

# Cavity-mediated collisionless sympathetic cooling of molecules with atoms

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## Abstract

Cooling a range of molecules to ultracold temperatures ( $<1$  mK) is a difficult but important challenge in molecular physics and chemistry. Collective cavity cooling of molecules is a promising method that doesn't rely on molecular energy level and thus can be applied to all molecules in principle. However, the initial lack of cold molecules leads to the difficulty in its experimental implementation. We show that efficient collective sympathetic cooling of molecules to sub-mK temperatures using a large ensemble of atoms within a cavity is feasible. This approach is a new type of sympathetic cooling which does not rely on direct collisions between atoms and molecules, but utilizes thermalization via their mutual interaction with a cavity field. Two important mechanisms are identified. This include: (1) giant enhancement of cavity optical field from the efficient scattering of the pump light by the atoms; (2) cavity-mediated collective interaction between the atoms and the molecules. We show an optimal cavity detuning for maximizing cooling, which is dependent on the atom and molecule numbers. We determine a threshold for the molecular pump strength and show that it is independent of molecule number when the number of atoms is much greater than the molecules. This can be reduced by orders of magnitude when compared to cavity cooling of single molecular species only. Using this new sympathetic cavity cooling technique, cooling molecules to sub-mK within a high-Q cavity could be within reach of experimental demonstration.

PACS numbers: 37.10.Mn, 37.10.Vz

The creation and application [1–3] of cold ( $<1$ K) and ultracold ( $<1$  mK) molecules is now being pursued within the molecular physics [4] and quantum chemistry [5, 6]. Cold molecules could be used for molecular clocks [7] covering broader frequency range than atom clocks. They offer the prospect of testing the time dependence fundamental constants [8]. Already cold molecules have been used in precision molecular spectroscopy with great potential for tests beyond the standard model [9]. Ultracold polar molecules with well controlled long-range anisotropic dipole-dipole interactions are of particular interest for their application in quantum computation [3] and in quantum simulation of exotic strongly-correlated many-body phenomena [1]. In addition, quantum matter-wave optics beyond Kerr nonlinear optics has been proposed [10]. The cold and ultracold regime offers a new frontier in chemistry where quantum tunneling dominates in chemical reaction [6], opening up a new approach to the creation of new materials [11].

Cold molecules can be generated using many techniques, such as buffer gas cooling [12], electric Stark deceleration [13], optical lattice deceleration [14], Zeeman deceleration [15], and velocity filtering [16]. However, ultracold molecules, except for ultracold alkali molecules via association of ultracold atoms [17, 18], are hardly produced. Now, developing general ways of producing ultracold molecules is a big theoretical and experimental challenge in ultracold molecule research.

Recently, sympathetic cooling has been attempted for generating ultracold molecules through collision with pre-cooled atoms [19]. In this cooling process, elastic collision can lead to translational cooling of molecules and even the internal rotation cooling. However, inelastic collision leads to heating and trap loss of the molecules, thus, a

high ratio of elastic collision rate to inelastic collision rate is critical for efficient sympathetic cooling. Moreover, the rate of cold chemical reaction which can quench sympathetic cooling should also be low. Now experimental implementation of this technique for most of molecules faces the lack of the detailed collision knowledge.

Doppler cooling and Sisyphus cooling of SrF molecules [20] have been demonstrated with multiple lasers for controlling 44 energy levels. However, such a direct laser cooling is hardly applicable to most of molecules whose complex energy levels require many repumping lasers required by Doppler cooling. Collective cavity cooling [21–27], which uses cavity leakage to dissipate kinetic energy, is a candidate for cooling molecules to sub-mK regime. To avoid direct using molecular energy levels, a far-off resonant pump laser is required. Consequently, it is not capable of generating the sufficiently strong dispersive coupling to the cavity field, leading to slow cavity cooling and much higher pump thresholds for cooling. To overcome these difficulties, a huge number ( $\sim 10^9$  [27]) of cold molecules are needed. Unfortunately, it is difficult for current methods [12–16] to produce the large number of cold molecules required.

