

## Density-matrix equations and photon recoil for multistate atoms

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We derive a Fokker-Planck equation (FPE) for the center-of-mass Wigner function of an atom with an arbitrary level structure, moving in a given three-dimensional light field. This FPE is obtained by eliminating adiabatically the internal degrees of freedom from the expansion of the equation of motion of the atomic density operator into a power series of photon momentum. The force and the diffusion tensor in the FPE may be computed by solving a set of differential equations closely related to the time-evolution equations of the density matrix for the internal state of the atom, as encountered in the conventional recoilless spectroscopy. By studying spontaneous, induced, and free-evolution terms in the equations of motion of the density operator for various level configurations, we devise algorithms that enable a computer to build all the required equations automatically for an arbitrary atomic level scheme.

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### I. INTRODUCTION

Tunable lasers have fostered a surge of interest in the mechanical manifestations of light, and facilitated such landmark achievements as laser cooling of a single trapped ion [1], efficient longitudinal cooling of an atomic beam [2,3], optical molasses [4], optical trapping of neutral atoms [5], and crystallization of trapped ions [6,7]. At that time, until 1988, theory was thought to be ahead of experiments, in that the essential concepts of light pressure on a two-state atom [8–16] had been laid down by the early eighties. A comprehensive review of this work is given by Stenholm [17].

However, a major surprise was still forthcoming. All two-state cooling theories for which it has been possible to work out the final temperature unanimously agree that a lower limit for the achievable temperature is set by the linewidth  $\gamma$  of the optical transition used for laser cooling,

$$T_D = \hbar\gamma / k_B \quad (1.1)$$

to within a possible geometric factor close to unity. The experimental observation of temperatures nearly an order of magnitude below this Doppler limit by Lett *et al.* [18] and the subsequent confirmations [19,20] squarely challenged the prevailing theoretical understanding of light pressure.

It is now generally accepted that the temperatures below the Doppler limit result from the interplay between the position-dependent polarization in optical molasses and the internal level structure of real multistate atoms [21,22]. A quick argument says that multistate atoms possess an optical-pumping time scale  $\tau$  much longer than the relaxation time of a two-state system  $\gamma^{-1}$ , and  $\tau^{-1}$  should replace  $\gamma$  in the expression of the Doppler limit. More quantitative graphical models of polarization-gradient cooling have been presented [21,22], numerical studies have elucidated the effect of the optical-pumping time scale on the optical force [22],

analytical theories including both the force and the associated stochastic diffusion [21] have demonstrated temperatures far below those predicted by Eq. (1.1), and a spherical-tensor formalism for calculating the cooling force has been given [23]. Nonetheless, the original experimental configuration, three-dimensional optical molasses formed by three orthogonal pairs of counterpropagating laser beams, has remained beyond the reach of both analytical and numerical theories.

The purpose of the present paper is to outline a theory of light pressure with virtually no *a priori* restrictions regarding the level structure of the atom, the light field, or the motion of the atom. At all stages of our work we keep in mind the ultimate implementation of the theory on a computer.

Sodium, as prototypical for quantum optics experiments as is the two-state atom for theory, has a complicated level structure. For instance, if optical pumping is to be avoided in an experiment involving the  $D_2$  line, two different-frequency laser beams may be needed. The number of Zeeman sublevels of the hyperfine states involved in optical transitions then becomes 24, and in the worst case  $24^2 = 576$  density-matrix elements are needed to completely describe the internal state of the sodium atom. In quantum-optics experiments a lot of effort, tricks such as optical pumping, goes into keeping the multistate level structure from complicating the outcome; and conversely, some investigations account theoretically for the realistic Zeeman level structure of an atom [23–25]. However, no general scheme for handling the density-matrix equations for an arbitrary atomic level structure seems to have been formulated.

Alongside a comprehensive treatment of photon recoil, we therefore aim at the widest possible generality also in the case without recoil. It turns out that the conventional algebraic notation does not easily lend itself to cascades and V- and  $\Lambda$ -type configurations of atomic levels, and whatever combinations one might conceive of. Instead, we shall describe *algorithms* for creating the density-matrix equations of motion for any given atomic

level structure. The key idea is that we think of the time evolution of the density matrix as being generated by a Liouville operator or “supermatrix,” and fill in the elements of the supermatrix by working our way through the list of all possible *couplings* (spontaneous and induced) between the states. We have coined the attribute “coupling driven” for our approach.

Sections II and III concentrate on spontaneous and induced processes, respectively. We describe manifestations of light pressure, and devise coupling-driven algorithms for constructing the density-matrix equations of motion. In Sec. IV the focus is tighter on light pressure. We expand the atomic density operator into a power series of photon momentum, and eliminate adiabatically the internal degrees of freedom. As is well known, such a procedure leads to a Fokker-Planck equation (FPE) for the center-of-mass (c.m.) motion, which in turn identifies the light-pressure force and the diffusion that accounts for quantum fluctuations in the motion of the atom. We show how the computation of force and diffusion is reduced to solving a surprisingly simple set of differential equations closely related to the equations of motion of the density matrix of the atom in the *absence* of recoil.

Although the present paper is geared toward the FPE, we have occasionally given full sets of intermediate results that might serve as points of departure for alternative treatments. Moreover, to render the presentation reasonably self-contained and (hopefully) accessible to a reader who is not thoroughly familiar with the formal theory of light pressure, we have on some occasions presented explicit details of the derivations instead of just listing the (often subtle) differences from the earlier treatments. The price we have to pay is the bulk of the paper, which forces us to stop short of detailed discussions of the actual numerical calculations and their results. In Sec. V we briefly summarize the present paper, and sketch the follow-up work that will be reported in future presentations.

## II. RECOIL IN SPONTANEOUS EMISSION

Even if an atom is left on its own without any external driving field, it still is coupled to the vacuum of quantized electromagnetic fields. The ensuing spontaneous emission rearranges the internal state of the atom. Equally important for our purposes is that the departing spontaneous photon leaves behind a recoil kick that changes the c.m. motion. In this section we analyze the interplay of spontaneous emission, internal state, and c.m. motion for an atom with an arbitrary level structure.

Section II A starts from the microscopic Hamiltonian of the atom in the quantized radiation field. We derive a coupled set of Heisenberg equations of motion for suitable field and atomic operators, and subsequently eliminate the field operators with the aid of the conventional Markov and Born approximations [12,14,26,27]. We assume that the electromagnetic field is initially in the vacuum state. By taking the expectation values of the equations of motion for the atomic operators, we find relaxation terms for the reduced density operator that incorporates the internal state and the c.m. degrees of freedom of

the atom, but no more quantized fields.

The conventional algebraic notation is not well suited for writing down the relaxation terms in their full generality. Instead, in Sec. II B we discuss algorithms to streamline the generation of spontaneous-emission contributions to the density-matrix equations of motion.

In Sec. II C the development of recoil effects is resumed. Whereas in Sec. II A the c.m. motion is specified in the wave-vector representation (basically, the momentum representation), we now transform to the Wigner representation [11,14,28,29]. The density matrix is thereby converted into a form resembling a collection of distribution functions in the classical position-momentum phase space. Finally, as the thrust of this paper is toward the limit of small photon recoil, in Sec. II D we work out the expansion in photon momentum of spontaneous relaxation terms.

This Sec. II introduces two generalizations of the theory of spontaneous emission, as compared with the literature known to the author. First, we describe an explicit procedure to deal with an arbitrary atomic level scheme. Second, we retain the photon recoil for such an arbitrary level scheme. We arrive at Eqs. (2.27) and (2.41) below, which embody the randomness in the motion of the atom owing to the random directions of spontaneous recoil kicks. Only special cases of these results for certain field polarizations [13] and one-dimensional atomic motion [21] have been discussed before. For illustration, a few simple examples of spontaneous emission are given in Appendix A.

### A. Relaxation terms in wave-vector representation

Our starting point is the atom-field Hamiltonian with dipole coupling [29]

$$\hat{H} = \hat{H}_{\text{at}} + \hat{H}_f - \hat{\mathbf{d}} \cdot \hat{\mathbf{E}}(\hat{\mathbf{r}}). \quad (2.1)$$

We label internal atomic-energy eigenstates with doublets  $jm$ , where  $j$  incorporates angular momentum and possibly other quantum numbers such as the principal quantum number  $n$  in hydrogen, and  $m$  stands for the quantum number of the projection of the angular momentum onto the direction of the quantization axis. We refer to  $j$  as the “level,” and the doublet  $jm$  specifies a “state.” We distinguish between states both by adding primes as superscripts to  $j$  and  $m$ , and by subscripting  $j$  and  $m$  with numbers 1 and 2. The subscripts are a remnant of two-state notation: a level carrying the subscript 2 is assumed to be higher in energy, i.e., spontaneous emission always proceeds in the direction  $2 \rightarrow 1$ . On the other hand, the c.m. motion of the atom is represented using the wave vector  $\mathbf{k}$ . The unperturbed atomic Hamiltonian thus reads

$$\hat{H}_{\text{at}}/\hbar = \sum_{j,m,\mathbf{k}} [\varepsilon_m(j) + \varepsilon_{\mathbf{k}}] |jm\mathbf{k}\rangle \langle jm\mathbf{k}|. \quad (2.2)$$

We tacitly assume that the magnetic field  $\mathbf{B}$  (if any) is in the direction of the quantization axis, so that the internal-state eigenfrequencies can be written in terms of the zero-field value  $\omega_j$  and the Landé factor  $g(j)$  as

$$\varepsilon_m(j) = \omega_j + \frac{\mu_B g(j) B}{\hbar} m. \quad (2.3)$$

The frequency associated with the c.m. motion is given by the ordinary dispersion relation for a massive particle,

$$\varepsilon_{\mathbf{k}} = \frac{\hbar \mathbf{k}^2}{2M}. \quad (2.4)$$

The unperturbed field Hamiltonian is just

$$\hat{H}_f / \hbar = \sum_q \Omega_q \hat{b}_q^\dagger \hat{b}_q. \quad (2.5)$$

It is understood that the sum runs over both the wave vectors  $\mathbf{q}$  and the two polarizations  $\mathbf{e}_q^\lambda$  ( $\lambda=1,2$ ) associated with each wave vector. To conform to our operator notation we have equipped even the creation and annihilation operators of the photon modes  $q \equiv (\mathbf{q}, \lambda)$  with roofs. The corresponding electric-field operator at the position  $\mathbf{r}$  is written

$$\hat{\mathbf{E}}(\mathbf{r}) = \sum_q (\mathbf{g}_q e^{i\mathbf{q}\cdot\mathbf{r}} \hat{b}_q + \text{H.c.}), \quad (2.6)$$

where the expansion coefficient has the conventional expression in terms of the quantization volume and vacuum permittivity,

$$\mathbf{g}_q = \left[ \frac{\hbar \Omega_q}{2\varepsilon_0 V} \right]^{1/2} \mathbf{e}_q^\lambda. \quad (2.7)$$

The spectral representation of the dipole interaction is easily worked out to be

$$\begin{aligned} \frac{-\hat{\mathbf{d}} \cdot \hat{\mathbf{E}}(\hat{\mathbf{r}})}{\hbar} = & - \sum_{\substack{\mathbf{k}, q \\ j_\alpha, m_\alpha, j_\beta, m_\beta}} [\chi_{m_\alpha m_\beta}^{j_\alpha j_\beta}(q) \hat{b}_q |j_\alpha m_\alpha \mathbf{k} + \mathbf{q}\rangle \langle j_\beta m_\beta \mathbf{k}| \\ & + \text{H.c.}] , \end{aligned} \quad (2.8a)$$

with

$$\chi_{m_\alpha m_\beta}^{j_\alpha j_\beta}(q) = \frac{1}{\hbar} \langle j_\alpha m_\alpha | \hat{\mathbf{d}} \cdot \mathbf{g}_q | j_\beta m_\beta \rangle. \quad (2.8b)$$

However, while no such difference is as yet apparent in (2.8a), the rotating-wave approximation (RWA) makes a distinction between the upper and lower level of a transition. For instance, a photon annihilation operator will only appear in conjunction with atomic dyads that have the upper state of a transition to the left and the lower state to the right. Within the RWA, the interaction thus reads

$$\begin{aligned} & \frac{-\hat{\mathbf{d}} \cdot \hat{\mathbf{E}}(\hat{\mathbf{r}})}{\hbar} \\ & \approx - \sum_{\substack{\mathbf{k}, q \\ j_1, m_1, j_2, m_2}} [\chi_{m_2 m_1}^{j_2 j_1}(q) \hat{b}_q |j_2 m_2 \mathbf{k} + \mathbf{q}\rangle \langle j_1 m_1 \mathbf{k}| \\ & \quad + \text{H.c.}] . \end{aligned} \quad (2.9)$$

Here the sums are interpreted as coupling-driven: They run over all *pairs of states* with a dipole coupling, and the level with subscript 2 is taken to be higher in energy than the level with subscript 1. We have not adopted any special notation for such a concept, but it is implicit in our subsequent calculations.

For our purposes the crux of the dipole interaction is that the field in  $-\hat{\mathbf{d}} \cdot \hat{\mathbf{E}}(\hat{\mathbf{r}})$  is evaluated at the c.m. position  $\hat{\mathbf{r}}$ , which is a dynamical operator within the present theory. The interaction (2.9) therefore contains terms that change the c.m. wave vector. Photon recoil is included at the outset in our formalism.

We next define Heisenberg picture operators  $\hat{O}$  that at the (arbitrary) initial time  $t=0$  coincide with certain dyadic products of the atomic states,

$$\hat{O}(j_p m_p \mathbf{k}_p; j_q m_q \mathbf{k}_q; t=0) \equiv |j_p m_p \mathbf{k}_p\rangle \langle j_q m_q \mathbf{k}_q|. \quad (2.10)$$

Because the RWA makes a difference between the upper and the lower level of a transition, there are three basically different types of Heisenberg equations of motion for the operators  $\hat{O}$ . First consider levels  $j_1$  and  $j_1'$  both occurring as lower levels of some transitions,

$$\begin{aligned} \dot{\hat{O}}(j_1 m_1 \mathbf{k}_1; j_1' m_1' \mathbf{k}_1') = & i[\varepsilon_{m_1}(j_1) + \varepsilon_{\mathbf{k}_1} - \varepsilon_{m_1'}(j_1') - \varepsilon_{\mathbf{k}_1'}] \hat{O}(j_1 m_1 \mathbf{k}_1; j_1' m_1' \mathbf{k}_1') \\ & - i \sum_{j_2, m_2, q} [\chi_{m_2 m_1}^{j_2 j_1}(q) \hat{O}(j_2 m_2 \mathbf{k}_1 + \mathbf{q}; j_1' m_1' \mathbf{k}_1') \hat{b}_q - \chi_{m_1' m_2}^{j_1' j_2}(q) \hat{b}_q^\dagger \hat{O}(j_1 m_1 \mathbf{k}_1; j_2 m_2 \mathbf{k}_1' + \mathbf{q})] . \end{aligned} \quad (2.11a)$$

Second, suppose  $j_2$  and  $j_2'$  are both upper levels of transitions,

$$\begin{aligned} \dot{\hat{O}}(j_2 m_2 \mathbf{k}_2; j_2' m_2' \mathbf{k}_2') = & i[\varepsilon_{m_2}(j_2) + \varepsilon_{\mathbf{k}_2} - \varepsilon_{m_2'}(j_2') - \varepsilon_{\mathbf{k}_2'}] \hat{O}(j_2 m_2 \mathbf{k}_2; j_2' m_2' \mathbf{k}_2') \\ & - i \sum_{j_1, m_1, q} [\chi_{m_1 m_2}^{j_1 j_2}(q) \hat{b}_q^\dagger \hat{O}(j_1 m_1 \mathbf{k}_2 - \mathbf{q}; j_2' m_2' \mathbf{k}_2') - \chi_{m_2' m_1}^{j_2' j_1}(q) \hat{O}(j_2 m_2 \mathbf{k}_2; j_1 m_1 \mathbf{k}_2' - \mathbf{q}) \hat{b}_q] . \end{aligned} \quad (2.11b)$$

Third, consider coherences between levels  $j_1$  and  $j_2$ ,

$$\begin{aligned} \dot{\hat{O}}(j_1 m_1 \mathbf{k}_1; j_2 m_2 \mathbf{k}_2) = & i[\varepsilon_{m_1}(j_1) + \varepsilon_{\mathbf{k}_1} - \varepsilon_{m_2}(j_2) - \varepsilon_{\mathbf{k}_2}] \hat{O}(j_1 m_1 \mathbf{k}_1; j_2 m_2 \mathbf{k}_2) \\ & - i \sum_{j_2', m_2', q} [\chi_{m_2' m_1}^{j_2' j_1}(q) \hat{O}(j_2' m_2' \mathbf{k}_1 + \mathbf{q}; j_2 m_2 \mathbf{k}_2) \hat{b}_q + i \sum_{j_1', m_1', q} \chi_{m_2 m_1'}^{j_2 j_1'}(q) \hat{O}(j_1 m_1 \mathbf{k}_1; j_1' m_1' \mathbf{k}_2 - \mathbf{q}) \hat{b}_q] . \end{aligned} \quad (2.11c)$$

Finally, there are the Heisenberg equations of motion of the photon annihilation operators

$$\begin{aligned} \hat{b}_q &= -i\Omega_q \hat{b}_q \\ &+ i \sum_{\mathbf{k}} \chi_{m_1 m_2}^{j_1 j_2}(q) \hat{O}(j_1 m_1 \mathbf{k}; j_2 m_2 \mathbf{k} + \mathbf{q}) . \end{aligned} \quad (2.12)$$

The key idea of the derivation of spontaneous decay

terms is that each transition sends out a field that reacts with the atom, both at the same transition and at other (nearly degenerate) transitions. The first few steps are quite standard [14,26,27,30].

(i) (2.12) and its Hermitian conjugate are formally solved, and the results are inserted into (2.11).

(ii) Presently we assume that at the initial time  $t=0$  the field is in the vacuum state; all terms with  $\hat{b}_q(0)$  furthest to the right or  $\hat{b}_q^\dagger(0)$  furthest to the left are dropped because in the end they will not contribute to the expectation values of the operators  $\hat{O}$ .

