Deriving spectroscopic information from intensity-intensity correlations

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(Received 2 June 1994; revised manuscript received 12 September 1994)

We present a very simple theoretical framework for extracting spectroscopic data on an atom via stochastic probing with a fluctuating laser source. By exploiting the fact that the linear susceptibility contains all the atomic structure information in it, we show that the power spectrum of the fluctuations in the intensity radiated from an atomic sample provides the relevant, atomic-level information. The analysis we present is very general and can be applied to a wide variety of atomic and molecular systems.

PACS number(s): 42.50.Md

In a recent paper, Yabuzaki, Mitsui, and Tanaka [1] demonstrated that stochastic fluctuations in the radiation from a diode laser can be used to derive spectroscopic data on an atom. These authors used the technique on Cs transitions, where the intensity fluctuations on a diode laser beam transmitted through an atomic sample were spectrally analyzed to reveal the atomic-level structure information. The method used appears to be an important spectroscopic technique, where the fluctuations in the radiation from a laser are utilized to get useful information. Related experiments have also been performed by McLean, Hannaford, and Fairchild [2] to probe atmospheric oxygen and by McIntyre *et al.* [3], where the diode laser field, modeled as a phase-diffusing field, was used to probe rubidium. The fact that laser field fluctuations can be used for spectroscopy is quite contrary to the rather accepted notion that fluctuations should be avoided. Previous theoretical studies [4–7] to understand the issue of laser noise in spectroscopy have usually relied on the solution of some form of the optical Bloch equations with the noise incorporated into the equations.

In this paper, we consider a sample of an atomic medium irradiated by a field whose amplitude, phase, frequency, or some combination of these, fluctuate, and present a very simple and yet general enough argument to decode the spectroscopic information contained in the intensity-intensity correlations of the laser beam transmitted through the atomic sample. The fluctuating laser field produces a fluctuating polarization in the medium, which we denote by $\vec{P}(r,\omega)$. This induced polarization can be written as

$$\vec{P}(r,\omega)=\vec{\chi}(\omega)\cdot\vec{E}(r,\omega)\ , \tag{1}$$

where $\vec{\chi}(\omega)$ is the linear response of the medium at the applied frequency. We will assume that the incident field is weak so that it is sufficient to work with the results of the linear-response theory. The induced polarization radiates a field which can be obtained from Maxwell's equations. In the far zone, this radiated field is given by

$$\vec{E}_R(\vec{r},\omega)\sim-\frac{e^{i(\omega/c)r}}{r}\frac{\omega^2}{c^2}\vec{n}\times\vec{n}\times\int_V\vec{P}(r',\omega)e^{-i(\omega/c)\vec{n}\cdot\vec{r}'}d^3r'\ , \tag{2}$$

where $\vec{n}$ is the unit vector in the direction of observation and the integral in Eq. (2) extends over the volume of the medium [8]. On combining Eqs. (1) and (2) we get

$$\vec{E}_R(\vec{r},\omega)\sim-\frac{e^{i(\omega/c)r}}{r}\frac{\omega^2}{c^2}\vec{n}\times\vec{n}\times\vec{\chi}(\omega)\times\int_V\vec{E}(r',\omega)e^{-i(\omega/c)\vec{n}\cdot\vec{r}'}d^3r'\ , \tag{3}$$

which for a plane wave in the direction $\vec{q}$

$$\vec{E}(\vec{r},\omega)=e^{i(\omega/c)\vec{q}\cdot\vec{r}}\vec{E}(\omega) \tag{4}$$

becomes

$$\vec{E}_R(\vec{r},\omega)\sim-\frac{e^{i(\omega/c)r}}{r}V\frac{\omega^2}{c^2}\vec{n}\times\vec{n}\times\vec{\chi}(\omega)\cdot\vec{E}(\omega)\Delta_\omega(\vec{n},\vec{q})\ , \tag{5}$$

where

$$\Delta_\omega(\vec{n},\vec{q})=\frac{1}{V}\int_V e^{i(\omega/c)(-\vec{n}+\vec{q})\cdot\vec{r}}d^3r\ . \tag{6}$$

