Collective transverse cavity cooling of a dense molecular beam

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Abstract. Based on a classical point particle description, we study light induced transverse collimation and phase space compression of a fast molecular beam traversing the crossover region of a high-$Q$ optical cavity and a standing-wave laser beam. The molecules scatter pump laser light, far detuned from any molecular transition, into the resonant cavity mode. We show that despite a very small probability for this process for a single particle, collective enhancement can lead to significant transverse cooling and collimation above the self-organization threshold. The analytical formulae for this threshold derived from homogeneous one-dimensional (1D) models give surprisingly good estimates for the pulsed 3D case. We compare ring and standing wave cavity geometries and show that phase space compression can be achieved even for extremely small particle field coupling if compensated by a large density and pump power. In the vicinity of the critical point the compression time can be reduced by tailored seeding of the cavity mode or using a second buildup cavity for the pump field.

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1. Introduction

Laser cooling has proven to be a tremendously successful tool to prepare and manipulate ultracold gaseous ensembles of many atomic species [24]. Unfortunately, standard methods like Doppler cooling [18] or refined sub-Doppler methods [5, 40] cannot be applied directly to molecules. Here unwanted optical pumping to the large manifolds of vibrational and rotational excitations hampers implementation of fast cyclic transitions needed to convey a significant dissipative light force [19]. To circumvent this problem, alternative and more complicated routes towards producing cold and dense molecular samples with lasers have been proposed [2] but to our knowledge not implemented yet. Nevertheless, in a few special cases, it was possible to first create a cold atomic ensemble and then convert this to binary molecules without heating [6, 29, 39]. While this allowed extremely low temperatures, the possible molecular species as well as particle numbers are quite restricted. In a more general but still quite complex approach, buffer-gas cooling employing e.g. liquid helium was applied to significantly cool down a molecular gas [12]. Slow molecules can also be obtained by Stark deceleration [3] or filtering of molecular beams [31], although only limited cooling and phase space density enhancement can be obtained in this way. Hence the search for a generally applicable method is still on and many routes are being pursued.

As a major hint that at the end lasers and light forces could play a vital role in this quest, it has been demonstrated that strong optical forces can be applied to molecules without destruction and internal heating, if very far detuned but intense light fields are used [13, 28, 33, 38]. Of course such optical potentials created in free space do not lead to cooling or phase space compression, but simply allow manipulation and trapping. In practice, the trap densities that can be achieved are, however, far too low to allow for evaporative or sympathetic cooling in such a trap. As an interesting alternative it was noted a decade ago that light induced dipole forces can become dissipative and useful for phase space compression, if the light fields are enclosed in optical resonators [10, 19, 36, 37]. As the fields are then dynamically modified depending on the particle positions, they can extract energy from the particle motion [8]. Subsequently, cavity decay provides for dissipative energy loss from the field and acts as an entropy and energy
sink for the cooling [15]. Using cold atoms the general cavity cooling effect has already been impressively demonstrated in several cases [17, 21]. Unfortunately, it turns out that in order to achieve significant effects on individual particles, strong particle-field coupling in the sense of cavity quantum electrodynamics (QED) is needed, which requires small resonator volumes or large dipole moments [14]. The required strong coupling nevertheless seems hard to achieve with molecules at sufficiently large detuning.

In some pioneering experiments, however, Vuletic and co-workers [4] were able to show that using a modified geometry these requirements can be substantially lowered by using collectively enhanced light scattering. Their findings fitted very well with theoretical predictions of self-organization and superradiant cavity cooling [9] and even worked surprisingly well at large detunings, where spontaneous emission can be neglected [19]. As a central finding one thus sees that increasing the number of particles lowers the required coupling strength, cavity $Q$ parameter and pump laser intensity to reach the threshold for collectively enhanced cooling [1].

Despite these strongly reduced technical requirements, it is still a challenge to achieve the required initial conditions for molecular gases. Molecules are hard to confine for sufficiently long times and with sufficient density within a small cavity volume as a starting point for such collective cavity cooling. Actually several important steps in this direction have been experimentally achieved in a wide range of setups from Stark decelerators [32], electrostatic traps [30] to pulsed dipole forces [20]. Nevertheless the phase space densities are far from a MOT as used by Vuletic and thus in order to make the scheme practically applicable, one still needs to expand the limits of cavity cooling towards allowing hotter samples and cooling on an even faster timescale. Fortunately, by a careful choice of geometry like in a confocal cavity, the limits to profit from collective enhancement and self-organization can be further lowered [1, 19]. In a first test, calculations and simulations were performed for parameters derived from existing dense molecular beam ovens [7]. Using somewhat heuristic scaling arguments these simulations showed that for sufficient initial density and available laser power, cavity cooling could even turn out to be a viable route to cold molecular ensembles of heavy and complex molecules up to clusters. In fact, the required parameters were not too far from current technological limits although the combination of very high powers and high $Q$ cavities is not too well tested.

In this work, we first want to substantiate these early rather promising results under more realistic and quite challenging conditions [7] and test various possible improvements towards a more realistic and practical implementation as milestone to full three-dimensional (3D) cavity cooling. Again we stick to transverse cooling of a fast and dense molecular beam via a single pass through the overlap region of an optical resonator and a strong far-of-resonance optical pump beam. The central goal in these setups is to reach conditions where the particles self-organize into a regular pattern with a Bragg-maximum pointing in the cavity direction. This is a prerequisite to get collective enhancement of scattering and cooling [22]. In order to stay close to experiments we will roughly keep the source parameters estimated from experiments with existing dense molecular sources as e.g. used in Vienna. Here, we first aim to solidify and better quantify previous results by studying transverse collimation and cooling over a wider range of parameters and configurations. As for the mirrors we also stay close to recent technological achievements, we expect not to present too spectacular results but rather to exhibit the minimum requirements to achieve something measurable and potentially useful. For the particles we only assume a given linear optical polarizability and mass. Hence the results are in principle applicable to a wide class of molecules or even polarizable nanoparticles and clusters [7, 34].
Figure 1. Scenario: the pulsed molecular beam propagates along the $y$-axis through a far red detuned standing wave laser along $x$ and a high finesse cavity along $z$.

Naturally, we want a high polarizability to mass ratio and low initial source temperature, while photon absorption at the chosen frequency should be as low as possible.