(iii) The result now contains terms such as

$$\begin{aligned} \hat{O}(j_1 m_1 \mathbf{k}_1; j_1' m_1' \mathbf{k}_1') &= i[\varepsilon_{m_1}(j_1) + \varepsilon_{\mathbf{k}_1} - \varepsilon_{m_1'}(j_1') - \varepsilon_{\mathbf{k}_1'}] \hat{O}(j_1 m_1 \mathbf{k}_1; j_1' m_1' \mathbf{k}_1') + \dots \\ &+ \sum_{\substack{\mathbf{k}, q, j_2, m_2 \\ j_1'', m_1'', j_2'', m_2''}} \chi_{m_2 m_1}^{j_2 j_1}(q) \chi_{m_1' m_2''}^{j_1' j_2''}(q) \hat{O}(j_2 m_2 \mathbf{k}_1 + \mathbf{q}; j_1' m_1' \mathbf{k}_1'; t) \\ &\times \int_0^t dt' e^{-i\Omega_q(t-t')} \hat{O}(j_1'' m_1'' \mathbf{k}; j_2'' m_2'' \mathbf{k} + \mathbf{q}; t') , \end{aligned} \quad (2.13a)$$

$$\begin{aligned} \hat{O}(j_2 m_2 \mathbf{k}_2; j_1 m_1 \mathbf{k}_1) &= i[\varepsilon_{m_2}(j_2) + \varepsilon_{\mathbf{k}_2} - \varepsilon_{m_1}(j_1) - \varepsilon_{\mathbf{k}_1}] \hat{O}(j_2 m_2 \mathbf{k}_2; j_1 m_1 \mathbf{k}_1) + \dots \\ &- \sum_{\substack{\mathbf{k}, q, j_1', m_1' \\ j_1'', m_1'', j_2'', m_2''}} \chi_{m_2'' m_1'}^{j_2'' j_1'}(q) \chi_{m_1' m_2}^{j_1' j_2}(q) \int_0^t dt' e^{i\Omega_q(t-t')} \hat{O}(j_2'' m_2'' \mathbf{k} + \mathbf{q}; j_1' m_1' \mathbf{k}; t') \\ &\times \hat{O}(j_1' m_1' \mathbf{k}_2 - \mathbf{q}; j_1 m_1 \mathbf{k}_1; t) . \end{aligned} \quad (2.13b)$$

As usual, we argue that the sums over  $q$  are rapidly oscillating and only times  $t \sim t'$  contribute inside the integrals (Markov approximation). Hence, inside the integrals we replace the true evolution of  $\hat{O}(t')$  by the free evolution such as described by the first terms on the right-hand sides of Eqs. (2.13) (Born approximation).

(iv) The resulting equal-time products of two operators  $\hat{O}$  can be combined into one  $\hat{O}$  by using product rules obtained from the definition (2.10) and from the orthonormality of the states  $|jm\mathbf{k}\rangle$ . The rules are derived easily at  $t=0$ , and the Heisenberg picture time evolution sustains them for all times.

(v) We use the familiar decomposition

$$\int_0^\infty e^{ixt} dt = \pi \delta(x) + i \rho \frac{1}{x} . \quad (2.14)$$

(vi) In the resulting  $\delta$  functions and principal-value integrals we ignore the magnetic and c.m. energies in com-

parison with the optical-level energies  $\hbar\omega_j$ .

(vii) Under the stated assumptions, the (infinite) level shifts derived from the principal-value integrals implied by (2.14) turn out to be the same for all Zeeman states  $m$  within a given manifold  $j$ . From now on we assume that the level shifts are absorbed in the internal-state energies [31].

In the next step we take the expectation values of the resulting Heisenberg equations of motion of the atomic operators  $\hat{O}$ . There is a simple connection between the Heisenberg operators  $\hat{O}$  and the Schrödinger picture (reduced) density matrix  $\rho$  for the internal and c.m. degrees of freedom of the atom, e.g.,

$$\langle \hat{O}(j_1 m_1 \mathbf{k}_1; j_1' m_1' \mathbf{k}_1') \rangle = \rho(j_1' m_1' \mathbf{k}_1'; j_1 m_1 \mathbf{k}_1) . \quad (2.15)$$

We thus obtain spontaneous relaxation terms for the density matrix. For instance, a double-lower-state contribution becomes

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j_1 m_1 \mathbf{k}_1; j_1' m_1' \mathbf{k}_1') = \pi \sum_q [\delta(\Omega_q - \omega_{j_2 j_1}) + \delta(\Omega_q - \omega_{j_2' j_1'})] \chi_{m_1 m_2}^{j_1 j_2}(q) \rho(j_2 m_2 \mathbf{k}_1 + \mathbf{q}; j_2' m_2' \mathbf{k}_1' + \mathbf{q}) \chi_{m_2' m_1'}^{j_2' j_1'}(q) , \quad (2.16a)$$

where we have adopted the obvious notation  $\omega_{j_2 j_1} = \omega_{j_2} - \omega_{j_1}$ . The relaxation terms of density-matrix elements 22, 12, and 21 are analogous, such as

$$\begin{aligned} \frac{d}{dt} \Big|_{\text{sp}} \rho(j_2 m_2 \mathbf{k}_2; j_2' m_2' \mathbf{k}_2') = & \pi \sum_q \delta(\Omega_q - \omega_{j_2' j_1}) \chi_{m_2 m_1}^{j_2 j_1}(q) \chi_{m_1 m_2'}^{j_1 j_2'}(q) \rho(j_2' m_2' \mathbf{k}_2'; j_2' m_2' \mathbf{k}_2') \\ & + \pi \sum_q \delta(\Omega_q - \omega_{j_2' j_1}) \rho(j_2 m_2 \mathbf{k}_2; j_2' m_2' \mathbf{k}_2') \chi_{m_2' m_1}^{j_2' j_1}(q) \chi_{m_1 m_2}^{j_1 j_2'}(q). \end{aligned} \quad (2.16b)$$

We are now faced with the numerous directions in the problem: outgoing photon, c.m. wave vectors, quantization axis, and so forth. Our first step toward a resolution is to introduce spherical components of an arbitrary vector  $\mathbf{V}$  and spherical unit vectors (slightly unconventionally) as

$$V_{\pm} = \frac{1}{\sqrt{2}}(V_1 \pm iV_2), \quad V_0 = V_3; \quad (2.17)$$

$$\mathbf{e}_{\pm} = \frac{1}{\sqrt{2}}(\mathbf{e}_1 \pm i\mathbf{e}_2), \quad \mathbf{e}_0 = \mathbf{e}_3.$$

An arbitrary vector  $\mathbf{V}$  may be decomposed in the form

$$\mathbf{V} = \sum_{\sigma=-1,0,1} V_{\sigma} \mathbf{e}_{-\sigma}. \quad (2.18)$$

By virtue of the Wigner-Eckart theorem, spherical matrix elements of the dipole operator can be factored in the form

$$\langle j_2 m_2 | \hat{d}_{\sigma} | j_1 m_1 \rangle = D_{j_2 j_1} \langle m_2 | \sigma m_1 \rangle_{j_2 j_1}, \quad (2.19)$$

where  $D_{j_2 j_1} (= D_{j_1 j_2})$ , always chosen real, stands for the reduced matrix element of the transition  $j_1 \rightarrow j_2$ , and the bracketed factor is a shorthand for a certain Clebsch-Gordan coefficient [32], except for a  $\sigma$ -dependent sign,

$$\langle m_2 | \sigma m_1 \rangle_{j_2 j_1} = \begin{cases} -(j_1 m_1 | \sigma | j_1 j_2 m_2), & \sigma = 1 \\ (j_1 m_1 | \sigma | j_1 j_2 m_2), & \sigma = -1 \text{ or } 0. \end{cases} \quad (2.20)$$

We write either  $\langle m_2 | \sigma m_1 \rangle_{j_2 j_1}$  or  $\langle \sigma m_1 | m_2 \rangle_{j_1 j_2}$  as it suits us best, and mostly omit the angular-momentum subscripts as well. The coefficient  $\langle m_2 | \sigma m_1 \rangle$  character-

izes the transition from state  $m_1$  to state  $m_2$  caused by  $\sigma$ -polarized light, and is nonzero only if the indices satisfy  $m_1 + \sigma = m_2$ . However, injecting this piece of information into our notation would bring no particular advantage.

Combining (2.7), (2.8b), and (2.17)–(2.19) we have, say, the result

$$\begin{aligned} \chi_{m_1 m_2}^{j_1 j_2}(q) \chi_{m_2' m_1'}^{j_2' j_1'}(q) = & \frac{\Omega_q D_{j_1 j_2} D_{j_2' j_1'}}{2\hbar \epsilon_0 V} \\ & \times \sum_{\sigma, \sigma'} \langle m_1 \sigma | m_2 \rangle \langle m_2' | m_1' \sigma' \rangle \\ & \times (\mathbf{e}_{\sigma} \cdot \mathbf{e}_q^{\lambda})(\mathbf{e}_{-\sigma'} \cdot \mathbf{e}_q^{\lambda}). \end{aligned} \quad (2.21)$$

Comparison of (2.21) with Eqs. (2.16) shows that the expression on the right of (2.21) should basically be multiplied by a  $\delta$  function that dictates the photon energy, summed over the two polarization directions  $\lambda$ , summed (i.e., integrated) over photon energies, and summed over the directions of outgoing photons. We define the wave number of resonant photons

$$q_{j_2 j_1} = \frac{\omega_{j_2} - \omega_{j_1}}{c} \quad (2.22)$$

and the Einstein  $A$  coefficient

$$\Gamma_{j_2 j_1} = \frac{D_{j_2 j_1}^2 q_{j_2 j_1}^3}{3\pi \hbar \epsilon_0} \quad (2.23)$$

for the transition  $j_1 \rightarrow j_2$ , and carry out all but the final integral over the direction  $\mathbf{n} = \mathbf{q}/|\mathbf{q}|$  of the photons. The result is

$$\begin{aligned} \pi \sum_{\lambda, |\mathbf{q}|} \delta(\Omega_q - \omega_{j_2 j_1}) \chi_{m_1 m_2}^{j_1 j_2}(q) \chi_{m_2' m_1'}^{j_2' j_1'}(q) \\ = \frac{3}{16\pi} \left[ \frac{q_{j_2 j_1}}{q_{j_2' j_1'}} \right]^{3/2} \text{sgn}(D_{j_2 j_1} D_{j_2' j_1'}) (\Gamma_{j_2 j_1} \Gamma_{j_2' j_1'})^{1/2} \sum_{\sigma, \sigma'} \langle m_1 \sigma | m_2 \rangle \langle m_1' \sigma' | m_2' \rangle [\delta_{\sigma \sigma'} - (\mathbf{e}_{\sigma} \cdot \mathbf{n})(\mathbf{e}_{-\sigma'} \cdot \mathbf{n})], \end{aligned} \quad (2.24)$$

where  $\text{sgn}$  stands for the signum function. This form is immediately useful when tackling the ground-state-type relaxation (2.16a).

To make further progress with the excited-state relaxation term (2.16b) it is expedient to note the angular integral

$$\frac{3}{8\pi} \int d^2 n [\delta_{\sigma \sigma'} - (\mathbf{n} \cdot \mathbf{e}_{\sigma})(\mathbf{n} \cdot \mathbf{e}_{-\sigma'})] = \delta_{\sigma \sigma'} \quad (2.25)$$

and a sum property that follows from the orthonormality of the Clebsch-Gordan coefficients,

$$\sum_{m_1, \sigma} \langle m_2 | \sigma m_1 \rangle_{j_2 j_1} \langle \sigma m_1 | m_2' \rangle_{j_1 j_2} = \delta_{m_2 m_2'}. \quad (2.26)$$

With these ingredients, the final relaxation terms become

$$\frac{d}{dt} \Big|_{\text{sp}} \rho(j_1 m_1 \mathbf{k}_1; j_1' m_1' \mathbf{k}_1) = \sum_{j_2, m_2, j_2', m_2'} \text{sgn}(D_{j_2 j_1} D_{j_2' j_1'}) (\Gamma_{j_2 j_1} \Gamma_{j_2' j_1'})^{1/2} \times \int d^2 n K_{m_1 m_2; m_2' m_1'}^{j_1 j_2; j_2' j_1'}(\mathbf{n}) \rho(j_2 m_2, \mathbf{k}_1 + q_{j_2 j_1} \mathbf{n}; j_2' m_2', \mathbf{k}_1' + q_{j_2' j_1'} \mathbf{n}), \quad (2.27)$$

$$\frac{d}{dt} \Big|_{\text{sp}} \rho(j_2 m_2 \mathbf{k}_2; j_2' m_2' \mathbf{k}_2) = - \sum_{j_2'', m_2''} [C_{j_2 m_2; j_2'' m_2''} \rho(j_2'' m_2'' \mathbf{k}_2; j_2' m_2' \mathbf{k}_2) + \rho(j_2 m_2 \mathbf{k}_2; j_2'' m_2'' \mathbf{k}_2) C_{j_2'' m_2''; j_2' m_2'}], \quad (2.28)$$

$$\frac{d}{dt} \Big|_{\text{sp}} \rho(j_1 m_1 \mathbf{k}_1; j_2 m_2 \mathbf{k}_2) = - \sum_{j_2', m_2'} \rho(j_1 m_1 \mathbf{k}_1; j_2' m_2' \mathbf{k}_2) C_{j_2' m_2'; j_2 m_2}, \quad (2.29)$$

$$\frac{d}{dt} \Big|_{\text{sp}} \rho(j_2 m_2 \mathbf{k}_2; j_1 m_1 \mathbf{k}_1) = - \sum_{j_2', m_2'} C_{j_2 m_2; j_2' m_2'} \rho(j_2' m_2' \mathbf{k}_2; j_1 m_1 \mathbf{k}_1). \quad (2.30)$$

The angular-distribution kernel for spontaneous emission  $K$  is defined as

$$K_{m_1 m_2; m_2' m_1'}^{j_1 j_2; j_2' j_1'}(\mathbf{n}) = \frac{3}{8\pi} \sum_{\sigma, \sigma'} \langle m_1 \sigma | m_2 \rangle \langle m_2' | m_1' \sigma' \rangle [\delta_{\sigma \sigma'} - (\mathbf{n} \cdot \mathbf{e}_\sigma)(\mathbf{n} \cdot \mathbf{e}_{-\sigma'})], \quad (2.31a)$$

and the coupling chain  $C$  as

$$C_{j_2' m_2'; j_2 m_2} = \frac{1}{2} \sum_{\sigma, j_1, m_1} \text{sgn}(D_{j_2 j_1} D_{j_2' j_1'}) (\Gamma_{j_2 j_1} \Gamma_{j_2' j_1'})^{1/2} \langle m_2' | m_1 \sigma \rangle \langle m_1 \sigma | m_2 \rangle. \quad (2.31b)$$

In practice, cross-relaxation terms are significant only for level pairs for which  $\omega_{j_2 j_1} \sim \omega_{j_2' j_1'}$  holds true on a characteristic frequency scale (spontaneous damping rate, detuning, Rabi frequency) of matter-field interaction; otherwise, the radiation sent out by the transition  $j_1 \rightarrow j_2$  is far off resonance and inconsequential for the transition  $j_1' \rightarrow j_2'$ . Such a restriction is always understood in the sums in (2.27) and in (2.31b), and we have therefore dropped factors of the form  $(q_{j_2 j_1} / q_{j_2' j_1'})^{3/2}$  [cf. Eq. (2.24)].

### B. Algorithmic considerations

Our notation so far implicitly associates relaxation terms with atomic states that are one or the other of two alternatives, namely an upper or a lower state of a transition. Unfortunately, the notation does not easily lend itself to, say, cascade configurations, in which the intermediate level is simultaneously an upper and a lower level.

Instead, it is useful to ascend one level of generality, and rather describe an *algorithm* for generating the relaxation terms. The equation of motion of the density matrix is linear, and may be specified by giving a “supermatrix.” The elements of the supermatrix are labeled with pairs of density-matrix indices, which in turn consist of state pairs of the type  $(j_\alpha m_\alpha, j_\beta m_\beta)$ . Our aim is to interpret (2.27)–(2.30) as a template for building the relaxation-term elements of the supermatrix.

An inspection of (2.27)–(2.30) and their derivations [one particularly relevant detail is given explicitly in Eq. (2.13b)] reveals that in the most general case there are two different types of relaxation terms: (i) those for density-matrix elements in which the left or right state (or both) is treated as an upper state of a transition; (ii) those for density-matrix elements where both states are treated as lower states of transitions. The intermediate state of a cascade must simply receive both treatments. We therefore formulate an algorithm that loops over *transitions* rather than states, as follows.

(i) Specify all levels of the problem, and their coupling coefficients  $D_{j_1 j_2}$ ; for any coupled pair of levels retain a marker indicating which level is the upper one in this particular transition.

(ii) Divide the level pairs in groups such that the transition frequencies within each group are (nearly) degenerate. Notice that the statement “transition frequencies are nearly degenerate” can be taken to define an equivalence relation. The division is thus unique, and will destine each pair of levels to precisely one group. While forming the coefficients  $C$  and  $K$ , only transitions within the same group need be combined.

(iii) Working through all transitions, find all density-matrix elements whose left or right state is an upper state of a transition. For each such density-matrix element, form relaxation terms analogously to (2.28)–(2.30) and add them to the supermatrix. Notice that (2.28) is just a special case in which the substitution rule apparent in (2.29) and (2.30) is applied to both excited-state labels ap-

pearing on the left-hand side of (2.28).

(iv) Working through the equations of motion of the density-matrix elements whose left and right states both constitute lower states of some transitions, form the relaxation terms as in (2.27) and add them to the supermatrix.

In practice, in our coupling-driven approach this algorithm is turned inside out. The way it works can be appreciated by looking at, say, (2.29),

$$\frac{d}{dt} \Big|_{\text{sp}} \rho(j_1 m_1 \mathbf{k}_1; j_2 m_2 \mathbf{k}_2) = - \sum_{j'_2, m'_2} \rho(j_1 m_1 \mathbf{k}_1; j'_2 m'_2 \mathbf{k}_2) C_{j'_2 m'_2; j_2 m_2}.$$

Basically, from the known couplings  $D_{j_2 j_1}$  we first generate all nonzero chains of coupling  $C_{j'_2 m'_2; j_2 m_2}$ . Next, we generate a list of all possible states of the problem  $j_\alpha m_\alpha$ . Looping through all states  $j_\alpha m_\alpha$  and all chains  $C_{j'_2 m'_2; j_2 m_2}$ , we take the relaxation-term coupling of the density-matrix element  $\rho(j_\alpha m_\alpha \mathbf{k}_\alpha; j_2 m_2 \mathbf{k}_2)$  to the density

matrix element  $\rho(j_\alpha m_\alpha \mathbf{k}_\alpha; j'_2, m'_2, \mathbf{k}'_2)$ , namely  $C_{j'_2 m'_2; j_2 m_2}$ , and add it to the supermatrix element  $j_\alpha m_\alpha, j_2 m_2; j_\alpha m_\alpha, j'_2 m'_2$ . The relaxation terms for  $\rho(j_2 m_2 \mathbf{k}_2; j_\alpha m_\alpha \mathbf{k}_\alpha)$  are handled in exactly the same way. The remaining relaxation terms, rates-in with both states acting as the receiving states of transitions, are generated analogously from the list of coefficients  $K$ .