In this letter, we show that collective sympathetic cooling of molecules with atoms mediated by an optical cavity is capable of rapidly cooling essentially any molecular species. This cooling uses the effective enhancement of the cavity optical field by the large number of ultracold atoms that can be placed in the cavity which cools the much smaller number of molecules. We find that when both atoms and molecules within a high-Q cavity are side pumped with a strong far-off resonant light, the cooling rate of molecules can be significantly enhanced by increasing the number of atoms, while reducing the

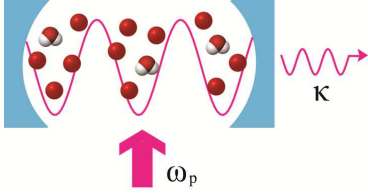


FIG. 1: Atoms and molecules are loaded in the cavity, and side pumped by a far-off resonant light with frequency  $\omega_p$ . Cavity optical field leaks from the cavity with a rate  $\kappa$ .

pump laser power by orders of magnitude. Our new cooling scheme does not need a large number of cold molecules and does not require knowledge of the complicated, typically inelastic interactions of conventional sympathetic cooling schemes. These simplifications put it firmly within reach of experimental realization.

Our scheme is shown in Fig. 1. Atoms of mass  $m_a$  with spontaneous emission rate  $\gamma_a$ , and molecules of mass  $m_m$  with spontaneous emission rate  $\gamma_m$  within a cavity with a leakage rate  $\kappa$  and a resonant frequency  $\omega_c$  are side pumped by an optical field with frequency  $\omega_p$ . In our following analysis, we assume that the pump frequency is far-off resonant from any transition of the atoms and molecules, for the purpose of developing a technique without using the detailed knowledge of molecular energy levels. For simplifying our analysis, we make two other assumptions: (1) the transverse motion of the atoms and molecules are frozen, so that we only need to investigate their motion along the cavity axis; (2) any potential collision between atoms and molecules has been neglected. With these assumptions and extension of the cavity cooling theory for atoms [21–23], the cavity optical field  $\alpha$  and the center-of-mass motion for the atoms and the molecules are described by the following equations:

$$\begin{aligned} \frac{d\alpha}{dt} = & (i\Delta_C - \kappa)\alpha - \sum_j [(iU_0^a + \Gamma_0^a) \cos^2(\tilde{z}_j^a) \alpha + \eta_{eff}^a \cos(\tilde{z}_j^a)] \\ & - \sum_l [(iU_0^m + \Gamma_0^m) \cos^2(\tilde{z}_l^m) \alpha + \eta_{eff}^m \cos(\tilde{z}_l^m)] + \xi_\alpha \end{aligned} \quad (1)$$

$$\frac{d\tilde{p}_j^a}{dt} = \Re U_0^a \sin(2\tilde{z}_j^a) + 2 \text{Im}(\eta_{eff}^a \alpha^*) \sin(\tilde{z}_j^a) + \xi_j^a \quad (2)$$

$$\frac{dz_j^a}{dt} = \frac{p_j^a}{m_a} \quad (3)$$

$$\frac{d\tilde{p}_l^m}{dt} = \Re U_0^m \sin(2\tilde{z}_l^m) + 2 \text{Im}(\eta_{eff}^m \alpha^*) \sin(\tilde{z}_l^m) + \xi_l^m \quad (4)$$

