

Cavity sideband cooling of trapped molecules

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The efficiency of cavity sideband cooling of trapped molecules is theoretically investigated for the case in which the infrared transition between two rovibrational states is used as a cycling transition. The molecules are assumed to be trapped either by a radiofrequency or optical trapping potential, depending on whether they are charged or neutral, and confined inside a high-finesse optical resonator that enhances radiative emission into the cavity mode. Using realistic experimental parameters and COS as a representative molecular example, we show that in this setup, cooling to the trap ground state is feasible.

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I. INTRODUCTION

With the field of cold and ultracold molecules coming of age in the past few years, efficient cooling methods are becoming more and more valuable. Optical methods have the advantage that they do not rely on collisional processes, which are hard to predict and which often open severe collisional loss channels. A number of ingenious optical cooling schemes for molecules have been devised: It has been realized, e.g., that translational cooling of molecules might be possible with light sources with a specifically designed set of lines [1,2]. Alternatively, in the almost-closed level scheme, a molecule like CaF has been exploited [3] to laser-cool molecules, as is usually done [4]. Even a three-dimensional (3D) scheme has been proposed resembling the popular magneto-optical trap for atoms [5]. Zeppenfeld *et al.* have proposed to use electro-optical forces to carry away up to 1 K per spontaneously decaying infrared (IR) photon [6]. Optical cavities have been predicted [7,8] and used for atoms [9] as a way to achieve translational cooling without relying on spontaneous emission, in principle allowing cooling of molecules. In Refs. [10,11], we have shown that cavity-enhanced Raman scattering can enable one to cool both the internal and the external degrees of freedom of molecules, using simulations based on *ab initio* calculations for OH and NO radicals. In this proposal, the cavity enhances the anti-Stokes cooling transitions over other heating transitions. In a recent paper, Vuletić and co-workers demonstrated sideband cooling of the motion of a single trapped ion to the ground state of the trap potential via cavity-enhanced scattering [12]. In the present paper, we take the next step and demonstrate theoretically cavity-enhanced sideband cooling of molecules and molecular ions in a strongly confining external potential, in the setup sketched in Fig. 1. We show that cooling to the ground state of the external potential can be achieved for experimentally feasible setups. The new scheme thus offers the possibility to reach lower final temperatures compared to the previously reported cooling schemes [8,10], where the limit is given by the cavity linewidth.

This paper is organized as follows. In Sec. II, the theoretical model is introduced and the derivation of the rate equations for cooling the external motion of the molecules is sketched. Moreover, the method used for obtaining the numerical results

is described. In Sec. III, the results are reported and discussed, while the conclusions are drawn in Sec. IV.

II. THE MODEL

We theoretically explore the efficiency of performing sideband cooling of a molecule of mass M , which is confined by a harmonic potential and couples to the mode of a high-finesse resonator. The level scheme is sketched in Fig. 2: a two-level transition of the molecule is driven transversally by a laser with coupling strength Ω (Rabi frequency), and emits photons at frequency ω_c . The bad-cavity limit is assumed, which implies that the molecule scatters prevalently photons into the cavity mode, which are then lost via cavity decay.

Let us now provide a specific example. The two-level transition, which is driven by electromagnetic radiation, is composed of two vibrational states $v = 0$ and 1 of the electronic ground state of a molecule, which we denote by $|g\rangle$ and $|e\rangle$, respectively. Let ω_0 be the transition frequency. This level scheme is closed inside the vibrational subspace. Spontaneous emission into different rotational substates is suppressed by the cavity, while any residual leakage can be handled by microwave repump schemes. These properties allow one to use this transition for cavity cooling.

We further note that this sort of vibrational transition is usually in the infrared regime ($100\text{--}4000\text{ cm}^{-1}$) and has typically a homogeneous linewidth below 1 kHz, which allows one to spectrally resolve the motional sidebands. In order to obtain convenient cooling rates, and hence efficient cooling, spontaneous emission is enhanced here by coupling with the mode of an infrared high-finesse resonator. The spatial confinement can be obtained by means of dipole or ion traps, depending on whether the molecule is neutral or charged [13–15].

