

## Spontaneous recoil effects of optical pumping on trapped atoms

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(Received 8 August 2008; published 14 October 2008)

The recoil effects of spontaneous photon emissions during optical pumping of a trapped three-level atom are exactly calculated. Without resort to the Lamb-Dicke approximation, and considering arbitrary detuning and saturation of the pump laser, the density of recoil shifts in phase space is derived. It is shown that this density is not of Gaussian shape, and that it becomes isotropic in phase space only for a branching ratio corresponding to fluorescence scattering but unfavorable for optical pumping. The dependence of its anisotropy on the laser saturation is discussed in the resonant case, and the mapping of moments of the atom's center-of-mass motion due to the pumping is presented. Moreover, it is shown how optimum parameters for protecting the center-of-mass quantum state from pump-induced disturbance depend on the specific property to be protected.

DOI: [10.1103/PhysRevA.78.043412](https://doi.org/10.1103/PhysRevA.78.043412)

PACS number(s): 37.10.Vz, 37.10.Ty, 32.80.Xx

### I. INTRODUCTION

Optical pumping (OP) [1,2] has proved to be a very powerful method for preparing specific electronic states of atoms. By application of a near resonant light field the atoms are excited and subsequently decay into the desired electronic state via spontaneous emission. In bulk media such as a solid, or a gas of atoms or molecules, optical pumping can be understood solely in terms of electronic or optical properties of the medium. Although in dense gases modifications arise due to atom-atom interactions mediated by the radiation field, the mechanical effects of optical pumping may be safely neglected in such situations, since momentum is transferred to the collective sample. However, any light interaction with free or weakly bound atoms not only is accompanied by a change of the internal electronic quantum state of the atoms, but unavoidably also affects their center-of-mass motion [3,4]. Examples of this type of interaction are found with the momentum diffusion in fluorescence scattering of free atoms [5–9], with the laser cooling of trapped atoms [10–14] or ions [15,16]. The mechanical effects are particularly apparent when we consider the interaction of a laser field with an individual particle, e.g., with a single trapped ion [17], with single atoms confined in a microcavity [18,19], as well as in atom optics [20] and laser-control of atoms [21]. When OP is applied to such a system, one has explicitly to take into account the mechanical effects of the laser-atom interaction. Sometimes, as with OP of low-lying vibronic states of weakly bound trapped ions, the radiative recoil generates a desired effect: cooling of the sample. However, more often than not, the mechanical effect modifies the momentum or energy distribution of the sample and the concomitant radiative spectrum in an undesirable way. In particular, the incoherent nature of spontaneous emission makes it add decoherence [22] to the quantum state of the center-of-mass motion of the atoms. In view of the possible application of such systems to quantum-state engineering [23,24], quantum logic, and computing [25–27], the introduction of any such decoherence is obviously problematic. Thus, the identification and avoidance of decoherent effects is of utmost importance.

From the experimental viewpoint, the optimum parameters must be found in order to obtain the smallest decoherence. Given a certain atomic species confined in a trapping potential, several parameters are predetermined such as the electronic transition frequencies, the relaxation rates, and the trap frequency. Apart from modifying the trap, the adjustable parameters are the laser intensity and detuning. One may consider it convenient to use fast optical pumping, faster than the vibrational period, since the negligible center-of-mass motion improves the efficiency of the pump process. On the other hand, the required laser intensity may saturate and broaden the employed atomic transition, which spoils OP's resolution of vibronic states. Thus, the interplay of saturation and pumping time requires close inspection.

The recoil upon atoms interacting with light has been treated in various contexts, such as, for example, in the light interaction of free [9] and trapped single atoms [28] or in the collective atomic recoil laser [29,30]. The applied models consider an atomic two-level system continuously irradiated by near-resonant monochromatic light. Such a treatment of free atoms leads to a Gaussian momentum distribution in accordance with the central limit theorem applied to the statistically independent individual photon recoils [8]. For trapped atoms this situation is different since here the waiting-time distribution of subsequent spontaneous photon emission correlates the individual photon recoils in phase space, and the validity of the central-limit theorem is doubtful. Additionally, OP requires a third atomic level, a lower target state. The process is intrinsically terminated by the spontaneous decay into this state. Thus, contributions with a small number of radiative events may dominate the interaction. The recoil effects of these contributions make the resulting distribution of the atom in vibrational phase space substantially deviate from being Gaussian—the more so, the less is the number of interaction events with the pump light.

In this paper, we address a single atom trapped in a harmonic potential. We model the motion of the atom's center of mass by a one-dimensional (1D) quantum-mechanical oscillation along a certain direction. This model can be viewed as a prototype system that provides us with the essential information to be applied to one of the real systems mentioned above. Using a quantum trajectory approach we obtain ana-

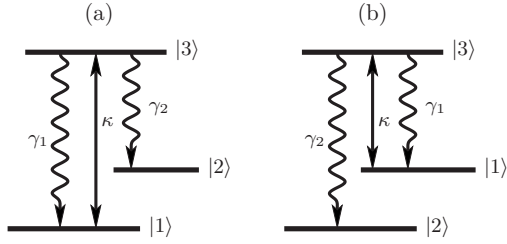


FIG. 1. Draft excitation schemes for the inversion of the two-level system  $|1\rangle \rightarrow |2\rangle$  by optical pumping via the third auxiliary level  $|3\rangle$ . Scheme (b) is identical to (a) with the ground state being replaced by a metastable state.

lytic results for the quantum statistical properties of the atomic center-of-mass motion. Moreover, we obtain insight into the decoherence properties by employing a phase-spatial picture, that lends to illustrative interpretation.

The method of quantum trajectories [31–37] is perfectly matched to studying OP with proper inclusion of recoil effects, and avoiding the Lamb-Dicke approximation: First, the trail of spontaneously emitted photons of a single experimental realization is finite, since a stationary state is reached when the system is no longer affected by the laser drive. This fact simplifies the structure of single quantum trajectories. Second, when using waiting-time distributions, an easy connection is made between the center-of-mass vibration and the statistics of photon emissions. Finally, an elegant formulation of recoil effects is found that characterizes the process of optical pumping, from the viewpoint of the vibrational motion, by just a single distribution function.

In Sec. II we introduce the model system under consideration and develop its evolution as a quantum trajectory in phase space. Section III outlines the vibronic state of the pumped atom in terms of the density of recoil shifts in phase space. This density is characterized in Sec. IV by its statistical moments that are expressed by quantities derived from photon-counting statistics. Section V is then devoted to a discussion of the results in limiting cases, such as fluorescence scattering and maximally anisotropic scattering in phase space, as well as to optimum laser parameters in order to minimize detrimental recoil effects of OP. Section VI places the results in the context of possible observations. Finally, Sec. VII provides a summary and conclusions.

## II. QUANTUM TRAJECTORIES OF OPTICAL PUMPING

### A. Quantum master equation

Our generic model for treating the motional effects of OP consists of a  $\Lambda$ -type three-level atom whose center-of-mass coordinate is bound in a harmonic trap potential. In this way, photonic-recoil effects can be described for various systems, such as single ions in rf traps or neutral atoms in optical-dipole or magneto-optical traps. The level scheme and setup of optical pumping is shown in Fig. 1.

In both equivalent schemes, a laser field is assumed to drive the electronic dipole transition  $|1\rangle \leftrightarrow |3\rangle$ , whereupon photons are spontaneously emitted connected with electronic dipole relaxation from the auxiliary level  $|3\rangle$  to both  $|1\rangle$  and

$|2\rangle$ . The latter process occurs only once, since in state  $|2\rangle$  the atom decouples from the driving laser and thus has reached its final electronic state. Both level schemes, Figs. 1(a) and 1(b), include pumping of population from the electronic state  $|1\rangle$  to state  $|2\rangle$ , with the corresponding direct transition being dipole forbidden. The optical-pumping rate is determined by the electronic relaxation rates  $\gamma_1$  and  $\gamma_2$  of the two dipole transitions, cf. Fig. 1, and by the laser Rabi frequency  $\kappa = -2dE/\hbar$ . Here,  $d$  is the electronic dipole moment of the transition  $|1\rangle \leftrightarrow |3\rangle$ , and  $E$  is the electric-field amplitude of the laser.

The subject to be addressed is the atom's center-of-mass vibration at frequency  $\nu$  along one principal axis of the trapping potential. Furthermore, only the motional recoil effects of spontaneous photon emissions are of interest here. The additional recoil effects due to the laser interaction have been discussed elsewhere [38] and can be separated by choosing the propagation direction of the laser beam perpendicular to the direction of motion under consideration. This motion may be thought to extend along the axis of a linear trap, while the orthogonal confinement of the particle be “strong,” such that the trap absorbs the momentum transmitted by the laser.

The dynamics of the vibronic density operator  $\hat{\rho}(t)$  is given by the quantum master equation [38]

$$\begin{aligned} \frac{\partial \hat{\rho}}{\partial t} = & \frac{1}{i\hbar} [\hat{H}_0 + \hat{V}(t), \hat{\rho}] \\ & + \sum_{a=1,2} \gamma_a \left[ \hat{\sigma}_{a,-} (\hat{\mathcal{R}}_a[\hat{\rho}]) \hat{\sigma}_{a,+} - \frac{1}{2} \{ \hat{\sigma}_{a,+} \hat{\sigma}_{a,-}, \hat{\rho} \} \right], \end{aligned} \quad (1)$$

where  $\{\hat{A}, \hat{B}\} = \hat{A}\hat{B} + \hat{B}\hat{A}$  denotes the anticommutator, and  $\hat{H}_0$  describes the free time evolution of the atom in the trap potential [49],

$$\hat{H}_0 = \hbar \nu \hat{b}^\dagger \hat{b} + \sum_{a=1,2} 2\hbar \omega_a \hat{\sigma}_{a,z}, \quad (2)$$

with  $\omega_a$  ( $a=1,2$ ) being the electronic transition frequencies. Moreover,  $\hat{b}$  and  $\hat{b}^\dagger$  are the annihilation and creation operators for the vibration along the chosen principal axis of the trap, and the electronic spin-type operators are given by

$$\hat{\sigma}_{a,-} = |a\rangle\langle 3|, \quad \hat{\sigma}_{a,+} = \hat{\sigma}_{a,-}^\dagger, \quad (3)$$

$$\sigma_{a,z} = \frac{1}{2} (|3\rangle\langle 3| - |a\rangle\langle a|). \quad (4)$$

Finally, the external drive of the laser with frequency  $\omega$  is described by the Hamiltonian

$$\hat{V}(t) = \frac{1}{2} \hbar \kappa \hat{\sigma}_{1,+} e^{-i\omega t} + \text{H.c.}, \quad (5)$$

with  $\kappa = 2\mathbf{d}_{31} \cdot \mathbf{E}/\hbar$  being the Rabi frequency,  $\mathbf{d}_{31} = \langle 3|\hat{\mathbf{d}}|1\rangle$  being the transition dipole moment, and  $\mathbf{E}$  being the electric-field amplitude of the laser.

In Eq. (1) the superoperators  $\hat{\mathcal{R}}_a$  are responsible for the momentum recoil due to spontaneous emission of a photon via the electronic decay channels  $|3\rangle \rightarrow |a\rangle$  ( $a=1,2$ ). They act on an arbitrary operator  $\hat{A}$  as

$$\hat{\mathcal{R}}_a[\hat{A}] = \int_{-1}^1 ds \mu_a(s) e^{isk_a \hat{x}} \hat{A} e^{-isk_a \hat{x}} \quad (a=1,2), \quad (6)$$

with the respective photon-momentum recoil  $\hbar k_a = \hbar \omega_a / c$  ( $a=1,2$ ) being projected on the considered axis according to the normalized dipole radiation characteristics,

$$\mu_a(s) = \frac{3}{8\pi} \int_0^{2\pi} d\phi \{1 - [\mathbf{n}_a \cdot \mathbf{n}(\Omega)]^2\}, \quad (7)$$

where  $s = \cos \theta$  is the projection of the photon-emission direction on the considered motional axis. The unit-normalized vectors  $\mathbf{n}_a$  and  $\mathbf{n}(\Omega)$  point into the directions of the transition dipole moment  $\langle 3|\hat{\mathbf{d}}|a\rangle$  and of the spontaneous emission  $\Omega = (\theta, \phi)$ , respectively. Assuming, for example, transition dipole moments orthogonal to the direction of motion under consideration, Eq. (7) reduces to

$$\mu_a(s) = \frac{3}{8}(1 + s^2). \quad (8)$$

The strength of the recoil effect of individual spontaneous photon emissions can be characterized by Lamb-Dicke parameters: The position operator  $\hat{x}$  is expressed in terms of the annihilation and creation operators as  $k_a \hat{x} = \eta_a (\hat{b} + \hat{b}^\dagger)$  ( $a=1,2$ ). The Lamb-Dicke parameters  $\eta_a$  of the corresponding transitions  $|3\rangle \rightarrow |a\rangle$  are defined as

$$\eta_a = k_a \Delta x_0 \quad (a=1,2), \quad (9)$$

where  $\Delta x_0 = \sqrt{\hbar / (2m\nu)}$  is the rms spread of the position of the atom of mass  $m$  in the ground state  $|0\rangle$  of the trapping potential.

To proceed, the master equation is transformed into a frame, whose center-of-mass part oscillates with the trap frequency  $\nu$  and whose electronic part rotates with the laser frequency  $\omega$ . The transformed master equation is written in terms of three superoperators,

$$\frac{\partial \hat{\rho}}{\partial t} = [\hat{\mathcal{N}} + \hat{\mathcal{J}}_1(t) + \hat{\mathcal{J}}_2(t)][\hat{\rho}], \quad (10)$$

where the evolution in the absence of spontaneous photon emissions is described by

$$\hat{\mathcal{N}}[\hat{\rho}] = \frac{1}{i\hbar} (\hat{H}_{\text{eff}} \hat{\rho} - \hat{\rho} \hat{H}_{\text{eff}}^\dagger). \quad (11)$$

The occurring non-Hermitian effective Hamiltonian reads

$$\hat{H}_{\text{eff}} = \frac{\hbar}{2} (\kappa \hat{\sigma}_{1,+} + \kappa^* \hat{\sigma}_{1,-}) + \hbar \Delta |1\rangle \langle 1| - \frac{i\hbar}{2} \sum_{a=1,2} \gamma_a \hat{\sigma}_{a,+} \hat{\sigma}_{a,-}, \quad (12)$$

with  $\Delta = \omega - \omega_1$  being the detuning of the laser.