Thus the positive-frequency part of the radiated field at the frequency ω can be written in the form

$$E_R(\omega)\sim f_\omega\chi(\omega)E(\omega)\ , \tag{7}$$

where for simplicity we have dropped the tensor and vector character of $\vec{\chi}(\omega)$ and $\vec{E}$. It should be borne in mind that the field $E(t)$ is centered around the frequency ω_L with a slowly varying envelope $\varepsilon(t)$. The radiated field is also expected to be a slowly varying function around the central frequency ω_L. We thus approximate Eq. (7) by

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$$E_R(\omega'+\omega_L)\sim f_{\omega_L}\chi(\omega'+\omega_L)E(\omega'+\omega_L)\ . \tag{8}$$

We rewrite Eq. (8) in terms of the Fourier transforms $\varepsilon(\omega)$ of the envelopes $\varepsilon(t)$

$$\varepsilon_R(\omega')\sim f_\chi(\omega'+\omega_L)\varepsilon(\omega')\ , \tag{9}$$

which when transformed to the time domain reads as $[\varepsilon(t)=\int_{-\infty}^{\infty}\varepsilon(\omega)e^{-i\omega t}d\omega]$

$$\varepsilon_R(t)\sim\frac{f}{2\pi}\int_{-\infty}^{\infty}X(t-t')\varepsilon(t')dt'\ . \tag{10}$$

Here $X(t)$ is the Fourier transform of

$$\Psi(\omega')\equiv\chi(\omega'+\omega_L)\ . \tag{11}$$

We next consider the detection of the radiated field by homodyning it with the incident field. The instantaneous intensity at the detector will be

$$\begin{aligned}I(t)&=|\varepsilon(t)e^{i\theta}+\varepsilon_R(t)|^2\\&=|\varepsilon(t)|^2+\frac{f}{2\pi}\int_{-\infty}^{\infty}X(t-t')\varepsilon^*(t)\varepsilon(t')e^{-i\theta}dt'\\&\quad+|\varepsilon_R(t)|^2+\frac{f^*}{2\pi}\int_{-\infty}^{\infty}X^*(t-t'')\varepsilon(t)\varepsilon^*(t'')e^{i\theta}dt''\ .\end{aligned} \tag{12}$$

Here we have included an extra phase factor in the homodyning process which would enable us to discriminate between different contributions to the observed signal. Since the field $\varepsilon(t)$ is fluctuating, the intensity becomes a stochastic variable. The quantity of interest is the intensity-intensity correlation, which is defined as

$$C_I(\tau)=\langle I(t+\tau)I(t)\rangle\ . \tag{13}$$

The correlation $C_I(\tau)$ has the significant property of being real and even, i.e., $C_I^*(\tau)=C_I(\tau)$ and $C_I(-\tau)=C_I(\tau)$. Using Eq. (12) we get

$$\begin{aligned}C_I(\tau)=&\langle|\varepsilon(t)|^2|\varepsilon(t+\tau)|^2\rangle+C_L(\tau)\\&+C_Q^{(1)}(\tau)+C_Q^{(2)}(\tau)+\cdots\ ,\end{aligned} \tag{14}$$

where

$$\begin{aligned}C_L(\tau)=\Bigg\langle|\varepsilon(t)|^2\Bigg\{&\int_{-\infty}^{\infty}\frac{f}{2\pi}X(t+\tau-t')\varepsilon^*(t+\tau)\varepsilon(t')e^{-i\theta}dt'+\int_{-\infty}^{\infty}\frac{f^*}{2\pi}X^*(t+\tau-t'')\varepsilon(t+\tau)\varepsilon^*(t'')e^{i\theta}dt''\Bigg\}\Bigg\rangle\\&+[\text{terms with } t \text{ and } (t+\tau) \text{ interchanged}]\ ,\end{aligned} \tag{15}$$

$$\begin{aligned}C_Q^{(1)}(\tau)=&\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{f^2}{4\pi^2}\langle\varepsilon^*(t+\tau)\varepsilon^*(t)X(t-t')X(t+\tau-t'')\varepsilon(t')\varepsilon(t'')\rangle e^{-2i\theta}dt'dt''\\&+\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{|f|^2}{4\pi^2}\langle\varepsilon^*(t'')\varepsilon^*(t)X(t-t')X^*(t+\tau-t'')\varepsilon(t')\varepsilon(t+\tau)\rangle dt'dt''+\text{c.c.}\ ,\end{aligned} \tag{16a}$$

and

$$C_Q^2(\tau)=\langle|\varepsilon(t)|^2|\varepsilon_R(t+\tau)|^2\rangle+\langle|\varepsilon(t+\tau)|^2|\varepsilon_R(t)|^2\rangle\ . \tag{16b}$$

