Atom-field interactions: Density-matrix equations including quantization of the center-of-mass motion

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Master equations are derived for the time evolution of density-matrix elements characterizing atoms interacting with classical radiation fields that drive transitions between electronic-state manifolds of levels. The atomic center-of-mass motion is quantized and recoil accompanying absorption, stimulated emission, and spontaneous emission is included. An adiabatic elimination of the optical coherences is used to obtain quantum rate equations for ground- and excited-state density-matrix elements. Subsequent adiabatic elimination of excited-state density-matrix elements results in rate equations for the slow evolution of ground-state density-matrix elements. It is shown that recoil during spontaneous emission mixes spatial and internal atomic degrees of freedom.

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I. INTRODUCTION

Rate equations were derived recently [1] to characterize the interaction of atoms with classical radiation fields which drive transitions between two electronic-state manifolds. The density-matrix equations derived in [1] include all complications arising from fine structure, hyperfine structure, and magnetic-state degeneracy. They can be applied to a wide range of spectroscopic problems. Solutions of the density-matrix equations enable one to calculate expectation values of physical observables that can be compared directly with experimental data for specific atomic transitions. The equations were used to calculate the transient response of atoms to a set of radiation pulses in both the weak [2] and strong [3] field limits. The equations derived in [1] were written using an irreducible tensor representation for density-matrix elements. In this representation the transformation properties of the equations under rotation are easily established. These transformation properties allow one to identify tensors associated with the polarization vectors of the incident fields.

The purpose of this article is to generalize the results of [1] to include quantization of the atomic center-of-mass motion. One can point out a number of problems where such quantum equations can be applied. Among them are different types of recoil-induced resonances (RIR) [4–9] , scattering of atoms from standing wave fields (so-called resonant Kapitza-Dirac effect) [10–14], wave-matter interference [15–19], atomic Talbot effect [19–22], grating stimulated echo (GSE) [23], and magnetic grating echo (MGE) [2]. For the most part, these phenomena have been observed using alkali-metal atoms, where hyperfine splitting and magnetic-level degeneracy can play an important role. The recoil an atom undergoes on the emission or absorption of radiation [24] lies at the heart of these effects (recoil is not critical to the GSE and MGE) and requires consideration of atomic center-of-mass motion for multilevel atoms.

One distinguishes two kind of effects relevant to atomic center-of-mass motion quantization: recoil during stimulated interaction with fields (STR) [24] and recoil during spontaneous emission (SPR) [25]. An inclusion of the STR in master equations for multilevel atoms can be carried out in the same manner as that for ''two-level'' atoms [24]. Spontaneous emission leads to both ''in'' terms and ''out'' terms in the density-matrix equations. The out term characterizes excited-state decay with a rate Γ that is independent of the magnetic quantum number and total angular momentum of the hyperfine sublevels. The in term, representing repopulation of ground-state levels resulting from spontaneous emission, is more complex [1,26,27]. The in terms depend on the total angular momenta of the initial and final hyperfine sublevels involved in the given radiative transition. In the absence of recoil, however, one finds that, after averaging over final states of the field, the in term is diagonal when the atomic density matrix is expanded in a spherical tensor basis $\{K,Q\}$. On average, spontaneous emission acts as a scalar.

When recoil is included, there is a correlation between the spontaneously emitted radiation and the atomic center-of-mass motion. As a consequence, it is no longer a simple matter to average over final states of the field. One finds that, with the inclusion of recoil, spontaneous emission mixes states having different $\{K,Q\}$ [28]. The resulting in term was obtained previously [28,29] for transitions between a ground and an excited state, each having a given total angular momentum. Even though the extension to manifolds of ground and excited states is straightforward, we present the derivation for the purpose of completeness. Moreover, a derivation of the in term has not been published previously, to our knowledge. Having obtained the master equations, we adiabatically eliminate certain density-matrix elements, to obtain rate equations for ground- and excited-state density-matrix elements.

This paper is arranged as follows. In Sec. II the in-term derivation is given. In Secs. III and IV rate equations and ground-state density-matrix equations are obtained, respectively. A discussion of the results is given in Sec. V.

II. IN TERM

Consider radiative decay of an excited atom to its ground state. If one neglects antiresonant terms and introduces an interaction representation for the field, he arrives at an atom-quantized field Hamiltonian

$$\mathscr{H}=(\epsilon_G+\epsilon_{\mathbf{p}})|G,m_g,\mathbf{p}\rangle\langle G,m_g,\mathbf{p}|+(\epsilon_H+\epsilon_{\mathbf{p}})|H,m_h,\mathbf{p}\rangle \times\langle H,m_h,\mathbf{p}|-\{i\hbar f_{\mathbf{k}}[\mathbf{e}(\lambda,\mathbf{k})\cdot\mathbf{d}]a(\lambda,\mathbf{k}) \times\exp(i\mathbf{k}\cdot\mathbf{r}-i\omega_{\mathbf{k}}t)+\text{H.c.}\},\tag{2.1}$$

where G,m_g and H,m_h specify the total angular momenta and magnetic quantum numbers of the ground and excited states, respectively, $\mathbf{p}$ is the atomic center-of-mass momentum, ϵ_F is the internal atomic energy of state $|F\rangle$ ($F=G$ or H), $\epsilon_{\mathbf{p}}=\mathbf{p}^2/2m$ is the kinetic energy of state $|\mathbf{p}\rangle$, m is the atomic mass, $a(\lambda,\mathbf{k})$ and $\mathbf{e}(\lambda,\mathbf{k})$ are the annihilation operator and unit polarization vectors for a radiation mode having wave vector $\mathbf{k}$, polarization λ ($\lambda=1$ or 2), and frequency $\omega_{\mathbf{k}}$, $\mathbf{d}$ is a dipole moment operator, $f_{\mathbf{k}}=[2\pi\omega_{\mathbf{k}}/\hbar\mathscr{V}]^{1/2}$, and $\mathscr{V}$ is a quantization volume. A summation convention is implicit in Eq. (2.1) that will be used in all subsequent equations, in which repeated indices and symbols appearing in the right-hand-side (rhs) of an equation are to be summed over, unless they also appear on the left-hand side (lhs) of the equation.

