Semiclassical laser-cooling theory for a trapped multistate ion

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(Received 26 May 1993)

We present a semiclassical laser-cooling theory for an ion bound in a quadratic potential well, allowing for an arbitrary internal level scheme of the ion and an arbitrary polarization of the light field. The special case that the ion moves in a region much smaller than the wavelength of the exciting light (Lamb-Dicke limit), as well as an additional low-intensity limit, are worked out in detail. Explicit general expressions are given for the damping and diffusion tensors of the center-of-mass motion of the ion, and the light-induced renormalization of the mechanical oscillation frequencies is discussed. The formalism is implemented analytically using a computer program MATHEMATICA for an ion with a $j=\frac{1}{2}\rightarrow j=\frac{3}{2}$ transition moving in a one-dimensional optical molasses consisting of two counterpropagating laser beams with perpendicular linear polarizations (lin $\perp$ lin). For a weakly bound ion at negligible saturation, the greatest lower limit of excitation energy turns out to be $(\frac{33}{20})^{1/2}-\frac{1}{2}\approx 0.78$ vibrational quanta above the zero-point energy.

PACS number(s): 32.80.Pj, 42.50.Vk

I. INTRODUCTION

Recent experiments [1] have prompted the realization that both the Zeeman degeneracy of the atomic levels and the spatial variations of the polarization of light often have to be taken into account in the theory of laser cooling of atoms [2,3]. The traditional two-state description of the atom may in fact give a qualitatively incorrect picture of the light pressure in, say, optical molasses. In response to the challenge, an extensive and rapidly growing theoretical literature has arisen on cooling and trapping of multistate atoms [2–10].

On the other hand, laser cooling of trapped ions has paved the way for progress both in fundamental physics, e.g., quantum jumps [11–13], and in applications such as frequency standards [14]. A theory of Doppler cooling of a trapped *two-state* ion was compiled some time ago [15–17], and tested against numerical solutions for the quantum motion of an ion in a trap [18,19]. Compared to Doppler cooling, polarization-gradient cooling of free atoms offers a stronger damping of the atomic motion and lower cooling temperature. These features might also be assets with trapped ions. Although there have been a few isolated studies of closely related subjects [20–22], our short paper [23] focuses on the polarization-gradient cooling of a trapped ion. Apparently no experiments have yet been reported.

The purpose of the present paper is to expand our earlier concise summary [23] into a detailed semiclassical (SC) approach to laser cooling of a harmonically bound ion. The ion may have an arbitrary internal-level scheme, and may move in a light field with an arbitrary position-dependent polarization. We have advanced such a theory for a free atom in Refs. [4] and [10], hereafter referred to as I and II. In Ref. [9], from now on referred to as III, we have developed additional theoretical methods for laser cooling of a free atom specifically for the limit of low light intensity. Here a major fraction of I–III will be adapted to a trapped ion.

Section II contains our formal development. In Sec. II A we outline the modifications of the cooling theory required to go over from a free atom to a trapped ion. For a trapped ion, the Lamb–Dicke limit, according to which the ion resides in a region much smaller than the wavelength of the exciting light, has proven both experimentally feasible and theoretically convenient. Section II B is thus dedicated to the Lamb-Dicke limit. The case with a simultaneous Lamb-Dicke limit and nonsaturating laser intensity is taken up in Sec. II C.

Section III presents two explicit applications of the theory. In Sec. III A we demonstrate that for a two-state ion the well-known results are regained. Section III B discusses the special case of an ion with a $j=\frac{1}{2}\rightarrow j=\frac{3}{2}$ transition moving in a one-dimensional (1D) optical molasses consisting of two counterpropagating laser beams with perpendicular linear polarizations (lin $\perp$ lin). It turns out that the interplay between the optical pumping time of the internal state of the ion and the period of the oscillations of the center of mass sets the lowest limit of the cooling temperature. In our example the ion can be cooled until its excitation energy is about one vibrational quantum above the zero-point energy.

In our SC approach the c.m. motion of the ion is treated classically. For instance, we simultaneously refer to the position and velocity of the ion. Nevertheless our cooling theory respects the quantum-mechanical zero-point limit of the energy of a harmonic oscillator. This issue is discussed in the final Sec. IV of the present paper.

II. COOLING THEORY FOR A TRAPPED ION

In this section we describe the changes of the SC theory of laser cooling and trapping as one goes from a free atom to a trapped ion. We state our main results in full and aim at self-containedness on a general level, but a

few technical details may only be appreciated by a reader familiar with I–III.

A. General semiclassical theory

Paper I opens with a full quantum treatment of both the internal degrees of freedom and the c.m. motion of a free atom. In I we use free-particle plane waves to represent the c.m. motion. It is possible to use the plane waves for the c.m. motion of a trapped ion just as well. Now, in the derivation of spontaneous relaxations in I we ignored the energy of the c.m. motion in comparison with the energy differences between the atomic levels. For that argument it does not matter whether the c.m. motion is free flight or bound oscillations of a trapped ion. All spontaneous damping terms for a trapped ion come out exactly the same as in I. Induced interactions with the external field are also the same. The c.m. motion appears only in the term $-(i/\hbar)[H_{\text{c.m.}},\rho]$ in the Liouville–von Neumann equation. The explicit form of this commutator in the plane-wave basis is quite involved for a trapped ion.

For definiteness we assume the c.m. Hamiltonian

$$H_{\text{c.m.}}=\sum_i\left[\frac{\hat{p}_i^2}{2M}+\frac{M\nu_i^2\hat{r}_i^2}{2}\right] . \tag{2.1}$$

Here $\hat{p}_i$ and $\hat{r}_i$ are the momentum and position operators of the ion in the three principal axis directions $i=1,2,3$ (or x,y,z) of the trap, M is the mass of the ion, and ν_i is the mechanical oscillation frequency of the ion in the direction i. $H_{\text{c.m.}}$ is the Hamiltonian of a generally anisotropic harmonic oscillator. The benefits are reaped in the next step of the development of I, in which spontaneous and induced processes as well as the evolution due to $H_{\text{c.m.}}$ are converted into the Wigner representation of the c.m. motion. Because of its form, $H_{\text{c.m.}}$ is taken into account in the fully quantum-mechanical equations of motion of the Wigner functions simply by using the classical convective derivative as appropriate for a trapped ion.

The net result is that the spontaneous emission contributions to the equations of motion of the Wigner functions, Eqs. (2.33)–(2.36) of I, apply as they stand. The remaining terms are given by Eqs. (3.8) of I, with the sole change that the convective derivative is to be interpreted as

$$\frac{d}{dt}=\frac{\partial}{\partial t}+\sum_i\left[\frac{p_i}{M}\frac{\partial}{\partial r_i}-M\nu_i^2 r_i\frac{\partial}{\partial p_i}\right] \tag{2.2}$$

instead of (3.9) of I. The free-atom theory is regained by setting $\nu_i=0$.

