

Collisions between electrostatically confined linear polar molecules

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Abstract. Collisions between linear polar molecules that were electrostatically trapped were investigated. The collisional transition from a low to a high field seeking state (inelastic collision) causes trap loss. The efficiency of evaporative cooling is improved at higher elastic collision rates. We calculated cross-sections of inelastic and elastic collision using a semi-classical treatment.

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

Trapping cold molecules has been considered difficult, mainly because of its difficulty to decelerate molecules using laser cooling. However, several groups have recently developed effective methods for preparing ultracold molecules and confining them in three-dimensional traps. A Harvard group used static magnetic fields to trap paramagnetic CaH molecules pre-cooled using buffer gas collisions [1]. Takekoshi *et al.* trapped cesium dimers produced using the photo-association of laser-cooled Cs atoms in focused CO₂ laser beams [2]. Bethlem *et al.* decelerated ND₃ molecular beams using a time-varying inhomogeneous electric field and loaded them into an electrostatic trap [3].

An electrostatic trap can only confine dipoles in the low-field seeking state because Maxwell's equation permits a local field minimum but not a maximum. The stability of the trap should be considered in relation to the Majorana effect (the transition between quantum states, caused by a change of the electric field direction) and inelastic collisions, which cause the transition to high-field seeking states. Both effects decrease as the energy gap between the low and high field seeking states increases. Bethlem *et al.* took advantage of the ND₃ characteristics of a relatively small inversion doublet and hence a large Stark effect [3]. Bohn calculated the collision loss rate, when diatomic polar molecules in the ¹ Π state (Λ -doublet plays a similar role to the inversion doublet) are electrostatically trapped [4].

To expand the utility of the molecular trapping technology, we discuss the loss rate of electrostatically trapped linear polar molecules (¹ Σ state). Linear polar molecules can be trapped inside a quadrupole electrode by the

second-order Stark effect. Alkali-halide molecules, in particular, have been considered to be advantageous in achieving high trap potential with a relatively low electric field strength (<30 kV/cm) [5,6]. We analyzed the Majorana effect with electrostatically confined linear polar molecules in the ($J = 1, M = 0$) state, where J denotes the quantum number of the total molecular angular momentum, and M is the quantum number of the trajectory of the molecular angular momentum parallel to the electric field. The loss rate from the trap is estimated by $4\pi\nu_0^2/\sqrt{\delta^2 + 4\nu_0^2}$, where ν_0 is the molecular secular motion frequency, and δ is the transition frequency between the ($J = 1, M = 0$) and ($J = 1, M = \pm 1$) states [7]. Taking $\nu_0 = 5$ kHz and $\delta = 500$ MHz, the rate of loss caused by the Majorana effect is 0.5 /s.

This paper discusses the collision between linear polar molecules in a ¹ Σ state, trapped in a quadrupole electrode. The inelastic collision rate should be decreased to increase the trap duration. However, the rate of elastic collisions should be increased to improve the efficiency of the evaporative cooling. We calculated the inelastic and elastic collision cross-sections of linear polar molecules in a ($J = 1, M = 0$) state. We took only the dipole-dipole interaction into account, as Bohn did [4]. The kinetic energy of the trapped molecules was taken to be several hundred mK, as presently obtained [1,3] (Bohn calculated taking the kinetic energy 1 μ K [4]). For linear polar molecules in a ¹ Σ state, the diagonal matrix element of the dipole moment is proportional to the strength of the electric field. Therefore, the dependence of a collision cross-section on the strength of the electric field is quite different from the dependence for polar symmetric-top molecules, which can be trapped in a hexapole electrode by the first-order Stark effect.

