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Optomechanical control of atoms and molecules

M. Bhattacharya, S. Singh, P.-L. Giscard, and P. Meystre
*B2 Institute, Department of Physics and College of Optical Sciences,
The University of Arizona, Tucson, Arizona 85721*

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In this article we present recent and ongoing developments in our group related to nanoscale optomechanics, an emerging area at the confluence of atomic, condensed matter and gravitational wave physics. The basic paradigm of optomechanics is the placement of a mechanical harmonic oscillator in its quantum ground state. First we discuss how the motion of such a macroscopic quantum oscillator can be squeezed, using the laser excitation of a high finesse optical cavity. Next we show how the placement of a ferroelectric tip on the oscillator allows it to manipulate polar molecules. Lastly we discuss how the role of the mechanical oscillator can be played by a quantum degenerate gas placed inside an optical cavity, giving rise to rich optomechanical physics.

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I. INTRODUCTION

Vladilen Letokhov's recent monograph "Laser Control of Atoms and Molecules" reviews some of the most important developments in atomic and molecular physics following the invention of the laser. He personally pioneered an astoundingly large fraction of these developments. His ground-breaking contributions are too numerous to list in any detail. They cover areas ranging from the laser control of atomic and molecular processes, including ionization and dissociation, to the detection of single atoms and molecules, from isotope separation to laser velocity selection and to laser trapping and cooling, and much more. According to the ISI Web of Knowledge he has published well over 700 papers. As just one illustration of his breadth of interests, in the last two years he has published work on astrophysical lasers, on scanning near-field optical microscopes, on the quantum theory of radiation by atoms placed near a microresonator, and on the excitation and dissociation of polyatomic molecules under the action of femtosecond laser pulses.

One of us (PM) first met Vladilen in the mid-1970s during one of his frequent visits to his close friend the late Peter Franken and to Marlan Scully at The University of Arizona. At the time, laser isotope separation was a major research topic and he was an undisputed leader in the field. Everybody wanted to learn his latest thoughts on the subject. We later met again at a number of occasions, sometimes in Europe, in particular at the Max-Planck Institute for Quantum Optics, or back in Tucson, where our interactions increased significantly following the break-up of the Soviet Union, and also in Moscow and in Troisk. It is therefore an honor and a pleasure to contribute an article to the Festschrift celebrating his seventieth birthday.

Returning to Dr. Letokhov's recent book, we remark that an important requirement to achieve further progress in the manipulation and control of atoms and molecules is the availability of neutral particle detectors that can operate at the single atom or single molecule level. With this in mind, this paper briefly reviews some

of our recent work toward that goal, concentrating on the emerging area of cavity optomechanics, a field that combines ideas of AMO physics, nanoscience, and condensed matter physics. It promises to open up new routes to the coherent control of atoms and molecules, relying on their coupling to nanomechanical oscillators laser-cooled close to their ground state of vibrations by techniques directly inspired from those originally developed and demonstrated in the laser cooling of neutral atoms and ions.

With the detailed understanding of mirror cooling gained in the last few years, we are now rapidly moving toward the longer-term goal of coupling nanoscale cantilevers to atomic or molecular samples. For instance the coupling of a cantilever carrying a magnetic domain with a Bose-Einstein condensate was recently described in Ref. [1]. In another example we have considered the coupling of a nanomechanical oscillator in the quantum regime with molecular (electric) dipoles [2]. We found theoretically that the cantilever can produce single-mode squeezing of the center-of-mass motion of an isolated trapped molecule and two-mode squeezing of the phonons of an array of molecules. This work opens up possibilities for manipulating "crystals" of ultracold dipolar molecules, which have been recently proposed as quantum memories [3]. More broadly, it indicates the promise of nanoscale cantilevers for the quantum detection and control of atomic and molecular systems.

II. CAVITY OPTOMECHANICS

A basic arrangement that can be used to understand optomechanics theory and experiment consists of a linear two-mirror optical cavity (2MC) driven by laser radiation tuned close to a resonance of the cavity. One of the cavity mirrors is movable, and the goal is to optically cool its vibrational motion to a point as close to its quantum mechanical ground state as possible. This procedure normally involves the use of two laser beams, the first one detuned above a cavity resonance so as to increase

the intrinsic oscillation frequency of the cantilever to a value ω_{eff} ; and the second one detuned below the cavity resonance, so as to (almost) independently increase the damping constant of the oscillating mirror from its field-free value Γ_m to Γ_{eff} [4]. Cooling of moving mirrors has now been achieved for a remarkably broad range of systems, from nanogram or picogram mirrors attached to nano-cantilevers to LIGO-class massive mirrors.

The laser fields change the properties of the moving mirror in two ways: they increase the energy level spacing of the harmonic mirror trap from $\hbar\omega_M$ to $\hbar\omega_{\text{eff}}$, and cool the mirror from its initial equilibrium temperature T_e to a lower value given by

$$T_{\text{eff}} = \left(\frac{\Gamma_m}{\Gamma_{\text{eff}}} \right) T_e. \quad (1)$$

The resultant trapping and cooling thus lowers the number of quanta of vibrational excitation of the oscillating mirror to

$$n_m = \frac{k_B T_{\text{eff}}}{\hbar\omega_{\text{eff}}} = \frac{k_B T_e}{\hbar\omega_{\text{eff}}} \left(\frac{\Gamma_m}{\Gamma_{\text{eff}}} \right), \quad (2)$$

where k_B is Boltzmann's constant. The goal is to achieve $n_M < 1$, i.e. to place the mirror in its quantum mechanical ground state.