In I we next engage in an adiabatic elimination of the internal degrees of freedom of the atom, so that eventually only the Wigner function representing the c.m. motion remains. The difference between the present case and the free-atom theory of I enters through the translation superoperator, which now reads

$$e^{\tau\mathcal{D}}\equiv\exp\left[-\tau\sum_i\left[\frac{p_i}{M}\frac{\partial}{\partial r_i}-M\nu_i^2 r_i\frac{\partial}{\partial p_i}\right]\right] . \tag{2.3}$$

The form of I, Eq. (4.20), is again obtained by setting $\nu_i=0$. The "τ shift" of an arbitrary function $f(\mathbf{r},\mathbf{p})$ is once more defined as

$$f_\tau\equiv e^{\tau\mathcal{D}}f . \tag{2.4}$$

One may see directly from the definition that the function f_τ satisfies the partial differential equation

$$\left[\frac{\partial}{\partial\tau}+\sum_i\left[\frac{p_i}{M}\frac{\partial}{\partial r_i}-M\nu_i^2 r_i\frac{\partial}{\partial p_i}\right]\right]f_\tau=0 . \tag{2.5a}$$

This is the Liouville equation for a harmonic oscillator, and can be solved easily with the proper initial condition

$$f_{\tau=0}(\mathbf{r},\mathbf{p})=f(\mathbf{r},\mathbf{p}) . \tag{2.5b}$$

The result (in somewhat abbreviated notation) reads

$$f_\tau(r_i,p_i)=f\left[r_i\cos(\nu_i\tau)-\frac{p_i}{M\nu_i}\sin(\nu_i\tau),\ p_i\cos(\nu_i\tau)+M\nu_i r_i\sin(\nu_i\tau)\right] . \tag{2.6}$$

The new form of the τ shift has two consequences. First, in the subsequent calculations in I finding the force and the diffusion tensor were connected to finding certain internal-state operators of an atom that moves along a straight free-flight trajectory. As one might surmise, a similar prescription works here. In the present case one just has to take the trajectory of a harmonic oscillator. The second change from I is more subtle. The fact of the matter is that in certain intermediate calculations one winds up taking partial derivatives of τ-shifted quantities with respect to momentum. An explicit example is provided by Eqs. (4.55)–(4.61) of I. Adapted to the trapped ion, we find intermediate expressions such as

$$\frac{\partial}{\partial p_i}f_{t-t'}=-\frac{\sin[\nu_i(t-t')]}{M\nu_i}\frac{\partial f}{\partial r_i}\bigg|_{t-t'}+\cos[\nu_i(t-t')]\frac{\partial f}{\partial p_i}\bigg|_{t-t'} . \tag{2.7}$$

The first term on the right-hand side of (2.7) is ignored by virtue of a similar argument as in I. However, the newly introduced cosine factor survives all the way to the final results.

We are ready to summarize our semiclassical laser cooling theory of a harmonically trapped ion. The c.m. distribution function $f(\mathbf{r},\mathbf{p},t)$ obeys the Fokker-Planck equation

$$\left[\frac{\partial}{\partial t}+\sum_i\left[\frac{p_i}{M}\frac{\partial}{\partial r_i}-M\nu_i^2 r_i\frac{\partial}{\partial p_i}\right]\right]f=-\sum_i\frac{\partial}{\partial p_i}(F_i f)+\sum_{i,j}\frac{\partial^2}{\partial p_i\partial p_j}(D_{ij}f) . \tag{2.8}$$

The term "semiclassical" refers to the state of affairs that in this formulation the c.m. motion is treated classically. The ion simply moves under the harmonic restoring force and the light pressure force $\mathbf{F}$. In addition, the ion is sub-

ject to random diffusion that does its best to take into account quantum fluctuations in ion-field interactions. The diffusion is quantified by the diffusion tensor D_{ij}. In the same vein, in the remainder of this paper we take the c.m. motion of the ion to be classical.

To calculate the force and the diffusion tensor at time t in the phase-space point $(\mathbf{r},\mathbf{p})$ one takes an ion moving along a harmonic-oscillator trajectory

$$r_i(\tau)-r_i^0=(r_i-r_i^0)\cos[\nu_i(\tau-t)] + \frac{p_i}{M\nu_i}\sin[\nu_i(\tau-t)] \ . \tag{2.9}$$

Here $\tau\in(-\infty,0]$ is the parameter of the trajectory, which we call the running time. For later convenience we have introduced the position of the center of the trap $\mathbf{r}^0$ that may differ from zero. Next one finds four internal-state operators of the ion, ρ and $\eta_{1,2,3}$, by solving for the moving ion a set of equations analogous to the ordinary density matrix equations:

$$\frac{\partial}{\partial\tau}\rho=\mathcal{Q}\mathfrak{L}\mathcal{Q}\rho+\mathcal{Q}\mathfrak{L}\mathcal{P}\frac{\mathbb{1}}{N} \ , \tag{2.10a}$$

$$\frac{\partial}{\partial\tau}\eta_i=\mathcal{Q}\mathfrak{L}\mathcal{Q}\eta_i+\cos[\nu_i(t-\tau)] \times\left[\frac{1}{N}\frac{\partial V}{\partial r_i}+\tfrac{1}{2}\mathcal{Q}\left[\frac{\partial V}{\partial r_i}\rho+\rho\frac{\partial V}{\partial r_i}\right] -\rho\,\mathrm{Tr}\left[\rho\frac{\partial V}{\partial r_i}\right]\right] \ . \tag{2.10b}$$

Here $\mathfrak{L}(\mathbf{r})$ is the Liouville superoperator (including spontaneous emission) that generates the evolution of the internal state of an ion at point $\mathbf{r}$, and $\mathcal{P}$ and $\mathcal{Q}$ are superoperators whose action on an arbitrary internal-state operator o is given by

$$\mathcal{P}o=\frac{\mathbb{1}}{N}\mathrm{Tr}(o), \quad \mathcal{Q}o=o-\mathcal{P}o \ . \tag{2.11}$$

$\mathbb{1}$ is the unit operator for the internal degrees of freedom of the ion, N is the number of internal states included in the model, and $V(\mathbf{r})$ is the ion-field dipole interaction operator rendered independent of time by a suitable rotating-wave approximation. In Eqs. (2.10), arbitrary traceless initial conditions are assumed for the operators ρ and $\eta_{1,2,3}$ in the distant past $\tau=-\infty$. The operator that we denote by ρ is in fact the ordinary density operator made traceless by subtracting $\mathbb{1}/N$ from it.

The differences from the free atom are the different trajectory (2.9), and the cosine factor in Eq. (2.10b) whose origin was discussed around Eq. (2.7). The rest of the calculations proceed exactly as before: Once the solutions to (2.10) are found at the running time $\tau=t$, the force $\mathbf{F}(\mathbf{r},\mathbf{p})$ and the diffusion tensor $D_{ij}(\mathbf{r},\mathbf{p})$ are

$$F_i=-\mathrm{Tr}\left[\frac{\partial V}{\partial r_i}\rho\right] \ , \tag{2.12}$$

$$D_{ij}=\tfrac{1}{2}\mathrm{Tr}\left[\frac{\partial V}{\partial r_i}\eta_i+\frac{\partial V}{\partial r_j}\eta_i\right]+\mathrm{Tr}\left[\mathcal{S}^{ij}\left[\rho+\frac{\mathbb{1}}{N}\right]\right] \ . \tag{2.13}$$

The superoperators $\mathcal{S}^{ij}$ governing the fluctuations in the motion of the ion due to random directions of spontaneously emitted photons are given in Eq. (4.65) of I.

