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Time-dependent spectrum in quantum theory of stimulated Raman scattering from a two-level system†

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Abstract. The time-dependent (physical) spectrum of stimulated Raman scattering (SRS) from a two-level system is calculated, proving the influence of the spectral width Γ of the detector on the obtained spectral line shape. In addition, the time evolution of the process leading to the Raman line is investigated. For a partially excited system, the scattered field intensity exhibits oscillations in time, and Γ is found to affect the amplitude of the oscillations in the Raman line intensity. A numerical analysis of the result is performed for the case of monochromatic external fields.

1. Introduction

Recently, numerous papers have been devoted to the time-dependent (physical) spectrum (TDS). Its definition [1, 2], established in 1977, takes into account the influence of the measuring device (the detector) on the spectral picture of the physical processes involved; it also eliminates various drawbacks inherent in earlier definitions, and permits the study of the time-evolution of the phenomena. Time-dependent spectra have already been applied in studies of resonant Raman scattering [1, 3], resonant fluorescence [4, 5], and super-fluorescence [6]. In [7], TDS has been applied to a new interpretation of the probability distribution function in phase space in quantum mechanics, whereas [8] provides the theoretical basis for time-dependent correlational spectroscopy.

Here, making use of the definition of TDS, we carry out an analysis of the spectral properties of the process of stimulated Raman scattering (SRS) on atomic two-level systems. Next, with recourse to the time-dependent properties of the spectrum, we study the process of the creation of the spectral line from the moment the perturbation is switched on ($t=0$) until the steady state sets in. New effects are found that are related to oscillations in intensity of the SRS spectral line on the radiatively shifted atomic level. In fact, the spectral width of the detector not only affects the measured spectral line-shape but also has an influence on the observed dynamics of its creation.

Our analysis of the problem is quantum mechanical throughout. The SRS process involves two strong external fields: a laser field (ω_L) and a Stokes field (ω_{k_0}).

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They determine the evolution of the two-level system (referred to henceforth as the 'atomic' or 'scattering' system). The evolution is described by the master equation, derived for the atomic quantities on averaging with respect to the strong external fields ω_L and ω_{k_0} (See § 2). The two fields concurrently cause a power shift and broadening, leading to the emergence of a field due to Raman scattering on the shifted levels of the system (figure 1).

The strong coupling between the external fields and the atomic system enable us to deal with the scattered field as a perturbation which has negligible effect on the scattering system. Thus, the correlation functions derived from the Maxwell equations can be appropriately de-correlated, leading to the spectrum of the SRS process (§ 3). Its numerical analysis is given in § 4.

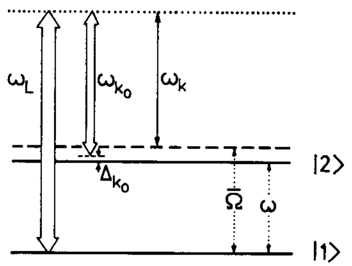

Figure 1. Raman scattering (SRS) from two-level atomic system with radiation-shifted transitions: Δ_{k_0}, detuning of the frequency ω_{k_0} from two-photon resonance; $\bar{\Omega}$, renormalized energy of the transition. The shift of the ground level is not shown.

2. The evolution of the scattering system

We consider a system of N non-interacting two-level atoms. The process of Raman scattering is described by the effective two-photon process Hamiltonian [9–13]:

$$\hat{H}=\hbar\omega\hat{S}^z+\hbar\sum_k\omega_k\hat{a}_k^+\hat{a}_k+\hbar\omega_L\hat{a}_L^+\hat{a}_L+\hbar\sum_k(g_k\hat{a}_L\hat{a}_k^+\hat{S}^+ +\text{h.c.}), \tag{1}$$

with $\hat{S}^z$, $\hat{S}^\pm$ the collective spin operators of the atomic system; and $\hat{a}_L(\hat{a}_L^+)$, $\hat{a}_k(\hat{a}_k^+)$ the photon annihilation (creation) operators for the laser and Stokes field, respectively. The coupling constant g_k, involving the Raman polarizability M_{12}, is of the form

$$g_k=\frac{2\pi c}{V}(k_L k)^{1/2}M_{12}\exp[\mathrm{i}(\mathbf{k}_L-\mathbf{k})\cdot\mathbf{r}]. \tag{2}$$

A similar, completely boson Hamiltonian has been applied to the description of the statistical properties of the Raman process on phonons [14–17].

In equations (1) and (2) we have assumed all the fields to be polarized linearly in the same direction, and that the scattering system of N atoms as isotropic. Thus, we shall be dealing with the field quantities as scalars. Anti-Stokes effects will not be considered.

The operators characterizing the scattering system are to be obtained from the master equation for the reduced density operator. The method of applying a projection operator to the atomic system [18, 19] involves the introduction of an operator for the form $\hat{P}(\ldots)=\hat{\rho}_R(0)\,\mathrm{Tr}_R(\ldots)$ and averaging the states of the whole system over the initial values of the electromagnetic fields. When this is applied to the Hamiltonian (1), it leads to the following equation of motion for the mean value of an operator $\hat{A}$ which describes the atomic system:

$$\begin{aligned}\frac{\partial\langle\hat{A}\rangle}{\partial t}= & -\gamma_{k_0}\bar{n}_L(\langle[[\hat{A},\hat{S}^+],\hat{S}^-]\rangle+\langle[[\hat{A},\hat{S}^-],\hat{S}^+]\rangle)\\ & -\gamma_{k_0}(\langle[\hat{A},\hat{S}^+]\hat{S}^-\rangle+\langle\hat{S}^+[\hat{S}^-,\hat{A}]\rangle)\\ & -\gamma_L(\langle[\hat{A},\hat{S}^-]\hat{S}^+\rangle+\langle\hat{S}^-[\hat{S}^+,\hat{A}]\rangle)\\ & +\mathrm{i}(2\Omega_{k_0}\bar{n}_L+\Omega_{k_0}+\Omega_L+\omega)\langle[\hat{S}^z,\hat{A}]\rangle,\end{aligned} \tag{3}$$

where

$$\gamma_{k_0(L)}=\pi\sum_{k_0}|g_{k_0}|^2\bar{n}_{k_0(L)}\delta(\Delta_{k_0}), \tag{4}$$

$$\Omega_{k_0(L)}=\sum_{k_0}|g_{k_0}|^2\bar{n}_{k_0(L)}\frac{1}{\Delta_{k_0}}, \tag{5}$$

and

$$\Delta_{k_0}=\omega_L-\omega_{k_0}-\omega, \tag{6}$$

$$\bar{n}_{k_0(L)}=\mathrm{Tr}_R\,(\hat{\rho}_R(0)\hat{a}^+_{k_0(L)}\hat{a}_{k_0(L)}). \tag{7}$$

Equation (3) has been derived with the assumption that a non-zero external laser field ω_L and a Stokes field ω_{k_0} are present at the initial time $t=0$. This equation, in fact, can be seen to be correct if we make the following assumption: the band-widths of the external fields are smaller than the energy separation of the two atomic levels. This equation describes the evolution of the atomic system and gives the radiation corrections to the spectrum. The quantities (4) and (5) represent, respectively, the radiation broadening and the shift of the spectral line due to the individual fields. Here, other effects that might lead to a broadening of the line (namely, atomic collisions and Doppler broadening) are neglected, i.e. we consider field-stimulated transitions only. The assumed model describes effects related to the presence, in the atom, of a metastable excited level with a lifetime much longer than the times characterizing the field-stimulated transitions.