In order to perform sideband cooling, the initial spatial localization of the molecule must be smaller than the wavelength of the driving laser. This requirement can be rewritten as $\eta\sqrt{2\langle n \rangle + 1} \ll 1$, with $\langle n \rangle$ the mean occupation of phononic states of the harmonic oscillator and η the Lamb-Dicke parameter, $\eta = \sqrt{\hbar k^2 / 2M\nu}$, with k the wave vector of the transition and ν the trap frequency in angular units [16,17]. One necessary condition is that the Lamb-Dicke parameter

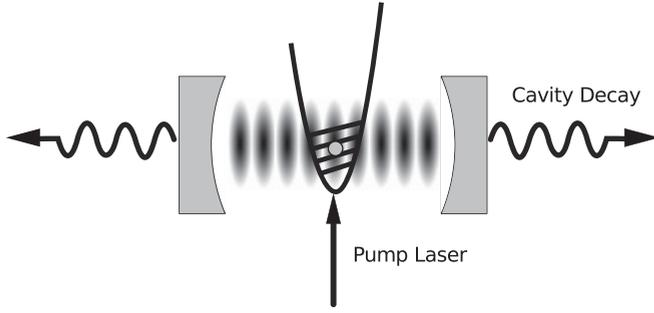


FIG. 1. Schematic representation of the setup. A molecule is confined by an external harmonic potential inside a high-finesse resonator. The transition between two molecular vibrational levels is driven transversally by a laser and scatters photons into a quasiresonant cavity mode. The scattering processes are tailored to enhance the transitions that cool the external motion.

is smaller than unity $\eta \ll 1$. Moreover, the molecule should be pre-cooled, e.g., by means of an optical cooling scheme [7,8] or other nonoptical methods [6]. Nevertheless, we note that sideband cooling can be efficient even for $\eta \sim 1$ [18]. Moreover, schemes for ground-state cooling for Lamb-Dicke parameters larger than unity have been theoretically proposed in [18], and can be extended to situations in which dissipation occurs via cavity decay.

A. Master equation for cavity sideband cooling

We now introduce the model, from which we extract the cooling rates and the final temperature expected by cavity sideband cooling the molecules. Below, we assume the Lamb-Dicke regime and hence expand in first order in the small parameter η [16,17]. The model has been reported on extensively, for instance in Refs. [19,20].

We study the coupled dynamics of the center-of-mass motion of the molecule, the two-level transition, and the cavity mode in the reference frame rotating at the frequency ω_L of the laser, driving the two-level molecular transition. Denoting by ρ the density matrix for molecular internal and external

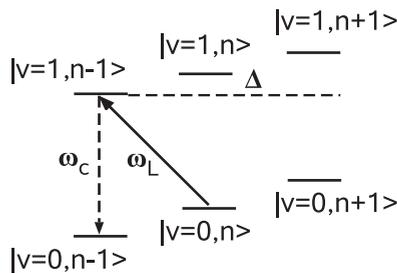


FIG. 2. Schematic representation of the scattering process leading to cooling. The molecular vibrational levels are labeled by $v = 0, 1$, and the phononic trap excitations by n . The pump laser at frequency ω_L drives resonantly the red sideband transition $|v = 0, n\rangle \rightarrow |v = 1, n - 1\rangle$. The laser photon is scattered into a cavity mode at frequency $\omega_c = \omega_0$, which couples resonantly with the molecular transition. Since the photon is then lost via cavity decay, the molecule is cooled by one phonon.

degrees of freedom (in this reduced Hilbert space) and for the cavity mode, its dynamics is governed by the master equation

$$\partial_t \rho = \frac{1}{i\hbar} [H, \rho] + \mathcal{L}_\kappa \rho + \mathcal{L}_\gamma \rho, \quad (1)$$

where the Hamiltonian H accounts for the coherent evolution, and the superoperators \mathcal{L}_κ and \mathcal{L}_γ describe cavity decay and spontaneous emission, respectively. We now give the specific form of each term. The full Hamiltonian can be written as the sum of the terms

$$H = H_M + H_T + H_C + V_I, \quad (2)$$

where

$$H_M = -\hbar \Delta |e\rangle \langle e| \quad (3)$$

is the Hamiltonian for the internal degrees of freedom, with $\Delta = \omega_L - \omega_0$. The Hamiltonian H_T describes the dynamics of the harmonic oscillator for the center-of-mass motion, which is assumed here to be one dimensional for convenience,