### C. Transformation to Wigner representation

It is common in light-pressure theory to express the density matrix in the Wigner representation (Refs. [8,9,11,14,28,29,33]) instead of the wave-vector representation (essentially the momentum representation) we have employed so far. Given an arbitrary function of two wave vectors  $f(\mathbf{k}_1, \mathbf{k}_2)$ , the corresponding Wigner function (WF) is defined through the transformation

$$f(\mathbf{r}, \mathbf{p}) = \frac{1}{(2\pi\hbar)^3} \int d^3q e^{i\mathbf{q}\cdot\mathbf{r}} f \left( \frac{\mathbf{p} + \mathbf{q}}{\hbar}, \frac{\mathbf{p} - \mathbf{q}}{\hbar} \right). \quad (2.32)$$

By simple manipulations the relaxation terms (2.27)–(2.30) are transformed to

$$\frac{d}{dt} \Big|_{\text{sp}} \rho(j_1 m_1; j'_1 m'_1; \mathbf{r}, \mathbf{p}) = \sum_{\substack{j_2 m_2 \\ j'_2 m'_2}} \text{sgn}(D_{j_2 j_1} D_{j'_2 j'_1}) (\Gamma_{j_2 j_1} \Gamma_{j'_2 j'_1})^{1/2} \int d^2n K_{m_1 m_2; m'_2 m'_1}^{j_1 j_2; j'_1 j'_2}(\mathbf{n}) \rho(j_2 m_2; j'_2 m'_2; \mathbf{r}, \mathbf{p} + \hbar \mathbf{q}_{j_2 j_1} \mathbf{n}), \quad (2.33)$$

$$\frac{d}{dt} \Big|_{\text{sp}} \rho(j_2 m_2; j'_2 m'_2; \mathbf{r}, \mathbf{p}) = - \sum_{j''_2, m''_2} [C_{j_2 m_2; j''_2 m''_2} \rho(j''_2 m''_2; j'_2 m'_2; \mathbf{r}, \mathbf{p}) + \rho(j_2 m_2; j''_2 m''_2; \mathbf{r}, \mathbf{p}) C_{j''_2 m''_2; j'_2 m'_2}], \quad (2.34)$$

$$\frac{d}{dt} \Big|_{\text{sp}} \rho(j_1 m_1; j_2 m_2; \mathbf{r}, \mathbf{p}) = - \sum_{j'_2, m'_2} \rho(j_1 m_1; j'_2 m'_2; \mathbf{r}, \mathbf{p}) C_{j'_2 m'_2; j_2 m_2}, \quad (2.35)$$

$$\frac{d}{dt} \Big|_{\text{sp}} \rho(j_2 m_2; j_1 m_1; \mathbf{r}, \mathbf{p}) = - \sum_{j'_2, m'_2} C_{j_2 m_2; j'_2 m'_2} \rho(j'_2 m'_2; j_1 m_1; \mathbf{r}, \mathbf{p}). \quad (2.36)$$

The advantage of the WF is that it is the closest counterpart of the classical phase-space density permitted by quantum mechanics [34–37]. The WF technically has the problem that it is not guaranteed to have the positivity properties of a true phase-space density. An interpretation along the lines of the probabilistic interpretation of the classical distribution function is therefore not permissible. Nonetheless, Eqs. (2.33) and (2.34) suggest the interpretation that an atom, while it decays from an excited state, receives a recoil kick opposite to the momentum of the photon. An atom starting in the excited level 2 with momentum  $\mathbf{p}$  will thus wind up in the lower level 1 with its momentum distributed over the sphere  $\mathbf{p} + \hbar \mathbf{q}_{j_2 j_1} \mathbf{n}$ . Here  $\mathbf{n}$  is a unit vector, whose direction is random and whose probability distribution is contained in the kernel  $K$ .

The new feature of multistate atoms, as opposed to the

traditional two-state model, is that during spontaneous emission a complicated change of the internal state takes place as well. The angular distribution in  $K$  depends on the states involved, indicating that the direction of the recoil photon and the rearrangement of the internal state are correlated. This might have been anticipated on the basis of conservation of angular momentum: the net change of the internal and c.m. angular momentum must compensate for the angular momentum taken away by the departing photon.

### D. Expansion in photon momentum

In the final step of our development of spontaneous-emission terms we assume that the photon momentum is small compared to the scale of momentum over which the WF's change considerably [8,9,11,14,33]. We carry

out the expansion

$$\rho(\mathbf{p} + \hbar \mathbf{q}_{j_2 j_1} \mathbf{n}) \approx \left[ 1 + \hbar \mathbf{q}_{j_2 j_1} \sum_{i=1}^3 n_i \frac{\partial}{\partial p_i} + \frac{1}{2} (\hbar \mathbf{q}_{j_2 j_1})^2 \sum_{i,j} n_i n_j \frac{\partial^2}{\partial p_i \partial p_j} + \dots \right] \rho(\mathbf{p}) \quad (2.37)$$

inside the integral in (2.33), whereupon the integral operator turns into a differential operator whose coefficients are deduced from the moments of  $K$ .

Using the explicit definition of the  $K$  coefficients, (2.31a), we find the zero-order moment of  $K$

$$\int d^2 n K_{m_1 m_2; m_2' m_1'}^{j_1 j_2; j_2' j_1'}(\mathbf{n}) = \sum_{\sigma} \langle m_1 \sigma | m_2 \rangle \langle m_2' | m_1' \sigma \rangle. \quad (2.38)$$

The first-order moments vanish by inversion symmetry, and the second-order moments are

$$\int d^2 n K_{m_1 m_2; m_2' m_1'}^{j_1 j_2; j_2' j_1'} n_i n_j = \sum_{\sigma, \sigma'} \langle m_1 \sigma | m_2 \rangle \langle m_2' | m_1' \sigma' \rangle \xi_{\sigma \sigma'}^{ij}, \quad (2.39a)$$

with

$$\xi_{\sigma \sigma'}^{ij} = \frac{1}{2} \delta_{\sigma \sigma'} \delta_{ij} - \frac{3}{8\pi} \int d^2 n (\mathbf{n} \cdot \mathbf{e}_{\sigma}) (\mathbf{n} \cdot \mathbf{e}_{-\sigma'}) n_i n_j. \quad (2.39b)$$

$$\frac{d}{dt} \Big|_{\text{sp}} \rho(j_1 m_1; j_1' m_1'; \mathbf{r}, \mathbf{p}) = \sum_{\sigma, \sigma'} \sum_{j_2, m_2; j_2', m_2'} \text{sgn}(D_{j_2 j_1} D_{j_2' j_1'}) (\Gamma_{j_2 j_1} \Gamma_{j_2' j_1'})^{1/2} \left[ \delta_{\sigma \sigma'} + \frac{1}{2} (\hbar \mathbf{q}_{j_2 j_1})^2 \sum_{i,j} \xi_{\sigma \sigma'}^{ij} \frac{\partial^2}{\partial p_i \partial p_j} \right] \times \langle m_1 \sigma | m_2 \rangle \rho(j_2 m_2; j_2' m_2'; \mathbf{r}, \mathbf{p}) \langle m_2' | m_1' \sigma' \rangle. \quad (2.41)$$

Other relaxation terms do not involve recoil at all, so (2.34)–(2.36) apply as they stand.

### III. RECOIL IN INDUCED PROCESSES

We now turn to induced absorption and emission in the equation of motion of the density operator, and also keep track of the free evolution of the internal states of the atom that persists even in the absence of electromagnetic fields.

In Sec. III A we write down the equations of motion of the density operator of the atom, internal and c.m. degrees of freedom included, for the case of a classical light field. To simplify the notation, we initially assume that there is only one laser frequency present in the system, and that the laser drives transitions between nearly degenerate sets of upper and lower levels. We convert the density-matrix equations into the Wigner representation, and again carry out an expansion in photon momentum.

Algorithms to build the free-evolution terms and the

TABLE I. Twenty-four independent coefficients  $\xi_{\sigma \sigma'}^{ij}$  that govern the correlations of the  $i$  and  $j$  (1, 2, or 3) components of the c.m. recoil and changes of the  $z$  component of internal angular momentum by  $\sigma$  and  $\sigma'$  ( $-1, 0,$  or  $+1$ ) in a process of spontaneous emission. These quantities are defined in Eq. (2.39b), and the remaining 57 coefficients  $\xi_{\sigma \sigma'}^{ij}$  may be generated by utilizing the symmetry relations (2.40).

$\xi_{+++}^{11} = \frac{3}{10}$	$\xi_{+0}^{11} = 0$	$\xi_{+-}^{11} = -\frac{1}{10}$	$\xi_{00}^{11} = \frac{2}{5}$
$\xi_{+++}^{12} = 0$	$\xi_{+0}^{12} = 0$	$\xi_{+-}^{12} = -\frac{i}{10}$	$\xi_{00}^{12} = 0$
$\xi_{+++}^{13} = 0$	$\xi_{+0}^{13} = -\frac{1}{10\sqrt{2}}$	$\xi_{+-}^{13} = 0$	$\xi_{00}^{13} = 0$
$\xi_{+++}^{22} = \frac{3}{10}$	$\xi_{+0}^{22} = 0$	$\xi_{+-}^{22} = \frac{1}{10}$	$\xi_{00}^{22} = \frac{2}{5}$
$\xi_{+++}^{23} = 0$	$\xi_{+0}^{23} = -\frac{i}{10\sqrt{2}}$	$\xi_{+-}^{23} = 0$	$\xi_{00}^{23} = 0$
$\xi_{+++}^{33} = \frac{2}{5}$	$\xi_{+0}^{33} = 0$	$\xi_{+-}^{33} = 0$	$\xi_{00}^{33} = \frac{1}{5}$

The coefficients  $\xi$  obviously have the following symmetries:

$$\xi_{\sigma \sigma'}^{ij} = \xi_{\sigma' \sigma}^{ji}, \quad (2.40a)$$

$$\xi_{\sigma' \sigma}^{ij} = (\xi_{\sigma \sigma'}^{ij})^*, \quad (2.40b)$$

$$\xi_{-\sigma -\sigma'}^{ij} = (\xi_{\sigma \sigma'}^{ij})^*. \quad (2.40c)$$

Only 24 of the 81 coefficients are independent. A full set of independent  $\xi$  coefficients is given in Table I.

When expanded up to second order in photon momentum, the relaxation term for a density-matrix element involving a pair of lower states of transitions, (2.33), finally becomes

induced terms of the density-matrix equations on a computer for an arbitrary atomic level structure are described in Sec. III B. Perhaps surprisingly, the RWA and the free evolution prove more delicate than the induced transitions themselves. Finally, in Sec. III C a few remarks are made on the connection between induced and spontaneous processes.

In addition to the algorithms, the main new result of Sec. III is Eq. (3.13), which turns out to be the key to the unexpectedly simple final results of the paper. In Appendix B we therefore discuss Eq. (3.13) from another viewpoint.

#### A. Derivation of induced recoil terms

For the time being we consider transitions driven by a classical external field between a nearly degenerate set of lower levels enumerated by the index  $j_1$ , and a nearly degenerate set of upper levels labeled by  $j_2$ . We factor out a representative average field frequency  $\omega$ , and write the field as a sum of plane wave modes,

$$\begin{aligned} \mathbf{E}(\mathbf{r}, t) &= \frac{1}{2} \sum_{\alpha} [E_{\alpha}(t) \mathbf{e}_{\alpha} e^{i(\mathbf{q}_{\alpha} \cdot \mathbf{r} - \omega t)} + \text{c.c.}] \\ &\equiv \frac{1}{2} [\mathcal{E}(\mathbf{r}, t) e^{-i\omega t} + \text{c.c.}] \end{aligned} \quad (3.1)$$

Here  $E_{\alpha}$ ,  $\mathbf{e}_{\alpha}$ , and  $\mathbf{q}_{\alpha}$  are the amplitude, polarization vector, and wave vector of the mode  $\alpha$ .  $\mathcal{E}(\mathbf{r}, t)$  is the positive-frequency part of the field, except that the dominant oscillations at the frequency  $\omega$  have been factored out.

The atomic Hamiltonian is exactly the same as in our study of spontaneous emission, viz. (2.2), except that in the present case  $\hat{\mathbf{E}}(\hat{\mathbf{r}})$  is an operator only through its dependence on the quantized c.m. position  $\hat{\mathbf{r}}$ . The interaction Hamiltonian is, again in the RWA,

$$\begin{aligned} \frac{-\hat{\mathbf{d}} \cdot \hat{\mathbf{E}}(\hat{\mathbf{r}})}{\hbar} &= - \sum_{\substack{\mathbf{k}, \alpha \\ j_1, m_1, j_2, m_2}} [\chi_{m_2 m_1}^{j_2 j_1}(\alpha) e^{-i\omega t} \\ &\quad \times |j_2 m_2 \mathbf{k} + \mathbf{q}_{\alpha}\rangle \langle j_1 m_1 \mathbf{k}| \\ &\quad + \text{H.c.}] \end{aligned} \quad (3.2)$$

The Rabi frequency for the transition  $j_1 m_1 \rightarrow j_2 m_2$  owing to the field mode  $\alpha$  is given in close analogy with (2.8b), (2.19), and (2.20) by

$$\chi_{m_2 m_1}^{j_2 j_1}(\alpha) = \frac{1}{2\hbar} \sum_{\sigma} D_{j_2 j_1} \langle m_2 | m_1 \sigma \rangle E_{\alpha} \mathbf{e}_{\alpha} \cdot \mathbf{e}_{-\sigma} \quad (3.3a)$$

and

$$\chi_{m_1 m_2}^{j_1 j_2}(\alpha) = [\chi_{m_2 m_1}^{j_2 j_1}(\alpha)]^* \quad (3.3b)$$

A representative component in the Liouville–von Neumann equation

$$\hat{\rho} = \frac{1}{i\hbar} [\hat{H}, \hat{\rho}] \quad (3.4)$$

for the density operator of the internal and c.m. degrees of freedom of the atom, in the wave-vector representation for the c.m. motion, runs as follows:

$$\begin{aligned} \dot{\rho}(j_2 m_2 \mathbf{k}_2; j_1 m_1 \mathbf{k}_1) &= -i[\Delta_{j_2 j_1} + \varepsilon_{m_2}(j_2) - \varepsilon_{m_1}(j_1) + \varepsilon_{\mathbf{k}_2} - \varepsilon_{\mathbf{k}_1}] \rho(j_2 m_2 \mathbf{k}_2; j_1 m_1 \mathbf{k}_1) \\ &\quad + i \sum_{j'_1, m'_1, \alpha, \sigma} \chi_{m_2 m_1}^{j_2 j'_1}(\alpha) \rho(j'_1 m'_1 \mathbf{k}_2 - \mathbf{q}_{\alpha}; j_1 m_1 \mathbf{k}_1) - i \sum_{j'_2, m'_2, \alpha, \sigma} \rho(j_2 m_2 \mathbf{k}_2; j'_2 m'_2 \mathbf{k}_1 + \mathbf{q}_{\alpha}) \chi_{m'_2 m_1}^{j'_2 j_1}(\alpha) \end{aligned} \quad (3.5)$$

In the customary way we have factored  $\exp(\pm i\omega t)$  out of density-matrix elements 12 and 21, so that, for instance,

$$\rho(j_2 m_2 \mathbf{k}_2; j_1 m_1 \mathbf{k}_1) \equiv e^{i\omega t} \langle j_2 m_2 \mathbf{k}_2 | \hat{\rho} | j_1 m_1 \mathbf{k}_1 \rangle \quad (3.6)$$

As a result, the detunings

$$\Delta_{j_2 j_1} = \omega_{j_2 j_1} - \omega \quad (3.7)$$

take the places of transition frequencies in the equations of motion.