The jump operators  $\hat{\mathcal{J}}_a(t)$  describe spontaneous photon emissions via the transitions  $|3\rangle \rightarrow |a\rangle$  ( $a=1,2$ ) and are given by

$$\hat{\mathcal{J}}_a(t)[\hat{\rho}] = \gamma_a \hat{S}_{a,-} [\hat{\mathcal{R}}_a(t)[\hat{\rho}]] \quad (i=1,2), \quad (13)$$

where the spin superoperator reads

$$\hat{S}_{a,-}[\hat{\rho}] = \hat{\sigma}_{a,-} \hat{\rho} \hat{\sigma}_{a,+}, \quad (14)$$

and the now time-dependent recoil operators read

$$\hat{\mathcal{R}}_a(t)[\hat{\rho}] = \int_{-1}^1 ds \mu_a(s) \hat{D}(i\eta_a s e^{i\nu t})[\hat{\rho}]. \quad (15)$$

The action of the right-hand side superoperator is defined as

$$\hat{D}(\alpha)[\hat{\rho}] = \hat{D}(\alpha) \hat{\rho} \hat{D}^\dagger(\alpha), \quad (16)$$

with the coherent displacement operator,

$$\hat{D}(\alpha) = \exp(\alpha \hat{a}^\dagger - \alpha^* \hat{a}). \quad (17)$$

Thus, in Eq. (15) the length of the shift in motional phase space depends on both the Lamb-Dicke parameter  $\eta_a$  and the direction of photon emission  $s$ , whereas the phase of the shift is determined by  $\pi/2 + \nu t$ , i.e., it depends on the time of the spontaneous emission event.

## B. Formal solution of the propagator

The formal solution of the master equation (10) in the rotating frame reads

$$\hat{\rho}(t) = \hat{\mathcal{M}}_{012}(t, t_0)[\hat{\rho}(t_0)], \quad (18)$$

where  $\hat{\rho}(t_0)$  is the initial density operator, and the propagator is given by the time-ordered expression

$$\hat{\mathcal{M}}_{012}(t, t_0) = \mathcal{T} \exp \left\{ \int_{t_0}^t d\tau [\hat{\mathcal{N}} + \hat{\mathcal{J}}_1(\tau) + \hat{\mathcal{J}}_2(\tau)] \right\}. \quad (19)$$

Equation (19) can be rewritten as a sum over all possible trajectories where the spontaneous emissions  $|3\rangle \rightarrow |2\rangle$ , i.e., the operator  $\hat{\mathcal{J}}_2(t)$ , occurs at the random times  $t_1, \dots, t_n$  [50],

$$\begin{aligned} & \hat{\mathcal{M}}_{012}(t, t_0) \\ &= \sum_{n=0}^{\infty} \int_{t_0}^t dt_n \dots \int_{t_0}^{t_2} dt_1 \hat{\mathcal{M}}_{01}(t, t_n) \hat{\mathcal{J}}_2(t_n) \hat{\mathcal{M}}_{01}(t_n, t_{n-1}) \dots \\ & \quad \times \hat{\mathcal{J}}_2(t_1) \hat{\mathcal{M}}_{01}(t_1, t_0). \end{aligned} \quad (20)$$

The operators  $\hat{\mathcal{M}}_{01}(t, t')$  determine the time evolution between the spontaneous emissions  $|3\rangle \rightarrow |2\rangle$  and contain both the laser drive and the spontaneous emissions  $|3\rangle \rightarrow |1\rangle$ ,

$$\hat{\mathcal{M}}_{01}(t, t') = \mathcal{T} \exp \left\{ \int_{t'}^t d\tau [\hat{\mathcal{N}} + \hat{\mathcal{J}}_1(\tau)] \right\}. \quad (21)$$

After a spontaneous emission  $|3\rangle \rightarrow |2\rangle$  at a time  $t_k$ , the atom is in its final (pumped) electronic state  $|2\rangle$  where it

decouples from the laser drive, and thus  $\hat{\mathcal{M}}_{01}(t_{k+1}, t_k) \hat{\mathcal{J}}_2(t_k) = \hat{\mathcal{J}}_2(t_k)$ . Since  $\hat{\mathcal{J}}_2(t_{k+1}) \hat{\mathcal{J}}_2(t_k) = 0$  a second spontaneous emission is impossible—it would require the electronic state to be  $|3\rangle$ . Accordingly the propagator (20) reduces to only two parts,

$$\hat{\mathcal{M}}_{012}(t, t_0) = \hat{\mathcal{M}}_{01}(t, t_0) + \int_{t_0}^t dt' \hat{\mathcal{J}}_2(t') \hat{\mathcal{M}}_{01}(t', t_0). \quad (22)$$

The first term on the right-hand side in Eq. (22) corresponds to the quantum trajectory when no spontaneous transition from state  $|3\rangle$  to  $|2\rangle$  occurred. The second term represents the trajectory with one final spontaneous transition  $|3\rangle \rightarrow |2\rangle$ . Only one such spontaneous transition can take place.

Defining now the projection operators into the electronic states  $|a\rangle$ ,

$$\hat{\mathcal{P}}_a[\hat{\mathcal{Q}}] = (\hat{\sigma}_{a,-} \hat{\sigma}_{a,+}) \hat{\mathcal{Q}} (\hat{\sigma}_{a,-} \hat{\sigma}_{a,+}), \quad (23)$$

and using the facts that (i) the projection into the pumped state  $|2\rangle$  is not affected by the dynamics on the transition  $|3\rangle \leftrightarrow |1\rangle$ , that is  $\hat{\mathcal{P}}_2 \hat{\mathcal{M}}_{01}(t, t') = \hat{\mathcal{P}}_2$ , and that (ii)  $\hat{\mathcal{P}}_2 \hat{\mathcal{J}}_2(t') = \hat{\mathcal{J}}_2(t')$ , the part of the propagator taking the atom to the pumped electronic state  $|2\rangle$  is obtained from Eq. (22) as

$$\hat{\mathcal{P}}_2 \hat{\mathcal{M}}_{012}(t, t_0) = \hat{\mathcal{P}}_2 + \int_{t_0}^t dt' \hat{\mathcal{J}}_2(t') \hat{\mathcal{M}}_{01}(t', t_0). \quad (24)$$

The first term represents the initial population in level  $|2\rangle$ , and the second one the effect of the pumping.

Analogous to Eq. (20) the propagator  $\hat{\mathcal{M}}_{01}(t, t')$  can be formally rewritten as a sum of sequences of spontaneous processes  $|3\rangle \rightarrow |1\rangle$ . Denoting by  $t_n = t'$  the time of the final spontaneous emission that leads into the electronic state  $|2\rangle$ , Eq. (24) thus can be rewritten as

$$\begin{aligned} \hat{\mathcal{P}}_2 \hat{\mathcal{M}}_{012}(t, t_0) &= \hat{\mathcal{P}}_2 + \sum_{n=1}^{\infty} \int_{t_0}^t dt_n \dots \int_{t_0}^{t_2} dt_1 \hat{\mathcal{J}}_2(t_n) \hat{\mathcal{M}}_0(t_n, t_{n-1}) \\ &\quad \times \hat{\mathcal{J}}_1(t_{n-1}) \dots \hat{\mathcal{J}}_1(t_1) \hat{\mathcal{M}}_0(t_1, t_0). \end{aligned} \quad (25)$$

Here the operator  $\hat{\mathcal{M}}_0(t, t')$  describes the evolution exclusive of any spontaneous emission,

$$\hat{\mathcal{M}}_0(t, t') = \exp[\hat{\mathcal{N}}(t - t')]. \quad (26)$$

A nonvanishing action of  $\hat{\mathcal{J}}_1(t_1)$  requires population in level  $|3\rangle$ . Since  $\hat{\mathcal{M}}_0(t_1, t_0)$  does only couple levels  $|1\rangle$  and  $|3\rangle$ , the second term in Eq. (25) only acts on populations in levels  $|1\rangle$  and  $|3\rangle$ . Assuming zero initial population of  $\hat{\mathcal{Q}}(t_0)$  in the rapidly decaying state  $|3\rangle$ , the action is restricted to population in level  $|1\rangle$ , and we may thus multiply the second term in Eq. (25) on its right-hand side with the projector  $\hat{\mathcal{P}}_1$ ,

$$\begin{aligned} \hat{\mathcal{P}}_2 \hat{\mathcal{M}}_{012}(t, t_0) &= \hat{\mathcal{P}}_2 + \sum_{n=1}^{\infty} \int_{t_0}^t dt_n \dots \int_{t_0}^{t_2} dt_1 \hat{\mathcal{J}}_2(t_n) \hat{\mathcal{M}}_0(t_n, t_{n-1}) \\ &\quad \times \hat{\mathcal{J}}_1(t_{n-1}) \dots \hat{\mathcal{M}}_0(t_1, t_0) \hat{\mathcal{P}}_1. \end{aligned} \quad (27)$$

The second term describes sequences of  $n$  consecutive spontaneous transitions. The laser interaction is restricted to  $n$  excitation processes from level  $|1\rangle$  to level  $|3\rangle$ , as shown by the sequences of operators  $\hat{\mathcal{M}}_0$  alternating with jump operators  $\hat{\mathcal{J}}_1 \propto \hat{\mathcal{S}}_{1,-} = |1\rangle\langle 3| \dots |3\rangle\langle 1|$ , see Eqs. (3), (4), and (14). Each of these excitation processes results therefore in the waiting-time distribution [39–42]

$$\begin{aligned} w(t - t') &= (\gamma_1 + \gamma_2) \langle 3 | \{ \hat{\mathcal{M}}_0(t, t') [ |1\rangle\langle 1| ] \} | 3 \rangle \\ &= (\gamma_1 + \gamma_2) | \langle 3 | \hat{U}_{\text{eff}}(t - t') | 1 \rangle |^2, \end{aligned} \quad (28)$$

obtained as the probability (density) of the atom being in level  $|3\rangle$  at time  $t$ , on the condition that it was in  $|1\rangle$  at time  $t'$ , and that no photons have been emitted in the interval  $[t', t]$ . Here we have defined  $\hat{U}_{\text{eff}}(t) = \exp(-i\hat{H}_{\text{eff}}t/\hbar)$ . The waiting-time distribution is calculated for resonant pumping in Appendix D. Using definition (28) the formal solution (27) can therefore be written as

$$\begin{aligned} \hat{\mathcal{P}}_2 \hat{\mathcal{M}}_{012}(t, t_0) &= \hat{\mathcal{P}}_2 + \sum_{n=1}^{\infty} \int_{t_0}^t dt_n \dots \int_{t_0}^{t_2} dt_1 \\ &\quad \times [\lambda_2 \hat{\mathcal{R}}_2(t_n) w(t_n - t_{n-1})] \\ &\quad \times \{ [\lambda_1 \hat{\mathcal{R}}_1(t_{n-1}) w(t_{n-1} - t_{n-2})] \dots \\ &\quad \times [\lambda_1 \hat{\mathcal{R}}_1(t_1) w(t_1 - t_0)] \} \hat{\mathcal{P}}_{21} \end{aligned} \quad (29)$$

where

$$\lambda_a = \gamma_a / (\gamma_1 + \gamma_2) \quad (a = 1, 2) \quad (30)$$

are the branching ratios, and the operator that induces the transition into the final level  $|2\rangle$  is defined as

$$\hat{\mathcal{R}}_{21}[\hat{\mathcal{Q}}] = (\hat{\sigma}_{2,-} \hat{\sigma}_{1,+}) \hat{\mathcal{Q}} (\hat{\sigma}_{1,-} \hat{\sigma}_{2,+}). \quad (31)$$

Defining the joint probability density for  $n$  spontaneous transitions at times  $t_1, \dots, t_n$  as

$$\begin{aligned} w_n(t_n, \dots, t_1) &= \lambda_2 \lambda_1^{n-1} w(t_n - t_{n-1}) \dots w(t_2 - t_1) \\ &\quad \times w(t_1 - t_0) \quad (n > 0), \end{aligned} \quad (32)$$

the result (29) is rewritten as

$$\begin{aligned} \hat{\mathcal{P}}_2 \hat{\mathcal{M}}_{012}(t, t_0) &= \hat{\mathcal{P}}_2 + \sum_{n=1}^{\infty} \int_{t_0}^t dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \hat{\mathcal{R}}_2(t_n) \\ &\quad \times \hat{\mathcal{R}}_1(t_{n-1}) \dots \hat{\mathcal{R}}_1(t_1) \hat{\mathcal{P}}_{21}. \end{aligned} \quad (33)$$

### III. THE VIBRATIONAL EFFECTS OF THE OPTICAL TRANSITIONS

In Eq. (33), a sequence of recoil operators, cf. Eq. (15), appears, which can be combined as

$$\begin{aligned} \hat{\mathcal{R}}_2(t_n) \dots \hat{\mathcal{R}}_1(t_1) &= \int ds_n \mu_2(s_n) \dots \int ds_1 \mu_1(s_1) \\ &\quad \times \hat{\mathcal{D}}(i\eta_2 s_n e^{i\nu t_n}) \\ &\quad \times \hat{\mathcal{D}}(i\eta_1 s_{n-1} e^{i\nu t_{n-1}}) \dots \hat{\mathcal{D}}(i\eta_1 s_1 e^{i\nu t_1}) \end{aligned}$$

$$= \int ds_n \mu_2(s_n) \dots \int ds_1 \mu_1(s_1) \times \hat{D}[\alpha_n(\{t_n, s_n\}, \dots, \{t_1, s_1\})], \quad (34)$$

where the total recoil shift is

$$\alpha_n(\{t_n, s_n\}, \dots, \{t_1, s_1\}) = i\eta_2 s_n e^{i\nu t_n} + \sum_{m=1}^{n-1} i\eta_1 s_m e^{i\nu t_m}. \quad (35)$$

The  $n$ -fold integration represents the averaging over the directions of recoil in the  $n$  consecutive spontaneous emissions at times  $t_1, \dots, t_n$ . In order to consider any number of spontaneous emissions at arbitrary moments of time within the interval  $[t_0, t]$ , we must average over the joint photon-emission probability at the consecutive emission times. In this way we obtain the operator of the mean recoil shift in phase space,

$$\begin{aligned} \hat{D}(t, t_0) &= \sum_{n=1}^{\infty} \int_{t_0}^t dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\ &\quad \times \hat{\mathcal{R}}_2(t_n) \hat{\mathcal{R}}_1(t_{n-1}) \dots \hat{\mathcal{R}}_1(t_1) \\ &= \sum_{n=1}^{\infty} \int_{t_0}^t dt_n \int ds_n \mu_2(s_n) \dots \\ &\quad \times \int_{t_0}^{t_2} dt_1 \int ds_1 \mu_1(s_1) w_n(t_n, \dots, t_1) \\ &\quad \times \hat{D}[\alpha_n(\{t_n, s_n\}, \dots, \{t_1, s_1\})]. \end{aligned} \quad (36)$$

One may define the density of recoil shifts in phase space that contains all the recoil effects of the spontaneous emissions at random times into random directions,

$$\begin{aligned} p(\alpha; t, t_0) &= \sum_{n=1}^{\infty} \int_{t_0}^t dt_n \int ds_n \dots \int_{t_0}^{t_2} dt_1 \int ds_1 \\ &\quad \times \mu_2(s_n) \mu_1(s_{n-1}) \dots \mu_1(s_1) w_n(t_n, \dots, t_1) \\ &\quad \times \delta[\alpha - \alpha_n(\{t_n, s_n\}, \dots, \{t_1, s_1\})]. \end{aligned} \quad (37)$$

The operator of the mean recoil shift in phase space (36) is expressed as an integral over  $\hat{D}$  weighted by this distribution  $p(\alpha; t, t_0)$  in phase space:

$$\hat{D}(t, t_0) = \int d^2 \alpha p(\alpha; t, t_0) \hat{D}(\alpha). \quad (38)$$

Since  $p(\alpha; t, t_0)$  is an integral over positive probabilities, it may be viewed as a blurring distribution that smoothes the initial vibrational phase-space distribution. It contains all the characteristics of the random process that acts on the center-of-mass degree of freedom of the electronic population that is transferred from state  $|1\rangle$  to state  $|2\rangle$ .