Equation (3) provides an expression of the one-time atomic correlation functions. By quantum regression theory [20], we can obtain multi-time functions from them. In the present case, we obtain the two-time correlation functions:

$$\langle\hat{S}^-(t+\tau)\hat{S}^+(t)\rangle=(\sigma_1\exp(-2\bar{\gamma}t)+\sigma_2)\exp[(-\mathrm{i}\bar{\Omega}-\bar{\gamma})\tau], \tag{8}$$

$$\langle\hat{S}^z(t+\tau)\hat{S}^z(t)\rangle=(\sigma_3\exp(-2\bar{\gamma}t)+\sigma_4)+\sigma_5[1-\exp(-2\bar{\gamma}\tau)]\exp(-2\bar{\gamma}t), \tag{9}$$

$$\langle\hat{S}^z(t+\tau)\hat{S}^+(t)\rangle=(\sigma_6\exp(-2\bar{\gamma}\tau)+\sigma_7)\exp(\mathrm{i}\bar{\Omega}-\bar{\gamma})t, \tag{10}$$

$$\langle\hat{S}^-(t+\tau)\hat{S}^z(t)\rangle=\sigma_8\exp[-(\mathrm{i}\bar{\Omega}+\bar{\gamma})(t+\tau)], \tag{11}$$

with

$$\left.\begin{array}{ll}\sigma_1=\sigma_0-\sigma_\infty, & \sigma_2=\sigma_\infty,\\ \sigma_3=\tfrac{1}{4}-(\sigma_z^\infty)^2, & \sigma_4=(\sigma_z^\infty)^2,\\ \sigma_5=\sigma_z^\infty(\sigma_z-\sigma_z^\infty), & \sigma_6=\langle\hat{S}^z(0)\hat{S}^+(0)\rangle-\sigma_7,\\ \sigma_7=\sigma_z^\infty\langle\hat{S}^+(0)\rangle, & \sigma_8=\langle\hat{S}^-(0)\hat{S}^z(0)\rangle,\\ \sigma_0=\langle\hat{S}^-(0)\hat{S}^+(0)\rangle, & \sigma_z=\langle\hat{S}^z(0)\rangle,\\ \sigma_\infty=\dfrac{\gamma_{k_0}(\bar{n}_L+1)}{\bar{\gamma}}, & \sigma_z^\infty=\dfrac{\gamma_{k_0}\bar{n}_L+\gamma_L}{2\bar{\gamma}}.\end{array}\right\} \tag{12}$$

The expressions

$$\bar{\gamma}=2\gamma_{k_0}n_L+\gamma_{k_0}+\gamma_L, \tag{13}$$

$$\bar{\Omega}=2\Omega_{k_0}\bar{n}_L+\Omega_{k_0}+\Omega_L+\omega, \tag{14}$$

describe, respectively, the total broadening and shift of the spectral line in the Raman process. One notes that the effect of the two fields cumulate and that, moreover, a component appears resulting from superposition of the effects of the two fields. Similar expressions for the radiation corrections have been obtained earlier when dealing with two-photon effects within the Heisenberg equation formalism, both with regard to the Raman process [21] and two-photon absorption [22]. In our approach to Raman scattering, the situation is the following: the scattering system, subjected to the action of the external fields, is modified by radiation corrections and, simultaneously, becomes the source of a scattered field. Since the two-photon scattering process under consideration occurs by way of a virtual level, the picture differs from that of Raman processes of the near-resonance type [23–26]. Moreover, from the way in which we introduce the rotating-wave approximation (RWA) when deriving equation (3) it results that we neglect any shift of the ground level.

3. The spectrum of the Raman process

The equations of motion of SRS corresponding to the Hamiltonian (1) are these:

$$\dot{\hat{a}}_k(t)=-i\omega_k\hat{a}_k(t)-ig_k\hat{a}_L(t)\hat{S}^+(t), \tag{15}$$

$$\dot{\hat{S}}^+(t)=i\omega\hat{S}^+(t)-2i\sum_k g_k^*\hat{a}_L^+(t)\hat{a}_k(t)\hat{S}^z(t), \tag{16}$$

$$\dot{\hat{S}}^z(t)=-i\sum_k(g_k\hat{a}_L(t)\hat{a}_k^+(t)\hat{S}^+(t)-g_k^*\hat{a}_L^+(t)\hat{a}_k(t)\hat{S}^-(t)). \tag{17}$$

The laser field being monochromatic and very strong, we neglect its variations in the course of the scattering process, i.e. we have

$$\hat{a}_L(t)=\hat{a}_L(0)\exp(-i\omega_L t). \tag{18}$$

Differentiation of (15) and the recourse to (16) gives

$$\ddot{\hat{a}}_k(t)=-\omega_k^2\hat{a}_k(t)-g_k\hat{a}_L(t)(\omega_k+\omega_L-\omega)\hat{S}^+(t)$$
$$-2(\bar{n}_L+1)g_k\sum_{k'}g_{k'}^*\hat{a}_{k'}(t)\hat{S}^z(t). \tag{19}$$

Obviously, for the conjugate operators we obtain equations that are Hermitian-conjugate to (15)–(19). One readily notes that (19) is equivalent to the following Maxwell wave equation:

$$\nabla^2 \hat{E}^{(-)}(t) - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\hat{E}^{(-)}(t) = \frac{4\pi}{c}\hat{J}^{(-)}(t), \tag{20}$$

where

$$\hat{J}^{(-)}(t) = -\frac{\mathrm{i}}{4\pi c}\sum_k \left(\frac{2\pi c k\hbar}{V}\right)^{1/2} g_k \exp(\mathrm{i}\mathbf{k}\cdot\mathbf{r}) \times\left[\hat{a}_{\mathrm{L}}(t)(\omega_k+\omega_{\mathrm{L}}-\omega)\hat{S}^+(t) + 2(\bar{n}_{\mathrm{L}}+1)\sum_{k'} g_{k'}^* \hat{a}_{k'}(t)\hat{S}^z(t)\right] \tag{21}$$

is the displacement current operator, and

$$\hat{E}^{(-)}(t) = \mathrm{i}\sum_k \left(\frac{2\pi c k\hbar}{V}\right)^{1/2} \hat{a}_k(t)\exp(\mathrm{i}\mathbf{k}\cdot\mathbf{r}). \tag{22}$$

The sum over k' in equation (21)—as one notes on comparison with (22)—is, in fact, the amplitude of the scattered field. With this in mind and applying the expression for the field induced by the displacement current,

$$\hat{E}_r^{(-)}(t) = \frac{1}{c}\int \frac{\hat{J}^{(-)}(r, t-r/c)}{r}\,\mathrm{d}V, \tag{23}$$

we immediately obtain from equation (21) the following linear operator equation for the scattered field amplitude:

$$\hat{E}^{(-)}(t) = A_1 \hat{E}_{\mathrm{L}}^{(-)}(t)\hat{S}^+(t) + A_2 \hat{E}^{(-)}(t)\hat{S}^z(t). \tag{24}$$

One readily notes that the term $\sim\hat{S}^z$ is a straightforward consequence of the nonlinearity in the effective Hamiltonian (1) (see (16) and (19)). A linear Hamiltonian would lead to a result involving terms $\sim\hat{S}^+$ and $\sim\hat{S}^-$ only [19]. The coefficients A_1 and A_2 are of the form

$$A_1 = -\sum_k \frac{1}{2\pi c r}\left(\frac{k^3}{k_{\mathrm{L}}}\right) g_k(\omega_k+\omega_{\mathrm{L}}-\omega), \tag{25}$$

$$A_2 = -\sum \frac{1}{2\pi c^2 r}|g_k|^2(\bar{n}_{\mathrm{L}}+1), \tag{26}$$

where $r=|\mathbf{r}|$ is the distance between the scattering system and the detector. Since the laser field is monochromatic, its amplitude has the form

$$\hat{E}_{\mathrm{L}}^{(-)}(t) = \mathrm{i}\left(\frac{2\pi c k_{\mathrm{L}}\hbar}{V}\right)^{1/2} \hat{a}_{\mathrm{L}}(t)\exp(\mathrm{i}\mathbf{k}\cdot\mathbf{r}). \tag{27}$$

In (23), integration extends over the volume of the source. As when deriving equation (3), the scattering system at the origin of the coordinates system is assumed point-like, and retarded effects are neglected. The extent of the sample (in comparison with $\lambda_{k(\mathrm{L})}$) rules out any considerations of Stokes beam amplification in the medium. The primary aim of this work is to elucidate the evolution in time of the spectral line in the elementary scattering process. In deriving (24) we also neglected the field resulting from the vacuum solution of the Maxwell equations, since the effects related to the presence of the strong external fields are predominant; in this sense, the phenomenon under consideration has to be dealt with as SRS. The role of spontaneous scattering and its effect on the evolution of SRS has been studied thoroughly in recent years [27].

