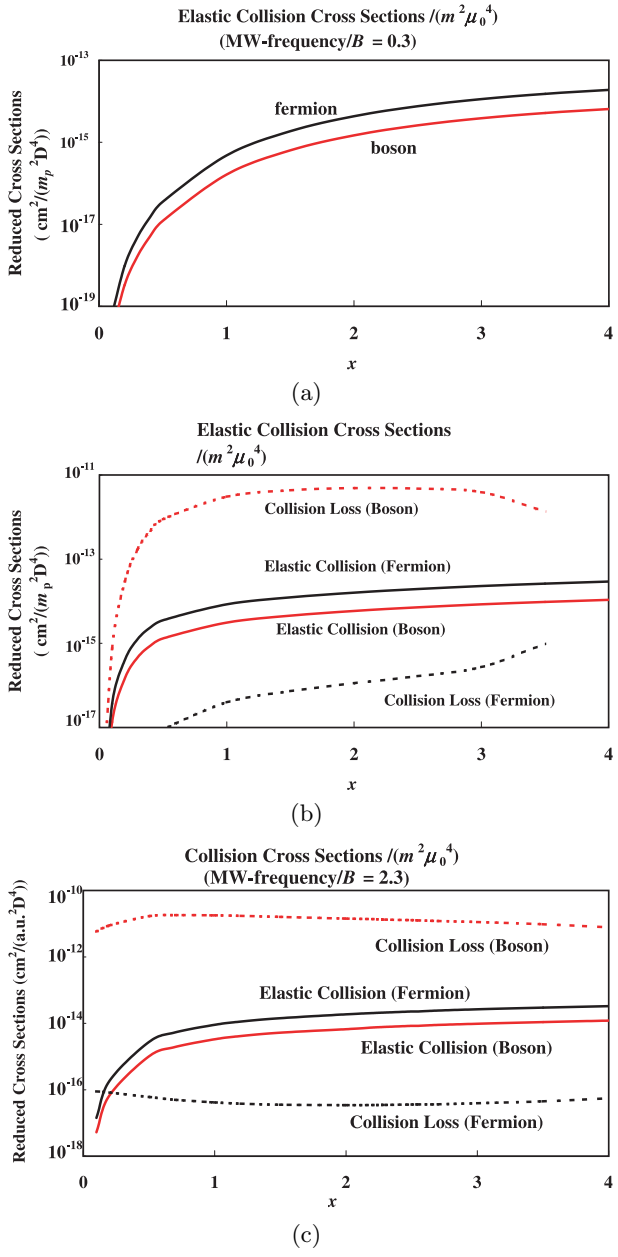

Fig. 5. (Color online) The reduced collision cross-sections for the trapped molecules in the Φ_i state (see the text) versus x for the microwave frequency of (a) $0.3B$, (b) 1.8 and (c) $2.3B$. The collision kinetic energy is taken as $10^{-6}\hbar B$. The solid lines are for the reduced elastic collision cross sections and the dotted lines are for the reduced cross section of the $\Phi_i \to \Phi_f$ transition. The thick (black) and thin (red) curves are for fermionic and bosonic molecules respectively.

are proportional to x^4 at $x \lesssim 0.5$, as derived through the first-order perturbation theory. When $3 < x < 5$, the reduced elastic collision cross section is almost proportional to x. One can see that the significant inelastic cross section appears both for red detuning (Fig. 5b) where $\nu/B = 1.8, \Delta = \nu - 2B/h < 0$ and blue detuning (Fig. 5c) where $\nu/B = 2.3, \Delta = \nu - 2B/h > 0$.

As for the practical size and depth of the trap the conditions are $x \gtrsim 1$ and $\nu/B \approx 1$ [36] we conclude that evaporative cooling will always be successful for the fermionic species for the above considered AC-fields. The evaporative cooling for bosons can only be successful at $\nu/B < 1$ and the inelastic cross section is quite sensitive to x near such a frequency.

4 Conclusion

In this paper we have discussed collision cross sections for polar $^1\Sigma$ type molecules trapped in a microwave field. We have analyzed the collisional dynamics in dependence on external AC-field within the Born approximation, which have rather a simple dependence on parameters such as the permanent dipole moment, mass, and rotational constant.

We have shown that the trap loss caused by the inelastic collision can be significant for bosonic molecules not only for fields with $\nu > 2B$ but and for fields with $B < \nu < 2B$. So the inelastic collision is only suppressed in cases when $\nu/B < 1$. Moreover it is quite sensitive to the amplitude of the AC field near this point. It has been demonstrated that evaporative cooling at ultracold energies should be successful at almost any parameters of the AC-field for fermionic species.

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