In the propagator of Eq. (33), leading to the pumped electronic state  $|2\rangle$ , the second term on the right-hand side contains the averaged displacement as given in Eq. (36), and thus Eq. (38) may be employed to yield

$$\hat{\mathcal{P}}_2 \hat{\mathcal{M}}_{012}(t, t_0) = \hat{\mathcal{P}}_2 + \int d^2 \alpha p(\alpha; t, t_0) \hat{D}(\alpha) \hat{\mathcal{P}}_{21}. \quad (39)$$

Applying Eq. (39) on the initial vibronic density operator  $\hat{\rho}(t_0)$  and tracing over the electronic degree of freedom, we obtain the reduced motional density operator for the population in the pumped electronic state,

$$\langle 2 | \hat{\rho}(t) | 2 \rangle = \langle 2 | \hat{\rho}(t_0) | 2 \rangle + \int d^2 \alpha p(\alpha; t, t_0) \hat{D}(\alpha) [\langle 1 | \hat{\rho}(t_0) | 1 \rangle]. \quad (40)$$

A vibrational density operator  $\langle a | \hat{\rho} | a \rangle$ , that corresponds to the population in electronic level  $|a\rangle$ , is related to the  $s$ -ordered phase-space distribution

$$P_a^{(s)}(\alpha) = \frac{1}{P_a} \text{Tr}[\hat{\delta}^{(s)}(\alpha - \hat{b}) \langle a | \hat{\rho} | a \rangle] \quad (a = 1, 2), \quad (41)$$

where the  $s$ -ordered  $\delta$  operator is formally defined via the displacement operator (17) as [43,44]

$$\hat{\delta}^{(s)}(\alpha - \hat{b}) = \frac{1}{\pi^2} \int d^2 \beta \exp(\alpha \beta^* - \beta \alpha^* + s|\beta|^2/2) \hat{D}(\beta), \quad (42)$$

and the probability to find the atom in the electronic state  $|a\rangle$  is defined by

$$P_a = \text{Tr}(\langle a | \hat{\rho} | a \rangle). \quad (43)$$

We want to finally come up with a mapping of the initial vibrational phase-space distribution on the distribution generated by the optical pumping process. For this purpose we apply Eq. (41) on the mapping of vibrational density operators, Eq. (40), making use of the definition (16) and obtain

$$\begin{aligned} P_2^{(s)}(\alpha, t) &= P_2(t_0) P_2^{(s)}(\alpha, t_0) + \int d^2 \alpha' p(\alpha'; t, t_0) \\ &\quad \times \text{Tr}[\hat{\delta}^{(s)}(\alpha - \hat{b}) \hat{D}(\alpha') \langle 1 | \hat{\rho}(t_0) | 1 \rangle \hat{D}^\dagger(\alpha')]. \end{aligned} \quad (44)$$

Employing the cyclic property of the trace, one rewrites the trace as

$$\begin{aligned} &\text{Tr}[\hat{D}^\dagger(\alpha') \hat{\delta}^{(s)}(\alpha - \hat{b}) \hat{D}(\alpha') \langle 1 | \hat{\rho}(t_0) | 1 \rangle] \\ &= \text{Tr}\{\hat{\delta}^{(s)}[\alpha - (\hat{b} + \alpha')] \langle 1 | \hat{\rho}(t_0) | 1 \rangle\}, \end{aligned} \quad (45)$$

where the displacement operation results in a shift of the vibrational operator  $\hat{b} \rightarrow \hat{b} + \alpha'$ . This result, with substitution of integration variable  $\alpha' \rightarrow \alpha - \alpha'$  in Eq. (44), yields

$$\begin{aligned} P_2^{(s)}(\alpha, t) &= P_2(t_0) P_2^{(s)}(\alpha, t_0) + \int d^2 \alpha' p(\alpha - \alpha'; t, t_0) \\ &\quad \times \text{Tr}\{\hat{\delta}^{(s)}[\alpha' - \hat{b}] \langle 1 | \hat{\rho}(t_0) | 1 \rangle\}. \end{aligned} \quad (46)$$

With the use of definition (41) this phase-space distribution writes

$$P_2^{(s)}(\alpha, t) = P_2(t_0)P_2^{(s)}(\alpha, t_0) + P_1(t_0) \int d^2\alpha' p(\alpha - \alpha'; t, t_0)P_1^{(s)}(\alpha', t_0). \quad (47)$$

Thus the initial phase-space distribution of the population in electronic level  $|1\rangle$  is mapped onto a contribution to the phase-space distribution of the pumped level  $|2\rangle$  via a convolution with the density of recoil shifts in phase-space (37).

#### IV. THE PHASE SPACE DENSITY OF RECOIL SHIFTS

##### A. Moments of the density of recoil shifts

The density  $p$  of recoil shifts in phase space multiplied by an infinitesimal phase-space volume,  $p(\alpha; t, t_0)d^2\alpha$ , is the probability of displacing, by  $\alpha$ , the initial phase-space distribution within the time interval  $[t_0, t]$ . The observable features of the collective recoil shift will be characterized by this density's statistical moments of all orders  $(k, l)$ ,

$$\langle \alpha^{*k} \alpha^l \rangle_{p(\alpha; t, t_0)} = \int d^2\alpha \alpha^{*k} \alpha^l p(\alpha; t, t_0). \quad (48)$$

To evaluate these moments we may employ the characteristic function  $\underline{p}(\alpha, t, t_0)$ , which is the Fourier transform of the density of recoil shifts,

$$\underline{p}(\alpha; t, t_0) = \int d^2\beta \exp(\alpha\beta^* - \beta\alpha^*)p(\beta; t, t_0). \quad (49)$$

With the help of this characteristic function the statistical moments, Eq. (48), are obtained as [45]

$$\langle \alpha^{*k} \alpha^l \rangle_{p(\alpha; t, t_0)} = (-1)^l [\partial_{\alpha^*}^k \partial_{\alpha}^l \underline{p}(\alpha, t, t_0)]_{\alpha=0}. \quad (50)$$

Inserting Eq. (37) into Eq. (49) and performing the integral over  $\beta$  yields

$$\begin{aligned} \underline{p}(\alpha; t, t_0) &= \sum_{n=1}^{\infty} \int_{t_0}^t dt_n \int ds_n \dots \int_{t_0}^{t_2} dt_1 \int ds_1 \\ &\times \mu_2(s_n) \mu_1(s_{n-1}) \dots \mu(s_1) w_n(t_n, \dots, t_1) \\ &\times \exp[\alpha \alpha_n^* (\{t_n, s_n\}, \dots, \{t_1, s_1\}) - \text{c.c.}], \end{aligned} \quad (51)$$

where  $\alpha_n(\{t_n, s_n\}, \dots, \{t_1, s_1\})$  is defined in Eq. (35). With this definition, the exponential factor writes

$$\begin{aligned} &\exp[\alpha \alpha_n^* (\{t_n, s_n\}, \dots, \{t_1, s_1\}) - \text{c.c.}] \\ &= \exp \left[ \alpha \left( i\eta_2 s_n e^{i\nu t_n} + \sum_{m=1}^{n-1} i\eta_1 s_m e^{i\nu t_m} \right)^* - \text{c.c.} \right] \\ &= \exp(-\alpha i \eta_2 s_n e^{-i\nu t_n} - \text{c.c.}) \\ &\times \prod_{m=1}^{n-1} \exp(-\alpha i \eta_1 s_m e^{-i\nu t_m} - \text{c.c.}). \end{aligned} \quad (52)$$

Each exponential factor on the right-hand side can be rewritten as

$$\begin{aligned} \exp(-\alpha i \eta s e^{-i\nu t} - \text{c.c.}) &= \exp[-2i\eta s |\alpha| \text{Re}(e^{i\nu t - i \arg \alpha})] \\ &= \exp[-2i\eta s |\alpha| \cos(\nu t - \arg \alpha)], \end{aligned} \quad (53)$$

so that Eq. (51) reads

$$\begin{aligned} \underline{p}(\alpha; t, t_0) &= \sum_{n=1}^{\infty} \int_{t_0}^t dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\ &\times \int ds_n \mu_2(s_n) e^{-2i\eta_2 s_n |\alpha| \cos(\nu t_n - \arg \alpha)} \\ &\times \int ds_{n-1} \mu_1(s_{n-1}) e^{-2i\eta_1 s_{n-1} |\alpha| \cos(\nu t_{n-1} - \arg \alpha)} \dots \\ &\times \int ds_1 \mu_1(s_1) e^{-2i\eta_1 s_1 |\alpha| \cos(\nu t_1 - \arg \alpha)}. \end{aligned} \quad (54)$$

Each integral over a dipole characteristics is a directional spectrum of recoils; it is formally expressed by the Fourier transform

$$\underline{\mu}_a(\xi) = \int_{-1}^1 ds \mu_a(s) \exp(-is\xi). \quad (55)$$

In Eq. (54) this directional spectrum appears  $n$  times with the arguments  $2\eta_a |\alpha| \cos(\nu t_m - \arg \alpha)$ , where  $m=1, \dots, n$ , and  $a=1, 2$  depending on the decay channel. Each of these spectra defines a characteristic function associated to the instantaneous probability density for a single recoil shift at time  $t$ ,

$$\underline{\mu}_a(\alpha, t) = \underline{\mu}_a[2\eta_a |\alpha| \cos(\nu t - \arg \alpha)] \quad (a=1, 2). \quad (56)$$

With these replacements Eq. (54) takes on the form

$$\begin{aligned} \underline{p}(\alpha; t, t_0) &= \sum_{n=1}^{\infty} \int_{t_0}^t dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\ &\times \underline{p}_2(\alpha, t_n) \underline{p}_1(\alpha, t_{n-1}) \dots \underline{p}_1(\alpha, t_1). \end{aligned} \quad (57)$$

The moments of the density of recoil shifts are evaluated according to Eq. (50). The characteristic function (57) depends on  $\alpha$  via the factors  $\underline{p}_a(\alpha, t)$ . The derivatives of these factors (Appendix A) are

$$[\partial_{\alpha^*}^k \partial_{\alpha}^l \underline{p}_a(\alpha, t)]_{\alpha=0} = (-i\eta_a)^{k+l} e^{i\nu t(l-k)} \langle (\cos \theta)^{k+l} \rangle_a \quad (58)$$

with  $a=1, 2$ . The moments of the projection of the spontaneously emitted photon's wave vector on the dipole axis are

$$\langle (\cos \theta)^k \rangle_a = \int_{-1}^1 ds s^k \mu_a(s). \quad (59)$$

Since the dipole radiation characteristics shows even spatial symmetry,  $\mu_a(-s) = \mu_a(s)$ , cf. Eq. (A4), all odd moments vanish,  $\langle (\cos \theta)^{2k+1} \rangle_a = 0$  ( $k=0, 1, 2, \dots$ ). In consequence all odd-order derivatives of  $\underline{p}_a(\beta, t)$  vanish, see Eq. (58), and via Eq. (57) the odd-order moments of  $p$  vanish likewise,

$$\langle \alpha^{*k} \alpha^l \rangle_{p(\alpha; t, t_0)} = 0 \quad \text{if } k+l = 2n+1. \quad (60)$$

This result could have been anticipated, since the incoherent scattering process connected with optical pumping cannot generate a coherent vibrational amplitude.

The nonvanishing *even* moments of a complete optical-pumping process ( $t \rightarrow \infty$ ) are

$$\begin{aligned} \langle \alpha^{*k} \alpha^l \rangle_p &= (-1)^l \sum_{n=1}^{\infty} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\ &\times \{ \partial_{\alpha}^k \partial_{\alpha^*}^l [ \underline{p}_2(\alpha, t_n) \underline{p}_1(\alpha, t_{n-1}) \dots \underline{p}_1(\alpha, t_1) ] \}_{\alpha=0}. \end{aligned} \quad (61)$$

For the lowest-order nontrivial even moment  $\langle \alpha^* \alpha \rangle_p$  two derivatives have to be performed on any same factor  $\underline{p}_a(\alpha, t)$  which results in a sum of  $n$  terms, where the first one reads

$$\begin{aligned} - \sum_{n=1}^{\infty} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\ \times \{ [ \partial_{\alpha} \partial_{\alpha^*} \underline{p}_2(\alpha, t_n) ] \underline{p}_1(\alpha, t_{n-1}) \dots \underline{p}_1(\alpha, t_1) \}_{\alpha=0}. \end{aligned} \quad (62)$$

Given that  $\underline{p}_a(0, t) = \mu_a(0) = 1$  [Eqs. (55) and (56)] and  $[ \partial_{\alpha} \partial_{\alpha^*} \underline{p}_2(\alpha, t_n) ]_{\alpha=0} = -\eta_2^2 \langle (\cos \theta)^2 \rangle_2$ , the above term is

$$\begin{aligned} \eta_2^2 \langle (\cos \theta)^2 \rangle_2 \sum_{n=1}^{\infty} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\ = \eta_2^2 \langle (\cos \theta)^2 \rangle_2 \sum_{n=1}^{\infty} P_n = \eta_2^2 \langle (\cos \theta)^2 \rangle_2. \end{aligned} \quad (63)$$

Here the statistics of the number of spontaneously emitted photons during the pump process is introduced as time-ordered integral of the joint probability density of photon emissions,

$$P_n = \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \quad (n \geq 1), \quad (64)$$

and  $P_0 = 0$ . The moments of this photon statistics are evaluated in Appendix B.