This work is organized as follows: in section 2, we recapitulate a classical point particle model as the basis of our cavity cooling analysis. In principle, these equations allow to derive analytical formulae for the necessary conditions to get self-ordering and cooling, as they have been derived for homogeneous 1D setups \[11\]. However, it is not clear how useful these limits are for a practical 3D setup with bunched operation. Hence in section 3, we will simulate a realistic scenario involving a fast and dense bunch of particles with realistic velocities, densities and masses. Our first interest here will focus on the minimum requirements for a successful experimental demonstration. In the following sections 4 and 5, we discuss a couple of new ideas to lower the experimental requirements for collective cooling under given density and laser power restrictions via injection seeding or pump power recycling.

2. Polarizable point particles moving in a cavity field

Let us develop a simple classical point particle model for a large molecular ensemble inside a high-quality cavity. The molecular dipoles interact with the cavity electric field as well as with an external driving laser tuned to the far red of any optical molecular resonance. It is applied from the side of the cavity as shown in figure 1. The dipole-field interaction induces spatially dependent Stark shifts generating the required light forces to influence the particle motion. Simultaneously the induced dipoles are the dominant source terms of the cavity field. In some cases, we will also allow for direct light injection into the cavity modes from an external pump field. To provide for dissipation we include cavity field damping due to losses via the mirrors.

2.1. General model

In the following, we present a classical model for such a scenario involving $N$ point particles. The dynamical quantities of our model are the cavity electric field $E(x, t)$, the dipole moment vectors $p_j(t)$ and the center-of-mass positions $x_j$ of the particles. Starting from Maxwell’s equations we thus obtain a wave equation for the field $E$ with a driving term proportional to the
total polarization given by (for point-like particles)

\[ P(x, t) = \sum_{j=1}^{N} p_j(t) \delta^{(3)}(x - x_j(t)). \]  

On the other hand, the evolution of the molecular dipoles \( p_j(t) \) is determined by a linear response to the total electromagnetic field at the particle position \( E^{tot}(x_j, t) \). We model their fast internal dynamics \( p_j \) as Lorentz oscillators and use the time-averages of the Lorentz force to determine the equations of motion for \( x_j \),

\[ m \ddot{x}_j = \nabla_E \left[ p_j(t) \cdot E^{tot}(x_j, t) \right], \]

where \( m \) denotes the particle mass and the index \( E \) on the nabla operator restricts its action on the variables of \( E^{tot}(x_j, t) \). As we will later see, this provides a self-consistent set of closed equations.

We will use here a few simplifying assumptions. First, each molecular dipole moment might be composed of several electrons harmonically bound to the particle with eigenfrequencies \( \omega_k \), so that we can write \( p_j = \sum_k p_{j,k}(\omega_k) \). The equations for the Lorentz oscillators including damping \( (\Gamma_k) \) thus read

\[ \ddot{p}_{j,k} + 2\Gamma_k \dot{p}_{j,k} + \omega_k^2 p_{j,k} = \frac{q^2}{m_e} \left[ E(x_j(t), t) + H^{ext}(x_j(t), t) \right], \]

where we added an external driving field, \( H^{ext}(x, t) \), while \( m_e \) and \( q \) are the electron mass and charge.

As the involved fields vary slowly in the transverse direction, we can set \( \nabla \cdot P \approx 0 \). Further we assume that \( P \) contributes only to a single polarization component of the electric field and we thus can ignore the vector nature of the fields and the polarization. Finally, we model the losses and the external driving of the cavity field by a current density

\[ J(x, t) = \sigma \left[ E(x, t) - E^{ext}(x, t) \right], \]

where \( E^{ext}(x, t) \) denotes the pump field.

Altogether the cavity field and the dipole moments can be shown to satisfy the equations (dropping the vector character, from now on)

\[ \left(-c^2 \nabla^2 + \frac{\partial^2}{\partial t^2} + \frac{\sigma}{\varepsilon_0} \frac{\partial}{\partial t} \right) E(x, t) = -\frac{1}{\varepsilon_0 \partial t^2} P(x, t) + \frac{\sigma}{\varepsilon_0} \frac{\partial}{\partial t} E^{ext}(x, t), \]

\[ \ddot{p}_{j,k} + 2\Gamma_k \dot{p}_{j,k} + \omega_k^2 p_{j,k} = \frac{q^2}{m} \left[ E(x_j(t), t) + H^{ext}(x_j(t), t) \right]. \]

Note at this point that both driving fields are set to have the same frequency of oscillation (significantly smaller than the molecular resonance frequencies, \( \omega_p < \omega_k \)). In the spatial coordinates the fields can decomposed into left and right traveling waves as

\[ E^{ext}(x, t) = E^{ext}(x) \exp(-i \omega_p t) + c.c., \]

\[ H^{ext}(x, t) = H^{ext}(x) \exp(-i \omega_p t) + c.c. \]

Let us expand the cavity field into the normal modes \( E_n(t) \) via the spatial overlap with a set of real orthogonal mode functions \( f_n(x) \) with eigenfrequencies \( \omega_n \) and equal mode volume

\[ \int d^3x \ f_n(x)f_m(x) = V \delta_{n,m}, \]

\[ E_n(t) \equiv \frac{1}{V} \int d^3x \ E(x, t) f_n(x). \]
Note that it is straightforward to generalize the model for imaginary mode functions. Let us further include only quasi-resonant modes (i.e. transverse modes with equal frequencies $\omega_n \equiv \omega_c \approx \omega_p$) while neglecting all other longitudinal mode families. For molecules this assumptions needs some care and in some cases, other longitudinal modes can be used for ro-vibrational cooling [26]. Within this approximation the field can be written as

$$E(x, t) = \sum_n E_n(t) f_n(x).$$

(9)

It is then straightforward to show that the mode amplitudes follow the equations

$$\dot{E}_n + \frac{\sigma}{\varepsilon_0} E_n + \omega_c^2 E_n = -\frac{1}{\varepsilon_0} \dot{P}_n + \frac{\sigma}{\varepsilon_0} E_{\text{ext}}^n$$

(10)

with the spatial overlap of the mode functions with the polarizability ($P_n$) and the cavity driving field ($E_{\text{ext}}^n$) as defined in (8). We have

$$P_n(t) = \frac{1}{V} \sum_j p_j(t) f_n(x_j),$$

(11a)

$$E_{\text{ext}}^n(t) = \mathcal{E}_{\text{ext}}^n \exp(-i\omega_p t) + \text{c.c.},$$