The conditions of validity of the SC theory are the same as before. First, as attested to by the appearance of finite-order momentum derivatives in the theory, we have replaced the discrete momentum transfer between the light and the ion with a (nearly) continuous flow. The momentum of a characteristic photon must therefore be much smaller than the characteristic momentum scale of the ion. Second, our derivation is an adiabatic elimination of the internal degrees of freedom, which are supposed to be slaved by the c.m. motion. There is one infinite time scale for the internal degrees of freedom of the ion corresponding to conservation of the population. We have explicitly taken care of it in the formalism. The condition then remains that the light-induced change in the motion of the ion during all *other* internal time scales must be "small." We emphasize that at this point no assumptions regarding the oscillation frequencies ν_i have been made.

B. Lamb-Dicke limit

As usual in laser-cooling theory of a trapped ion, a major simplification results when we assume that the ion stays in a region much smaller the wavelength λ of the exciting light. This is called the Lamb-Dicke limit. The ensuing expansions are the trapped-ion analogs of the velocity expansions pursued in Sec. II C of II.

In Eqs. (2.10) the right-hand side depends on the running time via the position dependence: $\mathfrak{L}=\mathfrak{L}(\mathbf{r}(\tau))$ and $V=V(\mathbf{r}(\tau))$. When the motion is confined to a region much smaller than the wavelength, we may expand these operators around the center of the trap $\mathbf{r}^0$. Using (2.9) we have

$$\mathfrak{L}(\mathbf{r}(\tau))=\mathfrak{L}(\mathbf{r}^0)+\sum_i\left[(r_i-r_i^0)\cos[\nu_i(\tau-t)] + \frac{p_i}{M\nu_i}\sin[\nu_i(\tau-t)]\right]\frac{\partial\mathfrak{L}(\mathbf{r}^0)}{\partial r_i^0} + \cdots \ . \tag{2.14}$$

The expansion parameter obviously is the ratio of the amplitude of the oscillations of the ion to the wavelength. Assuming a solution of (2.10a) in the form of a power series of this ratio,

$$\rho=\rho^0+\rho^1+\cdots \ , \tag{2.15}$$

and equating terms order by order, we find a succession of equations. The first two read

$$\frac{\partial}{\partial\tau}\rho^0=\mathcal{Q}\mathfrak{L}(\mathbf{r}^0)\mathcal{Q}\rho^0+\mathcal{Q}\mathfrak{L}(\mathbf{r}^0)\mathcal{P}\frac{\mathbb{1}}{N} \ , \tag{2.16a}$$

$$\frac{\partial}{\partial\tau}\rho^1=\mathcal{Q}\mathfrak{L}(\mathbf{r}^0)\mathcal{Q}\rho^1 +\sum_i\left[(r_i-r_i^0)\cos[\nu_i(\tau-t)] + \frac{p_i}{M\nu_i}\sin[\nu_i(\tau-t)]\right]\mathcal{Q}\frac{\partial\mathfrak{L}(\mathbf{r}^0)}{\partial r_i^0}\left[\rho^0+\frac{\mathbb{1}}{N}\right] \ . \tag{2.16b}$$

The solutions to Eqs. (2.16) at time $\tau=t$ with arbitrary traceless initial conditions are

$$\rho^0=-[\mathcal{Q}\mathfrak{L}(\mathbf{r}^0)\mathcal{Q}]^{-1}\mathcal{Q}\mathfrak{L}(\mathbf{r}^0)\mathcal{P}\frac{\mathbb{1}}{N}\ , \tag{2.17a}$$

$$\rho^1=-\sum_i \frac{p_i/M+(r_i-r_i^0)\mathcal{Q}\mathfrak{L}(\mathbf{r}^0)\mathcal{Q}}{[(\mathcal{Q}\mathfrak{L}(\mathbf{r}^0)\mathcal{Q})^2+\mathcal{Q}\nu_i^2]}\mathcal{Q}\frac{\partial\mathfrak{L}(\mathbf{r}^0)}{\partial r_i^0}\left[\rho^0+\frac{\mathbb{1}}{N}\right]\ . \tag{2.17b}$$

As usual (see II), the inverses are taken in the projection space of $\mathcal{Q}$.

To the lowest (second) nontrivial order in the expansion parameter, we also have to take into account the position dependence of the interaction operator,

$$\frac{\partial V}{\partial r_i}=\frac{\partial V(\mathbf{r}^0)}{\partial r_i^0}+\sum_j (r_j-r_j^0)\frac{\partial^2 V(\mathbf{r}^0)}{\partial r_i^0\partial r_j^0}+\cdots\ . \tag{2.18}$$

Combining (2.12), (2.17), and (2.18), we have the following expansion of the force to second order in the ratio of the oscillation amplitude to the wavelength:

$$F_i(\mathbf{r},\mathbf{p})=\phi_i(\mathbf{r}^0)-\sum_j \alpha_{ij}(\mathbf{r}^0)\frac{p_j}{M}-M\sum_j N_{ij}(\mathbf{r}^0)(r_j-r_j^0)\ . \tag{2.19}$$

Each term in the force merits a separate discussion. First,

$$\phi_i=-\mathrm{Tr}\left[\frac{\partial V}{\partial r_i}\rho^0\right] \tag{2.20}$$

is a constant force that can in principle be canceled by a suitable choice of the trapping electromagnetic fields. Henceforth, we usually ignore ϕ_i. Second, the matrix

$$\alpha_{ij}=-\mathrm{Tr}\left\{\frac{\partial V}{\partial r_i}[(\mathcal{Q}\mathfrak{L}\mathcal{Q})^2+\mathcal{Q}\nu_j^2]^{-1}\mathcal{Q}\frac{\partial\mathfrak{L}}{\partial r_j}\left[\rho^0+\frac{\mathbb{1}}{N}\right]\right\} \tag{2.21}$$

consisting of the derivatives of the force with respect to velocity at $\mathbf{v}=0$ is called the damping tensor. It indicates the presence of a velocity-dependent force that may be dissipative and therefore lead to cooling of the ion. Third, the force coefficient tensor

$$N_{ij}=\frac{1}{M}\left\{\mathrm{Tr}\left[\frac{\partial^2 V}{\partial r_i\partial r_j}\rho^0\right]-\mathrm{Tr}\left[\frac{\partial V}{\partial r_i}\frac{\mathcal{Q}\mathfrak{L}\mathcal{Q}}{[(\mathcal{Q}\mathfrak{L}\mathcal{Q})^2+\mathcal{Q}\nu_i^2]}\mathcal{Q}\frac{\partial\mathfrak{L}}{\partial r_j}\left[\rho^0+\frac{\mathbb{1}}{N}\right]\right]\right\} \tag{2.22}$$

implies that light-induced forces are present that are linear in the displacement from the trap center. The restoring forces responsible for the trapping are of the same form, so such forces lead to renormalizations of the mechanical oscillation frequencies of the ion. The principal axes of the trap may also be rotated. However, the tensor N_{ij} is of the order λ^{-2}, and the formal Lamb-Dicke limit $\lambda\to\infty$ implies that the renormalizations must be small compared to the original trapping frequencies which are independent of λ. Until further notice we therefore omit the renormalizations of the trap frequencies in our discussions. To shorten the formulas, in Eqs. (2.20)–(2.22) we have adopted the convention that all position-dependent quantities are calculated for the position of the center of the trap $\mathbf{r}^0$. We often resort to this convention without explicitly mentioning it.