The sum of the other  $n-1$  terms is

$$\begin{aligned} - \sum_{n=1}^{\infty} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \sum_{p=1}^{n-1} \{ \underline{p}_2(\alpha, t_n) \\ \times \underline{p}_1(\alpha, t_{n-1}) \dots [ \partial_{\alpha} \partial_{\alpha^*} \underline{p}_1(\alpha, t_p) ] \dots \underline{p}_1(\alpha, t_1) \}_{\alpha=0}. \end{aligned} \quad (65)$$

As the differentiated factor is  $-\eta_1^2 \langle (\cos \theta)^2 \rangle_1$  in all  $n-1$  terms, they sum up to yield

$$\begin{aligned} \eta_1^2 \langle (\cos \theta)^2 \rangle_1 \sum_{n=1}^{\infty} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) (n-1) \\ = \eta_1^2 \langle (\cos \theta)^2 \rangle_1 \sum_{n=1}^{\infty} P_n (n-1) = \eta_1^2 \langle (\cos \theta)^2 \rangle_1 (\langle \hat{n}_{\text{ph}} \rangle - 1), \end{aligned} \quad (66)$$

where the mean number of emitted photons is given in Eq. (B4). Thus, the lowest-order nontrivial moment of the density of recoil shifts is

$$\langle \alpha^* \alpha \rangle_p = \eta_2^2 \langle (\cos \theta)^2 \rangle_2 + \eta_1^2 (\langle \hat{n}_{\text{ph}} \rangle - 1) \langle (\cos \theta)^2 \rangle_1. \quad (67)$$

Upon insertion of the mean number of spontaneously emitted photons, see Eq. (B4), this moment becomes

$$\bar{n}_p = \langle \alpha^* \alpha \rangle_p = \eta_2^2 \langle (\cos \theta)^2 \rangle_2 + \frac{\lambda_1}{\lambda_2} \eta_1^2 \langle (\cos \theta)^2 \rangle_1, \quad (68)$$

where the mean recoil-induced excitation  $\bar{n}_p = \langle \alpha^* \alpha \rangle_p$  is introduced. This moment is the width of the density distribution of recoil shifts to which the system has been subjected in the optical pumping process.

In the same order one obtains the moment  $\langle \alpha^2 \rangle_p$  which gives information on the rotational symmetry of the density of recoil shifts in phase space. Analogously to the above calculation one obtains this moment—see Appendix C—as

$$\langle \alpha^2 \rangle_p = - \left( \eta_2^2 \langle (\cos \theta)^2 \rangle_2 + \frac{\lambda_1}{\lambda_2} \eta_1^2 \langle (\cos \theta)^2 \rangle_1 \right) \times \frac{\lambda_2 \underline{w}(2\nu)}{1 - \lambda_1 \underline{w}(2\nu)}, \quad (69)$$

where  $t_0 = 0$  was set to cancel the trivial phase factor  $e^{2i\nu t_0}$ , and the spectral waiting-time distribution is defined as

$$\underline{w}(\omega) = \int_0^{\infty} dt w(t) e^{i\omega t}, \quad (70)$$

with  $w(t)$  defined in Eq. (28).

The last complex-valued factor in Eq. (69),

$$\frac{\lambda_2 \underline{w}(2\nu)}{1 - \lambda_1 \underline{w}(2\nu)} = A e^{i\phi_A}, \quad (71)$$

where  $A$  is a real-valued semipositive number, depends on the branching ratio, and, via the waiting-time distribution, on the trap frequency and the laser parameters. This function characterizes the saturation of the recoil distribution's anisotropy. With the definition

$$\underline{w}(2\nu) = \underline{w} \exp(i\phi_w), \quad (72)$$

and since  $\underline{w}$  is semipositive, the modulus  $A$  and the phase  $\phi_A$  of the anisotropy (71) can be written as

$$A = \frac{\lambda_2 \underline{w}}{\sqrt{(1 - \lambda_1 \underline{w})^2 + 4\lambda_1 \underline{w} \sin^2(\phi_w/2)}}, \quad (73)$$

$$\tan \phi_A = \frac{\sin \phi_w}{\cos \phi_w - \lambda_1 \underline{w}}. \quad (74)$$

Employing Eqs (68) and (71), we rewrite the moment (69) as

$$\langle \alpha^2 \rangle_p = -\bar{n}_p A e^{i\phi_A}. \quad (75)$$

The phase-dependent quadrature is defined as

$$q(\phi) = \frac{1}{\sqrt{2}} (\alpha e^{i\phi} + \alpha^* e^{-i\phi}) = \sqrt{2} \text{Re}(\alpha e^{i\phi}). \quad (76)$$

The above considerations suggest that the average quadrature vanishes,  $\langle q(\phi) \rangle_p = 0$ . Its variance, however, is in general nonvanishing and phase dependent. It is expressed in terms of the moments  $[\Delta q(\phi) = q(\phi) - \langle q(\phi) \rangle_p]$  as

$$\langle [\Delta q(\phi)]^2 \rangle_p = \langle \alpha^{*2} \alpha^2 \rangle_p + \text{Re}(\langle \alpha^2 \rangle_p e^{2i\phi}), \quad (77)$$

which, upon insertion of Eqs. (68) and (75), writes

$$\delta q_{p,\phi}^2 = \langle [\Delta q(\phi)]^2 \rangle_p = \bar{n}_p [1 - A \cos(2\phi + \phi_A)], \quad (78)$$

where the abbreviation  $\delta q_{p,\phi} = \sqrt{\langle [\Delta q(\phi)]^2 \rangle_p}$  has been introduced.

At the values of the phase  $\phi_- = -\phi_A/2$  and  $\phi_+ = \phi_- + \pi/2$ , the minimum and maximum variances, respectively, are attained,

$$\delta q_{p,\phi_{\pm}}^2 = \bar{n}_p (1 \pm A), \quad (79)$$

and the product of the unequal rms uncertainties is

$$\delta q_{p,\phi_+} \delta q_{p,\phi_-} = \bar{n}_p \sqrt{1 - A^2}. \quad (80)$$

In general the density of recoil shifts is not rotationally symmetric, as shows the ratio of the difference of maximum and minimum quadrature fluctuations over the mean fluctuation, i.e., the anisotropy in the quadratures,

$$\frac{\delta q_{p,\phi_+}^2 - \delta q_{p,\phi_-}^2}{\delta q_{p,\phi_+}^2 + \delta q_{p,\phi_-}^2} = A. \quad (81)$$

There are, however, two special cases where the density is approximately isotropic, i.e., where  $A \approx 0$ . This situation is obtained either with negligible spectral waiting-time distribution at twice the trap frequency,  $w = |w(2\nu)| \approx 0$ , or with a very large value of the branching ratio,  $\lambda_1 \approx 1$  and  $\lambda_2 \approx 0$ , see Eq. (73). The first case represents a situation where the spectral waiting-time distribution lacks a frequency component at twice the trap frequency. This condition means that the waiting times of the probabilistic sequence of spontaneous emissions of photons are not synchronized to the half period of the trap oscillation. The latter case simply corresponds to a situation where the decay rate to state  $|1\rangle$  much exceeds that to state  $|2\rangle$ , so that the optical pumping process requires a very long sequence of spontaneous photon emissions, each one randomizing the density of recoil shifts that eventually becomes isotropic. This case corresponds to fluorescence scattering, as discussed for a free atom in Refs. [6,8]. In general, however, a finite sequence of photon emissions during the optical pumping generates a ‘‘squashed’’ density of recoil shifts with a predefined orientation in phase space that is determined by the phase of the spectral waiting-time distribution.

Finally, for the characterization of the density of recoil shifts, the variance

$$\delta n_p^2 = \langle \alpha^{*2} \alpha^2 \rangle_p - \bar{n}_p^2, \quad (82)$$

that corresponds to the mean  $\bar{n}_p$ , is evaluated. The first moment on the right-hand side is obtained as [see Eq. (C12)]

$$\begin{aligned} \langle \alpha^{*2} \alpha^2 \rangle_p &= \left( \eta_2^4 \langle (\cos \theta)^4 \rangle_2 + \frac{\lambda_1}{\lambda_2} \eta_1^4 \langle (\cos \theta)^4 \rangle_1 \right) \\ &+ 2\bar{n}_p [\bar{n}_p - \eta_2^2 \langle (\cos \theta)^2 \rangle_2] (1 + A \cos \phi_A), \end{aligned} \quad (83)$$

so that the variance (82) becomes

$$\begin{aligned} \delta n_p^2 &= \left( \eta_2^4 \langle (\cos \theta)^4 \rangle_2 + \frac{\lambda_1}{\lambda_2} \eta_1^4 \langle (\cos \theta)^4 \rangle_1 \right) - \bar{n}_p^2 \\ &+ 2\bar{n}_p (\bar{n}_p - \eta_2^2 \langle (\cos \theta)^2 \rangle_2) (1 + A \cos \phi_A). \end{aligned} \quad (84)$$

## B. Mapping the moments of the atomic phase-space distribution

So far, the density of recoil shifts has been characterized by its statistical moments. To characterize the vibrational quantum state of the ion after completion of optical pumping, the initial vibrational phase-space distribution of the ion has to be convolved with the density of recoil shifts, as given in Eq. (47). Fourier transforming Eq. (47), one obtains the mapping of the characteristic function, that corresponds to the vibrational phase-space distribution, as

$$P_2^{(s)}(\alpha, t) = P_2(t_0) P_2^{(s)}(\alpha, t_0) + P_1(t_0) p(\alpha; t, t_0) P_1^{(s)}(\alpha, t_0), \quad (85)$$

where the Fourier transform of the density of recoil shifts is given by Eq. (57).

From the  $s$ -ordered phase-space distributions,  $P_a^{(s)}(\alpha, t)$ , the  $s$ -ordered quantum-statistical moments are represented by integrals [43,44],

$$\begin{aligned} \langle \{\hat{b}^{\dagger k}(t) \hat{b}^l(t)\}_s \rangle_a &= \frac{1}{P_a} \text{Tr}[\{\hat{b}^{\dagger k} \hat{b}^l\}_s \langle a | \hat{\rho}(t) | a \rangle] \\ &= \int d^2 \alpha \alpha^{*k} \alpha^l P_a^{(s)}(\alpha, t), \end{aligned} \quad (86)$$

where  $\{\hat{b}^{\dagger k} \hat{b}^l\}_s$  denotes the  $s$ -ordered product of operators and  $a=1, 2$  specifies the electronic level. As a result of the Fourier transform, these moments are obtained equivalently as derivatives of the corresponding characteristics function,

$$\langle \{\hat{b}^{\dagger k}(t) \hat{b}^l(t)\}_s \rangle_a = (-1)^l [\partial_{\alpha^*}^k \partial_{\alpha}^l P_a^{(s)}(\alpha, t)]_{\alpha=0}. \quad (87)$$

Inserting the mapping of characteristic functions (85) into (87) and applying the Leibniz formula yields the mapping of initial quantum-statistical moments of the ion's vibration on the final moments ( $t_0=0$  and  $t \rightarrow \infty$ ),

$$\begin{aligned} \langle \{\hat{b}^{\dagger k}(\infty) \hat{b}^l(\infty)\}_s \rangle_2 &= P_2(0) \langle \{\hat{b}^{\dagger k}(0) \hat{b}^l(0)\}_s \rangle_2 + P_1(0) \sum_{n=0}^k \sum_{m=0}^l \binom{k}{n} \\ &\times \binom{l}{m} \langle \alpha^{*(k-n)} \alpha^{l-m} \rangle_p \langle \{\hat{b}^{\dagger n}(0) \hat{b}^m(0)\}_s \rangle_1. \end{aligned} \quad (88)$$

The moments of the density of recoil shifts,  $\langle \alpha^{*k} \alpha^l \rangle_p$ , act here as weight factors for contributions to the sum of initial vibrational moments on the right-hand side.

Setting  $k=0$  and  $l=1$  in Eq. (88), the mapping of the coherent vibrational amplitude is obtained as

$$\langle \hat{b}(\infty) \rangle_2 = \langle \hat{b}(0) \rangle, \quad (89)$$

where the complete quantum-statistical average on the right-hand side is defined as



$$\langle \dots \rangle = \sum_{a=1,2} P_a \langle \dots \rangle_a = \text{Tr}[\hat{\rho} \dots]. \quad (90)$$

Thus the coherent vibration is unaffected by the pump process, as is the expectation value of the phase-dependent quadrature operator

$$\hat{q}_\phi = \frac{1}{\sqrt{2}}(\hat{b}e^{i\phi} + \hat{b}^\dagger e^{-i\phi}), \quad (91)$$

which reveals the mapping

$$\langle \hat{q}_\phi(\infty) \rangle_2 = \langle \hat{q}_\phi(0) \rangle. \quad (92)$$

Although optical pumping is a highly incoherent process, the initial coherent amplitude of the ion's oscillation in the trapping potential is perfectly preserved.

The mean vibrational excitation, however, is altered. Using Eq. (88) with values  $k=l=1$  and specifying normally ordered operator products, i.e., setting  $s=-1$ , the mapping of the mean vibrational excitation is obtained as

$$\langle \hat{n}(\infty) \rangle_2 = \langle \hat{n}(0) \rangle + P_1(0)\bar{n}_p, \quad (93)$$

where  $\hat{n}=\hat{b}^\dagger\hat{b}$  is the number of vibrational quanta and the mean number of vibrational quanta added by the pump process,  $\bar{n}_p$ , is given by Eq. (68). Thus, as seen from Eq. (68), the addition of mean vibrational excitation by the optical pump process does entirely depend on the Lamb-Dicke parameters and on the mean number of spontaneously emitted photons, that is determined by the branching ratio,  $\langle \hat{n}_{\text{ph}} \rangle = \lambda_1/\lambda_2 + 1$ . It does *not* depend on the speed of optical pumping that is determined by the parameters of the pump laser.

Let us now turn to the rms spreads of these properties. Whereas the expectation value of the phase-dependent quadrature is preserved during the optical pumping, see Eq. (92), the corresponding rms spread is not. The square of this rms spread, i.e., the variance, is constructed as

$$\langle [\Delta \hat{q}_\phi]^2 \rangle = \langle \hat{n} \rangle + \frac{1}{2} + \text{Re}[\langle \hat{b}^2 \rangle e^{2i\phi}] - \langle \hat{q}_\phi \rangle^2, \quad (94)$$

where only the last two terms are phase dependent. From Eq. (88) the mapping of the moment  $\langle \hat{b}^2 \rangle$  is obtained with the choice  $k=0, l=2$  as

$$\langle \hat{b}^2(\infty) \rangle_2 = \langle \hat{b}^2(0) \rangle + P_1(0)\langle \alpha^2 \rangle_p. \quad (95)$$

Thus, with the mappings (92) and (93), and after completion of pumping, the variance of the quadrature in the pumped electronic state becomes

$$\langle [\Delta \hat{q}_\phi(\infty)]^2 \rangle_2 = \langle [\Delta \hat{q}_\phi(0)]^2 \rangle + P_1(0)\delta q_{p,\phi}^2, \quad (96)$$

where the additional noise  $\delta q_{p,\phi}^2$  is given in Eq. (78). In accordance with the moments of the density of recoil shifts, minimum noise is added to the rms spread of the quadrature at the phase  $\phi_- = -\phi_A/2$ , and maximum noise is added at phase  $\phi_+ = \phi_- + \pi/2$ .