We now proceed to define the correlation function of the electromagnetic field incident on the detector:

$$\begin{aligned} G(t,\tau) &= \langle \hat{E}^{(+)}_{\text{Det}}(t+\tau)\hat{E}^{(-)}_{\text{Det}}(t)\rangle \\ &= \langle (\hat{E}^{(+)}_{\text{L}}(t+\tau)+\hat{E}^{(+)}_{k_0}(t+\tau)+\hat{E}^{(+)}(t+\tau))(\hat{E}^{(-)}_{\text{L}}(t)+\hat{E}^{(-)}_{k_0}(t)+\hat{E}^{(-)}(t))\rangle. \end{aligned} \tag{28}$$

The correlation function $G(t,\tau)$ comprises a variety of components, describing the intensities of the individual fields as well as their mutual correlation. We find from the Heisenberg equations (15)–(17) that

$$\langle \hat{E}^{(+)}_{k_0}(t+\tau)\hat{E}^{(-)}(t)\rangle = \langle \hat{E}^{(+)}(t+\tau)\hat{E}^{(-)}_{k_0}(t)\rangle = 0, \tag{29}$$

i.e. that there is no correlation between the stimulating and scattered Stokes fields. Next, the assumption of zero correlation between the external fields ω_{L} and ω_{k_0} leads to

$$\langle \hat{E}^{(+)}_{k_0}(t+\tau)\hat{E}^{(-)}_{\text{L}}(t)\rangle = \langle \hat{E}^{(+)}_{\text{L}}(t+\tau)\hat{E}^{(-)}_{k_0}(t)\rangle = 0. \tag{30}$$

Thus, the problem reduces to the calculation of $G(t,\tau)$ in the form

$$\begin{aligned} G(t,\tau) &= \langle \hat{E}^{(+)}_{\text{L}}(t+\tau)\hat{E}^{(-)}_{\text{L}}(t)\rangle + \langle \hat{E}^{(+)}_{k_0}(t+\tau)\hat{E}^{(-)}_{k_0}(t)\rangle \\ &\quad + \langle \hat{E}^{(+)}_{\text{L}}(t+\tau)\hat{E}^{(-)}(t)\rangle + \langle \hat{E}^{(+)}(t+\tau)\hat{E}^{(-)}_{\text{L}}(t)\rangle \\ &\quad + \langle \hat{E}^{(+)}(t+\tau)\hat{E}^{(-)}(t)\rangle. \end{aligned} \tag{31}$$

To achieve this aim, we have to de-correlate the atomic and field-$\hat{E}^{(+)}(t)$ variables. This approximation is justified in our case since, at $\Delta_{k_0} \cong 0$, the fields ω_{L} and ω_{k_0} are strongly coupled to the atomic system by way of the two-photon process. It is this coupling that determines the evolution of the atomic system as described by the master equation (3). The radiation shift is small (of the order of 1 cm^{-1} for a laser field of 100 MW cm^{-2} and a Stokes field of 250 kW cm^{-3} [28]) enough not to destroy this coupling. In this situation the scattering process may be assumed negligibly to perturb the evolution of the atomic system.

On insertion of (24) into (31) we now obtain

$$G(t,\tau) = G_1(t,\tau) + G_2(t,\tau) + G_3(t,\tau), \tag{32}$$

where

$$G_1(t,\tau) = \varepsilon_{\text{L}}\bar{n}_{\text{L}} \exp(\mathrm{i}\omega_{\text{L}}\tau) + \sum_{k_0} \varepsilon_{k_0}\bar{n}_{k_0} \exp(\mathrm{i}\omega_{k_0}\tau), \tag{33}$$

$$G_2(t,\tau) = A_1\varepsilon_{\text{L}}\bar{n}_{\text{L}} \exp(\mathrm{i}\omega_{\text{L}}\tau)\left(\frac{\langle \hat{S}^{+}(t)\rangle}{1-A_2\langle \hat{S}^{z}(t)\rangle} + \frac{\langle \hat{S}^{-}(t+\tau)\rangle}{1-A_2\langle \hat{S}^{z}(t+\tau)\rangle}\right), \tag{34}$$

$$\begin{aligned} G_3(t,\tau) &= \frac{A_1^2\varepsilon_{\text{L}}\bar{n}_{\text{L}}}{1-A_2^2\langle \hat{S}^{z}(t+\tau)\hat{S}^{z}(t)\rangle} \exp(\mathrm{i}\omega_{\text{L}}\tau)\Bigg[\langle \hat{S}^{-}(t+\tau)\hat{S}^{+}(t)\rangle \\ &\quad + \left(\frac{\langle \hat{S}^{-}(t+\tau)\rangle\langle \hat{S}^{z}(t+\tau)\hat{S}^{+}(t)\rangle}{1-A_2\langle \hat{S}^{z}(t+\tau)\rangle} + \frac{\langle \hat{S}^{+}(t)\rangle\langle \hat{S}^{-}(t+\tau)\hat{S}^{z}(t)\rangle}{1-A_2\langle \hat{S}^{z}(t)\rangle}\right)A_2\Bigg] \end{aligned} \tag{35}$$

and

$$\varepsilon_{L(k_0)}=\frac{2\pi ck_{L(0)}\hbar}{V}, \quad \bar{n}_{L(k_0)}=\mathrm{Tr}_R\,(\hat{\rho}_R(0)\hat{a}^+_{L(k_0)}\hat{a}_{L(k_0)}). \tag{36}$$

In (33), summation over k_0 reflects the circumstance in which the external Stokes beam is non-monochromatic. The first component of $G(t,\tau)$ describes the external field intensity (33); the second component (34) accounts for the mutual correlation between the scattered field and laser field; and the third component (35) is the correlation function of the scattered field.

If the beams ω_L and ω_{k_0} are collinear, measurement will bear on the total correlation function $G(t,\tau)$ or on $G_3(t,\tau)$ only, depending on the configuration of the detector (figure 2). According to the configuration of the detector, we shall be observing the spectrum of the process as a whole (configuration D_1), or the spectrum of scattered light only (configuration D_2).

We now have recourse to the definition of the time-dependent spectrum. Assume the detector is set up to measure the electromagnetic field as a function of a frequency D to which it can be tuned. In a detector having intrinsic spectral width Γ we obtain the signal (the measured spectrum) in the following form:

$$S(D,\Gamma,t)=4\Gamma\mathrm{Re}\int_0^t \mathrm{d}t_1\exp(-2\Gamma(t-t_1))\int_0^{t-t_1}\mathrm{d}\tau\, G(t_1,\tau)\exp(\Gamma-\mathrm{i}D)\tau. \tag{37}$$

The above definition takes into account that measurement starts at $t=0$. We now insert the atomic mean values (8)–(11) into (33)–(35) and, next, the correlation function into (37). We thus obtain an analytical expression for TDS in SRS involving non-elementary integrals. Luckily, all the integrands can be expanded as power series in A_2. The expansion coefficients are of the form $A_2/(1-A_2\sigma_z^\infty)$ or $A_2^2/(1-A_2^2(\sigma_3+\sigma_4))$. Owing to the fact that even at very high laser-field strengths ($\bar{n}_L$) we still have $A_2\ll 1$, we are justified in retaining only the first two terms of the expansions without incurring an error greater than 1 per cent (for the numerical values assumed in our computations). On integration and after some cumbersome transformations, we arrive at the TDS for the SRS process in the following form:

$$\begin{aligned} S(D,\Gamma,t)=&\,S_L(D,\Gamma,t)+S_{k_0}(D,\Gamma,t)\\ &+S_1(D,\Gamma,t)+S_2(D,\Gamma,t)\\ &+S_3(D,\Gamma,t) \end{aligned} \tag{38}$$