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2 Calculation of collision cross-section

The cross-section that a molecule can achieve in a transition $(J, M) = (J_f, M_f) \leftarrow (J_i, M_i)$ through the collision is described by

$$\sigma_{(J_f, M_f, J_i, M_i)} = \frac{\pi}{k^2} \sum (2L + 1) P[L, (J_f, M_f \leftarrow J_i, M_i)] \quad (1)$$

where k is the incident wave number, L is the quantum number for the angular momentum of the molecular relative motion, and P is the opacity function [8]. When the intermolecular interaction is small enough, P is obtained using the first Born approximation [9].

When $1/k$ is much smaller than the scale size of the intermolecular potential (d), (1) is rewritten as

$$\sigma_{(J_f, M_f, J_i, M_i)} = \int_0^\infty 2\pi b P_{(J_f, M_f, J_i, M_i)}(b) db$$

$$b = \frac{1}{k} \left(L + \frac{1}{2} \right) \quad (2)$$

here b is called the impact parameter [8].

Here we calculate the collision cross-section using (2), taking the molecular relative velocity (v) 19 m/s or 8.5 m/s ($1/k$ is on the order of 1–5 Å). If the value of σ ($= \pi d^2$) obtained by the semi-classical treatment is much larger than π/k^2 , this treatment is reasonable. If $\sigma < \pi/k^2$ ($d < 1/k$) is obtained by the semi-classical treatment, the collision cross-section should be obtained by (1). In this case, the actual value of the collision cross-section is in maximum π/k^2 because of $P(L: L \geq 1) \ll P(L = 0) \leq 1$.

Assuming the classical path, P is obtained using

$$P_{(J_f, M_f, J_i, M_i)}(b) = \sum_{J_{\text{pi}}, M_{\text{pi}}} \rho(J_{\text{pi}}, M_{\text{pi}})$$

$$\times \sum_{J_{\text{pf}}, M_{\text{pf}}} |(1/\hbar) \int_{-\infty}^{\infty} \langle J_f, M_f, J'_{\text{pf}}, M'_{\text{pf}} | V(r) | J_i, M_i, J_{\text{pi}}, M_{\text{pi}} \rangle dt|^2$$

$$r = \sqrt{b^2 + (vt)^2} \quad (3)$$

where $(J_{\text{pi}}, M_{\text{pi}})$ and $(J_{\text{pf}}, M_{\text{pf}})$ denote the (J, M) state of the collision partner before and after the collision, respectively. $\rho(J_{\text{pi}}, M_{\text{pi}})$ is the distribution of the $(J_{\text{pi}}, M_{\text{pi}})$ state, taking $\sum_{J_{\text{pi}}, M_{\text{pi}}} \rho(J_{\text{pi}}, M_{\text{pi}}) = 1$. $V(r)$ is the intermolecular potential. Assuming that the intermolecular potential is determined only by the dipole-dipole interaction (this assumption is valid for the collision between molecules with dipole moment larger than 0.7 D [10]), V is described by

$$V = \frac{1}{r^3} \left(\boldsymbol{\mu}_a \cdot \boldsymbol{\mu}_p - \frac{3(\boldsymbol{\mu}_a \cdot \mathbf{r})(\boldsymbol{\mu}_p \cdot \mathbf{r})}{r^2} \right) \quad (4)$$

where $\boldsymbol{\mu}_{a,p}$ denotes the dipole moment of the probing molecule and the collision partner, respectively. We performed the numerical calculation of collision cross-sections

using the Quantum-Fourier-transform (QFT) theory [11], with which the momentum and the energy are conserved strictly

$$P_{(J_f, M_f, J_i, M_i)}(b) = \min(1, Q_{(J_f, M_f, J_i, M_i)}(b)). \quad (5)$$