(11b)

with constant driving strengths $\mathcal{E}_{\text{ext}}^n = (1/V) \int d^3 x \mathcal{E}_{\text{ext}}(x) f_n(x)$. We now make the following ansatz:

$$E_n(t) = \mathcal{E}_n(t) \exp(-i\omega_p t) + \text{c.c.},$$

(12a)

$$P_n(t) = \mathcal{P}_n(t) \exp(-i\omega_p t) + \text{c.c.},$$

(12b)

where the amplitudes $\mathcal{E}_n(t)$ vary slowly on the timescale defined by $\omega_p^{-1}$, i.e. we have $|\ddot{\mathcal{E}}_n| \ll \omega_p |\dot{\mathcal{E}}_n|$ and analogously for $\mathcal{P}_n(t)$. With these approximations we find for the mode amplitudes

$$\dot{\mathcal{E}}_n + (\kappa - i\Delta_c) \mathcal{E}_n \approx \frac{i\omega_p \mathcal{P}_n}{2\varepsilon_0} + \kappa \mathcal{E}_{\text{ext}}^n,$$

(13)

where $\Delta_c = \omega_p - \omega_c$ and $\kappa = \sigma/(2\varepsilon_0)$.

In order to calculate $\mathcal{P}_n$, we assume that the temporal evolution of $\mathcal{E}_n(t)$ as well as of $x_j(t)$ is much slower than the relaxation time of $\pi_{j,k}(t)$. We thus introduce the slowly varying amplitudes of the dipole moments as

$$p_{j,k}(t) = \pi_{j,k}(t) \exp(-i\omega_p t) + \text{c.c.},$$

(14)

by inserting (14) into equation (6) and solving for the steady state $\pi_{j,k}$. This approximation implies that the dipole moments and thus the polarization ($\mathcal{P}_n(t) = (1/V) \sum_j \pi_{j}(t) f_n(x_j)$ with $\pi_j = \sum_k \pi_{j,k}$) adiabatically follow the electric field. After some manipulation we get

$$\pi_j(t) = \alpha(\omega_p) \left[ \sum_n \mathcal{E}_n(t) f_n(x_j) + \mathcal{H}_{\text{ext}}(x_j) \right],$$

(15)

where we have introduced the combined molecular polarizability

$$\alpha(\omega_p) = \frac{q^2}{m_e} \sum_k \frac{1}{\omega_k^2 - \omega_p^2 - 2i\Gamma_k \omega_p}.$$  

(16)
As \( \omega_p \ll \omega_k \) the reactive response of the molecular dipoles to the driving fields dominates the dissipative response. In order to avoid being close to any of the molecular electronic resonances, the driving laser preferentially should operate in the red or infrared region so that
\[
\omega_k - \omega_p \gg \Gamma_k
\]
is very well fulfilled. In this case, we will have negligible saturation and we can neglect the imaginary part of \( \alpha(\omega_p) \) and write
\[
\alpha(\omega_p) = -\frac{2\varepsilon_0 V}{\omega_p} U_0,
\]
where \( U_0 < 0 \) is the light potential per photon.

As a last self-consistent step in this model we have to calculate the light-induced mechanical forces \( m \ddot{x}_j = F_j \) on the particles center-of-mass. The time-averaged force on the \( j \)th particle is given by equation (2) so that, neglecting the fast oscillating terms, we have
\[
F_j = \pi_j(t) \nabla \left[ \sum_n \mathcal{E}_n(t) f_n(x_j) + \mathcal{H}^{\text{ext}}(x_j) \right] + \text{c.c.}
\]

It is convenient to rewrite these equations in terms of dimensionless field amplitudes according to
\[
\alpha_n(t) = \sqrt{\frac{2\varepsilon_0 V}{\hbar \omega_p}} \mathcal{E}_n(t), \tag{20a}
\]
\[
\xi_n = \sqrt{\frac{2\varepsilon_0 V}{\hbar \omega_p}} \mathcal{E}_n^{\text{ext}}, \tag{20b}
\]
\[
\eta - U_0 h(x_j) = \sqrt{\frac{2\varepsilon_0 V}{\hbar \omega_p}} \mathcal{H}^{\text{ext}}(x_j), \tag{20c}
\]
where we have introduced the cavity pump amplitude \( \xi_n \) as well as the (transverse) pump amplitude of the particles \( \eta > 0 \). We also define the dimensionless total electric field amplitude at the position of the \( j \)th particle as
\[
e(x_j) = \sum_n \alpha_n f_n(x_j) + \frac{\eta}{-U_0} h(x_j). \tag{21}
\]

Altogether, we get the following set of coupled differential equations, generalized here to the case of complex mode functions,
\[
\dot{\alpha}_n = (i\Delta_c - \kappa)\alpha_n - iU_0 \sum_{j=1}^N e(x_j) f_n^*(x_j) + \xi_n, \tag{22a}
\]
\[
\dot{x}_j = v_j, \tag{22b}
\]
\[
\dot{v}_j = -\frac{\hbar U_0}{m} \nabla_j |e(x_j)|^2. \tag{22c}
\]

Note that the sum in equation (22a) can be split into three parts with different physical interpretation. These are: (i) the effective refractive index of the particles (terms proportional
to $|f_n|^2$ giving rise to a dynamically modified effective eigenfrequency (detuning) $\Delta_{n,\text{eff}} = \Delta_n - U_0|f_n(x_j)|^2$ for each mode. (ii) Stimulated photon scattering between different modes via the particles (terms proportional to $f_m f_n^*$ with $m \neq n$), and (iii) ‘Brillouin-type’ scattering from the driving laser in to the cavity mode which reads: 

$$i\eta \sum_j h(x_j) f_n^*(x_j).$$

These equations agree well with the equations derived via semiclassical expansion from a model involving quantized radiation modes and particle motion [8] and will be at the heart of our following considerations and numerical simulations. These equations should also be applicable to the dynamics of larger polarizable particles, clusters or droplets as long as they are significantly smaller than the wavelength used. As the (static) polarizability only decreases slowly with wavelength it might be even favorable to use a far infrared laser, where high power is more easily available.