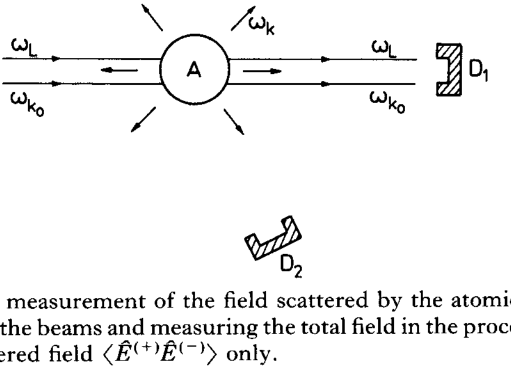

Figure 2. Set-up for the measurement of the field scattered by the atomic system A: D_1, detector collinear to the beams and measuring the total field in the process; D_2, detector measuring the scattered field $\langle \hat{E}^{(+)}\hat{E}^{(-)}\rangle$ only.

The first two components of (38) represent the shape of the external fields; in our case, this gives the following expressions:

$$S_L(D,\Gamma,t)=\varepsilon_L\bar{n}_L\frac{2\Gamma}{\Gamma^2+(D-\omega_L)^2}, \tag{39}$$

$$S_{k_0}(D,\Gamma,t)=\varepsilon_{k_0}\bar{n}_{k_0}\frac{2\Gamma}{\Gamma^2+(D-\omega_{k_0})^2}. \tag{40}$$

Next, we have

$$\begin{aligned} S_1(D,\Gamma,t)=\frac{\mathscr{A}_1\sigma_+}{\tilde{M}_0}&[X_1'(t)\cos\bar{\Omega}t+X_1''(t)\sin\bar{\Omega}t\\ &+X_2'(t)\cos(D-\omega_L)t+X_2''(t)\sin(D-\omega_L)t+X_3(t)]\\ &+\mathscr{A}_1'\sigma_-[X_4'(t)\cos\bar{\Omega}t+X_4''(t)\sin\bar{\Omega}t\\ &+X_5'(t)\cos\tilde{\Omega}t+X_5''(t)\sin\tilde{\Omega}t+X_6(t)]. \end{aligned} \tag{41}$$

The expression (41) is the spectral counterpart of the correlation function $G_2(t,\tau)$. The greatest component of the correlation function (the expression proportional to $\langle S^-S^+\rangle$) leads to the following spectral component:

$$S_2(D,\Gamma,t)=\mathscr{A}_2[Y_1'(t)\cos\tilde{\Omega}t+Y_1''(t)\sin\tilde{\Omega}t+Y_2(t)]. \tag{42}$$

The last two components of $G_3(t,\tau)$ give

$$\begin{aligned} S_3(D,\Gamma,t)=\mathscr{A}_3\exp(-2\Gamma t)\{&[\sigma_-(\sigma_6Z_1'(t)+\sigma_5Z_3'(t))+\sigma_+\sigma_7Z_5'(t)]\cos\tilde{\Omega}t\\ &+[\sigma_-(\sigma_6Z_1''(t)+\sigma_5Z_3''(t))+\sigma_+\sigma_7Z_5''(t)]\sin\tilde{\Omega}t\\ &+\sigma_-(\sigma_6Z_2(t)+\sigma_5Z_4(t))+\sigma_+\sigma_7Z_6(t)\}. \end{aligned} \tag{43}$$

Here,

$$\tilde{\Omega}=D-\omega_L-\bar{\Omega}. \tag{44}$$

The analytical form of the coefficients $X_i(t)$, $Y_i(t)$ and $Z_i(t)$ and the quantities determining them, are given in the Appendix. The exact formulae show that, for an atomic system in its ground state or excited state at $t=0$, the spectrum is described by $S_2(D,\Gamma,t)$ alone. For partial population inversion at $t=0$, the other terms of the spectrum $S(D,\Gamma,t)$ are non-zero and modify the Raman spectrum. The TDS also provides a picture of the time-evolution of the SRS process. In the next section, we perform a numerical analysis of the obtained spectrum.

4. Numerical analysis of $S(D,\Gamma,t)$

We consider the following situation: the two stimulating beams (the Stokes ω_{k_0} and laser ω_L beams) are monochromatic and collinear, and fulfil the condition $\Delta_{k_0}\cong 0$. The detector is positioned in the direction of the collinear stimulating beams (configuration D_1 in figure 2); obviously, it records all the fields occurring in the process. Figure 3 shows the spectrum of the process as 'seen' by the detector D_1 at $t=0{\cdot}7$ for $\omega_L=5000$ and $\omega=100$ (all quantities are in arbitrary units). The laser line (ω_L) and Stokes line (ω_{k_0}) are accompanied by the Raman-scattered line, originating in the radiation-shifted atomic levels. Our numerical analysis concerns the Raman line. Figure 4 shows the process of its creation from $t=0$ onwards.

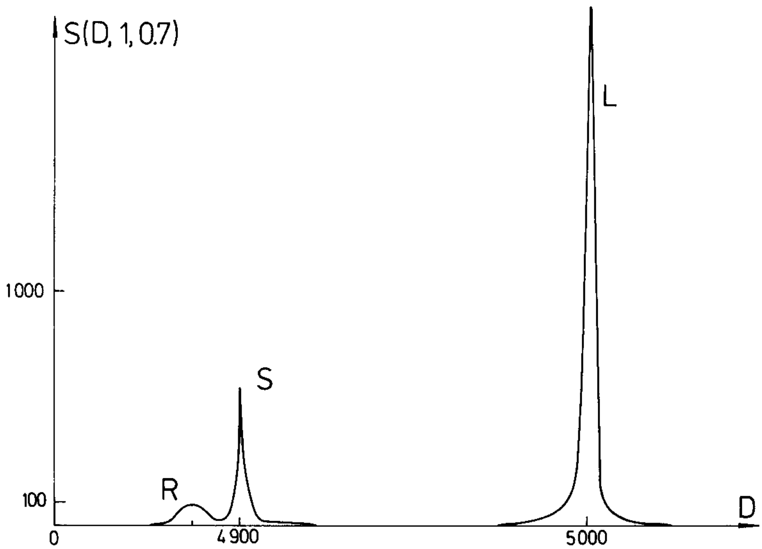

Figure 3. Spectrum measured by detector D_1: L, laser line ($\omega_L = 5000$); S, line of the external Stokes field ($\omega_{k_0} = \omega_L - \omega - \Delta_{k_0}$); R, SRS line ($\omega = 100$; $\Gamma = 1$; $\bar{n}_L/\bar{n}_{k_0} = 10$; $t = 0{\cdot}7$; $\sigma(0) = -1$; $\Delta_{k_0} \cong 0$). All quantities are in arbitrary units.

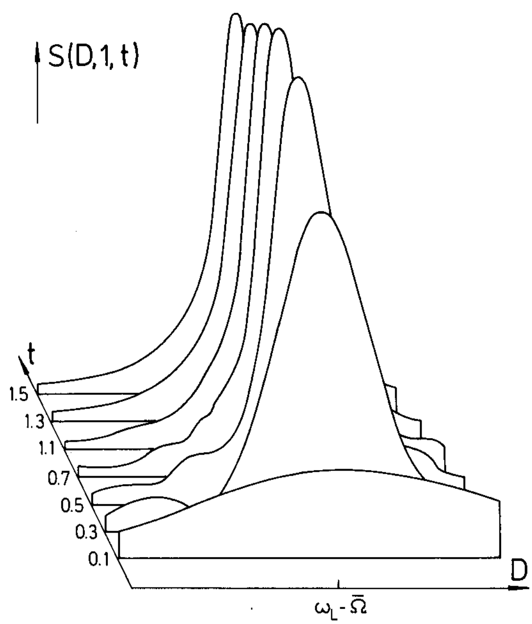

Figure 4. Time-evoluiton of the Raman line R. The conditions are those of figure 3.