Here C_L and C_Q are, respectively, linear and quadratic in the scattered field. The ellipsis in Eq. (14) gives terms of higher order in the scattered field and we will ignore such higher-order contributions. Note that by choosing $\theta=0$ and π (relative to the phase of f) and by adding or subtracting [9] the two measurements, we can study separately the linear and quadratic terms. Thus, in what follows we will study the behavior of linear and quadratic terms separately, and since the two contributions are studied separately, we drop the scale factor.

The power spectrum of the intensity-intensity correlations is defined as

$$P(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}C_I(\tau)e^{i\omega\tau}d\tau\ . \tag{17}$$

Replacing τ by $-\tau$ in Eq. (17) and using the evenness of $C_I(\tau)$ we get

$$P(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}C_I(\tau)e^{-i\omega\tau}d\tau\ . \tag{18}$$

From Eqs. (17) and (18), it is evident that

$$P(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}C_I(\tau)\cos(\omega\tau)d\tau\ . \tag{19}$$

To simplify Eq. (14), we adopt a simple model for the amplitude $\varepsilon(t)$ and assume that it is a Gaussian stochastic process. Needless to say, other models of stochastic noise can also be dealt with within the framework described here. In what follows we will drop from the spectrum the dc terms along with the peak at $\omega=0$. We first analyze the linear terms $C_L(\tau)$. Using the momentum theorem for Gaussian random processes Eq. (15) reduces to

$$C_L(\tau)=\frac{1}{2\pi}\int_{\infty}^{\infty}\langle|\varepsilon(t)|^2\rangle\langle\varepsilon^*(t+\tau)\varepsilon(t')\rangle X(t+\tau-t')e^{-i\theta}dt'$$
$$+\frac{1}{2\pi}\int_{-\infty}^{\infty}\langle\varepsilon(t)\varepsilon^*(t+\tau)\rangle\langle\varepsilon^*(t)\varepsilon(t')\rangle X(t+\tau-t')e^{-i\theta}dt'+\text{c.c.}+[\text{terms with } t \text{ and } (t+\tau) \text{ interchanged}]\ . \tag{20}$$

Note that the first term in Eq. (20) corresponds to the dc term [10] and can be dropped. We next introduce the second-order correlation function $\Gamma(\tau)$ for the field and its Fourier transform,

$$\Gamma(\tau)=\langle\varepsilon^*(t)\varepsilon(t+\tau)\rangle \tag{21}$$

and

$$\Gamma(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}\Gamma(\tau)e^{i\omega\tau}d\tau\ , \tag{22}$$

as well as the Fourier transform of $X(t)$

$$X(t)=\int_{-\infty}^{\infty}\Psi(\omega)e^{-i\omega t}d\omega\ . \tag{23}$$

Using Eq. (21), Eq. (20) reduces to

$$C_L(\tau)=\frac{1}{2\pi}\int_{-\infty}^{\infty}\Gamma(t-(t+\tau))\Gamma(t-t')$$
$$\times X(t+\tau-t')e^{-i\theta}dt'+\text{c.c.}$$
$$+[\text{terms with } t \text{ and } (t+\tau) \text{ interchanged}]\ . \tag{24}$$

On substituting Eq. (23) in Eq. (24) and using Eqs. (19) and (22), algebraic calculations lead to the following contribution to the spectrum:

$$P_L(\omega)=2\int_{-\infty}^{\infty}\Gamma(\omega')\{\Gamma(\omega'+\omega)$$
$$+\Gamma(\omega'-\omega)\}\text{Re}[\Psi(\omega')]d\omega'\ . \tag{25}$$