### 2.2. Standing-wave and ring-cavity geometry

Let us explicitly write down the equations of motion for generic examples in 3D. We start with a ring-cavity setup, where the field consists of two counter propagating modes with amplitudes $\alpha_\pm$ and mode functions

$$f_\pm(x) = \exp[-(x^2 + y^2)/w_0^2] \exp(\pm ikz).$$

The transverse pump laser is described by a Gaussian standing-wave with spot size $w_p$ and field distribution

$$h(x) = \exp[-(y^2 + z^2)/w_p^2] \cos(kx).$$

Using these functions, equation (22a) yields already quite complex expressions, so that we content ourselves at this point with the equations in 1D (along the cavity axis),

$$\dot{\alpha}_\pm = (i\delta - \kappa)\alpha_\pm - iNU_0\alpha_\pm (e^{\mp 2ikz}) + iN\eta \langle e^{\pm ikz} \rangle + \zeta_\pm,$$

$$\dot{v}_j = v_j,$$

$$\dot{z}_j = \frac{2\hbar k}{m} \text{Im} \left[ 2U_0\alpha_+\alpha_-^* e^{2ikz} - \eta \left( \alpha_+ e^{ikz} - \alpha_- e^{-ikz} \right) \right],$$

where $\delta = \Delta_c - NU_0$ and we have introduced $\langle \cdot \rangle$ as the abbreviation for the average value according to

$$\langle \xi(z) \rangle = \frac{1}{N} \sum_{j=1}^N \xi(z_j).$$

In equations (26a) and (26c), we have for the moment also neglected the shape of the driving laser along the z-axis. One clearly sees the collective action of the particles on the modes in equation (26a) while the origin of the forces as intensity redistribution between the modes as well as between the pump and the modes is evident in equation (26c).

Similarly one obtains the following equations (again in 1D) for a standing-wave cavity with mode function $f(z) = \cos(kz)$:

$$\dot{\alpha} = (i\Delta_c - \kappa)\alpha - iNU_0\alpha \langle \cos^2(kz) \rangle + iN\eta \langle \cos(kz) \rangle + \zeta,$$
\[ \dot{z}_j = v_j, \quad \text{(28b)} \]
\[ \dot{v}_j = \frac{2\hbar k}{m} \left[ |U_0|^2 \sin(kz_j) \cos(kz_j) - \eta \text{Re}\{\alpha}\sin(kz_j) \right]. \quad \text{(28c)} \]

Using various analytical approaches and simulations it has been shown that these (1D) equations predict self-organization of the particles into a regular pattern maximizing pump–cavity energy flow, whenever a certain pump amplitude threshold is reached [27]. Reaching this condition is essential to achieve significant phase space compression. In order to relate this threshold to a real physical quantity at this point, we can directly express \( \eta \) in terms of the power \( P \) of the driving laser. For a focused Gaussian beam with waist \( w_L \) and frequency \( \omega_p \) we get:

\[ |\eta| = \frac{\alpha(\omega_p)}{\varepsilon_0} \sqrt{\frac{\omega_p P}{\hbar c V w_L^2 \pi}}. \quad \text{(29)} \]

For negative detuning \( \delta < 0 \), the threshold pumping strength for self-organization in 1D reads

\[ \eta_c = \sqrt{\frac{k^2 + \delta^2}{2N}} \frac{k_B T}{\hbar (-\delta)}, \quad \text{(30)} \]

which is valid for both standing-wave [1] and traveling-wave [27] geometry; \( k_B T \) denotes here the initial kinetic energy of the particles. Equation (30) then yields the following threshold condition for the laser power:

\[ \frac{P}{w_L^2 \pi} > k_B T \left( \frac{k^2 + \delta^2}{-\delta} \right) \left( \frac{\alpha}{\varepsilon_0} \right)^{-2} \frac{c}{\omega_p} \left( \frac{N}{V} \right)^{-1}. \quad \text{(31)} \]

While enough power always seems to work, as an important feature we want to emphasize that the threshold power scales inversely with particle number \( N \). Let us see what this means for a concrete experimental example. Using a confocal cavity with a high finesse \( F = 50000 \) and length \( L = 5 \) cm results in a line width of \( \kappa = 188 \) kHz. The example molecules here are \( \text{C}_{60} \) compounds with mass \( m = 720 \) AMU and a measured fairly low polarizability \( \alpha/\varepsilon_0 = 83 \text{ Å}^3 \) [7] at the driving laser wavelength \( \lambda_p = 2\pi c/\omega_p = 1560 \) nm. For these parameters the number of necessary particles to achieve a significant refractive index in the mode, \( \kappa = |U_0|N \), is of the order of \( N \approx 10^9 \). Although this sounds rather high, source densities allowing numbers of this order have been reported recently [7].

So for the moment we fix \( \delta = NU_0 = -\kappa \) and calculate the threshold power for kinetic energy corresponding to a mean transverse velocity \( v_0 = \lambda \kappa \approx 1/3 \text{ m s}^{-1} \), i.e. \( kT \). We then find

\[ P_c = k_B T \left( \frac{\alpha}{\varepsilon_0} \right)^{-1} w_L^2 \pi c, \quad \text{(32)} \]

which amounts to about 6 kW for a Gaussian laser beam focused to a waist of \( w_L = 100 \) \( \mu \)m. This power can be injected directly or equivalently provided by a resonant Fabry–Perot-type pump cavity of the same size containing about \( 10^{12} \) photons. In the latter case, the required input power is reduced by factor given by the finesse of the pump cavity. Although still challenging, such a setup seems at least conceivable. In the next section, we see how this translates to a 3D case.

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3. Numerical studies

3.1. Collective enhancement and scaling of cavity cooling forces

Since the coupling of a single molecule to the electric field is tiny, standard cavity cooling is too slow in this case [14]. Even for very high-\(Q\) cavities the resulting light potentials/photoncs are typically many orders of magnitude smaller than the cavity linewidth, yielding a very long cooling time, which is in particular true for rather massive particles. One would, for example, predict cooling times of the order of seconds for individual \(C_{60}\) molecules in such a cavity.

However, using very high number densities, collective effects can be expected to strongly enhance the cooling efficiency [1]. If one wants to get cooling on a microsecond timescale, up to \(10^{10}\) molecules are needed in the cavity to profit enough from the \(N^2\) scaling of the total cooling force. In particular, the number of particles \(N\) is crucial in beam-type experiments [7] where the cooling time is limited by the molecule transit speed. It is, of course, not possible to numerically integrate all these equations \((28a)\) for such large particle numbers. Unfortunately, in particular in the transient case, it not so obvious how to scale parameters and particle numbers to reliably predict the dynamics of such a large number of particles. While the average scattering effect can be mimicked by using fewer particles with bigger dipole moments, this is not so easy for forces as simply increasing the mass of each particle to simulate more particles leads to a slowing of the motion of individual particles, while keeping the mass the same changes the center-of-mass dynamics of the cloud.