Here $Q_{(J_f, M_f, J_i, M_i)}(b)$ is obtained using the first Born approximation seen below. With a small b (strong interaction), the first Born approximation is not valid and $P_{(J_f, M_f, J_i, M_i)}(b) = 1$ is used for simplicity ($P(b) \leq 1$ must always hold) [12]

$$Q_{(J_f, M_f, J_i, M_i)}(b) = \frac{32\alpha^2}{27\pi\hbar^2 v^2 b^4} \sum_{J_{\text{pi}}, M_{\text{pi}}} \rho(J_{\text{pi}}, M_{\text{pi}})$$

$$\times \sum_{J_{\text{pf}}, M_{\text{pf}}} |\langle J_f, M_f | \boldsymbol{\mu}_a | J_i, M_i \rangle|^2$$

$$\times |\langle J_{\text{pf}}, M_{\text{pf}} | \boldsymbol{\mu}_p | J_{\text{pi}}, M_{\text{pi}} \rangle|^2 f_{(J_{\text{pi}}, M_{\text{pi}})}^{(J_{\text{pf}}, M_{\text{pf}})}(\Delta)$$

$$f_{(J_{\text{pi}}, M_{\text{pi}})}^{(J_{\text{pf}}, M_{\text{pf}})}(\Delta) = \exp \left[-\frac{4\Delta^2}{\pi\alpha^2} \right]$$

$$\Delta = \frac{b}{\hbar v} [R(J_f, M_f) + R(J_{\text{pf}}, M_{\text{pf}})$$

$$- R(J_i, M_i) - R(J_{\text{pi}}, M_{\text{pi}})]$$

$$R(J, M) = hBJ(J+1) + S(J, M). \quad (6)$$

Here α is an arbitrary (dimensionless) constant, which cannot be determined just by QFT theory. Davies recommends to take $\alpha = \sqrt{3\pi/4}$ (QFT-I), so that the formula of $P(b)$ converges to the Anderson-Tsao-Curnutte (ATC) theory with $\Delta \rightarrow 0$ (with ATC theory, the energy is conserved only with $\Delta = 0$) [11–13]. E is the strength of the electric field and B is the molecular rotational constant. $S(J, M)$ is the electric potential energy. When the strength of the electric field is low ($S(J, M) \ll 2hBJ$), $S(J, M)$ is obtained by the second-order perturbation as

$$S(J, M) \simeq \frac{DE^2}{2J(J+1)} \frac{J(J+1) - 3M^2}{(2J-1)(2J+3)}$$

$$D = \frac{\mu^2}{hB}. \quad (7)$$

3 Results of calculation

Here, the collision cross-sections ($\sigma_{(J_f, M_f)}$) of the linear polar molecules are discussed with $(J_i, M_i) = (J_{\text{pi}}, M_{\text{pi}}) = (1, 0)$, assuming that the fractions of the molecules in $J > 2$ states are negligibly small. This assumption is reasonable as $(J, M) = (1, 0)$ is the lowest energy state of the low-field seeking states, and the trapping force for molecules in $J > 2$ states is much smaller than in the $(J, M) = (1, 0)$ state when the strength of the electric field is low ($S(1, 0) \ll 2hB$). Table 1 gives the formulas for $|\langle J_f, M_f | \boldsymbol{\mu} | 1, 0 \rangle|^2$. The values of μ and B are given in Table 2 for the OCS, HCN, and NaCl molecules. The non-zero matrix elements are $\langle 0, 0 | \boldsymbol{\mu} | 1, 0 \rangle$, $\langle 2, 0 | \boldsymbol{\mu} | 1, 0 \rangle$, and

Table 1. Square of the absolute value of the matrix elements of the dipole moment.

$ \langle 1, 0 \mu 1, 0 \rangle ^2$	$\mu^4 E^2 / 25 h^2 B^2$
$ \langle 1, \pm 1 \mu 1, 0 \rangle ^2$	$\mu^4 E^2 / 50 h^2 B^2$
$ \langle 0, 0 \mu 1, 0 \rangle ^2$	$\mu^2 / 3$
$ \langle 2, 0 \mu 1, 0 \rangle ^2$	$4\mu^2 / 15$
$ \langle 2, \pm 1 \mu 1, 0 \rangle ^2$	$\mu^2 / 5$

Table 2. Cross-section of collisional transition from the $(J, M) = (1, 0)$ state to the $(0, 0)$ state for OCS, HCN, and NaCl molecules. The relative velocity is taken to be 19 m/s and 8.5 m/s key.

