We investigate the coupling of a nanomechanical oscillator in the quantum regime with molecular (electric) dipoles. We find 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 the possibility of manipulating dipolar crystals, which have been recently proposed as quantum memory, and more generally, is indicative of the promise of nanoscale cantilevers for the quantum detection and control of atomic and molecular systems.

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Recent experimental advances have brought macroscopic oscillators closer than ever before to operating in the quantum regime [1–6]. Technically progress has been enabled by improvements in nanofabrication and nonequilibrium cooling. Foundational interest in this frontier lies in the fact that quantum mechanics has never been tested at such a macroscopic scale, particularly with respect to counterintuitive effects such as superposition and entanglement. From a practical point of view it is important to explore the behavior of mechanical oscillators in the quantum regime since they serve as sensors whose precision is fundamentally restricted by quantum mechanics [7,8]. As part of an ongoing merger between atomic and condensed matter physics, it has become realistic to explore the interaction of cold atomic systems with quantum nanomechanical oscillators. Examples include the coupling of cold ions to vibrating electrodes [9], of a nanomechanical cantilever to a Bose-Einstein condensate [10], of an atomic vapor to an oscillating mirror [11], etc.

As a first step in this direction, this Letter investigates the coupling of a laser-cooled nanomechanical cantilever to an ultracold lattice of polar molecules. Because of the anisotropic, long-range interaction between these molecules, ensembles of ultracold polar molecules are believed to have a rich phase diagram and are the subject of intense theoretical and experimental interest [12]. The cantilever-molecule coupling is assumed to be enabled by a ferroelectric domain mounted on the former leading to a strong dipole-dipole interaction, and a strong polarizing dc field (of up to 100 kV/cm) freezes out the rotational freedom of the dipoles [13].

To set the stage for the discussion we consider first the simple case of a single molecule, see setup A of Fig. 1, and demonstrate that its coupling to the cantilever leads to the parametric squeezing of its center-of-mass motion. These considerations are then generalized to the situation of a linear chain of electric dipoles (setup B). Such a self-arranged crystal has recently been proposed as memory for quantum information processing [14]. We find that for an appropriate choice of cantilever frequency, the phonons in the crystal can be two-mode squeezed, i.e., entangled [15,16], hinting at the possibility of exploiting such a setup for the coherent control of the quantum state of the dipolar lattice.

The Hamiltonian describing the coupling of the cantilever to a single molecule is $H = H_c + H_m + V_I$, where

$$H_c = \hbar \omega_c (a^\dagger a + \frac{1}{2}),$$

(1)
describes the quantized (single mode of) vibration of the cantilever of effective mass $m_c$ at frequency $\omega_c$, $a$ and $a^\dagger$ being bosonic annihilation and creation operators obeying the commutation rules $[a, a^\dagger] = 1$. In terms of the displacement $y_c$ of the cantilever along the $y$ axis, we have

FIG. 1 (color online). Arrangement considered for coupling a nanomechanical oscillator to a dipolar “crystal.” In setup A, a single molecule is coupled to the oscillator. In setup B, the oscillator is again at a distance $R$ from the linear chain of molecules. A weakly confining harmonic trap for the dipoles is shown along the $x$ axis.
The Hamiltonian
\[ H_m = \hbar \omega_m (b^\dagger b + \frac{1}{2}) \]
describes the center-of-mass motion of the trapped dipole of mass \( m \), where \( \omega_m \) is the trap frequency and \( b, b^\dagger \) are bosonic annihilation and creation operators with
\[ x_m = \sqrt{\frac{\hbar}{2\omega_m m}} (b + b^\dagger) \]
x\(_m\) being the displacement of the molecule along the \( x \) axis. Finally, the interaction between the molecule and the oscillator is given by
\[ V_I = \frac{d_m d_c}{4\pi \epsilon_0 R^6} \left[ 1 - \frac{3(R + y_c)^2}{r^2} \right] \]
for the dipole interaction can be approximated as
\[ V_I = \frac{d_m d_c}{2\pi \epsilon_0 R^6} (-R^3 + 3y_c R^2 + 3x_m^2 R - 15y_c x_m^2) \]
The typical trap level spacing is much larger than the thermal energy of the ultracold molecule, which justifies its zero-temperature description. We will be including thermal effects for the cantilever below.

