

Low-energy scattering of molecules and ions in a magnetic field

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(Received 19 January 2014; revised manuscript received 14 May 2014; published 3 June 2014)

We describe calculations of elastic and inelastic scattering of neutral molecules and cold ions in a magnetic field. The molecule is assumed to have a magnetic and electric dipole moment. The external magnetic field splits the ground rovibrational energy levels of the molecule. The highest energy state within the ground rovibrational manifold increases in energy as the distance to the ion decreases leading to a repelling potential. At low energy, inelastic collisions are strongly suppressed due to the large distance of closest approach. Thus, a collision between a neutral molecule and a cold ion will lead to a decrease in the molecule's kinetic energy with no change in internal energy. We present results for the specific case of OH molecules cooled by Be^+ , Mg^+ , or Ca^+ ions. Also, we perform molecular dynamics simulations of ions and molecules in a combined Paul trap and time-averaged orbiting potential trap. Our results suggest that sympathetic cooling of neutral molecules by ions would be possible if cold ions and molecules could be simultaneously trapped.

DOI: [10.1103/PhysRevA.89.062701](https://doi.org/10.1103/PhysRevA.89.062701)

PACS number(s): 34.50.Cx, 37.10.Mn, 37.10.Pq

I. INTRODUCTION

For over ten years, there has been a substantial experimental effort to cool molecules to μK temperatures. This interest is sparked by the possibility of collective effects in a cold, dense molecular gas [1,2], interesting collisional mechanisms at low energy [3,4], or to enhance the spectroscopic accuracy needed for precision measurements [5,6]. A variety of techniques have been explored. The authors of Ref. [7] used buffer gas cooling to trap CaH at a temperature of ~ 400 mK; by a specific choice of scattering conditions, a single Ar-NO collision produced NO molecules at ~ 400 mK [8]. The Stark effect has been used to slow and trap a variety of molecules with electric dipole moments [9,10]. The authors of Ref. [11] took advantage of the favorable Franck-Condon factors in SrF to perform one-dimensional laser cooling. Evaporative cooling was able to decrease the temperature of trapped OH from an initial 45 to 5.1 mK [12]. The authors of Refs. [13,14] proposed variations of a Sisyphus cooling where each photon removes a large fraction of translational energy of the molecule; the method in Ref. [13] was realized in Ref. [15] to cool CH_3F from 390 to 29 mK. The authors of Ref. [16] were able to remove 95% of the translational energy of an O_2 beam using a “molecular coilgun.” Mechanical effects, as in a spinning nozzle [17], can produce colder molecules by having the molecules exit in a moving frame of reference. This is not a complete list, but the experimental limit for cooling preexisting molecules is still above 1 mK ten years after Ref. [18]. There are also wide varieties of theoretical proposals for cooling molecules into the ultracold regime; some examples are in Refs. [19–25]. This activity inspired us to study scattering between cold molecules and various targets.

The purpose of this paper is to present the results of calculations that show a class of neutral molecules will predominantly scatter from cold ions elastically when both are at temperatures less than a few tens of mK. The specific situation we examine is the case where the molecule has both a magnetic and an electric dipole moment with the molecule in a specific internal state. The specific internal state can be achieved through the trapping process, the natural cooling near the nozzle of a molecular beam, or active cooling as in

Ref. [26]. We assume the collision takes place in a uniform B field; for molecules held in a magnetic trap, the B field varies in space but can be considered to be uniform over distance scales that characterize the scattering with the ion. The E field from the ion at the molecule leads to a repulsive $1/r^4$ interaction while the splitting of the energy levels due to the B field leads to a “high” frequency scale which allows the collision to be adiabatic. The resulting collision rate between the molecule and the ion is a few *orders of magnitude* larger than molecule-molecule collision rates and is almost perfectly elastic at low energy.

The essential difference between the current mechanism and sympathetic cooling from atoms (e.g., Ref. [27]) is that the collisions take place at longer range, which vastly increases the elastic collision rate while decreasing the rate for changing the internal state of the molecule. If ions and cold molecules could be simultaneously trapped, the result would be most similar to the sympathetic cooling of molecular ions by laser-cooled atomic ions (e.g., Ref. [28]) except the long-range repulsive $1/r$ interaction is replaced with a repulsive $1/r^4$ interaction; it is well known that sympathetic cooling leads to translationally cold molecular ions while leaving the internal state unchanged. All of our calculations are performed for OH molecules, but we expect the basic cooling mechanism to work for other molecules as well.