Nevertheless in the following, we try to simulate the scenario by a significantly smaller number of equations as follows. As obvious from equation \((26c)\), the dynamics of a single particle \(j\) is solely determined by the momentary electric field at its position, \(e(x_j)\), while the total field corresponds to an average over all particles. Hence one possible route is to approximate these averages from the motion of a representative sample of \(N^* = N/\epsilon\) particles, with \(\epsilon > 1\). The crucial point here is to correctly estimate the scaling of the averages with the particle number. We simply achieve this here by imposing an effective weighting factor \(\epsilon\) multiplying the contribution of each particle in the sum of equation \((27)\). This method somehow amounts to opinion polling among the involved particles but of course amplifies fluctuations which often scale as \(\sqrt{N}\). Hence as a first goal we will test this scaling method and see how far it can be applied.

The equation for the mode amplitudes including the extrapolation of the sum can be written as

\[
\dot{\alpha}_n = (i\Delta_c - \kappa_n)\alpha_n - iNU_0 \langle e(x)f_n^*(x) \rangle + \zeta_n, \tag{33}
\]

where \(\langle \cdot \rangle\) indicates the average value

\[
\langle \xi(x) \rangle = \frac{1}{N^*} \sum_{j=1}^{N^*} \xi(x_j). \tag{34}
\]

The latter is essentially given by the particle position distribution but can in general depend on \(N^*\). As an example, this happens for a homogeneous gas where the average value of the pumping term vanishes. Let us define the parameter

\[
\sigma(k) = \langle \exp[-2i\mathbf{k} \cdot \mathbf{x}] \rangle \tag{35}
\]

describing spatial order of period \(\pi/|k|\). For a homogeneous gas, \(|\sigma(k)| = 0\) for any value \(k\) (direction and period). Mathematically, the pumping term \((23)\) can be expanded into plane
Figure 2. Demonstration of the scaling method for $N = 10\,000$ molecules in a 1D ring cavity for different values of the weighting factor $\epsilon$, as indicated in (a). The plots show the photon number (a), the order parameter $|\sigma|$ (b) and the RMS velocity (c) after a short time evolution as functions of $\eta$. The relative deviation of $\Delta v_z$ for $\epsilon = 10$ (diamonds) and 100 (squares) is shown in (d). The insets in (a) and (b) depict the relative deviations of $|\alpha|$ and $|\sigma|$, respectively. The parameters are $\delta = NU_0 = -\kappa$.

waves—i.e. expressed in terms of $\sigma(k/2)$—and one finds that $\langle h(x) f^*_n(x) \rangle = 0$ for plane wave mode functions. Statistical density fluctuations, however, yield the general scaling $|\sigma|^2 \sim 1/N^s$. For higher spatial order of the particles, $|\sigma|$ increases up to unity for a perfect lattice with period of an integer multiple of $\lambda_p/2$. The contribution of density fluctuations diminishes then, which should reduce the error of the estimate in equation (34). Our scaling method should therefore perform well at least above $\eta_c$, where we can expect higher spatial order of the molecules. As for an individual particle the effect of the other particles only acts via the total field, the correct scaling of the field amplitudes is crucial here.

Figure 2 gives an accuracy check of the scaling method for $N = 10\,000$ simulated particles in a 1D ring-cavity configuration, where we can simulate all cases explicitly. More precisely, we simulated equations (26a)–(26c) with the replacement (34) for the averages using values of $\epsilon = 1$ (blue circles), $\epsilon = 10$ (green diamond markers) and $\epsilon = 100$ (squares).

The molecules were initially randomly distributed along the axis with relatively low initial kinetic energy (Maxwellian velocity distribution with $k v_0 = \kappa$). After a time $t = 10\kappa^{-1}$ the simulations were stopped and the photon number (a), the absolute value of the order parameter...
\( \sigma(k) \) (b) and the RMS velocity (c, in units of the initial mean velocity) were calculated for various values of the pumping strength \( \eta \), as indicated in (a).

As expected, the results agree well for sufficient high \( \eta \) where the curves tend to cover each other. We therefore have also calculated the deviations for \( \epsilon = 10 \) and 100 by the ratio of the difference and the sum of the corresponding curves with the values for \( \epsilon = 1 \), respectively. Figure 2(d) and the insets in (a) and (b) show the relative deviations of \( \Delta v_z, |\alpha_\pm|^2 \) and \( \sigma \) for \( \epsilon = 10 \) (diamonds) and 100 (squares), respectively. The curves strongly reflect the threshold characteristics where below \( \eta_c \), the values of the photon number and the parameter \( \sigma \) are, however, too large for \( \epsilon = 10 \) and 100. It is remarkable, on the other hand, that the average kinetic energy shows very little deviation over the whole range of the pumping strength.

We infer from these results good reliability of our method above \( \eta_c \) as long as \( \epsilon < \sqrt{N} \). Even below the critical self-organization value, where fluctuations dominate, there seems to be a systematic dependence on \( \epsilon \) from which we could extract predictions for higher \( N \).

3.2. Transverse beam compression

Let us now turn to practical examples of molecular cooling. While steady-state trapping and cooling of molecules certainly is a realistic long term goal for cavity enhanced laser cooling, first-generation experiments will aim to employ cavity cooling to increase the phase-space density of a particle beam traversing the cavity. Here, as depicted in figure 1, the cavity and the vertical pump laser are intersected by the beam from the side. Naturally, the cooling time of the particles is limited by its rather short time of flight through the fields. Still, for large enough densities, collective enhancement can be used to get noticeable results once the self-organization threshold laser power has been reached.

