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Ultrashort coherence times in partially polarized stationary optical beams measured by two-photon absorption

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Abstract: We measure the recently introduced electromagnetic temporal degree of coherence of a stationary, partially polarized, classical optical beam. Instead of recording the visibility of intensity fringes, the spectrum, or the polarization characteristics, we introduce a novel technique based on two-photon absorption. Using a Michelson interferometer equipped with polarizers and a specific GaAs photocount tube, we obtain the two fundamental quantities pertaining to the fluctuations of light: the degree of coherence and the degree of polarization. We also show that the electromagnetic intensity-correlation measurements with two-photon absorption require that the polarization dynamics, i.e., the time evolution of the instantaneous polarization state, is properly taken into account. We apply the technique to unpolarized and polarized sources of amplified spontaneous emission (Gaussian statistics) and to a superposition of two independent, narrow-band laser beams of different mid frequencies (non-Gaussian statistics). For these two sources femtosecond-range coherence times are found that are in good agreement with the traditional spectral measurements. Although previously employed for laser pulses, two-photon absorption provides a new physical principle to study electromagnetic coherence phenomena in classical and quantum continuous-wave light at extremely short time scales.

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the reduced Planck constant), which at optical frequencies is on the order of a few femtoseconds. A pair of photons is absorbed within the Heisenberg time, \( \tau_0 \), which is on the order of a few femtoseconds.

In quantum optics it has been used to demonstrate the basic notions of bunching [5] and antibunching [6] of photons. In Young’s two-pinhole interferometer, temporal coherence is accessed with optical Holography [37], dealing with intensity rather than field correlations, has had a seminal impact on astronomy and quantum physics. Michelson-type interferometers. Hanbury Brown–Twiss interferometry [4], dealing with intensity correlations [7, 8], we choose the one utilizing two-photon absorption in a photomultiplier tube.

1. Introduction

Coherence in classical optics quantifies correlations of light fluctuations at two or more spatial points or instants of time [1, 2]. Often the random variations take place on femtosecond time scales and so are far too rapid to be directly seen by any detector. The observable quantities in optics therefore are correlation functions (time averages) [3]. Coherence affects many optical phenomena, especially those involving interference of light. While spatial coherence manifests itself in Young’s two-pinhole interferometer, temporal coherence is accessed with Michelson-type interferometers. Hanbury Brown–Twiss interferometry [4], dealing with intensity rather than field correlations, has had a seminal impact on astronomy and quantum physics. In quantum optics it has been used to demonstrate the basic notions of bunching [5] and antibunching [6] of photons.

Among a variety of techniques to measure ultrafast optical transients based on field and intensity correlations [7, 8], we choose the one utilizing two-photon absorption in a photomultiplier tube. Here for the first time we apply the technique in the context of stationary, partially polarized electromagnetic beams. Two-photon absorption is a nonlinear optical effect in which a pair of photons is absorbed within the Heisenberg time, \( \hbar / E_g \) (\( E_g \) is the bandgap energy, \( \hbar \) is the reduced Planck constant), which at optical frequencies is on the order of a few femtosec-
onds. The rate of two-photon absorption is proportional to the average of intensity squared, indicating that the phenomenon can be used to measure statistical properties of light [9, 10], such as intensity autocorrelation [11, 12]. Indeed, quite recently, the technique was employed to demonstrate femtosecond-scale photon bunching in thermal light [13], extrabunching in twin beams [14], and pulse compression by two-photon gain [15]. However, in these studies scalar light was considered omitting the polarization properties.