To obtain a finite temperature, it suffices to calculate the diffusion only to zeroth order in the Lamb-Dicke parameter, for $\mathbf{r}=\mathbf{r}^0$, $\mathbf{p}=0$, as if the ion stood still at the center of the trap. Nevertheless, due to the explicit cosine factor in (2.10b), the result is not the same as for a free atom. Solving (2.10b) to zeroth order in the Lamb-Dicke parameter, we find

$$\eta_i^0=-\frac{\mathcal{Q}\mathfrak{L}\mathcal{Q}}{[(\mathcal{Q}\mathfrak{L}\mathcal{Q})^2+\mathcal{Q}\nu_i^2]}\left[\frac{1}{N}\frac{\partial V}{\partial r_i}+\tfrac{1}{2}\mathcal{Q}\left[\frac{\partial V}{\partial r_i}\rho^0+\rho^0\frac{\partial V}{\partial r_i}\right]-\rho^0\,\mathrm{Tr}\left[\rho^0\frac{\partial V}{\partial r_i}\right]\right]\ . \tag{2.23}$$

Combination of (2.17a), (2.23), and (2.13) now gives for the diffusion tensor the expression

$$D_{ij}=\tfrac{1}{2}\,\mathrm{Tr}\left[\frac{\partial V}{\partial r_i}\eta_j^0+\frac{\partial V}{\partial r_j}\eta_i^0\right]+\mathrm{Tr}\left[\mathcal{S}^{ij}\left[\rho^0+\frac{\mathbb{1}}{N}\right]\right]\ . \tag{2.24}$$

Equations (2.19)–(2.24) are our main results. However, before embarking on explicit calculations, we develop another slightly different expansion that is particularly useful in the analysis of polarization gradient cooling at low light intensities.

C. Adiabatic low-intensity limit

The most peculiar consequences of polarization-gradient cooling come to the open in the limit of low light intensity. Physically, an atom whose ground state is degenerate then has a long optical pumping time scale. This leads to a singular response to laser cooling.

Although it is possible to expand the quantities given in Eqs. (2.19)–(2.24) further into a power series in light intensity, in practice such expansions require degenerate perturbation theory and are awkward; see, e.g., [20]. Here we take a slightly different route that was already traveled with a free atom in III. We will use the same notation as in III, even though it partly clashes with the notation of I and II. In the present paper we may thus use the same symbol for two different objects. However, the conflicting notations do not appear simultaneously in any physical case discussed hereunder.

To begin with, we assume that the density operator ρ has unit trace, instead of zero trace as in Secs. II A and II B. However, the operators η_i still have zero trace. Also, no projection operators are introduced as yet. Noting that the ion-field interaction operator V is traceless, and that the Liouvillean $\mathfrak{L}$ preserves the trace of its argument operator, we may rewrite (2.10) as

$$\frac{\partial}{\partial\tau}\rho=\mathfrak{L}\rho\ , \tag{2.25a}$$

$$\frac{\partial}{\partial\tau}\eta_i=\mathfrak{L}\eta_i+\cos[\nu_i(t-\tau)]\left[\frac{1}{2}\left[\frac{\partial V}{\partial r_i}\rho+\rho\frac{\partial V}{\partial r_i}\right]-\rho\,\mathrm{Tr}\left[\rho\frac{\partial V}{\partial r_i}\right]\right]\ . \tag{2.25b}$$

The prescriptions to calculate force and diffusion, (2.12) and (2.13), keep their forms under the new convention, except that the $\mathbb{1}/N$ in (2.13) is to be dropped.

The projection superoperator $\mathcal{P}$ introduced for the present case will be different from the projector $\mathcal{P}$ of Eq. (2.11). For simplicity we assume that the ion has only two angular momentum degenerate levels g and e, and denote by P_g and P_e the orthogonal projections to the ground- and excited-state manifolds. At low intensity the populations of, and coherences between, the Zeeman substates of the ground level occupy a special position in the theory. After all, they are only subject to the slow evolution due to optical pumping. We therefore define the new projector $\mathcal{P}$ and its complement $\mathcal{Q}$ as

$$\mathcal{P}o=P_goP_g,\quad \mathcal{Q}o=o-\mathcal{P}o\ . \tag{2.26}$$

The projector $\mathcal{P}$ restricts the domain and range of its argument operator to the ground-state manifold. We also split the Liouvillean into two parts:

$$\mathfrak{L}=\mathfrak{L}^{(0)}+\mathfrak{L}^{(1)}\ , \tag{2.27}$$

where the superscript indicates order in field strength. $\mathfrak{L}^{(0)}$ conveys spontaneous damping and detuning, while the induced transitions are contained in $\mathfrak{L}^{(1)}$. The operator V is automatically of first order in field strength.

Next we write from Eq. (2.25a) the equations of motion for the projections $\mathcal{P}\rho$ and $\mathcal{Q}\rho$ using the field decomposition (2.27). The equation for $\mathcal{Q}\rho$ reads

$$\frac{\partial}{\partial\tau}\mathcal{Q}\rho=\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{P}\mathcal{P}\rho+(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q}+\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{Q})\mathcal{Q}\rho\ . \tag{2.28}$$

The projector $\mathcal{Q}\rho$ is subject to spontaneous damping contained in $\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q}$. We assume that the rate of spontaneous emission Γ and possibly the field-ion detuning δ make the fastest time scales in the problem. This implies a formal low-intensity limit, just as was the case for a free atom. However, for the trapped ion we must also require that the spontaneous decay rate of the excited level is much larger than the ionic oscillation frequencies,

$$\Gamma\gg\nu_i\ . \tag{2.29}$$

Contrary to the models of the preceding Secs. II A and II B, a condition on the oscillator frequencies is thereby built into our successive development. At any rate, with our assumptions we may (as in III) solve Eq. (2.28) adiabatically by setting the τ derivative equal to zero. We insert the result for $\mathcal{Q}\rho$ into the equation of the projection $\mathcal{P}\rho$, and expand to the lowest nontrivial (second) order in field strength. The result is

$$\frac{\partial}{\partial\tau}\mathcal{P}\rho=\mathcal{O}\mathcal{P}\rho\ , \tag{2.30}$$

with

$$\mathcal{O}=[\mathcal{P}\mathfrak{L}^{(0)}\mathcal{Q}(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{Q}-\mathcal{P}\mathfrak{L}^{(1)}\mathcal{Q}]\times(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{P}\ . \tag{2.31}$$

The inverses are once more carried out in the projection space of $\mathcal{Q}$.