In an interaction picture, the amplitude for an atom to decay to the ground state via radiation in a given mode $(\lambda,\mathbf{k})$ evolves as

$$\dot{b}(\mathbf{k},\lambda;G,m_g,\mathbf{p},t)=f_{\mathbf{k}}^{*}\exp[i(\omega_{\mathbf{k}}-\omega_{HG}-\omega_{\mathbf{p}+\hbar\mathbf{k},\mathbf{p}})t] \times[\mathbf{e}^{*}(\lambda,\mathbf{k})\cdot\langle G,m_g|\mathbf{d}|H,m_h\rangle] \times b(H,m_h,\mathbf{p}+\hbar\mathbf{k},t),\tag{2.2}$$

where $\omega_{HG}=(\epsilon_H-\epsilon_G)/\hbar$ is the $H\rightarrow G$ transition frequency,

$$\omega_{\mathbf{pp}'}=(\epsilon_{\mathbf{p}}-\epsilon_{\mathbf{p}'})/\hbar$$

is a transition frequency between atomic center-of-mass momenta states, and $b(H,m_h,\mathbf{p},t)$ is an excited-state amplitude. In the Weisskopf-Wigner approximation, the in term

$$\bar{\rho}(G,m_g,\mathbf{p};G',m_g',\mathbf{p}';\text{in})\equiv\frac{d}{dt}[b(\mathbf{k},\lambda;G,m_g,\mathbf{p},t) \times b^{*}(\mathbf{k},\lambda;G',m_g',\mathbf{p}',t)]\tag{2.3}$$

is given by

$$\bar{\rho}(G,m_g,\mathbf{p};G',m_g',\mathbf{p}';\text{in})\equiv 2\pi\Phi(t)|f_{\mathbf{k}}|^2\delta(\omega_{\mathbf{k}}-\bar{\omega})e_q(\lambda,\mathbf{k})e_{q'}^{*}(\lambda,\mathbf{k})\langle G,m_g|d_{q'}|H,m_h\rangle \times\langle G',m_g'|d_q|H',m_h'\rangle^{*}\rho(H,m_h,\mathbf{p}+\hbar\mathbf{k};H',m_h',\mathbf{p}'+\hbar\mathbf{k}),\tag{2.4}$$

$$\Phi(t)=\exp(i\Delta\omega t),\tag{2.4a}$$

where $\Delta\omega=\omega_{H'G'}+\omega_{\mathbf{p}'+\hbar\mathbf{k},\mathbf{p}'}-\omega_{HG}-\omega_{\mathbf{p}+\hbar\mathbf{k},\mathbf{p}}$, $\bar{\omega}$ is some average value of transition frequencies $\omega_{HG}+\omega_{\mathbf{p}+\hbar\mathbf{k},\mathbf{p}}$ and $\omega_{H'G'}+\omega_{\mathbf{p}'+\hbar\mathbf{k},\mathbf{p}'}$, and e_q and d_q are spherical components of vectors $\mathbf{e}$ and $\mathbf{d}$, respectively, defined by

$$e_{\pm1}(\lambda,\mathbf{k})=\mp[e_x(\lambda,\mathbf{k})\pm ie_y(\lambda,\mathbf{k})]/\sqrt{2},$$

$$e_0(\lambda,\mathbf{k})=e_z(\lambda,\mathbf{k}),$$

$$d_{\pm1}=\mp(d_x\pm id_y)/\sqrt{2},\quad d_0=d_z.\tag{2.5}$$

In the "normal" representation, $\rho(F,m_f,\mathbf{p};F',m_f',\mathbf{p}')=\exp[-i(\omega_{FF'}+\omega_{\mathbf{pp}'})t]\bar{\rho}(F,m_f,\mathbf{p};F',m_f',\mathbf{p}')$, the phase factor Φ is absent in Eq. (2.4).

The sum over magnetic quantum numbers can be carried out using density-matrix elements in an irreducible tensor basis defined by

$$\rho_Q^K(F,F';\mathbf{p},\mathbf{p}')=(-1)^{F-m}[K]\begin{pmatrix}F & F' & K\\ m & -m' & -Q\end{pmatrix} \times\rho(F,m_f,\mathbf{p};F',m_f',\mathbf{p}'),\tag{2.6}$$

where ( ) is a $3-J$ symbol and

$$[X_1^{(n_1)}\cdots X_s^{(n_s)}]\equiv[(2X_1+1)^{(n_1)}\cdots(2X_s+1)^{(n_s)}]^{1/2}.\tag{2.7}$$

In this representation one finds

$$\rho_Q^K(G,G';\mathbf{p},\mathbf{p}';\text{in})=(-1)^{K+\bar{K}+Q'+1}\begin{pmatrix}K & K' & \bar{K}\\ Q & -Q' & \bar{Q}\end{pmatrix} \times[H,H',K,K',\bar{K}]\gamma(H,H';G,G')\frac{3}{8\pi}\begin{Bmatrix}K & K' & \bar{K}\\ G & H & 1\\ G' & H' & 1\end{Bmatrix}\int d\mathbf{n}\mathscr{E}_{\bar{Q}}^{\bar{K}}(\mathbf{n})\rho_{Q'}^{K'}(H,H';\mathbf{p}+\hbar\mathbf{k},\mathbf{p}'+\hbar\mathbf{k}),\tag{2.8}$$

where

$$\gamma(H,H';G,G')=4d_{GH}d^*_{G'H'}\bar{\omega}^3/3[H,H']\hbar c^3, \quad (2.9)$$

$\{\}$ is a $9-J$ symbol,

$$\mathbf{n}=\mathbf{k}/k,$$

$$\mathcal{E}^K_Q(\mathbf{n})=\boldsymbol{\epsilon}^K_Q(\mathbf{e}(\lambda,\mathbf{k}),\mathbf{e}(\lambda,\mathbf{k})), \quad (2.10)$$

$$\boldsymbol{\epsilon}^K_Q(\mathbf{A},\mathbf{B})=(-1)^{K+q}[K]\begin{pmatrix}1 & 1 & K\\ q & q' & Q\end{pmatrix}A_{-q}B^*_{q'} \quad (2.11)$$

and A_q and B_q are spherical components of the vectors $\mathbf{A}$ and $\mathbf{B}$. There is no summation over $\mathbf{k}$ in Eq. (2.10) (since $\mathbf{n}=\mathbf{k}/k$, the vector $\mathbf{k}$ appears implicitly in the lhs of this equation).