Finally most relevant is how broad grows the final vibrational number distribution. The variance of the vibrational quantum number is obtained from normally ordered moments as

$$\langle [\Delta \hat{n}]^2 \rangle = \langle \hat{b}^{\dagger 2} \hat{b}^2 \rangle - \langle \hat{n} \rangle (\langle \hat{n} \rangle - 1). \quad (97)$$

From Eq. (88), using  $k=l=2$  and  $s=-1$ , the mapping of the required normally ordered moment is obtained as

$$\langle \hat{b}^{\dagger 2}(\infty) \hat{b}^2(\infty) \rangle_2 = \langle \hat{b}^{\dagger 2}(0) \hat{b}^2(0) \rangle + P_1(0)\{2 \text{Re}[\langle \alpha^2 \rangle_p \langle \hat{b}^{\dagger 2}(0) \rangle_1] + 4\langle \alpha^* \alpha \rangle_p \langle \hat{n}(0) \rangle_1 + \langle \alpha^{*2} \alpha^2 \rangle_p\}. \quad (98)$$

Thus using Eqs. (93) and (98) the mapping of the variance of the vibrational quantum number, Eq. (97), results as

$$\langle [\Delta \hat{n}(\infty)]^2 \rangle_2 = \langle [\Delta \hat{n}(0)]^2 \rangle + P_1(0)\{\delta n_p^2 + 2\bar{n}_p[m_1 + P_2(0)m_2]\}. \quad (99)$$

The numbers  $m_1$  and  $m_2$  depend on the initial state,

$$m_1 = \left[ \langle \hat{n}(0) \rangle_1 + \frac{1}{2} \right] - A \text{Re}[\langle \hat{b}^2(0) \rangle_1 e^{-i\phi_A}], \quad (100)$$

$$m_2 = \frac{\bar{n}_p}{2} + \langle \hat{n}(0) \rangle_1 - \langle \hat{n}(0) \rangle_2, \quad (101)$$

and  $\delta n_p^2$  is given by Eq. (84).

For example, an atom being initially laser cooled to its trap ground state  $[\langle \hat{n}(0) \rangle_{1,2} = \langle \hat{n}(0) \rangle = 0, \langle \hat{b}^2(0) \rangle = 0]$ , and also in its electronic ground state  $[P_1(0)=1, P_2(0)=0]$ , has  $m_1 = 1/2$  and  $m_2 = \bar{n}_p/2$ , the latter not entering the final variance. The mapping of the variance of the vibrational excitation then simplifies to

$$\langle [\Delta \hat{n}(\infty)]^2 \rangle_2 = \bar{n}_p + \delta n_p^2, \quad (102)$$

i.e., the variance of the final vibrational distribution equals the mean plus variance of the recoil displacements.

## V. RESULTS

### A. Fluorescence scattering

As noted before the case of fluorescence scattering is covered by the model in the limit  $\lambda_2 \rightarrow 0$ , when the trail of spontaneously emitted photons becomes infinite so that the directions of recoils in vibrational phase space are efficiently averaged to result in an isotropic recoil density. However, the resulting recoil density would correspond to the limit  $t \rightarrow \infty$ , when the diffusion of fluorescence scattering would have infinitely broadened the density distribution. Therefore, a direct comparison is impossible, and we restrict ourselves to a discussion of the asymptotic behavior for small  $\lambda_2$ .

In the limit  $\lambda_2 \rightarrow 0$  the anisotropy parameter becomes

$$A \rightarrow \begin{cases} 1, & \text{if } w = 1 \text{ and } \phi_w = 0, \\ 0, & \text{else.} \end{cases} \quad (103)$$

Apart from the exceptional case  $w(2\nu)=1$  [51], where the waiting times are perfectly synchronized to half the trap period so that a highly directional scattering occurs ( $A \rightarrow 1$ ), the recoil density is isotropic with respect to the variance of its phase-dependent quadrature ( $A \rightarrow 0$ ). In more detail, for small  $\lambda_2$  the anisotropy's asymptotic behavior is given by  $A \asymp a\lambda_2$  and  $\phi_A \asymp \text{const}$ , where the coefficient  $a$  is

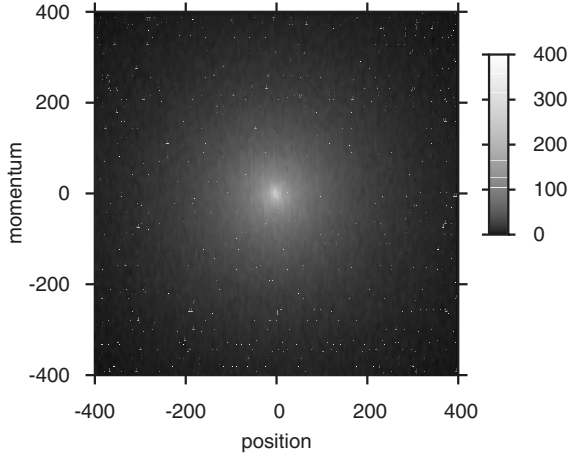


FIG. 2. Recoil density  $p(\alpha)$  in dependence on position and momentum scaled as  $x/(\eta_1\Delta x_0)$  and  $p/(\eta_1\Delta p_0)$ , respectively, with trap ground-state uncertainties being  $\Delta x_0\Delta p_0=\hbar/2$ . Parameters are  $\lambda_2=10^{-5}$ ,  $\eta_2/\eta_1=0.75$ ,  $\tilde{\nu}=0.16$ ,  $\Delta=0$ , and  $S=25$  (1 000 000 phase-space shifts sampled on a  $200\times 200$  grid). Transition dipole moments are assumed to be perpendicular to the chosen motional axis.

$$a = \frac{\underline{w}}{\sqrt{(1-\underline{w})^2 + 4\underline{w}\sin^2(\phi_w/2)}}. \quad (104)$$

The asymptotic form of the rms spread of the quadrature becomes then phase independent, cf. Eq. (78), and results as

$$\delta q_{p,\phi} \asymp \sqrt{\bar{n}_p}. \quad (105)$$

Like the mean vibrational excitation (68), the rms spread of the quadrature is now independent of the laser parameters. As the mean vibrational excitation asymptotically behaves as

$$\bar{n}_p \asymp \frac{\eta_1^2}{\lambda_2} \langle (\cos \theta)^2 \rangle_1, \quad (106)$$

the rms spread of the quadrature (105) asymptotically takes on the form

$$\delta q_{p,\phi} \asymp \eta_1 \sqrt{\langle (\cos \theta)^2 \rangle_1 / \lambda_2}. \quad (107)$$

Correspondingly, the rms spread is asymptotically  $\delta n_p \asymp \bar{n}_p$  and also turns independent of the laser parameters. Thus, we expect an isotropic recoil density in phase space that just weakly depends on the laser parameters through higher-order moments.

Numerical evaluation of Eq. (37) for  $t \rightarrow \infty$  ( $t_0=0$ ) and  $\lambda_2=10^{-5}$  in resonant excitation ( $\Delta=0$ ) reveals the isotropic recoil density in phase space as shown in Fig. 2, which is independent of the preset laser saturation  $S=(|\kappa|/\gamma)^2$ , in agreement with previous considerations. Given the infinite trail of spontaneously emitted photons, each photon contributing to a random recoil, one might presume the validity of the central-limit theorem that would predict a perfectly Gaussian profile of the recoil density. However, the individual recoil shifts in phase space are not precisely independent random variables. The phases, i.e., directions in phase space, of subsequent recoils are correlated by means of the waiting-time distribution (28). The detailed form of this dis-

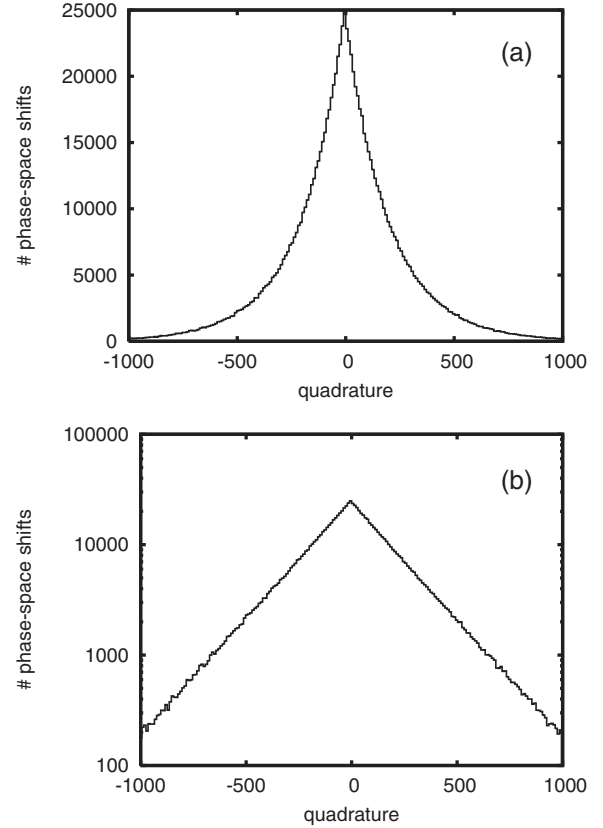


FIG. 3. (a) Phase-independent distributions of quadratures of  $p(\alpha)$  for (resonant) fluorescence scattering; parameters are the same as in Fig. 2 (1 000 000 phase-space shifts sampled in 200 bins in the interval  $[-1000:1000]$ ); (b) logarithmic plot of the same function.

tribution seems to be lost in the averaging process, its main characteristics, however, a finite mean waiting time between subsequent photon emissions, leaves a correlation between subsequent recoil shifts. This effect appears as substantial deviation from a Gaussian profile in the distribution of quadratures shown in Fig. 3(a). In fact, this recoil density perfectly approaches an  $\exp(-|x|)$  function as can be seen from the corresponding logarithmic plot in Fig. 3(b).

It has been shown that fluorescence scattering of a free atom generates, after many photon emissions, a Gaussian momentum distribution [6,8]. The above results show that this situation corresponds to the limit  $\nu \rightarrow 0$ , i.e., when switching off the trap potential. Then, the spectral waiting-time distribution enters Eqs. (73) and (74) at zero frequency and equals unity due to the normalization of the waiting-time distribution,  $\underline{w}(0)=1$ . Thus, in this particular case the laser parameters do not appear, and neither does the waiting-time distribution—except its normalization property. Each photon recoil shifts the atomic momentum in the momentum's original direction, and the length of each shift is a statistically independent random number, whose statistics is given by the dipole radiation characteristics. The application of the central-limit theorem is here perfectly justified given a sufficiently large number of spontaneous photon emissions.

## B. Optical pumping with few photon emissions

On the other hand, in the opposite limit  $\lambda_2 \rightarrow 1$ , the atom spontaneously emits one photon only and reaches its final

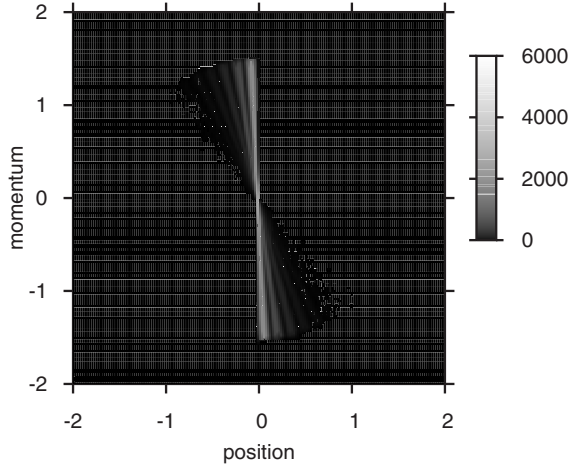


FIG. 4. Recoil density  $p(\alpha)$  versus position and momentum scaled as  $x/(\eta_1\Delta x_0)$  and  $p/(\eta_1\Delta p_0)$ , respectively, with trap ground-state uncertainties being  $\Delta x_0\Delta p_0=\hbar/2$ . Parameters are  $\lambda_1=10^{-5}$ ,  $\eta_2/\eta_1=0.75$ ,  $\tilde{\nu}=0.16$ ,  $\Delta=0$ , and  $S=25$  (1 000 000 phase-space shifts sampled on a  $200\times 200$  grid). Transition dipole moments are assumed to be perpendicular to the selected motional axis.

pumped state. In this limit, the joint photon emission probability density (32) becomes

$$\lim_{\lambda_2\rightarrow 1} w_n(t_n, \dots, t_1) = \delta_{n,1} w(t_1 - t_0), \quad (108)$$

from which the recoil density (37) is derived for completed pumping ( $t\rightarrow\infty$ ,  $t_0=0$ ) and making use of Eq. (35) as

$$p(\alpha) = \int_0^\infty dt \int ds \mu_2(s) w(t) \delta[\alpha - i\eta_2 s \exp(i\nu t)]. \quad (109)$$

For high enough laser saturation ( $S>1$ ) the waiting-time distribution decays at the rate on the order of  $\gamma$ . In an experiment, the trap frequency is typically much lower,  $\nu\ll\gamma$ . Thus, to very good approximation we may discard in Eq. (109) the comparably slow time dependence of the direction of the recoil shift,  $e^{i\nu t}\approx 1$ , perform the integral over the two-dimensional  $\delta$  function, and obtain

$$p(\alpha) \approx \delta(q) \mu_2 \left( \frac{p}{\eta_2} \right) / \eta_2, \quad (110)$$

where  $\alpha=q+ip$ . Thus the recoil density extends only in direction of momentum in phase space, reproducing the profile of the dipole radiation characteristics. This density distribution agrees with the profile associated with a free atom after single-photon scattering [8].

In Fig. 4 we approach this limiting case for the branching ratio  $\lambda_1=10^{-5}$ . The almost unidirectional scattering in phase space is well observed. Moreover, for the corresponding quadrature, the distribution of maximum rms fluctuation shows a dipole radiation characteristics, see solid curve in Fig. 5. The secondary and tertiary lobes in the recoil density of Fig. 4 mark delayed spontaneous emission, modulated by the laser-driven damped Rabi cycles on the  $|1\rangle\leftrightarrow|3\rangle$  transition.