In this work, we report the first measurement of the time-domain degree of coherence of stationary, partially polarized electromagnetic (vector-valued) light beams. Instead of considering the visibilities of interference fringes or performing spectral measurements we employ polarization selective Michelson interferometer and two-photon absorption detection that is particularly suitable for optical fields with ultrashort coherence times. The two-photon signal results in the electromagnetic intensity correlation functions that specify two fundamental quantities pertaining to random light beams: the electromagnetic degree of coherence and the degree of polarization. The method employed in this work thus provides a completely new physical approach to find these parameters. As examples we consider, on one hand, light generated in amplified spontaneous emission (ASE) obeying Gaussian statistics and, on the other, a superposition of two independent laser beams having different center frequencies. For both types of sources the measurements yield the correct degrees of polarization and coherence times that are in good agreement with values obtained by traditional spectral measurements. The electromagnetic detection we present differs substantially from the previous works on scalar fields [13], since partially polarized light exhibits polarization dynamics [16], i.e., the instantaneous polarization state evolves in time. This has an essential contribution to the two-photon absorption signal.

2. Electromagnetic degree of coherence

A realization of a random, statistically stationary, uniformly polarized light beam at time \( t \) is given by a two-component column vector \( \mathbf{E}(t) \). It is a zero-mean complex analytic signal that plays a key role in the analysis of photodetection [1]. The temporal second-order coherence of the field is characterized by the electric mutual coherence matrix \( \Gamma(\tau) = \langle \mathbf{E}^\dagger(t)\mathbf{E}^T(t + \tau) \rangle \) [1], where \( \tau \) is a time delay (path difference in Michelson’s interferometer), the angle brackets represent time averaging, and the asterisk and symbol \( T \) denote complex conjugation and matrix transpose, respectively. The temporal degree of coherence, \( \gamma(\tau) \), associated with a vectorial random optical beam was recently introduced as [17, 18]

\[
\gamma^2(\tau) = \frac{\text{tr} [\Gamma(\tau)\Gamma(-\tau)]}{I(t)^2},
\]

where \( \text{tr} \) stands for matrix trace and \( I(t) = \mathbf{E}^T(t)\mathbf{E}^\dagger(t) \) is the instantaneous intensity of the field. The quantity \( \gamma(\tau) \) is the temporal analog of the spatial degree of coherence, which in electromagnetic context describes the strength of the modulation of the four Stokes parameters (representing intensity and polarization state) in Young’s interferometer [18–20].

The degree of coherence in Eq. (1) obeys \( 0 \leq \gamma(\tau) \leq 1 \), with the lower and upper limits identifying, respectively, complete temporal incoherence and full coherence at \( \tau \). Complete coherence throughout a temporal (or spatial) domain is equivalent with the factorization of the coherence matrix [21, 22], consistently with the celebrated results in quantum optics [23]. An estimate for the coherence time \( \tau_c \) of the beam is obtained as a time interval over which \( \gamma(\tau) \) drops from \( \gamma(0) \) to a sufficiently small value. One finds that \( \gamma^2(0) = (P^2 + 1)/2 \) [17], where \( P \) is the degree of polarization [1]. Thus, unlike with scalar fields, \( \gamma(0) \) for vectorial fields is not necessarily equal to unity, since the two orthogonal electric-field components of a partially
polarized beam are not fully correlated and do not produce maximal polarization modulation in self-interference [18].

For many light sources the electric field can be regarded as a Gaussian random process [1]. This holds for thermal and chaotic light produced by a collection of independent radiators (central limit theorem), for a single-mode laser well below the threshold [2, 24], and for laser light with at least a few independent longitudinal modes [25, 26]. The ASE source we consider falls in this category and we may apply the moment theorem [1] to write the higher-order field correlations in terms of the second-order ones. In this case, $\gamma^2(\tau)$ in Eq. (1) can be expressed as [17, 27]

$$\gamma^2(\tau) = \frac{\langle \Delta I(t) \Delta I(t + \tau) \rangle}{\langle I(t) \rangle^2} = \frac{\langle I(t) I(t + \tau) \rangle}{\langle I(t) \rangle^2} - 1,$$

where $\Delta I(t) = I(t) - \langle I(t) \rangle$ describes fluctuation of the intensity around the average value. The first expression demonstrates that in the case of Gaussian statistics the electromagnetic degree of coherence can also be understood as the correlation of intensity fluctuations at two instants of time.