In the next step we transform the Liouville–von Neumann equation into the Wigner representation using (2.32). The full set of WF equations reads

$$\begin{aligned} \frac{d}{dt} \rho(j_1 m_1; j'_1 m'_1; \mathbf{r}, \mathbf{p}) &= -i[\varepsilon_{m_1}(j_1) - \varepsilon_{m'_1}(j'_1)] \rho(j_1 m_1; j'_1 m'_1; \mathbf{r}, \mathbf{p}) + i \sum_{j_2, m_2, \alpha} \chi_{m_1 m_2}^{j_1 j_2}(\alpha) e^{-i\mathbf{q}_{\alpha} \cdot \mathbf{r}} \rho \left[ j_2 m_2; j'_1 m'_1; \mathbf{r}, \mathbf{p} + \frac{\hbar \mathbf{q}_{\alpha}}{2} \right] \\ &\quad - i \sum_{j_2, m_2, \alpha} \rho \left[ j_1 m_1; j_2 m_2; \mathbf{r}, \mathbf{p} + \frac{\hbar \mathbf{q}_{\alpha}}{2} \right] \chi_{m_2 m_1}^{j_2 j'_1}(\alpha) e^{i\mathbf{q}_{\alpha} \cdot \mathbf{r}} \end{aligned} \quad (3.8a)$$

$$\begin{aligned} \frac{d}{dt} \rho(j_2 m_2; j'_2 m'_2; \mathbf{r}, \mathbf{p}) &= -i[\varepsilon_{m_2}(j_2) - \varepsilon_{m'_2}(j'_2)] \rho(j_2 m_2; j'_2 m'_2; \mathbf{r}, \mathbf{p}) + i \sum_{j_1, m_1, \alpha} \chi_{m_2 m_1}^{j_2 j_1}(\alpha) e^{i\mathbf{q}_{\alpha} \cdot \mathbf{r}} \rho \left[ j_1 m_1; j'_2 m'_2; \mathbf{r}, \mathbf{p} - \frac{\hbar \mathbf{q}_{\alpha}}{2} \right] \\ &\quad - i \sum_{j_1, m_1, \alpha} \rho \left[ j_2 m_2; j_1 m_1; \mathbf{r}, \mathbf{p} - \frac{\hbar \mathbf{q}_{\alpha}}{2} \right] \chi_{m_1 m_2}^{j_1 j'_2}(\alpha) e^{-i\mathbf{q}_{\alpha} \cdot \mathbf{r}} \end{aligned} \quad (3.8b)$$

$$\begin{aligned} \frac{d}{dt} \rho(j_2 m_2; j_1 m_1; \mathbf{r}, \mathbf{p}) &= -i[\Delta_{j_2 j_1} + \varepsilon_{m_2}(j_2) - \varepsilon_{m_1}(j_1)] \rho(j_2 m_2; j_1 m_1; \mathbf{r}, \mathbf{p}) \\ &\quad + i \sum_{j'_1, m'_1, \alpha} \chi_{m_2 m_1}^{j_2 j'_1}(\alpha) e^{i\mathbf{q}_{\alpha} \cdot \mathbf{r}} \rho \left[ j'_1 m'_1; j_1 m_1; \mathbf{r}, \mathbf{p} - \frac{\hbar \mathbf{q}_{\alpha}}{2} \right] \\ &\quad - i \sum_{j'_2, m'_2, \alpha} \rho \left[ j_2 m_2; j'_2 m'_2; \mathbf{r}, \mathbf{p} + \frac{\hbar \mathbf{q}_{\alpha}}{2} \right] \chi_{m'_2 m_1}^{j'_2 j_1}(\alpha) e^{i\mathbf{q}_{\alpha} \cdot \mathbf{r}} \end{aligned} \quad (3.8c)$$

$$\begin{aligned}
\frac{d}{dt}\rho(j_1 m_1; j_2 m_2; \mathbf{r}, \mathbf{p}) &= -i[-\Delta_{j_2 j_1} + \varepsilon_{m_1}(j_1) - \varepsilon_{m_2}(j_2)]\rho(j_1 m_1; j_2 m_2; \mathbf{r}, \mathbf{p}) \\
&+ i \sum_{j'_2, m'_2, \alpha} \chi_{m_1 m_2}^{j_1 j'_2}(\alpha) e^{-i\mathbf{q}_\alpha \cdot \mathbf{r}} \rho \left[ j'_2 m'_2; j_2 m_2; \mathbf{r}, \mathbf{p} + \frac{\hbar \mathbf{q}_\alpha}{2} \right] \\
&- i \sum_{j'_1, m'_1, \alpha} \rho \left[ j_1 m_1; j'_1 m'_1; \mathbf{r}, \mathbf{p} - \frac{\hbar \mathbf{q}_\alpha}{2} \right] \chi_{m'_1 m_2}^{j'_1 j_2}(\alpha) e^{-i\mathbf{q}_\alpha \cdot \mathbf{r}}.
\end{aligned} \tag{3.8d}$$

In these expressions

$$\frac{d}{dt} = \frac{\partial}{\partial t} + \frac{\mathbf{p}}{M} \cdot \frac{\partial}{\partial \mathbf{r}} \tag{3.9}$$

is the convective derivative that accounts for the free flight of the center of mass. For instance, the position derivative in (3.8c) originates from the transformation to the Wigner representation of the part in (3.5) proportional to  $\varepsilon_{\mathbf{k}_2} - \varepsilon_{\mathbf{k}_1}$ .

Recoil effects in induced absorption and emission enter with the momentum displacements  $\pm \frac{1}{2} \hbar \mathbf{q}_\alpha$  in (3.8), including the somewhat peculiar factor  $\frac{1}{2}$ . Under the assumption that  $\hbar \mathbf{q}_\alpha$  is small compared to the characteristic momentum scale of the WF's, we may expand the WF's in photon momenta just as in the case of spontaneous emission. For instance,

$$\begin{aligned}
\frac{d}{dt}\rho(j_1 m_1; j'_1 m'_1; \mathbf{r}, \mathbf{p}) &= \dots + \dots - i \sum_{j_2, m_2, \alpha} \left[ 1 + \frac{\hbar}{2} \sum_i q_{\alpha, i} \frac{\partial}{\partial p_i} + \frac{\hbar^2}{8} \sum_{i, j} q_{\alpha, i} q_{\alpha, j} \frac{\partial^2}{\partial p_i \partial p_j} + \dots \right] \\
&\quad \times \rho(j_1 m_1; j_2 m_2; \mathbf{r}, \mathbf{p}) \chi_{m_2 m'_1}^{j_2 j'_1}(\alpha) e^{i\mathbf{q}_\alpha \cdot \mathbf{r}} \\
&= \dots + \dots - i \sum_{j_2, m_2} \left[ X_{m_2 m'_1}^{j_2 j'_1} - i \frac{\hbar}{2} \sum_i \left[ \frac{\partial}{\partial r_i} X_{m_2 m'_1}^{j_2 j'_1} \right] \frac{\partial}{\partial p_i} + \dots \right] \rho(j_1 m_1; j_2 m_2; \mathbf{r}, \mathbf{p}).
\end{aligned} \tag{3.10}$$

We have made use of (3.1), and defined the Rabi frequency of the field as it acts on the transition  $j_1 m_1 \rightarrow j_2 m_2$ ,

$$X_{m_2 m_1}^{j_2 j_1}(\mathbf{r}, t) = \frac{1}{2\hbar} \sum_{\sigma} D_{j_2 j_1} \langle m_2 | m_1 \sigma \rangle \mathbf{e}_{-\sigma} \cdot \mathcal{E}(\mathbf{r}, t) \tag{3.11a}$$

and

$$X_{m_1 m_2}^{j_1 j_2}(\mathbf{r}, t) = [X_{m_2 m_1}^{j_2 j_1}(\mathbf{r}, t)]^*. \tag{3.11b}$$

Position and momentum of the center of mass, represented by the  $\mathbf{r}$  and  $\mathbf{p}$  arguments of  $\rho$  in Eqs. (3.8), are quantum-mechanical dynamical variables. Nonetheless, we shall to an increasing extent think of them as classical parameters. Thus, apart from the positivity pitfall of the Wigner functions, we may formally interpret the set of Wigner functions  $\{\rho_{j_\alpha m_\alpha, j_\beta m_\beta}(\mathbf{r}, \mathbf{p})\}$  as the components of the density operator for the internal state of an atom that resides at the phase-space point  $(\mathbf{r}, \mathbf{p})$ . Throughout the rest of this paper we will use the tilde to emphasize the viewpoint that the operator in question pertains to the internal degrees of freedom of an atom residing at  $\mathbf{r}, \mathbf{p}$ . For instance, we define the dipole interaction operator through its matrix elements as

$$\tilde{V}_{j_\alpha m_\alpha, j_\beta m_\beta}(\mathbf{r}, t) = -\hbar X_{m_\alpha m_\beta}^{j_\alpha j_\beta}(\mathbf{r}, t). \tag{3.12}$$

With the new notation, the expansion of density-matrix

equations in photon momentum becomes

$$\begin{aligned}
\frac{d}{dt}\tilde{\rho} &= \frac{\partial}{\partial t}\tilde{\rho} \Big|_f - \frac{i}{\hbar} (\tilde{V}\tilde{\rho} - \tilde{\rho}\tilde{V}) + \frac{1}{2} \sum_i \left[ \frac{\partial \tilde{V}}{\partial r_i} \frac{\partial \tilde{\rho}}{\partial p_i} + \frac{\partial \tilde{\rho}}{\partial p_i} \frac{\partial \tilde{V}}{\partial r_i} \right] \\
&+ i \frac{\hbar}{8} \sum_{i, j} \left[ \frac{\partial^2 \tilde{V}}{\partial r_i \partial r_j} \frac{\partial^2 \tilde{\rho}}{\partial p_i \partial p_j} - \frac{\partial^2 \tilde{\rho}}{\partial p_i \partial p_j} \frac{\partial^2 \tilde{V}}{\partial r_i \partial r_j} \right] \\
&+ \dots
\end{aligned} \tag{3.13}$$

The first term on the right stands for the time evolution of the internal state in the absence of electromagnetic fields, i.e., for the time evolution due to the internal-state energies. In this formulation photon recoil enters via position derivatives of the interaction operator  $\tilde{V}$ , i.e., ultimately via the derivatives of the electric field.

## B. Algorithmic considerations

The development of the induced terms so far assumes that the transitions take place between (nearly) degenerate sets of lower and upper levels. We have thus been able to pull out a common frequency of the light, implement the RWA for all transitions at once, and write down the induced contributions to the WF equations without notational difficulties that would arise for, say, an intermediate state in a cascade.

The major complication in an attempt to generalize the

calculations to an arbitrary atomic coupling scheme has to do with RWA and free-evolution terms of the internal degrees of freedom of the atom. The idea associated with the RWA for a transition  $j_1 \rightarrow j_2$  is to subtract the characteristic “laser photon energy”  $\hbar\omega(j_2, j_1)$  from the energy of the upper level. Thus, if the lower level is redefined to have the energy 0, the upper level will have the energy  $\hbar\Delta_{j_2 j_1}$  determined by the detuning. If there is yet another level labeled  $j_3$  on top of level  $j_2$  and the corresponding transition is driven by a laser with frequency  $\omega(j_3, j_2)$ , we may repeat the RWA: the energy of level  $j_3$  will now lie by the amount  $\hbar\Delta_{j_3 j_2}$  above the energy of level  $j_2$ . Thus, if the energy of level  $j_1$  is fixed as 0, after the RWA’s the energy of level  $j_3$  is  $\hbar(\Delta_{j_2 j_1} + \Delta_{j_3 j_2})$ .

The way this process can go wrong may be appreciated in a scheme in which another independent intermediate level  $j'_2$  is present, and two more optical fields drive the cascade  $j_1 \rightarrow j'_2 \rightarrow j_3$ . Carrying out the RWA’s along both branches separately, we obtain *two* energies for the final state  $j_3$ ,

$$E_3 = \hbar(\Delta_{j_2 j_1} + \Delta_{j_3 j_2}) \quad (3.14a)$$

and

$$E'_3 = \hbar(\Delta_{j'_2 j_1} + \Delta_{j_3 j'_2}). \quad (3.14b)$$

Unless these energies coincide, we have come to a contradiction, and the RWA is not consistent. In fact, the energies coincide if and only if the explicit laser frequencies subtracted in conjunction with the RWA’s satisfy

$$\omega(j_3 j_2) + \omega(j_2 j_1) = \omega(j_3 j'_2) + \omega(j'_2 j_1). \quad (3.15)$$

Given the levels and the induced couplings between them, an algorithm to find the energies of the levels after the RWA might go as follows.

(i) Start from an arbitrary initial level, and call it a newly found level.

(ii) For all newly found levels, find all levels that are directly coupled to the newly found levels by a laser field. Two possibilities may arise. First, it may be that all ensuing coupled levels have been considered earlier in the process. In such a case all levels that can be coupled via induced processes to the initial level, directly or in multiple steps, have been found already. Second, new levels may be found that have not yet been encountered during the previous rounds of the algorithm. In such a case execute another round of the algorithm by using those coupled levels as the newly found levels.

(iii) If there are levels present that are not accounted for after the process of part (ii) has exited, choose one of the remaining levels as the new initial level and repeat the procedure (i), (ii).

In the end we will have divided the levels into groups so that there is a path of induced emissions and absorptions between every pair of levels within a group, and there are no induced-coupling pathways between the groups. As far as induced processes and RWA are concerned, each group is treated separately. The algorithm may now continue.

(iv) Within each group, arbitrarily assign the energy of one level as, say, 0.

(v) Starting from each newly assigned level, find all coupled levels that are accessible from the newly assigned levels by an induced one-photon transition. One of three possibilities arises regarding each coupled level thus found: First, the coupled level may have an energy assigned already, and the energy is incompatible with the result of the RWA starting from the current newly assigned level. In such a case the RWA is inconsistent, and the procedure is terminated with an error. Second, the coupled level may have an energy assigned that is compatible with the result of the RWA. In such a case the coupled level is skipped. Third, the coupled level has not yet been assigned an energy. In such a case, it is assigned the energy calculated from the energy of the current newly assigned level using the RWA. The coupled level then becomes a newly assigned level for the next round of the iteration.

The procedure terminates after assigning a unique energy to every level, or fails if the RWA cannot be made consistently. However, the expansion coefficient  $\mathcal{E}(\mathbf{r}, t)$  in (3.1) is permitted to depend on time, so the average laser frequencies [denoted by  $\omega$  in (3.1)] can always be redefined in such a way that a global RWA can be made. On the other hand, if each transition in the atom is driven by a monochromatic field, it may be possible to choose the average frequencies in the RWA in such a way that they absorb all time dependence of the fields. In the “rotating frame” the atom-field interaction matrix  $\tilde{V}$  then becomes independent of time. Unless otherwise noted, we tacitly assume this in everything that follows.

In comparison with the free evolution, the induced contributions in the equation of motion of the density operator are easily handled. We consider the recoilless case only. We have, for instance, terms of the form

$$\begin{aligned} & \frac{d}{dt} \rho(j_1 m_1; j'_1 m'_1) \\ & = \cdots - i \sum_{j_2, m_2} [\rho(j_1 m_1; j_2 m_2) X_{m_2 m'_1}^{j_2 j'_1} \\ & \quad - X_{m_1 m_2}^{j_1 j_2} \rho(j_2 m_2; j'_1 m'_1)]. \end{aligned} \quad (3.16)$$

This turns out to be a special case of the general rule: a given  $\rho(j_\alpha m_\alpha; j_\beta m_\beta)$  is coupled to all density-matrix elements of the form  $\rho(j_\gamma m_\gamma; j_\beta m_\beta)$  via the coefficients  $iX_{m_\alpha m_\gamma}^{j_\alpha j_\gamma}$ , and to all  $\rho(j_\alpha m_\alpha; j_\gamma m_\gamma)$  via the coefficients  $-iX_{m_\gamma m_\beta}^{j_\gamma j_\beta}$ . This observation immediately translates into a coupling-driven algorithm to build the induced terms in the equation of motion of the density operator, no matter what is the configuration of atomic levels.

### C. Induced versus spontaneous processes

In the spirit of semiclassical theory of laser spectroscopy we assume that, whether a classical driving field is present or not, spontaneous relaxation is described by the same decay constants. This approximation presumably is

good at the laser intensities used in laser cooling and trapping, but it interferes in a subtle way with the general RWA.

The RWA is an issue in connection with induced processes, but ostensibly not so with spontaneous emission: One has to account for the frequency of the external driving field, while for the purpose of discussing atomic relaxation terms, spontaneously emitted radiation may be regarded as having the resonance frequency of the transition. The redefinition of level energies accompanying the RWA for induced processes in itself does not invalidate spontaneous relaxation terms. For instance, an excited state may clearly be taken to decay to a lower state at the same rate before and after the redefinition.

However, spontaneous relaxations of nearly degenerate transitions are entangled, and how this interference manifests itself depends on the differences of the transition frequencies. The redefinition of level energies that goes with the RWA must preserve the differences of the transition frequencies within each group of transitions whose frequencies are treated as nearly degenerate in spontaneous emission. For example, if one lower level and several upper levels make a set of near-degenerate transitions, the same photon energy must be subtracted from all excited-state energies.

#### IV. ADIABATIC ELIMINATION OF INTERNAL EVOLUTION

We are ultimately interested in the position and momentum distributions of an atom, and their slow modifications brought about by photon recoil. Our plan, then, is to *eliminate adiabatically* the evolution of the internal degrees of freedom of the atom, and thereby obtain a closed evolution equation for the center-of-mass Wigner function.

We start in Sec IV A by defining a projection superoperator  $\mathcal{P}$  that selects the c.m. WF out of the collection of all WF's  $\tilde{\rho}$ , and the complementary projection  $\mathcal{Q}=1-\mathcal{P}$ . We then write down the equations of motion for  $\mathcal{P}\tilde{\rho}$  and  $\mathcal{Q}\tilde{\rho}$ , and eliminate  $\mathcal{Q}\tilde{\rho}$  adiabatically. The result is a closed operator equation for the projection  $\mathcal{P}\tilde{\rho}$ , valid to second order in photon momentum.

The operator formalism pursued in Sec. IV A is handy in systematic derivations, but not immediately useful for practical calculations. The goal of our subsequent manipulations is to relate the complicated operator objects encountered in Sec. IV A to solutions of differential equations. The general plan is introduced in Sec. IV B, while the application to the equation of  $\mathcal{P}\tilde{\rho}$  and the additional approximations made to simplify the outcome are detailed in Secs. IV C and IV D.

In Sec. IV E we are ready to write down the Fokker-Planck equation (FPE) for the c.m. WF of the atom. The force and the diffusion tensor in the FPE are indeed expressed in terms of solutions of certain differential equations, which are variants of the density-matrix equations one has to solve in the conventional recoilless theory of atom-field interactions. Finally, in Sec. IV F we discuss the FPE in detail.

The main new result of this Sec. IV is the method to

calculate the force and the diffusion tensor expressed in Eqs. (4.63)–(4.65). It may be noted that the present section is closely patterned after Refs. [15] and [38] concerned with a trapped two-state ion. The key new innovations are the form of the projector  $\mathcal{P}$  in Eq. (4.1) and the recoil expansion in Eq. (3.13), which together lead us to the unexpectedly compact results. Nonetheless, the added generality of the present paper calls for an extensive fine tuning of the previous argument. We have therefore written this Sec. IV from the ground up, so that familiarity with Refs. [15] and [38] is not assumed.

##### A. Formal equation of the center-of-mass motion

Inasmuch as recoil effects are concerned, the quantity of immediate interest is the sum of the populations of the internal states of the atom. To gain access to it, we define the superoperator  $\mathcal{P}$  that acts on an arbitrary atomic operator  $\tilde{\rho}$  in the following way:

$$\mathcal{P}\tilde{\rho} = \frac{\bar{1}}{N} \text{Tr}\tilde{\rho} . \quad (4.1)$$

Here, like in everything that follows, the operators acting on the internal degrees of freedom of the atom are denoted by tildes, and the conventional operator product, trace, etc., are implied. In particular,  $\bar{1}$  is the unit operator for the internal degrees of freedom of the atom. These are quantum operators acting on atomic state vectors whose components are labeled by level-state doublets  $j_\alpha m_\alpha$ , so that in practice the components of operators could be labeled by state pairs such as  $j_\alpha m_\alpha, j_\beta m_\beta$ . The components of a superoperator acting on operators are then labeled with pairs of state pairs, and the superoperator  $\mathcal{P}$  reads in its full glory

$$\mathcal{P}_{j_\alpha m_\alpha, j_\beta m_\beta; j_\delta m_\delta, j_\gamma m_\gamma} = \frac{1}{N} \delta_{j_\alpha m_\alpha, j_\beta m_\beta} \delta_{j_\delta m_\delta, j_\gamma m_\gamma} . \quad (4.2)$$

$N$  is the total number of internal states included in the description of the atom.