Using the relation

$$\sum_\lambda e_q(\lambda,\mathbf{k})e^*_{q'}(\lambda,\mathbf{k})=\delta_{qq'}-n_q n_{q'} \quad (2.12)$$

one can reexpress $\mathcal{E}^K_Q(\mathbf{n})$ as

$$\mathcal{E}^K_Q(\mathbf{n})=-\sqrt{3}\,\delta_{K0}\delta_{Q0}-\boldsymbol{\epsilon}^K_Q(\mathbf{n},\mathbf{n}). \quad (2.13)$$

For a real vector $\mathbf{n}$, having spherical components $n^*_q=(-1)^q n_{-q}$, one can show that

$$\mathcal{E}^K_Q(\mathbf{n})=(-1)^K\mathcal{E}^K_Q(\mathbf{n}), \quad (2.14)$$

which implies that only even multipoles ($K=0$ or 2) are nonvanishing. The function $\mathcal{E}^K_Q$ is an irreducible tensor of rank K. The only irreducible tensor of rank K, which is a function of $\mathbf{n}$, is the spherical harmonic $Y_{KQ}(\mathbf{n})$; consequently $\mathcal{E}^K_Q(\mathbf{n})$ has to be proportional to $Y_{KQ}(\mathbf{n})$. The coefficients of proportionality can be obtained if one compares these tensors for $\mathbf{n}=\mathbf{n}_0=(0,0,1)$, where $\mathcal{E}^K_Q(\mathbf{n})=-\sqrt{2/3}(\sqrt{2}\,\delta_{K0}+\delta_{K2})\delta_{Q0}$, $Y_{KQ}(\mathbf{n})=\sqrt{(2K+1)/4\pi}\,\delta_{Q0}$. As a result one arrives at an expression for the in term (2.8):

$$\rho^K_Q(G,G';\mathbf{p},\mathbf{p}';\text{in})=(-1)^{Q'}\begin{pmatrix}K & K' & \bar{K}\\ Q & -Q' & \bar{Q}\end{pmatrix}\times B_{\bar{K}}I(K,K',\bar{K};G,G';H,H')\times\frac{3}{8\pi}\int d\mathbf{n}Y_{\bar{K}\bar{Q}}(\mathbf{n})\times\rho^{K'}_{Q'}(H,H';\mathbf{p}+\hbar\mathbf{k},\mathbf{p}'+\hbar\mathbf{k}), \quad (2.15)$$

$$I(K,K',\bar{K};G,G';H,H')=(-1)^K[H,H',K,K',\bar{K}]\times\gamma(H,H';G,G')\times\begin{Bmatrix}K & K' & \bar{K}\\ G & H & 1\\ G' & H' & 1\end{Bmatrix}, \quad (2.15a)$$

$$B_{\bar{K}}=\sqrt{16\pi/3}\,\delta_{\bar{K}0}+\sqrt{8\pi/15}\,\delta_{\bar{K}2}. \quad (2.15b)$$

The integration over directions $\mathbf{n}$ can be carried out if one transforms to an "$\mathbf{s}-\mathbf{u}$" "coordinate" representation defined by

$$\rho^K_Q(F,F';\mathbf{s},\mathbf{u})=\int\frac{d\mathbf{p}d\mathbf{p}'}{(2\pi\hbar)^3}\exp\left\{\frac{i}{\hbar}\left[(\mathbf{p}-\mathbf{p}')\cdot\mathbf{s}+\frac{\mathbf{p}+\mathbf{p}'}{2}\cdot\mathbf{u}\right]\right\}\rho^K_Q(F,F';\mathbf{p},\mathbf{p}'). \quad (2.16)$$

Using Eqs. (2.15) and (2.16), and expanding the spherical harmonics in terms of plane waves [30], one finds

$$\rho^K_Q(G,G';\mathbf{s},\mathbf{u};\text{in})=(-1)^{Q'}\begin{pmatrix}K & K' & \bar{K}\\ Q & -Q' & \bar{Q}\end{pmatrix}\times\bar{B}_{\bar{K}}I(K,K',\bar{K};G,G';H,H')j_{\bar{K}}(ku)\times Y_{\bar{K}\bar{Q}}(\mathbf{n}_\mathbf{u})\rho^K_Q(H,H';\mathbf{s},\mathbf{u}), \quad (2.17)$$

$$\bar{B}_{\bar{K}}=\sqrt{12\pi}\,\delta_{\bar{K}0}-\sqrt{6\pi/5}\,\delta_{\bar{K}2}, \quad (2.17a)$$

where $j_s(x)$ is a spherical Bessel function of order s and $\mathbf{n}_\mathbf{u}=\mathbf{u}/u$.

When recoil during spontaneous emission is negligible [$\hbar k\ll\min(p,p')$ in Eq. (2.15) or $k\ll u^{-1}$ in Eq.(2.17)], only the term with $K=0$ contributes in Eqs. (2.15) or (2.17), and one finds

$$\rho^K_Q(G,G';\mathbf{p},\mathbf{p}';\text{in})=(-1)^{K+H+G'+1}[H,H']\times\begin{Bmatrix}H & H' & K\\ G' & G & 1\end{Bmatrix}\gamma(H,H';G,G')\times\rho^K_Q(H,H';\mathbf{p},\mathbf{p}') \quad (2.18)$$

($\{\ \}$ is a $6-J$ symbol), coinciding with the equation obtained for atoms moving along classical trajectories [1]. The in term is now diagonal with respect to K and Q—spontaneous emission acts as a scalar, on average.

Owing to recoil, the scalar nature of spontaneous emission is broken for given excited-state momenta $\mathbf{p},\mathbf{p}'$. As a result, the in term mixes different $\{K,Q\}$ multipoles. For transitions in which there is only one value of G and only one value of H ($H=H',G=G'$) the in terms (2.15) or (2.17) can be derived from that obtained in [28]. In this case, the $9-J$ symbol is invariant if multiplied by a factor $(-1)^{K+K'+\bar{K}}$. Since $\bar{K}$ is even, only transitions satisfying

$$K-K'=0 \text{ or } 2 \quad (2.19)$$

are allowed [28]. When coherences between sublevels having different angular momenta are included ($H\neq H'$ or $G\neq G'$), the selection rule (2.19) no longer holds and transitions between multipoles with opposite parity ($K-K'=\pm1$) are also possible.