	$\mu(\text{D})$	$B(\text{GHz})$	$\sigma_{(0,0)}^{v=19 \text{ m/s}}(\text{Å}^2)$	$\sigma_{(0,0)}^{v=8.5 \text{ m/s}}(\text{Å}^2)$
OCS	0.71	6.09	43.4	19.0
HCN	2.94	44.4	6.02	1.34
NaCl	8.50	6.53	287	62.8

$\langle 2, \pm 1 | \mu | 1, 0 \rangle$. Also, $\langle 1, 0 | \mu | 1, 0 \rangle$ and $\langle 1, \pm 1 | \mu | 1, 0 \rangle$ become non-zero when the electric field is given. The formulas for $\langle 1, 0 | \mu | 1, 0 \rangle$ and $\langle 1, \pm 1 | \mu | 1, 0 \rangle$ are obtained using the first-order perturbation theory. The collision loss rate is determined by $\sigma_{(0,0)}$ and $\sigma_{(1,\pm 1)}$, and the elastic collision rate is determined by $\sigma_{(1,0)}$.

Here, we discuss the correlation between the collision cross-sections and the electrostatic potential of trapped molecules (U). When the strength of the electric field is low enough ($U \ll 2hB$), U is obtained by

$$U = S(1, 0) \simeq \frac{DE^2}{10}. \quad (8)$$

$\sigma_{(0,0)}$ is almost constant while $U \ll 2hB$, because $f_{(1,0)}^{(J_{\text{pf}}, M_{\text{pf}})}(\Delta)$ are determined mostly by Bb/v for $(J_{\text{pf}}, M_{\text{pf}}) = (1, 0), (1, \pm 1), (0, 0), (2, 0)$, and $(2, \pm 1)$. Table 2 shows the values of $\sigma_{(0,0)}$ in a zero-electric field for OCS, HCN and NaCl molecules, taking $v = 19$ m/s or 8.5 m/s.

Figure 1 shows $\sigma_{(1,\pm 1)}$ as a function of U for OCS, HCN, and NaCl molecules. In this case, (6) is approximately rewritten as (with $U \ll B$)

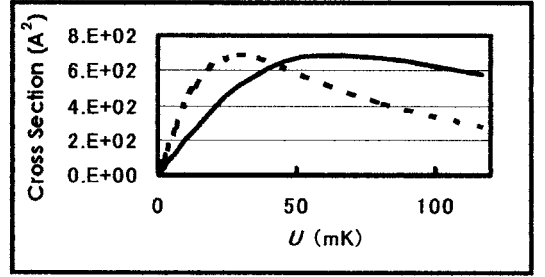
$$Q_{(1,0,1,\pm 1)}(b) \simeq \frac{64}{9h^2v^2b^4} U^2 D^2 \left[\exp \left[-\frac{12U^2b^2}{\pi^2h^2v^2} \right] + \exp \left[-\frac{48U^2b^2}{\pi^2h^2v^2} \right] \right]$$

with $\frac{Ub}{hv} \ll 1$

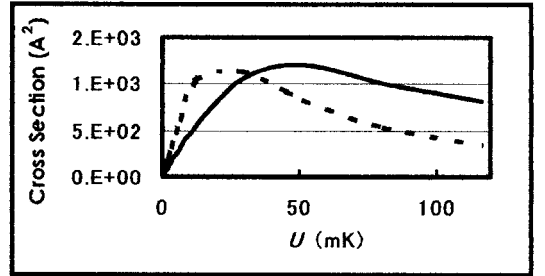
$$\simeq \frac{64}{9h^2v^2b^4} U^2 D^2 \exp \left[-\frac{12U^2b^2}{\pi^2h^2v^2} \right]. \quad (9)$$

