Rate equations between electronic-state manifolds

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Rate equations are derived to describe the interaction with an ensemble of atoms of a number of optical fields having arbitrary polarizations. The fields drive transitions between two manifolds of levels, each manifold consisting of magnetically degenerate fine and hyperfine levels. The rate equations are written in an irreducible tensor notation using a coupled tensor basis for the fields' polarizations, which significantly simplifies the equations. Validity conditions for the rate equations are discussed, an expression for the friction force of laser cooling is given, and specific values for elements of the coupled-basis polarization tensor are tabulated.

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In considering the interaction of radiation with matter, it is sometimes possible to obtain rate equations for atomic-state populations [1]. For example, the optical Bloch equations for a "two-level" atom interacting with a radiation field can be reduced to rate equations if the atomic-state coherence between the two levels decays or oscillates at a rate which is much larger than that at which the atomic-state populations evolve. The two-level approximation is inadequate if one wishes to include effects relating to magnetic-state degeneracy or radiatively induced coupling between fine and hyperfine levels within the electronic-state manifolds. In this paper, we derive rate equations that describe a situation in which optical fields of arbitrary polarization drive transitions between two manifolds of levels, each manifold consisting of a number of fine and hyperfine levels. In deriving these equations, we introduce a coupled polarization-tensor basis which facilitates the calculation.

DENSITY-MATRIX EQUATIONS

The density-matrix equations describing the system of interest have been given previously [2,3], but we present them here to make this paper self-contained and to introduce the notation [4]. We consider the interaction of several radiation fields with an ensemble of "active" atoms. The incident laser fields drive transitions between a ground-state manifold characterized by quantum numbers L_G (total orbital angular momentum), S_G (total spin angular momentum), J_G (coupling of L_G and S_G), I (total nuclear-spin angular momentum), G (coupling of J_G and I), and an excited-state manifold characterized by quantum numbers L_H, S_H, J_H, I, and H. In the resonance or rotating-wave approximation,

$$\begin{aligned}\dot{\tilde{\rho}}^K_Q(G,G') = {} & \gamma^{(K)}(H,H';G,G')e^{i(\omega_{GG'}-\omega_{HH'})t}\tilde{\rho}^K_Q(H,H') \\ & +i\chi^{(j)}_{HG'}(-1)^{G+G'}e^{i(\mathbf{k}_j\cdot\mathbf{R}-\Delta^{(j)}_{HG'}t)}(-1)^q\epsilon^{(j)}_{-q}\Lambda^{K'1K}_{Q'qQ}(G',G,H)\tilde{\rho}^{K'}_{Q'}(G,H) \\ & -i(\chi^{(j)}_{HG})^*(-1)^{2H+1+K'-K+Q'}e^{-i(\mathbf{k}_j\cdot\mathbf{R}-\Delta^{(j)}_{HG}t)}(\epsilon^{(j)}_q)^*\Lambda^{K'1K}_{Q'qQ}(G,G',H)[\tilde{\rho}^{K'}_{-Q'}(G',H)]^* ,\end{aligned} \tag{1a}$$

$$\begin{aligned}\dot{\tilde{\rho}}^K_Q(H,H') = {} & -\Gamma\tilde{\rho}^K_Q(H,H')-i\chi^{(j)}_{HG}(-1)^{H+H'+K'-K+1}e^{i(\mathbf{k}_j\cdot\mathbf{R}-\Delta^{(j)}_{HG}t)}(-1)^q\epsilon^{(j)}_{-q}\Lambda^{K'1K}_{Q'qQ}(H,H',G)\tilde{\rho}^{K'}_{Q'}(G,H') \\ & +i(\chi^{(j)}_{H'G})^*(-1)^{2G+Q'}e^{-i(\mathbf{k}_j\cdot\mathbf{R}-\Delta^{(j)}_{H'G}t)}(\epsilon^{(j)}_q)^*\Lambda^{K'1K}_{Q'qQ}(H',H,G)[\tilde{\rho}^{K'}_{-Q'}(G,H)]^* ,\end{aligned} \tag{1b}$$

$$\begin{aligned}\dot{\tilde{\rho}}^K_Q(G,H) = {} & -(\Gamma/2)\tilde{\rho}^K_Q(G,H)+i(\chi^{(j)}_{HG'})^*(-1)^{G+G'}e^{-i(\mathbf{k}_j\cdot\mathbf{R}-\Delta^{(j)}_{HG'}t)}(\epsilon^{(j)}_q)^*\Lambda^{K'1K}_{Q'qQ}(H,G,G')\tilde{\rho}^{K'}_{Q'}(G,G') \\ & -i(\chi^{(j)}_{H'G})^*(-1)^{2G+Q'+K'-K+1}e^{-i(\mathbf{k}_j\cdot\mathbf{R}-\Delta^{(j)}_{H'G}t)}(\epsilon^{(j)}_q)^*\Lambda^{K'1K}_{Q'qQ}(G,H,H')[\tilde{\rho}^{K'}_{-Q'}(H,H')]^* ,\end{aligned} \tag{1c}$$

$$\tilde{\rho}^K_Q(F',F)=(-1)^{F-F'+Q}[\tilde{\rho}^K_{-Q}(F,F')]^* . \tag{1d}$$

The symbols appearing in Eq. (1) are defined as follows: The $\tilde{\rho}^K_Q(F,F')$ ($F,F'=G$ or H) are atomic density matrix elements written in an irreducible tensor basis. They are written in an interaction representation and are related to density-matrix elements $\rho^K_Q(F,F')$ in the "normal" representation by

$$\tilde{\rho}^K_Q(F,F')=\rho^K_Q(F,F')\exp(i\omega_{FF'}t) , \tag{2}$$

where

$$\omega_{FF'}=(E_F-E_{F'})/\hbar \tag{3}$$

and E_F is the energy of state F. The density-matrix elements in the irreducible tensor basis are related to those in the m basis by

$$\rho_Q^K(F,F')=\sum_{m,m'}(-1)^{F'-m'}\langle F,m;F',-m'|K,Q\rangle \times\rho(F,m;F',m') , \quad (4)$$

where $\langle F_1,m_1;F_2;m_2|F,m\rangle$ is a Clebsch-Gordan coefficient [5]. The quantity $\gamma^{(K)}(H,H';G,G')$, which characterizes the rate of spontaneous emission of the Kth "multipole moment" from levels H and H' to G and G', is defined as

$$\gamma^{(K)}(H,H';G,G')=(-1)^{H+K+G'+1} \times[(2H+1)(2H'+1)]^{1/2} \times\begin{Bmatrix} H & H' & K \\ G' & G & 1\end{Bmatrix}\gamma(H,H';G,G') , \quad (5a)$$

