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$$\text{InertiaTensor} := \sum_{k=1}^n m_k \left(\|\vec{r}_k\|^2 \mathbf{I} - \vec{r}_k \vec{r}_k^T \right)$$

$$\Gamma_{2,2}^1 = -\frac{(1 + 2\epsilon)}{\partial r}$$

Analytical solution for the change of Raman transitions quantum statistics

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By using the Laplace transform method we give exact analytical solutions for the time development of the diagonal and off-diagonal elements of the photon density matrix in Raman transitions.

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In recent publications¹⁻³ the change of photon statistics in multiphoton absorption has been considered. Two different approaches have been thus proposed to obtain an exact solution of the corresponding master equation. Zubairy and Yeh¹ and Zubairy² used a density matrix procedure while Voigt, Bandilla, and Ritze³ used a Laplace transform method.

The photon statistics of the stimulated and hyper Raman effects was dealt with early by Simaan.⁴ The purpose of this paper is to give an exact analytical solution for the time development of the diagonal and off-diagonal elements of the radiation density matrix for Raman transitions (simultaneous absorption and emission) between localized states. Here, one photon is absorbed while the other is emitted. The case for antistokes luminescence (i.e., two photons being absorbed and one emitted) will be studied in a following paper.

The interaction Hamiltonian for Raman transitions is⁵

$$H_I = \sum_i [d_R c_{2i}^+ c_{1i} E_k^-(r_i) E_q^+(r_i) + \text{adj}], \quad (1)$$

where c_{2i}^+ and c_{1i} are creation and annihilation operators for the i th atom in states 2 and 1, respectively, d_R is the electric dipole matrix element for Raman transition, and $E_k^-(r_i)$ and $E_q^+(r_i)$ are the negative- and positive-frequency parts of, respectively, the input and output fields

$$E_k^-(r_i) = -i(2\pi\hbar\omega_k)^{1/2} u_k(r_i) a_k, \\ E_q^+(r_i) = i(2\pi\hbar\omega_q)^{1/2} u_q^*(r_i) a_q^+.$$

Here, ω is frequency, and a_k and a_q^+ are the creation and annihilation operators for the k th and q th modes, respectively. The spatial functions $u_k(r_i)$ and $u_q(r_i)$ are normalized functions for the respective mode in the i th atom.

In the single mode case, the time evolution of the photon density matrix becomes⁵

$$\dot{\rho}_{ph} = -\beta_R [(a_k^+ a_q a_k a_q^+ \rho_{ph} - 2a_k a_q^+ \rho_{ph} a_k^+ a_q + \rho_{ph} a_k^+ a_q a_k a_q^+) p_{1A}^0 + (a_k a_q^+ a_k^+ a_q \rho_{ph} - 2a_k^+ a_q \rho_{ph} a_k a_q^+ + \rho_{ph} a_k a_q^+ a_k^+ a_q) p_{2A}^0], \quad (2)$$

where p_{1A}^0 and p_{2A}^0 are the thermal populations for the ground and first-excited states and the remaining symbols have their conventional meaning.⁵

The diagonal matrix elements of the photon density operator in the Fock representation is immediately obtained from (2):

$$\dot{P}_{l,m}(T) = -P_{l,m}(T) [l(m+1)p_{1A}^0 + m(l+1)p_{2A}^0] + P_{l+1,m-1}(T)m(l+1)p_{1A}^0 + P_{l-1,m+1}(T)l(m+1)p_{2A}^0, \quad (3)$$

where $T = 2\beta_R t$ and $P_{l,m}(T) = \langle l, m | \rho_{ph} | l, m \rangle$ is the probability to simultaneously find l photons in the input mode k and m photons in the Stokes output mode q . Then, according to the method given in Ref. 3, we obtain

$$\mathcal{L}_{l,m}(s) = \int_0^\infty P_{l,m}(T) \exp(-sT) dT = \frac{p_{1A}^0 m(l+1) \mathcal{L}_{l+1,m-1}(s) + p_{2A}^0 l(m+1) \mathcal{L}_{l-1,m+1}(s) + P_{l,m}(0)}{s + p_{1A}^0 l(m+1) + p_{2A}^0 m(l+1)}. \quad (4)$$