$\mathcal{P}$  is a projector, i.e., its own square,

$$\mathcal{P}\mathcal{P} = \mathcal{P} . \quad (4.3)$$

By the same token we define the projector

$$\mathcal{Q} = 1 - \mathcal{P} , \quad (4.4)$$

whose effect is to render its argument operator  $\tilde{\rho}$  traceless by subtracting  $\text{Tr}(\tilde{\rho})/N$  from each diagonal element. Of course,  $\mathcal{Q}\mathcal{P} = \mathcal{P}\mathcal{Q} = 0$ .

Now consider the action of the superoperator  $\mathcal{P}$  on the matrix of atomic WF's,  $\tilde{\rho}(\mathbf{r}, \mathbf{p})$ . ( $\mathcal{P}\tilde{\rho} = \bar{1} \text{Tr}\tilde{\rho}/N$  is an atomic operator proportional to the unit operator  $\bar{1}$ , and carries information worth just one real number. But this piece of information is exactly what we are after, for  $\text{Tr}[\tilde{\rho}(\mathbf{r}, \mathbf{p})]$  is the WF of the c.m. motion of the atom at the phase-space point  $\mathbf{r}, \mathbf{p}$ , irrespective of the internal state.

The equation of motion of the atom up to second order in photon momentum formally reads

$$\frac{\partial}{\partial t} \tilde{\rho} = (\mathcal{D} + \mathcal{L}^{(0)} + \mathcal{L}^{(1)} + \mathcal{L}^{(2)}) \tilde{\rho} , \quad (4.5)$$

where we have defined

$$\mathcal{D} = -\frac{\mathbf{p}}{M} \cdot \frac{\partial}{\partial \mathbf{r}}. \quad (4.6a)$$

For notational convenience we have elevated the combination of position derivatives in (4.6a) to a status of a superoperator. This simply entails that, with respect to the matrix structure inherited from the internal states of the atom,  $\mathcal{D}$  is proportional to the unit superoperator. In other words,  $\mathcal{D}$  treats all components of its argument operator the same way,

$$(\mathcal{D}\bar{\sigma})_{j_\alpha m_\alpha, j_\beta m_\beta} = -\frac{\mathbf{p}}{M} \cdot \frac{\partial}{\partial \mathbf{r}} \bar{\sigma}_{j_\alpha m_\alpha, j_\beta m_\beta}. \quad (4.6b)$$

The expressions of the remaining superoperators may be deduced from the previous sections, but we do not write them down because we will simplify the formulas as we go along. For the time being it is sufficient to bear in mind that  $\mathcal{L}^{(p)}$  corresponds to  $p$ th order in recoil effects. We use the recoil velocity of a transition,  $v_r$ , as a symbolic expansion parameter, so that  $\mathcal{L}^{(p)} \propto v_r^p$ . To simplify our notation we assume that none of the Liouville operators  $\mathcal{L}^{(p)}$  depends explicitly on time, which is tantamount to the assumption that the RWA has absorbed all explicit time dependence of the driving fields.

We next insert the identity  $\mathcal{P}\bar{\rho} + Q\bar{\rho} = \bar{\rho}$  into the right-hand side of (4.5), and multiply the result from the left by  $\mathcal{P}$ . To simplify the expressions further we first note that  $\mathcal{L}^{(0)}$  is the time-evolution operator of the internal state in the absence of photon recoil, so that the time-evolution equation for the internal-state density operator reads

$$\dot{\bar{\rho}} = \mathcal{L}^{(0)} \bar{\rho}. \quad (4.7)$$

Since the trace of the density operator is preserved in the evolution governed by  $\mathcal{L}^{(0)}$ , we obtain

$$\frac{\partial}{\partial t} \text{Tr}(\bar{\rho}) = \text{Tr}(\dot{\bar{\rho}}) = \text{Tr}(\mathcal{L}^{(0)} \bar{\rho}) = 0, \quad (4.8)$$

for all density operators  $\bar{\rho}$ . But by linearity we then have

$$\mathcal{P}\mathcal{L}^{(0)}\bar{\sigma} = \frac{\tilde{1}}{N} \text{Tr}(\mathcal{L}^{(0)}\bar{\sigma}) = 0 \quad (4.9)$$

for all atomic operators  $\bar{\sigma}$ , not just density operators. All told, we have proven the superoperator identity

$$\mathcal{P}\mathcal{L}^{(0)} = 0. \quad (4.10a)$$

Similarly, by (3.13), the second-order recoil contribution from induced processes preserves the trace of the internal-state density matrix, so that only the spontaneous contribution survives the trace in  $\mathcal{P}$  and gives

$$\mathcal{P}\mathcal{L}^{(2)} = \mathcal{P}\mathcal{L}_{\text{sp}}^{(2)}. \quad (4.10b)$$

Next, as it comes to the internal state, (3.13) shows that  $\mathcal{L}^{(1)}\mathcal{P}$  is proportional to the dipole operator. But in an atom parity is a good quantum number and the dipole operator is traceless, so we obtain

$$\mathcal{P}\mathcal{L}^{(1)}\mathcal{P} = 0. \quad (4.10c)$$

Noting that

$$[\mathcal{P}, \mathcal{D}] = 0, \quad \left[ \mathcal{P}, \frac{\partial}{\partial t} \right] = 0, \quad (4.10d)$$

we finally have

$$\begin{aligned} \frac{\partial}{\partial t} \mathcal{P}\bar{\rho} &= \mathcal{P}\mathcal{D}\mathcal{P}\bar{\rho} + \mathcal{P}\mathcal{L}^{(1)}Q\bar{\rho} + \mathcal{P}\mathcal{L}_{\text{sp}}^{(2)}\mathcal{P}\bar{\rho} \\ &\quad + \mathcal{P}\mathcal{L}_{\text{sp}}^{(2)}Q\bar{\rho}. \end{aligned} \quad (4.11)$$

In an analogous manner, by multiplying (4.5) from the left by  $Q$ , we obtain

$$\begin{aligned} \frac{\partial}{\partial t} Q\bar{\rho} &= Q\mathcal{D}Q\bar{\rho} + Q\mathcal{L}^{(0)}\mathcal{P}\bar{\rho} + Q\mathcal{L}^{(0)}Q\bar{\rho} \\ &\quad + Q\mathcal{L}^{(1)}\mathcal{P}\bar{\rho} + Q\mathcal{L}^{(1)}Q\bar{\rho}. \end{aligned} \quad (4.12)$$

We have written (4.12) up to first order in  $v_r$  only, because the coupling of  $Q\bar{\rho}$  back to  $\mathcal{P}\bar{\rho}$  in (4.11) already involves at least one power of  $v_r$  and our final results below will come out consistent to second order in  $v_r$ .

We are now going to embark on increasingly complicated maneuvers with operators and superoperators. It is useful to bear in mind that, barring mathematical pathologies which are fortunately infrequent in physics, the usual rules of algebra, analysis, and calculus apply to operators just as they apply to scalar quantities. The important exception is that any manipulation depending on the commutativity of different operators is forbidden, unless the operators actually commute. Another common technique, already used above in the derivation of (4.10a), is to demonstrate operator equations by showing that the action of the presumably equal operators on an arbitrary argument vector is the same.

Thus, by direct insertion it is easy to show that, given arbitrary time independent operators  $\tilde{F}(\mathbf{r}, \mathbf{p})$  and  $\tilde{U}(\mathbf{r}, \mathbf{p})$ , the quantities formally defined as

$$\mathcal{P}\bar{\rho} = e^{t\mathcal{P}\mathcal{D}\mathcal{P}}\tilde{F}, \quad (4.13a)$$

$$Q\bar{\rho} = e^{tQ(\mathcal{D} + \mathcal{L}^{(0)})Q}\tilde{U}$$

$$+ \left[ \int_0^t dt' e^{(t-t')Q(\mathcal{D} + \mathcal{L}^{(0)})Q} Q\mathcal{L}^{(0)}\mathcal{P}e^{t'\mathcal{P}\mathcal{D}\mathcal{P}} \right] \tilde{F} \quad (4.13b)$$

satisfy Eqs. (4.11) and (4.12) to zeroth order in  $v_r$ . We will frequently use the shorthand notation

$$\mathcal{T}(t) = e^{tQ(\mathcal{D} + \mathcal{L}^{(0)})Q}, \quad (4.13c)$$

$$\mathcal{M}(t) = e^{t\mathcal{P}\mathcal{D}\mathcal{P}}$$

for the two oft-repeated operator exponentials.

Conversely, if we let  $\tilde{F}$  and  $\tilde{U}$  depend on time and attempt to write an ansatz of the form (4.13) for the solution of (4.11) and (4.12), the time dependence of  $\tilde{F}$  and  $\tilde{U}$  must be at least of the order  $v_r$ . Indeed, Eqs. (4.11) and (4.13a) give right away

$$\frac{\partial \tilde{F}}{\partial t} = e^{-t\mathcal{P}\mathcal{D}\mathcal{P}} [\mathcal{P}\mathcal{L}^{(1)}Q\bar{\rho} + \mathcal{P}\mathcal{L}_{\text{sp}}^{(2)}(\mathcal{P}\bar{\rho} + Q\bar{\rho})]. \quad (4.14)$$

We next insert (4.13b) into (4.12). Using (4.14), and without any other approximation than restriction to first order in  $v_r$ , we find the equation

$$\left[ \frac{\partial}{\partial t} - \mathcal{R} \right] \tilde{U}(t) = \left[ \mathcal{T}(-t) \left[ Q\mathcal{L}^{(1)}\mathcal{P}\mathcal{M}(t) + Q\mathcal{L}^{(1)}Q \int_0^t dt' \mathcal{T}(t-t') Q\mathcal{L}^{(0)}\mathcal{P}\mathcal{M}(t') \right] \right. \\ \left. - \int_0^t dt' \mathcal{T}(-t') Q\mathcal{L}^{(0)}\mathcal{P}\mathcal{M}(t'-t) \mathcal{P}\mathcal{L}^{(1)}Q \int_0^t dt'' \mathcal{T}(t-t'') Q\mathcal{L}^{(0)}\mathcal{P}\mathcal{M}(t'') \right] \tilde{F}(t), \quad (4.15a)$$

with

$$\mathcal{R} = \mathcal{T}(-t) \left[ Q\mathcal{L}^{(1)}Q - \int_0^t dt' \mathcal{T}(t-t') Q\mathcal{L}^{(0)}\mathcal{P}\mathcal{M}(t'-t) \mathcal{P}\mathcal{L}^{(1)}Q \right] \mathcal{T}(t). \quad (4.15b)$$

In order to proceed without entering the probably extremely tedious mathematical proofs, we at this point make a few heuristic arguments inspired by the properties of the exponential functions of ordinary scalar variables. First, by (4.12),  $\mathcal{T}(t) = e^{tQ(\mathcal{D} + \mathcal{L}^{(0)})Q}$  would be the time-evolution operator for the traceless part of the atomic density operator if there were no recoil and no coupling to the conserved trace. We assume that  $\mathcal{T}(t)$  is a “shrinking” superoperator that reduces every traceless operator to zero on a time scale  $\tau$  as  $t \rightarrow \infty$ ;  $\tau$  might be the spontaneous decay rate for a two-state atom, or the time scale of optical pumping for a multistate atom. Hence  $\mathcal{T}(-t)$  is an “expanding” superoperator that tends to increase its argument exponentially. Finally, the

superoperator  $\mathcal{R}$  is bracketed by  $\mathcal{T}(-t)$  and  $\mathcal{T}(t)$ , and has an expression analogous to the exponential  $\exp[-(t-t')/\tau]$  inside the integral. We therefore assume that  $\mathcal{R}$  is a “bounded” operator that does not expand its argument without a bound as  $t \rightarrow \infty$ .

The right-hand side of (4.15a) is proportional to  $v_r$ , and at least with zero initial conditions so will be the solution to (4.15a). But then the assumedly bounded operator  $\mathcal{R} \propto v_r$  on the left would give a correction to  $\tilde{U}$  proportional to  $v_r^2$ , inconsistent with the accuracy of our approximations. We thus omit the superoperator  $\mathcal{R}$  on the left-hand side of (4.15a), and integrate the resulting equation to give a solution that presumably grows exponentially,

$$\tilde{U}(t) = \mathcal{T}(-t) \int_{-\infty}^t dt' \mathcal{T}(t-t') \left[ \left[ Q\mathcal{L}^{(1)}\mathcal{P}\mathcal{M}(t') + Q\mathcal{L}^{(1)}Q \int_{-\infty}^{t'} dt'' \mathcal{T}(t'-t'') Q\mathcal{L}^{(0)}\mathcal{P}\mathcal{M}(t'') \right] \right. \\ \left. - \int_{-\infty}^{t'} dt'' \mathcal{T}(t'-t'') Q\mathcal{L}^{(0)}\mathcal{P}\mathcal{M}(t''-t') \mathcal{P}\mathcal{L}^{(1)}Q \int_{-\infty}^{t'} dt''' \mathcal{T}(t'-t''') Q\mathcal{L}^{(0)}\mathcal{P}\mathcal{M}(t''') \right] \tilde{F}(t'). \quad (4.16)$$

We aim at a solution over times much longer than the longest time scale of internal evolution  $\tau$ . The lower limits of the integrals have been moved from 0 to  $-\infty$  because for  $t \gg \tau$  the difference is immaterial by virtue of the shrinking character of the superoperator  $\mathcal{T}(t)$ . Also, we have assumed that the homogeneous solution carrying the initial conditions has become insignificant compared to the terms retained in (4.16). Finally, as one more consequence of the shrinking character of the internal-evolution superoperator, the dominant contribution to the integral comes from the vicinity of the upper limit of integration. Since the time variation of  $\tilde{F}$  is proportional to  $v_r$ , consistent with the order of accuracy of (4.16), we replace  $\tilde{F}(t')$  by  $\tilde{F}(t)$  in all subsequent calculations.

But the expression (4.16) is formally first order in  $v_r$ . By inserting  $\tilde{U}$  from (4.16) into (4.13b) we obtain an expression for  $Q\tilde{\rho}$  in which the growth of  $\tilde{U}(t)$  is curbed by  $\mathcal{T}(t)$ , and which is correct up to first order in  $v_r$ . By inserting this  $Q\tilde{\rho}$  in its turn into (4.11), we obtain a closed equation for the quantity  $\mathcal{P}\tilde{\rho}$  that is correct to the order  $v_r^2$ ,

$$\left[ \frac{\partial}{\partial t} - \mathcal{D} \right] \mathcal{P}\tilde{\rho} = \mathcal{P}\mathcal{L}^{(1)}Q \int_{-\infty}^t dt' \mathcal{T}(t-t') Q\mathcal{L}^{(0)}\mathcal{P} \\ \times \mathcal{M}(t'-t) \mathcal{P}\tilde{\rho} + \dots \quad (4.17)$$

We introduce the c.m. distribution for the atoms irrespective of the internal state,

$$f \equiv \text{Tr}(\tilde{\rho}) = \text{Tr}(\mathcal{P}\tilde{\rho}), \quad (4.18a)$$

and conversely write

$$\mathcal{P}\tilde{\rho} = \frac{f}{N} \tilde{1}. \quad (4.18b)$$

By taking the trace of (4.17) and using Eqs. (4.18), the equation for  $\mathcal{P}\tilde{\rho}$  may finally be rewritten as the following closed equation of motion for the c.m. distribution function  $f = f(\mathbf{r}, \mathbf{p}, t)$ :

$$\begin{aligned}
\left[ \frac{\partial}{\partial t} - \mathcal{D} \right] f &= \frac{1}{N} \text{Tr} \left[ \mathcal{L}^{(1)} \mathcal{Q} \int_{-\infty}^t dt' \mathcal{T}(t-t') \mathcal{Q} \mathcal{L}^{(0)} \mathcal{P} \mathcal{M}(t'-t) \bar{\mathbb{I}} \right] f \\
&+ \frac{1}{N} \text{Tr} \left[ \mathcal{L}^{(1)} \mathcal{Q} \int_{-\infty}^t dt' \mathcal{T}(t-t') \left[ \mathcal{Q} \mathcal{L}^{(1)} \mathcal{P} + \mathcal{Q} \mathcal{L}^{(1)} \mathcal{Q} \int_{-\infty}^{t'} dt'' \mathcal{T}(t'-t'') \mathcal{Q} \mathcal{L}^{(0)} \mathcal{P} \mathcal{M}(t''-t') \right. \right. \\
&\quad \left. \left. - \int_{-\infty}^{t'} dt'' \mathcal{T}(t'-t'') \mathcal{Q} \mathcal{L}^{(0)} \mathcal{P} \mathcal{M}(t''-t') \mathcal{P} \mathcal{L}^{(1)} \mathcal{Q} \right. \right. \\
&\quad \left. \left. \times \int_{-\infty}^{t'} dt'' \mathcal{T}(t'-t'') \mathcal{Q} \mathcal{L}^{(0)} \mathcal{P} \mathcal{M}(t''-t') \right] \mathcal{M}(t'-t) \bar{\mathbb{I}} \right] f \\
&+ \frac{1}{N} \text{Tr} \left[ \mathcal{L}_{\text{sp}}^{(2)} \left[ \bar{\mathbb{I}} + \int_{-\infty}^t dt' \mathcal{T}(t-t') \mathcal{Q} \mathcal{L}^{(0)} \mathcal{P} \mathcal{M}(t'-t) \bar{\mathbb{I}} \right] \right] f . \tag{4.19}
\end{aligned}$$

As a notational point it should be noticed that, while all matrix structure of (4.19) is confined within the traces, position and velocity derivatives in  $\mathcal{D}$ ,  $\mathcal{L}^{(1)}$ , and  $\mathcal{L}_{\text{sp}}^{(2)}$  reach outside and act on  $f(\mathbf{r}, \mathbf{p}, t)$ , too. The first trace on the right-hand side is proportional to  $v_r$ , while the other two are proportional to  $v_r^2$ . The last trace is obviously associated with the angular distribution of spontaneous emission, while we for the time being leave the interpretation of the other traces open.