In recent work we proposed an alternative to the “traditional” two-mirror geometry that allows one to reach and detect lower n_m 's for comparable parameters [5] by suspending a nearly perfectly reflecting mirror inside a high-finesse two-mirror cavity. This three-mirror cavity (3MC) arrangement was shown to possess at least three advantages. First, it provides a higher value of ω_{eff} for the mirror [6–9], leading to fewer quanta of excitation, see Eq. (2). Second it removes bistability problems completely for the trapping laser fields, and partially for the cooling fields. Lastly, it increases the time available for observing the quantum dynamics of the suspended mirror before thermal decoherence sets in. In addition, the coupling of the mirror to the intracavity field can be either linear (or *dissipative*) or quadratic (or *dispersive*) in the mirror position. This has several important implications, including the possibility to measure an energy eigenstate of the mirror and that of preparing nonclassical squeezed states of mirror motion.

An experiment that demonstrates the working of the 3MC and points out some of its additional virtues was recently reported in Ref. [10]. In that work a 50nm thick dielectric membrane placed inside a high-finesse optical cavity was cooled down from room temperature (294K) down to 6.82mK, i.e. by a factor of 4.4×10^4 .

III. SQUEEZING THE MIRROR MOTION

There is little doubt that the laser cooling of vibrating mirrors will soon be able to lower one or several modes of vibration to their quantum mechanical ground state

[5, 11–13]. In addition to metrological applications, this will represent an important first step in exploring characteristic features of quantum mechanics such as superposition [14] and entanglement in macroscopic systems [15]. This section discusses a possible way to realize a squeezed state of a mechanically moving mirror in a high finesse optical cavity.

Squeezed states of the harmonic oscillator have attracted much attention, due to their favorable quantum noise properties [17]. Squeezed states of light are expected to find applications in precision measurements [18] and optical communications [19, 20]. The squeezing of classical noise in mechanical oscillators has been demonstrated in optomechanical cavities [21], ion traps [22, 23] optical lattices [24] and other systems [25, 26]. Quantum squeezing of phonons has been achieved in ion traps [27] and in crystals [28]. Proposals to realize squeezed states of nanomechanical oscillators in the quantum regime have been made involving two-mirror cavities [29], parametric mixing in solid state circuits [30–32], microwave coupling to a charge qubit [33], quantum non-demolition measurements [34] and the parametric modulation of a mechanical spring [35, 36]. Their application to gravitational interferometry has also been discussed [37].

Previous proposals to achieve squeezing in optomechanical systems have relied on a mathematical analogy between an optical resonator with a moving mirror and light propagation in Kerr media, and the mechanism of squeezing has been *parametric driving*. Here we invoke *compression* as an alternative route to squeezing [16]. In that scheme squeezing of the mirror motion relies on coupling it dispersively with the cavity. More precisely, we consider a moving mirror coupled dispersively to an optical field and the modes and dissipatively to a second one, a configuration that was recently proposed as an efficient cooling and trapping configuration for semi-transparent mirrors [39].

This system is described by the model Hamiltonian [39]

$$H = H_f + H_m + V \quad (3)$$

where the free field Hamiltonian H_f is

$$H_f = \hbar\omega_D a^\dagger a + \hbar\omega_S b^\dagger b, \quad (4)$$

the oscillating middle mirror Hamiltonian H_m is

$$H_m = \frac{p^2}{2m} + \frac{1}{2}m\omega_m^2 q^2, \quad (5)$$

and the interaction Hamiltonian V is

$$V = \hbar\xi_D a^\dagger a q + \hbar\xi_S b^\dagger b q^2. \quad (6)$$

Here ω_m is the oscillation frequency of the middle mirror of transmittivity T and equilibrium position q_0 ,

$$|\xi_D| = \frac{\sin 2k_n q_0}{\sqrt{(1-T)^{-1} - \cos^2 2k_n q_0}} \xi, \quad (7)$$

$\omega_n = n\pi c/L$, $\xi = \omega_n/L$, $k_n = \omega_n/c$, and

$$|\xi_S| = \frac{\tau\xi^2}{2} \left(\frac{1-T}{T} \right)^{1/2}, \quad (8)$$

where $\tau = 2L/c$.

The frequencies of the optical fields can be chosen such that $\xi_{D,S}$ are either positive or negative. For ξ_S this corresponds to the use of trapping and anti-trapping modes, respectively [39]. For low values of ξ_S the spring potential energy dominates the anti-trapping due to radiation pressure, hence the middle mirror still behaves as a harmonic oscillator. However, for ξ_S negative, increasing $|\xi_S|$ leads to a point

$$C_S = -\omega_m/2, \quad (9)$$

where the mirror behaves as a free particle. For even higher values of $|\xi_S|$ radiation pressure-induced anti-trapping dominates and the mirror behaves like an inverted harmonic oscillator [40]. We do not consider that regime here.