Figure 1 shows how the four highest internal energies of OH vary with E -field strength when a 300 G B field is also applied. The two different line types are for different angles between the fields. If the OH starts in the uppermost state, an increasing electric field causes the internal energy to increase. In a collision, the E field from the ion at the OH increases as the distance decreases which leads to an increasing internal energy. This increasing internal energy is equivalent to a potential energy that increases with decreasing separation, i.e., a repelling force between the ion and the neutral molecule which keeps them separated. If the angle between the fields is not too close to 90° the collision will be adiabatic and the OH will remain in the uppermost state. If there is a close approach *and* the angle is near 90° the near degeneracy between the upper two levels could allow a transition between the states. Fortunately, if the OH and the ion have small relative kinetic

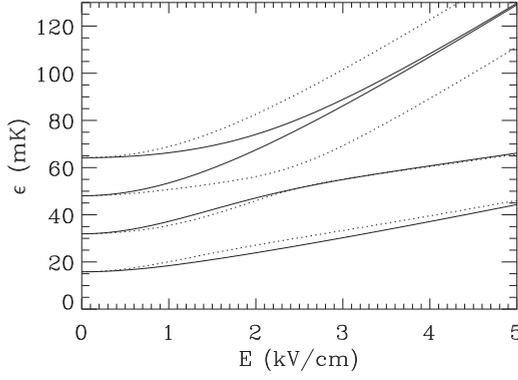


FIG. 1. The four largest energies of OH in a B field of 300 G as a function of applied E field. The solid lines are for an angle of 90° between the fields; the dotted lines are for 45° .

energy they will not be able to approach close enough for a transition between the states to occur. For a 300-G B field, if the OH kinetic energy is less than 20 mK, then the inelastic cross section is 10^5 times smaller than the elastic cross section. The ratio of collision rates for different B fields are shown in Fig. 3.

II. NUMERICAL METHOD

Our calculations were performed using a mixture of classical and quantum mechanics. The relative motion of the ion and molecule is treated classically. This should be a good approximation over the energy range presented here. We discuss the limitations of our calculation below and argue that the classical rates should be a good approximation down to μK temperatures. For the internal states of the molecule, we solved the full time-dependent Schrödinger equation using the leapfrog algorithm. By solving the Schrödinger equation for this eight-state system, we ensure that no approximation causes the inelastic processes to be erroneously suppressed.

For the B -field strengths considered in this paper, the OH molecule has eight internal states of interest: four spin states times two parity states. The Hamiltonian we use is the explicit form found in Ref. [29] Appendix A.2, but modified to include the possibility for the electric field from the ion to *not* lie in the xz plane. This modification is accomplished by replacing $E \cos \theta_{EB}$ with E_z , $E \sin \theta_{EB}$ with $E_x + iE_y$ in H_{16} , H_{27} , and H_{38} , and $E \sin \theta_{EB}$ with $E_x - iE_y$ in H_{25} , H_{36} , and H_{47} ; the matrix elements below the diagonal are obtained from those above the diagonal by complex conjugation.

In what follows $\vec{\psi}(t)$ is the eight-element vector holding the amplitudes of each state, $\vec{r}(t)$ is the relative position vector from the ion to the molecule, and $\vec{v}(t)$ is the relative velocity vector. We solve for both the classical and quantum dynamics using a leapfrog algorithm. For the quantum state, knowing $\vec{\psi}(t)$ and $\vec{\psi}(t - \delta t)$ and knowing the $\vec{r}(t)$ the wave function at time $t + \delta t$ is found from

$$\vec{\psi}(t + \delta t) = \vec{\psi}(t - \delta t) - 2i\delta t \underline{H}(\vec{r}(t))\vec{\psi}(t)/\hbar, \quad (1)$$

where $\underline{H}(\vec{r}(t))$ is the 8×8 matrix evaluated for the E field arising from the separation $\vec{r}(t)$. At this point in the algorithm, the classical force can be calculated using the