In fact $\sigma_{(1,\pm 1)}$ reaches a maximum at a specific value of U (U_p). As D/v increases, U_p decreases. $\sigma_{(1,\pm 1)}$ is much

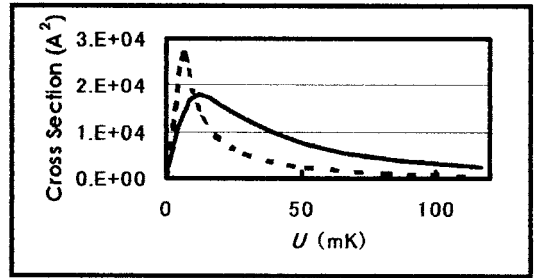
OCS



HCN



NaCl

**Fig. 1.** Cross-sections of collisional $(J, M) = (1, 0) \rightarrow (1, \pm 1)$ transition of OCS, HCN, and NaCl molecules as a function of the electrostatic potential of trapped molecules (U). The solid and dotted line show cases with a mean relative velocity of 19 m/s and 8.5 m/s, respectively.

larger than $\sigma_{(0,0)}$ at $U > 3$ mK, so the collision loss rate is actually determined mostly by $\sigma_{(1,\pm 1)}$.

Figure 2 shows $\sigma_{(1,0)}$ as a function of U for the OCS, HCN, and NaCl molecules. The figure shows that $\sigma_{(1,0)}$ is almost proportional to UD/v . This result can be explained as follows. With $2Bb/hv \gg Ub/hv \gg 1$, (6) is approximately rewritten as (see Tab. 1)

$$Q_{(1,0,1,0)}(b) = \frac{128}{9h^2v^2b^4} U^2 D^2. \quad (10)$$

As b_0 is proportional to $\sqrt{UD/v}$ (b_0 is the value which $Q(b_0) = 1$), $\sigma_{(1,0)} = 2\pi b_0^2$ should be proportional to UD/v while $Ub_0/hv \gg 1$ [12]. As the elastic collision rate is determined by $nv\sigma_{(1,0)}$ (n is the molecular density), the elastic collision rate should be constant with any value of v . But when $2B \lesssim U$ (high electric field) or $Ub_0/hv \lesssim 1$ (low electric field), the proportionality between $\sigma_{(1,0)}$ and UD/v is not valid.

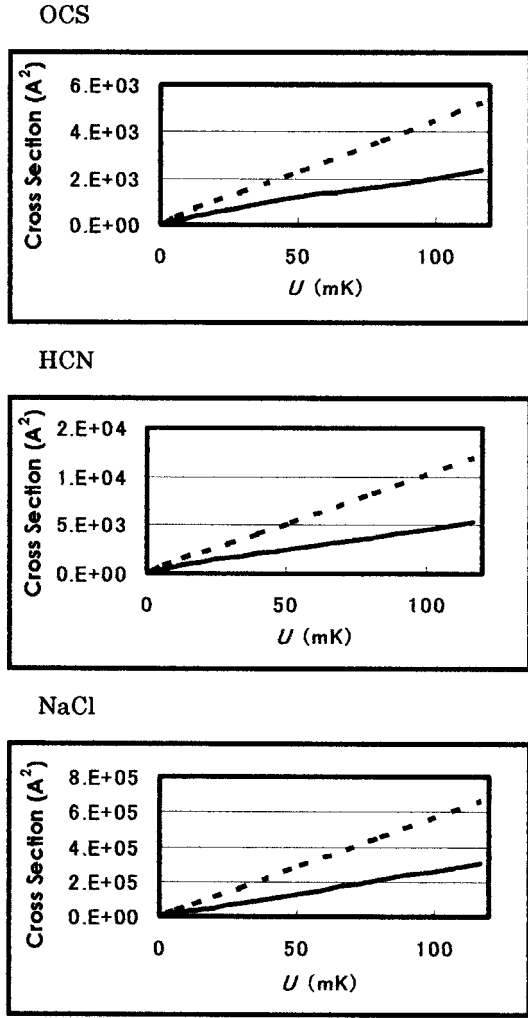


Fig. 2. Elastic collision cross-section of OCS, HCN, and NaCl molecules in the $(J, M) = (1, 0)$ state as a function of the electrostatic potential energy of the trapped molecules (U). The solid and dotted lines show the cases with a mean relative velocity of 19 m/s and 8.5 m/s, respectively.