$$\frac{dz_l^m}{dt} = \frac{p_l^m}{m_m} \quad (5)$$

in which  $\Delta_C = \omega_p - \omega_C$ ,  $p_j^a$  and  $p_l^m$  are respectively the momentum for the  $j$ -th atoms and the  $l$ -th molecules, and their corresponding positions are denoted with  $z_j^a$  and  $z_l^m$ . Their momentums and coordinates are scaled by  $\tilde{p}_j^a = p_j^a / (\hbar k_c)$ ,  $\tilde{p}_l^m = p_l^m / (\hbar k_c)$ ,  $\tilde{z}_j^a = k_c z_j^a$ ,  $\tilde{z}_l^m = k_c z_l^m$  ( $k_c$  is the wave number of the cavity optical field). Parameter  $\Re$  is given by  $\Re = |\alpha|^2 - 1/2$ . Parameters  $U_0$  and  $\Gamma_0$  respectively denote the dispersive and absorption effect, and  $\eta_{eff}$  is the effective pumping strength. For the atoms and the molecules respectively with the detuning  $\Delta_a$  and  $\Delta_m$  under the pump optical field, these parameters are given by  $U_0^a = g_a^2 \Delta_a / (\Delta_a^2 + \gamma_a^2)$ ,  $U_0^m = g_m^2 \Delta_m / (\Delta_m^2 + \gamma_m^2)$ ,  $\Gamma_0^a = g_a^2 \gamma_a / (\Delta_a^2 + \gamma_a^2)$ ,  $\Gamma_0^m = g_m^2 \gamma_m / (\Delta_m^2 + \gamma_m^2)$ ,  $\eta_{eff}^a = \eta_a g_a / (-i\Delta_a + \gamma_a)$  and  $\eta_{eff}^m = \eta_m g_m / (-i\Delta_m + \gamma_m)$  with atom/molecule-cavity coupling  $g_{a/m}$  and the pump strength  $\eta_{a/m}$  [28]. The noise terms can be decomposed into  $\xi_\alpha = \xi_\alpha^0 + \xi_\alpha^1$ ,  $\xi_j^a = \xi_j^{a,0} + \xi_j^{a,1}$ ,  $\xi_l^m = \xi_l^{m,0} + \xi_l^{m,1}$  which are given by

$$\langle |\xi_\alpha^0|^2 \rangle = \kappa + \frac{\Gamma_0^a}{2} \sum_j \cos^2(\tilde{z}_j^a) + \frac{\Gamma_0^m}{2} \sum_i \cos^2(\tilde{z}_i^m), \quad (6)$$

$$\langle |\xi_j^{a,0}|^2 \rangle = 2\bar{u}^2 \Gamma_0^a [\Re \cos^2(\tilde{z}_j^a) + 2\tilde{\eta}_a \alpha_r \cos(\tilde{z}_j^a) + \tilde{\eta}_a^2], \quad (7)$$

$$\langle |\xi_l^{m,0}|^2 \rangle = 2\bar{u}^2 \Gamma_0^m [\Re \cos^2(\tilde{z}_l^m) + 2\tilde{\eta}_m \alpha_r \cos(\tilde{z}_l^m) + \tilde{\eta}_m^2], \quad (8)$$

$$\xi_\alpha^1 = \frac{i\alpha}{|\alpha|} \left[ \sqrt{\frac{\Gamma_0^a}{2}} \sum_j \cos(\tilde{z}_j^a) \zeta_j^a + \sqrt{\frac{\Gamma_0^m}{2}} \sum_l \cos(\tilde{z}_l^m) \zeta_l^m \right], \quad (9)$$

$$\xi_j^{a,1} = -\sqrt{2\Gamma_0^a} |\alpha| \sin(\tilde{z}_j^a) \zeta_j^{(a)}, \quad (10)$$

$$\xi_l^{m,1} = -\sqrt{2\Gamma_0^m} |\alpha| \sin(\tilde{z}_l^m) \zeta_l^{(m)}, \quad (11)$$

in which  $\tilde{\eta}_a = \eta_a / g_a$ ,  $\tilde{\eta}_m = \eta_m / g_m$ , and  $\alpha_r = \text{Re}(\alpha)$ ,  $k_c \sqrt{\bar{u}^2}$  represents the mean projection of the momentum recoil along the cavity axis due to spontaneous emission.

Before numerically solving above equations of motion, we first qualitatively analyze the possible mechanisms for the collisionless sympathetic cooling of molecules with the atoms. First, this collisionless sympathetic cooling uses the giant enhancement of the cavity optical field from the efficient scattering of the pump light by the atoms. As shown in Eq. (1), the cavity optical field is induced by the scattered light from both the atoms and the molecules. When the molecules are pumped by a very far-off resonant light (typically,  $\Delta_m \sim 10^{15}$  Hz), the scattering light from a small number of molecules is negligibly weak; however the cavity optical field can

be effectively enhanced by adding the atoms with a off-resonant detuning ( $\Delta_a \sim 10$  GHz), and thus cooling efficiency of the molecules can be improved with the assistance of the atoms. Second, this collisionless sympathetic cooling profits from cavity-mediated collective interaction between the atoms and molecules. Eqs. (2) and (4) show that the optical forces on an atom or a molecule are functions of the cavity optical field. Whereas, the cavity optical field, as shown in Eq. (1), is influenced by the motion of all the atoms and molecules, thus the optical force exerted on a single atom or a single molecule is contributed from the influence from all other atoms and molecules, which means that cavity photon-mediated interaction between atoms and molecules exists [29], leading to possibility of sympathetic cooling of molecules with fast cooled atoms. It is hard for us to single out familiar two-body interaction between one single atom and one single molecule, but collective interaction between atoms and molecules exists. So, a large number of atoms and molecules can enhance the collective interaction and further speed up the sympathetic cooling of molecules.

