Detection of photon statistics and multimode field correlations by Raman processes

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ABSTRACT

Glauber’s $g^{(2)}$-function provides a common measure of quantum field statistics through two-photon coincidence counting in Hanbury Brown–Twiss measurements. Here, we propose to use nonlinear optical signals as a tool for the characterization of quantum light. In particular, we show that Raman measurements provide an alternative direct probe for a different component of the four-point correlation function underlying the $g^{(2)}$-function. We illustrate this capacity for a specific quantum state obtained from a frequency conversion process. Our work points out how the analysis of controlled optical nonlinear processes can provide an alternative window toward the analysis of quantum light sources.

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The photon counting formalism developed by Glauber lies at the very heart of quantum optics. It has been instrumental in the characterization of light—whether it is quantum fluctuations of light fields, the particle nature of Fock states, or the violation of Bell’s inequality for photons. On a formal level, photon correlation measurements characterize quantum fields in terms of normally ordered multipoint functionals of the form $\langle T E_1(\tau_1) E_2(\tau_2) E_3(\tau_3) E_4(\tau_4) \rangle$, where $T$ denotes the time-ordering operator and $E_1, \ldots, E_4$ are the various light fields in the signal, which are convoluted with the sample’s nonlinear response function. The time ordering—in combination with “filter” of the matter system—yields different types of correlation functions, depending on the chosen experimental setup. These include the normally ordered correlation functions of Glauber’s theory, which give rise to the $g^{(2)}$-measurement, for instance, in two-photon absorption measurements. Consequently, such measurements may equally reveal information on the incident photonic state, such as time-energy entanglement, provided that the sample
The broadband, ultrafast fields. We thus write the dimensionless field formalism. We shall be concerned with the characterization of information, which is not accessible in the standard photon counting experiments.

To set the stage, let us first briefly outline the photon counting formalism. We shall be concerned with the characterization of transition probabilities in terms of polarizabilities, where

\[ P_{gg'}(t; \Gamma) = \left( \frac{i}{\hbar} \right)^4 \int_0^{\tau_2} dt_2 \int_0^{\tau_1} dt_1 \int_0^{\tau_1} dt_{11} \int_0^{\tau_{11}} dt_{111} \times \left( V(\tau_1) V^\dagger(\tau_2) P_{gg'}(t) V(\tau_2) V^\dagger(\tau_1) \right) \]

where \( V \) denotes the positive-frequency component of the material dipole operator, describing the annihilation of excitations in the material, and \( E^\dagger \) denotes the negative-frequency component of the electric field operator, corresponding to the creation of photons. In writing Eq. (3), we have also employed the rotating wave approximation. To leading order perturbation theory in the light–matter interaction, the population in \( g' \) is given by (see, e.g., Ref. 22)

\[ P_{gg'}(t; \Gamma) = \left( \frac{i}{\hbar} \right)^4 \int_0^{\tau_2} dt_2 \int_0^{\tau_1} dt_1 \int_0^{\tau_1} dt_{11} \int_0^{\tau_{11}} dt_{111} \times \left( E^\dagger(\tau_1) E(\tau_2) P_{gg'}(t) E(\tau_2) E^\dagger(\tau_1) \right). \]

where \( P_{gg'} = |g'\rangle \langle g'| \) is the projector onto state \( g' \). We consider off-resonant Raman transitions, in which case we may express the transition probabilities in terms of polarizabilities.
The population in $p_E$ may be expressed as the modulus square of transition amplitude operators acting on the field Hilbert space, $p_E(t;\Gamma) = \langle \hat{T}_{g'g}(t) \rangle^2$, with the transition amplitude operator

$$T_{g'g}(t) = -\frac{i\alpha_{gg'}}{\hbar} \int_0^t d\tau e^{i\omega_{gg'}\tau} E_g^*(\tau)E_{g'}(\tau),$$

where $\alpha_{gg'}$ denotes the polarizability of the $g \to g'$ transition and $\omega_{gg'}$ denotes its transition frequency. We only consider times $t$ long after the pulses have passed through the sample. In this case, the populations are constant, and we can extend the time integration in Eq. (7) to infinity.\textsuperscript{\textendash}\textsuperscript{26} Decomposing the two fields in the Schmidt modes introduced above, the transition amplitude simplifies to

$$T_{g'g}(t) = \sum_{k,k'} d_{kk'} A_k^* B_{k'},$$

with

$$d_{kk'} = -\frac{i\alpha_{gg'}}{\hbar} \int_{-\infty}^\infty d\tau e^{i\omega_{gg'}\tau} \psi_{A_k}^*(\tau)\psi_{B_{k'}}(\tau).$$