Hellman-Feynman theorem

$$\vec{F}(t) = -\langle \vec{\psi}(t) | \vec{\nabla} \underline{H}(\vec{r}(t)) | \vec{\psi}(t) \rangle, \quad (2)$$

which will allow us to update the position and velocity vectors. Knowing $\vec{v}(t - \delta t/2)$ and $\vec{r}(t)$, the position and velocity at the next time step is computed from

$$\begin{aligned} \vec{v}(t + \delta t/2) &= \vec{v}(t - \delta t/2) + \vec{a}(t)\delta t, \\ \vec{r}(t + \delta t) &= \vec{r}(t) + \vec{v}(t + \delta t/2)\delta t, \end{aligned} \quad (3)$$

where the acceleration $\vec{a}(t) = \vec{F}(t)/\mu$ and μ is the reduced mass. These steps are iterated, giving an algorithm that gives the motion of the particles and time evolution of the internal states.

Since we assume the ion is much colder than the molecule, the initial relative speed is the speed of the molecule. To compute the cross section, we need to have a random set of initial positions and impact parameters. We do *not* assume the initial velocity is from a specific direction because there is an external B field which picks a specific form for the Hamiltonian. The direction of the velocity vector is chosen from a uniform sampling on the surface of a sphere. The initial position is chosen to be $\vec{r}(0) = \vec{b} - \hat{v}R$ where R is a large initial distance and \vec{b} is a random point within a circle delimited by a b_{\max} such that $\vec{b} \cdot \hat{v} = 0$. The initial conditions are such that we start the quantum state in the highest energy eigenstate of $\underline{H}(\vec{r}(0))$. The final time is chosen to be $2R/v$. For all parameters, we test convergence with respect to R (increasing R until the results do not change) and the b_{\max} (increasing the maximum until the results do not change).

The physical parameters of interest are the inelastic and energy transfer collision rates. The inelastic collision rate is found from

$$\beta = v\pi b_{\max}^2 (1 - P_8), \quad (4)$$

where $P_8 = |\langle \vec{\psi}_8 | \vec{\psi}(t_f) \rangle|^2$ is the probability the molecule is still in the highest-energy *eigenstate* at the final time t_f . The energy transfer collision rate is found from

$$\eta = v\pi b_{\max}^2 (E_{K_i} - E_{K_f}) / E_{K_i}, \quad (5)$$

where E_{K_i} is the initial kinetic energy of the molecule and E_{K_f} is the final kinetic energy of the molecule. To find the final kinetic energy of the molecule, we use center-of-mass coordinates $\vec{v} = \vec{v}_m - \vec{v}_i$ and $\vec{V} = (M_m \vec{v}_m + M_i \vec{v}_i) / (M_m + M_i)$ where the m, i subscripts refer to the molecule or ion, respectively. The final velocity of the molecule is found from $\vec{v}_{m,f} = \vec{V} + M_i \vec{v}(t_f) / (M_m + M_i)$, where $\vec{v}(t_f)$ is the final relative velocity from the calculation and the center-of-mass velocity \vec{V} is a conserved quantity. From these rates, we can solve for the rate of kinetic energy lost by the molecule and the rate of population lost if we know the density of ions. Taken as average quantities $dP/dt = -n\beta P$ and $dE_K/dt = -n\eta E_K$ where n is the ion density, P is the population of trapped molecules, and E_K is the average kinetic energy.

III. RESULTS

Figure 2 shows the rates as a function of OH kinetic energy for the case where the ion is Mg^+ . For these parameters, the energy loss rate is higher than the inelastic collision