Our second source is composed of two independent, fully polarized, quasi-monochromatic (single-mode) laser beams with different center frequencies and either the same or orthogonal polarizations. The lasers obey Poissonian photon statistics and the quantity $\gamma^2(\tau)$ in Eq. (1) can, for $\tau$ much smaller than the laser coherence times, be related to the intensity correlations as (see Appendix A)

$$\gamma^2(\tau) = \frac{2\langle I(t) I(t + \tau) \rangle + g\langle I(t) I(t + \tau) \rangle_{\text{min}}}{\langle I(t) I(t + \tau) \rangle_{\text{max}} + \langle I(t) I(t + \tau) \rangle_{\text{min}}} - 1,$$

where $g = 2$ if the two lasers have the same polarization state while $g = 1$ when the lasers are orthogonally polarized with the same intensity. These two cases correspond to fully polarized and completely unpolarized superposition beams, respectively. Subscripts min/max indicate the minimum/maximum values.

3. Experimental setup and two-photon absorption signal

Equations (2) and (3) imply that $\gamma(\tau)$, and the related coherence time $\tau_c$, can be found by measuring the intensity correlations as a function of $\tau$, which for $\tau = 0$ yields the degree of polarization $P$. For this purpose, in the case of temporally incoherent broadband light, femtosecond resolution is needed. This is achieved with a Michelson interferometer and a photodetector operating in two-photon absorption regime as shown in [11, 13] (see also Fig. 1). Whereas these papers deal with scalar light, we analyze partially polarized vector fields and take into account the intensity cross-correlations between orthogonal polarization components as well, i.e., we also consider the polarization dynamics of electromagnetic light. This is accomplished by using polarizers in the interferometer to prevent time evolution of the instantaneous polarization state, as will be explained shortly.

In our setup (see Fig. 1), light is guided in an optical fiber and transmitted through a beam collimator (BC). The beam is split into two with a polarization-insensitive beam splitter (BS). The beams then traverse polarizers (P), reflect from mirrors (M), are combined by the beam splitter, and focused with a lens (L) onto a GaAs photomultiplier tube (PMT). We use Hamamatsu photomultiplier H7421-50 with a band gap corresponding to wavelength $\lambda_g = 900$ nm. Thus, the two-photon absorption range is $900 \text{ nm} < \lambda < 1800 \text{ nm}$. We have experimentally verified that the response of the detector is nearly isotropic and for our measurements can be considered as fully polarization independent. The temporal resolution of the technique, $\tau_c = \lambda_g/(2\pi c)$, is below 1 fs ($c$ is the vacuum speed of light). The length of interferometer arm $b$ is tuned by...
translating the mirror with a piezoelectric transducer, operating with 100 nm steps for an overall scanning range of 2.7 cm. The path difference of the arms, $\Delta l$, determines $\tau$ in Eq. (2) as $2\Delta l = c\tau$. The light-spot diameter on the photodetector was about 5 $\mu$m. The required quadratic dependence of the photocurrent $J$ on the input optical power, indicating two-photon absorption, was obtained for powers less than 2 mW. On the other hand we could not obtain a proper signal at powers $< 100$ nW because of the thermal background noise. Hence, in all our experiments, the input power was set to be on the order of 100 $\mu$W.

We employ two light sources to demonstrate the technique. The first is amplified spontaneous emission (ASE) of an Er-doped fiber amplifier. The second is constructed by combining with a polarizing beam splitter two independent, orthogonally polarized, equal-power, single-mode laser beams. Their wavelengths are $\lambda_1 = 1545$ nm (Agilent 81689A) and $\lambda_2 = 1575$ nm (Tunics Plus CL/WB), with linewidths of about 100 MHz. The spectra of the two sources are shown in the inset of Fig. 1 and they fall within the two-photon absorption regime.

