Collisions between molecules in the J = 0 state

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Abstract. Collisions between electrostatically trapped molecules in the J = 0 state are theoretically investigated. Molecules in this state are advantageous for the evaporative cooling, because inelastic collision is impossible at ultra-low temperature. However, inelastic collision is more significant than elastic collision at high temperature, because the off-diagonal dipole matrix elements are much larger than the diagonal elements. The elastic and inelastic collision cross-sections are calculated under the condition that the collisional kinetic energy is larger than the energy gap between the rotational states. The dependencies of the collision cross-sections on the collisional kinetic energy and molecular constants are examined.

PACS. 31.15.Qg Molecular dynamics and other numerical methods -33.80.Ps Optical cooling of molecules; trapping -33.90.+h Other topics in molecular properties and interactions with photons

1 Introduction

It has traditionally been considered difficult to produce ultra-cold molecules, mainly because it is difficult to decelerate molecules by laser cooling. It seems possible, however, to cool molecules by evaporative cooling. Using only evaporative cooling, Fried et al. cooled hydrogen atoms enough to obtain Bose-Einstein-Condensation (BEC) [1]. To perform evaporative cooling, it is necessary to trap molecules with high density beforehand. Several groups have recently developed effective methods for preparing ultra-cold molecules and confining them in threedimensional traps. A Harvard group used static magnetic fields to trap paramagnetic CaH molecules that had been pre-cooled by buffer-gas collisions [2,3]. Takekoshi et al. trapped cesium dimers produced by photo-associating laser-cooled Cs atoms with focused CO_2 laser beams [4]. Bethlem et al. decelerated ND₃ molecular beams with a time-varying inhomogeneous electric field and loaded them into an electrostatic trap [5].

To perform evaporative cooling, the elastic collision rate should be high and the trap loss rate should be low [6]. When molecules are trapped by a dc-electric field, only those in the low-field-seeking states are trapped, and trap loss is caused by the transitions to the high-field-seeking states. We have analyzed the loss rate of the linear polar molecules in the $(J, M_J) = (1, 0)$ state, which result from the Majorana effect (the transition between quantum states caused by a change in the electric field direction) [7] and the inelastic collision [8,9]. Here, J denotes the quantum number of the total molecular angular momentum and M_J is the quantum number of the trajectory of the molecular angular momentum parallel to the electric field. It seems possible to perform evaporative cooling with fermion molecules, as the ratio of the collision loss rate to the elastic collision rate (R) decreases as the collisional kinetic energy (k_BT) decreases, for any range of T [10]. With boson molecules in low-field-seeking states, however, it is rather difficult to perform evaporative cooling, since R increases as T decreases with $T < 100 \ \mu \text{K}$ [9–12].

Molecules in high-field-seeking states can be trapped with a storage ring formed by alternate-gradient-focusing electrodes [13]. It has also been proposed to trap molecules in the high-field-seeking states in a microwave cavity [14]. These approaches are used mainly to trap molecules in the J = 0 state. When J = 0, the Majorana effect does not occur, and inelastic collision is also impossible when $k_BT < 2hB$, where B is the molecular rotational constant. Therefore, the collision loss rate is much lower than with molecules in the $J \ge 1$ states for both boson and fermion isotopes. This paper applies a semi-classical treatment to investigates the elastic and inelastic collision of molecules in the J = 0 state for 5 K < T < 100 K. When J = 0, the non-linear molecules can also be analyzed by using the same formula as that for linear polar molecules [10].