where $\{\ \}$ is a 6-J symbol,

$$\gamma(H,H';G,G')=[4/(3\hbar)](\omega_{HG}\omega_{H'G'}/c^2)^{3/2} \times[(2H+1)(2H'+1)]^{-1/2} \times p_{GH}(p_{G'H'})^* , \quad (5b)$$

and p_{GH} is the reduced density-matrix element of the dipole moment operator between states G and H. [Note that $p_{GH}=(-1)^{G-H}(p_{HG})^*$.] The Rabi frequency $\chi_{HG}^{(j)}$ associated with field j is defined in terms of the (complex) amplitude of field j. The incident field $\mathbf{E}$ written as

$$\mathbf{E}(\mathbf{R},t)=\sum_{j=1}^{N}\tfrac{1}{2}\mathcal{E}^{(j)}\hat{\boldsymbol{\epsilon}}^{(j)}e^{i(\mathbf{k}_j\cdot\mathbf{R}-\Omega_j t)}+\text{c.c.} , \quad (6)$$

where N is the number of incident fields and $\mathcal{E}^{(j)}$ is the (complex) amplitude, $\hat{\boldsymbol{\epsilon}}^{(j)}$ is the (complex) polarization, $\mathbf{k}_j$ is the propagation vector, and Ω_j is the frequency of field j. The Rabi frequency $\chi_{HG}^{(j)}$ is defined as

$$\chi_{HG}^{(j)}=p_{HG}\mathcal{E}^{(j)}/(2\sqrt{3}\hbar) , \quad (7)$$

while the spherical components of $\epsilon_q^{(j)}$ of the polarization $\hat{\boldsymbol{\epsilon}}^{(j)}$ are defined by

$$\epsilon_{\pm1}^{(j)}=\mp(\epsilon_x^{(j)}\pm\epsilon_y^{(j)})/\sqrt{2}, \quad \epsilon_0^{(j)}=\epsilon_z^{(j)} . \quad (8)$$

The detunings $\Delta_{HG}^{(j)}$ are defined as

$$\Delta_{HG}^{(j)}=\Omega_j-\omega_{HG} \quad (9)$$

and $\Lambda_{Q'qQ}^{K'kK}(A,B,C)$ is defined as

$$\Lambda_{Q'qQ}^{K'kK}(A,B,C)=(-1)^{k+K}[(2k+1)(2K'+1)]^{1/2} \times\langle K',Q';k,q|K,Q\rangle\begin{Bmatrix} K' & k & K \\ A & B & C\end{Bmatrix} . \quad (10)$$

The spontaneous decay rate of each of the excited states is equal to Γ. The time derivatives in Eqs. (1) are total time derivatives in the sense that

$$\frac{d}{dt}=\frac{\partial}{\partial t}+\mathbf{v}\cdot\boldsymbol{\nabla} , \quad (11)$$

where $\mathbf{v}$ is the atomic velocity. [The modification of the density-matrix equations resulting from any light-induced atomic acceleration is *not* included in Eq. (1).] Finally, there is a summation convention implicit in Eq. (1) that will be used in all subsequent equations. Repeated indices appearing on the right-hand side (rhs) of an equation are to be summed over, *except* if these indices also appear on the left-hand side (lhs) of the equation.

For specific experimental conditions, it might be necessary to add additional terms to Eqs. (1) such as those arising from external magnetic fields, collisions, "source" terms which bring atoms into the interaction volume, or loss terms resulting from atoms leaving the interaction volume. In obtaining Eqs. (1), it has been assumed implicitly that all the magnetic sublevels within a state of given F are degenerate. Generalization of the results to allow for a splitting of the magnetic-state sublevels will be discussed following a derivation of the rate equations.

RATE EQUATIONS

To obtain rate equations it is necessary to express the electronic state coherences in terms of the atomic state populations. It is not possible to do this directly in Eq. (1c) owing to the exponential factors in those equations. To remove these factors, a trial solution of the form

$$\tilde{\rho}_Q^K(G,H)=\tilde{\rho}_Q^K(G,H;j,G',H')e^{-i(\mathbf{k}_j\cdot\mathbf{R}-\Delta_{H'G'}^{(j)}t)} \quad (12)$$

is substituted into Eq. (1c). The $\tilde{\rho}_Q^K(G,H;j,G',H')$ satisfy an equation similar to Eq. (1c), except that the exponential factors are missing and $(\Gamma/2)$ is replaced by $\{(\Gamma/2)+i[\Delta_{H'G'}^{(j)}-\mathbf{k}_j\cdot\mathbf{v}]\}$. If the atomic state "populations" $\tilde{\rho}_Q^K(G,G')$ and $\tilde{\rho}_Q^K(H,H')$ are slowly varying (both spatially and temporally) with respect to $\tilde{\rho}_Q^K(G,H;j,G',H')$, an approximate solution of Eq. (1c) is

$$\tilde{\rho}_{Q'}^{K'}(G,H)=i(-1)^{G+G''}(\chi_{HG''}^{(j')})^*e^{-i(\mathbf{k}_{j'}\cdot\mathbf{R}-\Delta_{HG''}^{(j')}t)}(\epsilon_q^{(j')})^*\Lambda_{Q''qQ'}^{K''1K'}(H,G,G'')[(\Gamma/2)+i\Delta_{HG''}^{(j')}(v)]^{-1}\tilde{\rho}_{Q''}^{K''}(G,G'')$$
$$-i(\chi_{H''G}^{(j')})^*(-1)^{2G+Q''+K''-K'+1}e^{-i(\mathbf{k}_{j'}\cdot\mathbf{R}-\Delta_{H''G}^{(j')}t)}(\epsilon_q^{(j')})^*$$
$$\times\Lambda_{Q''qQ'}^{K''1K'}(G,H,H'')[(\Gamma/2)+i\Delta_{H''G}^{(j')}(v)]^{-1}[\tilde{\rho}_{-Q''}^{K''}(H,H'')]^* , \quad (13)$$

where

$$\Delta_{HG}^{(j)}(v)=\Delta_{HG}^{(j)}-\mathbf{k}_j\cdot\mathbf{v} . \quad (14)$$