A zero eigenvalue finally makes its appearance on an attempt to solve (2.30). Physically, optical pumping at a nonzero intensity leads to a (usually) unique steady-state density operator. The associated zero eigenvalue of $\mathcal{O}$ corresponds to conservation of population in the ground-state manifold. We therefore define yet another projector $\mathfrak{p}$ as

$$\mathfrak{p}o=\frac{\mathbb{1}}{n}\mathrm{Tr}(o)\ . \tag{2.32a}$$

In this case $\mathbb{1}$ is the unit operator within the ground-state manifold ($\mathbb{1}=P_g$), and n is the number of substates of the ground level ($n=\mathrm{Tr}P_g$). In our present approach $\mathfrak{p}$ is always applied within the projection space of $\mathcal{P}$, so the complement is defined as

$$\mathfrak{q}=\mathcal{P}-\mathfrak{p}\ . \tag{2.32b}$$

The superoperator $\mathfrak{q}\mathcal{O}\mathfrak{q}$ governs the optical pumping among the Zeeman states of the ground level.

The course for the calculation of the force is now clear. First, we solve Eq. (2.30) for the density operator $\mathcal{P}\rho$ of the ground-state manifold of the oscillating ion using the additional projectors $\mathfrak{p}$ and $\mathfrak{q}$, then resort to the adiabatic approximation to obtain the projection $\mathcal{Q}\rho$, and finally calculate the force from $\mathcal{Q}\rho$. During this process the ground-state evolution operator $\mathcal{O}$ is taken to be a function of position, and for an oscillating ion therefore a function of time τ. The technicalities of the analysis are similar to those in III and in Sec. II B above, and we only cite the results. First, we define the optically pumped density operator to zeroth order in field strength

$$\rho^{(0)}=[1-(\mathfrak{q}\mathcal{O}\mathfrak{q})^{-1}\mathfrak{q}\mathcal{O}\mathfrak{p}]\frac{\mathbb{1}}{n}\ . \tag{2.33}$$

The velocity-independent part of the force, and the damping and force coefficient tensors up to the lowest nontrivial (second) order in field strength, in the limit $\Gamma\gg\nu_i$, are

$$\phi_i=\mathrm{Tr}\left[\frac{\partial V}{\partial r_i}(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{P}\rho^{(0)}\right]\ , \tag{2.34}$$

$$\alpha_{ij}=\mathrm{Tr}\left\{\frac{\partial V}{\partial r_i}(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{P}[(\mathfrak{q}\mathcal{O}\mathfrak{q})^2+\mathfrak{q}\nu_j^2]^{-1}\mathfrak{q}\frac{\partial\mathcal{O}}{\partial r_j}\rho^{(0)}\right\}, \tag{2.35}$$

$$N_{ij}=-\frac{1}{M}\mathrm{Tr}\left\{\left[\frac{\partial}{\partial r_j}\left[\frac{\partial V}{\partial r_i}(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{P}\right]\right]\rho^{(0)}\right\}+\frac{1}{M}\mathrm{Tr}\left[\frac{\partial V}{\partial r_i}(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{P}\frac{\mathfrak{q}\mathcal{O}\mathfrak{q}}{[(\mathfrak{q}\mathcal{O}\mathfrak{q})^2+\mathfrak{q}\nu_j^2]}\mathfrak{q}\frac{\partial\mathcal{O}}{\partial r_j}\rho^{(0)}\right]. \tag{2.36}$$

While the expression for α_{ij} is not particularly transparent, juxtaposition with the free-atom result (obtained by setting $\nu_i=0$) is telling. The difference is the replacement

$$\frac{1}{(\mathfrak{q}\mathcal{O}\mathfrak{q})^2}\rightarrow\frac{1}{(\mathfrak{q}\mathcal{O}\mathfrak{q})^2+\mathfrak{q}\nu_j^2}. \tag{2.37}$$

This means that the time scales associated with optical pumping and with the mechanical oscillations of the ion are being compared.

The same comparison takes place with the diffusion tensor. As analyzed in detail in III, the operators η_i in (2.25b) have both a slow component that evolves on the optical pumping time scale, and a fast component that within our adiabatic assumption locks instantaneously to the inhomogeneous terms on the right-hand side of (2.25b). We employ the projectors $\mathcal{P}$ and $\mathcal{Q}$ to separate the slow and fast components in η_i. Correspondingly, we obtain an "anomalous" and a "normal" component in the diffusion.

At the interesting time $\tau=t$ the right-hand side of Eq. (2.25b) is the same as for a free atom. The normal diffusion is therefore the same for a trapped ion and a free atom. However, the anomalous diffusion depends directly on the comparison between the optical pumping time and the mechanical oscillation period of the ion. The technical difference from the free-atom case turns out to be the single replacement

$$\frac{1}{\mathfrak{q}\mathcal{O}\mathfrak{q}}\rightarrow\frac{\mathfrak{q}\mathcal{O}\mathfrak{q}}{(\mathfrak{q}\mathcal{O}\mathfrak{q})^2+\mathfrak{q}\nu_j^2} \tag{2.38}$$

in Eq. (4.13) of III. Nonetheless, for completeness we state here the entire algorithm for obtaining the diffusion tensor for a stationary ion ($\mathbf{r}=\mathbf{r}^0,\mathbf{p}=0$). While at that, we add some parentheses to the results of III to clarify details that might otherwise be misinterpreted.

First we complete Eq. (2.33) into basically an expansion of the steady-state density operator up to second order in field strength,

$$\begin{aligned}\rho^{(1)}&=-(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{P}\rho^{(0)},\\ \rho^{(2)}&=-(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{Q}\rho^{(1)}.\end{aligned} \tag{2.39}$$

Next, we define the expansion in field strength of the inhomogeneous terms in (2.25b):

$$\begin{aligned}\mathcal{P}R_i^{(2)}&=\tfrac{1}{2}\mathcal{P}\left[\frac{\partial V}{\partial r_i}(\mathcal{Q}\rho^{(1)})+(\mathcal{Q}\rho^{(1)})\frac{\partial V}{\partial r_i}\right]\\&\quad-\mathrm{Tr}\left[\frac{\partial V}{\partial r_i}\rho^{(1)}\right]\mathcal{P}\rho^{(0)},\\ \mathcal{Q}R_i^{(1)}&=\tfrac{1}{2}\mathcal{Q}\left[\frac{\partial V}{\partial r_i}(\mathcal{P}\rho^{(0)})+(\mathcal{P}\rho^{(0)})\frac{\partial V}{\partial r_i}\right],\\ \mathcal{Q}R_i^{(2)}&=\tfrac{1}{2}\mathcal{Q}\left[\frac{\partial V}{\partial r_i}(\mathcal{Q}\rho^{(1)})+(\mathcal{Q}\rho^{(1)})\frac{\partial V}{\partial r_i}\right].\end{aligned} \tag{2.40}$$