### B. Connection to internal time evolution

In order to unravel the operator formalism in a manner that facilitates a concrete scheme for numerical computations, we first look at the operator

$$e^{\tau \mathcal{D}} = \exp \left[ -\tau \frac{\mathbf{p}}{M} \cdot \frac{\partial}{\partial \mathbf{r}} \right]. \tag{4.20}$$

By Taylor's expansion, the action of  $e^{\tau \mathcal{D}}$  on an arbitrary position- and momentum-dependent argument function or operator is to shift the position by  $-\tau \mathbf{p}/M$ :

$$\begin{aligned}
e^{\tau \mathcal{D}} f(\mathbf{r}, \mathbf{p}) &= f(\mathbf{r} - \tau \mathbf{p}/M, \mathbf{p}) \\
&\equiv f_{\tau}(\mathbf{r}, \mathbf{p}) \equiv f(\mathbf{r}, \mathbf{p})|_{\tau} \equiv f_{\tau}. \tag{4.21}
\end{aligned}$$

This equation also serves to define the “ $\tau$  shift” operation denoted by the subscript  $\tau$  in the function  $f$ .

As long as the matrix  $\tilde{M}(\mathbf{r}, \mathbf{p})$  is proportional to the unit matrix so that  $\mathcal{P} \tilde{M} = \tilde{M}$ , the only effect of the opera-

tor  $e^{\tau \mathcal{D} \mathcal{P}}$  on it is the same shift of the position argument. Similarly, the only effect of  $e^{\tau \mathcal{Q} \mathcal{D} \mathcal{Q}}$  on a traceless matrix is the shift of position.

We next focus on the operator  $e^{\tau \mathcal{Q}(\mathcal{D} + \mathcal{L}^{(0)}) \mathcal{Q}}$ . Usually the Liouville operator depends on position, so that the operators  $\mathcal{L}^{(0)}$  and  $\mathcal{D}$  in the exponent do not commute. We want to find a Baker-Campbell-Hausdorff formula of the type

$$e^{(t-t') \mathcal{Q}(\mathcal{D} + \mathcal{L}^{(0)}) \mathcal{Q}} = \mathcal{O}(t, t') e^{(t-t') \mathcal{Q} \mathcal{D} \mathcal{Q}}. \tag{4.22}$$

By taking the derivative of (4.22) with respect to  $t$  one can see that the new operator  $\mathcal{O}$  satisfies the equations

$$\begin{aligned}
\frac{\partial}{\partial t} \mathcal{O}(t, t') &= \mathcal{O}(t, t') e^{(t-t') \mathcal{Q} \mathcal{D} \mathcal{Q}} \mathcal{Q} \mathcal{L}^{(0)} \mathcal{Q} e^{-(t-t') \mathcal{Q} \mathcal{D} \mathcal{Q}}, \\
\mathcal{O}(t', t') &= 1. \tag{4.23}
\end{aligned}$$

But the Liouville operator in the middle is a function of position and, in some approximations, maybe a function of momentum too. We arrive at the equation

$$\begin{aligned}
\frac{\partial}{\partial t} \mathcal{O}(t, t') &= \frac{\partial}{\partial t} \mathcal{O}(\mathbf{r}, \mathbf{p}; t, t') \\
&= \mathcal{O}(\mathbf{r}, \mathbf{p}; t, t') \mathcal{Q} \mathcal{L}^{(0)}(\mathbf{r} - (t-t') \mathbf{p}/M, \mathbf{p}) \mathcal{Q} \\
&\equiv \mathcal{O}(t, t') \mathcal{Q} \mathcal{L}_{t-t'}^{(0)} \mathcal{Q}, \tag{4.24}
\end{aligned}$$

whose formal solution may be found by iteration,

$$\begin{aligned}
\mathcal{O}(t, t') &= 1 + \int_{t'}^t dt_1 \mathcal{Q} \mathcal{L}_{t_1-t'}^{(0)} \mathcal{Q} + \int_{t'}^t dt_1 \int_{t'}^{t_1} dt_2 \mathcal{Q} \mathcal{L}_{t_2-t'}^{(0)} \mathcal{Q} \mathcal{L}_{t_1-t'}^{(0)} \mathcal{Q} + \dots \\
&= \dots + \int_{t'}^t dt_1 \int_{t'}^t dt_2 \theta(t_1 - t_2) \mathcal{Q} \mathcal{L}_{t_2-t'}^{(0)} \mathcal{Q} \mathcal{L}_{t_1-t'}^{(0)} \mathcal{Q} + \dots \\
&= \dots + \int_{t'}^t dt_1 \int_{t'}^t dt_2 \theta(t_2 - t_1) \mathcal{Q} \mathcal{L}_{t_1-t_2}^{(0)} \mathcal{Q} \mathcal{L}_{t_1-t_1}^{(0)} \mathcal{Q} + \dots \\
&= 1 + \int_{t'}^t dt_1 \mathcal{Q} \mathcal{L}_{t-t_1}^{(0)} \mathcal{Q} + \int_{t'}^t dt_1 \int_{t'}^{t_1} dt_2 \mathcal{Q} \mathcal{L}_{t-t_1}^{(0)} \mathcal{Q} \mathcal{L}_{t_1-t_2}^{(0)} \mathcal{Q} + \dots . \tag{4.25}
\end{aligned}$$

In the second step we have introduced the Heaviside step function  $\theta$ ; in the third step we have changed the integration variables from  $t_i$  to  $t + t' - t_i$ ; and finally we have reversed the numbering of integration variables.

On the other hand, let us fix the coordinates  $(\mathbf{r}, \mathbf{p})$  and

time  $t$ , and consider the differential equation

$$\frac{\partial}{\partial \bar{t}} \tilde{U}^{(0)}(\bar{\mathbf{r}}; t) = \mathcal{Q} \mathcal{L}_{t-\bar{t}}^{(0)} \mathcal{Q} U^{(0)}(\bar{\mathbf{r}}; t) + \mathcal{Q} \mathcal{L}_{t-\bar{t}}^{(0)} \mathcal{P} \tilde{\mathbb{I}}. \tag{4.26}$$

The Green's function for the initial time  $t'$  of this equa-

tion satisfies

$$\frac{\partial}{\partial \bar{t}} \mathcal{G}(\bar{t}, t'; t) = Q \mathcal{L}_{\bar{t}-\bar{t}}^{(0)} Q \mathcal{G}(\bar{t}, t'; t); \mathcal{G}(t', t'; t) = 1, \quad (4.27)$$

with the solution

$$\begin{aligned} \mathcal{G}(\bar{t}, t'; t) = & 1 + \int_{t'}^{\bar{t}} dt_1 Q \mathcal{L}_{\bar{t}-t_1}^{(0)} Q \\ & + \int_{t'}^{\bar{t}} dt_1 \int_{t'}^{t_1} dt_2 Q \mathcal{L}_{\bar{t}-t_1}^{(0)} Q \mathcal{L}_{t_1-t_2}^{(0)} Q + \dots \end{aligned} \quad (4.28)$$

In terms of the Green's function, the general solution of (4.26) reads

$$\begin{aligned} \tilde{U}^{(0)}(\bar{t}; t) = & \mathcal{G}(\bar{t}, t_0; t) \tilde{U}^{(0)}(t_0; t) \\ & + \int_{t_0}^{\bar{t}} dt' \mathcal{G}(\bar{t}, t'; t) Q \mathcal{L}_{\bar{t}-t'}^{(0)} \mathcal{P} \tilde{I}. \end{aligned} \quad (4.29)$$

where the initial condition has been specified at time  $t_0$ .

We are interested in the case when the initial condition is given in the distant past, and in the special time for the solution  $\bar{t} = t$ . As  $e^{\tau Q(\mathcal{D} + \mathcal{L}^{(0)})Q}$  is to be a shrinking superoperator, we surmise the same of the closely related Green's function. In the limit  $t_0 \rightarrow -\infty$  any traceless initial value therefore vanishes from the result, and we obtain

$$\tilde{U}^{(0)}(t; t) = \int_{-\infty}^t dt' \mathcal{G}(t, t'; t) Q \mathcal{L}_{t-t'}^{(0)} \mathcal{P} \tilde{I}. \quad (4.30)$$

To gain more insight into the meaning of Eq. (4.30), we note that for fixed  $\mathbf{r}$ ,  $\mathbf{p}$ , and  $t$  the operator  $\mathcal{L}_{t-\bar{t}}^{(0)}$  is the generator of time evolution for the internal state of an atom whose trajectory has taken it to  $\mathbf{r}(\bar{t}) = \mathbf{r} - (t - \bar{t})\mathbf{p}/M$  at time  $\bar{t}$ . Denoting the density operator for the internal state of such an atom by  $\tilde{\rho}$ , we have the time-evolution equation

$$\frac{\partial}{\partial \bar{t}} \tilde{\rho}(\bar{t}) = \mathcal{L}_{\bar{t}-\bar{t}}^{(0)} \tilde{\rho}(\bar{t}). \quad (4.31)$$

We project out separately the total population and the traceless part of the density matrix, and find the equations

$$\begin{aligned} \frac{\partial}{\partial \bar{t}} \mathcal{P} \tilde{\rho} &= 0, \\ \frac{\partial}{\partial \bar{t}} Q \tilde{\rho} &= Q \mathcal{L}_{\bar{t}-\bar{t}}^{(0)} Q Q \tilde{\rho} + Q \mathcal{L}_{\bar{t}-\bar{t}}^{(0)} \mathcal{P} \mathcal{P} \tilde{\rho} \\ &= Q \mathcal{L}_{\bar{t}-\bar{t}}^{(0)} Q Q \tilde{\rho} + \frac{\text{Tr}(Q \tilde{\rho})}{N} Q \mathcal{L}_{\bar{t}-\bar{t}}^{(0)} \tilde{I}. \end{aligned} \quad (4.32)$$

Except for the overall factor  $N^{-1} \text{Tr}(Q \tilde{\rho})$ , the solutions of (4.26) and (4.32) coincide. Hence, the operator  $\tilde{U}^{(0)}$  in Eq. (4.30) is essentially the traceless part of the density operator for the internal state of an atom that has started out in the distant past and whose straight-line trajectory has taken it to the phase-space point  $(\mathbf{r}, \mathbf{p})$  at time  $t$ .

For the special case  $\bar{t} = t$  a comparison of (4.25) and (4.28) shows that  $\mathcal{G}(t, t'; t) = \mathcal{O}(t, t')$ . The solution of Eq. (4.26) for the special choice  $\bar{t} = t$  with any traceless initial conditions in the distant past may therefore be written

$$\tilde{U}^{(0)}(t; t) = \int_{-\infty}^t dt' \mathcal{O}(t, t') Q \mathcal{L}_{t-t'}^{(0)} \mathcal{P} \tilde{I}. \quad (4.33)$$

The significance of this result may be appreciated after noting that (4.22) enables us to write the operator inside the first order (in  $v_r$ ) contribution in (4.19) as

$$\begin{aligned} \int_{-\infty}^t dt' \mathcal{T}(t-t') Q \mathcal{L}^{(0)} \mathcal{P} \mathcal{M}(t'-t) \tilde{I} \\ = \int_{-\infty}^t dt' \mathcal{O}(t, t') Q \mathcal{L}_{t-t'}^{(0)} \mathcal{P} \tilde{I} = \tilde{U}^{(0)}(t; t). \end{aligned} \quad (4.34)$$

The exceedingly abstract object on the left can be evaluated simply by solving the tangible differential equation (4.26), and the result is basically just the traceless part of the internal density operator of the atom whose straight-line trajectory has brought the atom to the phase-space point  $(\mathbf{r}, \mathbf{p})$  at time  $t$ . The plan of our upcoming development is to express similarly *all* the complicated operator functions in (4.19) in terms of solutions to differential equations.

### C. Force in the Fokker-Planck equation

To make further progress we need the explicit form of the first-order Liouville operator from (3.13). When  $\mathcal{L}^{(1)}$  acts on an arbitrary atomic operator  $\tilde{\sigma}(\mathbf{r}, \mathbf{p})$ , the result is

$$\mathcal{L}^{(1)} \tilde{\sigma} = \frac{1}{2} \sum_i \left[ \frac{\partial \tilde{V}}{\partial r_i} \frac{\partial \tilde{\sigma}}{\partial p_i} + \frac{\partial \tilde{\sigma}}{\partial p_i} \frac{\partial \tilde{V}}{\partial r_i} \right]. \quad (4.35)$$

Here  $\tilde{V}$  is the interaction Hamiltonian between the atom and the light field, granted that the position of the atom is a parameter and not a dynamical variable anymore.

Using Eqs. (4.34) and (4.35), the term linear in  $v_r$  on the right of (4.19) may be written

$$\begin{aligned} \frac{1}{N} \text{Tr} \left[ \mathcal{L}^{(1)} Q \int_{-\infty}^t dt' \mathcal{T}(t-t') Q \mathcal{L}^{(0)} \mathcal{P} \mathcal{M}(t'-t) \tilde{I} \right] f \\ = \frac{1}{N} \sum_i \text{Tr} \left[ \frac{\partial \tilde{V}}{\partial r_i} \frac{\partial}{\partial p_i} [\tilde{U}^{(0)}(t; t) f] \right] \\ = - \sum_i \frac{\partial}{\partial p_i} (F_i f), \end{aligned} \quad (4.36)$$

where we have defined the functions

$$F_i(\mathbf{r}, \mathbf{p}, t) = - \frac{1}{N} \text{Tr} \left[ \frac{\partial \tilde{V}}{\partial r_i} \tilde{U}^{(0)}(t; t) \right]. \quad (4.37)$$

When taken up to first order in recoil effects only, Eq. (4.19) reads

$$\left[ \frac{\partial}{\partial t} + \frac{\mathbf{p}}{M} \cdot \frac{\partial}{\partial \mathbf{r}} + \sum_i \frac{\partial}{\partial p_i} F_i \right] f = 0. \quad (4.38)$$

This is exactly the Liouville equation of the phase-space distribution of a particle subject to the force  $\mathbf{F}$ . The quantities  $F_i(\mathbf{r}, \mathbf{p})$  may be interpreted as the three Cartesian components of the light-pressure force acting on an atom at the phase-space point  $(\mathbf{r}, \mathbf{p})$ .

In view of the discussions after Eqs. (4.32) and (4.4), the quantity

$$\tilde{\rho}_a = \frac{1}{N} [\tilde{U}^{(0)}(t;t) + \tilde{I}] \quad (4.39)$$

is exactly the internal density matrix of an atom whose free flight has taken it to the phase-space position  $(\mathbf{r}, \mathbf{p})$  at time  $t$ . Since the interaction operator  $\tilde{V}$  is traceless, we may write

$$F_i = -\text{Tr} \left[ \tilde{\rho}_a \frac{\partial \tilde{V}}{\partial r_i} \right]. \quad (4.40)$$

This is the obvious quantum-mechanical analog of the net force of the electromagnetic field on a dipole [17]. While Eq. (4.40) may not have been derived at the present level of generality before, for an arbitrary number of atomic states and driving fields, the two-state form of this equation is one of the starting points of a well-known version of light-pressure theory [12].

#### D. Diffusion in the Fokker-Planck equation

Now that light pressure has been successfully extracted from the unwieldy looking Eqs. (4.19), we may obviously implement a similar strategy for higher-order recoil terms.

Let us consider as an example part of (4.19), a term called  $T_1$ ,

$$\begin{aligned} T_1 &= \frac{1}{N} \text{Tr} \left[ \mathcal{L}^{(1)} Q \int_{-\infty}^t dt' \mathcal{T}(t-t') \sum_i \frac{\partial \tilde{V}}{\partial r_i} \frac{\partial}{\partial p_i} f(\mathbf{r} - (t'-t)\mathbf{p}/M, \mathbf{p}, t) \right] \\ &= \frac{1}{N} \sum_i \text{Tr} \left[ \mathcal{L}^{(1)} Q \int_{-\infty}^t dt' \mathcal{T}(t-t') \frac{\partial \tilde{V}}{\partial r_i} \left[ \frac{\partial f}{\partial p_i} \Big|_{t'-t} + \frac{t-t'}{M} \frac{\partial f}{\partial r_i} \Big|_{t'-t} \right] \right], \end{aligned} \quad (4.44)$$

where the subscript in, say,  $\partial f / \partial p_i$  denotes the partial derivative of the function  $f(\mathbf{r}, \mathbf{p}, t)$  with respect to  $p_i$  at the appropriately shifted position argument. Equation (4.22) yields

$$T = \frac{1}{N} \sum_i \text{Tr} \left[ \mathcal{L}^{(1)} Q \int_{-\infty}^t dt' \mathcal{O}(t, t') \frac{\partial \tilde{V}}{\partial r_i} \Big|_{t-t'} \left[ \frac{\partial f}{\partial p_i} + \frac{t-t'}{M} \frac{\partial f}{\partial r_i} \right] \right]. \quad (4.45)$$

In (4.45) the partial derivatives of  $f$  are taken at the original arguments  $(\mathbf{r}, \mathbf{p})$ , and  $t$ , so that they are multiplicative constant as far as the rest of the integrand is concerned. Second, exactly like (4.33) gives a solution to (4.26), we find that

$$\tilde{U}_i^{(1,1)}(t;t) = \int_{-\infty}^t dt' \mathcal{O}(t, t') \frac{\partial \tilde{V}}{\partial r_i} \Big|_{t-t'} \quad (4.46a)$$

and

$$\tilde{U}_i^{(1,2)}(t;t) = \int_{-\infty}^t dt' \mathcal{O}(t, t') \frac{\partial \tilde{V}}{\partial r_i} \Big|_{t-t'} \frac{t-t'}{M} \quad (4.46b)$$

are the solutions at time  $\bar{t} = t$  to the respective equations

$$\frac{\partial}{\partial \bar{t}} \tilde{U}^{(1,1)}(\bar{t}; t) = Q \mathcal{L}_{t-\bar{t}}^{(0)} Q \tilde{U}^{(1,1)}(\bar{t}; t) + \frac{\partial \tilde{V}}{\partial r_i} \Big|_{t-\bar{t}} \quad (4.47a)$$

$$T_1 = \frac{1}{N} \text{Tr} \left[ \mathcal{L}^{(1)} Q \int_{-\infty}^t dt' \mathcal{T}(t-t') Q \mathcal{L}^{(1)} \mathcal{P} \mathcal{M}(t'-t) \tilde{I} \right] f. \quad (4.41)$$