The validity conditions for this approximation are discussed below. When the expression for $\tilde{\rho}_{Q'}^{K'}(G,H)$ is substituted into Eqs. (1a) and (1b), one arrives at rate equations for $\tilde{\rho}_Q^K(G,G')$ and $\tilde{\rho}_Q^K(H,H')$. These equations can be simplified considerably if one introduces a coupled tensor basis defined by

$$\epsilon_{\bar{Q}}^{\bar{K}}(j,j')=(-1)^{q'}\epsilon_q^{(j)}(\epsilon_{-q'}^{(j')})^*\langle 1,q;1,q'|\bar{K},\bar{Q}\rangle \ , \tag{15}$$

from which it follows that

$$(-1)^q\epsilon_{-q}^{(j)}(\epsilon_{q'}^{(j')})^*=(-1)^{\bar{Q}}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')\langle 1,-q;1,-q'|\bar{K},\bar{Q}\rangle \tag{16a}$$

and

$$(-1)^{q'}\epsilon_{-q'}^{(j)}(\epsilon_q^{(j')})^*=(-1)^{\bar{Q}+\bar{K}}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')\times\langle 1,-q;1,-q'|\bar{K},\bar{Q}\rangle \ . \tag{16b}$$

If one substitutes Eq. (13) into Eqs. (1a) and (1b) and uses Eqs. (10) and (16), it is possible to carry out the summations over q and q' in the products of the three Clebsch-Gordon coefficients which appear [5]; the resultant expression can then be summed over K' using properties of the 6-J symbols [5]. In this manner one obtains

$$\begin{aligned}\dot{\rho}_Q^K(G,G')=&-i\omega_{GG'}\rho_Q^K(G,G')+\gamma^{(K)}(H,H';G,G')\rho_Q^K(H,H')+S_{KQ}^{K'Q'}(G,G';G'',G''';\mathbf{R},t)\rho_{Q'}^{K'}(G'',G''')\\&+S_{KQ}^{K'Q'}(G,G';H,H';\mathbf{R},t)\rho_{Q'}^{K'}(H,H') \ ,\end{aligned} \tag{17a}$$

$$\dot{\rho}_Q^K(H,H')=-(\Gamma+i\omega_{HH'})\rho_Q^K(H,H')+S_{KQ}^{K'Q'}(H,H';H'',H''';\mathbf{R},t)\rho_{Q'}^{K'}(H'',H''')+S_{KQ}^{K'Q'}(H,H';G,G';\mathbf{R},t)\rho_{Q'}^{K'}(G,G') \ , \tag{17b}$$

where

$$\begin{aligned}S_{KQ}^{K'Q'}(G,G';G'',G''';\mathbf{R},t)=&-3[(2K+1)(2K'+1)]^{1/2}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')(-1)^{Q+H-G}e^{i\phi_{jj'}(\mathbf{R},t)}\\&\times\langle K',Q';K,-Q|\bar{K},\bar{Q}\rangle\left[(-1)^{K+\bar{K}+K'}\chi_{HG'}^{(j)}[\chi_{HG'''}^{(j')}]^*\begin{Bmatrix}K & K' & \bar{K}\\ G''' & G' & G\end{Bmatrix}\begin{Bmatrix}1 & 1 & \bar{K}\\ G''' & G' & H\end{Bmatrix}\right.\\&\times[(\Gamma/2)+i\Delta_{HG'''}^{(j')}(v)]^{-1}\delta_{G'',G}\\&+(-1)^{G''-G'}\chi_{HG''}^{(j)}[\chi_{HG}^{(j')}]^*\begin{Bmatrix}K & K' & \bar{K}\\ G'' & G & G'\end{Bmatrix}\begin{Bmatrix}1 & 1 & \bar{K}\\ G'' & G & H\end{Bmatrix}\\&\left.\times[(\Gamma/2)-i\Delta_{HG''}^{(j)}(v)]^{-1}\delta_{G''',G'}\right] \ ,\end{aligned} \tag{18a}$$

$$\begin{aligned}S_{KQ}^{K'Q'}(G,G';H,H';\mathbf{R},t)=&\,3[(2K+1)(2K'+1)]^{1/2}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')(-1)^{Q+1+G'-G+H'-H+K'}e^{i\phi_{jj'}(\mathbf{R},t)}\\&\times\langle K',Q';K,-Q|\bar{K},\bar{Q}\rangle\chi_{H'G'}^{(j)}[\chi_{HG}^{(j')}]^*\begin{Bmatrix}K & K' & \bar{K}\\ G & H & 1\\ G' & H' & 1\end{Bmatrix}\\&\times\{[(\Gamma/2)+i\Delta_{HG}^{(j')}(v)]^{-1}+[(\Gamma/2)-i\Delta_{H'G'}^{(j)}(v)]^{-1}\} \ ,\end{aligned} \tag{18b}$$

$$\begin{aligned}S_{KQ}^{K'Q'}(H,H';H'',H''';\mathbf{R},t)=&-3[(2K+1)(2K'+1)]^{1/2}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')(-1)^{Q+G-H'-\bar{K}}e^{i\phi_{jj'}(\mathbf{R},t)}\\&\times\langle K',Q';K,-Q|\bar{K},\bar{Q}\rangle\\&\times\left[(-1)^{K+\bar{K}-K'-2G-H-H'''}\chi_{H'''G}^{(j)}[\chi_{H'G}^{(j')}]^*\right.\\&\times\begin{Bmatrix}K & K' & \bar{K}\\ H''' & H' & H\end{Bmatrix}\begin{Bmatrix}1 & 1 & \bar{K}\\ H''' & H' & G\end{Bmatrix}[(\Gamma/2)-i\Delta_{H'''G}^{(j)}(v)]^{-1}\delta_{H'',H}\\&\left.+\chi_{HG}^{(j)}[\chi_{H''G}^{(j')}]^*\begin{Bmatrix}K & K' & \bar{K}\\ H'' & H & H'\end{Bmatrix}\begin{Bmatrix}1 & 1 & \bar{K}\\ H'' & H & G\end{Bmatrix}[(\Gamma/2)+i\Delta_{H''G}^{(j')}(v)]^{-1}\delta_{H''',H'}\right] \ ,\end{aligned} \tag{18c}$$

$$S_{KQ}^{K'Q'}(H,H';G,G';\mathbf{R},t)$$
$$=3[(2K+1)(2K'+1)]^{1/2}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')(-1)^{Q+1+\bar{K}+K'}e^{i\phi_{jj'}(\mathbf{R},t)}$$
$$\times\langle K',Q';K,-Q|\bar{K},\bar{Q}\rangle\chi_{HG}^{(j)}[\chi_{H'G'}^{(j')}]^{*}\begin{Bmatrix}K & K' & \bar{K}\\ H & G & 1\\ H' & G' & 1\end{Bmatrix}\{[(\Gamma/2)-i\Delta_{HG}^{(j)}(v)]^{-1}+[(\Gamma/2)+i\Delta_{H'G'}^{(j')}(v)]^{-1}\}\ , \tag{18d}$$

$$\phi_{jj'}(\mathbf{R},t)=(\mathbf{k}_j-\mathbf{k}_{j'})\cdot\mathbf{R}-(\Omega_j-\Omega_{j'})t\ , \tag{19}$$

and the term in large curly brackets is a 9-j symbol. Equation (17) is written in the "normal" rather than the interaction representation. Consistent with the summation convention adopted in this work, Eqs. (18) contain a sum over j, j', $\bar{K}$, and $\bar{Q}$. As a check on the equations, one can verify that Eqs. (17a), (18a), and (18b) go over into Eqs. (17b), (18c), and (18d) on the interchange of G and H, provided one replaces $\Delta_{GH}^{(j)}$ by $-\Delta_{HG}^{(j)}$ and $\epsilon_{\bar{Q}}^{\bar{K}}(j',j)$ by $(-1)^{\bar{K}}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')$ [6].

