# Reply to “Comment on ‘Nonlinear fluctuations and dissipation in matter revealed by quantum light’ ”

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# Reply to Comment on “Nonlinear fluctuations and dissipation in matter revealed by quantum light”

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We discuss the proper definition of the term”quantum spectroscopy”. Since the response of matter to coherent states of the field is not necessarily classical, positivity of the Glauber-Sudarshan P representation, as used by M. Kira, S. Koch, R. Smith, A. Hunter, and S. Cundi, Nature Physics 7, 799 (2011), does not imply that the response is classical. We show that classical and quantum spectroscopy may be unambigously distinguished by formulating the signals in terms of multipoint field correlation functions.

Quantum spectroscopy which monitors the response of matter to a quantum optical fields provides many types of control knobs (parameters of the photon wavefunction) that may be used to generate new signals, thus offering a unique novel window into the matter response [1–11]. In this reply we address two issues raised in the comment [12]

1. How should the term “quantum spectroscopy” be defined?
2. How should quantum spectroscopy signals be calculated?

In a series of publications the Marburg group [7, 10, 13] had referred to spectroscopy performed with coherent states of light as “classical”, whereas signals obtained with true quantum light (an ill defined term) have been labeled “quantum spectroscopy”. Their starting point is the relation

$$R_{QM} = \int d^2\beta P(\beta) R_{|\beta\rangle}. \qquad (1)$$

Here the response to a coherent state $|\beta\rangle$ $R_{|\beta\rangle}$ represents the classical response, $R_{QM}$ is the quantum response and $P(\beta)$ is the Glauber-Sudarshan $P$ distribution which represents the field density matrix. Taking this terminology literally, and making the obvious assumption that classical spectroscopy refers to signals obtained with classical light, raises some difficulties with the Marburg results which have been pointed out in [11]. Eq. (1) implies that the underlying matter information in quantum spectroscopy is the same as in the classical field case. The outcomes of a complete set of experiments performed with various classical fields can thus be combined to generate the quantum response. In that picture the quantum field simply provides a different gating window for the classical response function (CRF). A complete knowledge of the CRF is enough to compute the response to any quantum field, and the quantum response function (QRF) may be then recovered from the CRF by simple data processing. If correct, this makes quantum spectroscopy less exciting since it does not carry fundamentally new matter information.

We had argued in [11] that this picture is false. CRF, which by definition, describe the response of a quantum system to classical fields, are causal; the field affects the system but the system does not affect the field. The situation is fundamentally different when two quantum systems (matter and field in our case) interact. Now the response and classical fluctuations of both systems mix, and causality is violated. The bottom line is that matter information other that the CRF is provided by quantum signals and consequently data processing of CRF is not enough to account for quantum spectroscopy. We had further argued that the fact that quantum spectroscopy signals may not be predicted from classical spectroscopy as suggested by Eq. (1) is intimately related to the absence of a nonlinear fluctuation-dissipation theorem: spontaneous fluctuations and response are uniquely related only in the linear regime, but not when they are nonlinear.

In their comment [12] the authors now revise the terminology of $R_{|\beta\rangle}$ from “classical” to “coherent state spectroscopy”. This is a welcome improvement. They correctly point out that coherent states are quantum and can sometimes give signals different from classical light. They now define spectroscopy as “classical” when the $P$-representation is positive definite. This is a legitimate but not very practical or useful definition. They justify this by arguing that classical fields are never realized exactly in laser spectroscopy experiments that typically use coherent states. We take issue with their following statement:

> “In this Comment, we show that this interpretation is correct only when classical spectroscopy is perceived as a theoretical description which neglects quantum fluctuations of light altogether. While such an assumption can be a good approximation and useful for comparing theoretical results, it is never realized exactly in laser spectroscopy experiments that typically use coherent states.”

