Comment on "Discrepancies in the resonance-fluorescence spectrum calculated with two methods"

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There are two alternative methods used in the literature to calculate the incoherent part of the spectrum of light scattered by an atomic system. In the first, one calculates the spectrum of the total light scattered by the system and obtains the incoherent part by subtracting the coherent part. In the second method, one introduces the fluctuation operators and obtains the incoherent part of the spectrum by taking the Fourier transform of the two-time correlation function of the fluctuation operators. These two methods have been recognized for years as completely equivalent for evaluating the incoherent part of the spectrum. In a recent paper, Xu *et al.* [Phys. Rev. A **78**, 013407 (2008)] showed that there are discrepancies between the incoherent parts of the stationary spectrum of a three-level Λ-type system calculated with these two methods. The predicted discrepancies can be severe that over a wide range of the Rabi frequencies and atomic decay rates, the spectrum calculated with the variance method can have negative values. In this Comment, we show that there are no discrepancies between these two methods. We show the equivalence of these two methods that leads to the same incoherent spectra which are positive for all frequencies independent of values of the parameters involved. We also identify the source of the discrepancy, that is, in an incorrect treatment of the incoherent part of the spectrum calculated with the two-time correlation function of the fluctuation operators.

The incoherent part of the fluorescence spectrum of an atomic system driven by laser fields is commonly studied with two, often called "limit" and "variance," methods. In the limit method, one calculates the total spectrum from which the coherent part is subtracted to obtain the incoherent part alone. In the variance method, one defines the fluctuation operators and directly obtains the incoherent part of the spectrum by a Fourier transformation of the two-time correlation function of the fluctuation operators. These methods are generally recognized as completely equivalent and have been used for decades to calculate spectra of the stationary electromagnetic field radiated by variety of atomic systems [1–8]. The variance method has been particularly popular in the calculations of the incoherent spectrum of nonstationary fields due to some difficulties in the treatment of the coherent part of the spectrum [9–16].

In a recent paper, Xu *et al.* [17] calculated the incoherent spectrum of the fluorescence field emitted by a three-level Λ system driven by two laser fields and claimed that these two methods produce different results, and the method of the calculation of the spectrum involving the fluctuation operators may even lead to negative values of the spectrum. This statement, however, is incorrect. In this Comment we clarify this point, directly showing that both methods lead to the same incoherent part of the spectrum and the negative values of the spectrum simply arise from an incorrect treatment by Xu *et al.* [17] of the variance method. Specifically, the explanation of the source of the discrepancies given by Xu *et al.* is based on the general solution of the set of differential equations describing the time evolution of the system. Therefore, it is necessary to show that there are no discrepancies

between these alternative methods since similar discrepancies could be found in other systems described by a similar set of differential equations.

As it is well known, the spectrum of the fluorescence light emitted by an atomic system is obtained by the Fourier transformation of the two-time correlation function of the emitted electromagnetic field that is usually expressed in terms of the two-time correlation function of the atomic operators. In the case of the Λ-type atom considered by Xu *et al.* [17], the spectrum of the fluorescence light emitted on the atomic $|3\rangle - |1\rangle$ transition can be written as

$$S_T(\omega) = \gamma_1 u(\vec{r}) \mathrm{Re} \int_0^\infty d\tau e^{-i(\omega-\omega_1)\tau} \lim_{t\to\infty} \langle \sigma_{31}(t+\tau)\sigma_{13}(t)\rangle,$$

(1)

where γ_1 is the spontaneous emission rate of the transition, $u(\vec{r})$ is a geometrical factor corresponding to the radiation pattern of the dipole moment, and the expectation values of the atomic dipole operators are evaluated in the rotating frame oscillating with the frequency ω_1 of the atomic transition involved.

Usually the scattered field is composed of a coherent component, corresponding to a field elastically scattered by the source atoms, and an incoherent (noise) component, corresponding to a field produced by fluctuations of the atomic dipoles. Therefore, it is often distinguished between these two contributions to the scattered field by expressing an atomic dipole operator as the sum of its expectation value and its fluctuations so that

$$\sigma_{ij}(t) = \langle \sigma_{ij}(t)\rangle + \Delta\sigma_{ij}(t),$$

(2)

where $\langle \Delta\sigma_{ij}(t)\rangle = 0$ by definition. In terms of the fluctuation operators the spectrum is of the form

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$$S_{in}(\omega) = \gamma_1 u(\vec{r}) \mathrm{Re} \int_0^\infty d\tau e^{-i(\omega-\omega_1)\tau} \lim_{t\to\infty} \langle \Delta\sigma_{31}(t+\tau)\Delta\sigma_{13}(t)\rangle.$$

$$(3)$$

We illustrate the equivalence of the two methods by considering the general case of systems whose dynamics are described by a set of inhomogeneous linear differential equations

$$\frac{d}{dt}\vec{X}(t) = \mathbf{Q}\vec{X}(t) + \vec{R}, \qquad (4)$$

where $\mathbf{Q}$ is a finite-dimensional $n \times n$ matrix of time-independent coefficients, Rabi frequencies, damping rates, and detunings, involved in the dynamics of a given system, $\vec{X}(t)$ is a column vector of the one-time correlation functions, and $\vec{R}$ is a column vector of inhomogeneous terms. The dimension n of the vector $\vec{X}$ is equal to the dimension of the Hilbert space of a given system. The general case includes, as a special case, the dynamics of Λ-type atom driven by two laser fields, considered by Xu *et al.* [17] for which $n=8$.