The general solution of these rate equations is very complicated, involving spatial and temporal harmonics of the differences of field propagation vectors and atom-field detunings, respectively. On the other hand, these equations *are* exact to second order in the applied fields and can be solved to give a perturbative result correct to this order.

In the case of transitions between a single G and a single H state, Eqs. (17) and (18) reduce to

$$\dot{\rho}_Q^K(G)=\gamma^{(K)}(H;G)\rho_Q^K(H)+S_{KQ}^{K'Q'}(G;G;\mathbf{R},t)\rho_{Q'}^{K'}(G)$$
$$+S_{KQ}^{K'Q'}(G;H;\mathbf{R},t)\rho_{Q'}^{K'}(H)\ , \tag{20a}$$
$$\dot{\rho}_Q^K(H)=-\Gamma\rho_Q^K(H)+S_{KQ}^{K'Q'}(H;H;\mathbf{R},t)\rho_{Q'}^{K'}(H)$$
$$+S_{KQ}^{K'Q'}(H;G;\mathbf{R},t)\rho_{Q'}^{K'}(G)\ , \tag{20b}$$

where

$$S_{KQ}^{K'Q'}(\alpha,\beta;\mathbf{R},t)=A(K,K',\bar{K};Q,Q';G,H;\mathbf{R},t)T_{\alpha\beta}(G,H;K,K',\bar{K})\quad(\alpha=G,H;\beta=G,H)\ , \tag{21}$$

$$A(K,K',\bar{K};Q,Q';G,H;\mathbf{R},t)=(-1)^{Q'}\begin{pmatrix}K' & K & \bar{K}\\ -Q' & Q & \bar{Q}\end{pmatrix}\chi_{HG}^{(j)}[\chi_{HG}^{(j')}]^{*}\epsilon_{\bar{Q}}^{\bar{K}}(j,j')e^{i\phi_{jj'}(\mathbf{R},t)}$$
$$\times\{[(\Gamma/2)+i\Delta_{HG}^{(j')}(v)]^{-1}+(-1)^{K+\bar{K}+K'}[(\Gamma/2)-i\Delta_{HG}^{(j)}(v)]^{-1}\}\ , \tag{22}$$

$$T_{GG}(G,H;K,K',\bar{K})=-3(-1)^{K+K'+H-G}[(2K+1)(2K'+1)(2\bar{K}+1)]^{1/2}\begin{Bmatrix}K & K' & \bar{K}\\ G & G & G\end{Bmatrix}\begin{Bmatrix}1 & 1 & \bar{K}\\ G & G & H\end{Bmatrix}\ , \tag{23a}$$

$$T_{GH}(G,H;K,K',\bar{K})=3(-1)^{\bar{K}+K'+1}[(2K+1)(2K'+1)(2\bar{K}+1)]^{1/2}\begin{Bmatrix}K & K' & \bar{K}\\ G & H & 1\\ G & H & 1\end{Bmatrix}\ , \tag{23b}$$

$$T_{HH}(G,H;K,K',\bar{K})=(-1)^{K+K'}T_{GG}(H,G;K,K',\bar{K})\ , \tag{23c}$$

$$T_{HG}(G,H;K,K',\bar{K})=(-1)^{\bar{K}}T_{GH}(H,G;K,K',\bar{K})\ , \tag{23d}$$

$\rho_Q^K(G)\equiv\rho_Q^K(G,G)$, $\rho_Q^K(H)\equiv\rho_Q^K(H,H)$, $\gamma^{(K)}(H;G)\equiv\gamma^{(K)}(H,H;G,G)$, and the Clebsch-Gordan coefficients appearing in Eq. (18) have been converted to 3-J symbols in Eq. (22). Several values of T_{GG} and T_{GH} are tabulated in Appendix B. The fact that the 9-J symbol in Eq. (23b) vanishes unless $(K+K'+\bar{K})$ is even has been used in writing Eqs. (22) and (23).

Under certain conditions (see below), it is also possible to adiabatically eliminate the excited state density-matrix elements. If these conditions are satisfied and if, moreover, the incident fields are sufficiently weak, the approximate solution of Eq. (17b) correct to order $|\chi_{HG}^{(j)}|^2/\{(\Gamma/2)^2+[\Delta_{HG}^{(j)}(v)]^2\}$, obtained by rewriting Eq. (17b) in the interaction representation and neglecting the temporal and spatial variation of $\tilde{\rho}_Q^K(G,G')$, is

$$\rho_Q^K(H,H')=S_{KQ}^{K'Q'}(H,H';G,G';j,j';\mathbf{R},t)\rho_{Q'}^{K'}(G,G')$$
$$\times\{\Gamma-i[\Delta_{HG}^{(j)}(v)-\Delta_{H'G'}^{(j')}(v)]\}^{-1}\ , \tag{24}$$

where $S_{KQ}^{K'Q'}(H,H';G,G';j,j';\mathbf{R},t)$ is equal to the expression on the rhs of Eq. (18d) for $S_{KQ}^{K'Q'}(H,H';G,G';\mathbf{R},t)$ *without* the summation over j and j'. Such an adiabatic elimination of excited-state density-matrix elements is used routinely in theories of sub-Doppler laser cooling [7]. If Eq. (24) is substituted into Eq. (17a), one obtains an equation of motion involving ground-state density-matrix elements only. When the fields drive transitions between a single-G and a single-H state, the evolution of the ground-state density-matrix elements, obtained from Eqs. (17)–(24) and (5) is given by

$$\dot{\rho}_Q^K(G)=A(K,K',\bar{K};Q,Q';G,H;j,j';\mathbf{R},t)$$
$$\times[T_{GG}(G,H;K,K',\bar{K})+(\gamma(H;G)/\{\Gamma-i[\Delta_{HG}^{(j)}(v)-\Delta_{HG}^{(j')}(v)]\})R(G,H;K,K',\bar{K})]\rho_{Q'}^{K'}(G)\ , \tag{25}$$

where

$$R(G,H;K,K',\bar{K})=(-1)^{H+K+G+1}(2H+1)\begin{Bmatrix} H & H & K \\ G & G & 1 \end{Bmatrix} T_{HG}(G,H;K,K',\bar{K})\ , \tag{26}$$