The general expressions (2.15) or (2.17) for the in term must be used when one considers interaction with noncolinear radiation fields; however, when atoms interact with fields propagating along a given direction it is possible to reduce the equations to one dimension. Taking the quantization axis $\hat{z}$ along the field propagation direction and assuming that the atomic momentum distribution in the transverse directions is much wider than the recoil momentum, one can write

$$\rho_Q^K(F,F';\mathbf{p},\mathbf{p}')=[(2\pi\hbar)^2\mathscr{V}^{-2/3}]\delta(\mathbf{p}_\perp-\mathbf{p}'_\perp)\times W(\mathbf{p}_\perp)\rho_Q^K(F,F';p,p'), \quad (2.20)$$

where $\mathbf{p}_\perp=(p_x,p_y)$, $W(\mathbf{p}_\perp)$ is the transverse distribution function having width $\Delta p_\perp \gg \hbar k$, and $p=(\mathbf{p})_z$, $p'=(\mathbf{p}')_z$. In this limit, the transverse momentum distribution can be eliminated from all the equations. After the transverse distribution function is eliminated in Eq. (2.15), one can integrate over azimuthal angle ϕ to obtain

$$\rho_Q^K(G,G';p,p';\text{in})=(-1)^Q\begin{pmatrix} K & K' & \bar{K} \\ Q & -Q & 0\end{pmatrix}\times B_{\bar{K}}^a I(K,K',\bar{K};G,G';H,H')\times\int_{-1}^{1}dx P_{\bar{K}}(x)\times\rho_Q^{K'}(H,H';p+\hbar kx,p'+\hbar kx), \quad (2.21)$$

$$B_{\bar{K}}^a=\sqrt{3}/2\,\delta_{\bar{K}0}+\sqrt{3/8}\,\delta_{\bar{K}2}, \quad (2.21a)$$

where $P_\ell(x)$ is a Legendre polynomial of order ℓ. In the "$s-u$" representation the in term can be expressed as

$$\rho_Q^K(G,G';s,u;\text{in})=2(-1)^{Q+\bar{K}/2}\times\begin{pmatrix} K & K' & \bar{K} \\ Q & -Q & 0\end{pmatrix}\times B_{\bar{K}}^a I(K,K',\bar{K};G,G';H,H')j_{\bar{K}}(ku)\times\rho_Q^{K'}(H,H';s,u). \quad (2.22)$$

The one-dimensional in term in momentum space (2.21) has been used by Castin *et al.* [31] and Guo and Berman [8] for $G=0\rightarrow H=1$ and $G=1\rightarrow H=2$ transitions, respectively, and a term similar to Eq. (2.22) has been used by Tan and Walls [14] in a two-level model with an in term proportional to the Bessel function $j_2(ku)$.

The in term can be simplified also when fields propagate in a plane (two-dimensional case). For a $G=\frac{1}{2}\rightarrow H=\frac{3}{2}$ transition, the in term has been derived by Castin *et al.* [32] and used in their analysis of two-dimensional sub-Doppler cooling.

III. RATE EQUATIONS

Consider now interactions of atoms with a nearly resonant field having amplitude

$$\mathbf{E}(\mathbf{r},t)=\frac{1}{2}E^{(j)}\mathbf{e}^{(j)}\exp[i(\mathbf{k}_j\cdot\mathbf{r}-i\Omega_j t)+\text{c.c.} \quad (3.1)$$

The field consists of a set of N traveling waves having amplitude $E^{(j)}$, frequency Ω_j, propagation vector $\mathbf{k}_j$, and polarization vector $\mathbf{e}^{(j)}$ [$1\leq j\leq N$ in Eq. (3.1)]. In the rotating wave approximation, the Hamiltonian for the atom-field interaction $V=-\mathbf{d}\cdot\mathbf{E}(\mathbf{r},t)$ has matrix elements given by

$$\langle H,m_h,\mathbf{p}|V|G,m_g,\mathbf{p}'\rangle=\sqrt{3}(-1)^{H-m_g+1}\begin{pmatrix} H & 1 & G \\ -m_h & q & m_g\end{pmatrix}\times\hbar\chi_{HG}^{(j)}e_{-q}^{(j)}\exp[-i\Delta_{HG}^{(j)}t]\times\delta(\mathbf{p}-\mathbf{p}'-\hbar\mathbf{k}_j), \quad (3.2)$$

where $\chi_{HG}^{(j)}=d_{HG}E^{(j)}/2\sqrt{3}\hbar$ and $\Delta_{HG}^{(j)}=\Omega_j-\omega_{HG}$ are the Rabi frequency and detuning of field j. Matrix elements are written in an "interaction picture" with respect to the internal degrees of freedom only, defined by

$$\tilde{\rho}_Q^K(F,F';\mathbf{p},\mathbf{p}')=\exp(i\omega_{FF'}t)\rho_Q^K(F,F';\mathbf{p},\mathbf{p}'). \quad (3.3)$$

Using Schrödinger's equations, one finds that density-matrix elements evolve according to

$$\left(\frac{\partial}{\partial t}+i\omega_{\mathbf{pp}'}\right)\tilde{\rho}_Q^K(G,G';\mathbf{p},\mathbf{p}')=e^{i\omega_{GG'}t}\rho_Q^K(G,G';\mathbf{p},\mathbf{p}';in)+i[1,K,K'](-1)^{Q'}\begin{pmatrix} K & K' & 1 \\ Q & -Q' & q\end{pmatrix}\times\left((-1)^{G+G'+1}\chi_{HG'}^{(j)}e_q^{(j)}e^{-i\Delta_{HG'}^{(j)}t}\begin{Bmatrix} K & K' & 1 \\ H & G' & G\end{Bmatrix}\tilde{\rho}_{Q'}^{K'}(G,H;\mathbf{p},\mathbf{p}'+\hbar\mathbf{k}_j)-(-1)^{Q+2H+K+K'}[\chi_{HG}^{(j)}e_{-q}^{(j)}]^*e^{i\Delta_{HG}^{(j)}t}\begin{Bmatrix} K & K' & 1 \\ H & G & G'\end{Bmatrix}[\tilde{\rho}_{-Q'}^{K'}(G',H;\mathbf{p}',\mathbf{p}+\hbar\mathbf{k}_j)]^*\right), \quad (3.4)$$

$$\left(\frac{\partial}{\partial t}+i\omega_{\mathbf{pp}'}\right)\tilde{\rho}_Q^K(H,H';\mathbf{p},\mathbf{p}')=-\Gamma\tilde{\rho}_Q^K(H,H';\mathbf{p},\mathbf{p}')+i[1,K,K'](-1)^{Q'}\begin{pmatrix} K & K' & 1 \\ Q & -Q' & q\end{pmatrix}\times\left(-(-1)^{H+H'+K+K'}\chi_{HG}^{(j)}e_q^{(j)}e^{-i\Delta_{HG}^{(j)}t}\begin{Bmatrix} K & K' & 1 \\ G & H & H'\end{Bmatrix}\tilde{\rho}_{Q'}^{K'}(G,H';\mathbf{p}-\hbar\mathbf{k}_j,\mathbf{p}')+(-1)^{Q+2G+1}[\chi_{H'G}^{(j)}e_{-q}^{(j)}]^*e^{i\Delta_{H'G}^{(j)}t}\begin{Bmatrix} K & K' & 1 \\ G & H' & H\end{Bmatrix}[\tilde{\rho}_{-Q'}^{K'}(G,H;\mathbf{p}'-\hbar\mathbf{k}_j,\mathbf{p})]^*\right), \quad (3.4a)$$