2 Calculation of collision cross-section

When the molecular temperature is high enough that the broadening of the molecular wavepacket is negligibly small, the cross-section of the collisional process

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 $(J_1, M_{J_1}) + (J_2, M_{J_2}) = (0, 0) + (0, 0) \rightarrow (J_1^p, M_{J_1}^p) +$ where $|J, M\rangle_0$ denotes the wave function in field-free (J_1^p, M_2^p) is obtained as (J_2^p, M_{J2}^p) is obtained as

$$\sigma_{\left(J_{1}^{p},M_{J_{1}}^{p},J_{2}^{p},M_{J_{2}}^{p}\right)} = \int_{0}^{\infty} 2\pi b P_{\left(J_{1}^{p},M_{J_{1}}^{p},J_{2}^{p},M_{J_{2}}^{p}\right)}(b) \, db \quad (1)$$

where P is the opacity function [15] and b is called the impact parameter. Under the assumption of a classical path, P is given by

$$P_{\left(J_{1}^{p},M_{J_{1}}^{p},J_{2}^{p},M_{J_{2}}^{p}\right)}\left(b\right) = \left|\frac{1}{\hbar}\int_{-\infty}^{\infty}\left\langle0,0,0,0\left|H\left(r,\theta,\varphi\right)\right|J_{1}^{p},M_{J_{1}}^{p},J_{2}^{p},M_{J_{2}}^{p}\right\rangle dt\right|^{2}$$

$$r = \sqrt{b^2 + (vt)^2}$$
$$v = \sqrt{\frac{2k_BT}{m}}$$
(2)

where H is the intermolecular Hamiltonian. If the interaction between two linear polar molecules is mainly caused by the dipole-dipole interaction, the matrix element of His given by

$$\langle J_1, M_{J1}, J_2, M_{J2} | H(r, \theta, \varphi) | J'_1, M'_{J1}, J'_2, M'_{J2} \rangle = \frac{1}{4\pi\varepsilon_0 r^3} \bigg[\langle J_1, M_{J1} | \overrightarrow{\mu} | J'_1, M'_{J1} \rangle \cdot \langle J_2, M_{J2} | \overrightarrow{\mu} | J'_2, M'_{J2} \rangle - \frac{3}{r^2} (\langle J_1, M_{J1} | \overrightarrow{\mu} | J'_1, M'_{J1} \rangle \cdot \overrightarrow{r}) \times (\langle J_2, M_{J2} | \overrightarrow{\mu} | J'_2, M'_{J2} \rangle \cdot \overrightarrow{r}) \bigg].$$
(3)

For non-linear molecules, K_J , the quantum number of the trajectory of the molecular angular momentum parallel to the molecular axis, must also be considered. When J = 0, however, K_J is always zero. Also, if J is changed by the collision related dipole transition, K_J must also be zero after the collision. When $K_J = 0$, non-linear molecules can be analyzed by applying the same formula as with linear molecules. In field-free space, the following matrix elements $\langle 0, 0 | \overrightarrow{\mu} | J^p, M_J^p \rangle$ are non-zero:

$$\langle 0, 0 | \mu_z | 1, 0 \rangle = \frac{\mu}{\sqrt{3}}$$
$$\langle 0, 0 | \mu_x \pm i \mu_y | 1, \pm 1 \rangle = \mp \mu \sqrt{\frac{2}{3}}$$
(4)

where μ is the molecular permanent dipole moment. When the electric field is given, $\langle 0, 0 | \overrightarrow{\mu} | 0, 0 \rangle$ also becomes nonzero because of the mixture of the wave functions. According to the first-order perturbation theory, the wave function $|0,0\rangle$ under an electric field is given by

$$|0,0\rangle = |0,0\rangle_0 - \frac{\langle 0,0 |\mu| |1,0\rangle E}{2hB} |1,0\rangle_0$$
(5)

space and *E* is the electric field strength. $\langle 0, 0 | \overrightarrow{\mu} | 0, 0 \rangle$ is given by

$$\langle 0, 0 | \mu_z | 0, 0 \rangle = \frac{\mu^2 E}{3hB}.$$
 (6)

When the electric field is low enough $(\mu E < hB)$, (4) and (6) can be rewritten as follows by using the Stark energy shift of trapped molecules given by $U (= \mu^2 E^2/6hB)$:

$$\left|\left\langle 0,0\left|\overrightarrow{\mu}\right|1,0\right\rangle\right|^{2} = \left|\left\langle 0,0\left|\overrightarrow{\mu}\right|1,\pm1\right\rangle\right|^{2} = \frac{\mu^{2}}{3}$$
$$\left|\left\langle 0,0\left|\overrightarrow{\mu}\right|0,0\right\rangle\right|^{2} = \frac{2\mu^{2}}{3hB}U.$$
(7)