If we choose as initial conditions

$$\sum_{l=0}^\infty \sum_{m=0}^\infty P_{l,m}(0) = 1,$$

and $P_{r,z}(0) = 0$ for either $r > l$, $z > m$, or both (where $m > 0$ only for the case in which $P_{2A}^0 \neq 0$, so that there may be spontaneous emission of q th mode photons), we find

$$\mathcal{L}_{n,t}(s) = \frac{P_{n,t}(0)}{s + p_{1A}^0 n(t+1) + p_{2A}^0 t(n+1)} \quad (5)$$

so that, for single Raman transitions

$$\left[\frac{p_{1A}^* n(t+1) P_{n,t}(0)}{s + p_{1A}^* n(t+1) + p_{2A}^* t(t+1)} + \frac{p_{2A}^* (n-1)(t+2) P_{n-2,t+2}(0)}{s + p_{1A}^* (n-2)(t+3) + p_{2A}^* (n-1)(t+2)} + P_{n-1,t+1}(0) \right] \times [s + p_{1A}^* (n-1)(t+2) + p_{2A}^* n(t+1)]^{-1} = \mathcal{L}_{n-1,t+1}(s), \quad (6)$$

which can be eventually generalized into

$$\begin{aligned} \mathcal{L}_{n-j,t+k}(s) = & \sum_{r=0}^j \sum_{v=0}^k P_{n-j+r,t+k-v}(0) \times \prod_{a=0}^r \prod_{b=0}^v \frac{p_{1A}^* (n-j+a)(t+k+1-b)}{p_{1A}^* (n-j)(t+k+1) + p_{2A}^* (t+k)(n-j+1)} \\ & \times [s + p_{1A}^* (n-j+a)(t+k+1-b) + p_{2A}^* (n-j+1+a)(t+k-b)]^{-1} \\ & + \sum_{r=-j}^0 \sum_{v=-k}^0 P_{n-j+r,t+k-v}(0) \prod_{a=-r}^0 \prod_{b=-v}^0 \frac{p_{2A}^* (n-j+a+1)(t+k-b)}{p_{1A}^* (n-j)(t+k+1) + p_{2A}^* (t+k)(n-j+1)} \\ & \times [s + p_{1A}^* (n-j+a)(t+k+1-b) + p_{2A}^* (n-j+1+a)(t+k-b)]^{-1}, \end{aligned} \quad (7)$$

where it has been considered the most general case in which the number of emitted Stokes photons may not be the same as the number of absorbed input photons due to a variety of possible phenomena such as self-induced stimulated emission, nonradiative relaxation, etc. If all of these additional effects are disregarded, then we should make $k=j$, $r=v$, and $a=b$, obtaining the pure Raman transition equation.

Now, following the method of Ref. 3, we transform Eq. (7) back and obtain,⁶ for the most general case,

$$\begin{aligned} P_{n-j,t+k}(T) = & \sum_{r=0}^j \sum_{v=0}^k \prod_{a=0}^r \prod_{b=0}^v p_{1A}^* (n-j+a)(t+k+1-b) P_{n-j+r,t+k-v}(0) \\ & \times \sum_{\alpha=0}^r \sum_{\beta=0}^v [p_{1A}^* (n-j)(t+k-1) + p_{2A}^* (t+k)(n-j+1)]^{-1} \\ & \times \exp\{-[p_{1A}^* (n-j+\alpha)(t+k+1-\beta) + p_{2A}^* (n-j+\alpha+1)(t+k-\beta)]T\} \\ & \times \left\{ \prod_{\substack{v=0 \\ v \neq \alpha}}^r \prod_{\substack{\omega=0 \\ \omega \neq \beta}}^v \left[p_{1A}^* (n-j+v)(t+k+1-\omega) \right. \right. \\ & \left. \left. + p_{2A}^* (n-j+v+1)(t+k-\omega) - p_{1A}^* (n-j+\alpha)(t+k+1-\beta) - p_{2A}^* (n-j+1+\alpha)(t+k-\beta) \right] \right\}^{-1} \\ & + \sum_{r=-j}^0 \sum_{v=-k}^0 \prod_{a=-r}^0 \prod_{b=-v}^0 p_{2A}^* (n-j+a+1)(t+k-b) P_{n-j+r,t+k-v}(0) \\ & \times \sum_{\alpha=-r}^0 \sum_{\beta=-v}^0 [p_{1A}^* (n-j)(t+k-1) + p_{2A}^* (t+k)(n-j+1)]^{-1} \\ & \times \exp\{-[p_{1A}^* (n-j+\alpha)(t+k+1-\beta) + p_{2A}^* (n-j+\alpha+1)(t+k-\beta)]T\} \\ & \times \left\{ \prod_{\substack{v=-r \\ v \neq \alpha}}^0 \prod_{\substack{\omega=-v \\ \omega \neq \beta}}^0 \left[p_{1A}^* (n-j+v)(t+k+1-\omega) + p_{2A}^* (n-j+v+1)(t+k-\omega) \right. \right. \\ & \left. \left. - p_{1A}^* (n-j+\alpha)(t+k+1-\beta) - p_{2A}^* (n-j+1+\alpha)(t+k-\beta) \right] \right\}^{-1}, \end{aligned} \quad (8)$$

where we also have $j=k$, $r=v$, $a=b$, $\alpha=\beta$, and $v=\omega$ for pure Raman transitions.