We deal with the case when, at $t=0$, the atomic system is in its ground state, i.e. population inversion amounts to

$$\sigma(0)=2\sigma_z(0)=-1. \tag{45}$$

At time $t=0{\cdot}7$ the intensity of the Raman line attains its maximum value; after that, it decreases slightly and becomes stationary. The tails of the line exhibit some influence of the harmonic components of the spectrum which, at high values of time t, quench out owing to the presence of damping factors. A similar effect of 'wiggles' has been obtained for the spectral lines of resonance fluorescence and superfluorescence [5, 6]. The interpretation given is that of intra-detector transitions, related with interference in the Fabry–Pérot interferometer The time-evolution of the SRS spectral line is portrayed more clearly in figure 5, where the intensity of the central part of the line is shown for $D=\omega_L-\bar{\Omega}$ ($\tilde{\Omega}=0$) as a function of time. One notes that the time-evolution of the line intensity is very strongly dependent on the intensity of the external fields. For a strong laser field $\bar{n}_L$ ($n_{k_0}=\text{const.}$), the maximum of the intensity curve tends to vanish and the intensity of the central part decreases. This is due to the fact that, for strong fields, radiation broadening of the line is considerable and at the expense of its central part. It is moreover worth noting that in the case of strong fields the process leading to the emergence of the line proceeds more rapidly (is shorter) than in that in weak fields. Here, we assume the intensity of the Stokes field (ω_{k_0}) to be much weaker than that of the laser field (ω_L) in order to eliminate effects of higher-order Raman scattering ($\omega_{2k}=\omega_{k_0}-\omega$).

The situation is much more interesting if the initial population inversion $\sigma(0)$ corresponds to partial excitation of the atomic system at $t=0$. All the components of $S(D, \Gamma, t)$ are then non-zero, and the picture of the creation of the Raman line is quite different. The scattered light intensity exhibits oscillations: figure 6 show the initial evolution of the line intensity for several values of $\sigma(0)$. The amplitude of the oscillations grows with increasing initial population inversion.

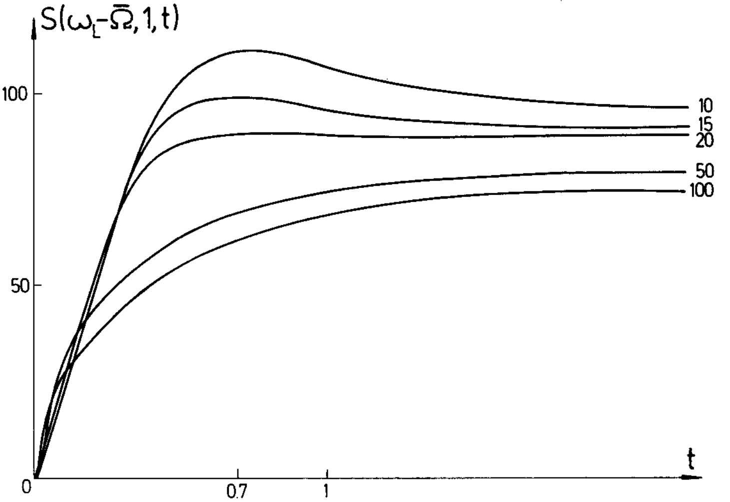

Figure 5. Intensity of the line R in its central part ($D=\omega_L-\bar{\Omega}$) versus the time, for different values of the ratio $\bar{n}_L/\bar{n}_{k_0}$. The other conditions are those of figure 3.

We also performed a numerical analysis for the effect of the detector on the SRS process. In our case it was a Fabry–Pérot detector (cf. equation (37)); the influence of its spectral width Γ on the SRS line-shape is shown in figure 7. With increasing Γ the line exhibits diffluence, as would be expected. The same has been proved for resonance fluorescence [5]. Moreover—and this is a new finding of especial interest—Γ affects the observation of the time-dependent (dynamical) changes in intensity of the SRS line. From figure 8, the magnitude of Γ affects the amplitude of

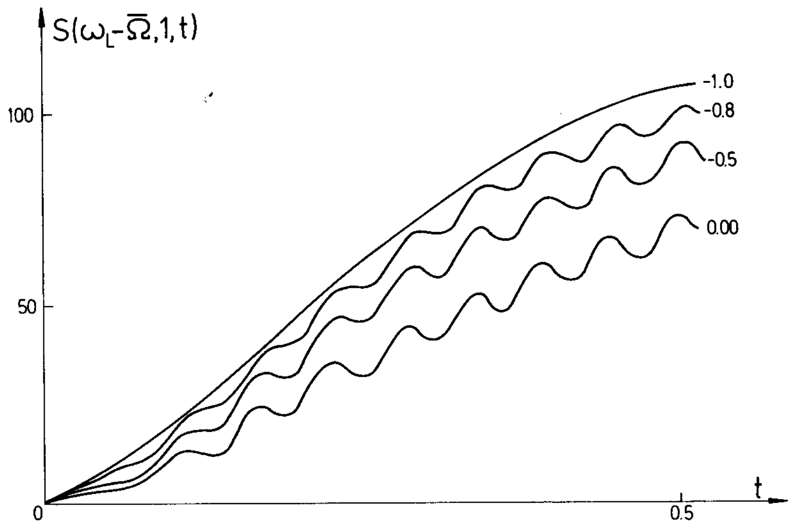

Figure 6. Influence of population inversion $\sigma(0)$ on the process leading to the Raman line R.

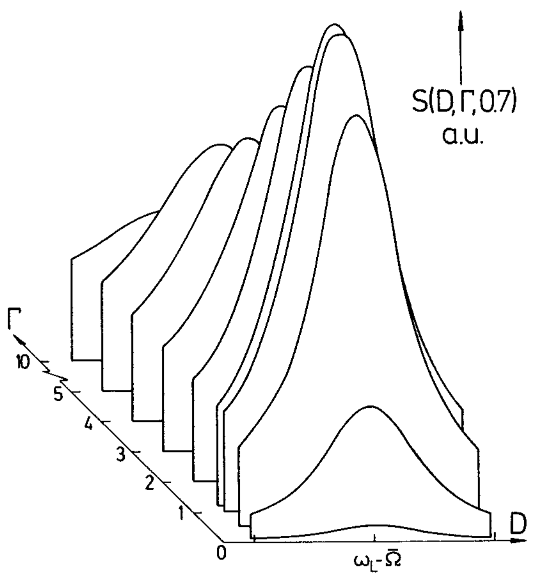

Figure 7. Shape of the spectral line R as a function of the spectral width Γ.

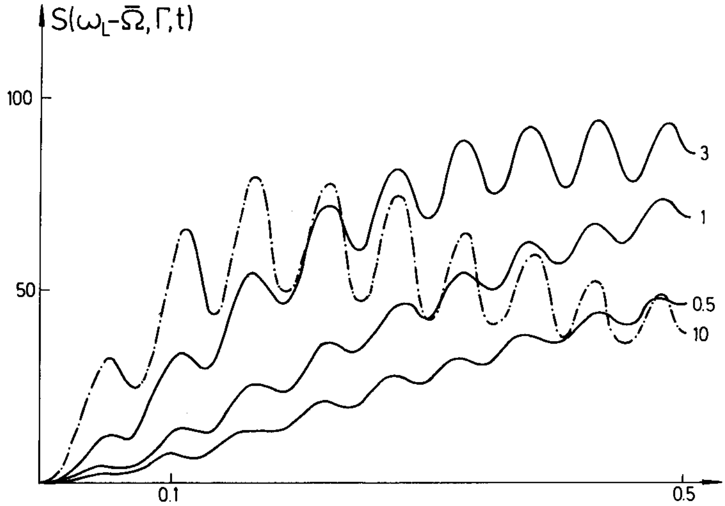

Figure 8. The oscillations in intensity of the line R for different values of Γ.

the oscillations in intensity of the scattered field measured by the detector. Also, the greater Γ the shorter is the time required for the mean intensity of the line to become maximal.