$$\left(\frac{\partial}{\partial t}+i\omega_{\mathbf{pp}'}\right)\tilde{\rho}_Q^K(G,H;\mathbf{p},\mathbf{p}')=-\gamma\tilde{\rho}_Q^K(G,H;\mathbf{p},\mathbf{p}')-i[1,K,K'](-1)^{Q'}\begin{pmatrix}K & K' & 1\\ Q & -Q' & q\end{pmatrix}[e_{-q}^{(j)}]^* \times\Bigg((-1)^{q+G+G'}[\chi_{HG'}^{(j)}]^*e^{i\Delta_{HG'}^{(j)}t}\begin{Bmatrix}K & K' & 1\\ G' & H & G\end{Bmatrix}\tilde{\rho}_{Q'}^{K'}(G,G';\mathbf{p},\mathbf{p}'-\hbar\mathbf{k}_j) +(-1)^{Q+2G+K+K'}[\chi_{H'G}^{(j)}]^*e^{i\Delta_{H'G}^{(j)}t}\begin{Bmatrix}K & K' & 1\\ H' & G & H\end{Bmatrix}[\tilde{\rho}_{-Q'}^{K'}(H,H';\mathbf{p}',\mathbf{p}+\hbar\mathbf{k}_j)]^*\Bigg), \tag{3.4b}$$

where Γ is the (H-independent) excited-state decay rate and γ is the (G,H-independent) decay rate of the optical coherences. If state G (H) is a hyperfine sublevel of the ground- (excited-) state manifold, having electronic angular moment J (J') and radiative decay occurs only between the H and G manifolds (“closed” system), then

$$\gamma=\Gamma/2 \tag{3.5}$$

and $\Gamma=4|d_{JJ'}|^2\bar{\omega}^3/3(2J'+1)\hbar c^3$. When recoil is negligible the quantum transport Eqs. (3.4) can be reduced to those given in Refs. [26,27] (see also [1]) for atoms moving classically.

Under appropriate conditions, Eqs. (3.4) can be reduced to rate equations for ground- and excited-state density-matrix elements. When the ground- and excited-state density-matrix elements vary slowly with respect to the optical coherences (see discussion of the validity conditions in [1]), and γ and Γ are larger than the Doppler width and recoil frequency $\omega_k=\hbar k^2/2m$ [33],

$$\min\{\Gamma,\gamma\}\gg\max\{\omega_{\mathbf{pp}'},\mathbf{k}_j\cdot\mathbf{p}/\mathbf{m},\omega_k\}, \tag{3.6}$$

an approximate solution of Eq. (3.4b) is

$$\tilde{\rho}_{Q'}^{K'}(G,H;\mathbf{p},\mathbf{p}')=-i[1,K',K''](-1)^{Q''}\begin{pmatrix}K' & K'' & 1\\ Q' & -Q'' & q'\end{pmatrix}[e_{-q'}^{(j')}]^*\Bigg((-1)^{q'+G+G''}[\chi_{HG''}^{(j')}]^*e^{i\Delta_{HG''}^{(j')}t}(\gamma+i\Delta_{HG''}^{(j')})^{-1} \times\begin{Bmatrix}K' & K'' & 1\\ G'' & H & G\end{Bmatrix}\tilde{\rho}_{Q''}^{K''}(G,G'';\mathbf{p},\mathbf{p}'-\hbar\mathbf{k}_{j'})+(-1)^{Q'+2G+K'+K''}[\chi_{H''G}^{(j')}]^*e^{i\Delta_{H''G}^{(j')}t}(\gamma+i\Delta_{H''G}^{(j')})^{-1} \times\begin{Bmatrix}K' & K'' & 1\\ H'' & G & H\end{Bmatrix}[\tilde{\rho}_{-Q''}^{K''}(H,H'';\mathbf{p}',\mathbf{p}+\hbar\mathbf{k}_{j'})]^*\Bigg). \tag{3.7}$$

Substituting this expression in Eqs. (3.4),(3.4a) one finds bilinear forms with respect to the fields' polarization vectors. It is convenient [1] to expand them in a coupled tensor basis (2.11) as

$$e_q^{(j)}[e_{q'}^{(j')}]^*=(-1)^q[\bar{K}]\begin{pmatrix}1 & 1 & \bar{K}\\ q & -q' & -\bar{Q}\end{pmatrix}\epsilon_{\bar{Q}}^{\bar{K}}(j,j'), \tag{3.8}$$

where simplified notation, $\epsilon_{\bar{Q}}^{\bar{K}}(j,j')\equiv\epsilon_{\bar{Q}}^{\bar{K}}[\mathbf{e}^{(j)},\mathbf{e}^{(j')}]$, is used. The sums over q,q',Q' in Eqs. (3.4,3.4a) lead to $6-J$ symbols and the sum over K' can be carried out analytically. As a result, one finds rate equations for ground- and excited-state density matrixes which, in the normal representation, are given by

$$\left(\frac{\partial}{\partial t}+i\omega_{GG'}+i\omega_{\mathbf{pp}'}\right)\rho_Q^K(G,G';\mathbf{p},\mathbf{p}')=\rho_Q^K(G,G';\mathbf{p},\mathbf{p}';\text{in})+(-1)^{Q'}\begin{pmatrix}K & K' & \bar{K}\\ Q & -Q' & \bar{Q}\end{pmatrix}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')e^{-i\Omega_{jj'}t} \times[S_+(K,K',\bar{K};G,G',G'';j,j')\rho_{Q'}^{K'}(G,G'';\mathbf{p},\mathbf{p}'+\hbar\mathbf{k}_{jj'}) +S_-(K,K',\bar{K};G,G',G'';j,j')\rho_{Q'}^{K'}(G'',G';\mathbf{p}-\hbar\mathbf{k}_{jj'},\mathbf{p}') +S(K,K',\bar{K};G,G',H,H';j,j')\rho_{Q'}^{K'}(H,H';\mathbf{p}+\hbar\mathbf{k}_{j'},\mathbf{p}'+\hbar\mathbf{k}_j)], \tag{3.9}$$

$$\left(\frac{\partial}{\partial t}+\Gamma+i\omega_{HH'}\right)\rho_Q^K(H,H';\mathbf{p},\mathbf{p}')=(-1)^{Q'}\begin{pmatrix}K & K' & \bar{K}\\ Q & -Q' & \bar{Q}\end{pmatrix}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')e^{-i\Omega_{jj'}t}[S_+(K,K',\bar{K};H,H',H'';j,j')$$
$$\times\rho_{Q'}^{K'}(H,H'';\mathbf{p},\mathbf{p}'+\hbar\mathbf{k}_{jj'})+S_-(K,K',\bar{K};H,H',H'';j,j')\rho_{Q'}^{K'}(H'',H';\mathbf{p}-\hbar\mathbf{k}_{jj'},\mathbf{p}')$$
$$+S(K,K',\bar{K};H,H',G,G';j,j')\rho_{Q'}^{K'}(G,G';\mathbf{p}-\hbar\mathbf{k}_j,\mathbf{p}'-\hbar\mathbf{k}_{j'})], \tag{3.9a}$$