$\gamma(H;G)\equiv\gamma(H,H;G,G)$, and $A(K,K',\bar{K};Q,Q';G,H;j,j';\mathbf{R},t)$ is equal to the expression on the rhs of Eq. (22) for $A(K,K',\bar{K};Q,Q';G,H;\mathbf{R},t)$ *without* the summation over j and j'. Several values of $R(G,H;K,K',\bar{K})$ are tabulated in Appendix B. For equal frequencies of the incident fields and for $\gamma(H,G)=\Gamma$, Eq. (25) becomes

$$\dot{\rho}_Q^K(G)=A(K,K',\bar{K};Q,Q';G,H;\mathbf{R},t)P(G,H;K,K',\bar{K})\rho_{Q'}^{K'}(G)\ , \tag{27a}$$

where

$$P(G,H;K,K',\bar{K})=T_{GG}(G,H;K,K',\bar{K})+R(G,H;K,K',\bar{K})$$
$$=-3(-1)^{K+K'+H-G}[(2K+1)(2K'+1)(2\bar{K}+1)]^{1/2}$$
$$\times\left[\begin{Bmatrix} K & K' & \bar{K} \\ G & G & G \end{Bmatrix}\begin{Bmatrix} 1 & 1 & \bar{K} \\ G & G & H \end{Bmatrix}-(-1)^{2G}(2H+1)\begin{Bmatrix} H & H & K \\ G & G & 1 \end{Bmatrix}\begin{Bmatrix} K & K' & \bar{K} \\ H & G & 1 \\ H & G & 1 \end{Bmatrix}\right]\ . \tag{27b}$$

From conservation of population, it follows that $P(G,H;0,K',\bar{K})=0$.

VALIDITY CONDITIONS

In obtaining Eq. (13) for $\tilde{\rho}_Q^K(G,H)$ in terms of $\tilde{\rho}_Q^K(G,G')$ and $\tilde{\rho}_Q^K(H,H')$, we used Eqs. (1c) and (12), along with the assumption that

$$\left|\left[\frac{\partial}{\partial t}+\mathbf{v}\cdot\nabla\right]\tilde{\rho}_Q^K(F,F')\right|\ll|\{\Gamma+i[\Delta_{HG}^{(j)}(v)]\}\times\tilde{\rho}_Q^K(F,F')|\ , \tag{28}$$

where $(F,F')=(G,G'),\ (H,H')$. In all cases, it is assumed that any transient effects resulting from temporal variations of the field amplitudes can be ignored. The quantities $\tilde{\rho}_Q^K(G,G')$ and $\tilde{\rho}_Q^K(H,H')$ contain spatial harmonics of the form $n\mathbf{k}_{jj'}\cdot\mathbf{R}\equiv n(\mathbf{k}_j-\mathbf{k}_{j'})\cdot\mathbf{R}$ and temporal harmonics of the form $n\Delta_{HG;H'G'}(j,j')\equiv n(\Delta_{HG}^{(j)}-\Delta_{H'G'}^{(j')})$, where n is an integer determined by the specifics of the problem. For example, in strong fields where $|\chi_{HG}^{(j)}|\gg\Gamma$, one may have n of order $|\chi_{HG}^{(j)}|^2/\{\Gamma^2+[\Delta_{HG}^{(j)}(v)]^2\}$ for atom-field detunings $|\Delta_{HG}^{(j)}(v)|\le\Gamma$ [8]. In that case, it follows from Eq. (28) that a sufficient condition for the validity of the adiabatic elimination of $\tilde{\rho}_Q^K(G,H)$ is

$$|\chi_{HG}^{(j)}/\{\Gamma+i[\Delta_{HG}^{(j)}(v)]\}|^2|\mathbf{k}_{jj'}\cdot\mathbf{v}-\Delta_{HG;H'G'}(j,j')|\ll|(\Gamma/2)+i\Delta_{HG}^{(j)}(v)|\ . \tag{29}$$

Condition (29) is sufficient but may not be necessary for the validity of the adiabatic elimination of $\rho_Q^K(G,H)$. For example, when an atom having $\mathbf{v}=0$ is subjected to a standing-wave field proportional to $\cos(kZ)$, the atomic populations are proportional to $[1+r\cos(2kZ)]^{-1}$, where $r=I/(1+I)$ and I is a saturation parameter proportional to $|\chi_{HG}^{(j)}/[\Gamma+i\Delta_{HG}^{(j)}]|^2$ [8]. Although the populations contain harmonics up to $n\approx I$, it is possible to show by direct substitution into Eq. (28) that the validity condition (29) can be replaced by the less severe condition

$$|\chi_{HG}^{(j)}/\{\Gamma+i[\Delta_{HG}^{(j)}(v)]\}||\mathbf{k}_{jj'}\cdot\mathbf{v}-\Delta_{HG;H'G'}(j,j')|\ll|(\Gamma/2)+i\Delta_{HG}^{(j)}(v)|\ . \tag{30}$$

For weak fields, where $|\chi_{HG}^{(j)}|\ll\Gamma$, the number of spatial and temporal harmonics of the fields that enter is roughly equal to the number N of excitation-emission cycles needed to establish equilibrium via optical pumping, *multiplied* by a factor α. The number N is typically of order $G_{\max}$, the largest value of ground-state angular momentum, while the quantity α depends on the specific field configuration. For orthogonally polarized fields j and j', α is of order unity; on the other hand, for linearly polarized fields j and j' whose polarization directions differ by a finite angle $\theta\ll1$, α is of order $\theta^{-2}\gg1$ [9]. The validity condition for the adiabatic elimination of $\tilde{\rho}_Q^K(G,H)$ becomes

$$\alpha G_{\max}|\mathbf{k}_{jj'}\cdot\mathbf{v}-\Delta_{HG;H'G'}(j,j')|\ll|(\Gamma/2)+i\Delta_{HG}^{(j)}(v)|\ . \tag{31}$$