We next simplify the quadratic contributions to $P(\omega)$. We examine the two quadratic contributions separately. On using the moment theorem $C_Q^{(1)}(\tau)$ can be written as

$$C_Q^{(1)}(\tau)=\frac{1}{4\pi^2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\Gamma(-t'-\tau)\Gamma(\tau-t'')$$
$$\times X(t')X(t'')dt'dt''$$
$$+\frac{1}{4\pi^2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\Gamma(-t'-\tau+t'')\Gamma(\tau)X(t')$$
$$\times X^*(t'')dt'dt''+\text{c.c.}\ , \tag{26}$$

which on using Eqs. (22) and (23) simplifies to

$$C_Q^{(1)}(\tau)=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\Psi(\omega')\Psi(\omega'')\Gamma(\omega')\Gamma(\omega'')$$
$$\times e^{i(\omega'-\omega'')\tau}d\omega'd\omega''$$
$$+\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}|\Psi(\omega')|^2\Gamma(\omega')\Gamma(\omega'')$$
$$\times e^{i(\omega'-\omega'')\tau}d\omega'd\omega''+\text{c.c.} \tag{27}$$

Assuming that $\Gamma(\omega)$ is real and using the real nature of $C_I(\tau)$ we can write Eq. (27) as

$$C_Q^{(1)}(\tau)=\left\{\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\Psi(\omega')\Psi(\omega'')\Gamma(\omega')\Gamma(\omega'')\right.$$
$$\left.\times\cos(\omega'-\omega'')\tau d\omega'd\omega''+\text{c.c.}\right\}$$
$$+2\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}|\Psi(\omega')|^2\Gamma(\omega')\Gamma(\omega'')$$
$$\times\cos(\omega'-\omega'')\tau d\omega'd\omega''\ . \tag{28}$$

Using the relation $\int_{-\infty}^{\infty}\cos(\omega'-\omega'')\tau\cos(\omega\tau)d\tau=\pi[\delta(\omega'-\omega''+\omega)+\delta(\omega'-\omega''-\omega)]$ and substituting for $C_Q^{(1)}(\tau)$ from Eq. (28) into Eq. (19), we get

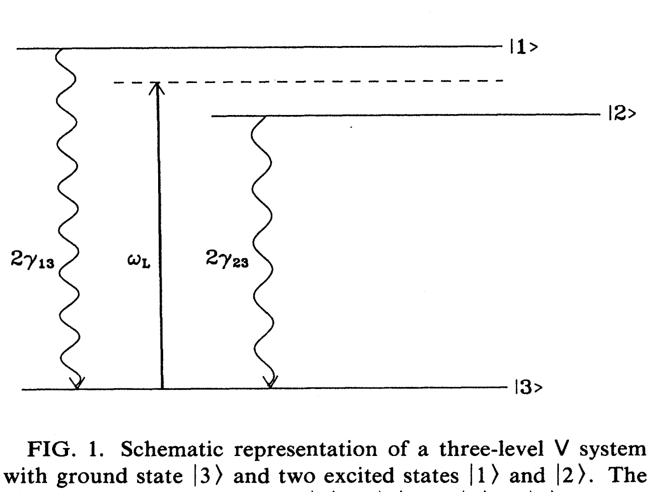

FIG. 1. Schematic representation of a three-level V system with ground state $|3\rangle$ and two excited states $|1\rangle$ and $|2\rangle$. The spontaneous decay rates from $|1\rangle$ to $|3\rangle$ and $|2\rangle$ to $|3\rangle$ are $2\gamma_{13}$ and $2\gamma_{23}$, respectively. The transition from $|1\rangle$ to $|2\rangle$ is not allowed. ω_L is the central frequency of the exciting field.

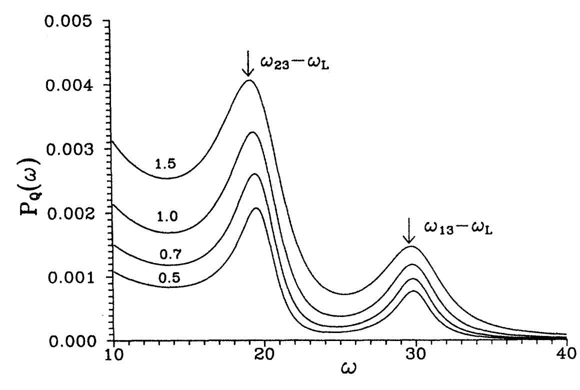

FIG. 2. Quadratic contribution $P_Q(\omega)$ (in arbitrary units) to the power spectrum of the intensity-intensity correlations is plotted as a function of frequency ω for values of γ_c of 0.5, 0.7, 1, and 1.5. The other parameters are $\chi_1=\chi_2$, $\omega_{13}-\omega_L=30$, and $\omega_{23}-\omega_L=20$. All frequencies and widths are in units of $\gamma_{13}=\gamma_{23}$.