where

$$\mathbf{k}_{jj'}=\mathbf{k}_j-\mathbf{k}_{j'},\Omega_{jj'}=\Omega_j-\Omega_{j'},$$

$$S_+(K,K',\bar{K};G,G',G'';j,j')=-3(-1)^{H-G+\bar{K}}[K,K',\bar{K}]\chi_{HG'}^{(j)}[\chi_{HG''}^{(j')}]^*(\gamma+i\Delta_{HG''}^{(j')})^{-1}\begin{Bmatrix}K & K' & \bar{K}\\ G'' & G' & G\end{Bmatrix}\begin{Bmatrix}1 & 1 & \bar{K}\\ G' & G'' & H\end{Bmatrix}, \tag{3.10}$$

$$S_-(K,K',\bar{K};G,G',G'';j,j')=-3(-1)^{H-G+G''-G'+K+K'}[K,K',\bar{K}]\chi_{HG''}^{(j)}[\chi_{HG}^{(j')}]^*(\gamma-i\Delta_{HG''}^{(j)})^{-1}$$
$$\times\begin{Bmatrix}K & K' & \bar{K}\\ G'' & G & G'\end{Bmatrix}\begin{Bmatrix}1 & 1 & \bar{K}\\ G & G'' & H\end{Bmatrix}, \tag{3.10a}$$

$$S(K,K',\bar{K};G,G',H,H';j,j')=3(-1)^{G'-G+H'-H+K+1}[K,K',\bar{K}]\chi_{H'G'}^{(j)}[\chi_{HG}^{(j')}]^*[(\gamma+i\Delta_{HG}^{(j')})^{-1}+(\gamma-i\Delta_{H'G'}^{(j)})^{-1}]$$
$$\times\begin{Bmatrix}K & K' & \bar{K}\\ G & H & 1\\ G' & H' & 1\end{Bmatrix}; \tag{3.10b}$$

$$S_+(K,K',\bar{K};H,H',H'';j,j')=-3(-1)^{G+H+H'+H''}[K,K',\bar{K}]\chi_{H''G}^{(j)}[\chi_{H'G}^{(j')}]^*(\gamma-i\Delta_{H''G}^{(j)})^{-1}\begin{Bmatrix}K & K' & \bar{K}\\ H'' & H' & H\end{Bmatrix}\begin{Bmatrix}1 & 1 & \bar{K}\\ H' & H'' & G\end{Bmatrix}, \tag{3.11}$$

$$S_-(K,K',\bar{K};H,H',H'';j,j')=-3(-1)^{H'-G+K+K'}[K,K',\bar{K}]\chi_{HG}^{(j)}[\chi_{H''G}^{(j')}]^*(\gamma+i\Delta_{H''G}^{(j')})^{-1}\begin{Bmatrix}K & K' & \bar{K}\\ H'' & H & H'\end{Bmatrix}\begin{Bmatrix}1 & 1 & \bar{K}\\ H & H'' & G\end{Bmatrix}, \tag{3.11a}$$

$$S(K,K',\bar{K};H,H',G,G';j,,j')=3(-1)^{1+K+\bar{K}}[K,K',\bar{K}]\chi_{HG}^{(j)}[\chi_{H'G'}^{(j')}]^*[(\gamma+i\Delta_{H'G'}^{(j')})^{-1}+(\gamma-i\Delta_{HG}^{(j)})^{-1}]\begin{Bmatrix}K & K' & \bar{K}\\ H & G & 1\\ H' & G' & 1\end{Bmatrix}. \tag{3.11b}$$

IV. GROUND-STATE DENSITY-MATRIX EVOLUTION

In many problems associated with laser cooling or optical pumping, the ground state evolves on a time scale τ' that is slow compared with Γ^{-1}, i.e.,

$$\Gamma\tau'\gg 1, \tag{4.1}$$

and the Rabi frequencies are smaller than Γ, i.e.,

$$|\chi_{HG}^{(j)}|\ll\Gamma. \tag{4.2}$$

These conditions allow one to adiabatically eliminate both the optical coherences and excited-state density-matrix elements in Eqs. (3.4) or (3.9). Condition (4.2) allows one to neglect all excited-state density-matrix elements in the rhs of Eq. (3.9a). If ground-state density-matrix elements [in the "interaction picture" (3.3)] vary slowly with respect to Γ^{-1}, the approximate solution of Eq. (3.9a) is

$$\rho_{Q'}^{K'}(H,H';\mathbf{p},\mathbf{p}')=(-1)^{Q''}\begin{pmatrix}K' & K'' & \bar{K}\\ Q' & -Q'' & \bar{Q}\end{pmatrix}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')e^{-i\Omega_{jj'}t}(\Gamma-i\Delta_{HG''}^{(j)}+i\Delta_{H'G'''}^{(j')})^{-1}$$
$$\times S(K',K'',\bar{K};H,H',G'',G''';j,j')\rho_{Q''}^{K''}(G'',G''';\mathbf{p}-\hbar\mathbf{k}_j,\mathbf{p}'-\hbar\mathbf{k}_{j'}). \tag{4.3}$$

Substituting this expression in Eqs. (2.15) and then in Eq. (3.9), and introducing higher-order tensor products

$$\lambda_{\tilde{Q}}^{\tilde{K}}(\mathbf{n};\bar{K},\bar{K}';j,j')=(-1)^{\bar{K}+\bar{K}'+\tilde{Q}}[\tilde{K}]\begin{pmatrix} \bar{K} & \bar{K}' & \tilde{K} \\ \bar{Q} & \bar{Q}' & -\tilde{Q} \end{pmatrix} Y_{\bar{K}\bar{Q}}(\mathbf{n})\epsilon_{\bar{Q}'}^{\bar{K}'}(j,j') \tag{4.4}$$

in carrying out the summation over magnetic quantum numbers, one finds that the ground-state density matrix evolves as