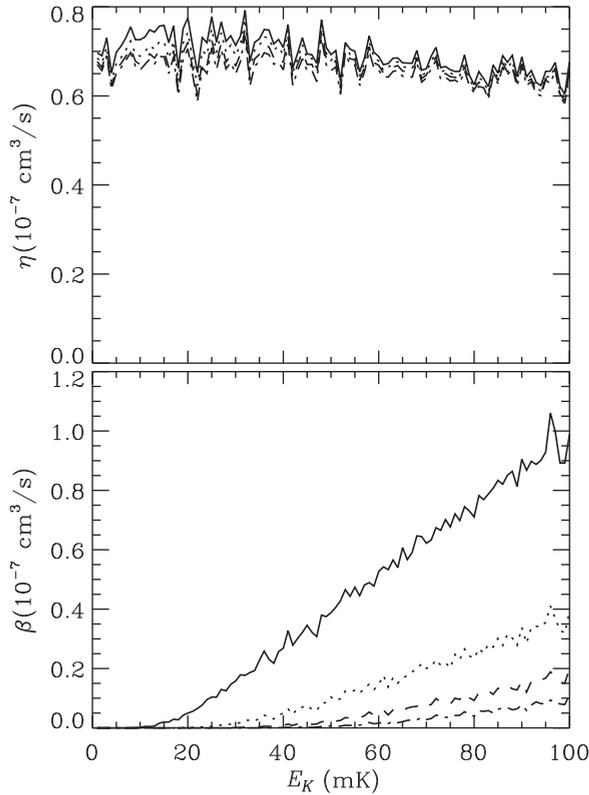


FIG. 2. The energy loss rate η and population loss rate β for neutral OH molecules colliding with cold Mg^+ ions. The ions are assumed to be much colder than the molecules and E_K is the kinetic energy of the molecules. The OH are assumed to start in their highest internal state in a B field of 100 (solid line), 200 (dotted line), 300 (dashed line), and 400 G (dash-dot line).

rate over the whole energy range except for the 100 G case. Another obvious feature is that the inelastic collisions are more suppressed as the B field is increased, and the inelastic rate has a threshold which is at larger E_K as the B field increases. Both effects are because the collisions become more adiabatic as the splitting from the B field increases. Most importantly, the energy loss rate η is approximately constant, which results from the fact that the repelling potential is approximately $1/r^4$.

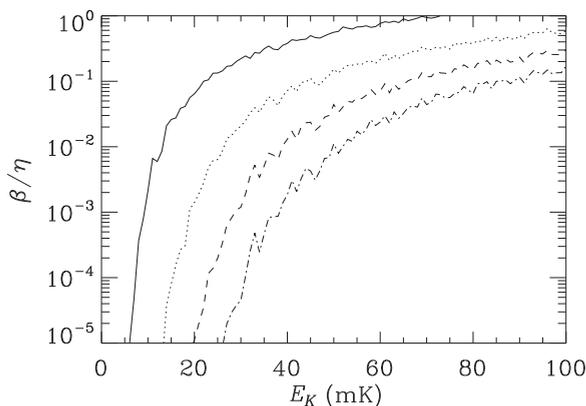


FIG. 3. The ratio of parameters for Fig. 2; the line types are the same as Fig. 2.

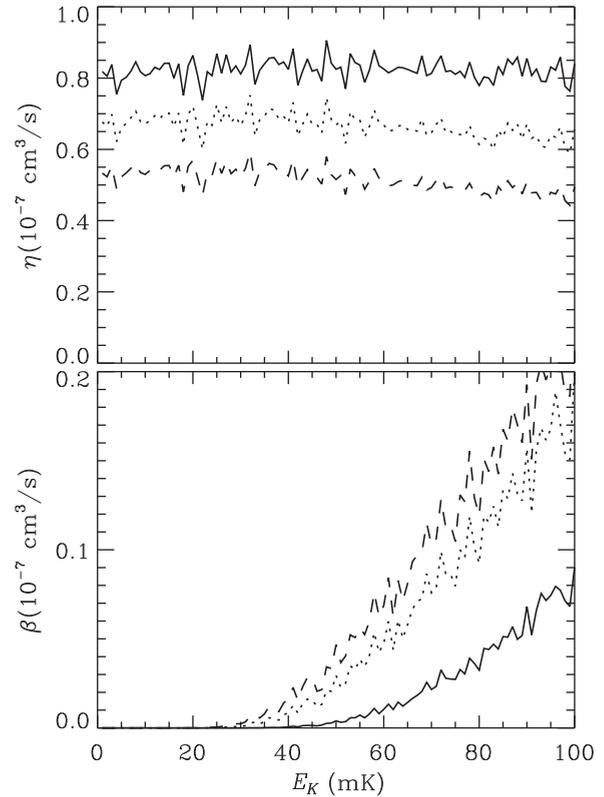


FIG. 4. Similar to Fig. 2, except all lines are for 300 G. The difference is the solid line is for OH collisions with ${}^9\text{Be}^+$, ${}^{24}\text{Mg}^+$, and ${}^{40}\text{Ca}^+$. This shows lighter ions do a better job cooling the OH molecules although the difference is not qualitative.