We solved the above equations of motion for Rb atoms and CN molecules at initial temperatures 10 mK with  $\Delta_a = -20$  GHz,  $\Delta_m = 2.45 \times 10^{15}$  Hz, cavity detuning  $\Delta_c = -\kappa + U_0^a N_a + U_0^m N_m$  ( $N_a$  and  $N_m$  are the total number of the atoms and the molecules respectively), and  $\kappa = 20$  MHz. Figure 1 shows the time evolution of temperatures  $T$  of the atoms and the molecules at a pump strength  $\eta_a = 5000\kappa$  for three cases: (1), cavity cooling of atoms only ( $N_m = 0$ ,  $N_a = 10^6$ , black line is for the atomic temperature); (2) cavity cooling of molecules only ( $N_m = 10^4$ ,  $N_a = 0$ , red line is for the molecular temperature); (3) cavity cooling of atoms and molecules mixture ( $N_m = 10^4$ ,  $N_a = 10^6$ , the blue line is for the atomic temperature, and the yellow line is for molecule temperature). As shown in Fig. 1, cavity cooling efficiency of molecules is much enhanced by adding a large number of atoms into the cavity. The atoms are cooled faster than the molecules, so the relatively cooler atoms can cool the relatively hotter molecules through cavity-mediated collective interaction between atoms and molecules. Consequently, comparing with black line (only atoms), cooling of the atoms is slowed down when the molecules is added. This slowing down is a signal of collisionless sympathetic cavity cooling of molecules with atoms.

We further study the relation of sympathetic cavity cooling efficiency to the pumping power. Figure 3 shows the time evolution of temperature for  $10^4$  molecules assisted by  $10^6$  atoms for pumping strength  $\eta_a = 500\kappa$  (red line), and  $5000\kappa$  (black line). Figure 3 shows that cooling time increases with the decreasing of the pump power. We can imagine that the sympathetic cavity cooling of molecules with atoms has minimum requirement for the pump laser power.

We now discuss the threshold of the pump laser power. Similar to cavity cooling of single species, sympathetic

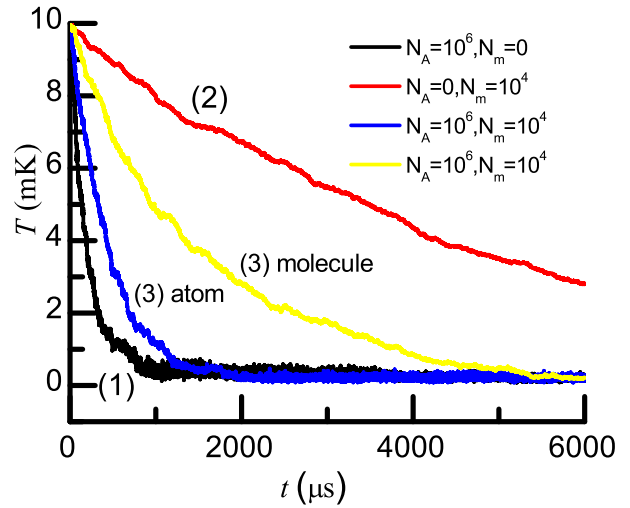


FIG. 2: Times evolution of the temperatures for the atom ensemble and the molecule ensemble for three cases: (1) only  $10^6$  atoms are in the cavity (black line); (2) only  $10^4$  molecules are in the cavity (red line); (3)  $10^6$  atoms and  $10^4$  molecules are in the same cavity (yellow line for molecules, and blue line for atoms).