It follows from Eq. (9) that Raman scattering mixes different modes in beams $A$ and $B$. This can be simplified by making further assumptions: by choosing a basis in $B$, which is shifted by $\omega_{gg'}$ with respect to $A$,

$$\psi_{A_k}^*(t) = \psi_{B_k}(t) e^{-i\omega_{gg'}t},$$

the time integration simply yields a delta-function, $d_{kk'} = -\frac{i\omega_{gg'}}{\hbar} \delta_{kk'}$. Hence, apart from an irrelevant prefactor, the Raman transition may be described by the two-photon field operator

$$\hat{T} = \sum_k A_k^* B_k.$$  \hfill (11)

In addition, we require the phonon frequency $\omega_{gg'}$ to be larger than the bandwidth of the pulses such that the field operators commute, $[A_k, B_k^+] = 0$. The basis $\psi_{A_k}^*$ may still be chosen such as to simplify the description of a particular quantum state. Yet once we pick this basis, it also fixes the corresponding basis for field $B$. The anti-Stokes process, in which a phonon is destroyed, $\sim A_k^* B_k^+$ can be neglected since the vibrational state is not excited thermally.

In contrast to intensity measurements, Eq. (1), Raman signals correlate the corresponding high-energy destruction of a photon in mode $B$ (i.e., the positive frequency component of mode $B$) with a photon creation in mode $A$ (with its negative frequency component). To appreciate the difference, let us first calculate the expectation value of the Raman transition operator $T$ when both fields $A$ and $B$ are in coherent states and temporal amplitudes $a_{A/B}(t)$. By expanding these amplitudes in terms of the eigenmodes $\phi_{A/B}^{(2)}$, we find

$$\langle T \rangle_{coh} = \int dt a_A^*(t)a_B(t).$$  \hfill (12)

Hence, the Raman transition operator quantifies the overlap between two fields’ modes. In contrast to Eq. (1), it is inherently phase-dependent. The introduction of a time delay between the two fields will quickly erode this overlap. In the following, we turn to question of how this phase-dependence is reflected in Raman-based photon correlation measurements.

The vibrational population created by the Raman transition is described by the modulus square of the Raman operator (11), which is normalized by the two fields’ mean photon numbers,

$$\delta_{coh}^{(2)} = \frac{\langle T^\dagger T \rangle}{\langle \sum_k A_k^* A_k \rangle \langle \sum_k B_k^* B_k \rangle} = 1 + \frac{\sum_{k,k'} \langle A_k^* B_{k'} \rangle \langle A_{k'}^* B_k \rangle}{\langle \sum_k A_k^* A_k \rangle \langle \sum_k B_k^* B_k \rangle}.$$  \hfill (13)

In the second line, Eq. (14), we have normally ordered the numerator of Eq. (13). Consequently, the first term stems from the field commutator, and the second term stems from the normally ordered contribution. We find that the off-resonant Raman detection (11) yields a different pairing of operators $A_k$ and $B_k$ in the second term of Eq. (14) compared to two-photon counting (2). This unusual pairing correlates positive frequency contributions from one field with negative frequency parts of the other field, which in turn has important consequences for the information about the quantum state of light that can be obtained with this type of measurement.

To illustrate the information provided by Raman signals compared to conventional photon counting, we now construct input states, which will then be analyzed with either photon counting or Raman measurements. In particular, we consider two fields created by frequency conversion (FC).\textsuperscript{27} An initial state $|\phi\rangle_A$ in beam $A$ interacts with a (narrowband) pump pulse via a $\chi^{(2)}$-nonlinearity to populate a new field $C$, which is shifted by the pump frequency with respect to the initial state, to create the output state $|\phi_{out}\rangle = \exp(-iH_{FC})|\phi\rangle_A$. The FC Hamiltonian reads\textsuperscript{11,27}

$$H_{FC} = \int d\omega_a \int d\omega_a f_{FC}(\omega_a, \omega_b) a_{\omega_a}^+ b_{\omega_b} + H.c.$$  \hfill (15)