As in the strong-field case, condition (31) is sufficient, but may not be necessary. For example, in the case alluded to above, the ground-state density-matrix elements of atoms having $\mathbf{v}=0$ vary as $-[\sin\theta\sin(2kZ)/\{1+\cos\theta\cos(2kZ)\}]$ [9]. For $\theta\ll1$, they contain spatial harmonics of order $n=\alpha\approx\theta^{-2}$, but the validity condition for the adiabatic elimination of $\tilde{\rho}_Q^K(G,H)$, obtained directly from Eq. (28), is given by the less-restrictive condition [9]

$$\sqrt{\alpha}G_{\max}|\mathbf{k}_{jj'}\cdot\mathbf{v}-\Delta_{HG;H'G'}(j,j')|\ll|(\Gamma/2)+i\Delta_{HG}^{(j)}(v)|\ . \tag{32}$$

Inequalities (29)–(32) can be satisfied for large atom-field detunings. They can also be satisfied in the Doppler limit of laser cooling where $|\mathbf{k}_{jj'}\cdot\mathbf{v}| \ll \Gamma$. Inequalities (29)–(32), with the right-hand side replaced by $|\Gamma + i\Delta_{HG;H'G'}(j,j')|$, also provide the validity criteria for the adiabatically elimination of the excited-state density-matrix elements $\rho_Q^K(H,H')$.

Consistent with the adiabatic elimination of $\tilde{\rho}_Q^K(G,H)$ [and $\rho_Q^K(H,H')$, where appropriate], it is reasonable to replace $\Delta_{HG}^{(j)}(v)$ by $\Delta_{HG}^{(j)}$ in Eqs. (13), (18), (22), (24), and (25) whenever $|\mathbf{k}_{j'}\cdot\mathbf{v}| \simeq |\mathbf{k}_{jj'}\cdot\mathbf{v}|$, since inequalities (29)–(32) require that $|\mathbf{k}_{jj'}\cdot\mathbf{v})| \ll |(\Gamma/2)+i\Delta_{HG}^{(j)}(v)|$. The v dependence in these equations has been retained, however, to ensure that these equations are exact to second order in the applied fields, regardless of the validity of the adiabatic elimination procedure.

If the magnetic sublevels within a state of given G or H have different energies owing to the presence of an external magnetic field, the adiabatic elimination of $\tilde{\rho}_Q^K(G,H)$ and $\tilde{\rho}_Q^K(H,H')$ remains valid provided that the magnetic-field-induced frequency splitting of the levels is small compared with $|(\Gamma/2)+i\Delta_{HG}^{(j)}(v)|$ and Γ, respectively. One can add terms to the right-hand sides of Eqs. (17), (20), and (25) to account for this splitting [3].

Finally, we note that inequalities (29)–(32) may not be valid for certain rapidly varying density-matrix elements. For example if $j=j'$, $H=H'$, and $G\neq G'$, then $\Delta_{HG;HG'}(j,j)=\omega_{GG'}$, which may be much larger than $|(\Gamma/2)+i\Delta_{HG}^{(j)}|$. It may still be possible to obtain rate equations of the form (20) in this limit, however, provided that the rapid temporal variation of $\tilde{\rho}_Q^K(G,G')$ results in negligible values for these density-matrix elements.

FRICTION FORCE OF LASER COOLING

The force on the atoms resulting from the incident fields is given by [10]

$$\mathbf{f}=\mathrm{Tr}[\rho\nabla(\boldsymbol{\rho}\cdot\mathbf{E})]\ , \tag{33}$$

where $\mathbf{p}$ is the dipole moment operator. Using Eq. (6) for the field, expressing the dipole moment and density-matrix operators in terms of their spherical tensor components, and using Eq. (7) for the Rabi frequency, one arrives at

$$f=i\hbar\mathbf{k}_j(-1)^{H-G}\chi_{HG}^{(j)}\epsilon_q^{(j)}\tilde{\rho}_q^1(G,H)e^{i(\mathbf{k}_j\cdot\mathbf{R}-\Delta_{HG}^{(j)}t)}+\mathrm{c.c.} \tag{34}$$

Terms varying as $\exp(2i\Omega_j t)$ have been omitted from Eq. (34). When Eq. (13) is substituted into Eq. (34), one can use the coupled tensor basis (15) and carry out the sum over q to obtain

$$\begin{aligned}\mathbf{f}=&(-1)^{K+H}3\hbar\mathbf{k}_j\exp\{i[\phi_{jj'}(\mathbf{R},t)]\}\chi_{HG}^{(j)}\epsilon_Q^K(j,j')\\ &\times\Bigg[(\chi_{HG'}^{(j')})^*(-1)^{G'}\begin{Bmatrix}K&1&1\\H&G&G'\end{Bmatrix}[(\Gamma/2)+i\Delta_{HG'}^{(j')}(v)]^{-1}\rho_Q^K(G,G')\\ &\quad-(\chi_{H'G}^{(j')})^*(-1)^{G+H-H'+K}\begin{Bmatrix}K&1&1\\G&H&H'\end{Bmatrix}[(\Gamma/2)+i\Delta_{H'G}^{(j')}(v)]^{-1}\rho_Q^K(H',H)\Bigg]+\mathrm{c.c.}\end{aligned} \tag{35}$$

In the weak-field limit, the term proportional to $\rho_Q^K(H',H)$ can be dropped from this equation. Let us consider the weak-field limit for transitions between a single-G and single-H state; moreover, let us assume that $|\mathbf{k}_j\cdot\mathbf{v}| \ll \Gamma$. The evolution of $\rho_Q^K(G)$ is then given by Eqs. (27) and (22) with $\Delta_{HG}^{(j)}(v)$ replaced by $\Delta_{HG}^{(j)}$. If there are two incident fields having $\mathbf{k}_1=-\mathbf{k}_2$, $\Omega_1=\Omega_2$, and the *same* polarization, it follows from Eqs. (27) and (35) that the steady-state solution for $\rho_Q^K(G)$ is not spatially modulated and the spatially averaged friction force vanishes. This result is consistent with the fact that there is no sub-Doppler laser cooling when there is no polarization gradient for the incident fields [7]. On the other hand, if the atoms spend a finite time in the interaction region owing to transit-time effects or other loss mechanisms (such as being optically pumped out of the interacting levels), the atomic ground-state polarization *is* spatially modulated. Owing to finite-lifetime effects, the $\rho_Q^K(G)$ of atoms in strong-field regions will differ from those in weak-field regions. The spatial modulation is directly responsible for narrow structures seen in four-wave-mixing experiments on Na[11].

CONCLUSION

We have derived rate equations that determine the evolution of ground- and excited-state density-matrix elements when a number of radiation fields drive transitions between two electronic state manifolds. The equations are valid whenever one is justified in adiabatically eliminating the electronic state coherence. Moreover, the equations are exact to second order in the applied fields so that they may be used in perturbative solutions such as those needed in theories of four-wave mixing. The introduction of a coupled polarization tensor has significantly simplified the equations from those obtained previously [3]. In the future, we plan to extend this approach to transient problems and to problems requiring the quantization of the center-of-mass motion of the atoms.