The collision cross-sections were calculated by applying the Quantum-Fourier-transform (QFT) theory [16]

$$P_{\left(J_{1}^{p},M_{J_{1}}^{p},J_{2}^{p},M_{J_{2}}^{p}\right)}\left(b\right) = \min\left(1,Q_{\left(J_{1}^{p},J_{2}^{p}\right)}\left(b\right)\right).$$
 (8)

Here $Q_{\left(J_{1}^{p},J_{2}^{p}\right)}(b)$ is obtained using the first-order Born approximation shown below. With small b (i.e., strong interaction), the first Born approximation is not valid and $P_{\left(J_{1}^{p},M_{J_{1}}^{p},J_{2}^{p},M_{J_{2}}^{p}
ight)}\left(b\right)=1$ is used for simplicity $\left(P\left(b\right)\leq1\right)$ must always hold) [17]. Assuming $\mu E < hB$, $Q_{(J_1^p, J_2^p)}(b)$ is obtained as follows:

(1)

л

$$\begin{aligned} Q_{\left(J_{1}^{p},J_{2}^{p}\right)}\left(b\right) &= D_{\left(J_{1}^{p},J_{2}^{p}\right)}\left(b\right)f_{\left(J_{1}^{p},J_{2}^{p}\right)}\left(b\right)\\ D_{\left(J_{1}^{p},J_{2}^{p}\right)}\left(b\right) &= \frac{32\alpha^{2}\pi m}{27h^{2}k_{B}Tb^{4}}\left|\langle0,0\left|\overrightarrow{\mu}\right|J_{1}^{p},M_{J1}^{p}\rangle\right|^{2}\\ &\times\left|\langle0,0\left|\overrightarrow{\mu}\right|J_{2}^{p},M_{J2}^{p}\rangle\right|^{2}\\ f_{\left(J_{1}^{p},J_{2}^{p}\right)}\left(b\right) &= \exp\left[-\frac{4\Delta_{\left(J_{1}^{p},J_{2}^{p}\right)}^{2}}{\pi\alpha^{2}}b^{2}\right]\\ \Delta_{\left(J_{1}^{p},J_{2}^{p}\right)} &= 2\pi B\sqrt{\frac{m}{2k_{B}T}}\left[J_{1}^{p}\left(J_{1}^{p}+1\right)+J_{2}^{p}\left(J_{2}^{p}+1\right)\right]. \end{aligned}$$
(9)

Here, m is the molecular reduced mass and α is an arbitrary (dimensionless) constant, that cannot be determined by QFT theory alone. Davies recommends taking $\alpha = \sqrt{3\pi/4}$ (QFT-I), so that the formula for P(b) converges to that of the Anderson-Tsao-Curnutte (ATC) theory with $\Delta \to 0$ [16–18]. Q and $\sigma_{(J_1^p, M_{J_1}^p, J_2^p, M_{J_2}^p)}$ depend only on J_1^p and J_2^p , as $|\langle 0, 0 | \overrightarrow{\mu} | 1, M_J^p \rangle|^2$ does not depend on M^p_{I} .

When the collision is between the same kinds of molecules, the wave function must be transformed to the symmetric form

boson:

$$|J_1, M_{J1}, J_2, M_{J2}\rangle \to \frac{1}{\sqrt{2(1 + \delta(J_1, J_2)\delta(M_{J1}, M_{J2}))}} \times (|J_1, M_{J1}, J_2, M_{J2}\rangle + |J_2, M_{J2}, J_1, M_{J1}\rangle)$$

Table 1. Molecular permanent dipole moment (μ), rotational constant (B), the constant (γ and γ') with which the elastic collision cross-section is determined from (14) and (16), and T_c at which (20–23) are valid with $T \ll T_c$, for various molecules.