The lowest-order contributions in field strength to the operators η_i as determined by optical pumping may now be written

$$\mathcal{P}\eta_i^{(0)}=\frac{\mathfrak{q}\mathcal{O}\mathfrak{q}}{(\mathfrak{q}\mathcal{O}\mathfrak{q})^2+\mathfrak{q}\nu_i^2}\mathfrak{q}\{[\mathcal{P}\mathfrak{L}^{(1)}\mathcal{Q}-\mathcal{P}\mathfrak{L}^{(0)}\mathcal{Q}(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{Q}](\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}R_i^{(1)}+\mathcal{P}\mathfrak{L}^{(0)}\mathcal{Q}(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}R_i^{(2)}-\mathcal{P}R_i^{(2)}\}. \tag{2.41}$$

The diffusion tensor to second order in field strength and to zeroth order in the ratio of the ionic oscillation amplitude to the wavelength of light, in the limit $\Gamma\gg\nu_i$, finally is

$$\begin{aligned}D_{ij}&=-\tfrac{1}{2}\mathrm{Tr}\left[\frac{\partial V}{\partial r_i}(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{P}\mathcal{P}\eta_j^{(0)}+\frac{\partial V}{\partial r_j}(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}\mathfrak{L}^{(1)}\mathcal{P}\mathcal{P}\eta_i^{(0)}\right]\\&\quad-\tfrac{1}{2}\mathrm{Tr}\left[\frac{\partial V}{\partial r_i}(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}R_j^{(1)}+\frac{\partial V}{\partial r_j}(\mathcal{Q}\mathfrak{L}^{(0)}\mathcal{Q})^{-1}\mathcal{Q}R_i^{(1)}\right]+\mathrm{Tr}(\mathcal{S}^{ij}\rho^{(2)}).\end{aligned} \tag{2.42}$$

The first term describes the anomalous diffusion, the second term the normal diffusion, and the third term the diffusion due to the angular distribution of spontaneously emitted photons.

III. EXPLICIT RESULTS

A. Two-state ion

For the sake of illustration we develop in some detail the simplest possible special case, a two-state ion in a 1D trap driven by a traveling wave, using the Lamb-Dicke formalism of Sec. II B. By assumption there is only one direction in the problem, call it z. The center of the trap is correspondingly denoted by z^0. We take a traveling wave of the form $\mathcal{E}\cos(kz-\omega t)$. Given the dipole moment matrix element $\mathcal{D}$ and the resonance frequency ω_0, the ion-field interaction is parametrized by Rabi frequency $\Omega=\mathcal{D}\mathcal{E}/2\hbar$ and detuning $\delta=\omega-\omega_0$. The spontaneous decay rate of the excited state of the ion $|2\rangle$ back to the ground state $|1\rangle$ is Γ, which implies the dipole damping rate $\gamma=\Gamma/2$.

We represent an arbitrary internal-state operator o as a four-dimensional column vector consisting of the matrix elements in the basis $\{|1\rangle,|2\rangle\}$ in the following order:

$$o=[o_{11},o_{12},o_{21},o_{22}]^T . \tag{3.1}$$

Besides possessing the obvious linear algebra, these operators inherit the product and the trace of 2×2 matrices:

$$uv=[u_{11}v_{11}+u_{12}v_{21},\ldots]^T , \tag{3.2a}$$

$$\mathrm{Tr}(o)=o_{11}+o_{22} . \tag{3.2b}$$

In particular, the dipole interaction operator is

$$V(z)=\hbar[0,-\Omega e^{-ikz},-\Omega e^{ikz},0]^T . \tag{3.3}$$

With these conventions superoperators, linear operators acting on operators, become 4×4 matrices. The premier one is the Liouvillian

$$\mathfrak{L}(z)=\begin{bmatrix} 0 & -i\Omega e^{ikz} & i\Omega e^{-ikz} & \Gamma \\ -i\Omega e^{-ikz} & -i\delta-\gamma & 0 & i\Omega e^{-ikz} \\ i\Omega e^{ikz} & 0 & i\delta-\gamma & -i\Omega e^{ikz} \\ 0 & i\Omega e^{ikz} & -i\Omega e^{-ikz} & -\Gamma \end{bmatrix} , \tag{3.4}$$

while the nonzero elements of the projector $\mathcal{P}$ are

$$\mathcal{P}_{11}=\mathcal{P}_{14}=\mathcal{P}_{41}=\mathcal{P}_{44}=\tfrac{1}{2} . \tag{3.5}$$

As it comes to spontaneously emitted photons, only one superoperator $\mathcal{S}^{zz}$ is relevant. The matrix representing $\mathcal{S}^{zz}$ has precisely one nonzero element, namely

$$\mathcal{S}^{zz}_{14}=\frac{\beta}{2} . \tag{3.6}$$

Here β is a constant that depends on the angular distribution of spontaneous photons. Typical values are $\beta=\frac{1}{3}$ for hypothetical isotropic spontaneous emission, and $\beta=\frac{2}{5}$ ($\frac{3}{10}$) that applies to a $j=0\to j=1$ transition driven by linearly (circularly) polarized light [4,24].

The final technicality is the inverses of operators restricted to the projection space of $\mathcal{Q}$. For $\Gamma\neq0$ the null space of $\mathcal{Q}$ is spanned by the vector $[1,0,0,1]^T$, and the nonzero elements of this null space are also the only vectors outside the range of the mapping $\mathfrak{L}$. This implies that the projection space of $\mathcal{Q}$ consists precisely of the traceless operators, and that $\mathfrak{L}$ is one to one if restricted to act on traceless operators. Given a traceless operator o, the operator $p=(\mathcal{Q}\mathfrak{L}\mathcal{Q})^{-1}o$ is therefore the unique traceless solution to the equation $\mathcal{L}p=o$. In practice we solve for p by making sure that o is traceless, then replacing the first element of o by zero and the first row of the superoperator $\mathfrak{L}$ by [1,0,0,1], and finally tackling the ensuing set of equations without any restrictions remaining. In short, one of the linear equations to be solved for p is replaced by the condition that p is traceless.

Obtaining the quantities in Eqs. (2.21)–(2.24) is now a plug-in-the-formula assignment, albeit tedious. We employ MATHEMATICA [25] in our calculations. All tensors only have one relevant component. We drop the directional indices, and present the results as rational expressions of the single trapping frequency remaining in the problem, ν:

$$\alpha=\frac{-4\hbar\delta\gamma k^2\Omega^2[4\gamma^2(\delta^2+\gamma^2+2\Omega^2)+\nu^2(\delta^2+\gamma^2+6\Omega^2)]}{d} , \tag{3.7}$$

$$N=\frac{2\hbar\delta k^2\Omega^2\nu^2[4\gamma^2(\delta^2-3\gamma^2)+\nu^2(\delta^2-7\gamma^2+4\Omega^2)]}{Md} , \tag{3.8}$$

$$\begin{aligned} D=\hbar^2\gamma k^2\Omega^2\{&4\gamma^2[\delta^4+2\delta^2\gamma^2+\gamma^4+6\delta^2\Omega^2-2\gamma^2\Omega^2+4\Omega^4+\beta(\delta^4+2\delta^2\gamma^2+\gamma^4+4\delta^2\Omega^2+4\gamma^2\Omega^2+4\Omega^4)] \\ &+\nu^2[\delta^4+6\delta^2\gamma^2+5\gamma^4+10\delta^2\Omega^2-6\gamma^2\Omega^2+16\Omega^4+\beta(\delta^4-6\delta^2\gamma^2+9\gamma^4+8\delta^2\Omega^2+8\gamma^2\Omega^2+16\Omega^4)] \\ &+\nu^4[\delta^2+\gamma^2+2\Omega^2+\beta(-2\delta^2+6\gamma^2-8\Omega^2)]+\beta\nu^6\}d^{-1} , \end{aligned} \tag{3.9}$$

where the common denominator is

$$d=(\delta^2+\gamma^2+2\Omega^2)\{4\gamma^2(\delta^2+\gamma^2+2\Omega^2)^2+\nu^2(\delta^4-6\delta^2\gamma^2+9\gamma^4+8\delta^2\Omega^2+8\gamma^2\Omega^2+16\Omega^4)-2\nu^4(\delta^2-3\gamma^2+4\Omega^2)+\nu^6\} . \tag{3.10}$$