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APPENDIX A: VALUES FOR $\epsilon_Q^K(j,j')$

Several values for elements of the coupled-basis polarization tensor are given below for reference. Additional values may be obtained directly from the relationship

$$\epsilon_Q^K(j',j)=(-1)^Q[\epsilon_{-Q}^K(j,j')]^* \ . \tag{A1}$$

The following individual field polarizations are considered: Linearly polarized along the $\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$, or $\hat{\mathbf{z}}$ directions:

$$\begin{aligned}\epsilon_q^{(z)}&=\delta_{q,0} \ ,\\ \epsilon_q^{(x)}&=(\delta_{q,-1}-\delta_{q,1})/\sqrt{2} \ ,\\ \epsilon_q^{(y)}&=-i(\delta_{q,-1}+\delta_{q,1})/\sqrt{2} \ .\end{aligned} \tag{A2a}$$

Linearly polarized at an angle θ_x relative to the x axis:

$$\epsilon_q^{(\theta_x)}=(e^{-i\theta_x}\delta_{q,-1}-e^{i\theta_x}\delta_{q,1})/\sqrt{2} \ . \tag{A2b}$$

Circularly polarized, σ^+ or σ^-:

$$\epsilon_q^{+}=\delta_{q,-1}, \quad \epsilon_q^{+}=\delta_{q,1} \ . \tag{A2c}$$

TABLE I. Values for $T_{GG}(G,H;K,K',\bar{K})$, $T_{GH}(G,H;K,K',\bar{K})$, and $R(G,H;K,K',\bar{K})$.

K	K'	$\bar{K}$	$T_{GG}(\frac{1}{2},\frac{1}{2};K,K',\bar{K})$	$T_{GH}(\frac{1}{2},\frac{1}{2};K,K',\bar{K})$	$R(\frac{1}{2},\frac{1}{2};K,K',\bar{K})$
0	0	0	$\sqrt{3}/2$	$-\sqrt{3}/2$	$-\sqrt{3}/2$
0	1	1	$-\sqrt{3/2}$	$-\sqrt{3/2}$	$\sqrt{3/2}$
1	0	1	$-\sqrt{3/2}$	$\sqrt{3/2}$	$1/\sqrt{6}$
1	1	0	$-3/2$	$-\frac{1}{2}$	$1/6$
1	1	1	$-\sqrt{3}$	0	0
1	1	2	0	$\sqrt{5}$	$-\sqrt{5}/3$

K	K'	$\bar{K}$	$T_{GG}(\frac{1}{2},\frac{3}{2};K,K',\bar{K})$	$T_{GH}(\frac{1}{2},\frac{3}{2};K,K',\bar{K})$	$R(\frac{1}{2},\frac{3}{2};K,K',\bar{K})$
0	0	0	$\sqrt{3}/2$	$-\sqrt{3/8}$	$-\sqrt{3}/2$
0	1	1	$\sqrt{3/8}$	$-\sqrt{15}/4$	$-\sqrt{3/8}$
1	0	1	$\sqrt{3/8}$	$-\sqrt{3}/4$	$-5/\sqrt{24}$
1	1	0	$-3/2$	$\sqrt{5/8}$	$5/6$
1	1	1	$\sqrt{3}/2$	0	0
1	1	2	0	$-1/\sqrt{8}$	$-\sqrt{5}/6$

K	K'	$\bar{K}$	$T_{GG}(1,2;K,K',\bar{K})$	$T_{GH}(1,2;K,K',\bar{K})$	$R(1,2;K,K',\bar{K})$
0	0	0	$1/\sqrt{3}$	$-1/\sqrt{5}$	$-1/\sqrt{3}$
0	1	1	$\frac{1}{2}$	$-3/(2\sqrt{5})$	$-\frac{1}{2}$
0	2	2	$1/(2\sqrt{15})$	$-\sqrt{7}/(2\sqrt{5})$	$-1/(2\sqrt{15})$
1	0	1	$\frac{1}{2}$	$-\sqrt{3}/(2\sqrt{5})$	$-\frac{3}{4}$
1	1	0	-1	$3/(2\sqrt{5})$	$\frac{3}{4}$
1	1	1	$\sqrt{3}/4$	0	0
1	1	2	$1/(4\sqrt{5})$	$-\frac{3}{10}$	$-3/(4\sqrt{5})$
1	2	1	$-\sqrt{5}/4$	$\sqrt{21}/(2\sqrt{5})$	$3/(4\sqrt{5})$
1	2	2	$\sqrt{3}/(4\sqrt{5})$	0	0
1	2	3	0	0	0
2	0	2	$1/(2\sqrt{15})$	$-\frac{1}{10}$	$-7/(4\sqrt{15})$
2	1	1	$-\sqrt{5}/4$	$\frac{3}{10}$	$7/(4\sqrt{5})$
2	1	2	$\sqrt{3}/(4\sqrt{5})$	0	0
2	1	3	0	0	0
2	2	0	$\sqrt{5/3}$	$-\sqrt{7}/(2\sqrt{5})$	$-7/(4\sqrt{15})$
2	2	1	$-\sqrt{15}/4$	0	0
2	2	2	$\sqrt{7}/(4\sqrt{15})$	$1/(2\sqrt{5})$	$\sqrt{7}/(4\sqrt{15})$
2	2	3	0	0	0
2	2	4	0	0	0

It then follows from Eq. (15) that the $\epsilon_Q^K(j,j')$ are given by

$$\epsilon_Q^K(z,z)=[-(1/\sqrt{3})\delta_{K,0}+\sqrt{2/3}\delta_{K,2}]\delta_{Q,0}\ , \tag{A3a}$$

$$\epsilon_Q^K(x,x)=-(1/\sqrt{3})\delta_{K,0}\delta_{Q,0}+\delta_{K,2}[-(1/\sqrt{6})\delta_{Q,0}+\tfrac{1}{2}(\delta_{Q,2}+\delta_{Q,-2})]\ , \tag{A3b}$$

$$\epsilon_Q^K(y,y)=-(1/\sqrt{3})\delta_{K,0}\delta_{Q,0}-\delta_{K,2}[(1/\sqrt{6})\delta_{Q,0}+\tfrac{1}{2}(\delta_{Q,2}+\delta_{Q,-2})]\ , \tag{A3c}$$