It should be stated once again that the above concerns the picture of the process as 'seen' by the detector. Naturally, the properties of the latter are unable to exert an influence on the course of the scattering process itself.

5. Summary

The two-photon process, in our case SRS, when measured with a physical detector, is found not to be an instantaneous event but rather to evolve in time. The formation time of the spectral line is finite. We carried out a numerical study of the time-evolution of the Raman line both with regard to its spectral properties and with regard to the time variations of its intensity, and determined the effect of the external fields as well as the parameters of the scattering system on the creation process of the SRS line.

For a partially excited scattering system we obtained a line of oscillating intensity, with an observed oscillation amplitude dependent on the spectral properties of the detector. Thus, the measuring device affects not only the spectral properties of the observed SRS line but also the time-evolution of its intensity as well.

Our calculations concerned the case of a two-level system without damping, with higher-order effects and anti-Stokes effect neglected. Work along these lines, taking into consideration the above effects, is proceeding.

Appendix

The explicit form of the coefficients $X_i(t)$, $Y_i(t)$ and $Z_i(t)$ and the quantities determining them are

$$X'_1(t)=\left(\frac{q_1}{\tilde{M}_1^1}-\frac{q_3}{\tilde{M}_1^2}\right)\exp(-\bar{\gamma}t)+d\exp(-3\bar{\gamma}t)\left(\frac{q_9}{\tilde{M}_1^1\tilde{M}_3^1}-\frac{q_7}{\tilde{M}_1^2\tilde{M}_3^2}\right),$$

$$X''_1(t)=\left(\frac{q_4}{\tilde{M}_1^2}-\frac{q_2}{\tilde{M}_1^1}\right)\exp(-\bar{\gamma}t)+d\exp(-3\bar{\gamma}t)\left(\frac{q_8}{\tilde{M}_1^1\tilde{M}_3^1}-\frac{q_{10}}{\tilde{M}_1^2\tilde{M}_3^2}\right),$$

$$X'_2(t)=\frac{\exp(-\Gamma t)}{\tilde{M}_1^1}\left(\frac{q_7}{\tilde{M}_3^1}-q_1\right),$$

$$X''_2(t)=\frac{\exp(-\Gamma t)}{\tilde{M}_1^1}\left(\frac{q_8}{\tilde{M}_3^1}-q_2\right),$$

$$X_3(t)=\frac{\exp(-2\Gamma t)}{\tilde{M}_1^2}\left(q_3-\frac{q_9 d}{\tilde{M}_3^2}\right),$$

$$X'_4(t)=\frac{\exp(-\bar{\gamma}t)}{\tilde{M}_1^1}\left(\frac{q_1}{\tilde{M}_0}-\frac{r_1}{\tilde{M}_1^2}\right)+\frac{d\exp(-3\bar{\gamma}t)}{\tilde{M}_3^1}\left(\frac{r_5}{\tilde{M}_3^2}-\frac{r_3}{\tilde{M}_0}\right),$$

$$X''_4(t)=-\frac{\exp(-\bar{\gamma}t)}{\tilde{M}_1^1}\left(\frac{q_2}{\tilde{M}_0}+\frac{r_2}{\tilde{M}_1^2}\right)-\frac{d\exp(-3\bar{\gamma}t)}{\tilde{M}_3^1}\left(\frac{r_6}{\tilde{M}_3^2}-\frac{r_4}{\tilde{M}_0}\right),$$

$$X'_5(t)=\frac{dr_3}{\tilde{M}_3^1\tilde{M}_0}\exp(-\Gamma-3\bar{\gamma})t-\frac{q_1}{\tilde{M}_1^1\tilde{M}_0}\exp(-\Gamma-\bar{\gamma})t,$$

$$X''_5(t)=\frac{dr_4}{\tilde{M}_3^1\tilde{M}_0}\exp(-\Gamma-3\bar{\gamma})t+\frac{q_2}{\tilde{M}_1^1\tilde{M}_0}\exp(-\Gamma-\bar{\gamma})t,$$

$$X_6(t)=\exp(-2\Gamma t)\left(\frac{r_1}{\tilde{M}_1^1\tilde{M}_1^2}-\frac{dr_5}{\tilde{M}_3^2\tilde{M}_3^1}\right),$$

$$Y'_1(t)=-P_1\left(\frac{C_1p_1}{\tilde{M}_1^1}-\frac{C_2p_5}{\tilde{M}_{-1}^1}-\frac{\sigma_2\sigma_\infty p_7}{\tilde{M}_{-3}^1}-2b\sigma_3\sigma_1\right)$$
$$\times\exp(-\Gamma-3\bar{\gamma})t+P_2\left(b\left(\frac{p_1}{\tilde{M}_3^1}+\frac{\sigma_\infty}{\sigma_1}\right)-\frac{\sigma_\infty p_3}{\sigma_1\tilde{M}_{-1}^1}-1\right)\exp(-\Gamma-\bar{\gamma})t,$$

$$Y''_1(t)=-P_1\left(\frac{C_1p_2}{\tilde{M}_1^1}-\frac{C_2p_6}{\tilde{M}_{-1}^1}-\frac{\sigma_2\sigma_\infty 3p_2}{\tilde{M}_{-3}^1}\right)\exp(-\Gamma-3\bar{\gamma})t$$
$$-P_2\left(\frac{b}{\tilde{M}_3^1}+\frac{\sigma_\infty}{\sigma_1\tilde{M}_{-1}^1}\right)p_2\exp(-\Gamma-\bar{\gamma})t,$$

$$Y_2(t)=-P_1b\sigma_1\sigma_3\exp(-6\bar{\gamma}t)+\left[P_1\left\{bC_0\left(\frac{2p_1}{\tilde{M}_1^1}+\frac{\Gamma-3\bar{\gamma}}{2\bar{\gamma}-\Gamma}\right)+\sigma_3\sigma_1\right.\right.$$
$$\left.\left.\times\left(\frac{\Gamma-3\bar{\gamma}}{2(2\bar{\gamma}-\Gamma)}+\frac{p_1}{\tilde{M}_1^1}\right)\right\}+P_2\left(b\frac{\Gamma-\bar{\gamma}}{2(\Gamma-2\bar{\gamma})}-\frac{bp_1}{\tilde{M}_3^1}\right)\right]\exp(-4\bar{\gamma}t)$$
$$+\left[P_1C_2\left(\frac{p_5}{\tilde{M}_{-1}^1}-\frac{\Gamma-3\bar{\gamma}}{2(\bar{\gamma}-\Gamma)}\right)+P_2\left(1-\frac{\sigma_\infty b}{\sigma_1}\right)\Big/2\right]\exp(-2\bar{\gamma}t)$$
$$-\left[P_1\left\{C_0\left(\frac{b}{2\bar{\gamma}-\Gamma}-\frac{1}{2(\bar{\gamma}-\Gamma)}\right)+\sigma_2\sigma_\infty\right.\right.$$
$$\left.\times\left(\frac{b}{\bar{\gamma}-\Gamma}+\frac{1}{2\Gamma}\right)+\sigma_3\sigma_1\left(\frac{b}{\Gamma-3\bar{\gamma}}+\frac{1}{2(2\bar{\gamma}-\Gamma)}\right)\right\}$$