In the following we consider a semiclassical version of the Hamiltonian H valid for situations where the optical fields can be treated classically. In that case

$$a \rightarrow \alpha, \quad b \rightarrow \beta. \quad (10)$$

Expressing the mirror displacement in terms of raising and lowering operators,

$$q = \sqrt{\frac{\hbar}{2m\omega_m}}(c^\dagger + c), \quad (11)$$

H becomes

$$H = \hbar C_D(c + c^\dagger) + 2\hbar C_R K_0 + \hbar C_S(K_- + K_+) \quad (12)$$

where we have removed a constant energy $E_0 = \omega_D|\alpha|^2 + \omega_S|\beta|^2$, and

$$\begin{aligned} C_D &= \frac{\xi_D|\alpha|^2}{\sqrt{2m\omega_m/\hbar}}, \\ C_S &= \frac{\hbar\xi_S|\beta|^2}{m\omega_m}, \\ C_R &= C_S + \omega_m. \end{aligned} \quad (13)$$

We have also introduced the operators

$$K_0 = (c^\dagger c + c c^\dagger)/4, \quad K_- = c^2/2, \quad K_+ = c^{\dagger 2}/2. \quad (14)$$

which together with c and c^\dagger form the basis of a Lie algebra [41] with

$$\begin{aligned} [K_0, K_\pm] &= \pm K_\pm, \quad [K_-, K_+] = 2K_0, \\ [K_-, c] &= [K_+, c^\dagger] = 0, \\ [K_-, c^\dagger] &= c, \quad [K_0, c^\dagger] = c^\dagger/2. \end{aligned} \quad (15)$$

The Hamiltonian (12) was previously studied in the context of molecular translational-vibrational interactions

[42], of laser-plasma scattering [43], and of atomic vapors inside resonators [44].

Using the Lie-algebraic symmetries of H , the associated evolution operator can be disentangled as [41]

$$U = \exp[-iHt/\hbar] = e^{i\delta} D(\nu) R(\phi) S(\kappa), \quad (16)$$

where δ is an unimportant overall phase. The operator

$$D(\nu) = e^{\nu c^\dagger - \nu^* c}, \quad (17)$$

is the displacement operator, with [41]

$$\nu = \frac{C_D}{\chi} \left[\frac{\omega_m}{\chi} (\cos \chi t - 1) - i \sin \chi t \right] \quad (18)$$

and

$$\chi = \sqrt{C_R^2 - C_S^2} = [\omega_m(\omega_m + 2C_S)]^{1/2}. \quad (19)$$

In the bound oscillator regime, i.e. for $C_S > -\omega_m/2$, we have $\chi^2 > 0$, and we can choose $\chi > 0$ without loss of generality. That parameter largely determines the time scale of the mirror dynamics; in the absence of squeezing ($C_S = 0$) it is just the harmonic oscillator period. The second operator,

$$R(\phi) = e^{i\phi K_0} \quad (20)$$

is a rotation operator, and the third operator,

$$S(\kappa) = e^{\kappa^* K_- - \kappa K_+} \quad (21)$$

is a squeezing operator, with

$$|\kappa| = \left| \sinh^{-1} \left(\frac{C_S}{\chi} \sin \chi t \right) \right|. \quad (22)$$

As expected that operator does not depend on C_D , i.e. displacement does not affect squeezing.

It turns out that the rotation angle ϕ in Eq. (20) is exactly opposite the angle at which the squeeze operator tilts the error ellipse of the moving mirror in phase space [45], i.e.

$$\phi = -\frac{1}{2} [\text{phase}(\kappa) + \pi]. \quad (23)$$

The two rotations therefore cancel each other out and ϕ effectively plays no role in the dynamics. It is in fact intuitively clear that the effects of rotation should cancel, i.e. the axes of the final error ellipse should be aligned along p and q in phase space. This is because Eq. (3) stipulates that position is the only quadrature of the oscillating middle mirror that can be squeezed or anti-squeezed, the latter situation corresponding to momentum squeezing.

Figure 1 shows $|\kappa|$ versus time for typical experimental parameters. The squeezing first grows linearly in time, as can be confirmed by analytically expanding Eq. (22) for the case of $t \ll 1/\chi$

$$|\kappa| \simeq \left| C_S \left[t - \frac{(\omega_m + C_S)^2}{6} t^3 \right] + \mathcal{O}[t^5] \right|. \quad (24)$$

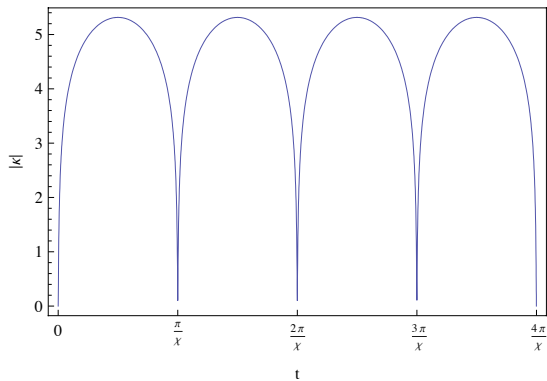


FIG. 1: Modulus of $|\kappa|$ from Eq. (22) as a function of time for $L = 5\text{mm}$, $\lambda = 514\text{nm}$, $m = 1\mu\text{g}$, $\omega_m = 2\pi \times 1\text{kHz}$, mechanical quality factor $Q = 5 \times 10^6$, end mirrors transmissivity $T = 10^{-4}$, $q_0 = \lambda/10$, background temperature $T_e = 100\text{mK}$, and incident power 10mW . The period of the oscillations is $2\pi/\chi \simeq 5\text{ms}$.