$$\begin{aligned}\left(\frac{\partial}{\partial t}+i\omega_{GG'}+i\omega_{\mathbf{pp}'}\right)\rho_Q^K(G,G';\mathbf{p},\mathbf{p}')=(-1)^{Q'}\begin{pmatrix} K & K' & \tilde{K} \\ Q & -Q' & \tilde{Q} \end{pmatrix} e^{-i\Omega_{jj'}t}\Bigg\{ & T(K,K',\tilde{K},\bar{K},\bar{K}';G,G',G'',G''',j,j')\frac{3}{8\pi} \\ & \times\int d\mathbf{n}\lambda_{\tilde{Q}}^{\tilde{K}}(\mathbf{n};\bar{K},\bar{K}';j,j')\rho_{Q'}^{K'}(G'',G''';\mathbf{p}+\hbar(\mathbf{k}-\mathbf{k}_j),\mathbf{p}'+\hbar(\mathbf{k}-\mathbf{k}_{j'}))+\epsilon_{Q}^{\tilde{K}}(j,j') \\ & \times[S_+(K,K',\tilde{K};G,G',G'';j,j')\rho_{Q'}^{K'}(G,G'';\mathbf{p},\mathbf{p}'+\hbar\mathbf{k}_{jj'}) \\ & +S_-(K,K',\tilde{K};G,G',G'';j,j')\rho_{Q'}^{K'}(G'',G';\mathbf{p}-\hbar\mathbf{k}_{jj'},\mathbf{p}')]\Bigg\},\end{aligned} \tag{4.5}$$

where

$$\begin{aligned}T(K,K',\tilde{K},\bar{K},\bar{K}';G,G',G'',G''';j,j')=3(-1)^{1+\bar{K}+K'}B_{\bar{K}}[H,H',K,K',\tilde{K},\bar{K},\bar{K}']\gamma(H,H',G,G')\chi_{HG''}^{(j)}[\chi_{H'G'''}^{(j')}]^* \\ \times(\gamma-i\Delta_{HG''}^{(j)})^{-1}(\gamma+i\Delta_{H'G'''}^{(j')})^{-1}A(K,K',\tilde{K},\bar{K},\bar{K}';G,G';H,H';G'',G''')\end{aligned} \tag{4.6}$$

and

$$A(K,K',\tilde{K},\bar{K},\bar{K}';G,G';H,H';G'',G''')=[X^2]\begin{Bmatrix} K & K' & \tilde{K} \\ \bar{K}' & \bar{K} & X \end{Bmatrix}\begin{Bmatrix} K & X & \bar{K} \\ G & H & 1 \\ G' & H' & 1 \end{Bmatrix}\begin{Bmatrix} X & K' & \bar{K}' \\ H & G'' & 1 \\ H' & G''' & 1 \end{Bmatrix}. \tag{4.6a}$$

For simplicity, we have set $\gamma=\Gamma/2$ in deriving Eq. (4.5).

Equation (4.6a) is a representation of the $15-J$ symbol of the third kind [34],

$$A(K,K',\tilde{K},\bar{K},\bar{K}';G,G';H,H';G'',G''')=(-1)^{G+G'+H+H'+K+K'+\tilde{K}}\left\{\begin{matrix} G & G'' & K & K' & G' \\ G''' & H & \tilde{K} & H' & 1 \\ 1 & \bar{K} & \bar{K}' & 1 & 1 \end{matrix}\middle|\, 3\right\}. \tag{4.7}$$

In the absence of recoil, Eq. (4.5) reduces to ground-state density-matrix equations derived previously [1] [see also Eq. (2A) in [2]].

One can simplify the rhs of the density matrix equations (4.5) in the "$\mathbf{s}-\mathbf{u}$" representation (2.16). Replacing the in-term in Eq. (4.5) by that given in (2.17), one can obtain

$$\begin{aligned}\left[\frac{\partial}{\partial t}+i\omega_{GG'}+\frac{i}{m}\left(\frac{\partial}{\partial\mathbf{s}}\cdot\frac{\partial}{\partial\mathbf{u}}\right)\right]\rho_Q^K(G,G';\mathbf{s},\mathbf{u})=(-1)^{Q'}\begin{pmatrix} K & K' & \tilde{K} \\ Q & -Q' & \tilde{Q} \end{pmatrix}\exp[i(\mathbf{k}_{jj'}\cdot\mathbf{s}-\Omega_{jj'}t)] \\ \times P_{\tilde{Q}}^{\tilde{K}}(K,K';G,G';G'',G''';j,j')\rho_{Q'}^{K'}(G'',G''';\mathbf{s},\mathbf{u}),\end{aligned} \tag{4.8}$$

where

$$P_{\tilde{Q}}^{\tilde{K}}(K,K';G,G';G'',G''';j,j')=\lambda_{\tilde{Q}}^{\tilde{K}}(\mathbf{n}_u,\bar{K},\bar{K}';j.j')3(-1)^{1+\bar{K}+K'}\bar{B}_{\bar{K}}j_{\bar{K}}(ku)\exp(i\mathbf{u}\cdot\overline{\mathbf{k}_{jj'}})$$
$$\times[H,H',K,K',\tilde{K},\bar{K},\bar{K}']\gamma(H,H',G,G')\chi_{HG''}^{(j)}[\chi_{H'G'''}^{(j')}]^*(\gamma-i\Delta_{HG''}^{(j)})^{-1}$$
$$\times(\gamma+i\Delta_{H'G'''}^{(j)})^{-1}A(K,K',\tilde{K},\bar{K},\bar{K}';G,G';H,H';G'',G''')$$
$$+\epsilon_{\tilde{Q}}^{\tilde{K}}(j,j')[\exp(-i\mathbf{u}\cdot\mathbf{k}_{jj'}/2)S_+(K,K',\tilde{K};G,G',G''';j,j')\delta_{G''G}$$
$$+\exp(i\mathbf{u}\cdot\mathbf{k}_{jj'}/2)S_-(K,K',\tilde{K};G,G',G'';j,j')\delta_{G'''G'}] \qquad (4.9)$$

and $\overline{\mathbf{k}_{jj'}}=(\mathbf{k}_j+\mathbf{k}_{j'})/2$.