$$H_T = \frac{P^2}{2M} + \frac{1}{2} M v^2 X^2 = \hbar v \left(b^\dagger b + \frac{1}{2} \right), \quad (4)$$

with X and P the position and canonical conjugate momentum of the molecule, respectively, and $b = (iP + MvX)/\sqrt{2\hbar m v}$ annihilates a quantum of motional excitations $\hbar v$ such that $[b, b^\dagger] = 1$. We denote by $\{|n\rangle\}$ the number states with $n = 0, 1, 2, \dots$, which are eigenstates of the Hamiltonian in Eq. (4). The Hamiltonian for the cavity mode is

$$H_C = -\hbar \delta_c a^\dagger a, \quad (5)$$

where operators a and a^\dagger annihilate and create, respectively, a cavity photon at frequency ω_c , and $\delta_c = \omega_L - \omega_c$ is the detuning between the laser and the cavity mode.

In the bad-cavity regime, we may reduce the Hilbert space of the cavity mode to the states $|0_c\rangle$ and $|1_c\rangle$, namely zero and one photon in the cavity mode, respectively. The coupling of the molecule with the laser and with the cavity mode are given by operator V_I , which is reported here in the rotating wave approximation (RWA), as the pump is assumed to be sufficiently weak to neglect the effect of counter-rotating terms. The interactions in the Lamb-Dicke expansion are given by

$$V_I = V_I^{(0)} + V_I^{(1)}(b + b^\dagger) + O(\eta^2),$$

where the superscript gives the corresponding perturbative order. The zero-order term reads [19,20]

$$V_I^{(0)} = \hbar(\Omega + g a^\dagger \cos \phi) \sigma + \text{H.c.}, \quad (6)$$

where Ω is the Rabi frequency, g is the vacuum Rabi frequency, and ϕ is the phase between a maximum of the cavity standing wave and the center of the trap. Operators $\sigma = |g\rangle \langle e|$ and its adjoint σ^\dagger flip the molecular excitation.

The term in first order in the Lamb-Dicke expansion includes the coupling with the external degrees of freedom and reads [19,20]

$$V_I^{(1)} = \hbar \eta (i \Omega \cos \Theta_L - g a^\dagger \cos \Theta_C \sin \phi) \sigma + \text{H.c.}, \quad (7)$$

where Θ_L (Θ_C) is the angle between the axis of the motion and the laser (cavity) wave vectors.

The superoperators, describing dissipation in Eq. (1), take the form

$$\mathcal{L}_\kappa \rho = \frac{\kappa}{2} (2a\rho a^\dagger - \{a^\dagger a, \rho\}), \quad (8)$$

$$\mathcal{L}_\gamma \rho = \frac{\gamma}{2} (2\sigma \tilde{\rho} \sigma^\dagger - \{\sigma^\dagger \sigma, \rho\}), \quad (9)$$

with the molecular (cavity) linewidth γ (κ) and $\tilde{\rho}$ accounting for the mechanical effects of the spontaneously emitted photon on the motion [19]. Since spontaneous decay is very slow with respect to the other rates, in the simulations we will take $\tilde{\rho} \simeq \rho$, hence discarding terms of the order $\eta^2 \gamma$, which describe the diffusion due to the recoil of the spontaneously emitted photons.

B. Rate equations

We characterize the efficiency of cavity-enhanced sideband cooling by means of a rate equation, which gives the dynamics of the occupation of the oscillator levels $|n\rangle$, $p_n = \text{Tr}\{|n\rangle\langle n|\rho\}$. This treatment is valid in the limit in which the dynamics of internal and cavity degrees of freedom is faster than the external motion, which requires $\eta \ll 1$. In this case, one can assume that the coupled system, given by the cavity mode and two-level transition, reaches a steady state given by the density matrix ϱ_S , which is the solution of the equation $\mathcal{L}_0 \varrho_S = 0$. Here, ϱ_S is defined over the Hilbert space of the cavity mode and the internal degrees of freedom of the molecule, and