Consider next the structure of the two-photon absorption signal. The field arriving at the detector is a sum of the fields from the two arms, i.e., $E(t) = E_a(t) + E_b(t + \tau)$. We introduce the complex envelope representation [1] as $E_m(t) = A_m(t)e^{-i\omega_0 t}$, $m \in (a, b)$, where the amplitude $A_m(t)$ is slowly varying and $\omega_0$ is an angular frequency within the spectrum. The two-photon absorption (TPA) signal then takes on the form (see Appendix B)

$$S(\tau) \propto \langle I^2(t) \rangle = \langle I_a^2(t) \rangle + \langle I_b^2(t) \rangle + 2\langle I_a(t)I_b(t + \tau) \rangle + 2\langle |A_a^m(t)A_b^m(t + \tau)|^2 \rangle + \text{Re}[F^{(1)}(\tau)e^{i\omega_0 \tau}] + \text{Re}[F^{(2)}(\tau)e^{2i\omega_0 \tau}],$$

where $F^{(1)}(\tau)$ and $F^{(2)}(\tau)$ are slowly varying functions of $\tau$ whose explicit forms are not relevant for this work. The first two and the third term above are due to photons from the same and different interferometer arms, respectively, while the last two terms essentially represent ordinary field interference. The fourth term originates from the vector nature of light and is
not present in the previous scalar works [12, 13, 15]. It has a clear physical meaning as characterizing polarization dynamics in partially polarized light [16]. More precisely, it describes how rapidly, on average, the intensity in a certain polarization state is transferred to the orthogonal state. The effect of polarizers is to allow access to the third term representing intensity auto-correlations and cross-correlation.

Filtering from the signal the high-frequency components around $\omega_0$ and $2\omega_0$, assuming that the fields in the arms are either $x$ or $y$ polarized (due to the polarizers), we find from Eq. (4) that

$$\langle I_m(t)I_n(t+\tau)\rangle = C[S_{ij}(\tau) - S_{ii} - S_{nj}]/\eta_{ij}. \quad (5)$$

Above, $I_m(t)$ is the intensity of $i \in \{x,y\}$ polarized component in arm $m \in \{a,b\}$, $S_{ii} = \langle I_m^2(t)\rangle$ is the related TPA signal, $C$ is a polarization-independent coefficient, and $\eta_{ij} = 4$ for $i = j$ while $\eta_{ij} = 2$ if $i \neq j$. We thus find that, for any unknown field at the interferometer input, measuring the two-photon absorption signal $S_{ij}(\tau)$ for $(i,j) \in \{x,y\}$ by moving the mirror in arm $b$, and recording the signals $S_{ai}$ and $S_{bj}$ when one arm is blocked, one obtains the intensity correlation functions of the input field as $\langle I_i(t)I_j(t+\tau)\rangle = D(\langle I_{ai}(t)I_{bj}(t+\tau)\rangle)$, where $D$ is a proportionality constant. These, in turn, lead to the correlation function of the total intensity $I(t) = I_i(t) + I_j(t)$ present in Eqs. (2) or (3). The term $\langle I(t)\rangle^2$ in the denominator of Eq. (2) is for ASE obtained from $\langle I_i(t)I_j(t+\tau)\rangle$ at long $\tau$. For the two-laser beam its value is not needed, as evidenced by Eq. (3). Note that the coefficients $C$ and $D$ are cancelled since $\gamma(\tau)$ is a normalized quantity.