As practical quantitative measures for the effect of the setup we will use the change in the transverse phase-space density \( \rho \) (along the cavity axis) as well as the total particle number emerging from the interaction region through a adjustable aperture behind the cavity. Hence, we define \( \rho \) as the inverse of the product \( \Delta z \Delta v_z \), where \( \Delta z \) is the beam width (RMS distance from beam center) and \( \Delta v_z \) denotes the RMS velocity which is proportional to the square root of the total kinetic energy. It is clear that an increase in phase space density can be achieved either via spatial compression of \( \Delta z \) or momentum compression \( \Delta v_z \). This includes a standard notion of laser cooling via reduction of the mean kinetic energy (\( \Delta v_z \)). Note that for any freely evolving particle cloud with nonzero mean kinetic energy, the beam width will grow linearly in time with \( \partial_t \Delta z(t) = \Delta v_z \). Hence a linearly growing \( \rho \) should serve as a reference here and we will compare \( \rho \) with the phase-space density for the case without pumping—i.e. with \( \rho_0 = \rho|_{\eta=0} \).

Let us now look at some typical results. Figure 3(a) shows the time evolution of \( \rho/\rho_0 \) for a cloud of \( N = 10^8 \) molecules passing through a ring cavity with mode functions given by (24) and mode amplitudes \( \alpha_\pm \). Here we have neglected the radial spatial variation of the pump laser so that \( h(x) = \cos(kx) \) is assumed to be broader than the cavity waist. The three curves obtained by integrating equations (22a)–(22c) correspond to different values of the pump strength, \( \eta = 2\eta_c \) (red), \( \eta = 3\eta_c \) (green), and \( \eta = 5\eta_c \) (black). In the simulation, we have explicitly used \( N^* = 10^5 \) particles (i.e. \( \epsilon = 1000 \)) with initial kinetic energy according to Maxwell’s velocity distribution with \( v_0 = 4\pi\kappa/k \). For the abscissa we have chosen here the distance \( y \) of the cloud center from the cavity axis.

Note the drop in \( \rho \) during the passage of the cloud through the cavity, which is due to fast particle oscillations in the optical potentials. Large kinetic energies are reached near the
Figure 3. Evolution of the phase-space density $\rho$ (upper row) and corresponding photon numbers $|\alpha|_\pm^2$ (lower row) of a cloud of $N = 10^8$ molecules passing through a ring cavity for different values of the transverse pumping strength $\eta$, as indicated. In (c) and (d), an additional external field directly drives the cavity with one per cent of the transverse laser power. The other parameters are $NU_0 = \delta = -\kappa$, $\epsilon = 1000$ and $v_0 = 4\pi \kappa/k$.

To distinguish cooling from mere optical beam focusing we study $\rho/\rho_0$ for various pump strengths and at different times or distances from the cavity axis after passing. Cavity-induced cooling occurs if we get a relative enhancement of the phase-space density $\rho$ diminishing slower than $\rho_0$ after the interaction, which shows that the kinetic energy of the particles was reduced. In these simulations, we have chosen parameters where we typically find an increase of the relative phase space density up to a factor of two, where the transverse kinetic energy $E_z = m \Delta v_z^2/2$ is compressed by only about $10\%$. This is rather modest but should still be clearly measurable and shows the minimum requirements for a successful experimental demonstration. Note that we have to be significantly above the 1D self-organization threshold to obtain a measurable result.
Much higher compressions can be obtained for a stronger pump or better cavities. Note as well that we get a similar final kinetic energy for various values of $\overline{N}$ without qualitative differences in the results as long as we keep $NU_0$ constant.

Figure 3(b) depicts the corresponding time evolution of the cavity photon numbers $|\alpha_\pm|^2$ during transit of the bunch divided by the pump power $(\eta/\eta_c)^2$. This scaling also allows a better comparison of the results as for a fixed particle position distribution the scaled values should coincide for different $\eta$. When the particles are perfectly ordered we get a maximum attainable value of $|\alpha_\pm|^2 \approx 2.6 \times 10^{11}$, while randomly distributed particles yield a strong reduction by $1/N$. Hence the scaled curves give a direct indication of the particle ordering, which thus is relatively high for $\eta = 5\eta_c$ but relatively poor for $\eta = 2\eta_c$. Note that apart from any cooling the cavity photon number can be used for nondestructive bunch density detection, which could be useful by itself.

Interestingly, the maximum of $|\alpha_\pm|^2$ occurs slightly after the molecules have passed the cavity center with increasing offset for reduced pumping powers. Although the effective rate of light scattering into the cavity is not maximum there, localization effects lead to enhanced scattering. In fact, the scattered light can cancel due to destructive interference for a homogeneous particle distribution or when each single site of maximum coupling strength (distance $\lambda_p/2$) is occupied [23]. A checkerboard pattern, however, emerges due to positive feedback of statistical density fluctuations on the particle positions via the cavity dipole force. In this configuration, where the particles arrange in a tilted square lattice (with period $\lambda_p/\sqrt{2}$), constructive interference leads to efficient Bragg scattering of pump photons into the cavity.

Here the finite time for the pattern to establish causes the time offset in the maxima with respect to the cavity symmetry. It can be shown [27] that for short times, the cavity field amplitude as well as the localization parameter $\sigma (k)$ increase exponentially in time, $\sim e^{\gamma t}$. For a flat (quasi-continuous) position distribution the initial growth rate $\gamma$ can be estimated to have an upper bound according to

$$\frac{\gamma^2}{N} < \frac{\kappa^2 + \delta^2}{N} \left[ \left( \frac{\eta}{\eta_c} \right)^2 - 1 \right],$$

for $\eta > \eta_c$ and $\delta < 0$. Although the upper bound of $\gamma$ can in principle be increased arbitrarily via $\eta$, having fixed the operating point (by the ratio $\eta/\eta_c$) yields the scaling $\sim 1/\sqrt{N}$. It is now interesting to note that the predictions for this critical exponent could be tested using the growth rate and shift of the photon number evolution.

4. Cavity seeding

Obviously self-ordering and the corresponding critical exponents are crucial to obtain useful results. From a practical point of view one should look for possibilities to enhance them. One rather obvious possibility for this seems injection seeding of the cavity field in order to guide the molecules entering the cavity mode in a way to assist the process of self-organization in its initial phase. As the initial growth rate $\gamma$ is typically tiny for a homogeneous gas, a proper seeding field attracts the particles to high-intensity regions of constructive interference of cavity and pump field. This tends to reduce the ordering startup time and thus increases the effective exponent $\gamma$. In particular, close to threshold, a pre-existing cavity field could mimic the presence of more particles and thus reduce the threshold condition for self-organization.
Figure 4. Comparison of the phase-space density (a) between the cases with (diamonds) and without (circles) an additional cavity pumping laser. The curves were calculated for a cloud of \( N = 10^8 \) molecules after having passed the cavity at distance \( y = 2w_0 \) (at the screen, see figure 1). The cavity-driving laser power was adjusted to only 1% of the transverse laser power, i.e. \( \zeta = \eta/10 \). The other parameters are \( NU_0 = \delta = -\kappa \). Panel (b) shows the maximum photon number in the cavity during the passage of the cloud.