$$\mathcal{L}_0 \varrho = \frac{1}{i\hbar} [H_M + H_C + V_I^{(0)}, \varrho] + \mathcal{L}_\gamma \varrho + \mathcal{L}_\kappa \varrho \quad (10)$$

is the Liouville operator for the internal and cavity degrees of freedom. Under these assumptions, the rate equation determining the dynamics of the trap states takes the form

$$\begin{aligned} \dot{p}_n = & -[nA_- + (n+1)A_+]p_n \\ & + (n+1)A_- p_{n+1} + nA_+ p_{n-1}, \end{aligned} \quad (11)$$

with [19]

$$A_\pm = -2 \text{Re} \left[\text{Tr} \left\{ V_I^{(1)} \frac{1}{\mathcal{L}_0 \mp i\nu} V_I^{(1)} \varrho_S \right\} \right]. \quad (12)$$

Here, A_+ (A_-) is the heating (cooling) rate and $V_I^{(1)}$ is defined in Eq. (7). These are found by assuming that the Rabi frequency Ω is the smallest parameter, $\Omega \ll g, \gamma, \kappa, \nu$, which implies that the relevant internal and cavity states that are involved in the dynamics are the ground state $|v=0, 0_c\rangle$ and the excited states $|v=0, 1_c\rangle, |v=1, 0_c\rangle$. In the bad-cavity limit, state $|v=0, 1_c\rangle$ decays very fast, while $|v=1, 0_c\rangle$ is characterized by a relatively smaller rate of decay, such that $\kappa \gg g \gg \gamma$ [19,20]. An effective cooling rate W can be defined for the case $A_- > A_+$ [16],

$$W = A_- - A_+, \quad (13)$$

which gives the rate at which phonons are exponentially damped toward the steady-state value of the trap. The steady-state expectation value of the phonon occupation number is given by [16]

$$\langle n \rangle_{\text{st}} = \frac{A_+}{A_- - A_+}. \quad (14)$$

TABLE I. Parameters of the chosen molecules and their IR cooling transitions. The point group of the molecule and the irreducible representation of the corresponding vibrational normal mode are given. The transition frequencies, transition dipole moments (1 a.u. = 2.54 debye), and Einstein A coefficients γ are approximated from the *ab initio* frequency analysis.

	PG	Irred.	$\tilde{\nu}_{12}$ (cm ⁻¹)	μ_{12} (a.u.)	γ (s ⁻¹)
CHBr ₃	C_{3v}	E	635	0.10	5.6
HCCCF ₃	C_{3v}	A_1	1234	0.15	80
TMA	C_{3v}	A_1	2910	0.067	226
COS	$C_{\infty v}$	Σ_g	2108	0.15	424
CFI ₃	C_{3v}	A_1	1038	0.087	17.1
CSCI ₂	C_{2v}	A_1	1131	0.13	5.12
MgH ⁺	$C_{\infty v}$	Σ_g	1609	0.036	11

C. Numerical evaluation of the master equation

We check the analytical predictions by means of numerical calculations, which consists of performing the time evolution of the density matrix of the cavity and the molecule internal and external degrees of freedom according to Eq. (1). This is performed taking the Hilbert space of the cavity mode to be reduced to the photon states $|0_c\rangle, |1_c\rangle$, which is a reasonable choice considering that in the bad-cavity limit, the cavity mode is either in the vacuum state or has an occupation of one photon. Moreover, the molecular vibrational levels are approximated by a two-level system, which is resonantly driven by radiation. Finally, the center-of-mass oscillator is approximated by the first five levels. A convergence check for an increasing number of trap states has been carried out to ensure that the error due to the truncation does not significantly affect the results. The resulting density matrix has dimensions 20×20 . The integration of Eq. (1) is done with a fourth-order Runge-Kutta scheme to obtain the time evolution of the coupled quantum system. The time evolution of the center-of-mass motion is found by taking the partial trace of the total density matrix, as a function of time, over the internal and cavity degrees of freedom. In order to determine the cooling rate, it is assumed that the rate equations, Eq. (11), apply. Thus the time evolution of the trap states is fitted against the populations p_n to obtain A_+ and A_- , which allows us then to extract W and $\langle n \rangle_{\text{st}}$.