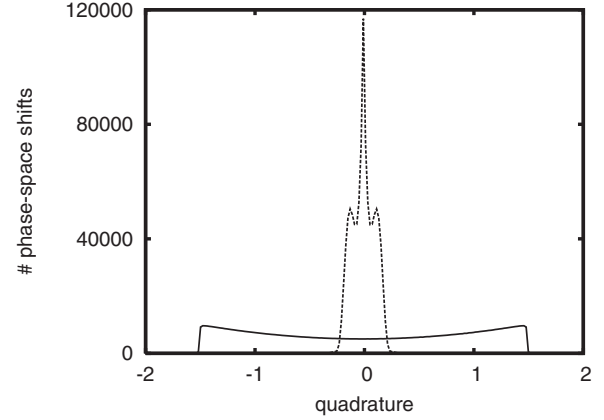


FIG. 5. Distributions of quadratures of  $p(\alpha)$  versus the scaled quadrature corresponding to maximum (solid) and minimum (dashed) rms quadrature fluctuations. Parameters are those of Fig. 4 (1 000 000 phase-space shifts sampled in 200 bins on the interval  $[-2, 2]$ ).

### C. Laser-saturation dependence of the anisotropy

The anisotropy can be written as a function of three variables,

$$A(\lambda_1, \underline{w}, \phi_w) = \frac{(1-\lambda_1)\underline{w}}{\sqrt{(1-\lambda_1\underline{w})^2 + 4\lambda_1\underline{w} \sin^2(\phi_w/2)}}. \quad (111)$$

Two of these variables,  $\underline{w}$  and  $\phi_w$ , depend on the laser parameters, i.e., on detuning and saturation of transition  $|1\rangle\leftrightarrow|3\rangle$ . For resonant pumping ( $\Delta=0$ ), these functions of laser saturation  $S$  are obtained as [see Eqs. (D13) and (D14)]

$$\underline{w}(S) = \frac{S}{\sqrt{(1+\tilde{\nu}^2)[(S-\tilde{\nu}^2)^2 + 4\tilde{\nu}^2]}}, \quad (112)$$

$$\phi_w(S) = \arctan \left[ \frac{\tilde{\nu}(S+2-\tilde{\nu}^2)}{S-3\tilde{\nu}^2} \right], \quad (113)$$

with  $\tilde{\nu}=2\nu/\gamma$ .

Thus, the anisotropy  $A(S)$  depends on laser saturation with the branching ratio  $\lambda_2$  and the scaled trap frequency  $\tilde{\nu}$  being parameters. With Eqs. (112) and (113) inserted into Eq. (111), this function is

$$A(S) = \frac{\lambda_2 S}{\sqrt{(\lambda_2 S - 3\tilde{\nu}^2)^2 + \tilde{\nu}^2(S+2-\tilde{\nu}^2)^2}}. \quad (114)$$

Its saturated value is

$$\lim_{S\rightarrow\infty} A(S) = \frac{\lambda_2}{\sqrt{\lambda_2^2 + \tilde{\nu}^2}}, \quad (115)$$

which vanishes for resonant fluorescence scattering,  $\lambda_2\rightarrow 0$ . Likewise, the phase may be written as the saturation-dependent function

$$\tan \phi_A(S) = \frac{\sin \phi_w(S)}{\cos \phi_w(S) - \lambda_1 \underline{w}(S)}. \quad (116)$$

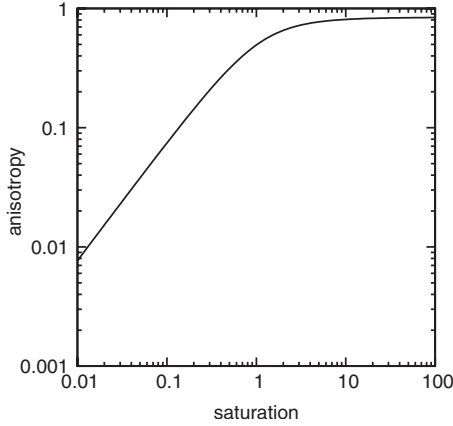


FIG. 6. Anisotropy  $A$  as a function of the laser saturation  $S$  for resonant pumping ( $\Delta=0$ ); parameters are  $\tilde{\nu}=0.16$ ,  $\lambda_1=0.75$ .

The anisotropy, an example of which is shown in Fig. 6, may have a local maximum in form of a typical underdamped peak. However, it can be shown that this maximum appears at the saturation value

$$S_{\max} = \frac{\tilde{\nu}^4 + 5\tilde{\nu}^2 + 4}{\tilde{\nu}^2 + 3\lambda_2 - 2} \quad (117)$$

only if the condition

$$\lambda_2 > \frac{2 - \tilde{\nu}^2}{3} \quad (118)$$

holds. Otherwise the global maximum is attained at full saturation  $S \rightarrow \infty$ . For example, given the typical value  $\tilde{\nu}=2\nu/\gamma=0.16$ , a branching ratio  $\lambda_2 \geq 0.66$  is required to observe the maximum anisotropy at extremely high laser saturation. Typically, this regime is inaccessible in experiment, so that usually the anisotropy is a monotonically increasing function of the saturation, as shown in Fig. 6.

#### D. Optimized optical pumping

In many applications one intends to minimize the disturbance of optical pumping on an initially given vibrational quantum state, in order to only affect the electronic system in the desired way. Optimization depends, however, on the specific type of disturbance that is supposed to be minimized. As we have seen in Sec. IV B, the various vibrational moments depend quite differently on the laser parameters, some even being independent of the kind of laser excitation. Thus, we may present optimum values for optical pumping in a common case. To discuss the worst-case scenario, we start from an atom with initial population in the unpumped state  $P_1(0)=1$ .

For keeping the rise of the mean vibrational excitation (i.e., heating) as small as possible during optical pumping, as indicated by Eq. (93), the moment  $\bar{n}_p$ , i.e., the mean recoil-induced excitation, has to be minimized. This moment does not depend on laser parameters, cf. Eq. (68), but is a fixed value determined by the Lamb-Dicke parameters, the branching ratio, and the second moment of the dipole radi-

ation characteristics. It is thus irrelevant whether optical pumping is performed resonantly or off-resonantly, or whether a low or high saturation is employed.

The width of the resulting distribution over vibrational quantum numbers, however, does depend on the laser parameters. While the atom is initially in its vibrational ground state  $|n=0\rangle$ , the final spread of vibrational quantum numbers in the pumped state is given by Eq. (102) as

$$\begin{aligned} \langle [\Delta\hat{n}(\infty)]^2 \rangle_2 = & \bar{n}_p^2 + \frac{\lambda_1}{\lambda_2} \eta_1^4 \langle (\cos\theta)^2 \rangle_1^2 + \left( \eta_2^4 C_2 + \frac{\lambda_1}{\lambda_2} \eta_1^4 C_1 \right) \\ & + 2\bar{n}_p \frac{\lambda_1}{\lambda_2} \eta_1^2 \langle (\cos\theta)^2 \rangle_1 A \cos\phi_A, \end{aligned} \quad (119)$$

where we have defined

$$C_{1,2} = \langle (\cos\theta^4) \rangle_{1,2} - \langle (\cos\theta^2) \rangle_{1,2}^2. \quad (120)$$

This is minimized for  $A \cos\phi_A$  being as negative as possible. For resonant pumping ( $\Delta=0$ ) the anisotropy parameters are only functions of the saturation  $S$ . Thus, we may search for the minimum of  $A(S)\cos\phi_A(S)$ . Extrema of this function follow from the condition

$$\frac{A'(S)}{A(S)} = \phi'_A(S) \tan\phi_A(S), \quad (121)$$

where the primes indicate derivatives. The equation can be shown to have only negative solutions, whereas the saturation by definition is a semipositive number. Since the global minimum is located at  $S=0$ , where  $A \cos\phi_A=0$ , a minimum spread of the vibrational numbers is obtained when saturation is kept as low as possible. Thus, slow optical pumping is required for minimum width of the vibrational number distribution. Optimization with respect to other moments, however, requires other values of saturation.

## VI. MOTIVATION AND CONTEXT

### A. The center-of-mass effects in optical pumping

Optical pumping is a time-honored subject. A wealth of experiments have required various approaches of modeling. However, these theories usually limited their models to the description of the atom's electronic dynamics driven by the light. The center-of-mass dynamics, modified by the unavoidable radiative recoil, was usually neglected on the basis of the assumption that atomic collisions and in particular the spontaneous emission intrinsically tied to the pumping process would average an atom's state to an unspectacular slightly broadened Gaussian distribution in phase space that might at most render energy levels and transitions broadened by an almost unobservable amount.

The results of the above calculations show that this attitude is an oversimplification. For complete description of OP, the demonstrated modeling of the recoil effects is indispensable. These effects leave, in general, a pumped atom in a non-Gaussian state with an anisotropic distribution in phase space. The model reveals a continuous transit from maximum asymmetry to full symmetry going along with the num-

ber of reiterated scattering events increasing up to the final pumping event. Consequently, the center-of-mass effect of optical pumping is linked in a natural way to that of resonance scattering on the pump-excited line: The latter phenomenon is described by an infinite series of scattering events, lacking the final spontaneous decay into the pumped state. When the pump light is detuned off resonance, the accumulated recoil turns out to be a detrimental heating contribution to the effect of laser sideband cooling of the atom weakly bound in a trap. (The wanted contribution is from the recoil by the pump light.) For an atom strongly bound to the trap, however, the momentum of recoil is transferred to the entire trap. This process is analogous with the origin of narrow-band fluorescence emission in the Mössbauer effect.

### B. Optical pumping of vapors

Optical pumping of trapped or free atomic vapors can be described by our model, except for ultralow temperature when the gas is degenerate. Elastic collisions will then generate scattering among vibrational levels, to effect thermalization of the vapor.

The atoms will be in a nonequilibrium state after optical pumping. As with the electronic inversion, this also applies to the vibrational state. Both the particular shape and the anisotropy of the density of recoil shifts is transmitted in optical pumping to the final vibrational phase-space distribution. Such a distribution, in general, is far from a thermal distribution corresponding to equilibrium. In particular, the anisotropy in phase space indicates that kinetic and potential energies of the ensemble are unbalanced, which manifests itself by the atomic cloud breathing at twice the trap frequency.

Atomic collisions redistribute populations in the vibrational levels and thus rebalance kinetic versus potential energy to arrive at a thermalized equilibrium state. This process happens on a time scale that is determined by the atomic scattering cross section. Given that this time scale is large enough to be detected experimentally, the observation of the cloud should reveal damping of the cloud's breathing.

### C. Observability of anisotropy and non-Gaussian shape in phase space

The prominent features of the results presented here are the anisotropy and the non-Gaussian shape of the density of recoil shifts in phase space. They can be directly observed by using a simplified version of a method, proposed for the reconstruction of the vibrational quantum state [46,47], that has been implemented experimentally in recent years [48].

After optical pumping a ground-state atom whose initial phase-space distribution is rotationally symmetric (e.g., any vibrational number state or incoherent mixtures of these states), the  $|1\rangle \leftrightarrow |2\rangle$  transition is bichromatically driven in stimulated Raman configuration on the first red and the first blue vibrational sideband. Application of this laser light for a finite and adjustable time duration  $\tau$  maps information on the distribution of quadratures onto the occupation of the ground state  $|1\rangle$ . Given a laser phase shift  $\phi$  introduced in one of the

two Raman transitions, the ground state occupation reads [46,47]

$$P_1(\tau, \phi) = \frac{1}{2} - \frac{1}{2} \int dq e^{-iq\tau} p(q, \phi), \quad (122)$$

where the distribution  $p(q, \phi)$  is the probability density of observing the quadrature  $q$  at phase  $\phi$ , as shown in Figs. 3 and 5 in Sec. V.

The occupation in the ground state  $P_1$  is obtained by probing the atom for fluorescence of the transition from  $|1\rangle$  to an auxiliary level. Thus, varying the time duration  $\tau$  and Fourier transforming the measured time-dependent ground state occupation, all distributions of the phase-dependent quadrature are obtained. The appearance of phase-dependent distributions of quadratures would prove the predicted anisotropy of the density of recoil shifts. In addition, the shape of the measured distributions could be tested for the deviation from a Gaussian profile.

## VII. SUMMARY AND CONCLUSIONS

In this paper it was shown how recoil effects that naturally appear with optical pumping are analytically modeled for a harmonically bound atom. Due to the trail of spontaneously emitted photons being finite, a quantum trajectory-type of approach is well adapted to this analytic procedure. All effects on the quantum motion of the atom depend on a single function, the recoil density in phase space. Its moments are obtained analytically from the dipole radiation characteristics and the waiting-time distribution of the atom's light scattering.

It was shown that even approaching the case of fluorescence scattering the recoil density does not become a Gaussian function in the motional phase space, as one would expect when taking resort to the central-limit theorem. Instead, it is a distribution of the form  $\exp(-|x|)$ , that emerges from correlations of emission times and the concomitant correlations of the recoil directions in phase space of subsequent spontaneous photon emissions. In the opposite extreme, when only few photons are scattered during the full pump cycle, the recoil density is characterized by the spatial distribution of dipole radiation.

In general, the recoil density is not isotropically distributed but reveals a pronounced directional structure. For resonant pumping, this anisotropy can be maximized adjusting the laser saturation—in most relevant cases by increasing it.

Optimum laser saturation and detuning for minimum detrimental heating depend on the particular property that is to be protected from the recoil effects. Whereas the mean increase of vibrational excitation cannot be varied, the rms fluctuations of the vibrational number can be affected by the laser parameters, as is the case also for the rms fluctuations of the phase-dependent quadrature. However, optimum laser parameters depend on the initial motional quantum state and on the particular phase of a quadrature to become protected. There exists a complex interplay among the initial motional quantum state, the particular fluctuations to be minimized, and the laser parameters that fit in with the given requirements.

## ACKNOWLEDGMENTS

S.W. and P.E.T. acknowledge support by FONDECYT Grant No. 7060187, S.W. acknowledges support by FONDECYT Grant No. 1051042.