The positive effect of seeding is clearly shown in figure 3(c), where we have plotted the results for simulations as in (a) but with an additional cavity-driving laser with power \( P \) of 1% of the transverse laser power \( P_t \). While this is quite a small amount, the final phase space density is increased for all three parameter sets. For the parameters chosen the additional pump laser yields photon numbers of about \( 10^7 (\eta/\eta_c)^2 \) per mode in the cavity. Note in particular that cavity seeding has a significant impact on the cooling performance particularly for pumping close to the threshold \( \eta = 2\eta_c \), where we already get some visible effect now. Generally, seeding accelerates the particle self-organization such that the maximum of the photon number occurs earlier, when the particles are still much closer to the cavity axis, see figure 3(d). Hence the effective time the cooling works can be significantly increased. In our simulations we find that the offset time is reduced from about \( 3\kappa^{-1} \) to about half that value. This is nevertheless still larger than \( t_{\text{min}} = \lambda^{-1} \) obtained from a rather rough estimate from the 1D exponent \( (36) \) predicting \( t > 0.25\kappa^{-1} \) for \( \eta = 2\eta_c \).

A more general comparison of the phase-space density gain as a function of pump power for the two cases of no seeding and seeding can be found in figure 4. Figure 4(a) depicts \( \rho/\rho_0 \) after passing the cavity (at \( y = 2w_0 \)) as a function of the transverse driving laser strength \( \eta/\eta_c \) without cavity pump (blue circles) and with 1% seeding laser power (green diamond markers). In addition in figure 4(b), we have also plotted the maximum photon number \( |\alpha_\pm|^2 \) in the cavity mode during the passage of the molecules. As one might expect it is increased by orders of magnitude for low pumping strength, where self-organization is too slow without seeding. The effect becomes less important for larger \( \eta \) where the particles have ordered anyway before reaching the cavity center. This clearly shows that in particular, when power and density are only marginally enough for ordering, seeding is essential for getting good results.

How strong should this seeding be? As we know from previous simulations [7], cavity pumping alone does not lead to significant cooling. In our simulations we find that up to 10% seeding power the effect still grows but then starts saturating as is illustrated in figure 5(a).
Figure 5. Dependence of the phase-space density (upper plots) and the position and momentum RMS values (lower) of a molecular beam at the screen on the power (left plots) and the phase $\phi$ (right) of the cavity-driving laser. In (b) and (d), we have plotted the values of $\Delta z/z_0$ and $\Delta v_z/v_0$ where $z_0$ is the width of the nonperturbed beam. The transverse driving strength is $\eta = 2\eta_c$ while the other parameters are as before. In (a) and (b), $\phi = 0$, whereas in (c) and (d) we have used seeding laser powers as large as 5% of the transverse laser power. The points of coincidence are indicated by the arrows.

for $\eta = 2\eta_c$. This plot shows the phase-space density as a function of the seeding ratio, i.e. the ratio of the cavity driving laser power $P$ and the transverse pumping power $P_{tr}$. Another interesting piece of information is revealed if we split the effect of beam spreading and velocity compression. The corresponding values of the spatial width and the velocity spread are depicted in figure 5(b), displaying $\Delta z/\Delta z_0$ and $\Delta v_z/\Delta v_0$ where $\Delta z_0$ is the final beam width for the case of free particles and $\Delta v_0$ denotes the initial mean velocity. For larger seeding the phase space compression gets more and more due to spatial compression via guiding and less due to velocity compression.

Let us finally discuss one more subtlety in this setup. Whereas the phase of the scattered light from the transverse pump laser is determined by the particle positions (‘position’ of the checkerboard pattern), the phase of the seeding laser light directly fed into the cavity can be geometrically adjusted. This can be crucial in particular for a ring-cavity geometry, where the checkerboard pattern can form at any arbitrary position along the cavity axis in principle.
Figure 6. Phase-space plots at distance $y = 3w_0$ away from the cavity center for $\eta = \eta_c$ (a), $\eta = 5\eta_c$ (b) and (c). The other parameters are the same as in figure 4 but an additional cavity-driving laser with 5% seeding power was used in (c). The rectangles indicate $z = \Delta z$ and $v_z = \Delta v_z$, respectively.

Hence seeding requires the correct relative phase. The dependence of $\rho$ and $\Delta z$, $\Delta v_z$ on the phase difference $\phi$ of the cavity pump laser and the transverse laser is shown in figures 5(c) and (d), respectively. The arrows indicate points of coinciding parameters. Note that for these parameters, carefully adjusting the phase $\phi$ between the two lasers yields higher phase-space density than doubling the seeding laser power. On the other hand, this phase dependence clearly exhibits the relevance of seeding.

As a further step to exhibit the cavity effects we have plotted the positions and velocities of a representative sample of 1000 molecules as they would impinge at a screen 2.5 $w_0$ away from the cavity axis as depicted in figure 6 for $\eta = \eta_c$ (a) and $\eta = 5\eta_c$ (b). The red rectangles exhibit the region for which $|z| < \Delta z$ and $|v_z| < \Delta v_z$, respectively. The reduced tilt of the distribution clearly shows the improved source brightness behind the resonator induced by spatial and velocity compression of the particles.

5. Pump power recycling with a second cavity

From the simulations above we clearly see that enough pump power is a central requirement for a significant effect. Naturally instead of using a stronger and stronger laser, we can think of enhancing the transverse driving laser within a second cavity oriented along the $x$-axis. As long as the pump cavity has a very low finesse (large damping $\kappa$) its field will be stationary and the physical situation is almost identical to the previous one, except that we need less power or we can allow for a larger interaction region. However, as we already need a high $Q$ cooling cavity, we can think of using the same or similar mirrors again, so that due to the high finesse, new dynamics is introduced into the system. Here we can get a dynamic response of the pump cavity field to the presence of the molecular cloud which acts as a dynamical refractive index and a scattering loss into the original cooling cavity.