Here, $a_{\omega_a}^+ (b_{\omega_a})$ denotes the photon (annihilation) creation operator in field $A$ ($B$), respectively, and $f_{FC}$ is the phase-matching function of the FC process that depends on the crystal properties and the pump field (see, e.g., the discussion in Ref. 27). The discussion of its properties becomes most transparent by using the Schmidt decomposition of the phase-matching function\textsuperscript{19}
where \( r_k > 0 \) gives the weight, with which mode \( k \) participates in the FC process, and the functions \( \{ \psi^{(+)}_A \} \) and \( \{ \psi^{(+)}_B \} \) are sets of orthonormal functions, which depend on the phase-matching conditions inside the nonlinear crystal.\(^{28,30}\) These define the basis set, for which the Raman correlation function (14) can be evaluated most easily.

With the initial state \(|\phi\rangle_A \otimes |0\rangle_B\), the output state in the weak conversion limit, when only one photon is exchanged between the two fields, may be written as

\[
|\phi_{\text{out}}\rangle = \sum_k r_k A_k |\phi\rangle_A \otimes B_k^\dagger |0\rangle_B, \tag{17}
\]

where we introduce renormalized mode weights \( r_k \), satisfying \( \langle \phi_{\text{out}} | \phi_{\text{out}} \rangle = \sum_k r^2_k |\phi_k\rangle^2 = 1 \). Note that due to the normalization, the factors \( r_k \) depend on the input state coefficients \( c_k \) [see, for example, Eq. (19) below]. This state cannot generally be factorized. However, when we calculate the conventional cross correlation function (2), we obtain

\[
\delta^{(2)}_{\text{RC}} = 1, \tag{18}
\]

regardless of the input state \(|\phi\rangle\) or the structure of the FC process. Hence, photon counting does not provide information on the properties of state (17). In contrast, the Raman correlation function does. In the following, we will consider several states of light, where the Raman correlation provides new information. Our first example will be a coherent state of the form

\[
|\phi\rangle_A = \prod_k \exp \left( c_k A_k^\dagger - c_k^\ast A_k \right) |0\rangle_A, \tag{19}
\]

with \( \sum_k |c_k|^2 = 1 \), such that \( \langle \sum_k A_k^\dagger A_k \rangle = \langle \sum_k B_k^\dagger B_k \rangle = 1 \); we arrive at

\[
\delta^{(2)}_{\text{RC}} = 1 + \left( \sum_k r_k |c_k|^2 \right)^2, \tag{20}
\]

which depends on both initial states as well as the FC process. Hence, while photon counting cannot reveal any properties of the frequency conversion process, Raman measurements do. Let us first assume that the input state (19) has a large bandwidth with \( \Delta \omega_k = 1/\sqrt{M} \) for \( k \leq M \). If the FC only affects a single mode \( k_0 \), i.e., \( r_{k_0} = \text{const.} \) and \( = 0 \) otherwise, we obtain the limiting value for the correlation function \( \delta^{(2)}_{\text{RC}} \) \( \to 1 + 1/M \). If, on the other hand, many modes participate in the FC process, \( r_k = \text{const} \) for \( k \leq M \), we obtain \( \delta^{(2)}_{\text{RC}} \to 2 \).

This behavior is illustrated in Fig. 2, where we simulate the Raman correlation function \( \delta^{(2)}_{\text{RC}} \) using a bi-Gaussian phase-matching function \( f_{\text{FC}}(\omega_a, \omega_b) = a \exp[-\omega^2/(2\sigma_a^2)] \exp[-(\Delta \omega(\omega_a, \omega_b)L/2)^2]/\sqrt{2\pi\sigma_p^2} \), where \( \Delta \omega(\omega_a, \omega_b)L = (\omega_a - \omega_1 - \omega_b)T_1 + (\omega_b - \omega_1)T_2 \) describes the phase-matching in the crystal by the two inverse bandwidths \( T_1 \) and \( T_2 \) and \( \sigma_p \) describes the pump bandwidth. In this case, the Schmidt decomposition (16) can be carried out analytically, and the eigenmodes are simply given by Hermite functions.\(^{25,26}\)