	μ (D)	B (GHz)	$\gamma \ (\mathrm{cm}^2 \mathrm{K}^{-1/2})$	$\gamma'~({\rm cm}^2{\rm K}^{1/5})$	T_c (K)
OCS	0.7	6.1	2.54×10^{-12}	1.83×10^{-13}	15.3
$\mathrm{CH}_{3}\mathrm{F}$	1.8	26	2.86×10^{-12}	4.01×10^{-13}	201
HCN	2.9	44	3.91×10^{-12}	6.88×10^{-13}	589
NaCl	8.5	6.5	3.27×10^{-10}	9.26×10^{-12}	425

fermion:

$$J_{1}, M_{J1}, J_{2}, M_{J2} \rangle \to \frac{1}{\sqrt{2 (1 + \delta (J_{1}, J_{2}) \delta (M_{J1}, M_{J2}))}} \times (|J_{1}, M_{J1}, J_{2}, M_{J2}\rangle - |J_{2}, M_{J2}, J_{1}, M_{J1}\rangle).$$
(10)

The elastic collision cross-section is given by

$$\sigma_{elastic} = \sigma_{(0,0,0,0)}.\tag{11}$$

Using alternate-gradient-focusing electrodes, molecules in both the high-field- and low-field-seeking states are trapped. The trapping force for molecules with $J \ge 1$, however, is lower than that for molecules in the J = 0state (with a factor of 0.6 for the (J, M) = (1, 0) state and 0.3 for the $(1, \pm 1)$ states). We therefore consider the trap loss to result from the collisional transition to the J = 1 state. The collision loss cross-section is given by

$$\sigma_{loss} = 2\sigma_{(0,0,1,0)} + 2\sigma_{(0,0,1,1)} + 2\sigma_{(0,0,1,-1)} + 4\sigma_{(1,0,1,1)} + 4\sigma_{(1,0,1,-1)} + 2\sigma_{(1,0,1,0)} + 4\sigma_{(1,1,1,-1)} + 2\sigma_{(1,1,1,1)} + 2\sigma_{(1,-1,1,-1)} = 6\sigma_{(0,0,1,0)} + 18\sigma_{(1,0,1,0)}.$$
(12)

The coefficients in (12) are obtained by the treatment given by (10) and under the condition that two molecules are lost through one collision when $(J_1^p, J_2^p) = (1, 1)$. The values of $\sigma_{elastic}$ and σ_{loss} for OCS, CH₃F, HCN, and NaCl molecules can be obtained using the values of μ and *B* listed in Table 1.

The elastic collision cross-section is obtained by taking $f_{(0,0)}(b) = 1$ and $Q_{(0,0)}(b) = D_{(0,0)}(b)$. The elastic collision cross-section obtained considering just the dipoledipole interaction is given by

$$\sigma_0 = 2\pi b_0^2 = \frac{8\pi^2}{9h^2} \sqrt{\frac{2m}{k_B T}} \frac{\mu^2}{B} U \tag{13}$$

holds [17] where b_0 is the value of b for which $D_{(0,0)}(b_0) = 1$. Equation (13) can be rewritten as

$$\sigma_0 \left(\mathrm{cm}^2 \right) = \gamma \frac{U}{\sqrt{T}} \left(\mathrm{K}^{1/2} \right) \tag{14}$$

where γ is a molecular constants listed in Table 1 for OCS, CH₃F, HCN and NaCl molecules. When the electric field is low, the elastic collision cross-section is actually more dominated by the dipole-induced dipole interaction ($\propto r^6$) than the dipole-dipole interaction. Considering just

the dipole-induced dipole interaction, the elastic collision cross-section is obtained as follows [17]:

$$\sigma_s = \frac{5}{4}\pi \left[\frac{7\pi^4}{960h^4} \frac{m}{k_B T} \frac{\mu^8}{B^2}\right]^{1/5}$$
(15)