In principle, a properly tuned pump cavity can get out of resonance when a sufficiently large number of particles couple to its field; i.e. as soon as the total light shift $N|U_p|$ is on the order of the pump-cavity line width $\kappa_p$. As a striking example we assume that two identical cavities are used for pump enhancement and cooling. Hence the pump field itself becomes a dynamical
Figure 7. (a) Sample trajectory of the mean kinetic energy $E_{\text{kin}}$ of one million molecules for a setup with two identical, perpendicularly oriented cavities. One of them is resonantly driven with strength $\zeta = 3\eta_c$ while $E_{\text{kin}}$ is calculated along the direction of the other one. Panel (b) shows the corresponding photon numbers where the curve for the pump cavity is scaled down by ten. The parameters are as before.

The corresponding time evolution of the photon numbers in the two resonators is shown in figure 7(b) with the pump cavity intensity (green line) scaled down by a factor of ten. We seed a strong depletion effect of the molecules on the pump field, which is scattered into the cooling cavity. In general, we encounter significantly better cooling in this case than before. Although we have only explored a small range of possible values, such an improvement occurs for a wide range of pump strengths and detuning. Its physical origin is not completely clear but it could be related to the improvement one expects from multimode cooling setups in general [19]. Plots of the cavity-induced enhancement of the phase-space compression, $\rho/\rho_0$, and of $\Delta z$ and $\Delta v_z$ are shown in figures 8(a) and (b), respectively, as functions of the cavity driving strength $\zeta$ while the other parameters were left as before. The arrows indicate the values for the example parameters used to calculate the curves in figure 7. While technically certainly not easy, the use of a pump enhancement cavity could turn out to be a vital step towards a successful implementation of cavity cooling here.
Figure 8. Dependence of the phase-space density (a) and of $\Delta z$ and $\Delta v_z$ (b) on the driving strength of the pump cavity, $\zeta$, for the same parameters as before. The arrows indicate the driving strength used in figure 7.

6. Conclusions

The central challenge in applying light forces and in particular cavity cooling to molecules is to overcome the small effective ratio of single particle polarizability over mass at available frequencies far from any electronic resonance. In general, a very large detuning is required for suppression of spontaneous emission during optical manipulations to avoid optical pumping of the molecules to unwanted long-lived states. Thus one cannot profit from a resonantly enhanced dipole moment near an allowed optical transition. In practice, if lasers are available [19], one still can try to find a compromise between losses and resonant dipole moment enhancement by operating a bit closer to a molecular line. This will of course specifically depend on the chosen molecule and the number of open transitions in the scheme, which one might be able to close by a number of extra lasers. Here we have largely ignored such a possibility, which to our knowledge has not been tried experimentally either. Instead we opted to choose typical molecular parameters as one finds far from any resonance. To create noticeable forces we thus have to assume very high laser powers [13]. This choice also implies that for cooling we have to rely on collective enhancement of wanted versus unwanted scattering with the help of large particle numbers in the cavity mode [19]. Note that this limiting case is indeed also the basis for applying light forces to macroscopic objects as e.g. glass beads or cells in optical tweezers [16] or micromechanical mirror cooling setups [25]. Here billions of particles participate in the off-resonant light scattering. In many cases, the required conditions on laser powers or molecule densities in such setups can be achieved more easily using pulsed lasers [13], pulsed particle sources or even both. Realistic models involve millions of particles moving in laser fields up to kilowatts of laser power. The nonstationary time evolution and inhomogeneous geometry thus leaves numerical simulations as the only essential tool for reliable quantitative predictions.

In this work, we presented a simple classical model for the coupled particle–field dynamics in terms of optical polarizability and cavity damping rates. As a major step this model was extensively tested for suitable parameter rescaling to reduce the effective number of the degrees of freedom, i.e. the required particle number, for which we had to calculate the trajectories explicitly. As besides mean values, fluctuations play a central role in the initial growth of particle ordering and super radiance, special care is needed to determine the proper
scaling for the initial conditions. At the bottom line, finding the proper scaling is vital to study the interaction of a large number of polarizable particles with the electromagnetic field. By thorough testing and extrapolation we could reduce the required number of equations by several orders of magnitude through appropriate rescaling of interaction strength, mass, particle number and initial density fluctuations. This rescaling was then applied to a realistic cavity cooling configuration, where a fast and dense bunch of molecules propagates through the overlap region of a high-$Q$ cavity and a transverse pump laser.

Cavity-induced cooling and phase-space compression in the transverse direction occurs by stimulated photon scattering from the pump into the cavity. Although the effect is tiny for a single molecule, we found collective enhancement of the scattering probability for sufficient laser powers, where the particles organize in a periodic pattern maximizing collective photon scattering via the Bragg effect. Even though the particles spend only very little time in the cavity field and are only weakly excited to avoid spontaneous emission, our simulations predict a significant and measurable phase-space compression increasing the brightness of the beam source with currently available cavities, lasers and beam sources.

In order to get the best possible results, it is essential to speed up the self-organization phase during the entrance time of the pulse. While one could also think of structuring the particle beam, we show that additional phase controlled injection of a small amount of the laser power (seeding) even at relatively low power enhances the phase-space density of the transmitted particle beam. The effect of seeding can also be nicely demonstrated by varying the seeding phase, which turns the enhancement on and off. These early results suggest that even more effective enhancements could be expected by feedback control of the laser pump.

As the available pump power is one of the central experimental limitations, we introduce pump power recycling using a second resonator for the pump beam. In this configuration, we not only find much lower power requirements but we surprisingly also get better cooling at the same local intensity as for the free space pump. Though the origin of the additional cooling forces is not completely clear, pump depletion seems to play a vital role. We think that there are certainly many other clever ways to enhance the performance of such a beam compressor using, e.g. multimode geometries in a practical setup; for a first experimental demonstration of molecular laser cooling, the proposed geometry should nevertheless be sufficient.

From a theoretical point of view, proper rescaling of particle numbers is still an issue that is not completely resolved. One future possibility certainly is the use of continuous density description in a Vlasov-type approach. While this provides a more natural description for very large particle numbers, one still has to solve the issue of fluctuations in solving the corresponding 3D partial differential equations. Experimentally, it seems that it is mainly technical problems of resonator and beam source technology which remain to be solved for the scheme to work in a useful way. In this respect, several techniques for slowing and cooling will have to be combined in a hybrid setup, e.g. as recently proposed [20]. While this is certainly not easy, one finally would get a very general method to cool a great variety of polarizable objects from atoms and molecules even up to clusters, droplets and nanoparticles.

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