4. Measurement results

The signals $S(\tau)$ measured for the ASE light with the $x$ and $y$ polarized components selected in the interferometer arms are shown in Fig. 2(a). They are labeled as XX, YY, and XY, conforming to the polarizations in arms $a$ and $b$. Before the measurements we ensured that the ASE beam is fully unpolarized, supported by the almost indistinguishable blue XX and red YY curves. These curves also demonstrate a good reproducibility of our experimental results. They are both rapidly oscillating functions of $\tau$, as is seen in the inset, and have a major amplitude peak at $\tau = 0$ (equal-length arms). The secondary peaks at $\tau = \pm 310$ fs are caused by the presence of two maxima in the ASE spectrum (see Fig. 1). The rapid oscillations correspond to the last two (field-interference) terms in Eq. (4). The black XY curve is flat as the $x$ and $y$ components of the unpolarized ASE beam do not correlate. Since the mirror position is scanned...
symmetrically around $\tau = 0$, measurement of $\langle I_y(t)I_x(t+\tau) \rangle$ that would yield the YX curve is not necessary. We simply use the fact that $\langle I_y(t)I_x(t+\tau) \rangle = \langle I_x(t)I_y(t-\tau) \rangle$.

Applying a Fourier-transform-based low-pass filtering to the curves of Fig. 2(a) we obtain the signals $S_{ij}(\tau)$. Inserting these data and the measured values of $S_{ai}$ and $S_{bi}$ into Eq. (5) then yields the intensity correlation functions (ICFs) $\langle I_x(t)I_x(t+\tau) \rangle$, $\langle I_y(t)I_y(t+\tau) \rangle$, and $\langle I_x(t)I_y(t+\tau) \rangle$ shown in Fig. 2(b) with the blue XX, red YY, and black XY curves, respectively. The curves exhibit small oscillations with a period of about 70 fs (corresponding to scanning time of about two minutes), which presumably originate from weak ASE power fluctuations (< 2 %). The measured intensity correlation functions are seen to decrease significantly when $\tau$ increases from 0 to about 200 fs. This fast drop would be impossible to observe with ordinary single-photon absorption, because even ultrafast detectors have too long a response time (~100 ps). In our measurements, all intensity fluctuations that have a characteristic time longer than ~1 fs contribute to $\langle I(t)I(t+\tau) \rangle$ at each $\tau$.

Substituting the functions in Fig. 2(b) into Eq. (2), we obtain the electromagnetic degree of temporal coherence $\gamma(\tau)$ shown by the blue curve in Fig. 3. It has a peak value of about 0.7, close to the theoretical prediction of $1/\sqrt{2}$ for a fully unpolarized field. By Fourier transforming the ASE spectrum in Fig. 1 and assuming that the beam is fully unpolarized we find the values of $\gamma(\tau)$ indicated in Fig. 3 by the blue stars. The agreement between the two results is remarkable. The red curve shows the degree of coherence of the $x$-polarized component of the field, and the red stars represent the values of $\gamma(\tau)$ evaluated from the spectrum and assuming a fully polarized field. The coherence time $\tau_c$ shown in the figure is approximately 200 fs corresponding to $\gamma(\tau_c) = 0.3\gamma(0)$.

The second field that we consider is the two-laser beam whose spectrum is given in Fig. 1. The field constructed by combining two orthogonally linearly polarized laser beams is transmitted through the fiber that is not polarization maintaining. At the fiber output, the lasers are generally elliptically polarized but the polarization states are still nearly orthogonal. Consequently, the total field is effectively unpolarized and both the $x$ and $y$ polarized components within the interferometer contain contributions from both lasers. Figure 4(a) shows the two-photon absorption signal when the polarizers in the interferometer transmit $x$ polarization (blue XX curve), $y$ polarization (red YY curve), and $x$ and $y$ polarization in arms $a$ and $b$, respectively (black XY curve). In contrast to the ASE source, all curves exhibit a periodic structure illustrating the
wave-beating phenomenon; the observed period $T = 270$ fs is given by $T = \frac{\lambda_2 \lambda_1}{\lambda_2 - \lambda_1} c$.