For a classical Hamiltonian with a pure $1/r^4$ potential, there is an exact scaling of any length ($L \propto 1/E_K^{1/4}$) and speed ($v \propto E_K^{1/2}$). Thus, classical rates vL^2 do not depend on E_K . The smallest E_K calculated is 1 mK; see below for a discussion of the rates at smaller E_K .

Figure 3 shows the ratio of the population loss rate divided by the energy loss rate for different B fields. From this plot, it is clear that the inelastic collisions become completely unimportant as energy is removed from the molecules. As an interesting point of comparison, the ratio is 0.1 for OH kinetic energies of 22, 46, 66, and 90 mK for the 100, 200, 300, and 400 G B fields. The ratio is 0.01 for OH kinetic energies of 13, 26, 39, and 51 mK. The kinetic energy at which the ratio reaches a specific low value approximately scales with the B field.

Figure 4 shows the rates as a function of OH kinetic energy for the case of 300 G for Be^+ , Mg^+ , and Ca^+ . There is the interesting trend that the inelastic collision rate is smaller and the energy loss rate is larger as the ion mass decreases. This trend is not surprising. For lighter ions, the energy transfer rate increases because it is easier to accelerate ion. For lighter ions, the distance of closest approach is larger (for the same reason) which suppresses the inelastic rate.

IV. POSSIBILITY FOR SYMPATHETIC COOLING

In this section, we speculate about whether this scattering could be used for sympathetic cooling of neutral molecules

using cooled ions. We address how many ions are needed and present a simple electric and magnetic field geometry that could simultaneously trap ions and molecules.

An important question is how many ions will be needed to produce substantial cooling. The *fundamental* limiting factor is the inelastic collision between two molecules. The authors of Ref. [30] (Fig. 4) had an inelastic rate at 0 V/cm of $\sim 4 \times 10^{-10}$ cm³/s for 500 G and 50 mK, $\sim 1 \times 10^{-11}$ cm³/s for 500 G and 1 mK, and $\sim 1 \times 10^{-11}$ cm³/s for 500 G and 1 μ K. These rates are much smaller than those in Figs. 2 and 4 because the interaction distance is smaller for the OH-OH collisions. If the molecules and ions are equally distributed with an energy ~ 50 mK, then there can be somewhat less than 1 ion per 200 molecules. Note that once the molecule energy is ~ 1 mK or less, then there can be approximately 1 ion per 10 000 molecules. Since the OH-OH inelastic scattering rate decreases rapidly with energy, we estimate that approximately 1 ion per 1000 molecules would be sufficient for temperatures less than 25 mK. The authors of Ref. [12] estimated a peak density of OH of $\sim 5 \times 10^{10}$ cm⁻³ with $\sim 10^6$ molecules. For these parameters, sympathetic cooling should be effective with less than 1000 ions and, perhaps, with as few as 100. While these are few ions, we will address below whether trapping even this number of ions with neutral molecules is feasible.

One important question is the energy dependence of the energy loss rate η for energies below 1 mK. To get an idea, we performed a quantum calculation for the energy loss rate for a repelling $1/r^4$ potential. The actual potential depends on both r and $\cos \theta$, but we can obtain the trend with respect to energy just from the isotropic interaction. We fit the dependence of the highest-energy eigenstate as a function of the E field and the angle with respect to the B field. For low field strengths, the energy can be written as

$$\epsilon_8 = \epsilon_8(E = 0) + \frac{1}{2}[\alpha_0 + \alpha_2 P_2(\cos \theta)]E^2, \quad (6)$$

where P_2 is a Legendre polynomial, $\alpha_0 = 7.5 \times 10^{-10}$ mK/(V/m)², and $\alpha_2 = 7.0 \times 10^{-10}$ mK/(V/m)². For the quantum scattering calculation, we set $\alpha_2 = 0$ and used a potential C_4/r^4 with $C_4 = (\alpha_0/2)(e/4\pi\epsilon_0)^2$. The scattering could be treated using a method analogous to that found in Ref. [31], but, instead, we numerically solved the radial Schrödinger equation using a Numerov algorithm.

The quantum energy loss rate η changes by less than 1% between 10 mK and 10 nK, which is the behavior expected from *classical* scaling. The energy loss rate is

$$\eta \propto v \int_{-1}^1 (1 - \cos \theta) \frac{d\sigma}{d \cos \theta} d \cos \theta, \quad (7)$$

with the proportionality constant independent of energy. As the energy decreases, the cross section decreases but the differential cross section is less strongly peaked at $\cos \theta = 1$. The near constant quantum rate for the isotropic potential suggests the energy loss rate $\eta \sim 10^{-7}$ cm³/s for the full potential down to ~ 10 nK.