$$P_Q^{(1)}(\omega)=\left\{\tfrac{1}{2}\int_{-\infty}^{\infty}\Psi(\omega')\Psi(\omega'+\omega)\Gamma(\omega')\Gamma(\omega'+\omega)d\omega'+\tfrac{1}{2}\int_{-\infty}^{\infty}\Psi(\omega')\Psi(\omega'-\omega)\Gamma(\omega')\Gamma(\omega'-\omega)d\omega'+\text{c.c.}\right\}$$
$$+\int_{-\infty}^{\infty}|\Psi(\omega)|^2\Gamma(\omega')\{\Gamma(\omega'+\omega)+\Gamma(\omega'-\omega)\}d\omega' . \quad (29)$$

The calculation of the contribution $P_Q^{(2)}(\omega)$ proceeds along similar lines and we quote the final result

$$P_Q^{(2)}(\omega)=\int_{-\infty}^{\infty}\Gamma(\omega'+\omega)\Gamma(\omega')\Psi^*(\omega'+\omega)\Psi(\omega')d\omega'$$
$$+\int_{-\infty}^{\infty}\Gamma(\omega'-\omega)\Gamma(\omega')\Psi^*(\omega'-\omega)\Psi(\omega')d\omega' . \quad (30)$$

The complete quadratic contribution is obtained by adding (29) and (30).

In order to understand the information contained in Eqs. (25), (29), and (30) we consider the simple V system shown in Fig. 1, i.e., a three-level atom with ground state $|3\rangle$ and two closely spaced excited $|1\rangle$ and $|2\rangle$. The spontaneous decay rates from $|1\rangle\rightarrow|3\rangle$ and $|2\rangle\rightarrow|3\rangle$ are denoted by $2\gamma_{13}$ and $2\gamma_{23}$, respectively. Assuming that the only allowed transitions are between the levels $|1\rangle$ and $|3\rangle$ and between $|2\rangle$ and $|3\rangle$, the linear susceptibility of the atomic medium is found to be

$$\Psi(\omega)=\left[\frac{i\chi_1}{\gamma_{13}+i(\omega_{13}-\omega-\omega_L)}+\frac{i\chi_2}{\gamma_{23}+i(\omega_{23}-\omega-\omega_L)}\right] . \quad (31)$$

The constants χ_i are related to the densities and oscillator strengths of the transitions. A Lorentzian profile centered at zero is chosen for the spectrum of the envelope $\varepsilon(t)$ of the exciting field and is given by

$$\Gamma(\omega)=\frac{\gamma_c/\pi}{\gamma_c^2+\omega^2} , \quad (32)$$

where γ_c is the spectral width of the incident beam.

Substituting from Eqs. (31) and (32) into Eqs. (25), (29), and (30), $P(\omega)$ is calculated analytically. A typical term involves calculation of an integral of the type

$$\int_{-\infty}^{\infty}\{(\omega'+i\gamma_c)(\omega'-i\gamma_c)[\omega'-(-\omega-i\gamma_c)]\times[\omega'-(-\omega+i\gamma_c)][\omega'-(\omega_{13}-\omega_L-i\gamma_{13})]\times[\omega'-(\omega_{23}-\omega_L-\omega+i\gamma_{23})]\}^{-1}d\omega' ,$$