Substituting first $n-j=n'$, $t+k=t'$, making then $n \rightarrow \infty$, $t \rightarrow \infty$, and omitting finally the primes, one gets

$$\begin{aligned} P_{n,t}(T) = & \sum_{r=0}^{\infty} \sum_{v=-\infty}^0 P_{n+r,t-v}(0) \sum_{\alpha=0}^r \sum_{\beta=v}^0 p_{1A}^* (n+\alpha)(t-\beta+1) \frac{V(n,t,\alpha,\beta,T)}{\prod_{\substack{v=0 \\ v \neq \alpha}}^r \prod_{\substack{\omega=v \\ \omega \neq \beta}}^0 W(n,t,\alpha,\beta,v,\omega)} \\ & + \sum_{r=-\infty}^0 \sum_{v=0}^{\infty} P_{n+r,t-v}(0) \sum_{\alpha=r}^0 \sum_{\beta=0}^v p_{2A}^* (n+\alpha+1)(t-\beta) \frac{V(n,t,\alpha,\beta,T)}{\prod_{\substack{v=0 \\ v \neq \alpha}}^0 \prod_{\substack{\omega=v \\ \omega \neq \beta}}^v W(n,t,\alpha,\beta,v,\omega)}, \\ V(n,t,\alpha,\beta,T) = & \frac{\exp\{-[p_{1A}^* (n+\alpha)(t-\beta+1) + p_{2A}^* (n+\alpha+1)(t-\beta)]T\}}{p_{1A}^* n(t+1) + p_{2A}^* t(t+1)}, \\ W(n,t,\alpha,\beta,v,\omega) = & p_{1A}^* [(n+v)(t+1-\omega) - (n+\alpha)(t+1-\omega)] + p_{2A}^* [(n+v+1)(t-\omega) - (n+\alpha+1)(t-\beta)]. \end{aligned} \quad (9)$$

For calculating the off-diagonal density matrix elements in the Fock representation, we operate from the master equation (2) and obtain

$$\begin{aligned}
\dot{P}_{l,m;l',m'}(T) &= \langle l,m|\dot{\rho}_{ph}|l',m'\rangle \\
&= -P_{l,m;l',m'}(T)\{\dot{p}_{1A}[l(m+1)+l'(m'+1)]+\dot{p}_{2A}[m(l+1)+m'(l'+1)]\} \\
&\quad +P_{l+1,m-1;l'+1,m'-1}(T)[\dot{p}_{1A}m(l+1)+\dot{p}_{2A}l'(m'+1)] \\
&\quad +P_{l-1,m+1;l'-1,m'+1}(T)[\dot{p}_{1A}m'(l'+1)+\dot{p}_{2A}l(m+1)]
\end{aligned} \tag{10}$$

and

$$\begin{aligned}
\mathcal{L}_{l,m;l',m'}(s) &= [\dot{p}_{1A}m(l+1)+\dot{p}_{2A}l'(m'+1)]\mathcal{L}_{l+1,m-1;l'+1,m'-1}(s) \\
&\quad +[\dot{p}_{1A}m'(l'+1)+\dot{p}_{2A}l(m+1)]\mathcal{L}_{l-1,m+1;l'-1,m'+1}(s)+P_{l,m;l',m'}(0) \\
&\quad \times \{s+\dot{p}_{1A}[l(m+1)+l'(m'+1)]+\dot{p}_{2A}[m(l+1)+m'(l'+1)]\}^{-1}
\end{aligned} \tag{11}$$

Imposing now initial conditions similar to those for the diagonal matrix elements, we get

$$\begin{aligned}
\mathcal{L}_{n-j,t+k;n'-j',t'+k'}(s) &= \sum_{r=0}^j \sum_{v=0}^k \sum_{r'=0}^{j'} \sum_{v'=0}^{k'} P_{n-j+r,t+k-v;n'-j'+r',t'+k'-v'}(0) \\
&\times \prod_{a=0}^r \prod_{b=0}^v \prod_{a'=0}^{r'} \prod_{b'=0}^{v'} [\dot{p}_{1A}(n-j+a)(t+k+1-b)+\dot{p}_{2A}(n'-j'+1+a')(t'+k'-b')] R(a,b;a',b')^{-1} \\
&+ \sum_{r=-j}^0 \sum_{v=-k}^0 \sum_{r'=-j'}^0 \sum_{v'=-k'}^0 P_{n-j+r,t+k-v;n'-j'+r',t'+k'-v'}(0) \\
&\times \prod_{a=-r}^0 \prod_{b=-v}^0 \prod_{a'=-r'}^0 \prod_{b'=-v'}^0 [\dot{p}_{1A}(n'-j'+a')(t'+k'+1-b')+ \dot{p}_{2A}(n-j+1+a)(t+k-b)] R(a,b;a',b')^{-1},
\end{aligned} \tag{12}$$