## APPENDIX A: DERIVATIVES OF THE CHARACTERISTIC FUNCTION FOR INSTANTANEOUS RECOIL SHIFTS

The characteristic function associated with the probability density of instantaneous recoil shifts is

$$p_a(\alpha, t) = \underline{\mu}_a [2\eta_a |\alpha| \cos(\nu t - \arg \alpha)] = \underline{\mu}_a [\eta_a (\alpha^* e^{i\nu t} + \alpha e^{-i\nu t})],$$

where  $\nu$  is the vibrational frequency. Its  $(k, l)$ -fold derivatives are

$$\begin{aligned} & \left( \frac{\partial}{\partial \alpha^*} \right)^l \left( \frac{\partial}{\partial \alpha} \right)^k p_a(\alpha, t) \Big|_{\alpha=0} \\ &= \left( \frac{\partial}{\partial \alpha^*} \right)^l (\eta_a e^{-i\nu t})^k \underline{\mu}_a^{(k)} [\eta_a (\alpha^* e^{i\nu t} + \alpha e^{-i\nu t})] \Big|_{\alpha=0} \\ &= (\eta_a e^{i\nu t})^l (\eta_a e^{-i\nu t})^k \underline{\mu}_a^{(k+l)}(0), \end{aligned}$$

where

$$\underline{\mu}_a^{(k+l)}(0) = (-i)^{k+l} \int_{-1}^1 ds \mu_a(s) s^{k+l} = (-i)^{k+l} \langle (\cos \theta)^{k+l} \rangle_a,$$

so that

$$\begin{aligned} & \left( \frac{\partial}{\partial \alpha^*} \right)^l \left( \frac{\partial}{\partial \alpha} \right)^k p_a(\alpha, t) \Big|_{\alpha=0} \\ &= (-i \eta_a)^{k+l} e^{i\nu t(l-k)} \times \langle (\cos \theta)^{k+l} \rangle_a. \end{aligned} \quad (\text{A1})$$

The dipole radiation characteristics is obtained from Eq. (7) as

$$\mu_a(s) = \frac{3}{8\pi} \int_0^{2\pi} d\phi \{1 - \cos^2 \alpha\}, \quad (\text{A2})$$

where  $\alpha$  is the angle between  $\mathbf{n}_a$  and  $\mathbf{n}(\Omega)$  and thus

$$\cos \alpha = \cos \theta \cos \theta_a + \sin \theta \sin \theta_a \cos(\phi - \phi_a), \quad (\text{A3})$$

where  $\theta_a$  and  $\phi_a$  are the spherical angles of the transient dipole moment  $\mathbf{d}_{3a} = \langle 3|\hat{\mathbf{d}}|a \rangle$ . Inserting Eq. (A3) into Eq. (A2) and performing the integration the dipole characteristics results as

$$\mu_a(s) = \frac{3}{8} [1 + \cos^2 \theta_a + s^2(1 - 3 \cos^2 \theta_a)]. \quad (\text{A4})$$

It is thus a symmetric function,  $\mu_a(-s) = \mu_a(s)$ , that depends on the angle  $\theta_a$  between the transition dipole moment and the direction of motion.

## APPENDIX B: PHOTON-COUNTING STATISTICS

From the joint probability of spontaneous photon emissions, the photon-counting statistics derives from the prob-

ability for  $n$  spontaneous emissions during the time interval  $[t, t_0]$ ,

$$P_n(t, t_0) = \int_{t_0}^t dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \quad (n > 0), \quad (\text{B1})$$

and  $P_0(t, t_0) = 1 - \sum_{n=1}^{\infty} P_n(t, t_0)$ . The photon-counting statistics for a complete optical-pumping process,  $P_n$ , is obtained in the limit  $P_n = \lim_{t \rightarrow \infty} P(t, t_0)$  which reads

$$\begin{aligned} P_n &= \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\ &= \lambda_2 \lambda_1^{n-1} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w(t_n - t_{n-1}) \dots w(t_1 - t_0), \end{aligned} \quad (\text{B2})$$

where Eq. (32) has been used. The waiting-time distributions are defined for positive time arguments only, so that we set  $w(t) = 0$  for  $t < 0$  and extend the upper integration limits to infinity. With the substitution of integration variables  $\tau_k = t_k - t_{k-1}$  ( $k = 1, \dots, n$ ) the photon-count statistics writes

$$\begin{aligned} P_n &= \lambda_2 \lambda_1^{n-1} \int_0^{\infty} d\tau_n \dots \int_0^{\infty} d\tau_1 w(\tau_n) \dots w(\tau_1) \\ &= \lambda_2 \lambda_1^{n-1} \quad (n > 0), \end{aligned}$$

and  $P_0 = 0$ . Here, the unit normalization of the waiting-time distribution has been used.

The average number of spontaneously emitted photons is therefore

$$\langle \hat{n}_{\text{ph}} \rangle = \sum_{n=0}^{\infty} n P_n = \sum_{n=1}^{\infty} n \lambda_2 \lambda_1^{n-1}, \quad (\text{B3})$$

which can be expressed as the derivative

$$\lambda_2 \frac{\partial}{\partial \lambda_1} \sum_{n=1}^{\infty} \lambda_1^n = \lambda_2 \frac{\partial}{\partial \lambda_1} \frac{\lambda_1}{1 - \lambda_1} = \frac{1}{\lambda_2},$$

so that the average photon number is

$$\langle \hat{n}_{\text{ph}} \rangle = \frac{1}{\lambda_2}. \quad (\text{B4})$$

The rms spread of photon numbers is obtained via the second-order moment

$$\begin{aligned} \langle \hat{n}_{\text{ph}}^2 \rangle &= \sum_{n=1}^{\infty} n^2 \lambda_2 \lambda_1^{n-1} \\ &= \lambda_2 \frac{\partial}{\partial \lambda_1} \sum_{n=1}^{\infty} n \lambda_1^n \\ &= \lambda_2 \frac{\partial}{\partial \lambda_1} \left( \frac{\partial}{\partial \lambda_1} \lambda_1 - 1 \right) \sum_{n=1}^{\infty} \lambda_1^n \\ &= \lambda_2 \frac{\partial}{\partial \lambda_1} \left( \frac{\partial}{\partial \lambda_1} \lambda_1 - 1 \right) \frac{\lambda_1}{1 - \lambda_1} \end{aligned}$$

$$\begin{aligned}
 &= \lambda_2 \frac{\partial}{\partial \lambda_1} \left( \frac{\partial}{\partial \lambda_1} \frac{\lambda_1^2}{1 - \lambda_1} - \frac{\lambda_1}{1 - \lambda_1} \right) \\
 &= \lambda_2 \frac{\partial}{\partial \lambda_1} \frac{\lambda_1}{(1 - \lambda_1)^2} \\
 &= \frac{1 + \lambda_1}{\lambda_2^2}.
 \end{aligned}$$

Therefore, the rms spread reads

$$\langle \Delta \hat{n}_{\text{ph}}^2 \rangle = \frac{\lambda_1}{\lambda_2^2}.$$

Its relative value reads

$$\langle \Delta \hat{n}_{\text{ph}}^2 \rangle / \langle \hat{n}_{\text{ph}} \rangle = \frac{\lambda_1}{\lambda_2},$$

which indicates sub- or super-Poissonian photon statistics, dependent on the branching ratios.

### APPENDIX C: MOMENTS OF THE DENSITY OF RECOIL SHIFTS

The function to be evaluated is given by

$$\begin{aligned}
 \langle \alpha^{*k} \alpha^l \rangle_p &= (-1)^l \sum_{n=1}^{\infty} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\
 &\times \{ \partial_{\alpha^*}^k \partial_{\alpha^*}^l [p_2(\alpha, t_n) p_1(\alpha, t_{n-1}) \dots p_1(\alpha, t_{n-1})] \}_{\alpha=0}.
 \end{aligned} \tag{C1}$$

#### 1. Moment $\langle \alpha^{2l} \rangle_p$

From identity (C1), the moment  $\langle \alpha^{2l} \rangle_p$  is given by the expression

$$\begin{aligned}
 \langle \alpha^{2l} \rangle_p &= \sum_{n=1}^{\infty} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\
 &\times \{ \partial_{\alpha^*}^{2l} [p_2(\alpha, t_n) p_1(\alpha, t_{n-1}) \dots p_1(\alpha, t_{n-1})] \}_{\alpha=0}.
 \end{aligned} \tag{C2}$$

The nonvanishing second derivative has to be applied to all  $n$  factors resulting in  $n$  terms again. The first term is

$$\begin{aligned}
 &\sum_{n=1}^{\infty} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\
 &\times \{ [ \partial_{\alpha^*}^{2l} p_2(\alpha, t_n) ] p_1(\alpha, t_{n-1}) \dots p_1(\alpha, t_{n-1}) \}_{\alpha=0},
 \end{aligned}$$

where the derivative is  $(-i\eta_2)^{2l} \langle (\cos \theta)^{2l} \rangle_2 e^{2il\nu n}$ , cf. Eq. (A1), so that it reads

$$\begin{aligned}
 &(-i\eta_2)^{2l} \langle (\cos \theta)^{2l} \rangle_2 \sum_{n=1}^{\infty} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 \\
 &\times w_n(t_n, \dots, t_1) e^{2il\nu n}.
 \end{aligned} \tag{C3}$$

The sum of the other  $n-1$  terms is

$$\begin{aligned}
 &\sum_{n=1}^{\infty} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\
 &\times \sum_{p=1}^{n-1} \{ p_2(\alpha, t_n) \\
 &\times p_1(\alpha, t_{n-1}) \dots [ \partial_{\alpha^*}^{2l} p_1(\alpha, t_p) ] \dots p_1(\alpha, t_{n-1}) \}_{\alpha=0} \\
 &= (-i\eta_1)^{2l} \langle (\cos \theta)^{2l} \rangle_1 \sum_{n=1}^{\infty} \sum_{p=1}^{n-1} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 \\
 &\times w_n(t_n, \dots, t_1) e^{2il\nu p},
 \end{aligned} \tag{C4}$$

where the derivative was identified as  $(-i\eta_1)^{2l} \langle (\cos \theta)^{2l} \rangle_1 e^{2il\nu p}$ .

In both expressions (C3) and (C4) the  $n$ -fold integral

$$\int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_p, \dots, t_1) e^{2il\nu p}$$

with  $p=1, \dots, n$  appears. Inserting the definition of the joint probability density for  $n$  spontaneous emissions (32), we find

$$\lambda_2 \lambda_1^{n-1} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w(t_n - t_{n-1}) \dots w(t_1 - t_0) e^{2il\nu p}. \tag{C5}$$

In the vein of Eq. (B2), the upper limits of the integrals are set to infinity. With substitution of the  $n$  integration variables,  $\tau_n = t_n - t_{n-1}$ , expression (C5) gives

$$\lambda_2 \lambda_1^{n-1} e^{2il\nu t_0} \int_0^{\infty} d\tau_n \dots \int_0^{\infty} d\tau_1 w(\tau_n) \dots w(\tau_1) e^{2il\nu(\tau_p + \dots + \tau_1)},$$

where  $t_p = \tau_p + \tau_{p-1} + \dots + \tau_1 + t_0$  is used. Given the definition of the spectral waiting-time distribution

$$\underline{w}(\omega) = \int_0^{\infty} d\tau w(\tau) e^{i\omega\tau}, \tag{C6}$$

and the normalization of the waiting-time distribution, the  $n$ -fold integral therefore results as

$$\begin{aligned}
 &\int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_p, \dots, t_1) e^{2il\nu p} \\
 &= \lambda_2 \lambda_1^{n-1} e^{2il\nu t_0} [ \underline{w}(2l\nu) ]^p.
 \end{aligned} \tag{C7}$$

When Eq. (C7) is inserted into the first term (C3), this term writes

$$\begin{aligned}
 &(-i\eta_2)^{2l} \langle (\cos \theta)^{2l} \rangle_2 e^{2il\nu t_0} \frac{\lambda_2}{\lambda_1} \sum_{n=1}^{\infty} [ \lambda_1 \underline{w}(2l\nu) ]^n \\
 &= (-i\eta_2)^{2l} \langle (\cos \theta)^{2l} \rangle_2 e^{2il\nu t_0} \frac{\lambda_2 \underline{w}(2l\nu)}{1 - \lambda_1 \underline{w}(2l\nu)}.
 \end{aligned}$$

The sum of the other  $n-1$  terms [Eq. (C4)] becomes

$$\begin{aligned}
& (-i\eta_1)^{2l} \langle (\cos \theta)^{2l} \rangle_1 e^{2il\nu_0} \sum_{n=1}^{\infty} \lambda_2 \lambda_1^{n-1} \sum_{p=1}^{n-1} [\underline{w}(2l\nu)]^p \\
&= (-i\eta_1)^{2l} \langle (\cos \theta)^{2l} \rangle_1 e^{2il\nu_0} \frac{\lambda_2}{\lambda_1} \sum_{n=1}^{\infty} \lambda_1^n \frac{w(2l\nu) - [w(2l\nu)]^n}{1 - w(2l\nu)} \\
&= (-i\eta_1)^{2l} \langle (\cos \theta)^{2l} \rangle_1 e^{2il\nu_0} \frac{\lambda_1 w(2l\nu)}{1 - \lambda_1 w(2l\nu)}.
\end{aligned}$$

Thus, the moment (C2) of the density of recoil shifts is

$$\begin{aligned}
\langle \alpha^{2l} \rangle_p &= (-1)^l \left( \eta_2^{2l} \langle (\cos \theta)^{2l} \rangle_2 + \frac{\lambda_1}{\lambda_2} \eta_1^{2l} \langle (\cos \theta)^{2l} \rangle_1 \right) \\
&\times \frac{\lambda_2 w(2l\nu)}{1 - \lambda_1 w(2l\nu)} e^{2il\nu_0}. \quad (\text{C8})
\end{aligned}$$

## 2. Moment $\langle \alpha^{*2} \alpha^2 \rangle_p$

From Eq. (C1) the moment  $\langle \alpha^{*2} \alpha^2 \rangle_p$  has the form

$$\begin{aligned}
\langle \alpha^{*2} \alpha^2 \rangle_p &= \sum_{n=1}^{\infty} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\
&\times \{ \partial_{\alpha}^2 \partial_{\alpha}^2 [p_2(\alpha, t_n) p_1(\alpha, t_{n-1}) \dots p_1(\alpha, t_1)] \}_{\alpha=0}.
\end{aligned}$$

The nonvanishing derivatives acting on a single factor  $p_a(\alpha, t)$  are those of second and fourth order, i.e., the derivatives  $\partial_{\alpha}^2 p_a$ ,  $\partial_{\alpha}^4 p_a$ ,  $\partial_{\alpha} \partial_{\alpha}^3 p_a$ , and  $\partial_{\alpha}^2 \partial_{\alpha}^2 p_a$ . Therefore, the moment can be written as

$$\begin{aligned}
\langle \alpha^{*2} \alpha^2 \rangle_p &= \sum_{n=1}^{\infty} \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) \\
&\times \left\{ \sum_{p=1}^n \partial_{\alpha}^2 \partial_{\alpha}^2 p_{a_p}(\alpha, t_p) + \sum_{p=1}^n \sum_{q \neq p} \{ [\partial_{\alpha}^* \partial_{\alpha} p_{a_p}(\alpha, t_p)] \right. \\
&\times [\partial_{\alpha}^* \partial_{\alpha} p_{a_q}(\alpha, t_q)] + [\partial_{\alpha}^2 p_{a_p}(\alpha, t_p)] \\
&\left. \times [\partial_{\alpha}^2 p_{a_q}(\alpha, t_q)] \right\}_{\alpha=0}, \quad (\text{C9})
\end{aligned}$$

where  $a_p=1$  for  $p=1, \dots, n-1$  and  $a_p=2$  for  $p=n$ . The fourth-order derivatives that corresponds to the first term in the braces, produce terms such as  $\eta_a^4 \langle (\cos \theta)^4 \rangle_a$ . They occur once with  $a=2$  and  $n-1$  times with  $a=1$ , to yield, after time integrations, the contribution

$$\eta_2^4 \langle (\cos \theta)^4 \rangle_2 + \frac{\lambda_1}{\lambda_2} \eta_1^4 \langle (\cos \theta)^4 \rangle_1.$$