A standard analysis of the Fokker-Planck equation shows that under the present assumptions, namely constant diffusion and velocity-linearized force, the stationary state of laser cooling is thermal, and is characterized by the total c.m. energy $E=D/\alpha$. Our present expressions yield an energy that coincides with the thoroughly verified known result for this system [15–19]. Similarly, to the lowest order in the Lamb-Dicke parameter the correction to the force coefficient embodied in N leads to the renormalization of the trap frequency of the ion in agreement with Lindberg and Stenholm [15].

Here we expand on the renormalization of the oscillator frequency. The total position-dependent restoring force on the ion may be written

$$F=-M(\nu^2+N)(z-z^0)$$
$$\equiv -M[\nu(1+\xi)]^2(z-z^0)\ , \tag{3.11}$$

where the fractional change in the oscillation frequency is defined as

$$\xi=\sqrt{1+N/\nu^2}-1\ . \tag{3.12}$$

We take as an example the experimentally quite common case in which the trapping frequency ν is small compared to the linewidth of the transition γ. The parameter ξ then reads

$$\xi=\left[1+\frac{4\delta\gamma(\delta^2-3\gamma^2)\Omega^2}{(\delta^2+\gamma^2+2\Omega^2)^3}\frac{\varepsilon_R}{\gamma}\right]^{1/2}-1\ . \tag{3.13}$$

Here

$$\varepsilon_R=\frac{\hbar k^2}{2M} \tag{3.14}$$

is the recoil frequency. For an asymptotically small ratio ε_R/γ the largest fractional frequency renormalization occurs at $\delta/\gamma=-0.505$, $\Omega/\gamma=0.560$, and equals $\xi=0.130\,\varepsilon_R/\gamma$.

A final note about the implications of the Lamb-Dicke limit on the renormalization of the trapping frequency of a two-state ion is useful for contrast with the corresponding result for polarization-gradient cooling, to be described below. Let us take the quantum-mechanical length scale

$$l=(\hbar/2M\nu)^{1/2} \tag{3.15}$$

as the lowest limit of the size of the ion cloud, then by the Lamb-Dicke limit $l/\lambda\ll 1$. Moreover, the limit underlying Eq. (3.13) implies that $\nu/\gamma\ll 1$, hence $(l/\lambda)^2\nu/\gamma\sim\varepsilon_R/\gamma\ll 1$. Expression (3.13) is consistent with the Lamb-Dicke limit only if $\varepsilon_R/\gamma\ll 1$, and hence the renormalization must satisfy $\xi\ll 1$. Of course, this does not automatically mean that the renormalization cannot be observed in an experiment.

B. Polarization-gradient cooling

As our second example we take up polarization-gradient cooling of a $j=\frac{1}{2}\rightarrow j=\frac{3}{2}$ ion in one dimensional lin ⊥ lin molasses [23]. We now use the adiabatic low-intensity formalism of Sec. II C. Again only one direction of oscillations is assumed available, call it z, and the corresponding trapping frequency is denoted by ν.

The laser field consists of two counterpropagating waves in the z direction, one of which is linearly polarized in the x direction and the other in the y direction. We write the electric field as

$$\mathbf{E}(z,t)=\frac{\mathcal{E}}{2}(\hat{\mathbf{e}}_x e^{i(kz-\omega t)}+\hat{\mathbf{e}}_y e^{i(-kz-\omega t)}+\text{c.c.})\ . \tag{3.16}$$

The field is linearly polarized at the position $z=0$, circularly polarized at $z=\pm\lambda/8$, and so forth. Given the dipole moment matrix element for a transition whose Clebsch-Gordan coefficient equals unity, $\mathfrak{D}$, we define Rabi frequency Ω, saturation parameter s_0, and optical pumping rate r through

$$\Omega=\frac{\mathfrak{D}\mathcal{E}}{2\hbar},\quad s_0=\frac{2\Omega^2}{\delta^2+\Gamma^2/4},\quad r=\frac{2\Gamma s_0}{9}\ . \tag{3.17}$$

We also define a scaled detuning Δ (positive is the laser is tuned below resonance) and a parameter c that conveys the position of the trap center in the field,

$$\Delta=-\delta/\Gamma,\quad c=\cos(4kz^0)\ . \tag{3.18}$$

This problem was studied earlier with a free atom in III, where we also discussed the implementation of the calculations using MATHEMATICA. It turned out that the superoperator $\mathfrak{q}\mathcal{O}\mathfrak{q}$ is basically one-dimensional. In fact, independent of the position z, the effect of $\mathfrak{q}\mathcal{O}\mathfrak{q}$ is essentially to multiply its argument by the negative of the optical pumping rate r. Going over to the trapped ion, the key replacements (2.37) and (2.38) therefore boil down to the same operation: The damping coefficient and the anomalous diffusion coefficient are to be multiplied by the factor $r^2/(r^2+\nu^2)$. The rest of the calculations proceed as before.

Unfortunately, enough details were not published in III to allow one to write down the results by inspection. We produce the missing details here. The damping coefficient for a trapped ion is

$$\frac{\alpha}{\hbar k^2}=\frac{3r^2\Delta(1+c)}{\nu^2+r^2}\ , \tag{3.19}$$

and the diffusion coefficient is

$$\frac{D}{\hbar^2k^2r}=\frac{9(1+c)^2\Delta^2r^2}{4(\nu^2+r^2)}+\frac{3(3+c)}{8}+\frac{29-7c}{40}\ . \tag{3.20}$$

In (3.20) the three terms are the anomalous, the normal, and the spontaneous diffusion, in the same order as in (2.42). Once more the steady state of cooling is thermal, and could be characterized by a temperature. However, we prefer to specify the steady state in terms of the total energy of the c.m. harmonic oscillator expressed in units of an oscillator quantum. The result is

$$\frac{E}{\hbar\nu}=\frac{(37+4c)(r^2+\nu^2)+45\Delta^2r^2(1+c)^2}{60\Delta r\nu(1+c)}\ . \tag{3.21}$$

We first analyze the cooling energy as a function of the optical pumping rate, keeping other parameters fixed. In practice this would most closely correspond to varying the laser intensity. The minimum occurs at

$$\frac{r_m}{\nu}=\left[\frac{37+4c}{37+4c+45\Delta^2(1+c)^2}\right]^{1/2}, \tag{3.22}$$

and equals

$$\frac{E_m}{\hbar\nu}=\frac{\sqrt{(37+4c)[37+4c+45\Delta^2(1+c)^2]}}{30\Delta(1+c)} . \tag{3.23}$$

For, say, $\Delta=-\delta/\Gamma=2$ and $c=1$ (for a position at which the light is linearly polarized), we have the minimum $E_m=1.47\hbar\nu$ at $r_m=0.23\nu$. On an attempt at further minimization it becomes evident that the only dependence on detuning in (3.23) enters through the expression

$$x=\Delta(1+c) , \tag{3.24}$$

and that (3.23) is a monotonically decreasing function of x. It follows that (3.23) has no minimum, but instead a monotonic approach to the lowest limit $E_m/\hbar\nu=(\frac{33}{20})^{1/2}=1.28$ as $c\to-1$ and simultaneously $\Delta\to\infty$ in such a way that $x\to\infty$.