We open the manipulations by applying the last shift operator  $\mathcal{M}$  on the c.m. distribution  $f$ . Thus,

$$\begin{aligned} T_1 &= \frac{1}{N} \text{Tr} \left[ \mathcal{L}^{(1)} Q \int_{-\infty}^t dt' \mathcal{T}(t-t') Q \mathcal{L}^{(1)} \tilde{P} \tilde{I} \right. \\ &\quad \left. \times f(\mathbf{r} - (t'-t)\mathbf{p}/M, \mathbf{p}, t) \right]. \end{aligned} \quad (4.42)$$

Using the definition of the projector  $\mathcal{P}$ , (4.1), and Eq. (4.35) for the operator  $\mathcal{L}^{(1)}$ , we find

$$\begin{aligned} \mathcal{L}^{(1)} \mathcal{P} \tilde{\sigma} &= \frac{1}{2} \sum_i \left[ \frac{\partial \tilde{V}}{\partial r_i} \frac{\tilde{I}}{N} \frac{\partial \text{Tr}(\tilde{\sigma})}{\partial p_i} + \frac{\partial \text{Tr}(\tilde{\sigma})}{\partial p_i} \frac{\tilde{I}}{N} \frac{\partial \tilde{V}}{\partial r_i} \right] \\ &= \frac{1}{N} \sum_i \frac{\partial \tilde{V}}{\partial r_i} \frac{\partial \text{Tr}(\tilde{\sigma})}{\partial p_i}. \end{aligned} \quad (4.43)$$

This operator is traceless since  $\tilde{V}$  is, and further action of  $Q$  on it is an identity. Application of these observations to (4.42) gives

and

$$\frac{\partial}{\partial \bar{t}} \tilde{U}^{(1,2)}(\bar{t}; t) = Q \mathcal{L}_{t-\bar{t}}^{(0)} Q \tilde{U}^{(1,2)}(\bar{t}; t) + \frac{\partial \tilde{V}}{\partial r_i} \Big|_{t-\bar{t}} \frac{t-\bar{t}}{M}. \quad (4.47b)$$

Analogously to the discussion after Eq. (4.32), these are solutions for the internal state of a moving atom, albeit with some quite counterintuitive driving terms and traceless initial conditions specified in the distant past. In the final step we use (4.35), and find that

$$\begin{aligned} T_1 &= \frac{1}{N} \sum_{i,j} \frac{\partial}{\partial p_i} \left\{ \text{Tr} \left[ \tilde{U}_j^{(1,1)}(t;t) \frac{\partial \tilde{V}}{\partial r_i} \right] \frac{\partial}{\partial p_j} \right. \\ &\quad \left. + \text{Tr} \left[ \tilde{U}_j^{(1,2)}(t;t) \frac{\partial \tilde{V}}{\partial r_i} \right] \frac{\partial}{\partial r_j} \right\} f. \end{aligned} \quad (4.48)$$

To reduce the number of terms we will have to deal with, we at this point introduce two approximations.

(i) As before, we denote by  $\tau$  the internal evolution scale of the atom, and estimate the derivatives as

$$\frac{\partial}{\partial r_i} f \sim \frac{f}{\Delta r}, \quad \frac{\partial}{\partial p_i} f \sim \frac{f}{\Delta p}. \quad (4.49)$$

Equations (4.47) show that, to the order of magnitude, the ratio of  $\tilde{U}^{(1,2)}$  and  $\tilde{U}^{(1,1)}$  is  $\tau/M$ , and hence the ratio of the  $\partial/\partial r$  and  $\partial/\partial p$  terms in (4.48) is

$$r_1 \sim \frac{\Delta p}{\Delta r} \frac{\tau}{M} \quad (4.50)$$

We assume that  $r_1$  is much smaller than unity, and ignore the position derivatives of  $f$  in (4.48).

(ii) We develop the remaining terms in (4.48) further as

$$T_1 = \frac{1}{N} \sum_{i,j} \frac{\partial^2}{\partial p_i \partial p_j} \left[ \text{Tr} \left[ \tilde{U}_j^{(1,1)}(t;t) \frac{\partial \tilde{V}}{\partial r_i} \right] f \right] - \frac{1}{N} \sum_{i,j} \frac{\partial}{\partial p_i} \left[ f \frac{\partial}{\partial p_j} \text{Tr} \left[ \tilde{U}_j^{(1,1)}(t;t) \frac{\partial \tilde{V}}{\partial r_i} \right] \right]. \quad (4.51)$$

The second term is of the same form, a force in the FPE, as (4.36). Qualitatively, the ratio of the force in (4.51) to the force in (4.36) is

$$r_2 \sim \frac{1}{\tilde{U}^{(0)}} \frac{\partial \tilde{U}^{(1,1)}}{\partial p_i}. \quad (4.52)$$

But dimensionally  $\tilde{U}^{(0)} \sim 1$ , whereas by (4.47a)  $\tilde{U}^{(1,1)}$  is obtained by integrating an expression of the form  $\partial \tilde{V} / \partial r_i \sim F$ , the force, over the time  $\tau$ . We write

$$\tilde{U}^{(1,1)} \sim F \tau \phi, \quad (4.53)$$

where  $\phi$  is a dimensionless function. The rate of change of  $\tilde{U}^{(1,1)}$  with momentum comes from the Doppler shift  $qp/M$ , and the natural scale with which the Doppler shift should be compared is  $\tau^{-1}$ . Thus  $\phi = \phi(qp\tau/M)$ , and the estimate (4.52) becomes

$$r_2 \sim \frac{F \tau^2 q}{M}. \quad (4.54)$$

We assume that  $r_2 \ll 1$ , and ignore the second term in (4.51).

With the two approximations we rewrite the term  $T_1$ , (4.48), as

$$T_1 = \frac{1}{N} \sum_{i,j} \frac{\partial^2}{\partial p_i \partial p_j} \left[ \text{Tr} \left[ \tilde{U}_j^{(1,1)}(t;t) \frac{\partial \tilde{V}}{\partial r_i} \right] f \right]. \quad (4.55)$$

As far as  $T_1$  is concerned, we are dealing with momentum diffusion.

Unfortunately, the preceding example does not exhaust the list of complications met in an attempt to develop various terms in (4.19) to a more tractable form. This is illustrated by another contribution

$$T_2 = \frac{1}{N} \text{Tr} \left[ \mathcal{L}^{(1)} \int_{-\infty}^t dt' \mathcal{T}(t-t') Q \mathcal{L}^{(1)} Q \int_{-\infty}^{t'} dt'' \mathcal{T}(t'-t'') Q \mathcal{L}^{(0)} \mathcal{P} \mathcal{M}(t''-t) \tilde{\Gamma} \right] f. \quad (4.56)$$

With minor rearrangements we find

$$\left[ \int_{-\infty}^t dt' \mathcal{T}(t-t') Q \mathcal{L}^{(1)} Q \int_{-\infty}^{t'} dt'' \mathcal{T}(t'-t'') Q \mathcal{L}^{(0)} \mathcal{P} \mathcal{M}(t''-t) \tilde{\Gamma} \right] f = \int_{-\infty}^t dt' \mathcal{T}(t-t') Q \mathcal{L}^{(1)} Q f_{t'-t} \int_{-\infty}^{t'} dt'' \mathcal{T}(t'-t'') Q \mathcal{L}^{(0)} \mathcal{P} \mathcal{M}(t''-t) \tilde{\Gamma}, \quad (4.57)$$

since the net translation of the position argument of  $f$  inside the second integral is independent of  $t''$  and since the unit operator is insensitive to all position translations. But by Eqs. (4.22), (4.23), and (4.30) the second integral on the right of (4.57) is just the object  $\tilde{U}^{(0)}$ , except that the "final time" is called  $t'$  instead of  $t$ . We write the second integral as  $\tilde{U}^{(0)}(\mathbf{r}, \mathbf{p}; t'; t')$ , and obtain

$$T_2 = \frac{1}{N} \text{Tr} \left[ \mathcal{L}^{(1)} Q \int_{-\infty}^t dt' \mathcal{T}(t-t') Q \mathcal{L}^{(1)} Q f(\mathbf{r} + (t-t')\mathbf{p}/M, \mathbf{p}, t') \tilde{U}^{(0)}(\mathbf{r}, \mathbf{p}; t'; t') \right] = \frac{1}{2N} \sum_i \text{Tr} \left[ \mathcal{L}^{(1)} Q \int_{-\infty}^t dt' \mathcal{O}(t, t') Q \left\{ \frac{\partial \tilde{V}}{\partial r_i} \Big|_{t-t'}, \tilde{U}_{t-t'}^{(0)} \left[ \frac{t-t'}{M} \frac{\partial f}{\partial r_i} + \frac{\partial f}{\partial p_i} \right] + f \frac{\partial \tilde{U}^{(0)}}{\partial p_i} \Big|_{t-t'} \right\} \right]. \quad (4.58a)$$

In this expression the brace notation  $\{ \tilde{A}, \tilde{B} \}_+$  stands for the anticommutator, and we write

$$\tilde{U}_{t-t'}^{(0)} = \tilde{U}^{(0)}(\mathbf{r} - (t-t')\mathbf{p}/M, \mathbf{p}; t'; t'). \quad (4.58b)$$

Our analysis greatly depends on the realization that (4.58b) is the internal-state density operator for an atom that is at the position  $\mathbf{r} - (t-t')\mathbf{p}/M$  at time  $t'$ , i.e., that

will be at  $\mathbf{r}$  at time  $t$ . The operator in (4.58b) is just the running solution to (4.26) at time  $\bar{t} = t'$ . As usual, the operator

$$\tilde{U}_i^{(2,1)} = \frac{1}{2} \int_{-\infty}^t dt' \mathcal{O}(t, t') Q \left\{ \frac{\partial \tilde{V}}{\partial r_i} \Big|_{t-t'}, \tilde{U}_{t-t'}^{(0)} \right\}_+, \quad (4.59)$$

in turn, is the solution at time  $\bar{t}=t$  to

$$\frac{\partial}{\partial \bar{t}} \tilde{U}_i^{(2,1)}(\bar{t}; t) = Q \mathcal{L}_{t-\bar{t}}^{(0)} Q \tilde{U}_i^{(2,1)}(\bar{t}; t) + \frac{1}{2} Q \left\{ \frac{\partial \tilde{V}}{\partial r_i} \Big|_{t-\bar{t}} \tilde{U}^{(0)}(\bar{t}; t) \right\}_+, \quad (4.60)$$

with traceless initial conditions in the distant past.

Suppose the solution to (4.60) is given, along with the same two conditions  $r_1 \ll 1$ ,  $r_2 \ll 1$  as in the case of  $T_1$ . We carry out steps analogous to the steps from (4.48) to (4.55), and note that an estimate similar to the one following Eq. (4.52) allows us to discard also the  $\partial \tilde{U}^{(0)}/\partial p_i$  term in (4.58a). We finally write  $T_2$  as

$$T_2 = \frac{1}{N} \sum_{i,j} \frac{\partial^2}{\partial p_i \partial p_j} \left[ \text{Tr} \left[ \tilde{U}_i^{(2,1)}(t; t) \frac{\partial \tilde{V}}{\partial r_j} \right] f \right]. \quad (4.61)$$

We are ready to combine the results into a Fokker-Planck equation.

### E. Final Fokker-Planck equation

After investigating each term on the right-hand side of (4.19) in a manner outlined in the previous sections, we find the Fokker-Planck equation

$$\left[ \frac{\partial}{\partial t} + \frac{\mathbf{p}}{M} \cdot \frac{\partial}{\partial \mathbf{r}} \right] f = - \sum_i \frac{\partial}{\partial p_i} (F_i f) + \sum_{i,j} \frac{\partial^2}{\partial p_i \partial p_j} (D_{ij} f) \quad (4.62)$$

for the c.m. WF  $f(\mathbf{r}, \mathbf{p}, t)$  of an atom moving in the given light field.

In order to derive the components of the force and the diffusion tensor at time  $t$  in the given phase-space location  $(\mathbf{r}, \mathbf{p})$ , one imagines an atom coming from time  $-\infty$  along a straight-line path with momentum  $\mathbf{p}$  in such a way that the path takes the atom to the desired position  $\mathbf{r}$  at time  $t$ . One sets up the Liouville operator for the internal state of the atom,  $\mathcal{L}^{(0)}$ . As the field driving the atom depends on position, in a coordinate system moving with the atom  $\mathcal{L}^{(0)}$  also depends on time. One integrates the following equations of motion for the internal state of the moving atom until time  $t$ :

$$\dot{\tilde{\rho}} = \mathcal{L}^{(0)} \left[ \tilde{\rho} + \frac{\tilde{\mathbf{1}}}{N} \right], \quad \tilde{\rho}(-\infty) = 0, \quad (4.63a)$$

$$\begin{aligned} \dot{\tilde{\xi}}_i &= \mathcal{L}^{(0)} \tilde{\xi}_i + \frac{1}{N} \frac{\partial \tilde{V}}{\partial r_i} + \frac{1}{2} Q \left[ \frac{\partial \tilde{V}}{\partial r_i} \tilde{\rho} + \tilde{\rho} \frac{\partial \tilde{V}}{\partial r_i} \right] \\ &\quad - \tilde{\rho} \text{Tr} \left[ \frac{\partial \tilde{V}}{\partial r_i} \tilde{\rho} \right], \quad \tilde{\xi}_i(-\infty) = 0. \end{aligned} \quad (4.63b)$$

Here the interaction potential  $\tilde{V}$  is evaluated in the frame moving with the atom, too, so  $\tilde{V}$  appears to depend on time just like  $\mathcal{L}^{(0)}$  does. The force and the diffusion at time  $t$  are then given by

$$F_i = - \text{Tr} \left[ \frac{\partial \tilde{V}}{\partial r_i} \tilde{\rho} \right], \quad (4.64a)$$

$$D_{ij} = \frac{1}{2} \text{Tr} \left[ \frac{\partial \tilde{V}}{\partial r_i} \tilde{\xi}_j + \frac{\partial \tilde{V}}{\partial r_j} \tilde{\xi}_i \right] + \text{Tr} \left[ \mathcal{S}^{ij} \left[ \frac{\tilde{\mathbf{1}}}{N} + \tilde{\rho} \right] \right]. \quad (4.64b)$$

The required spontaneous-emission superoperator can be read out from (2.41),

$$\begin{aligned} \mathcal{S}_{j_1 m_1, j_1' m_1'; j_2 m_2, j_2' m_2'}^{ij} \\ = \frac{1}{2} \sum_{\sigma, \sigma'} \xi_{\sigma \sigma'}^{ij} (\hbar q_{j_2 j_1})^2 \text{sgn}(D_{j_2 j_1} D_{j_2' j_1'}) (\Gamma_{j_2 j_1} \Gamma_{j_2' j_1'})^{1/2} \\ \times \langle m_1 \sigma | m_2 \rangle_{j_1 j_2} \langle m_2' | m_1' \sigma \rangle_{j_2' j_1'}. \end{aligned} \quad (4.65)$$

While writing our final equations, we have somewhat simplified the notation. For instance, the operator we now call  $\tilde{\rho}$  is defined in terms of the operators of the preceding sections as  $\tilde{U}^{(0)}/N$ . Second, by utilizing linearity of the ensuing expressions, we have combined (4.47a), (4.60), and yet another undisplayed equation of the same type into the single equation (4.63b). Third, the observation that the time evolution governed by  $\mathcal{L}^{(0)}$  preserves the trace enables us to do away with all but one projection in Eqs. (4.63) and (4.64). Finally, we have symmetrized the diffusion tensor by hand. The difference between the symmetrized and unsymmetrized expressions is of the same order as, say, the discarded second term in Eq. (4.51), and hence the asymmetry (if any) was not significant to begin with.

### F. Discussion of the Fokker-Planck equation

The FPE (4.62) and the recipe to compute the force and the diffusion embodied in Eqs. (4.63) and (4.64) constitute the main result of the present paper. We therefore dwell on the assumptions and implications of the FPE at some length.

*Underlying approximations of the FPE.* Several approximations have been made during the derivation of the FPE. The fundamental ones are the following three.

(i) We have expanded finite-difference WF's to power series in photon momentum  $\hbar q$ . This can be warranted only if the momentum scale of the c.m. WF,  $\Delta p$ , satisfies  $\Delta p \gg \hbar q$ .

(ii) We have thrown out the initial conditions of the internal state of the atom. The FPE is only valid for times much longer than the longest time scale of the internal evolution,  $\tau$ . For a two-state atom  $\tau$  is comparable to the excited-state lifetime, whereas in multistate models a longer optical-pumping time may apply.

(iii) In the calculations of force and diffusion we have approximated the past trajectory of the atom by free flight. Such an approach can only make sense if the actual trajectory varies little over the memory time  $\tau$ . The rule-of-thumb rate of change of atomic motion in light-pressure theory is the recoil frequency  $\omega_r = M v_r^2 / 2 \hbar$  [17], and the FPE is expected to be applicable if  $\tau \omega_r \ll 1$ . In particular, for a two-state atom the linewidth  $\gamma$  of the optical transition has to satisfy the well-known condition  $\gamma \gg \omega_r$ .

There are also two unproven suppositions concerning the expansion characteristics of certain operators.

(iv) We have assumed that  $e^{tQ(\mathcal{D}+\mathcal{L}^{(0)})Q}$  contracts “every” traceless argument operator to zero on a time scale  $\tau$ . For a two-state system it can be proven explicitly that  $e^{tQ(\mathcal{D}+\mathcal{L}^{(0)})Q}$  is a shrinking superoperator [15], but we have not been able to construct a proof in the general case. Nonetheless, our assumption appears quite plausible. For instance, for a spatially homogeneous ensemble of zero-velocity atoms the condition that  $e^{tQ(\mathcal{D}+\mathcal{L}^{(0)})Q}$  be a shrinking operator can easily be shown to be equivalent to the statement that the density operator of the internal state of each atom, the object of ordinary recoilless spectroscopy, reaches a unique steady state. The unique steady state is equivalent to the state of affairs that the atom forgets its long-past states, so that the shrinking character of  $e^{tQ(\mathcal{D}+\mathcal{L}^{(0)})Q}$  is evidently required to make the fundamental assumption (iii) in the first place.