$$\sigma_s \left(\mathrm{cm}^2 \right) = \frac{\gamma'}{T^{1/5}} \left(\mathrm{K}^{-1/5} \right) \tag{16}$$

where γ' is a molecular constant listed in Table 1. The actual elastic collision cross-section is approximately obtained by

$$\sigma_{elastic} = \max\left(\sigma_0, \sigma_s\right). \tag{17}$$

The collision loss cross-section is obtained using

$$D_{(0,1)}(b) = \frac{16\pi^2 m}{81h^2 k_B T b^4} \frac{\mu^4}{hB} U$$
$$D_{(1,1)}(b) = \frac{8\pi^2 m}{81h^2 k_B T b^4} \mu^4$$
$$f_{(0,1)}(b) = \exp\left[-\frac{128mB^2}{3k_B T} b^2\right]$$
$$f_{(1,1)}(b) = \exp\left[-\frac{512mB^2}{3k_B T} b^2\right].$$
(18)

Equation (18) is valid only when $k_B T > 2hB$ so that the collisional transition $J = 0 \rightarrow 1$ is possible keeping the total energy constant. The dipole-induced dipole interaction (r^{-6}) cannot cause the collision loss with zero electric field. Also the dipole-induced dipole interaction can cause the collision loss when electric field is applied, but it is negligible when $\mu E \ll hB$. The following two parameters must be considered in discussing the role of f(b):

$$b_{c} = \frac{1}{8B} \sqrt{\frac{3k_{B}T}{2m}}$$
$$T_{c} = \frac{2^{17/3} \pi^{2/3} m}{9k_{B}} \left(\frac{\mu^{2}B^{2}}{h}\right)^{2/3}.$$
(19)

Here, b_c is the value of b satisfying $f_{(0,1)}(b_c) = 1/e$, and T_c is the value of T satisfying $D_{(1,1)}(b_c) = 1$. The values of T_c are listed in Table 1. When $T \ll T_c$, b_c is the value of the parameter giving

$$Q(b) \ll 1 \quad b \gg b_c$$

$$Q(b) \gg 1 \quad b \ll b_c$$
(20)

because

$$f(b) \approx 1 \quad b < b_c$$

$$f(b) \ll 1 \quad b > b_c.$$

Then

$$\sigma_{loss} \propto \pi b_c^2 = \frac{3k_B T}{32mB^2}.$$
 (21)

When $\sigma_{elastic} = \sigma_s$,

$$R = \frac{\sigma_{loss}}{\sigma_{elastic}} \propto \frac{T^{6/5}}{\mu^{8/5} m^{6/5} B^{8/5}}$$
(22)

and when $\sigma_{elastic} = \sigma_0$,

$$R \propto \frac{T^{3/2}}{\mu^2 m^{3/2} BU} \cdot \tag{23}$$

When $T \gg T_c$, σ_{loss} is obtained by taking $f(b) \approx 1$ and

$$\sigma_{loss} = \frac{8\pi^2\mu^2}{3h}\sqrt{\frac{2m}{k_BT}} + \frac{8\pi^2\mu^2}{h}\sqrt{\frac{mU}{k_BThB}}$$

with $U \ll hB$

$$\approx \frac{8\pi^2 \mu^2}{3h} \sqrt{\frac{2m}{k_B T}} \tag{24}$$

$$R \propto \frac{\mu^{2/5} m^{3/10} B^{2/5}}{T^{3/10}} \quad \sigma_{elastic} = \sigma_s \tag{25}$$

$$R \approx \frac{3hB}{U} \qquad \qquad \sigma_{elastic} = \sigma_0 \qquad (26)$$

is obtained.