If the middle mirror is initially prepared in its quantum mechanical ground state, the squeezing operator (21) produces a squeezed vacuum [46]. From Fig. 1, the maximum value of κ is approximately 5, which implies a maximum squeezing of $R \sim e^{-5} \sim 0.007$, or $\text{Log}_{10}(0.007) \sim 22\text{dB}$ of squeezing.

Any realistic model should also include the damping of the mirror. We estimate this effect by including noise and damping in the Heisenberg equations of the mirror in a manner consistent with the fluctuation-dissipation theorem. Setting $\alpha = 0$ for simplicity and concentrating therefore solely on the squeezing part of the Hamiltonian H this leads to the quantum Langevin equations

$$\begin{aligned} \dot{q} &= p/m, \\ \dot{p} &= -m\chi^2 q - \frac{\Gamma_m}{m} p + \epsilon(t). \end{aligned} \quad (25)$$

Here $\Gamma_m = m\omega_m/Q$ is the damping constant of the mirror of mechanical quality factor Q and $\epsilon(t)$ represents brownian noise with average zero and fluctuations correlated as

$$\begin{aligned} \langle \delta\epsilon(t)\delta\epsilon(t') \rangle &= \\ \Gamma_m \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega(t-t')} \hbar\omega \left[1 + \coth\left(\frac{\hbar\omega}{2k_B T_e}\right) \right] \end{aligned} \quad (26)$$

Linearizing all operators in Eq. (25) as sums of a semiclassical steady-state value and small quantum fluctuations (i.e. $q = q_s + \delta q$) we obtain linear dynamical equations for the fluctuations. Following Ref. [47], we find the equal-time correlation function for the position

$$\langle \delta q^2 \rangle = (2n_T + 1) \frac{\hbar\omega_m}{2m\chi^2}, \quad (27)$$

which is independent of time since the noise process is stationary [Eq. (26)]. In the absence of squeezing ($C_S =$

FIG. 2: (Color online). Nanomechanical oscillator coupled to A) a single polar molecule B) a one-dimensional dipolar crystal made of many such molecules. In both cases the molecules are weakly confined by a harmonic trap along the x axis and oriented along the y axis by a strong uniform electric field.

0), and at high temperatures, ($n_T \sim k_B T_e / \hbar\omega_m \gg 1$),

$$\langle \delta q^2 \rangle = k_B T_e / m\omega_m^2 \quad (28)$$

while at low temperatures ($n_T \ll 1$),

$$\langle \delta q^2 \rangle = \hbar / 2m\omega_m, \quad (29)$$

which is just the square of the oscillator length of the ground state of the moving mirror.

IV. MOTIONAL SQUEEZING OF DIPOLAR MOLECULES

We mentioned already that ultracold mirrors and cantilevers operating close to or in the quantum regime provide a novel class of quantum sensors that may find applications in matter-wave optics and in new approaches to the coherent and quantum control of ultracold atomic and molecular samples. A broad perspective on the subject can be gained from related successes in atomic physics where laser cooling and trapping techniques have enabled impressive coherent control of microscopic systems. A particularly promising research direction at the interface between atomic and solid state physics, is the interaction of cold atomic systems with quantum nanomechanical oscillators. Proposals along these lines have included the coupling of cantilevers to cold ions [48], Bose-Einstein condensates [1], and cold atomic gases [49], etc. In this section we review some recent work on the dipole-dipole coupling between ultracold nanomechanical cantilevers with a ferroelectric tip and ultracold polar molecules [2].

Consider first the case of a single molecule, see Fig. 2. Its coupling with the cantilever is described by the Hamiltonian

$$H = H_c + H_{\text{mol}} + V_I, \quad (30)$$

where

$$H_c = \hbar\omega_c a^\dagger a \quad (31)$$

describes a vibrational mode of the cantilever of effective mass m_c and frequency ω_c ,

$$H_{\text{mol}} = \hbar\omega_t b^\dagger b, \quad (32)$$

accounts for the center-of-mass motion of the trapped molecular dipole of mass m oscillating at the trap frequency ω_t , and the cantilever-molecule interaction is

$$V_I = \frac{d_{\text{mol}} d_c}{4\pi\epsilon_0 r^3} \left[1 - \frac{3(R + y_c)^2}{r^2} \right]. \quad (33)$$

Here

$$y_c = \sqrt{\frac{\hbar}{2\omega_c m_c}}(a + a^\dagger), \quad (34)$$

d_c is the dipole moment of the ferroelectric domain situated a distance $r = [(R + y_c)^2 + x_{\text{mol}}^2]^{1/2}$, and R is the equilibrium molecule-cantilever separation. In practice $R \gg x_{\text{mol}}, y_c$ and in this case the dipolar interaction is approximately

$$V_I \approx \frac{d_{\text{mol}} d_c}{2\pi\epsilon_0 R^6} (-R^3 + 3y_c R^2 + 3x_{\text{mol}}^2 R - 15y_c x_{\text{mol}}^2) \quad (35)$$

with

$$x_{\text{mol}} = \sqrt{\frac{\hbar}{2\omega_t m}}(b + b^\dagger). \quad (36)$$

For small R the presence of the cantilever results in a tightening of the trap, with $\omega_t \rightarrow \omega'_t$. In addition, it produces a parametric squeezing of the molecular motion. This can be seen by transforming to an interaction picture with respect to the free Hamiltonian $H_c + H_{\text{mol}}$, choosing $\omega_c = 2\omega'_t$. Performing the rotating-wave approximation and assuming that the cantilever amplitude can be treated classically, $a \rightarrow \alpha$, the potential V_I reduces to