(v) We have assumed that the operator  $\mathcal{R}$  defined in (4.15b) is bounded. This assumption promptly implies that in the limit  $v_r \rightarrow 0$  a fair approximation to the internal state of a moving atom is obtained from the assumption that the velocity of the atom has been constant all along; recoil effects and the associated past changes of the velocity only give small corrections, instead of completely dominating the internal state [15]. As a counterexample consider the possibility that  $\mathcal{R}$  grows without a bound and eventually dominates on the left-hand side of (4.15a). The outcome would under quite plausible assumptions be a singular long-time limit of  $\tilde{U}$  in which the recoil has canceled out, but which is nonetheless different from the operator  $\tilde{U}$  in the absence of recoil. No such singularity has been described in any form of optical spectroscopy.

In addition, we have made four approximations that probably could have been avoided.

(vi) We have assumed that a magnetic field, if any, is in the direction of the quantization axis. A magnetic field can cause cooling below the Doppler limit [39], and a magnetic field is also employed to assist trapping of atoms [40]. The requirement that the field points in a fixed direction might be overly restrictive in some cases of practical importance. We believe that our final results are independent of such an assumption: As long as the proper magnetic field evolution is used in the Liouvillean  $\mathcal{L}^{(0)}$ , our procedure to obtain the force and the diffusion should remain valid for an arbitrary position-dependent magnetic field.

(vii) We have assumed that, with the aid of a RWA, the interaction matrix  $\tilde{V}$  has been rendered independent of time. Otherwise the Liouvillean  $\mathcal{L}^{(0)}$  depends explicitly on time, a time-ordered exponential must be used in lieu of  $e^{tQ(\mathcal{D}+\mathcal{L}^{(0)})Q}$ , the notation becomes cumbersome, and the crucial relation between Eqs. (4.25) and (4.28) gets marred. Nonetheless, in Eqs. (4.63) and (4.64) it does not make any conceptual or practical difference whether  $\tilde{V}$  depends explicitly on time or not. We conjecture that (4.63) and (4.64) remain valid for an explicitly time dependent interaction potential  $\tilde{V}$ .

(viii) We have ignored cross diffusion between position and momentum in the FPE. Our somewhat heuristic condition for this is that the quantity  $r_1$  defined in Eq.

(4.50) has to be much less than unity. Presumably the worst case is deep modulation of the atom density as a function of position, whereupon  $\Delta r \sim \lambda$ . The condition  $r_1 \ll 1$  then requires that  $\tau \Delta p \ll \lambda$ , i.e., that a characteristic cooled atom must traverse only a small fraction of the wavelength  $\lambda$  during the internal-evolution time scale  $\tau$ . For a two-state system it is easily shown that for reasonable experimental parameters such a condition is satisfied by virtue of the customary validity condition of the FPE  $\gamma \gg \omega_r$ , whereas for a multistate atom we end up requiring that the velocity of a typical atom is smaller than the “velocity capture range” of Dalibard and Cohen-Tannoudji [21].

However, one should notice that if there is no substantial position modulation in the atomic distribution function,  $\partial/\partial r$  and hence the cross diffusion are negligible. Moreover, if the total  $6 \times 6$  diffusion tensor is positive definite, the crossed  $r$ - $p$  diffusion will rather smear both the position and the velocity distributions, and thereby further iron away any  $r$  dependence. Indefinite diffusion matrices sometimes occur in quantum problems [41] and could have dramatic consequences in the present problem, but positive-definite diffusion is the default. In such a case one might in principle judge the effect of the cross diffusion *a posteriori*, by checking the solution of the FPE obtained without cross diffusion for variation with  $r$ . If the variation is weak, a plausible assumption is that the cross diffusion will not make it stronger. The cross diffusion is then altogether negligible.

(ix) We have ignored corrections  $\propto v_r^2$  in the force. A qualitative validity condition for such an approximation is that the quantity  $r_2$  defined in (4.54) is much less than unity. Now, the approximation of the force  $F \sim M v_r \gamma$  converts the condition  $r_2 \ll 1$  to  $\sqrt{\omega_r \gamma \tau} \ll 1$ . In a two-state atom the latter is satisfied by virtue of the conditions  $\gamma \gg \omega_r$  and  $\tau^{-1} \sim \gamma$ , but in a multistate atom one should exercise more care.

A useful way of reading the condition  $r_2 \ll 1$  is that during the time interval  $\tau$  light-pressure forces should be able to move an initially stationary atom only a distance much less than one wavelength. On the other hand, if the contrary is true, the main condition (iii) is obviously violated. We surmise that the force corrections  $\propto v_r^2$  are negligible whenever the FPE is good, no matter whether we are dealing with a two-state or a multistate atom.

Incidentally, when the limit  $v_r \rightarrow 0$  is taken by keeping all other atomic and field parameters fixed, the steady-state temperature of laser-cooled two-state atoms becomes independent of  $v_r$ . For all we know, the same also applies to multistate atoms. Inclusion of contributions proportional  $v_r^2$  to the force would lead to a correction in the temperature proportional to  $v_r$ , but so would presumably also “diffusion” terms proportional to powers of  $v_r$  higher than two that have been discarded in our derivations. Related arguments have been given earlier [17] to the effect that it is inconsistent with the accuracy of the FPE to retain the  $v_r^2$  terms in the force.

At any rate, it is obvious that, keeping everything else fixed, the FPE is expected to become increasingly accurate as the size of the recoil kick of a photon on the atom

gets smaller. A convenient (theoretical) trick to force the limit of small recoil without touching other fundamental parameters of the atom-field system is to let the mass of the atom go to infinity.

*Interpretation of the FPE.* The FPE describes the motion of an ensemble of particles subject to a deterministic force  $\mathbf{F}$ , with an additional random component in the motion characterized by the diffusion tensor  $D_{ij}$ . This is a completely classical picture; quantum fluctuations in the matter-field interaction are accounted for by classical stochasticity in the motion of the atom. The reduction of the fully quantized coupled time evolution of the internal and c.m. degrees of freedom to a classical stochastic process for the c.m. motion is possible in the limit of small recoil velocity. In the contrary case the FPE is not valid, and we are faced with the full quantum problem [42–44].

The force in the FPE is a straightforward descendant of the classical dipole force [12,17], as discussed in Sec. IV C. By the same token, the diffusion proportional to  $\mathcal{S}^{ij}$  arises from the randomness of recoil kicks on the atom as a result of the randomness of the directions of spontaneous photons. The remaining terms in the diffusion reflect time statistics of absorption and emission processes. The connection between induced diffusion and photon statistics has been studied in detail for two-state systems [45,46], and a similar close relationship should exist for any multistate atom. However, to display it, we would first have to work out the photon statistics for an atom with an arbitrary level structure. This task we deem to be outside of the scope of the present paper.

## V. CONCLUDING REMARKS

In this paper we have derived the Fokker-Planck equation for the center-of-mass Wigner function, for an atom with an arbitrary level structure in an arbitrary light field. The force and the diffusion tensor may be obtained by solving a set of equations closely related to the density-matrix equations of recoilless spectroscopy. At this point the problem becomes one of writing down the ordinary density-matrix equations, i.e., the Liouville operator  $\mathcal{L}^{(0)}$ . We have described algorithms capable of building the free-evolution contributions, induced, and spontaneous terms in  $\mathcal{L}^{(0)}$ .

The natural next step is to implement algorithms to construct  $\mathcal{L}^{(0)}$  and methods to solve Eqs. (4.62)–(4.64) on a computer. We have written an extensive collection of C programs for this purpose, and compared the computation results with known analytical theories to validate both our theory and its numerical implementation. However, a wealth of new physical and computational issues arises in this context. We will have to defer the discussion of our numerical methods and results to forthcoming publications.

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## APPENDIX A: EXAMPLES OF SPONTANEOUS EMISSION

A computer algorithm for generating spontaneous relaxation terms does not require any external knowledge of the form of the results, or physical interpretation for them. However, in order to illustrate some features of spontaneous emission that must be understood in order to successfully program the algorithms, we discuss in this appendix a few special cases of spontaneous relaxation terms. We ignore photon recoil, a particularly apt approximation as the Liouville operator  $\mathcal{L}^{(0)}$  required to find the force and the diffusion for the FPE does not contain the recoil. We thus omit  $\mathbf{r}$  and  $\mathbf{p}$  labels of the c.m. motion in Eqs. (2.27)–(2.30), and remove the angular integral with the kernel  $K$  using (2.25). Spontaneous emission in the presence of level degeneracy has been the subject of much study both with [13,21] and without [23–25,31,47–52] photon recoil; our examples can be found in some form in the existing literature.

*Two levels with angular-momentum degeneracy.* We begin with the case of just two levels having the customary degeneracy with respect to angular momentum. Using (2.26) we find the relaxation terms

$$\begin{aligned} \left. \frac{d}{dt} \right|_{\text{sp}} \rho(m_1; m'_1) &= 2\gamma \sum_{\sigma, m_2, m'_2} \langle m_1 \sigma | m_2 \rangle \rho(m_2; m'_2) \langle m'_2 | m'_1 \sigma \rangle, \end{aligned} \quad (\text{A1a})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(m_1; m_2) = -\gamma \rho(m_1; m_2), \quad (\text{A1b})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(m_2; m_1) = -\gamma \rho(m_2; m_1), \quad (\text{A1c})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(m_2; m'_2) = -2\gamma \rho(m_2; m'_2). \quad (\text{A1d})$$

The relaxation terms for the density-matrix elements that refer to at least one excited state describe simple exponential decay determined by the Einstein  $A$  coefficient  $2\gamma$ . The double-ground-state relaxations are more interesting: “Zeeman coherences” between excited states are at least in part transmitted to the ground state (if the latter is degenerate). The transfer of coherence has a peculiar “parallel drop” property: An excited-state coherence between the states  $m_2$  and  $m'_2$  can be converted to ground-state coherences between states of the form  $m_2 - \sigma$  and  $m'_2 - \sigma$  only (with the same  $\sigma = -1, 0, +1$ ).

*Quantum-beat coherence.* Next consider a V-type transition from the state  $j_1=0, m_1=0$  to the excited states  $j_2=1, m_2=0$ , and  $j'_2=1, m'_2=0$ . Assuming for simplicity that both transitions are governed by the same Einstein  $A$  coefficient, choosing the phases of the state vectors so that the signum function in (2.27) equals unity, and using an obvious notation for the states, we find the full complement of relaxation terms in the form

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j_2; j_2) = -\gamma [2\rho(j_2; j_2) + \rho(j_2; j'_2) + \rho(j'_2; j_2)] , \quad (\text{A2a})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j_2; j'_2) = -\gamma [2\rho(j_2; j'_2) + \rho(j_2; j_2) + \rho(j'_2; j'_2)] , \quad (\text{A2b})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j'_2; j_2) = -\gamma [2\rho(j'_2; j_2) + \rho(j_2; j_2) + \rho(j'_2; j'_2)] , \quad (\text{A2c})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j'_2; j'_2) = -\gamma [2\rho(j'_2; j'_2) + \rho(j'_2; j_2) + \rho(j_2; j'_2)] , \quad (\text{A2d})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j_1; j_2) = -\gamma [\rho(j_1; j_2) + \rho(j_1; j'_2)] , \quad (\text{A2e})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j_1; j'_2) = -\gamma [\rho(j_1; j'_2) + \rho(j_1; j_2)] , \quad (\text{A2f})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j_1; j_1) = 2\gamma [\rho(j_2; j_2) + \rho(j_2; j'_2) + \rho(j'_2; j_2) + \rho(j'_2; j'_2)] . \quad (\text{A2g})$$

The spontaneous relaxations of the coherences 1-2 and 1-2' are coupled, and so are the relaxations of the populations 2-2, 2'-2', and the coherences 2-2'.

*Spontaneously generated coherence.* Finally, let us consider two nearly degenerate lower states in a  $\Lambda$  configuration. For notational simplicity we consider transitions between the lower states  $j_1=0, m_1=0, j'_1=0, m'_1=0$ , and the excited state  $j_2=1, m_2=0$ . With a proper choice of the phases of the state vectors, the relaxation terms become

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j_2; j_2) = -4\gamma(j_2; j_2) , \quad (\text{A3a})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j_2; j_1) = -2\gamma\rho(j_2; j_1) , \quad (\text{A3b})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j_2; j'_1) = -2\gamma\rho(j_2; j'_1) ,$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j_1; j_2) = -2\gamma\rho(j_1; j_2) , \quad (\text{A3c})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j'_1; j_2) = -2\gamma\rho(j'_1; j_2) ,$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j_1; j_1) = 2\gamma\rho(j_2; j_2) , \quad (\text{A3d})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j'_1; j_1) = 2\gamma\rho(j_2; j_2) , \quad (\text{A3e})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j_1; j'_1) = 2\gamma\rho(j_2; j_2) , \quad (\text{A3f})$$

$$\left. \frac{d}{dt} \right|_{\text{sp}} \rho(j'_1; j'_1) = 2\gamma\rho(j_2; j_2) . \quad (\text{A3g})$$

A population in one excited state may give rise to a coherent superposition of several receiving states.

#### APPENDIX B: SCALAR VERSUS MATRIX WIGNER FUNCTIONS

The WF equations of motion for the atom, (3.13), are a straightforward generalization of the WF equation of motion for a scalar particle (no internal degrees of freedom), in that the WF  $\bar{\rho}$  and the interaction potential  $\bar{V}$  are just treated as noncommuting numbers.

In order to expose this relation, we consider a particle that has no internal degrees of freedom and whose time evolution is governed by the Hamiltonian

$$\hat{H} = \frac{\hat{\mathbf{p}}^2}{2M} + V(\hat{\mathbf{r}}) . \quad (\text{B1})$$

This time we start from the position representation. The equation of motion for  $\rho(\mathbf{r}_1, \mathbf{r}_2) = \langle \mathbf{r}_1 | \hat{\rho} | \mathbf{r}_2 \rangle$  reads

$$\begin{aligned} \frac{\partial}{\partial t} \rho(\mathbf{r}_1, \mathbf{r}_2) &= -\frac{i}{\hbar} \left\langle \mathbf{r}_1 \left| \left[ \frac{\hat{\mathbf{p}}^2}{2M} + \hat{V}, \hat{\rho} \right] \right| \mathbf{r}_2 \right\rangle \\ &= \frac{i\hbar}{2M} \left[ \frac{\partial^2}{\partial \mathbf{r}_1^2} - \frac{\partial^2}{\partial \mathbf{r}_2^2} \right] \rho(\mathbf{r}_1, \mathbf{r}_2) \\ &\quad - \frac{i}{\hbar} [V(\mathbf{r}_1)\rho(\mathbf{r}_1, \mathbf{r}_2) - \rho(\mathbf{r}_1, \mathbf{r}_2)V(\mathbf{r}_2)] . \end{aligned} \quad (\text{B2})$$

The transformation from the position representation to the Wigner representation is carried out according to

$$\rho(\mathbf{r}, \mathbf{p}) = \frac{1}{(2\pi\hbar)^3} \int d^3r' \exp \left[ \frac{i}{\hbar} \mathbf{p} \cdot \mathbf{r}' \right] \rho \left[ \mathbf{r} - \frac{\mathbf{r}'}{2}, \mathbf{r} + \frac{\mathbf{r}'}{2} \right] . \quad (\text{B3})$$

The derivatives in (B2) complete the convective derivative, while the potential terms give

$$\begin{aligned}
& \int d^3r' \exp\left[\frac{i}{\hbar}\mathbf{p}\cdot\mathbf{r}'\right] \left[ V\left[\mathbf{r}-\frac{\mathbf{r}'}{2}\right] \rho\left[\mathbf{r}-\frac{\mathbf{r}'}{2}, \mathbf{r}+\frac{\mathbf{r}'}{2}\right] - \rho\left[\mathbf{r}-\frac{\mathbf{r}'}{2}, \mathbf{r}+\frac{\mathbf{r}'}{2}\right] V\left[\mathbf{r}+\frac{\mathbf{r}'}{2}\right] \right] \\
&= V\left[\mathbf{r}+\frac{i\hbar}{2}\frac{\partial}{\partial\mathbf{p}}\right] \int d^3r' \exp\left[\frac{i}{\hbar}\mathbf{p}\cdot\mathbf{r}'\right] \rho\left[\mathbf{r}-\frac{\mathbf{r}'}{2}, \mathbf{r}+\frac{\mathbf{r}'}{2}\right] - \dots \\
&= (2\pi\hbar)^3 \left[ V\left[\mathbf{r}+\frac{i\hbar}{2}\frac{\partial}{\partial\mathbf{p}}\right] \rho(\mathbf{r}, \mathbf{p}) - \rho(\mathbf{r}, \mathbf{p}) V\left[\mathbf{r}-\frac{i\hbar}{2}\frac{\partial}{\partial\mathbf{p}}\right] \right]. \quad (\text{B4})
\end{aligned}$$

We have formally pulled  $\mathbf{r}'$  out of the integrals by replacing it with  $-i\hbar(\partial/\partial\mathbf{p})$ , and the left arrow in the last form of (B4) indicates that the derivative exceptionally acts to the left.

We now combine (B2)–(B4) and expand the result into a power series of  $\hbar$ . We obtain

$$\begin{aligned}
\frac{d}{dt}\rho &= -\frac{i}{\hbar}(V\rho - \rho V) + \frac{1}{2} \sum_i \left[ \frac{\partial V}{\partial r_i} \frac{\partial \rho}{\partial p_i} + \frac{\partial \rho}{\partial p_i} \frac{\partial V}{\partial r_i} \right] + \frac{i\hbar}{8} \sum_{i,j} \left[ \frac{\partial^2 V}{\partial r_i \partial r_j} \frac{\partial^2 \rho}{\partial p_i \partial p_j} - \frac{\partial^2 \rho}{\partial p_i \partial p_j} \frac{\partial^2 V}{\partial r_i \partial r_j} \right] + O(\hbar^2) \\
&= \sum_i \frac{\partial V}{\partial r_i} \frac{\partial \rho}{\partial p_i} + O(\hbar^2). \quad (\text{B5})
\end{aligned}$$

This is the conventional equation of motion of the WF, taken up to order  $\hbar^2$  [34–37]. As we have made use of the fact that  $V$  and  $\rho$  are commuting scalars only at the last step of (B5), Eq. (3.13) readily emerges as the generalization of the ordinary WF equation of motion to the case

when the density matrix and the potential are not numbers but noncommuting objects instead. The free-evolution term in (3.13) describes the evolution of  $\bar{\rho}$  resulting from internal-state energies of the atom, independently of any external potential.

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