A process with $\delta\to-\infty$, etc., is not possible experimentally, but in 1D lin $\perp$ lin molasses a $j_1=\frac{1}{2}\to j_2=\frac{3}{2}$ ion might nevertheless be cooled until its excitation energy $E-\frac{1}{2}\hbar\nu$ is less than one oscillator quantum above the zero-point energy.

The optical pumping rate giving a minimum of temperature may be understood as reflecting a balance between optical pumping and mechanical oscillations of the ion. In the case $\nu\ll r$ the oscillations of the center of mass take place on a much longer time scale than optical pumping. For an ion with the present velocity v optical pumping is essentially instantaneous, and past velocities do not affect the internal state. The cooling takes place similarly to a free atom, except that the ion is confined to the neighborhood of the trap center. Suppose one then attempts to lower the temperature, as with a free atom, by decreasing the intensity; i.e., by decreasing the optical pumping rate r. By the time one has reached the case $\nu\gg r$ the ion oscillates back and forth many times during an optical pumping time, and many oscillation cycles are averaged over while the ion integrates its equations of motion to arrive at the internal state. The force therefore displays only a weak dependence on the *current* velocity of the ion. The damping coefficient $\alpha\sim F(v)/v$ is small, and the c.m. energy $E=D/\alpha$ is large. Obviously there must be a minimum of temperature somewhere around $r_m\sim\nu$.

The fractional renormalization of the oscillator frequency reads

$$\xi=\left[1-\frac{6\Delta r[2r^2c+\nu^2(c-1)]}{\nu(r^2+\nu^2)}\frac{\varepsilon_R}{\nu}\right]^{1/2}-1 . \tag{3.25}$$

Here $\varepsilon_R/\nu\sim(l/\lambda)^2$ is small by virtue of the Lamb-Dicke limit, but the multiplier of ε_R/ν need not be bounded. It appears that $|\xi|\sim1$ is possible even though the ion is in the Lamb-Dicke regime. The renormalizations of the trapping frequencies might be more prominent in polarization gradient cooling than in Doppler cooling.

IV. DISCUSSION

We have presented a semiclassical approach to laser cooling of a trapped multistate ion in an arbitrary light field, and developed in detail both the Lamb-Dicke limit and the additional low-intensity limit of the theory. For a two-state ion we have verified agreement with known results. This should not come as a surprise: The approach of I and II was a descendant of our earlier theory of Doppler cooling of a two-state ion [16], and in a manner of speaking we have simply closed the circle. For polarization-gradient cooling, insights have been gained. Basically, the competition between optical pumping and mechanical oscillations sets a limit on cooling. In our example the limit was such that the excitation energy of the ion is about one vibrational quantum above the zero-point energy.

In our final Fokker-Planck equation the c.m. motion is treated classically. The ion simply is subjected to a harmonic restoring force, a light pressure force, and a classical stochastic diffusion. The diffusion succeeds remarkably well, in that the present SC theory even respects the zero-point energy limit of a harmonic oscillator. We have pondered on this earlier [26] in the context of a two-state ion. Having adapted those discussions to the present case, we have come to believe that quantum mechanics apparently enters with the explicit cosine factor in Eq. (2.10b). Without the cosine the motion of the ion would contribute solely through the classical trajectory; with the cosine there is a piece of global knowledge about the nature of particle trajectories present in Eq. (2.10b). Were it not for the cosine, the SC theory would readily violate the zero-point energy.

Incidentally, the widely employed semiheuristic analysis of diffusion [2,7] initiated by Gordon and Ashkin [27] would miss the cosine factor. While this approach seems to work for a free atom, it fails dramatically when applied to a trapped ion.

Semiclassical cooling theory of a *two-state* ion not only respects the zero-point energy, but an even stronger statement holds true: The cooling temperature from our SC Lamb-Dicke calculations exactly coincides with the result of a fully quantized analysis based on an expansion in the parameter ε_R/ν [15]. In Sec. III A we have furthermore shown that the same applies to the light-induced renormalization of the c.m. oscillator frequency. In the remainder of the paper we consider the possible agreement between our SC approach and the eventual quantum theories in regard to polarization-gradient cooling.

We recall the basic conditions of validity of the SC theory: (i) the momentum scale of the c.m. motion must be larger than the momentum of a characteristic photon, and (ii) the internal equilibration time of the ion must be short compared to the time scale over which the c.m. motion changes appreciably. The first condition is readily satisfied in the Lamb-Dicke limit. If the length scale l of the c.m. motion has to be much smaller than the wavelength λ, then the corresponding momentum scales satisfy $\hbar/l\gg\hbar/\lambda$. Using the quantum-mechanical length scale as the size parameter l, we have an alternative inequality

$$\frac{\varepsilon_R}{\nu} \ll 1 \ . \tag{4.1}$$

The second condition is more subtle. Given the friction coefficient α of the c.m. motion for the $j=\frac{1}{2}\rightarrow j=\frac{3}{2}$ ion in 1D lin $\perp$ lin molasses, Eq. (3.19), the damping rate is α/M. The rate of optical pumping must thus satisfy $r \gg \alpha/M$. With the estimate $r \sim \nu$ we have the qualitative condition

$$\frac{\varepsilon_R |\delta|}{\nu\Gamma} \ll 1 \ . \tag{4.2}$$

Inequality (4.2) need not be satisfied even though (4.1) is. However, in a theory in which the ratio ε_R/ν is taken to be *asymptotically* small, (4.1) and (4.2) are both satisfied.

All told, we conjecture that our SC theory is equivalent to an expansion of the full quantum theory to the lowest nontrivial order in the parameter ε_R/ν. Strange as it may sound for an approach phrased in terms of classical particle trajectories, our cooling limit of the order of one quantum above the zero-point energy should have predictive power. We speculate that a qualitatively similar limit also applies to polarization-gradient cooling of ions with other $j\rightarrow j+1$ transitions and with other types of polarization gradients.

Note added in proof. After the present paper was submitted, an article by Cirac *et al.* [28] on polarization-gradient cooling of a trapped ion appeared. To the extent that we have made a comparison, their results agree with ours.

ACKNOWLEDGMENTS

This work is supported in part by the National Science Foundation and the Academy of Finland.

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