$$V_I = -\hbar C (b^2 + b^{\dagger 2}), \quad (37)$$

where

$$C = L_c \frac{15d_{\text{mol}} d_c}{4\pi\epsilon_0 m \omega'_t R^6}, \quad (38)$$

and L_c is the classical amplitude of oscillation of the cantilever given by

$$L_c = \sqrt{N} \left(\frac{\hbar}{2m_c \omega_c} \right)^{1/2}. \quad (39)$$

Equation (37) is the familiar quantum optics squeezing Hamiltonian of the degenerate parametric amplifier, see e.g. Ref. [58]. The squeezing resulting from the interaction in Eq. (37) is of course degraded by thermal noise, which affects the system in the form of phase fluctuations of the cantilever field. As in the previous section these fluctuations are related to the cantilever damping rate Γ_m by the fluctuation-dissipation theorem. For times t such that $\Gamma_m < t^{-1} < 2C$, the variance in the dimensionless molecule position quadrature is given by [58]

$$(\Delta x_1)_t^2 = \frac{1}{4} e^{-2u} + \frac{1}{8} e^{2u} \Gamma_m t, \quad (40)$$

where the squeezing parameter is $u = 2Ct$.

We now extend these considerations to a one-dimensional chain of N heteronuclear molecules contained in a harmonic potential V_t , see Fig. 2. A uniform polarizing electric field is once again applied along the

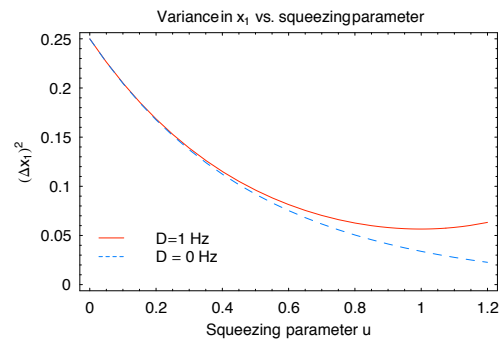


FIG. 3: Variance in the molecule position quadrature component x_1 as a function of the squeezing parameter u . The parameters are for a SrO molecule interacting with a cantilever. Dotted curve: single mode squeezing obtained in the absence of any thermal noise. Solid curve: effect of a cantilever damping rate $\Gamma_m = 1$ Hz. Motional squeezing of the molecular motion occurs for $(\Delta x_1)^2 < 1/4$, and is degraded by phase noise in the cantilever.

y -axis to align the dipoles. The dipole-dipole interaction being repulsive in that case, the molecules self-organize into a linear chain of period l described by the Hamiltonian

$$H_{\text{chain}} = \sum_i^N \frac{p_i^2}{2m} + \frac{d_{\text{mol}}^2}{4\pi\epsilon_0} \sum_{i < j}^N \frac{1}{|x_i - x_j|^3} + V_t, \quad (41)$$

where x_i, p_i are the position and momentum, of the i th molecule and V_t is the external trapping potential.

For small molecular oscillations the Hamiltonian (41) can be reexpressed in terms of acoustic phonon modes of momentum k and energy $\hbar\omega_k$ [59],

$$H_{\text{chain}} = \sum_k \hbar\omega_k b_k^\dagger b_k, \quad (42)$$

where b_k, b_k^\dagger are bosonic phonon annihilation and creation operators, $\omega_k = 2\omega_0 |\sin(kl/2)|$, and $\omega_0 = d_m (3/2\pi\epsilon_0 ml^5)^{1/2}$. Only purely harmonic terms have been retained in arriving at Eq. (42) from Eq. (41). Higher order terms represent phonon-phonon interactions and in particular determine the lifetime of the phonons in the crystal [60].

As in the case of a single molecule, a zero-temperature description is adequate in the regime $k_B T \ll \hbar\omega_0$. The nanomechanical cantilever energy is still correctly described by Eq. (31), and the interaction between the chain of molecules and the oscillator is given by

$$V_I = \sum_i \frac{d_{\text{mol}} d_c}{4\pi\epsilon_0 r_i^3} \left[1 - \frac{3(R + y_c)^2}{r_i^2} \right]. \quad (43)$$

Here y_c is the displacement of the cantilever along the y axis, R is now its distance from the center of the dipolar crystal and $r_i = [(R + y_c)^2 + (il + x_i)^2]^{1/2}$ is its distance from the i -th molecule.