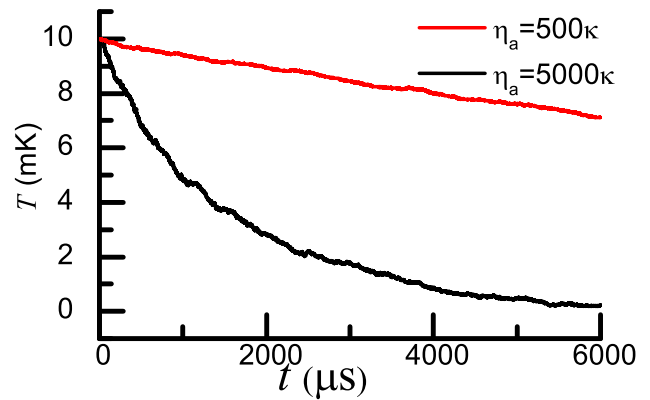


FIG. 3: Time evolution of the molecule temperature when  $10^6$  atoms and  $N_m = 10^4$  molecules are in the cavity for different pump strengths  $\eta_a = 5000\kappa$  (black) and  $500\kappa$  (red)

cavity cooling of molecules relies on self-organization of molecules and atoms in the cavity. The atoms and the molecules are initially distributed uniformly in the cavity with statistic fluctuation. When the pump laser is turned on, the cavity optical field starts to build up through light scattering from the molecules and atoms. The intra-cavity field and the scattering optical field will lead to localization of molecules and atoms in positions around the position  $k_c x = 2n\pi$  and  $(2n+1)\pi$  ( $n$  is an integer). Such a self-organization requires the intra-cavity field is strong enough to produce a deep potential well to suppress the thermal motion. Since the dispersion couplings

$U_0^a$  and  $U_0^m$  are weak in the far-off resonant optical field, the potential well depths for the atoms and molecules are respectively approximated by

$$U_{a/m} \approx 2\hbar\eta_{a/m}g_{a/m}\alpha_r/\Delta_{a/m}. \quad (12)$$

The real part of  $\alpha$ ,  $\alpha_r$ , can be estimated by the steady solution of the Eq. (1), which is given by

$$\alpha_r = \frac{2(\frac{\eta_m g_m}{\Delta_m} \delta N_m + \frac{\eta_a g_a}{\Delta_a} \delta N_a) [\Delta_c - \frac{U_0^a N_a + U_0^m N_m}{2}]}{\pi \left\{ \left[ \Delta_c - \frac{U_0^a N_a + U_0^m N_m}{2} \right]^2 + \left[ \kappa + \frac{\Gamma_0^a N_a + \Gamma_0^m N_m}{2} \right]^2 \right\}}. \quad (13)$$

In deducing Eq. (13), we have used  $\cos^2(k_C z_j^{(a)}) \approx N_a/2$ ,  $\cos^2(k_C z_i^{(a)}) \approx N_m/2$ ,  $\cos(k_C z_j^{(a)}) \approx 2N_a/\pi$ , and  $\cos(k_C z_i^{(a)}) \approx 2N_m/\pi$  in the vicinity of threshold [23, 27].

When

$$\Delta_c = \pm \left[ \kappa + \frac{\Gamma_0^a N_a + \Gamma_0^m N_m}{2} \right] + U_0^a N_a + U_0^m N_m, \quad (14)$$

$\alpha_r(\Delta_c)$  gets its maximum value, and thus achieving the deepest potential well depths for  $U_a$  and  $U_m$ . So, in the following, we keep the optimum cavity detuning given by Eq. (14), and now

$$\alpha_r = \frac{(\eta_m g_m / \Delta_m \delta N_m + \eta_a g_a / \Delta_a \delta N_a)}{\pi \left( \kappa + \frac{\Gamma_0^a N_a + \Gamma_0^m N_m}{2} \right)}. \quad (15)$$

Equation (15) shows that when  $\Delta_a \ll \Delta_m$  and  $N_m \ll N_A$ ,  $\eta_m g_m / \Delta_m \delta N_m \ll \eta_a g_a / \Delta_a \delta N_a$ , the cavity optical field dominantly comes from the scattering of the pump light by the atoms. In this situation, using the self-organization requirement  $U_m \geq k_B T_f$  [27], in which  $T_f = 2\hbar\kappa/k_B$  is the finally achieved cooling temperature [23], we get a formula for the threshold for the molecular pump strength,

$$\eta_m^{th} = \sqrt{\frac{\pi}{2Rg_m g_a} \frac{\kappa^2 \Delta_m \Delta_a}{\delta N_a}}. \quad (16)$$

in which  $R = g_a/g_m$  [30].