$$\times(\Gamma-3\bar{\gamma})-P_2\left\{\frac{b}{2(\Gamma-2\bar{\gamma})}-\frac{1}{2(\bar{\gamma}-\Gamma)}-\frac{\sigma_\infty}{\sigma_1}\right.$$
$$\left.\times\left(\frac{1}{2\Gamma}+\frac{b}{2(\bar{\gamma}-\Gamma)}\right)\right\}(\Gamma-\bar{\gamma})\Bigg]\exp(-2\Gamma t)$$
$$+P_1\sigma_2\sigma_\infty\left(\frac{\Gamma-3\bar{\gamma}}{2\Gamma}-\frac{p_7}{\tilde{M}^1_{-3}}\right)+P_2\frac{\sigma_\infty}{\sigma_1}\left(\frac{p_3}{\tilde{M}^1_{-1}}-\frac{\Gamma-\bar{\gamma}}{2\Gamma}\right),$$

$$Z'_1(t)=\frac{P_0}{B}\left(\frac{p_1 b}{\tilde{M}^1_3}-1\right)\exp(\Gamma-\bar{\gamma})t+\frac{1}{B}\Bigg[P_1\left\{\sigma_2\left(\frac{p_5}{\tilde{M}^1_{-1}}-\frac{bp_1}{\tilde{M}^1_1}\right)+\sigma_3\left(2b-\frac{p_1}{\tilde{M}^1_1}\right)\right\}$$
$$+\frac{P'_1}{B}\left(\frac{p_1}{\tilde{M}^1_1}-b\right)\Bigg]\exp(\Gamma-3\bar{\gamma})t,$$

$$Z''_1(t)=\frac{p_2}{B}\left(\frac{P_0 b}{\tilde{M}^1_3}\exp(\Gamma-\bar{\gamma})t-\frac{P'_1}{B\tilde{M}^1_1}\exp(\Gamma-3\bar{\gamma})t\right)+\frac{P_1}{B}$$
$$\times\left[\frac{\sigma_2 p_6}{\tilde{M}^1_{-1}}-\frac{p_2}{\tilde{M}^1_1}(\sigma_2 b+\sigma_3)\right]\exp(\Gamma-3\bar{\gamma})t$$

$$Z_2(t)=\frac{P_0}{B}\Bigg[\frac{1}{2}\exp(2(\Gamma-\bar{\gamma})t)-b\left(\frac{p_1}{\tilde{M}^1_3}-\frac{\Gamma-\bar{\gamma}}{2(\Gamma-2\bar{\gamma})}\right)\exp(2(\Gamma-2\bar{\gamma})t)$$
$$-\frac{1}{2}\left(\frac{b(\Gamma-\bar{\gamma})}{2(\Gamma-2\bar{\gamma})}-1\right)\Bigg]+\frac{P_1}{B}$$
$$\times\Bigg[\sigma_2\left\{\frac{\Gamma-3\bar{\gamma}}{2}\left(\frac{\exp(2(\Gamma-\bar{\gamma})t)-1}{\Gamma-\bar{\gamma}}+\frac{2b}{\Gamma-2\bar{\gamma}}(1-\exp(2(\Gamma-2\bar{\gamma})t))\right)\right]$$
$$\left.+\frac{bp_1}{\tilde{M}^1_1}\exp(2(\Gamma-2\bar{\gamma})t)-\frac{p_6}{\tilde{M}^1_{-1}}\exp(2(\Gamma-\bar{\gamma})t)\right\}$$
$$+\sigma_3\left\{\frac{\Gamma-3\bar{\gamma}}{2(\Gamma-2\bar{\gamma})}(1-\exp(2(\Gamma-2\bar{\gamma})t))\right.$$
$$\left.+\frac{p_1}{\tilde{M}^1_1}\exp(2(\Gamma-\bar{\gamma})t)-b(\exp(2(\Gamma-3\bar{\gamma})t)+1)\right\}\Bigg]$$
$$+\frac{P'_1 b}{B^2}\exp(2(\Gamma-3\bar{\gamma})t),$$

$$Z'_3(t)=\frac{P_0 b}{B}\left(\frac{p_2}{\tilde{M}^1_3}\exp(\Gamma-3\bar{\gamma})t-\frac{2bp_8}{\tilde{M}^1_5}\exp(\Gamma-5\bar{\gamma})t\right)-\frac{P_1 p_5}{A_2^2\tilde{M}^1_{-1}B}\exp(\Gamma-3\bar{\gamma})t$$
$$+\frac{P_5\sigma'_1}{B^2}\left(\frac{p_4}{\tilde{M}^1_{-1}}-\frac{p_8 b}{\tilde{M}^1_1}\right)\exp(\Gamma-5\bar{\gamma})t$$
$$+\frac{P_5^2\tilde{M}^1_5}{B}\left[\sigma_2\left(\frac{p_9}{\tilde{M}^1_{-3}}-\frac{2bp_8}{\tilde{M}^1_{-1}}\right)+\frac{\sigma_3 p_4}{\tilde{M}^1_{-1}}\right]\exp(\Gamma-5\bar{\gamma})t,$$

$$Z''_3(t)=\frac{P_0 b}{B}\left(\frac{p_2}{\tilde{M}^1_3}\exp(\Gamma-3\bar{\gamma})t-\frac{2bp_6}{\tilde{M}^1_5}\exp(\Gamma-5\bar{\gamma})t\right)$$

$$-\frac{P_1 p_6}{A_2^2 \tilde{M}_{-1}^1 B}\exp(\Gamma-3\bar{\gamma})t-\frac{P_5\sigma_1'}{B^2}\left(\frac{3p_2}{\tilde{M}_{-1}^1}-\frac{p_6 b}{\tilde{M}_1^1}\right)\exp(\Gamma-5\bar{\gamma})t$$

$$+\frac{P_5^2\tilde{M}_5^1}{B}\left(2p_6\sigma_2\left(\frac{1}{\tilde{M}_{-3}^1}-\frac{b}{\tilde{M}_{-1}^1}\right)+\frac{3\sigma_3 p_2}{\tilde{M}_{-1}^1}\right)\exp(\Gamma-5\bar{\gamma})t,$$

$$Z_4(t)=\frac{P_5\sigma_1'}{B^2}\left[\left(\frac{\Gamma-5\bar{\gamma}}{2(\Gamma-2\bar{\gamma})}-\frac{p_4}{\tilde{M}_{-1}^1}\right)\exp(2(\Gamma-2\bar{\gamma})t)\right.$$

$$\left.+\left(\frac{p_8 b}{\tilde{M}_1^1}-\frac{b(\Gamma-5\bar{\gamma})}{2(\Gamma-3\bar{\gamma})}\right)\exp(2(\Gamma-3\bar{\gamma})t)+\frac{\Gamma-5\bar{\gamma}}{2}\left(\frac{b}{\Gamma-3\bar{\gamma}}-\frac{1}{\Gamma-2\bar{\gamma}}\right)\right]$$

$$-\frac{P_5 b(\Gamma-5\bar{\gamma})}{2B(\Gamma-2\bar{\gamma})}(\exp(2(\Gamma-2\bar{\gamma})t)-1)$$

$$+\frac{P_5^2\tilde{M}_5^1}{B}\left[\sigma_2\left\{\left(\frac{2p_8}{\tilde{M}_{-1}^1}-\frac{\Gamma-5\bar{\gamma}}{\Gamma-2\bar{\gamma}}\right)b\exp(2(\Gamma-2\bar{\gamma})t)\right.\right.$$

$$+\left(\frac{\Gamma-5\bar{\gamma}}{2(\Gamma-\bar{\gamma})}-\frac{p_9}{\tilde{M}_{-3}^1}\right)\exp(2(\Gamma-\bar{\gamma})t)$$

$$\left.-(\Gamma-5\bar{\gamma})\left(\frac{1}{2(\Gamma-\bar{\gamma})}-\frac{b}{\Gamma-2\bar{\gamma}}\right)\right\}+\sigma_3\left\{\frac{b(\Gamma-5\bar{\gamma})}{B(\Gamma-3\bar{\gamma})}\right.$$

$$\left.\left.\times(1-\exp(2(\Gamma-3\bar{\gamma})t))-\frac{p_4}{B}\exp(2(\Gamma-2\bar{\gamma})t)\right\}\right]$$