Typically $x_i \ll l$, $Nl \ll R$, and we use these relations to keep only the lowest order terms in Eq. (43). As was the case for a single molecule, they result in a slight frequency shift $\omega_k \rightarrow \omega'_k$ – now in the phonon frequency – and to a squeezing interaction, now in the form of a two-mode squeezing Hamiltonian given in the interaction picture by

$$V_I = - \sum_k \hbar C'_k (a + a^\dagger) \left(b_k b_{-k} + b_k^\dagger b_{-k}^\dagger + b_k^\dagger b_k + b_{-k} b_{-k}^\dagger \right) \quad (44)$$

where

$$C'_k = - \frac{17d_m d_c}{16\pi\epsilon_o m \omega'_k R^6} \left(\frac{\hbar}{2m_c \omega_c} \right)^{1/2}. \quad (45)$$

We select for concreteness the cantilever frequency $\omega_c = 2\omega'_k$, with $k = \pi/l$, so that it couples mainly to excitations near the edge of the first Brillouin zone, where the density of phonon states is largest. Assuming as in the single-molecule case that the motion of the nanomechanical cantilever can be described classically, in the rotating wave approximation we then obtain the two-mode squeezing hamiltonian

$$V_I = -\hbar C_k \left(b_k b_{-k} + b_{-k}^\dagger b_k^\dagger \right). \quad (46)$$

where $C_k = \sqrt{\bar{N}} C'_k$ and \bar{N} is the average occupation number of the cantilever, related to the classical amplitude of oscillations L_c by Eq. (39).

Two-mode squeezing is conveniently described in terms of the two dimensionless quadratures

$$s_1 = \frac{1}{\sqrt{2}} (b_k + b_{-k} + b_k^\dagger + b_{-k}^\dagger) \quad (47)$$

$$s_2 = \frac{1}{\sqrt{2}i} (b_k - b_k^\dagger - b_{-k} + b_{-k}^\dagger). \quad (48)$$

The sum of variances in the two quadratures, taking into account the phase fluctuations in the cantilever motion resulting from thermal noise, is then [61]:

$$\begin{aligned} (\Delta s_1)^2 + (\Delta s_2)^2 = & \frac{e^{-\frac{\Gamma_m t}{2}}}{C_{k0}} \{ \Gamma_m \sinh(C_{k0} t) + 2C_{k0} \cosh(C_{k0} t) \} \\ & - \sum_{i,j,k,i \neq j, j \neq k} e^{\lambda_i t} \frac{2C_{k0}(\lambda_i + 4\Gamma_m)}{(\lambda_i - \lambda_j)(\lambda_i - \lambda_k)}, \end{aligned} \quad (49)$$

where u is the phonon squeezing parameter and is equal to $2C_{k0}t$, with $C_{k0} = \frac{1}{2}\sqrt{4C_k^2 - \Gamma_m^2}$ and the λ_i 's are the roots of the cubic equation:

$$\lambda^3 + 5\Gamma_m \lambda^2 + (4\Gamma_m^2 - C_{k0}^2)\lambda - 2C_{k0}^2 \Gamma_m = 0 \quad (50)$$

Consider for example a nanomechanical cantilever with frequency $\omega_c = 2$ MHz, effective mass $m_c = 10^{-16}$ kg, linewidth $\Gamma_m = 1$ Hz, average thermal occupation

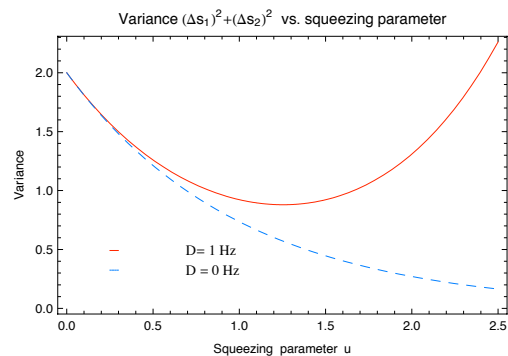


FIG. 4: Sum of the variances in s_1 and s_2 [see Eqs. (47-49)], as a function of the squeezing parameter u , for a dipolar crystal of SrO molecules interacting with a cantilever. Dotted curve: two-mode squeezing in the absence of thermal noise. Solid curve: effect of thermal noise producing a cantilever damping rate $\Gamma_m = 1$ Hz. Two-mode squeezing and phonon entanglement occurs for a variance less than 2.

$\bar{N}=100$, and a ferroelectric domain with dipole moment $d_c = 2.1 \times 10^{-23}$ C-m attached to the cantilever and placed at $R = 2\mu\text{m}$ from a linear chain of dipolar molecules with inter-molecular distance $l \approx 200$ nm. These parameters give a phonon frequency $\omega_o = 4$ MHz, and thus an interaction $C_k = 4.4$ Hz. Figure 4 gives the sum of variances of s_1 and s_2 as a function of the squeezing parameter.

V. CANTILEVER-BASED COHERENT CONTROL

Building upon these results, we now show that it is possible to use nanoscale cantilevers to engineer nonclassical center-of-mass states of an atomic or molecular system. These results are preliminary and involve two dipolar molecules only. An extension to larger systems will be the object of a future publication.

The Hamiltonian describing the coupling of the cantilever to the two molecules is

$$H = H_c + H_m + V_{dd}, \quad (51)$$

where as before

$$H_c = \hbar\omega_c c^\dagger c, \quad (52)$$

describes a single mode of vibration of the cantilever.

We assume that the two molecules are trapped tightly in the y and z -direction, such that they align themselves in the x -direction. As before, we also assume that a strong polarizing DC field freezes out the rotational freedom of the dipoles so that they align along the y -axis.