The three illustrated periods in each signal are identical because they are close to $\tau = 0$, but if $\tau$ approached the coherence times of the lasers ($\sim 10$ ns) the signal amplitudes would considerably decrease. At $\tau = 0$, the XX and YY signals have maximum amplitudes implying that the beating intensities from the interferometer arms oscillate in phase. The XY curve also exhibits beating, but it has a minimum at $\tau = 0$, reflecting the fact that the intensities of the $x$ and $y$ polarized fields are out of phase. The beating is accompanied by a high-frequency oscillation caused by a small coupling between the mutually partially correlated $x$ and $y$ polarization components of the field at the detector. This oscillation, however, is Fourier-filtered out and does not affect the final result. At $\tau = 0$ the curves indicate that the intensity ratio of the $x$ and $y$ polarized components is about 0.86 and they are weakly correlated. The two-laser beam is therefore rather unpolarized. Filtering out the high-frequency oscillations leads to the intensity correlation functions depicted in Fig. 4(b). The functions are shifted with respect to each other, but have the same shape consistently with the fact that the field is approximately unpolarized.

Using the intensity correlation functions of Fig. 4(b) in Eq. (3) with $g = 1$, we obtain the degree of coherence $\gamma(\tau)$ shown by the blue line in Fig. 5. The degree of coherence is seen to be independent of $\tau$ within the measured interval and approximately equal to $\gamma(0) = 1/\sqrt{2}$ of a fully unpolarized light. This value for small $\tau$ can be verified by analyzing Eq. (1) for orthogonally polarized, equal-intensity single-mode lasers. The coherence time for this field is roughly equal to that of each laser, i.e., about 10 ns, which is far beyond the scanning range of the interferometer. However, the situation is different for a polarized two-laser beam as evidenced by the red curve in Fig. 5 obtained from Eq. (3) with $g = 2$ and illustrating $\gamma(\tau)$ for the $x$-polarized component. The degree of coherence varies significantly as a function of $\tau$ with a period $T = 270$ fs. The red stars show $\gamma(\tau)$ calculated from the spectrum and taking that the contribution of the laser at $\lambda_1$ is 0.21 (this estimate can be found by an analysis of the two-laser beating) times that at $\lambda_2$. The match is excellent. The coherence time can be chosen as $\tau_c = 135$ fs corresponding to the fall from $\gamma(0) = 1$ to about 2/3. An analogous criterion has been used to specify the coherence time of a multi-mode laser [28]. The two-laser example also demonstrates that the method can be used to examine ultrafast polarization beating of light.
Fig. 5. Electromagnetic degree of coherence \( \gamma(\tau) \) for the nearly unpolarized two-laser field (blue line) and its \( x \)-polarized component (red line). The stars show the values of \( \gamma(\tau) \) obtained from the spectrum. The oscillation period is \( T = 270 \) fs and the coherence time is \( \tau_c = T/2 = 135 \) fs.

5. Conclusion

In summary, we have measured for the first time the electromagnetic degree of coherence of unpolarized and polarized stationary light beams with and without Gaussian statistics at ultra-short time scales. This was done by using the two-photon absorption effect in a photomultiplier tube which has never before been employed to analyze coherence characteristics of stationary, partially polarized electromagnetic light beams. We assessed the femtosecond-scale coherence times, revealed ultrafast polarization beating, and in the case of Gaussian statistics found also the degree of polarization. In contrast to the previous studies on intensity correlations of scalar light, the role of polarization dynamics in intensity interference and two-photon absorption was recognized theoretically and handled in practice by making use of polarizers in the interferometer arms. Our results can impact experimental studies of electromagnetic coherence and polarization in classical and quantum light.