$$+\frac{P_1}{A_2^2}\left[\left(\frac{\Gamma-3\bar{\gamma}}{2(\Gamma-\bar{\gamma})}+\frac{p_5}{\tilde{M}_{-1}^1 B}\right)\exp(2(\Gamma-\bar{\gamma})t)\right.$$

$$\left.+\frac{b(\Gamma-3\bar{\gamma})}{2B(\Gamma-2\bar{\gamma})}(\exp(2(\Gamma-2\bar{\gamma})t)-1)-\frac{\Gamma-3\bar{\gamma}}{2(\Gamma-\bar{\gamma})}\right]$$

$$+\frac{P_0 b}{B}\left(\frac{2bp_8}{\tilde{M}_5^1}\exp(2(\Gamma-3\bar{\gamma})t)-\frac{p_1}{\tilde{M}_3^1}\exp(2(\Gamma-2\bar{\gamma})t)\right),$$

$$Z_5'(t)=P_1 B\left[\sigma_2\left(\frac{p_6}{\tilde{M}_{-1}^1}+\frac{(2b-d)p_2}{\tilde{M}_1^1}\right)-\frac{\sigma_3 p_2}{\tilde{M}_1^1}\right]\exp(\Gamma-3\bar{\gamma})t$$

$$Z_5''(t)=\left(\frac{p_1(b+d)}{\tilde{M}_3^1}+1\right)\frac{1}{\tilde{M}_1^1}\exp(\Gamma-\bar{\gamma})t+P_1 B\left[\sigma_2\left(\frac{p_5}{\tilde{M}_{-1}^1}+\frac{2b-d}{\tilde{M}_1^1}p_1\right)\right.$$

$$\left.-\sigma_3\left(\frac{p_1}{\tilde{M}_1^1}-2b+d\right)\right]\exp(\Gamma-3\bar{\gamma})t,$$

$$Z_6(t)=\frac{1}{\tilde{M}_1^1}\left[(d+b)\left(\frac{2(\Gamma-\bar{\gamma})}{(\Gamma-3\bar{\gamma})}-\frac{p_1}{\tilde{M}_3^1}\right)\exp(2(\Gamma-2\bar{\gamma})t)\right.$$

$$\left.-\frac{2(\Gamma-\bar{\gamma})(d-b)}{\Gamma-3\bar{\gamma}}+\frac{\exp(2(\Gamma-\bar{\gamma})t)}{2}+1\right]$$

$$+P_1 B\left[\frac{\sigma_2(\Gamma-3\bar{\gamma})}{2}\left\{\frac{\exp(2(\Gamma-\bar{\gamma})t)-1}{\Gamma-\bar{\gamma}}+\frac{(2b-d)(\exp(2(\Gamma-2\bar{\gamma})t)-1}{\Gamma-2\bar{\gamma}}\right\}\right.$$

$$+\frac{\sigma_3}{2\bar{\gamma}}\left\{\frac{2b-d}{2}(\exp(-4\bar{\gamma}t)-1)+\exp(-2\bar{\gamma}t)-1\right\}$$

$$+\left\{\frac{p_1}{\tilde{M}_1^1}\exp(-2\bar{\gamma}t)(\sigma_2(2b-d)+\sigma_3)-\frac{\sigma_2 p_5}{\tilde{M}_{-1}^1}\right\}$$

$$\times\exp(2(\Gamma-\bar{\gamma})t)+\sigma_3(2b-d)\exp(2(\Gamma-3\bar{\gamma})t)\Bigg]$$

within

$$\mathscr{A}_1=\frac{4\Gamma A_1\varepsilon_L\bar{n}_L}{1-A_2\sigma_z},\quad \mathscr{A}'_1=\frac{4\Gamma A_1\varepsilon_L\bar{n}_L}{1-A_2\sigma_z^\infty},$$

$$\mathscr{A}_2=4\Gamma A_1^2\varepsilon_L\bar{n}_L,\quad \mathscr{A}_3=\mathscr{A}_2A_2,$$

$$P_0=\frac{1}{\tilde{M}_1^1B},\quad P_1=\frac{A_2^2}{\tilde{M}_3^1B^2},\quad P'_1=\frac{\sigma'_1A_2}{\tilde{M}_3^1B},$$

$$P_2=\frac{\sigma_1}{\tilde{M}_1^1B},\quad P_5=\frac{A_2^2}{\tilde{M}_5^1B},$$

$$B=1-A_2^2\sigma_4,\quad b=\frac{A_2^2\sigma_3}{B},\quad d=\frac{A_2\sigma'_1}{1-A_2\sigma_z^\infty},$$

$$\tilde{M}_0=\Gamma^2+(D-\omega_L)^2,\quad \bar{\tilde{M}}_n^m=(m\Gamma-n\bar{\gamma})^2+\bar{\tilde{\Omega}},$$

$$C_0=\sigma_1\sigma_2-\sigma_3\sigma_\infty,\qquad \sigma_\pm=\langle\hat{S}^\pm(0)\rangle,$$

$$C_1=2bC_0+\sigma_3\sigma_1,\qquad \sigma'_1=\sigma_z-\sigma_z^\infty$$

$$C_2=C_0-2b\sigma_2\sigma_\infty,$$

$$r_1=(\Gamma-\bar{\gamma})(2\Gamma-\bar{\gamma})-\bar{\Omega}\tilde{\Omega},\qquad r_2=(\Gamma-\bar{\gamma})\bar{\Omega}+\tilde{\Omega}(2\Gamma-\bar{\gamma}),$$

$$r_3=(\Gamma-3\bar{\gamma})\Gamma+\tilde{\Omega}(D-\omega_L),\qquad r_4=\tilde{\Omega}\Gamma-(D-\omega_L)(\Gamma-3\bar{\gamma}),$$

$$r_5=(\Gamma-3\bar{\gamma})(2\Gamma-3\bar{\gamma})-\bar{\Omega}\tilde{\Omega},\qquad r_6=(2\Gamma-3\bar{\gamma})\tilde{\Omega}+\bar{\Omega}(\Gamma-3\bar{\gamma}),$$

$$q_1=\Gamma(\Gamma-\bar{\gamma})+\tilde{\Omega}(D-\omega_L),\qquad q_2=(D-\omega_L)(\Gamma-\bar{\gamma})-\tilde{\Omega}\Gamma,$$

$$q_3=(2\Gamma-\bar{\gamma})+(D-\omega_L)\tilde{\Omega},\qquad q_4=(2\Gamma-\bar{\gamma})(D-\omega_L)-\Gamma\tilde{\Omega},$$

$$q_5=(2\Gamma-\bar{\gamma})(2\Gamma-3\bar{\gamma})+\tilde{\Omega}^2,\qquad q_6=2\tilde{\Omega}\bar{\gamma},$$

$$q_7=q_1p_1+q_2p_2,\qquad q_8=q_2p_1-p_2q_1,$$

$$q_9=q_3q_5+q_4q_6,\qquad q_{10}=q_4q_5-q_3q_6,$$

$$p_1=(\Gamma-\bar{\gamma})(\Gamma-3\bar{\gamma})+\tilde{\Omega}^2,\qquad p_2=2\bar{\gamma}\tilde{\Omega},$$

$$p_3=\Gamma^2+\bar{\gamma}^2+\tilde{\Omega}^2,\qquad p_4=(\Gamma+\bar{\gamma})(\Gamma-5\bar{\gamma})+\tilde{\Omega}^2,$$

$$p_5=(\Gamma+\bar{\gamma})(\Gamma-3\bar{\gamma})+\tilde{\Omega}^2,\qquad p_6=2p_2,$$

$$p_7=(\Gamma-3\bar{\gamma})(\Gamma+3\bar{\gamma})+\tilde{\Omega}^2,\qquad p_8=(\Gamma-5\bar{\gamma})(\Gamma-\bar{\gamma})+\tilde{\Omega}^2,$$

$$p_9=(\Gamma-5\bar{\gamma})(\Gamma+3\bar{\gamma})+\tilde{\Omega}^2.$$

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