The vibrations of the two-molecule system are conveniently described in terms of their center-of-mass (COM) and relative modes of motion (see inset in Fig. 5). Ignoring the trap potential, the two-molecule Hamiltonian is

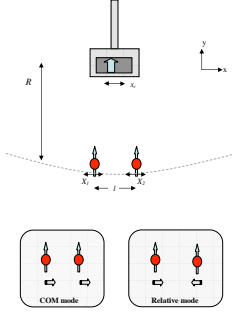


FIG. 5: (Color online). Schematic of the coupling a nanomechanical oscillator to two dipolar molecules. The molecules are along x -axis and interact with both the cantilever and with each other via dipole-dipole interaction. A weakly confining harmonic trap for the dipoles is shown along the x axis. The two insets show the center-of-mass and relative modes of vibration of the molecular system.

then

$$H_m = \frac{p_{\text{com}}^2}{4m} + \frac{p_{\text{rel}}^2}{m} + \frac{6d_m^2}{\pi\epsilon_0 l^5} x_{\text{rel}}^2. \quad (53)$$

where p_{com} and p_{rel} are the center-of-mass and relative momenta, and the third term in the Hamiltonian describes the dipole-dipole interaction between molecules in the harmonic approximation. Here m is the mass of each molecule, d_m its electric dipole moment, and l their equilibrium separation.

For $x_1, x_2 \ll l \ll R$ the dipole-dipole interaction between the ferroelectric domain on the cantilever of dipole moment d_c and the molecules is given approximately by

$$V_{dd} = \frac{d_m d_c}{8\pi\epsilon_0 R^7} \left(24R^2(x_{\text{com}}^2 + x_{\text{rel}}^2 + x_c^2) - 48R^2 x_c x_{\text{com}} - 140l x_{\text{rel}}^3 + 60l x_{\text{rel}} x_{\text{com}}^2 - 60l x_{\text{rel}} x_c^2 + 120l x_c x_{\text{rel}} x_{\text{com}} \right), \quad (54)$$

where

$$x_c = \sqrt{\frac{\hbar}{2\omega_c m_c}} (c + c^\dagger). \quad (55)$$

The design of appropriate fields for the internal control of systems of interacting atoms and molecules on their natural timescales is a difficult task. A ground-breaking approach using Genetic Algorithms and a pulse-shaping

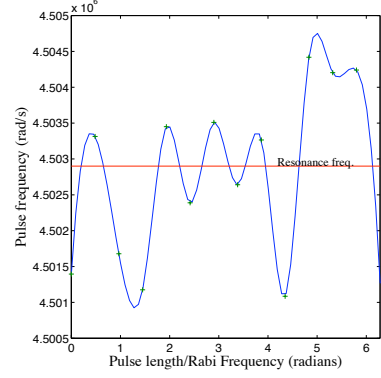


FIG. 6: (Color online) Frequency $\delta\omega(t)$ corresponding to the final generation of the genetic algorithm. That pulse shape results in an average occupation number of 0.98 in the COM mode, and 1.6 in the relative mode.

element inside a feedback loop to let atoms design their own optimal field was first proposed by Rabitz and Judson as early as 1992 [50]. In the present example we apply similar ideas to the center-of-mass motion of the pair of molecules. We proceed by modulating the frequency of oscillations of the cantilever so as to achieve specific populations of the center-of-mass and relative modes of vibration of the molecular system. We proceed by decomposing that frequency into a constant term and a time-dependent part to be determined iteratively via a genetic algorithm,

$$\omega_c = \omega_0 + \delta\omega(t). \quad (56)$$

In an interaction picture with respect to the molecular Hamiltonian and the ω_0 part of the cantilever Hamiltonian and assuming once more that the cantilever motion can be treated classically the interaction potential V_{cm} reduces in the rotating wave approximation to

$$V_{dd} = \frac{15\hbar L_c}{2\pi\epsilon_0 \sqrt{\omega_{\text{com}} \omega_{\text{rel}}}} \frac{d_m d_c l}{m R^7} (ab^\dagger e^{i(\omega_{\text{rel}} - \omega_{\text{com}} - \omega_c)t} + \text{h.c.}), \quad (57)$$

where ω_{com} and ω_{rel} have been redefined to include their shifts due to the presence of the cantilever. Here a and a^\dagger are the annihilation and creation operators of the COM mode, and b and b^\dagger are the annihilation and creation operators of the relative mode of vibration. The distances R and l are defined in Fig. 5, and L_c is defined in Eq. (39). We have also assumed that the dc cantilever frequency, ω_0 is close to $\omega_{\text{rel}} - \omega_{\text{com}}$.

Equation (57) indicates that the interaction between the molecules and the cantilever enables the transfer of population from one vibrational mode to the other. By changing the frequency of the cantilever in an iterative fashion determined by a genetic algorithm, it is then possible to drive any desired population transfer.

Figure 6 illustrates the design of a pulse that can be used to cool one of the normal modes of the molecular

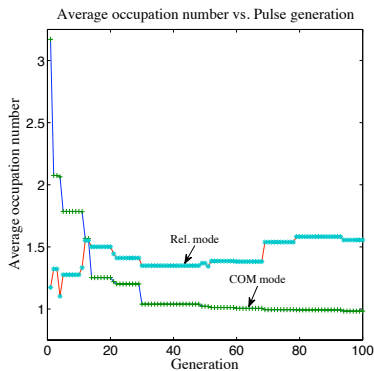


FIG. 7: (Color online) Average occupation number of the COM and relative modes as functions of the number of generations of the genetic algorithm.

system. In this example two SrO molecules are trapped 200nm apart from each other, and $2\mu\text{m}$ from the cantilever the specifications for which are the same as in the previous section. The center-of-mass and relative modes of the two-molecule system are both initially in a thermal state at a temperature of $100\ \mu\text{K}$. The specific time dependence of the cantilever frequency is chosen to minimize the average occupation number in the COM mode. The average occupation number for each genetic algorithm iteration is plotted in Fig. 7. We remark that in this example the total center-of-mass energy of the molecular system is decreased at the end of the pulse produced by the last generation of the algorithm (as seen in Fig. 8), and hence the method can demonstrably cool the molecular system.