The next term in the braces of Eq. (C9) results as the time-independent double sum

$$\begin{aligned}
& \sum_{p=1}^n \sum_{q \neq p} \eta_{a_p}^2 \eta_{a_q}^2 \langle (\cos \theta)^2 \rangle_{a_p} \langle (\cos \theta)^2 \rangle_{a_q} \\
&= 2 \eta_2^2 \eta_1^2 (n-1) \langle (\cos \theta)^2 \rangle_2 \langle (\cos \theta)^2 \rangle_1 \\
&\quad + \eta_1^4 (n-1)(n-2) \langle (\cos \theta)^2 \rangle_1^2,
\end{aligned}$$

which, after time integrations in Eq. (C9), adds to the moment the contribution

$$\begin{aligned}
& 2 \eta_2^2 \eta_1^2 (\langle \hat{n}_{\text{ph}} \rangle - 1) \langle (\cos \theta)^2 \rangle_2 \langle (\cos \theta)^2 \rangle_1 \\
&\quad + [\eta_1^4 (\langle \hat{n}_{\text{ph}} \rangle - 1) (\langle \hat{n}_{\text{ph}} \rangle - 2) + \eta_1^4 \langle (\Delta \hat{n}_{\text{ph}})^2 \rangle] \langle (\cos \theta)^2 \rangle_1^2 \\
&= 2 \eta_1^2 \frac{\lambda_1}{\lambda_2} \langle (\cos \theta)^2 \rangle_1 \left( \eta_2^2 \langle (\cos \theta)^2 \rangle_2 + \frac{\lambda_1}{\lambda_2} \eta_1^2 \langle (\cos \theta)^2 \rangle_1 \right).
\end{aligned}$$

The last term in the curved bracket of Eq. (C9) produces the double sum

$$\begin{aligned}
& \sum_{p=1}^n \sum_{q \neq p} \eta_{a_p}^2 \eta_{a_q}^2 \langle (\cos \theta)^2 \rangle_{a_p} \langle (\cos \theta)^2 \rangle_{a_q} e^{2i\nu(t_q - t_p)} \\
&= 2 \text{Re} \sum_{p=1}^n \sum_{q=1}^{p-1} \eta_{a_p}^2 \eta_{a_q}^2 \langle (\cos \theta)^2 \rangle_{a_p} \langle (\cos \theta)^2 \rangle_{a_q} e^{2i\nu(t_p - t_q)},
\end{aligned}$$

which generates the contribution

$$\begin{aligned}
& 2 \text{Re} \sum_{n=1}^{\infty} \sum_{p=1}^n \sum_{q=1}^{p-1} \eta_{a_p}^2 \eta_{a_q}^2 \langle (\cos \theta)^2 \rangle_{a_p} \langle (\cos \theta)^2 \rangle_{a_q} \\
&\quad \times \int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) e^{2i\nu(t_p - t_q)}. \quad (\text{C10})
\end{aligned}$$

The time integrals,

$$\int_{t_0}^{\infty} dt_n \dots \int_{t_0}^{t_2} dt_1 w_n(t_n, \dots, t_1) e^{2i\nu(t_p - t_q)},$$

with  $p > q$ , can be transformed as above into

$$\begin{aligned}
& \lambda_2 \lambda_1^{n-1} \int_0^{\infty} d\tau_n \dots \int_0^{\infty} d\tau_1 w(\tau_n) \dots w(\tau_1) e^{2i\nu(\tau_p + \dots + \tau_{q+1})} \\
&= \lambda_2 \lambda_1^{n-1} [w(2\nu)]^{p-q}.
\end{aligned}$$

Thus, the contribution (C10) becomes

$$\begin{aligned}
& 2 \text{Re} \sum_{n=1}^{\infty} \sum_{p=1}^n \sum_{q=1}^{p-1} \eta_{a_p}^2 \eta_{a_q}^2 \langle (\cos \theta)^2 \rangle_{a_p} \langle (\cos \theta)^2 \rangle_{a_q} \\
&\quad \times \lambda_2 \lambda_1^{n-1} [w(2\nu)]^{p-q}.
\end{aligned}$$

Using the substitution  $q \rightarrow q' = p - q$  and performing the sum over  $q'$ , we have

$$\begin{aligned}
& 2 \text{Re} \sum_{n=1}^{\infty} \sum_{p=1}^n \eta_{a_p}^2 \eta_1^2 \langle (\cos \theta)^2 \rangle_{a_p} \langle (\cos \theta)^2 \rangle_1 \lambda_2 \lambda_1^{n-1} \\
&\quad \times \frac{w(2\nu)}{1 - w(2\nu)} \{1 - [w(2\nu)]^{p-1}\}.
\end{aligned}$$

In the sum over  $p$  we separate the term with  $p=n$  from the terms  $p=1, \dots, n-1$  and obtain



$$2 \operatorname{Re} \sum_{n=1}^{\infty} \eta_1^2 \langle (\cos \theta)^2 \rangle_1 \lambda_2 \lambda_1^{n-1} \frac{w(2\nu)}{1-w(2\nu)} \\ \times \left( \eta_2^2 \langle (\cos \theta)^2 \rangle_2 \{1 - [w(2\nu)]^{n-1}\} \right. \\ \left. + \eta_1^2 \langle (\cos \theta)^2 \rangle_1 \left\{ (n-1) - \frac{1 - [w(2\nu)]^{n-1}}{1-w(2\nu)} \right\} \right).$$

The sum over  $n$  produces terms such as

$$\sum_{n=1}^{\infty} \lambda_2 (\lambda_1 x)^{n-1} = \frac{\lambda_2}{1 - \lambda_1 x},$$

where  $x$  may be 1 or  $w(2\nu)$ . Thus, after performing this sum, we arrive at

$$2 \operatorname{Re} \eta_1^2 \langle (\cos \theta)^2 \rangle_1 \frac{w(2\nu)}{1-w(2\nu)} \left\{ \eta_2^2 \langle (\cos \theta)^2 \rangle_2 \right. \\ \times \left[ 1 - \frac{\lambda_2}{1 - \lambda_1 w(2\nu)} \right] + \eta_1^2 \langle (\cos \theta)^2 \rangle_1 \\ \left. \times \left[ (\langle \hat{n}_{\text{ph}} \rangle - 1) - \frac{1}{1-w(2\nu)} \left( 1 - \frac{\lambda_2}{1 - \lambda_1 w(2\nu)} \right) \right] \right\}. \quad (\text{C11})$$

The brackets can be simplified to

$$\left[ 1 - \frac{\lambda_2}{1 - \lambda_1 w(2\nu)} \right] = \frac{\lambda_1 [1 - w(2\nu)]}{1 - \lambda_1 w(2\nu)},$$

and  $\langle \hat{n}_{\text{ph}} \rangle = 1/\lambda_2$ , see Eq. (B4), so that we obtain for the contribution (C11)

$$2 \eta_1^2 \frac{\lambda_1}{\lambda_2} \langle (\cos \theta)^2 \rangle_1 \left( \eta_2^2 \langle (\cos \theta)^2 \rangle_2 + \frac{\lambda_1}{\lambda_2} \eta_1^2 \langle (\cos \theta)^2 \rangle_1 \right) \\ \times \operatorname{Re} \left[ \frac{\lambda_2 w(2\nu)}{1 - \lambda_1 w(2\nu)} \right].$$

The complete results is therefore

$$\langle \alpha^{*2} \alpha^2 \rangle_p = \left[ \eta_2^4 \langle (\cos \theta)^4 \rangle_2 + \frac{\lambda_1}{\lambda_2} \eta_1^4 \langle (\cos \theta)^4 \rangle_1 \right] \\ + 2 \eta_1^2 \frac{\lambda_1}{\lambda_2} \langle (\cos \theta)^2 \rangle_1 \\ \times \left( \eta_2^2 \langle (\cos \theta)^2 \rangle_2 + \frac{\lambda_1}{\lambda_2} \eta_1^2 \langle (\cos \theta)^2 \rangle_1 \right) \\ \times \left\{ 1 + \operatorname{Re} \left[ \frac{\lambda_2 w(2\nu)}{1 - \lambda_1 w(2\nu)} \right] \right\}. \quad (\text{C12})$$

#### APPENDIX D: SPECTRAL WAITING-TIME DISTRIBUTION FOR RESONANT PUMPING

The waiting-time distribution  $w(t)$  is defined by

$$w(t) = (\gamma_1 + \gamma_2) |\langle 3 | \hat{U}_{\text{eff}}(t) | 1 \rangle|^2, \quad (\text{D1})$$

[Eq. (28)]. The time evolution  $\hat{U}_{\text{eff}}(t)$  is governed by the non-Hermitian Hamiltonian

$$\hat{H}_{\text{eff}} = \frac{\hbar}{2} (\kappa \hat{\sigma}_{1,+} + \kappa^* \hat{\sigma}_{1,-}) + \hbar \Delta |1\rangle \langle 1| - \frac{i\hbar}{2} \sum_{a=1,2} \gamma_a \hat{\sigma}_{a,+} \hat{\sigma}_{a,-} \quad (\text{D2})$$

[Eq. (9)]. The transition amplitude  $|1\rangle \rightarrow |3\rangle$  is the projection of the solution of the equation of motion

$$i\hbar \frac{\partial |\psi(t)\rangle}{\partial t} = \hat{H}_{\text{eff}} |\psi(t)\rangle, \quad (\text{D3})$$

on state  $|3\rangle$  with the initial condition  $|\psi(t=0)\rangle = |1\rangle$ . Equation (D3) is decomposed in the set of coupled equations for the components  $\psi_i(t) = \langle i | \psi(t) \rangle$ ,

$$\dot{\psi}_1(t) = -\frac{i\kappa^*}{2} \psi_3(t) - i\Delta \psi_1(t), \quad (\text{D4})$$

$$\dot{\psi}_2(t) = 0, \quad (\text{D5})$$

$$\dot{\psi}_3(t) = -\frac{i\kappa}{2} \psi_1(t) - \gamma \psi_3(t), \quad (\text{D6})$$

where  $\gamma = (\gamma_1 + \gamma_2)/2$ . The second derivative  $\ddot{\psi}_3(t)$  therefore results as

$$\ddot{\psi}_3(t) = -\frac{i\kappa}{2} \dot{\psi}_1(t) - \gamma \dot{\psi}_3(t),$$

which, with the use of Eq. (D4), writes in the resonant case ( $\Delta=0$ )

$$\left[ \partial_t^2 + \gamma \partial_t + \left| \frac{\kappa}{2} \right|^2 \right] \psi_3(t) = 0. \quad (\text{D7})$$

With the ansatz

$$\psi_3(t) = \phi_3(t) \exp \left[ -\frac{\gamma t}{2} \right],$$

the differential equation for  $\phi_3(t)$  is obtained,

$$\ddot{\phi}_3(t) = \left[ \left( \frac{\gamma}{2} \right)^2 - \left| \frac{\kappa}{2} \right|^2 \right] \phi_3(t). \quad (\text{D8})$$

The general solution is

$$\phi_3(t) = a e^{\lambda t} + b e^{-\lambda t}, \quad (\text{D9})$$

with

$$\lambda = \frac{\gamma}{2} \sqrt{1 - S}, \quad (\text{D10})$$

where  $S = |\kappa|^2 / \gamma^2$  is the laser-saturation parameter.

Thus, the time-evolved amplitude of state  $|3\rangle$  reads

$$\psi_3(t) = (a e^{\lambda t} + b e^{-\lambda t}) e^{-\gamma t/2}.$$

With the initial conditions  $\psi_3(0)=0$  and  $\psi_1(0)=1$ , one obtains, via Eq. (D6),  $\dot{\psi}_3(0) = -i\kappa/2$ , so that one finds the con-

stants of integration,  $a=-b=-i\kappa/(4\lambda)$ . Including these values, the above probability amplitude is

$$\psi_3(t) = \frac{\kappa}{i\gamma\sqrt{1-S}} \sinh\left(\frac{\gamma t}{2}\sqrt{1-S}\right) e^{-\gamma t/2}. \quad (\text{D11})$$

The waiting-time distribution  $w(t)=2\gamma|\psi_3(t)|^2$  is then given by

$$w(t) = \frac{2\gamma S}{|1-S|} \left| \sinh\left(\frac{\gamma t}{2}\sqrt{1-S}\right) \right|^2 e^{-\gamma t}. \quad (\text{D12})$$

Note that this distribution is normalized to unity,

$$\int_0^\infty dt w(t) = 1.$$

The spectral waiting-time distribution results from the Fourier transform of Eq. (D12),

$$\underline{w}(\omega) = \int_0^\infty dt w(t) e^{i\omega t}.$$

It is obtained by straightforward calculation as

$$\underline{w}(\omega) = \frac{\gamma^3 S}{(\gamma - i\omega)[\gamma^2(S-1) + (\gamma - i\omega)^2]} \times \begin{cases} 1, & S \leq 1, \\ (-1), & S > 1. \end{cases}$$

Thus, the required modulus and phase at twice the trap frequency become

$$\underline{w} = \frac{S \underline{w}^{(\text{sat})}}{\sqrt{(S-1)^2 + 2(S-1)(1-\tilde{\nu}^2) + (1+\tilde{\nu}^2)^2}}, \quad (\text{D13})$$

$$\tan \phi_{\underline{w}} = \frac{S+2-\tilde{\nu}^2}{S-3\tilde{\nu}^2} \tan \phi_{\underline{w}}^{(\text{sat})} \quad (\text{D14})$$

where the saturated values are

$$\underline{w}^{(\text{sat})} = \lim_{s \rightarrow \infty} \underline{w} = 1/\sqrt{1+\tilde{\nu}^2},$$

$$\phi_{\underline{w}}^{(\text{sat})} = \lim_{s \rightarrow \infty} \phi_{\underline{w}} = \arctan \tilde{\nu},$$

with  $\tilde{\nu}=2\nu/\gamma$ .

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- [49] Without loss of generality, the zero-energy level has been shifted here by  $-\hbar(\omega_1 + \omega_2)$  for simplicity.
- [50] This is a standard expansion, known in quantum optics by the term “unraveling” of the master equation.
- [51] This case is impossible to realize, at least for resonant pumping ( $\Delta=0$ ), as can be seen from the results of Appendix D.