R is obtained by numerical calculation using (1–18). Figure 1 shows R for the OCS, CH₃F, HCN and NaCl molecules as a function of T (6 K < T < 100 K). This area of T is chosen so that $k_B T > 2hB$ is satisfied and (18) is valid. When $k_BT < 2hB$, (18) is not valid and R becomes zero. U was taken as 0.03 K, which corresponds to the electric field strength 13.3 kV/cm (OCS), 11.0 kV/cm (CH_3F) , 8.80 kV/cm (HCN), and 1.16 kV/cm (NaCl), respectively. With this value of U, $\sigma_{elastic} = \sigma_s$ for all these molecules. For CH_3F , HCN, and NaCl molecules, R is roughly proportional to $T^{0.7}$, though (22) shows $R \propto T^{1.2}$. This is because T is not low enough that (22) is valid. For OCS molecule, whose T_c is quite low at 15.3 K, R decreases as T increases when T > 40 K. But $R \propto T^{-3/10}$ (shown in (25)) is actually not valid when T < 100 K. According to (22), R should be proportional to $(\mu^{-8/5}m^{-6/5}B^{-8/5})$. Figure 2 shows that R is actually roughly proportional to $(\mu^{-8/5}m^{-6/5}B^{-8/5})^{0.6}$ when U = 0.03 K and T = 10 K.

At ultra-low collisional kinetic energy ($T < 100 \ \mu \text{K}$), (1) is not valid because of the broadening of the molecular wavepacket. The elastic collision cross-sections with ultra-low collisional kinetic energy are discussed in reference [10] for boson and fermion isotopes.



Fig. 1. $R = \sigma_{loss}/\sigma_{elastic}$ for OCS, CH₃F, HCN, and NaCl molecule as a function of the collisional kinetic energy (T), where the electrostatic potential of the trapped molecule (U) is 0.03 K.



Fig. 2. $R = \sigma_{loss}/\sigma_{elastic}$ as a function of $\mu^{-8/5}m^{-6/5}B^{-8/5}$, where the electrostatic potential of the trapped molecule (U) is 0.03 K and the collisional kinetic energy (T) is 10 K.

3 Conclusion

The loss of electrostatically trapped molecules is caused by the Majorana effect and inelastic collision. Therefore, molecules in the J = 0 state seems to be advantageous for trapping with high stability, because the Majorana effect does not occur in this case. In addition, inelastic collision is prohibited when $k_BT < 2hB$. We have discussed the collision between electrostatically trapped molecules in the J = 0 state when $k_B T > 2hB$. The collision cross-sections can be obtained through a classical treatment under this condition, because T is high enough that the broadening of the molecular wavepacket is negligible. The elastic collision is more dominated by the dipole-induced dipole interaction than the dipole-dipole interaction except for when μE is very large. We have assumed that the collisional transitions to the $J \ge 1$ states cause trap loss, because the trapping force becomes weaker after these collisional transitions. The ratio of the collision loss rate to the elastic collision rate (R) increases with T, when T is lower than T_c shown in Table 1. Because

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of $|\langle 0, 0 | \overrightarrow{\mu} | 0, 0 \rangle|^2 \ll |\langle 0, 0 | \overrightarrow{\mu} | 1, 0 \rangle|^2$, $|\langle 0, 0 | \overrightarrow{\mu} | 1, \pm 1 \rangle|^2$, evaporative cooling seems difficult when $k_B T > 2hB$.

When the molecules in the J = 0 state are trapped in a stage ring, their tangential velocity component can be rather high. Therefore, it is worthwhile to analyze the collision with high kinetic energy.

In this paper, we have assumed that $\mu E < hB$. When $\mu E > hB$, $|\langle 0, 0 | \overrightarrow{\mu} | 0, 0 \rangle|^2$ becomes larger than the value given by (7) and σ_0 becomes larger than the values given by (13) and (14). On the other hand, $|\langle 0, 0 | \overrightarrow{\mu} | 1, 0 \rangle|^2$ and $|\langle 0, 0 | \overrightarrow{\mu} | 1, \pm 1 \rangle|^2$ becomes smaller than the value given by (7). Note also that the energy gap between the J = 0 and 1 states becomes larger and σ_{loss} becomes smaller as compared with the values obtained by ignoring the Stark energy shift. Giving a high electric field, R becomes smaller than the value estimated by assuming $\mu E \ll hB$.

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