The presence of the cantilever has two major effects on the molecule dynamics. First, it leads to a tightening of the trap for small distances \( R \), resulting in a shifted trapping frequency
\[ \omega'_I = \left[ \omega^2 + \frac{3d_m d_c}{\pi \epsilon_0 m R^5} \right]^{1/2} \]
The second, more interesting effect is parametric squeezing. In an interaction picture with respect to the free Hamiltonian \( H_c + H_m \), taking \( \omega_c = 2\omega'_I \), performing the rotating-wave approximation, and further assuming that the cantilever motion can be treated classically (operator \( a \mapsto \) complex number \( a \)), the interaction potential \( V_I \) reduces to
\[ V_I = -\hbar C (b^2 + b^12) \]
where
\[ C = \frac{L_c}{4\pi \epsilon_0 m \omega_c R^6} \]
Here, \( L_c \) is the amplitude of oscillation of the classical cantilever. For a temperature \( T_c \), and a mean number of excitations \( \bar{N} = k_B T_c / \hbar \omega_c \) (\( k_B \) being the Boltzmann constant), we have
\[ L_c = \sqrt{\bar{N}} \left( \frac{\hbar}{2m_c \omega_c} \right)^{1/2} \]
Equation (8) is the standard squeezing Hamiltonian familiar from studies of the degenerate parametric amplifier in quantum optics, see, e.g., Ref. [17].

Thermal noise can simply be introduced in the description of the system in the form of phase fluctuations in the cantilever field. These fluctuations are related to the cantilever damping rate, \( D \) by the fluctuation-dissipation theorem. For times \( t \) such that \( D < t^{-1} < 2C \), the variance in the dimensionless position quadrature for the molecule, \( x_1 = \frac{1}{2}(b + b^\dagger) \), is then given by [17]
\[ (\Delta x_1)^2 = \frac{1}{4} e^{-2u} + \frac{1}{8} e^{2u} Dt \]
where \( u = 2Ct \) is the squeezing parameter.

Consider, for example, a nanomechanical cantilever with frequency \( \omega_c = 4 \text{ MHz} \), effective mass \( m_c = 10^{-16} \text{ kg} \), and linewidth \( D = 1 \text{ Hz} \). A ferroelectric domain with dipole moment \( d_c = 2.1 \times 10^{-23} \text{ C m} \) is attached to the cantilever and is placed at \( R = 2 \mu \text{m} \) from a SrO molecule. These parameters give an oscillator frequency \( \omega'_c = 2 \text{ MHz} \). We assume \( \bar{N} = 100 \), yielding \( C = 20.4 \text{ Hz} \). Figure 2 shows the variance in \( x_1 \) as a function of the squeezing parameter for that example. We remark that the squeezing in single trapped ions [18] and atoms [19] has been experimentally demonstrated, and similar measurement techniques can possibly be implemented to detect squeezing in the present case.

We now extend these considerations to the case of a lattice of \( N \) heteronuclear molecules contained in a harmonic trap \( V_I \), as shown in Fig. 1(b). The trap is arranged so as to confine the sample weakly along \( x \), tightly along \( y \) and \( z \), so that \( x_1 \) is a function of the squeezing parameter for that example. We remark that the squeezing in single trapped ions [18] and atoms [19] has been experimentally demonstrated, and similar measurement techniques can possibly be implemented to detect squeezing in the present case.

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dimensional chain in the following.) A polarizing dc electric field is provided along $y$ so that all the dipoles align along that direction. The system can be described by the Hamiltonian

$$H_p = \sum_{i}^{N} \frac{p_i^2}{2m} + \frac{d_m^2}{4\pi\epsilon_r} \sum_{i,j}^{N} \frac{1}{|x_i - x_j|^3} + V_i,$$  \hspace{1cm} (12)$$

where $x_i, p_i$ are the position and momentum, of the $i$th molecule and $V_i$ is the external trapping potential. The first term in Eq. (12) corresponds to the kinetic energy of the dipoles, the second to their dipole-dipole interaction and the last term denotes the trap energy.

Because of their mutual repulsion along the $x$ direction the molecules self-organize into a linear lattice [14]. For small oscillations of the molecules about their equilibrium positions Eq. (12) can be expressed in terms of acoustic phonon modes of momentum $k$ and energy $\hbar\omega_k$,

$$H_p = \sum_{k} k \hbar \omega_k (b_k^\dagger b_k + \frac{1}{2}),$$ \hspace{1cm} (13)$$

where $b_k, b_k^\dagger$ are the phonon annihilation and creation operators obeying the bosonic commutation rules $[b_k, b_k^\dagger] = \delta_{kk'}$, the phonon frequencies are given by $\omega_k = 2\omega_0 \sin(kl/2)$, where $\omega_0 = d_m(3/2\pi\epsilon_r m^3)^{1/2}$, and $l$ is the lattice spacing. We note that only terms harmonic in the $x_i$ have been retained in deriving Eq. (13) from Eq. (12). Higher, anharmonic terms represent phonon-phonon interactions and, in particular, determine the phonon lifetime in the crystal [20].