It is well established that the vast majority of quantum optics experiments can be fully understood by treating

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the fields as classical. Many effects and signals can be perfectly understood by using the Bloch equations where the field is classical. There are of course examples where coherent states yield different results than classical fields. Indeed the experiments of Rempe [14] demonstrate the revival in the damped excited state probability due to the effect of quantum field - coherent state, which are missed by classical light. The Mollow resonance fluorescence spectrum [15] is another example. Saying that classical fields are never realized and that this is an artificial invention of theorists is factually incorrect. In fact, the pump-probe experiments which form the basis for the quantum spectroscopy applications of the Marburg group can be completely described with classical fields and do not carry any quantum field information.

The Glauber-Sudarshan $P$-representation implies that the field density matrix can be represented as a sum over coherent state density matrices. This implies that the response is a sum over coherent state responses. With their new definition of "classical", Eq. (1) thus merely expands quantum spectroscopy in a coherent state basis. It does not connect a classical and a quantum signal. It is highly misleading to define quantum spectroscopy as opposed to coherent state spectroscopy since both are quantum. Quantum calculations can be carried out in any basis set for the field. Coherent states constitute an overcomplete basis so that Eq. (1) should be interpreted as quantum calculations performed in a specific basis set. This is obviously true and needs no further proof .The following statement they make then becomes trivial:

> "We rigorously show that quantum spectroscopy can always be projected from the experimentally realized coherent-state spectroscopy regardless how nonlinear the system response is."

This is tantanomous to saying that the quantum field can be described by the coherent state basis. This is an undisputed fact that needs no further proof. It is of course possible to expand the response using this representation. Eq. (1) is therefore merely a straightforward expansion of the quantum response in a specific basis set.

In summary of our first point, we argue that it makes sense to label spectroscopy performed with classical light (rather that with coherent states) as classical.This definition is simple and well established both experimentally and theoretically and is independent on a specific basis set such as the P representation.Using this as a reference,any signal that cannot be described by the corresponding classical response functions should then naturally be labeled as quantum spectroscopy.

Leaving semantics aside, a more important issue is our second point: how to calculate quantum spectroscopy signals. The authors say

> "Therefore, it is interesting to determine whether spectroscopy performed with semi-classical vs. true quantum sources are connected."

This issue has been fully addressed and resolved in our earlier work [4]. In the spirit of Glauber's formalism, we had developed a general way to calculate quantum spectroscopy signals by expanding the response using multipoint correlation functions of the field [16]. These can then be calculated using the entire arsenal of quantum optics methods including quantum master equations and the Heisenberg-Langevin approach [17]. This formulation is not limited to a particular basis set. The expansion in coherent states is only one option. To extend Glauber's approach to spectroscopy we had used time ordered products of superoperators rather than normally ordered ordinary operators. In four wave mixing, the relevant multipoint functions are a natural generalization of Glauber's $g^{(2)}$ that depend on four rather than two time variables [18]. The correlation function formalism gives an unambiguous clear cut and practical answer to the question posed in the above quote. Classical spectroscopy is obtained by factorizing these correlation functions into field amplitudes. This is a much simpler and more transparent definition compared to that based on the positivity of the $P$-representation. It makes sense to denote the experimentally readily accessible response to classical fields as classical. In that case the normally ordered field correlation functions factorize into classical amplitudes. Quantum field effects can then naturally show up via two mechanisms:

i Expectation values of normally ordered products can have unusual time/frequency correlations stemming from the quantum state of light that correlates different field modes.

ii Non-normally ordered products can be brought into normal form by adding field commutators. These provide additional quantum contributions to the response.

The CRF is one specific combination of matter correlation functions. The quantum response is given by other combinations. An interesting question is how useful is the $P$-representation for practical calculations. This representation can be highly singular. It often suffers from numerical difficulties, diverges in some cases, and will be very hard to implement for multimode fields. The applications presented so far by the Marburg group are limited to a single mode; multiple broadband calculations of the $P$-representation will be extremely tedious and unnecessary. In contrast, we had calculated the multipoint correlation functions for realistic pulses using Heisenberg algebra, singular value decomposition for the quantum state amplitude and input-output relations for the field operators [19]. Quantum spectroscopy is very exciting since it carries information regarding quantum fluctuations that is not available from the classical response. This is obvious from the correlation function representation.

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