VI. OUTLOOK: CONDENSATES IN HIGH- Q CAVITIES

As we have seen, a central tenet of cavity optomechanics is the cooling of mechanical oscillators close to their ground state of vibration, and we have shown in a few examples that these systems have fascinating potential applications in basic and applied physics. In recent work, the groups of Esslinger [51] in Zurich and of Stamper-Kurn at Berkeley [52] have proposed and demonstrated an alternative approach to cavity optomechanics that uses Bose condensates instead of “traditional” mechanical oscillators. In these systems, the optical length of the cavity is controlled and modified via the dispersive properties of the condensate, just as in usual optical bistability. In contrast to the moving mirrors and membranes discussed in this paper, the mechanical oscillator is no longer based on the presence of an external spring. Rather, it is provided by condensate density excitations resulting from photon recoil, a situation closely related to CARL amplification [53–56].

Esslinger *et al* have shown that in the simplest case there is a one-to-one mathematical correspondence be-

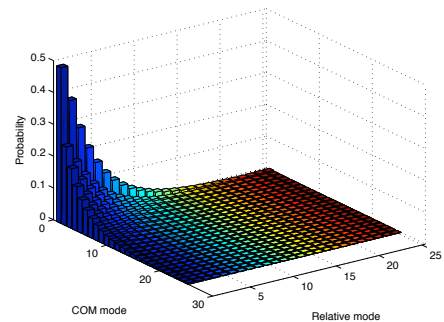
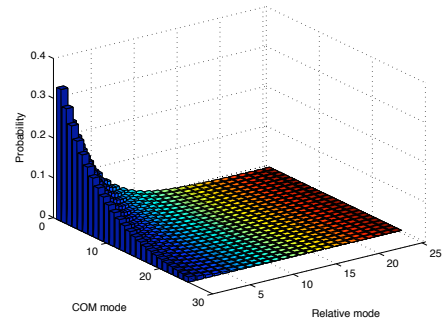


FIG. 8: (Color online) Excitation probability $P(n_{\text{rel}}, n_{\text{com}})$ of the molecule pair (a) initially (upper figure) and after the final iteration of the genetic algorithm (lower picture).

tween this system and the moving mirror situation. This can be seen simply by expanding the order parameter of the condensate as

$$\psi(x, t) = c_0(t) + \sqrt{2}c_2(t) \cos(2kx), \quad (58)$$

where the second term on the right-hand side accounts for the photon recoil of the atoms due to virtual transitions involving the absorption and reemission of cavity photons of momentum k . Assuming that the light-atom interaction does not significantly deplete the initial condensate one can replace the operator c_0 by a complex number, in which case the Hamiltonian describing the interaction between the atoms and the intracavity field reduces approximately to

$$H = \hbar \left[\Delta + g(c_2 + c_2^\dagger) \right] a^\dagger a, \quad (59)$$

where a and a^\dagger are the annihilation and creation operators of the optical field, g is a coupling constant, and Δ is the detuning between the frequency of the intracavity optical field and the cavity resonant frequency.

At this point, the analogy between this system and the case of a cavity with a suspended mirror is fully apparent:

The condensate side-mode is the analog of the moving mirror, and the intracavity field acts on this side mode amplitude exactly in the same way radiation pressure acts on the moving mirror.

Several experiments exploiting that analogy have already been performed, demonstrating in particular the appearance of optical bistability in these systems. But it is known from general principles that generically, bistability ceases to exist as systems become truly microscopic, due to quantum tunneling between the coexisting metastable state and true ground state that are responsible for the bistable behavior. In classical systems, this tunneling time is typically exceedingly long, certainly much longer than the duration of typical experiments. This results in the observed bistability. But as the systems become more microscopic this tunneling time decreases, leading to a behavior that, in the open systems at hand, is somewhat reminiscent of the Maxwell construction familiar from statistical physics. It will be interesting to study this transition by reducing the size of the initial condensate to a point where its approximate classical description ceases to be valid and quantum fluctuations dominate the system.

It should also be possible to study quantum phase transitions in ultracold atomic systems in cavity optomechanical environments. For example, it is known that radiation pressure induced bistability can occur in these sys-

tems. (This closely related to the more familiar bistability that occurs when a Kerr nonlinear medium is confined inside a Fabry-Pérot resonator. The difference is that instead of an intensity-dependent index of refraction, we are now in a situation where it is the true, physical length of the resonator that is changed when the light intensity inside the resonator is varied.) Radiation pressure induced optical bistability is generally considered as detrimental when trying to cool nanoscale cantilevers. However it can also result, at least in principle, in fascinating new effects such as a bistable quantum phase transition between a Mott insulator state and a superfluid state of the atoms [62].

These examples, and numerous other fascinating aspects of cavity optomechanics and of the optomechanical control of atomic and molecular systems, promise to lead to exciting developments in future years, further contributing to the merging of AMO physics, nanoscience and condensed matter physics.

Acknowledgments

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