If only molecules are cooled within the cavity, the threshold  $\eta_m^{th,s} = \sqrt{\pi\kappa^2\Delta_m^2/(2g_m^2\delta N_m)}$ . The ratio of  $\eta_m^{th}$  to  $\eta_m^{th,s}$  is

$$\eta_m^{th}/\eta_m^{th,s} = \sqrt{\frac{g_m}{Rg_a} \frac{\Delta_a}{\Delta_m} \frac{\delta N_m}{\delta N_a}}. \quad (17)$$

Using typical values  $\Delta_m \sim 10^{15}$  Hz,  $\Delta_a \sim 10^{10}$  Hz,  $R \approx 14.7$  and  $g_a/g_m=4.4$  in our numerical calculation, even when  $\delta N_a = \delta N_m$ ,  $\eta_m^{th}/\eta_m^{th,s} \approx 3.93 \times 10^{-4}$ . Since  $\delta N_{a/m} \sim \sqrt{N_{a/m}}$ , this ratio can be further lowered by

increasing the ratio  $N_A/N_m$ . Thus, the threshold for molecular pump strength  $\eta_m^{th}$  with the assistance of atoms can be reduced by at least four orders of magnitude, compared with  $\eta_m^{th,s}$  without using atoms. Physically, this greatly reduced threshold arises from the much enhanced cavity optical field from the scattering of the pump light by the atoms.

Equation (16) shows several ways to reduce the threshold of the pumping power for molecules: reducing the atom detuning and increasing the atom number. Also, using a cavity with a very low leakage rate  $\kappa$  is useful for reducing the threshold.

Similarly, the threshold for atomic pump strength is  $\eta_a^{th} = \sqrt{\pi\kappa^2\Delta_a^2/(2g_a^2\delta N_a)}$  which is in general much smaller than that for cooling molecules when  $\Delta_a \ll \Delta_m$ . Thus, we observe atoms are always cooled before molecules, as shown in Fig 2, and the cooled atoms can be further used to collective sympathetic cooling of molecules through cavity-mediated interaction between the atoms and the molecules.

In summary, we have shown that the efficiency of collective cavity cooling of molecules with the assistance of atoms within the same cavity can be greatly improved. We have obtained a formula determining the optimum cavity detuning for efficient cooling. We also have deduced an analytical formula for the pump power threshold for molecular cavity cooling, and found that the threshold, independent of the molecule number, can be reduced by several orders of magnitude. Two mechanisms contributed to the enhanced cavity cooling of molecules are (1) the enhancement of cavity optical field from the efficient scattering of the pump light by the atoms; (2) cavity-mediated collective interaction between atoms and molecules. Using the current cavity cooling scheme, cavity cooling of even a small number of cold molecules down to sub-mK temperatures is experiment feasible.

Our scheme presents a new kind of sympathetic cooling, which does not use direct collision between atoms and molecule. However, when a larger ensemble of atoms is loaded into the cavity with the molecules, the direct collision between the atoms and molecules, which has been neglected in our analysis, will come to play. In general, elastic collision will enhance the cavity cooling of molecules. However, the role of inelastic collision is unclear. Moreover, chemical reaction within the cavity could also be induced. To avoid these direct collision effects, we can use a potential barrier to separate atoms and molecules in the cavity.

Our results will have implications for a number of future theoretical and experimental issues, such as single molecule manipulation within a cavity, cavity-mediated cold collision and cold chemistry. Especially, with the cavity leakage rate is further reduced in future, ultra-cold dense degenerate molecular gases would be produced within a cavity directly, getting rid of evaporative cool-

ing.

**Acknowledgement 1** *In the preparation of this manuscript, from private communication with Prof. H. Ritsch, we learn that a similar idea has appeared in Ref. [31]. This work was supported by the National Basic Research Program of China (973 Program) under Grant Nos. 2011CB921604 and 2011CB921602, and the National Natural Science Foundation of China under Grants Nos. 10874045, 11034002 and 10828408.*

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