If we can achieve a small $K = \sigma_{(1,\pm 1)}/\sigma_{(1,0)}$, we can get highly efficient evaporative cooling effect with a low collision loss rate. Figure 3 shows K for the OCS, HCN, and NaCl molecules as a function of U , taking $v = 8.5$ m/s. As U increases, K decreases rapidly. With $U < 10$ mK, the values of K are almost in the same order for the three different molecules. But at $U > 10$ mK, K (NaCl) is much smaller than K (OCS) and K (HCN).

At $U \rightarrow 0$, the calculated values of $\sigma_{(1,\pm 1)}$ and $\sigma_{(1,0)}$ converge to zero, because only the dipole-dipole interaction is taken into account. In fact, they converge to certain values, as determined by the higher order interaction ($\propto r^{-6}$).

4 Discussion

The focus of our research is the collisions between molecules that are electrostatically trapped, which differ

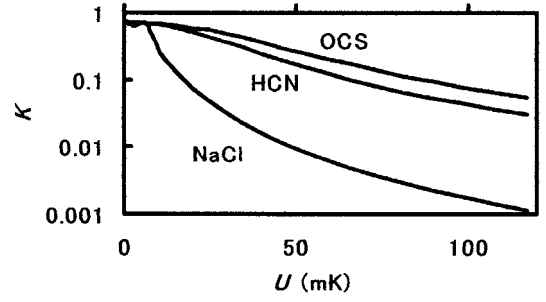


Fig. 3. $K = \sigma_{(1,\pm 1)}/\sigma_{(1,0)}$ of OCS, HCN, and NaCl molecules in the $(J, M) = (1, 0)$ state as a function of the electrostatic potential energy of trapped molecules. Here the mean relative velocity is 8.5 m/s.

from the collisions in cells at room temperature on the following points.

1. Without an electric field, $\langle J, M | \mu | J, M \rangle$ and $\langle J, M \pm 1 | \mu | J, M \rangle$ are zero for linear polar molecules. But these matrix elements are non-zero, because trapped molecules are under electric field. Both elastic and inelastic collisions are mainly caused by the dipole-dipole interaction.
2. Only low-field seeking molecules can be electrostatically trapped. The collision partner molecules may therefore be in limited quantum states.
3. The kinetic energy of molecules trapped in a conventional hyperboloid electrode is in the order of several hundred mK [7]. In a ring electrode, the kinetic energy of trapped molecules may be almost the same order at room temperature [14].

For linear polar molecules at $(J, M) = (1, 0)$ state that are electrostatically trapped, $\sigma_{(0,0)}$ is much smaller than $\sigma_{(1,0)}$ and $\sigma_{(1,\pm 1)}$ at $U > 3$ mK, although $|\langle 0, 0 | \mu | 1, 0 \rangle|$ is much larger than $|\langle 1, 0 | \mu | 1, 0 \rangle|$ and $|\langle 1, \pm 1 | \mu | 1, 0 \rangle|$. This is because the collision partner molecules are mostly in the $(J_{\text{pi}}, M_{\text{pi}}) = (1, 0)$ state. If the collision partner molecules were at $(J_{\text{pi}}, M_{\text{pi}}) = (0, 0)$ state, $\sigma_{(0,0)}$ becomes much larger than $\sigma_{(1,0)}$ and $\sigma_{(1,\pm 1)}$.