which is evaluated using contour integration. The resulting analytical expression for $P(\omega)$ is provided in the Appendix. The numerical behavior of $P(\omega)$, as a function of frequency for different values of γ_c, is shown in Figs. 2 and 3. There are two peaks in the susceptibility behavior corresponding to the two transitions from $|1\rangle\leftrightarrow|3\rangle$ and $|2\rangle\leftrightarrow|3\rangle$ in the atom. Both these resonances are observed in $P(\omega)$, at $\omega=\omega_{13}-\omega_L$ and $\omega_{23}-\omega_L$. It should be noted that the resonances in the linear term $P_L(\omega)$ have dispersive character. Hence the intensity fluctuations provide spectroscopic information of the atomic system. We emphasize here that the merits of the arguments presented in this paper are the simplicity of the approach and its general applicability. Our scheme exploits the fact that the linear susceptibility has all the atomic-level information built into it and that a fluctuating laser source can be used to exact that information. The analysis presented here is extendable to other complicated atomic and molecular systems and shows that, contrary to the usual belief, laser noise can be used for doing spectroscopy. Finally, we note that related earlier works [11] have utilized laser noise to determine atomic dephasing times in the context of four-wave-mixing schemes.

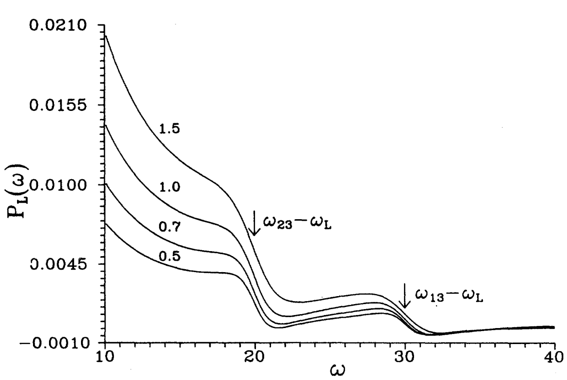

FIG. 3. Linear contribution $P_L(\omega)$ to the power spectrum of the intensity-intensity correlations for the same parameters as in Fig. 2.

This collaboration was supported by the National Science Foundation Grant No. INT-9100685. G.V. also acknowledges support from Purdue University. G.S.A. and G.V. acknowledge very useful correspondence with Professor J. Cooper.

APPENDIX

Analytic expressions representing various contributions to the power spectrum are

$$P_0(\omega)=\frac{2\gamma_c/\pi}{\omega^2+4\gamma_c^2}\ ,$$

$$\begin{aligned}P_L(\omega)=\frac{4\gamma_c^2 i}{\pi}&[2i\gamma_c\omega(\omega+2i\gamma_c)]^{-1}\{(\omega_A-i\gamma_c)[\omega_A-i(\gamma_c+\gamma_{13})]^{-1}[\omega_A-i(\gamma_c-\gamma_{13})]^{-1}\\&+(\omega_B-i\gamma_c)[\omega_B-i(\gamma_c+\gamma_{13})]^{-1}[\omega_B-i(\gamma_c-\gamma_{23})]^{-1}\}\\&+[2i\gamma_c\omega(\omega-2i\gamma_c)]^{-1}\{(\omega_A+\omega-i\gamma_c)[\omega+\omega_A-i(\gamma_c+\gamma_{13})]^{-1}[\omega+\omega_A-i(\gamma_c-\gamma_{13})]^{-1}\\&\qquad+(\omega_B+\omega-i\gamma_c)[\omega+\omega_B-i(\gamma_c+\gamma_{23})]^{-1}[\omega+\omega_B-i(\gamma_c-\gamma_{23})]^{-1}\}\\&-\tfrac{1}{2}\{[\omega_A+i(\gamma_c+\gamma_{13})]^{-1}[\omega_A-i(\gamma_c-\gamma_{13})]^{-1}[\omega+\omega_A+i(\gamma_c+\gamma_{13})]^{-1}\\&\quad\times[\omega+\omega_A-i(\gamma_c-\gamma_{13})]^{-1}+[\omega_B+i(\gamma_c+\gamma_{23})]^{-1}[\omega_B-i(\gamma_c-\gamma_{23})]^{-1}\}+(\text{terms with }\omega\to-\omega)\ ,\end{aligned}$$