We first find the steady-state solution of Eq. (4), which is obtained by taking the limit of $t\to\infty$, or more directly by setting the left-hand side of Eq. (4) equal to zero. Thus

$$\vec{X}(\infty) = -\mathbf{Q}^{-1}\vec{R}. \qquad (5)$$

Next, we consider two-time correlation functions involved in the calculation of the spectrum of a given system. These are simply found by applying the quantum regression theorem to Eq. (4) which states that for $\tau > 0$ the two-time correlation functions satisfy the same equations of motion as the one-time averages [18]. Thus, we find that

$$\frac{d}{d\tau}\vec{Y}(t,\tau) = \mathbf{Q}\vec{Y}(t,\tau) + \langle X_i(t)\rangle\vec{R}, \qquad (6)$$

where $\vec{Y}(t,\tau)$ is a column vector of components $\langle X_i(t)X_1(t+\tau)\rangle, \langle X_i(t)X_2(t+\tau)\rangle,\dots$.

Using the Laplace transform, we can transform Eq. (6) into a set of algebraic equations, which we can solve by the matrix inversion. Thus, applying the Laplace transform, we obtain

$$\vec{Y}(s) = (s\mathbf{I}-\mathbf{Q})^{-1}\vec{Y}(0) + [(s\mathbf{I})(s\mathbf{I}-\mathbf{Q})]^{-1}\langle X_i(t)\rangle\vec{R}, \quad (7)$$

where $\vec{Y}(s)$ is a column vector composed of the Laplace transforms of the correlation functions, s is a complex (Laplace transform) parameter, and $\mathbf{I}$ is the unit diagonal matrix.

We can rewrite Eq. (7) in the form

$$\vec{Y}(s) = (s\mathbf{I}-\mathbf{Q})^{-1}[\vec{Y}(0) + \mathbf{Q}^{-1}\langle X_i(t)\rangle\vec{R}] - (s\mathbf{I})^{-1}\mathbf{Q}^{-1}\langle X_i(t)\rangle\vec{R},$$

$$(8)$$

which, in the steady-state limit of $t\to\infty$, takes the form

$$\vec{Y}(s) = (s\mathbf{I}-\mathbf{Q})^{-1}\Delta\vec{Y}(0) + (s\mathbf{I})^{-1}\langle X_i(\infty)\rangle\vec{X}(\infty), \qquad (9)$$

where $\Delta\vec{Y}(0) = \vec{Y}(0) - \langle X_i(\infty)\rangle\vec{X}(\infty)$.

As we see from Eq. (9) that the Laplace transform of the two-time correlation functions is composed of two terms, one corresponds to contributions of terms with $s\neq 0$ that reflect the presence of incoherent-scattering resonances and the other contribution from the pole at $s=0$ that reflects the coherent scattering peak. This term is proportional to the delta function centered at the driving field frequency since the inverse Laplace transform of the term proportional to $(s\mathbf{I})^{-1}$ gives the delta function type contribution.

We now show that the first term on the right-hand side of Eq. (9) is equal to that obtained for the correlation functions of the fluctuation operators, i.e., leads to the same correlation function that is involved in the variance method of the calculation of the incoherent part of the spectrum. This will constitute that both methods of calculating the incoherent part of the spectrum are completely equivalent. We define a column vector of average values of the fluctuation operators $\Delta\vec{X}(t)$ whose the time evolution is determined by the homogeneous equation of motion

$$\frac{d}{dt}\langle\Delta\vec{X}(t)\rangle = \mathbf{Q}\langle\Delta\vec{X}(t)\rangle, \qquad (10)$$

from which we find, by applying the quantum regression theorem, that the vector $\Delta\vec{Y}(t,\tau)$ of the two-time correlation functions satisfies an equation of motion

$$\frac{d}{d\tau}\Delta\vec{Y}(t,\tau) = \mathbf{Q}\Delta\vec{Y}(t,\tau), \qquad (11)$$

where the components of the vector $\Delta\vec{Y}(t,\tau)$ are $\langle\Delta X_i(t)\Delta X_1(t+\tau)\rangle, \langle\Delta X_i(t)\Delta X_2(t+\tau)\rangle,\dots$.

Taking the Laplace transform of Eq. (11), we find

$$\Delta\vec{Y}(s) = (s\mathbf{I}-\mathbf{Q})^{-1}\Delta\vec{Y}(0), \qquad (12)$$

which is equal to the first term on the right-hand side of Eq. (9). Thus, the limit and variance methods both lead to the same solutions for the correlation functions involved in the definition of the incoherent part of the spectrum. This statement is true for an arbitrary system whose time evolution is described by the set of differential Eq. (4).