We depict the dependence of the Raman correlation function \( \delta^{(2)}_{\text{RC}} \) on the number of modes in the FC process (16) for different coherent input states. The FC process is controlled by the bandwidth \( \sigma_p \) of the pump pulse facilitating the FC, and the input state is described by \( c_k = 1/\sqrt{M} \) for \( k \leq M \). For \( M = 100 \), \( \delta^{(2)}_{\text{RC}} \) is close to two only for \( \sigma_p \times T_1 \ll 1 \), i.e., when the pump bandwidth is much smaller than that of the frequency converted pulse \( \sim 1/T_1 \). In this case, many modes contribute to the FC process. As soon as the bandwidths are comparable, \( \sigma_p \times T_1 \sim 1 \), the correlation function drops to \( \sim 1.1 \) and \( \sigma_p/T \sim 3 \) and then slowly rises again. For \( M = 10 \), the Raman correlation function \( \delta^{(2)}_{\text{RC}} \) shows a very similar behavior, yet less pronounced. It drops to 1.4 since only the behavior of the ten largest eigenvalues contributes to the signal. This may be explained by the close analogy between the FC Hamiltonian and the well studied Hamiltonian for parametric down conversion. If the pump bandwidth is much smaller than the pulse bandwidth, \( \sigma_p \ll 1/T_1 \), the outgoing light is strongly anti-correlated, while for \( \sigma_p \gg 1/T_1 \), it shows strong positive correlations.\(^{31}\) For intermediate values of \( \sigma_p \), the correlations naturally move through zero.

In our discussion so far, state (17) is separable and hence shows no quantum correlations. This is no longer the case when the input state to Eq. (17) is entangled. In this situation, we may write the input state, e.g., for the case of a type-I downconverted photon pair as

\[
|\phi\rangle_A = \frac{1}{\sqrt{2}} \sum_{k_i,k_2} d_{k_i,k_2} A_{k_i}^\dagger A_{k_2}^\dagger |0\rangle, \tag{21}
\]

where the symmetric coefficient matrix \( d_{k_i,k_2} \) stems from the decomposition of the two-photon wavefunction in the eigenbasis of the FC process [Eq. (16)]. A similar calculation to before yields

\[
\delta^{(2)}_{\text{RC}} = 1 + 2 \sum_{k_i,k_2} r_{k_i} r_{k_2} |d_{k_i,k_2}|^2. \tag{22}
\]
The Raman correlation function in this case is again controlled by the FC process as described by the coefficients $r_k$. However, it further depends on the two-photon wavefunction and hence on the entanglement of the two photons. The Raman measurement contains information on the nonseparability of state (17), which is missed by the two-photon counting measurements [see Eq. (18)].

To conclude, we have proposed that Raman measurements provide a tool for the characterization of the quantum state of multimode light and demonstrated that it offers complementary information content to the well-established photon counting formalism. As an example, we demonstrated how the multimode structure in a quantum state of light created by a frequency conversion process may be detected. We envision that such measurements might be particularly interesting for multimode, broadband fields considered here, where direct quantum state tomography becomes prohibitively expensive.

Our results point out the potential of nonlinear optical signals in the quantum regime as photon detectors: By replacing the two-level atoms of Glauber’s theory with more complex level detectors, additional information on the quantum state of the light may be extracted. The full extent to this approach to the characterization of multimode light fields has yet to be explored. The present formalism based on a quantum state that is generated as the result of $(\chi^{(3)})$ nonlinear frequency conversion. For optical wavelengths of the fields $A$ and $B$, the pump must be in the IR regime to facilitate the phase-matching. This, in principle, is limiting the spectral resolution and the degree of frequency correlations that are governed by $q_{AB}$. An alternative approach can utilize the $\chi^{(2)}$ nonlinearity in the four-wave-mixing conversion process. The case. This way all four fields are in the optical regime and the corresponding degree of correlations can be maintained with higher precision than in the $\chi^{(2)}$ case.

Conversely, the discussion of quantum properties of light in Raman transitions opens the possibility of exploiting quantum correlations in Raman spectroscopy. A connection should be established to older investigations into the role of photon fluctuations in stimulated Raman processes. The output state (17) obtained from an entangled input state (21) could be understood as an entangled state of a photon with a photon hole, and our result shows that such entanglement is reflected in Raman measurements. Future work will explore how these correlations may be tailored to control spectroscopic signals.

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### DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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