$$\begin{aligned}P_Q^{(1)}(\omega)=\frac{2\gamma_c^2 i}{\pi}(&[2i\gamma_c\omega(\omega+2i\gamma_c)]^{-1}\{[\omega_A-i(\gamma_c+\gamma_{13})]^{-1}+[\omega_B-i(\gamma_c+\gamma_{13})]^{-1}\}\\&\times\{[\omega_A-i(\gamma_c-\gamma_{13})]^{-1}+[\omega_B-i(\gamma_c-\gamma_{23})]^{-1}\}\\&+[2i\gamma_c\omega(\omega-2i\gamma_c)]^{-1}\{[\omega+\omega_A-i(\gamma_c+\gamma_{13})]^{-1}+[\omega+\omega_B-i(\gamma_c+\gamma_{23})]^{-1}\}\\&\times\{[\omega+\omega_A-i(\gamma_c-\gamma_{13})]^{-1}+[\omega+\omega_B-i(\gamma_c-\gamma_{23})]^{-1}\}\\&\quad+[\omega_A+i(\gamma_c+\gamma_{13})]^{-1}[\omega_A-i(\gamma_c-\gamma_{13})]^{-1}[\omega+\omega_A+i(\gamma_c+\gamma_{13})]^{-1}\\&\quad\times[\omega+\omega_A-i(\gamma_c-\gamma_{13})]^{-1}\{[2i\gamma_{13}]^{-1}+[\omega_A-\omega_B+i(\gamma_{23}+\gamma_{13})]^{-1}\}\\&+[\omega_B+i(\gamma_c+\gamma_{23})]^{-1}[\omega_B-i(\gamma_c-\gamma_{23})]^{-1}[\omega+\omega_B+i(\gamma_c+\gamma_{23})]^{-1}\\&\quad\times[\omega+\omega_B-i(\gamma_c-\gamma_{23})]^{-1}\{[2i\gamma_{23}]^{-1}+[\omega_B-\omega_A+i(\gamma_{23}+\gamma_{13})]^{-1}\})+(\text{terms with }\omega\to-\omega)\ ,\end{aligned}$$

$$\begin{aligned}P_Q^{(2)}(\omega)=\frac{2\gamma_c^2 i}{\pi}(&-[2i\gamma_c\omega(\omega+2i\gamma_c)]^{-1}\{[\omega_A-i(\gamma_c+\gamma_{13})]^{-1}+[\omega_B-i(\gamma_c+\gamma_{23})]^{-1}\}\\&\times\{[\omega-\omega_A+i(\gamma_c-\gamma_{13})]^{-1}+[\omega-\omega_B+i(\gamma_c-\gamma_{23})]^{-1}\}\\&+[2i\gamma_c\omega(\omega-2i\gamma_c)]^{-1}\{[\omega_A-i(\gamma_c-\gamma_{13})]^{-1}+[\omega_B-i(\gamma_c-\gamma_{23})]^{-1}\}\\&\times\{[\omega+\omega_A-i(\gamma_c+\gamma_{13})]^{-1}+[\omega+\omega_B-i(\gamma_c+\gamma_{23})]^{-1}\}\\&+[\omega-\omega_A-i(\gamma_c+\gamma_{13})]^{-1}[\omega-\omega_A+i(\gamma_c-\gamma_{13})]^{-1}[\omega_A-i(\gamma_c-\gamma_{13})]^{-1}\\&\times[\omega_A+i(\gamma_c+\gamma_{13})]^{-1}\{[-\omega+2i\gamma_{13}]^{-1}+[\omega_A-\omega_B-\omega+i(\gamma_{23}+\gamma_{13})]^{-1}\}\\&+[\omega-\omega_B-i(\gamma_c+\gamma_{23})]^{-1}[\omega-\omega_B+i(\gamma_c-\gamma_{23})]^{-1}[\omega_B-i(\gamma_c-\gamma_{23})]^{-1}\\&\quad\times[\omega_B+i(\gamma_c+\gamma_{23})]^{-1}\{[\omega+2i\gamma_{23}]^{-1}+[\omega_B-\omega_A-\omega+i(\gamma_{23}+\gamma_{13})]^{-1}\})\ ,\end{aligned}$$

where $\omega_A=\omega_{13}-\omega_L$, $\omega_B=\omega_{23}-\omega_L$, and $P_0(\omega)$ is the zeroth-order contribution that can be dropped. The terms with difference of decays can be shown to cancel out in each contribution.

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