As was noted above, evolution equations in one dimension can be obtained when all fields propagate along the quantization axis, i.e., when $\mathbf{k}_j=(0,0,k_j)$, $\mathbf{k}_{jj'}=(0,0,k_{jj'})$. The corresponding one-dimensional equations for ground-state density-matrix elements can be derived in the same manner as for the three-dimensional Eqs. (4.5,4.8). Using the in terms (2.21) and (2.22), one arrives at

$$\left(\frac{\partial}{\partial t}+i\omega_{GG'}+i\omega_{pp'}\right)\rho_Q^K(G,G';p,p')=(-1)^{Q'}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')e^{-i\Omega_{jj'}t}\left\{R_{QQ'\bar{Q}}^{KK'\bar{K}}(\bar{K}';G,G';G'',G''';j,j')\right.$$
$$\times\int_{-1}^{1}dx P_{\bar{K}'}(x)\rho_{Q'}^{K'}(G'',G''';p+\hbar(kx-k_j),p'+\hbar(kx-k_{j'}))$$
$$+\begin{pmatrix}K & K' & \bar{K}\\ Q & -Q' & \bar{Q}\end{pmatrix}[S_+(K,K',\bar{K};G,G',G'';j,j')\rho_{Q'}^{K'}(G,G'';p,p'+\hbar k_{jj'})$$
$$\left.+S_-(K,K',\bar{K};G,G',G'';j,j')\rho_{Q'}^{K'}(G'',G';p-\hbar k_{jj'},p')]\right\} \qquad (4.10)$$

and

$$\left(\frac{\partial}{\partial t}+i\omega_{GG'}+\frac{i}{m}\frac{\partial^2}{\partial s\partial u}\right)\rho_Q^K(G,G';s,u)=(-1)^{Q'}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')\exp[i(k_{jj'}\cdot s-\Omega_{jj'}t)]\left\{2(-1)^{\bar{K}'/2}\exp(iu\bar{k}_{jj'})\right.$$
$$\times j_{\bar{K}'}(ku)R_{QQ'\bar{Q}}^{KK'\bar{K}}(\bar{K}';G,G';G'',G''';j,j')+\begin{pmatrix}K & K' & \bar{K}\\ Q & -Q' & \bar{Q}\end{pmatrix}$$
$$\times[\exp(-iuk_{jj'}/2)S_+(K,K',\tilde{K};G,G',G''';j,j')\delta_{G''G}$$
$$\left.+\exp(iuk_{jj'}/2)S_-(K,K',\tilde{K};G,G',G'';j,j')\delta_{G'''G'}]\right\}\rho_{Q'}^{K'}(G'',G''';s,u), \qquad (4.11)$$

where

$$R_{QQ'\bar{Q}}^{KK'\bar{K}}(\bar{K}';G,G';G'',G''';j,j')=(-1)^Q\begin{pmatrix}K & X & \bar{K}'\\ Q & -Q & 0\end{pmatrix}\begin{pmatrix}X & K' & \bar{K}\\ Q & -Q' & \bar{Q}\end{pmatrix}(\Gamma-i\Delta_{HG''}^{(j)}+i\Delta_{H'G'''}^{(j')})^{-1}$$
$$\times S(X,K',\bar{K};H,H';G'',G''';j.j')B_{K'}^{a}I(K,X,\bar{K}';G,G';H,H'). \qquad (4.12)$$

V. CONCLUSION

It has seen that atomic recoil can be included in the equations for ground- and excited-states density-matrix elements when fields drive the transition between electronic-state manifolds. In this article we have focused on the effects of recoil associated with spontaneous emission. Since the in term (2.15) is a tensor product of the excited-state density-matrix elements in a spherical tensor basis and a spherical harmonic $Y_{\bar{K}\bar{Q}}(\mathbf{n})$, it is covariant with respect to rotation. The contribution to the in term from a given direction of spontaneous emission **n** depends on the momentum distribution of the excited states' populations and internal coherences through the factor $\rho_{Q'}^{K'}(H,H';\mathbf{p}+\hbar k\mathbf{n},\mathbf{p}'+\hbar k\mathbf{n})$. Covariance is established by the coupling of spatial (center-of-mass momenta) and internal (magnetic quantum numbers, total angular momenta) degrees of freedom. This coupling can lead to Faraday rotation of a probe field's polarization vector when the probe field passes through an atomic beam (considered for a $G=0\rightarrow H=1$ transition in Ref. [29]) or an atomic vapor that is also driven by a pump field (considered for a $G=\frac{1}{2}\rightarrow H=\frac{1}{2}$ transition in Ref. [9]). Equation (4.5) allows one to generalize these results to arbitrary manifolds, which we plan to present in a future paper.

The density-matrix elements appearing in the master equations (3.9, 4.5, 4.8) are functions of three-dimensional vector quantities. For collinear radiation fields these equations can be reduced to ones in which the density-matrix elements are functions of one-dimensional variables, with all dependence on transverse momenta $(\mathbf{p}_{\perp},\mathbf{p}_{\perp}')$ or transverse coordinates $(\mathbf{s}_{\perp},\mathbf{u}_{\perp})$ eliminated [see Eqs. (4.10, 4.11)]. The price one pays for this reduction is that Eqs. (4.10, 4.11) are no longer rotationally covariant; under rotation, the momenta or coordinates would be rotated outside of the reduced one-dimensional subspace. When the contribution from the in term is included, the multipoles ρ_Q^K no longer are tensor quantities; their properties with respect to rotation no longer obey simple transformation properties.

Two representations, momentum "$\mathbf{p}-\mathbf{p}'$" and "coordinate" "$\mathbf{s}-\mathbf{u}$," have been used in this article. The momentum representation is convenient for steady-state problems, such as pump-probe or nonlinear spectroscopy. In the momentum representation the in term contains an integral, which can be evaluated numerically or using various perturbation schemes. In the coordinate representation the in term is local; however, the corresponding equations (4.8, 4.11) include mixed second derivatives in the variables **s** and **u**. The situation improves for a number of transient problems, such as resonant Kapitza-Dirac scattering, wave-matter interference, atomic Talbot effect, GSE, or MGE. When the atom-field interaction time is smaller than inverse frequency transition $\omega_{\mathbf{pp}'}^{-1}$ one can use the Raman-Nath approximation and omit the second derivatives in Eqs. (4.8, 4.11), greatly simplifying the equations. For example, if the spatial or temporal envelope functions and amplitudes of the incident traveling wave electric fields coincide with one another and if the coherences $\rho_Q^K(G,G';s,u)$ for $G\neq G'$ can be neglected, one can use the method of Ref. [3] to solve Eqs. (4.8, 4.11). An important example where the field requirements can be met is the scattering of atoms by a standing wave field (resonance Kapitza-Dirac effect [10–14]). Using this technique one can calculate the total momentum distribution of the scattered particles $\Sigma_G[G]\rho_0^0(G,G;p,p)$, as well as the momentum distribution associated with the various ground-state multipoles.

Finally, we would like to point out that, in adiabatically eliminating the optical coherences and excited-state density-matrix elements, we neglect recoil shifts in the resonance denominators of the rhs of Eqs. (3.7, 4.3). These recoil shifts can be responsible for RIR when pump and probe fields are collinear [7]. To consider RIR for this case one needs to start from the exact master equations (3.4).

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