where

$$\begin{aligned}
R(a,b;a',b') &= \dot{p}_{1A}[(n-j)(t+k-1)+(n'-j')(t'+k'-1)]+\dot{p}_{2A}[(n-j+1)(t+k)+(n'-j'+1)(t'+k')] \\
&\times \{s+\dot{p}_{1A}[(n-j+a)(t+k+1-b)+(n'-j'+a')(t'+k'+1-b')] \\
&\quad +\dot{p}_{2A}[(n-j+1+a)(t+k-b)+(n'-j'+1+a')(t'+k'-b')]\}^{-1}.
\end{aligned}$$

Following the same procedure as for the diagonal matrix elements, one finally obtains

$$\begin{aligned}
P_{n,t,n',t'}(T) &= \sum_{r=0}^{\infty} \sum_{v=-\infty}^0 \sum_{r'=0}^{\infty} \sum_{v'=-\infty}^0 P_{n+r,t-v;n'+r',t'-v'}(0) \\
&\times \sum_{\alpha=0}^r \sum_{\beta=v}^0 \sum_{\alpha'=0}^{r'} \sum_{\beta'=v'}^0 \frac{[\dot{p}_{1A}(n+\alpha)(t+1-\beta)+\dot{p}_{2A}(n'+1+\alpha')(t'-\beta')]\exp[-B(\alpha,\beta;\alpha',\beta')T]}{A(n,t;n',t')\prod_{\substack{v\neq\alpha \\ \omega\neq\beta}}^r \prod_{\substack{v'=0 \\ \omega'\neq\beta'}}^0 \prod_{\substack{v'\neq\alpha' \\ \omega'\neq\beta'}}^0 C(\alpha,\beta,v,\omega;\alpha',\beta',v',\omega')} \\
&+ \sum_{r=-\infty}^0 \sum_{v=0}^{\infty} \sum_{r'=-\infty}^0 \sum_{v'=0}^{\infty} P_{n+r,t-v;n'+r',t'-v'}(0) \\
&\times \sum_{\alpha=r}^0 \sum_{\beta=0}^v \sum_{\alpha'=r'}^0 \sum_{\beta'=0}^{v'} \frac{[\dot{p}_{1A}(n'+\alpha')(t'+1-\beta')+ \dot{p}_{2A}(n+1+\alpha)(t-\beta)]\exp[-B(\alpha,\beta;\alpha',\beta')T]}{A(n,t;n',t')\prod_{\substack{v\neq\alpha \\ \omega\neq\beta}}^0 \prod_{\substack{v'=0 \\ \omega'\neq\beta'}}^v \prod_{\substack{v'\neq\alpha' \\ \omega'\neq\beta'}}^0 C(\alpha,\beta,v,\omega;\alpha',\beta',v',\omega')},
\end{aligned} \tag{13}$$

where

$$\begin{aligned}
A(n,t;n',t') &= \dot{p}_{1A}[n(t+1)+n'(t'+1)]+\dot{p}_{2A}[t(n+1)+t'(n'+1)], \\
B(\alpha,\beta;\alpha',\beta') &= \dot{p}_{1A}[(n+\alpha)(t+1-\beta)+(n'+\alpha')(t'+1-\beta')]+\dot{p}_{2A}[(n+1+\alpha)(t-\beta)+(n'+1+\alpha')(t'-\beta')], \\
C(\alpha,\beta,v,\omega;\alpha',\beta',v',\omega') &= B(v,\omega;v',\omega')-B(\alpha,\beta;\alpha',\beta').
\end{aligned}$$

Expressions (9) and (13) are our most general analytical solutions. The pure Raman transition case may be straightforwardly obtained by making in these expressions $r=v$, $a=b$, $\alpha=\beta$, $v=\omega$, and likewise for the primed indices in the off-diagonal case. Spontaneous emission of photons could be omitted if it is assumed that all the two-level systems are maintained in their ground states by some external influence, so that $P_{2A}^* = 0$. The well-known⁷ dependence of the yield of the Raman process on the statistical properties of the input field may be shown from Eqs. (9) and (13).

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