A. Derivation of Eq. (3)

Single-mode laser beams do not obey Gaussian field statistics but rather follow Poissonian photon-number distributions. The electric field of the superposition of two quasi-monochromatic laser beams with mid frequencies \( \omega_1 \) and \( \omega_2 \) is

\[
E(t) = A_1(t)e^{-i\omega_1t} + A_2(t)e^{-i\omega_2t},
\]

where the amplitudes \( A_1(t) \) and \( A_2(t) \) are slowly varying with time. The mutual coherence matrix becomes

\[
\Gamma(\tau) = \Gamma_1(\tau)e^{-i\omega_1\tau} + \Gamma_2(\tau)e^{-i\omega_2\tau},
\]

where \( \Gamma_i(\tau) = \langle A_i^*(t)A_i^T(t+\tau) \rangle, \) with \( i \in (1, 2) \), is the coherence matrix related to a single laser. In the derivation of Eq. (7) we used the fact that rapidly oscillating terms average to zero. The numerator in the degree of coherence of Eq. (1) then takes on the form

\[
\text{tr}[\Gamma(\tau)\Gamma(-\tau)] = \text{tr}[\Gamma_1(\tau)\Gamma_1(-\tau)] + \text{tr}[\Gamma_2(\tau)\Gamma_2(-\tau)]
+ \text{tr}[\Gamma_1(\tau)\Gamma_2(-\tau)]e^{i\Delta\omega\tau} + \text{tr}[\Gamma_2(\tau)\Gamma_1(-\tau)]e^{-i\Delta\omega\tau},
\]

\[\text{(8)}\]
where $\Delta \omega = \omega_2 - \omega_1$. The matrices $\mathbf{\Gamma}_1(\tau)$ and $\mathbf{\Gamma}_2(\tau)$ are slowly varying with $\tau$. For $\tau$ much smaller than the coherence times of the lasers we can write

$$\text{tr}[\mathbf{\Gamma}(\tau)\mathbf{\Gamma}(-\tau)] = \text{tr}(\mathbf{J}_1^2) + \text{tr}(\mathbf{J}_2^2) + 2\text{tr}(\mathbf{J}_1\mathbf{J}_2)\cos(\Delta \omega \tau),$$

where $\mathbf{J}_i = \mathbf{\Gamma}_i(0)$, with $i \in (1, 2)$, is the polarization matrix of a single laser. The squared degree of polarization of laser $i$ is $P_i^2 = 2\text{tr}(\mathbf{J}_i^2)/\text{tr}(\mathbf{J}_i) - 1/2$ [29], which for completely polarized light, $P_i = 1$, implies that $\text{tr}(\mathbf{J}_i^2) = \text{tr}^2(\mathbf{J}_i) = (I_i(t))^2$, $i \in (1, 2)$. Hence, for a superposition of two quasi-monochromatic, fully polarized laser beams we find that the degree of coherence in Eq. (1) is for small $\tau$ specified by

$$\gamma^2(\tau) = \frac{(I_1(t))^2 + (I_2(t))^2}{(I(t))^2} + \frac{2\text{tr}(\mathbf{J}_1\mathbf{J}_2)\cos(\Delta \omega \tau)}{(I(t))^2},$$

where $\langle I(t) \rangle = (I_1(t)) + (I_2(t))$ is the total beam intensity. This result shows that the degree of coherence behaves sinusoidally as a function of $\tau$ with period $T = 2\pi/\Delta \omega$.

Consider next the intensity correlations of the two-laser beam. The instantaneous intensity related to the field of Eq. (6) is

$$I(t) = E^T(t)E^*(t) = I_1(t) + I_2(t) + A_1^T(t)A_2^*(t)e^{i\Delta \omega t} + A_2^T(t)A_1^*(t)e^{-i\Delta \omega t},$$

where $I_i(t) = A_i^T(t)A_i^*(t)$ is the instantaneous intensity of laser $i \in (1, 2)$. The formula above implies that