We have calculated the collisional cross-section using the semi-classical treatment. This treatment is valid for $\sigma_{(1,0)}$ and $\sigma_{(1,\pm 1)}$ at $U > 10$ mK, where they become larger than 200 Å^2 ($\gg \pi/k^2$). The semi-classical treatment gives smaller values of $\sigma_{(0,0)}$ (HCN) than π/k^2 ($\approx 50 \text{ Å}^2$ with $v = 8.5$ m/s, 10 Å^2 with $v = 19$ m/s). So the semi-classical treatment does not give accurate values of $\sigma_{(0,0)}$ (HCN). The actual values of $\sigma_{(0,0)}$ (HCN) can be in maximum π/k^2 , which is still much smaller than $\sigma_{(1,\pm 1)}$. So the collision loss rate can be estimated well with the semi-classical treatment.

Taking a molecular density of $1 \times 10^{10} \text{ cm}^3$ and a mean relative velocity of 8.5 m/s, the collision loss rate of the OCS, HCN, and NaCl molecules reach a maximum of 1 /s ($U \approx 30$ mK), 2 /s ($U \approx 30$ mK), and 46 /s (at $U \approx 10$ mK), respectively. We can reduce the collision loss rate by giving a non-zero minimum value of U (U_{min}) at the trap center ($U_{\text{min}} > 30$ mK for the OCS and HCN molecules and $U_{\text{min}} > 10$ mK for the NaCl molecule).

Assuming that the molecules are trapped in an ideal hyperboloid electrode (DC-voltage is given to the ring electrode), a non-zero value of U_{\min} can be given using the following method. Given an AC-voltage between the two end-caps, the strength of the electric field becomes zero at $(0, 0, z_0 \sin(\omega t))$, where z_0 is the constant determined by the AC-voltage amplitude. The electric field is given by $(E_x, E_y, E_z) = C(x, y, -2z + 2z_0 \sin(\omega t))$, where C is the constant determined by the DC-voltage. The mean strength of the electric field becomes $C\sqrt{x^2 + y^2 + 4z^2 + 2z_0^2}$, whose minimum value is $\sqrt{2}Cz_0$. Given the appropriate values of $U_{\min} = (DC^2/5)z_0^2$, the efficiency of the evaporative cooling is improved keeping the collision loss rate low (K is low at $U > U_{\min}$). The trap loss caused by the Majorana effect is also reduced, given the non-zero values of U_{\min} [7].

When molecules (not linear) are confined by the first order Stark effect, trapped molecules are mostly in a $|KM| \approx J^2$ state (K is the angular momentum trajectory parallel to the molecular axis). The cross-section of the elastic collision is larger than that in a cell, where the molecules are uniformly distributed to all values of M between $-J$ and J . The elastic collision cross-section has a very small dependence on the strength of the electric field, because the diagonal matrix element of the dipole moment is almost constant within the feasible strength of the electric field. The inelastic collision cross-section decreases as the strength of the electric field increases because of the larger energy gap between different M states.

5 Conclusion

We calculated the inelastic and elastic collision cross-sections of linear polar molecules in the $(J, M) = (1, 0)$ state. For the electrostatically trapped molecules, the collision cross-section of the transition to the $(J, M) = (1, \pm 1)$ state is much larger than the cross-section of the transition to the $(J, M) = (0, 0)$. The collision cross-section of the transition to the $(J, M) = (1, \pm 1)$ state reaches a maximum at a specific strength of the electric

field. The elastic collision cross-section is proportional to the square of the electric field (proportional to the electrostatic potential energy). To perform evaporative cooling with a low collision loss rate, increasing the electric field strength is preferable.

We assumed that two colliding molecules experience the same electric field strength. This assumption is reasonable, as intermolecular interaction occurs only when the intermolecular distance is much less than $1 \mu\text{m}$ and the gradient of the electric field strength is actually less than 10 kV/mm^2 .

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