We expect that it is not trivial to simultaneously trap neutral molecules and atomic ions. Therefore, we performed simulations of one possible situation to show that the molecular temperature does approach that of the positive ions. In the simulations, we try to include all of the important processes

as accurately as possible while using simplified forces when the exact form seems less important. The calculation described below is a molecular dynamics simulation with N_m molecules (from 10 to 3000) and with N_i ions from (1 to 10). We use Ca⁺ as the example ion and OH as the example molecule.

The ion-ion force was done exactly by summing up the pair-wise Coulomb interactions. We felt this was necessary because we wanted to ensure that the trap was stable with many ions and we also wanted the energy transfer between ions to be accounted for correctly. The ion-molecule force was done approximately by using the expression in Eq. (6) as the potential energy between an ion and a molecule. We tested this by performing scattering calculations as in Figs. 2 and 4 and found good agreement with the η obtained by the more involved method of Sec. III. We did *not* include the molecule-molecule interaction as a force in the calculation. Instead we used a statistical treatment of elastic scattering so that on average each molecule was scattered with a rate of ~ 100 Hz (approximately the value found in Ref. [12]). For the laser cooling, we simulated an optical molasses using a Monte Carlo technique similar to the method we used in Ref. [32]: the ion's velocity gave a Doppler shift that affected the absorption of a photon. We simulated the case of three lasers with orthogonal orientations and each photon emission was in a random direction. We chose a laser intensity so that the photon scattering rate was 1/100 of the spontaneous emission rate when exactly on resonance.

The most difficult aspect will be to trap a large number of ions so that they stay cold. We simulated ions in a Paul trap with the fields from a time-averaged orbiting potential (TOP) trap also present. For the Paul trap, we used the equations of motion in the form found in Ref. [33]. For the trap parameters, we used $\Omega_{RF} = 2\pi \cdot 4$ MHz, $q_x = 0.1516$ and $a_x = -q_x/1000$. These parameters correspond to electric fields of $E' = 1$ V/cm² and $E'_{RF} = 2$ kV/cm². For these parameters, we found that the ions were all on axis for $N_i \leq 10$. For the TOP trap parameters, we used the formulation found in Ref. [34]. In an attempt to *destabilize* the ions, we used strong magnetic fields: $B_b = 0.03$ T (i.e., 300 G), $B_q = 50$ T/m (i.e., 0.5 T/cm), and $\omega_b = 2\pi \cdot 9$ kHz. We found that the TOP field did *not* have a substantial effect on the results of our simulation independent of the orientation of the TOP trap with respect to the Paul trap. When the ions were trapped on the Paul trap axis, the temperature computed from $T = \langle E_K \rangle / (3k_B)$ was approximately 1 mK, which is a factor of 2 larger than the temperature reached by laser cooling Ca⁺ without any external fields.

The least realistic aspect of our simulation was the trap potential for the molecules. To simulate reasonable densities with relatively few particles, we were forced to use a tighter trap for the molecules than was physically reasonable. We also used a simple harmonic force with noncommensurate frequencies to reduce the CPU time needed for the molecules' acceleration. Defining $\omega_{y,z} = \alpha_{y,z} \omega_x$, we chose $\alpha_y = \sqrt{0.7}$ and $\alpha_z = \sqrt{1/8}$ so the molecule density would more strongly overlap the ions. The molecules were initialized with random positions and velocities chosen from a Maxwell-Boltzmann distribution at 25 mK. We chose ω_x so that the number of ions divided by the average volume traversed by a thermal molecule was $\sim \rho = 10^7$ cm⁻³, which will lead to a thermalization