We consider ultracold molecules at a temperature $T$ such that $k_B T \ll \hbar\omega_0$, so that a $T = 0$ description is appropriate as before for the molecules. The energy of the nanomechanical cantilever is again given by Eq. (1), so that the coupling of the cantilever to the phonons is given by

$$V_I = -\sum_{i} \frac{d_m d_c}{4\pi\epsilon_r r_i^3} \left(1 - \frac{3(R + y_i)^2}{r_i^2}\right),$$ \hspace{1cm} (14)$$

where $d_c$ is the dipole moment of the ferroelectric domain attached to the tip of the cantilever. Here $y_i$ is the displacement of the cantilever along the $y$ axis, $R$ is its distance from the center of the dipolar crystal, and its distance from the $i$th molecule is given by $r_i = [(R + y_i)^2 + (i + x_i)^2]^{1/2}$. Exploiting the hierarchy of length scales, $x_i \ll l$, $Nl \ll R$, we expand Eq. (14) to find that the oscillator produces a slight shift in the phonon frequency,

$$\omega'_k = \left[\omega_k + \frac{3d_m d_c}{\pi\epsilon_r m R^5}\right]^{1/2},$$ \hspace{1cm} (15)$$

and the coupling of the cantilever to the phonons is given by

$$V_I = -\sum_{k} \hbar C'_k (a + a^\dagger)(b_k b_{-k} + b_k^\dagger b_{-k}^\dagger + b_k^\dagger b_k + b_{-k} b_{-k}^\dagger).$$ \hspace{1cm} (16)$$

We remark that for sufficiently small $R$ and/or sufficiently large $d_c$ the cantilever will couple to the individual dipoles rather than collectively to the acoustic phonons.

Following an approach that parallels the single-molecule description we work in an interaction picture with respect to the free Hamiltonian of the cantilever and the (frequency shifted) phonon mode. We further choose the cantilever frequency such that $\omega_k = 2\omega_0'$, with $k = \pi/l$, implying that the cantilever couples primarily to excitations near the edge of the first Brillouin zone, where the density of phonon states is largest. We assume for simplicity that the motion of the nanomechanical cantilever can be described classically just as in the single-molecule case (again, operator $a \to$ complex number $\alpha$), a reasonable approximation since it is still challenging to cool these systems to their quantum regime. Performing the rotating-wave approximation we then obtain the approximate interaction picture Hamiltonian

$$V_I = -\hbar C_k (b_k b_{-k} + b_k^\dagger b_{-k}^\dagger),$$ \hspace{1cm} (18)$$

where $C_k = \sqrt{N} C'_k$ and $\bar{N}$ is the average occupation number of the cantilever. This Hamiltonian is known from quantum optics to lead to the generation of two-mode squeezing between acoustic phonon modes of momenta $\pm k$ (within the bandwidth of the nanomechanical resonance), and hence their quantum entanglement.

In order to characterize two-mode squeezing we introduce the two dimensionless quadratures as follows:

$$s_1 = \frac{1}{\sqrt{2}}(b_k + b_{-k} + b_k^\dagger + b_{-k}^\dagger),$$ \hspace{1cm} (19)$$

$$s_2 = \frac{1}{\sqrt{2}i}(b_k - b_k^\dagger - b_{-k} + b_{-k}^\dagger).$$ \hspace{1cm} (20)$$

Taking into account the phase fluctuations in the cantilever motion resulting from thermal noise, the sum of variances in the two quadratures is then [21]:

$$(\Delta s_1)^2 + (\Delta s_2)^2 = e^{-D_{s1}/2C_{s0}}\{D\sinh(C_{s0} t) + 2C_{s0}\cosh(C_{s0} t)\}$$

$$-\sum_{ijk} e^{\lambda_i}\frac{2C_{s0}(\lambda_i + 4D)}{(\lambda_i - \lambda_j)(\lambda_i - \lambda_k)},$$ \hspace{1cm} (21)$$

where $u$ is the squeezing parameter and is equal to $2C_{s0}$, with $C_{s0} = \frac{1}{2}\sqrt{4C_k^2 - D^2}$ and the $\lambda_i$’s are the roots of the cubic equation:

$$\lambda^3 + 5D\lambda^2 + (4D^2 - C_k^2)\lambda - 2C_k^2D = 0. \hspace{1cm} (22)$$

For example, let us consider a nanomechanical cantilever with frequency $\omega_c = 2$ MHz, effective mass $m_e = 10^{-16}$ kg, and linewidth $D = 1$ Hz. We assume an average occupation number of 100. A ferroelectric domain with
In conclusion, we studied the coupling of nanomechanical cantilevers to dipolar molecules. We found that for a single trapped molecule, the presence of the cantilever leads to tighter confinement and parametric squeezing. We also demonstrated squeezing and entanglement of the phonon modes of a linear chain of dipolar molecules. These results open up the way to extremely promising novel methods for the quantum manipulation and control of the state of ultracold dipolar systems, and are indicative of the general use of nanoscale cantilevers in the detection and control of atomic and molecular systems.

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