$$\epsilon_Q^K(\theta_x,\theta_x)=-(1/\sqrt{3})\delta_{K,0}\delta_{Q,0}+\delta_{K,2}[-(1/\sqrt{6})\delta_{Q,0}+\tfrac{1}{2}(e^{2i\theta_x}\delta_{Q,2}+e^{-2i\theta_x}\delta_{Q,-2})]\ , \tag{A3d}$$

$$\epsilon_Q^K(x,y)=(i/\sqrt{2})[\delta_{K,1}\delta_{Q,0}+(i/2)\delta_{K,2}(\delta_{Q,2}-\delta_{Q,-2})]\ , \tag{A3e}$$

$$\epsilon_Q^K(x,z)=(1/2)[\delta_{K,1}(\delta_{Q,1}+\delta_{Q,-1})-\delta_{K,2}(\delta_{Q,1}-\delta_{Q,-1})]\ , \tag{A3f}$$

$$\epsilon_Q^K(y,z)=-(i/2)[\delta_{K,1}(\delta_{Q,1}-\delta_{Q,-1})+\delta_{K,2}(\delta_{Q,1}+\delta_{Q,-1})]\ , \tag{A3g}$$

$$\epsilon_Q^K(-\theta_x/2,\theta_x/2)=-\delta_{Q,0}[(1/\sqrt{3})\cos(\theta_x)\delta_{K,0}-(i/\sqrt{2})\sin(\theta_x)\delta_{K,1}+(1/\sqrt{6})\cos(\theta_x)\delta_{K,2}]+\tfrac{1}{2}\delta_{K,2}(\delta_{Q,2}+\delta_{Q,-2})\ , \tag{A3h}$$

$$\epsilon_Q^K(+,+)=-[(1/\sqrt{3})\delta_{K,0}-(1/\sqrt{2})\delta_{K,1}+(1/\sqrt{6})\delta_{K,2}]\delta_{Q,0}\ , \tag{A3i}$$

$$\epsilon_Q^K(-,-)=-[(1/\sqrt{3})\delta_{K,0}+(1/\sqrt{2})\delta_{K,1}+(1/\sqrt{6})\delta_{K,2}]\delta_{Q,0}\ , \tag{A3j}$$

$$\epsilon_Q^K(+,-)=-\delta_{K,2}\delta_{Q,-2}\ , \tag{A3k}$$

$$\epsilon_Q^K(+,x)=(1/\sqrt{2})\delta_{Q,0}[(1/\sqrt{3})\delta_{K,0}-(1/\sqrt{2})\delta_{K,1}+(1/\sqrt{6})\delta_{K,2}]-(1/\sqrt{2})(\delta_{K,2}\delta_{Q,-2})\ , \tag{A3l}$$

$$\epsilon_Q^K(+,y)=-(i/\sqrt{2})\delta_{Q,0}[(1/\sqrt{3})\delta_{K,0}-(1/\sqrt{2})\delta_{K,1}+(1/\sqrt{6})\delta_{K,2}]-(i/\sqrt{2})(\delta_{K,2}\delta_{Q,-2})\ . \tag{A3m}$$

APPENDIX B: VALUES FOR $T_{GG}(G,H;K,K',\bar{K})$, $T_{GH}(G,H;K,K',\bar{K})$, and $R(G,H;K,K',\bar{K})$

Values for $T_{GG}(G,H;K,K',\bar{K})$, $T_{GH}(G,H;K,K',\bar{K})$, and $R(G,H;K,K',\bar{K})$ are given in Table I for ($G=\frac{1}{2}$, $H=\frac{1}{2}$), ($G=\frac{1}{2}$, $H=\frac{3}{2}$), and ($G=1$, $H=2$). Note that the quantity $P(G,H;K,K',\bar{K})$ needed in Eq. (27) is equal to $T_{GG}(G,H;K,K',\bar{K})+R(G,H;K,K',\bar{K})$.

[1] See, for example, L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975), Chap. 6.

[2] See, for example, M. Dumont and B. Decomps, J. Phys. (Paris) **29**, 181 (1968); M. Dumont, *ibid.* **33**, 971 (1972); A. Omont, *Progress in Quantum Electronics* (Pergamon, Oxford, 1977), Vol. 5, pp. 69–138; C. Feuillade and P. R. Berman, Phys. Rev. A **29**, 1236 (1984); M. Ducloy and D. Bloch, *ibid.* **30**, 3107 (1984); K. Blum, *Density Matrix Theory and Applications* (Plenum, New York, 1981); B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley-Interscience, New York, 1990), Vol. 2, Secs. 21.9 and 21.10.

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[4] The basic equations for density-matrix elements were written in the "normal" representation for $\rho_Q^K(G,G')$ and $\rho_Q^K(H,H')$ in Ref. [3]. In adiabatically eliminating the electronic state coherence, it is important to write the equations in an interaction representation, since density-matrix elements $\rho_Q^K(G,G')$ for $G\neq G'$ and $\rho_Q^K(H,H')$ for $H\neq H'$ may not be slowly varying with respect to $\tilde{\rho}_Q^K(G,H)$. As a consequence, the frequency denominators in Eqs. (A23) and (A26) of Ref. [3] should be modified. The correct expressions are given by Eqs. (20)–(24) of this paper.

[5] See A. Messiah, *Quantum Mechanics* (Wiley, New York, 1960), Vol. II, Appendix C. In particular, the Wigner-Eckart theorem is taken as $\langle F,m|T_q^k|F',m'\rangle=(2F+1)^{-1/2}\langle F\|T^k\|F'\rangle\langle F',m';k,q|F,m\rangle$.

[6] Note that $\Delta_{GH}^{(j)}\neq-\Delta_{HG}^{(j)}$ and $\epsilon_Q^{\bar{K}}(j',j)\neq(-1)^{\bar{K}}\epsilon_Q^{\bar{K}}(j,j')$. Under these *replacements,* however, the two sets of equations are seen to be equal.

[7] See, for example, J. Dalibard and C. Cohen-Tannoudji, J. Opt. Soc. Am. B **6**, 2023 (1989); P. J. Ungar, D. S. Weiss, E. Riis, and S. Chu, *ibid.* **6**, 2058 (1989); D. Sheehy, S-Q. Shang, P. van der Staten, S. Hatamian, and H. Metcalf, Phys. Rev. Lett. **64**, 858 (1990). The preceding papers generally refer to level schemes involving specific values for angular momenta. Somewhat more general treatments, applicable to a range of angular momentum values, may be found in Ref. [3] and G. Neinhuis, P. van der Straten, and S.-Q. Shang, Phys. Rev. A **44**, 462 (1991); J. Javanainen, *ibid.* **44**, 5857 (1991); A. M. Steane, G. Hillenbrand, and C. J. Foot, J. Phys. B **25**, 4721 (1992).

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