III. RESULTS

In order to evaluate the efficiency of cavity sideband cooling of molecules, we first identify some good candidates. We note that a strong vibrational transition favors large scattering into the cavity. Moreover, for the case of neutral molecules trapped by a far-detuned standing-wave laser, a large polarizability is needed to generate a sufficiently deep potential well. Several molecular candidates have been selected and calculated with the software package GAUSSIAN [21]. The equilibrium geometry of the molecules has been optimized with density functional theory and a high-quality atomic basis set [Becke three-parameter Lee-Yang-Parr functional augmented correlation-consistent polarized valence triple-zeta basis set (B3LYP/aug-cc-pVTZ) [22–24]]. Based on these

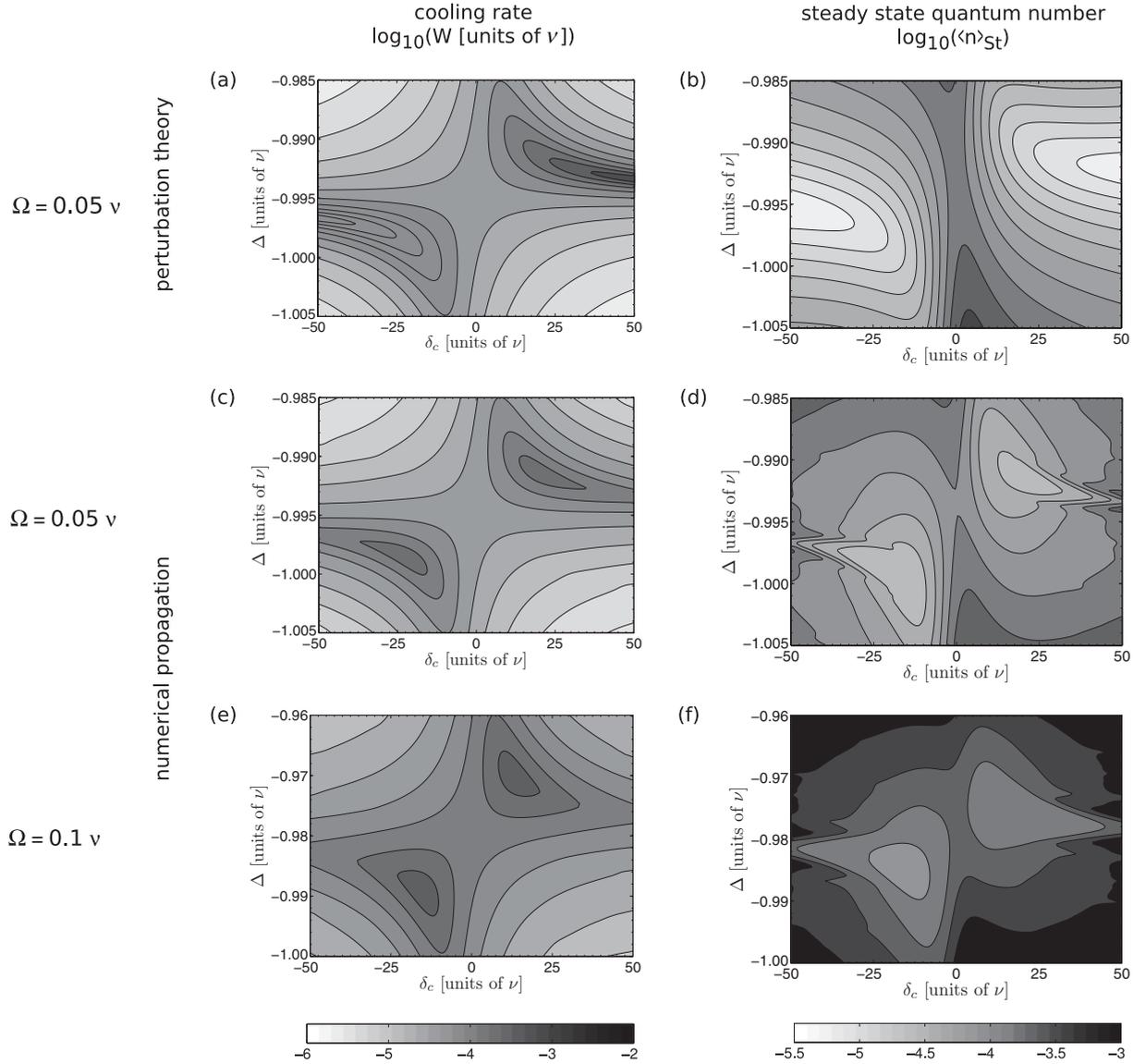


FIG. 3. Contour plots of the cooling rate (left column) and corresponding steady-state occupation (right column) of the motion of a trapped COS molecule driven at the asymmetric stretch vibration (2108 cm^{-1}) as a function of the laser-molecule detuning Δ and the laser-cavity detuning δ_c . The parameters are $g = 0.41\nu$, $\eta = 0.02$, $\kappa = 14\nu$, $\gamma = 1.9 \times 10^{-4}\nu$, and $\Theta_L = \Theta_C = \phi = \pi/4$. Subplots (a) and (b) correspond to the results found by applying perturbation theory Eq. (12) and taking $\Omega = 0.05\nu$. Subplots (c) and (d) are found for the same parameters as in (a) and (b), but are calculated from the density matrix found by numerical integration of Eq. (1). Subplots (e) and (f) are the same as (c) and (d) but with pump strength $\Omega = 0.1\nu$.

geometries, a frequency analysis and polarizability calculation have been carried out to determine the spontaneous emission rates γ of the excited vibrational state and polarizability tensors as the decisive molecular properties. A small choice of possible candidates that we have considered for implementing cavity sideband cooling is shown in Table I.