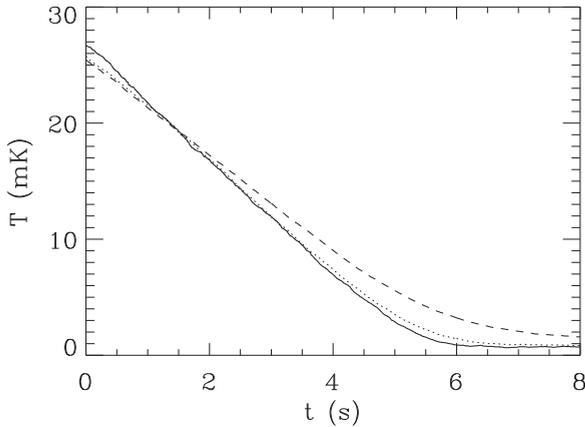


FIG. 5. The temperature of the molecules as a function of time for 10 Ca^+ ions for different number of molecules. The solid line is for 300 molecules, the dotted line is for 1000, molecules and the dashed line is for 3000 molecules. The asymptotic temperature of the 3000 molecule case would be lower if the laser cooling of the ions used a larger photon absorption rate.

time scale of a few seconds. The exact relation we used was $\omega_x = \sqrt{2\pi k_B T_m / M_m (\rho / [N_i \alpha_y \alpha_z])^{1/3}}$.

We performed calculations for $N_i = 1, 3,$ and 10 with results similar to those shown in Fig. 5. Figure 5 shows the temperature as a function of time for different number of molecules interacting with ten trapped ions. There are several important features worth considering. The first is that molecules scattering from the ions did not lead to a destabilization of the Paul trap even when the molecules outnumbered the ions by more than a factor of 100. When the number of molecules per ion was 100 or less, we found that the temperature reached by the molecules was approximately $700 \mu\text{K}$, which is somewhat larger than the field free temperature of Ca^+ ($500 \mu\text{K}$), but is smaller than the temperature of the ions computed from their average kinetic energy in the Paul trap ($T = \langle E_K \rangle / (3k_B) \simeq 1000 \mu\text{K}$). For the largest number of molecules, the final temperature was somewhat higher. This is because the collisions with molecules push the ions slightly off axis which increases their kinetic energy from the oscillating fields. We found the temperatures decreased when we increased the photon scattering rate from 1% of saturation used in Fig. 5. By increasing the laser intensity, we could recover the $700\text{-}\mu\text{K}$ temperature obtained for the calculation with fewer molecules. We do not expect any surprises due to larger number of molecules. For example, we simulated 3000 molecules with 3 ions and found that the final temperature was the expected value.

We also simulated the situation where the ions were not all on axis. For given Paul trap fields, there is a maximum

number of ions that will be trapped on axis. Increasing the number of ions leads to trapping off axis. This situation can give substantially higher ion temperature. While the molecule temperature was lower than this value for the cases we checked, the final molecule temperature was higher than the asymptotic value in Fig. 5.

These simulations were performed in a way where the ions, with no molecules present, were laser cooled to ~ 1 mK. The asymptotic temperature of the molecules tracked the temperature of the ions reached by laser cooling. Thus, a laser cooling scheme that led to colder ions would yield colder molecules.

V. CONCLUSION

We perform calculations of collisions between an OH molecule and a cold positive ion in a magnetic field. We find that the collision is adiabatic at low temperature, which means the inelastic cross section is exponentially suppressed at low energy. We present results for 100, 200, 300, and 400 G B fields and find that the cooling behavior is more favorable for larger fields. The limitation to the cooling will be the number of ions that can be trapped with the molecules and the inelastic collision between pairs of OH molecules. Since this inelastic collision rate also decreases rapidly with decreasing temperature, it seems likely that a small fraction of cold, positive ions could sympathetically cool neutral molecules.

We perform molecular dynamics simulation of ions and molecules in a combined Paul trap and TOP trap. We find that the asymptotic molecular temperature and the time scale needed to reach that temperature follows the simple estimates from the scattering rates. The *fundamental* lower bound on the OH temperature cannot be estimated from the data in this paper but we expect it to be well below 1 mK. Because the mechanism that suppresses the inelastic ion-molecule collision is a generic property of perturbed quantum systems, the results in this paper should be generally applicable for neutral molecules with magnetic and electric dipole moments. Furthermore, since the inelastic molecule-molecule transition is due to a shorter-range interaction, it seems likely that cooling will be possible for many, if not most, molecules with magnetic and electric dipole moments.

ACKNOWLEDGMENTS

We thank C. H. Greene for pointing out relevant earlier work and insightful conversations. This work was supported by the Chemical Sciences, Geosciences, and Biosciences Division of the Office of Basic Energy Sciences, US Department of Energy.

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