We now proceed to apply the above procedure to the case considered by Xu *et al.* [17], the incoherent fluorescence spectrum of Λ-type atom driven by two laser fields. In this case, the initial vector $\Delta\vec{Y}(0)$ is of the form

$$\Delta\vec{Y}(0) = \{-\langle\sigma_{13}\rangle_s^2, \langle\sigma_{33}\rangle_s - \langle\sigma_{31}\rangle_s\langle\sigma_{13}\rangle_s, -\langle\sigma_{23}\rangle_s\langle\sigma_{13}\rangle_s,$$
$$-\langle\sigma_{32}\rangle_s\langle\sigma_{13}\rangle_s, -\langle\sigma_{12}\rangle_s\langle\sigma_{13}\rangle_s, \langle\sigma_{23}\rangle_s$$
$$-\langle\sigma_{21}\rangle_s\langle\sigma_{13}\rangle_s, \langle\sigma_{13}\rangle_s - \langle\sigma_{11}\rangle_s\langle\sigma_{13}\rangle_s, -\langle\sigma_{33}\rangle_s\langle\sigma_{13}\rangle_s\},$$

$$(13)$$

and the matrix $\mathbf{Q}$ is of the same form as that given in Eq. (7) of Xu *et al.* The subscript s appearing in Eq. (13) stands for the steady-state values of the average atomic dipole operators.

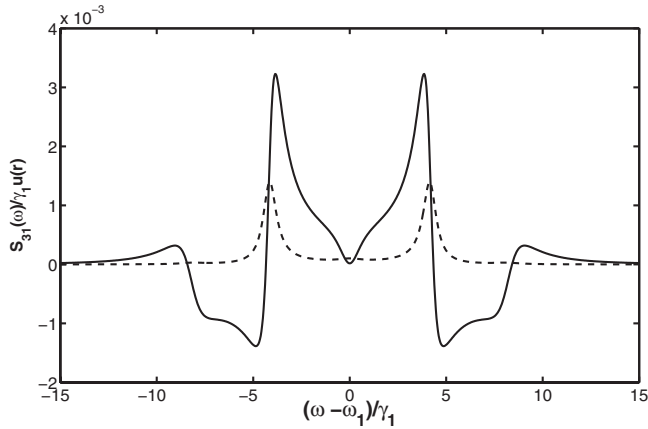However, according to the results of Xu *et al.*, the initial vector is of the form

FIG. 1. The incoherent part of the fluorescence spectrum $S_{31}(\omega)$ calculated with the variance method for the same parameters as in Fig. 2(b) of Ref. [17], and two different initial vectors $\Delta\vec{Y}_{Xu}(0)$ (solid line) and $\Delta\vec{Y}(0)$ (dashed line).

$$\Delta\vec{Y}_{Xu}(0) = \{0, \langle\sigma_{33}\rangle_s, 0, 0, 0, \langle\sigma_{23}\rangle_s, \langle\sigma_{13}\rangle_s, 0\}. \quad (14)$$

Comparing Eqs. (13) and (14), we see that the difference between our initial vector and that obtained by Xu *et al.* is in ignoring the product terms $\langle\sigma_{ij}\rangle_s\langle\sigma_{lk}\rangle_s$.

To resolve the problem of which initial vector should be used in the calculations, we plot in Fig. 1 the incoherent part

of the fluorescence spectrum $S_{31}(\omega)$ calculated numerically with the variance method for the same parameters as in Fig. 2(b) of Ref. [17]. Note that this is the figure where the most prominent negative values of the spectrum have been predicted. We plot the spectrum by taking the Laplace transform of Eq. (11) and solving numerically the resulting set of algebraic equations by matrix inversion for the two different initial vectors $\Delta\vec{Y}(0)$ and $\Delta\vec{Y}_{Xu}(0)$ [19].

Evidently, with the initial vector $\Delta\vec{Y}(0)$, the spectrum is positive for all frequencies and exhibits Lorentzian-type structures at the Rabi sidebands, that is, the incoherent spectrum is the same as that calculated with the limit method, shown as a dotted line in Fig. 2(b) of Ref. [17]. With the initial vector $\Delta\vec{Y}_{Xu}(0)$, we recover the spectrum with negative values predicted by Xu *et al.* [17]. Thus, the source of the discrepancy between the two methods is in the error in the initial vector derived by Xu *et al.*

In summary, we have presented a simple analysis against the statement of Xu *et al.* [17] that the incoherent part of the fluorescence spectrum of a three-level Λ-system calculated with the variance method can have negative values. The analysis clearly show that the incoherent part of the spectrum calculated with the variance method is positive for all frequencies independent of the system considered and values of the parameters involved. We have also identified the error in the calculations of Xu *et al.* that led to the discrepancies between the two methods.

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