The molecule COS is selected from the list as a neutral candidate for its remarkable strong IR transition in the asymmetric stretch vibration. Its large polarizability makes it also a good choice for optical trapping. For the two-level system, $v = 0$ and 1 of the asymmetric stretch mode with the parameters given in Table I are chosen. The test candidate COS is assumed to reside in a standing-wave optical potential that has a trap depth of $\approx 900 \mu\text{K}$ and a trap frequency

$\nu = 2\pi \times 350 \text{ kHz}$ in the harmonic approximation. Such a potential could, for example, be formed by a laser with a wavelength of 532 nm and an effective power of 500 W achieved by a build-up cavity. The resulting Lamb-Dicke parameter is then $\eta = 0.02$. Molecular samples below 1 mK may be achieved in the near future [6], making it plausible to assume the Lamb-Dicke regime in our system.

The cavity axis and the pump laser axis is chosen to be in a 45° arrangement with the trap axis ($\Theta_C = \Theta_L = \pi/4$). The amplitude of the electric field of the cavity mode is $\varepsilon_c = 150 \text{ V m}^{-1}$ and the corresponding cavity linewidth is $\kappa = 2\pi \times 5 \text{ MHz}$. The single-particle cooperativity $C_1 = g^2/\kappa\gamma = 61$ warrants the strong-coupling regime, which is needed to provide a sufficiently strong enhancement of the

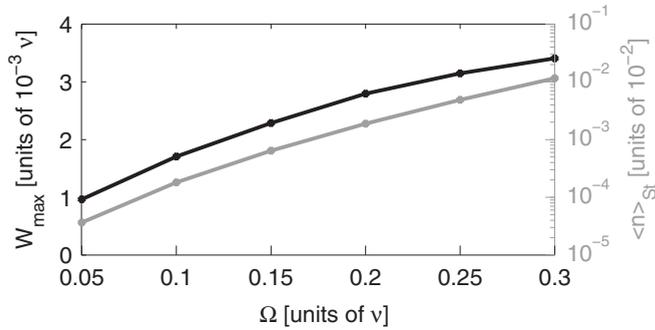


FIG. 4. Extremal values of the cooling rate (black line) and of the steady-state occupation (gray line) vs the laser Rabi frequency (in units of ν). The other parameters are the same as in Fig. 3.

scattering rate. However, it is important to notice that the cavity coupling g is smaller than the trap frequency ($g = 0.41\nu$). This is a major difference from previous theoretical works [20,25], which assumed significantly larger values of g with respect to ν , and it determines in our case the disappearance of most interference phenomena reported in [20,25].

In order to explore the parameter regime identified for the case of the COS molecule, the cooling and heating rates are calculated by the two different methods described in Sec. II C, namely the analytical predictions from perturbation theory and the numerical results found by integrating master equation (1).

Figure 3 displays the contour plots of cooling rate and steady-state phonon number as a function of laser-molecule detuning Δ and the laser-cavity detuning δ_c . The top row in Fig. 3 shows the predictions of the analytical model, while the second row reports the corresponding predictions of the numerical results for the same set of parameters. Their comparison shows discrepancies between analytics and numerics. This is understood: In fact, the perturbative treatment is based on the assumption that the Rabi frequency Ω is the smallest parameter, and that the excited state of the two-level transition is essentially empty [20], while for the parameters considered here with γ being the smallest rate, this is not fulfilled. We have also checked that in the limit in which the perturbative treatment is expected to be valid, the predictions of analytics and numerics are almost identical. In the following, we refer then to the numerical results. The analytical results, nevertheless, serve a double purpose. In the first place, they allow one to identify the parameter interval at which cooling is optimal (allowing us to quickly find the region where the numerical effort should be concentrated). Moreover, since the dynamics described by the analytical solution is well understood (see, e.g., [19,20]) in comparison with the numerical results, they permit one to identify the physical origin of the discrepancies.

We first notice that efficient cooling is found for the values of the detuning at which the red sideband of the narrow dressed state of the cavity and the molecule is driven. At large values of the detuning, $|\delta_c| \rightarrow \infty$, when the coupling with the cavity mode becomes irrelevant, this dressed state tends to the molecular vibrational state $v = 1$, and optimal cooling is thus found when $\Delta = -\nu$, which is the sideband cooling condition in free space [16,17]. Correspondingly, the temperature is minimal. We also find that the numerical results

predict that the cooling rate exhibits a maximum (and the finite occupation a minimum) about a finite value of $|\delta_c|$. Comparison with the analytical results shows that this behavior must be due to saturation effects, which are not accounted for in the perturbative treatment. Indeed, for the chosen parameters, the laser saturates the red sideband transition (being $\eta\Omega > \gamma$). As a consequence, the mean vibrational occupation at steady state, extracted from the numerical method and displayed in Fig. 3(d), shows oscillations as the parameters are varied across the sideband resonance. The cooling rate that we extract at the maximum is $W = 2 \times 10^3 \text{ s}^{-1}$.

Figures 3(e) and 3(f) display the numerical predictions for the cooling rate and steady-state occupation when the parameters are the same as in Figs. 3(c) and 3(d), but the strength of the pump is doubled, $\Omega = 0.1\nu$. Larger cooling rates are achieved in this case, and the minimal temperature is also increased. The observable power broadening leads also to a larger parameter space where efficient cooling is observed.

Figure 4 shows the maximum cooling rate as a function of Ω , beyond the validity of the perturbative regime, which predicts that the cooling rate scales quadratically with Ω [20]. We observe that with increasing pump strengths, the cooling rate becomes larger and increases almost linearly with Ω . Correspondingly, the steady-state phonon number increases exponentially with Ω . Thus the choice of the pump strength is a trade-off between the final temperature and the fast cooling process.

IV. CONCLUSION

In this paper, we have presented numerical evidence that cavity-enhanced sideband cooling of trapped molecules is feasible by performing the calculations for a particular molecule, COS. The chosen example of COS has favorable properties, such as a large polarizability and a strong vibrational transition. We have identified experimental parameters for trapping and cooling the COS molecule, which may be realized in the near future. The use of infrared transition well-resolved sidebands may easily be achieved in different types of harmonic traps. As long as the strong-coupling regime can be implemented with a chosen vibrational transition, concurrent decay channels are outrun by the enhancement. Moreover, the scheme is well-suited for ion traps due to their long storage times and their low anharmonicities. Under these considerations, ground-state cooling on a submillisecond time scale seems possible. More efficient cooling could be achieved by increasing the cavity coupling g , which could be achieved by exploiting collective enhancement in a molecular gas as proposed in Ref. [26]. Further improvement of the cooling process is conceivable by adaptive control of the Rabi frequency in time.

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