$$\langle I(t)I(t+\tau) \rangle = \langle I_1(t)I_1(t+\tau) \rangle + \langle I_1(t)I_2(t+\tau) \rangle + \langle I_2(t)I_1(t+\tau) \rangle + \langle I_2(t)I_2(t+\tau) \rangle + \langle [A_1^T(t)A_2^*(t)][A_2^T(t+\tau)A_1^*(t+\tau)] \rangle e^{-i\Delta \omega t} + \langle [A_2^T(t)A_1^*(t)][A_1^T(t+\tau)A_2^*(t+\tau)] \rangle e^{i\Delta \omega t},$$

where the rapidly oscillating terms again averaged to zero. Since the two single-mode lasers are independent, $\langle I_1(t)I_2(t+\tau) \rangle = \langle I_2(t)I_1(t+\tau) \rangle = \langle I_1(t) \rangle \langle I_2(t) \rangle$, and because they obey Poissonian photon-coincidence statistics, $\langle I_i^2(t) \rangle = (I_i(t))^2$ for $i \in (1, 2)$, we may write for small $\tau$ that

$$\langle I(t)I(t+\tau) \rangle = (I_1(t))^2 + (I_2(t))^2 + 2(I_1(t))(I_2(t)) + 2\langle |A_1^T(t)A_2^*(t)|^2 \rangle \cos(\Delta \omega \tau).$$

We further find for independent lasers that $\langle |A_i^T(t)A_2^*(t)|^2 \rangle = \text{tr}(\mathbf{J}_1\mathbf{J}_2)$, and hence

$$\langle I(t)I(t+\tau) \rangle = (I(t))^2 + 2\text{tr}(\mathbf{J}_1\mathbf{J}_2)\cos(\Delta \omega \tau),$$

whose minimum and maximum values are

$$\langle I(t)I(t+\tau) \rangle_{\text{min}} = (I(t))^2 - 2\text{tr}(\mathbf{J}_1\mathbf{J}_2),$$

$$\langle I(t)I(t+\tau) \rangle_{\text{max}} = (I(t))^2 + 2\text{tr}(\mathbf{J}_1\mathbf{J}_2),$$

leading to

$$\langle I(t)I(t+\tau) \rangle_{\text{min}} + \langle I(t)I(t+\tau) \rangle_{\text{max}} = 2(I(t))^2.$$
Using Eqs. (14) and (17) in Eq. (10) results in

\[
\gamma^2(\tau) = \frac{2\langle I_1(t) \rangle^2 + \langle I_2(t) \rangle^2 + 2\langle I(t)I(t + \tau) \rangle}{\langle I(t)I(t + \tau) \rangle_{\text{min}} + \langle I(t)I(t + \tau) \rangle_{\text{max}}} - 1. \tag{18}
\]

This equation holds for the superposition of two independent, quasi-monochromatic, fully polarized laser beams when \( \tau \) is much smaller than the coherence times of the lasers. Next we reduce the above expression for the specific cases considered in the main text.

**Polarized superposition beam: lasers with the same polarization state**

When the two fully polarized lasers have the same states of polarization but possibly unequal intensities, we can write the polarization matrices in the factored forms as [30]

\[
\mathbf{J}_1 = (\text{tr} \mathbf{J}_1) \mathbf{e} \mathbf{e}^T, \tag{19}
\]

\[
\mathbf{J}_2 = (\text{tr} \mathbf{J}_2) \mathbf{e} \mathbf{e}^T, \tag{20}
\]

where the complex unit vector \( \mathbf{e} \) describes the polarization state. In this case we find that \( \text{tr}(\mathbf{J}_1 \mathbf{J}_2) = \text{tr}(\mathbf{J}_1)\text{tr}(\mathbf{J}_2) \), and thus Eq. (15) yields \( \langle I(t)I(t + \tau) \rangle_{\text{min}} = \langle I_1(t) \rangle^2 + \langle I_2(t) \rangle^2 \). Consequently, Eq. (18) becomes

\[
\gamma^2(\tau) = \frac{2\langle I(t)I(t + \tau) \rangle + 2\langle I(t)I(t + \tau) \rangle_{\text{min}}}{\langle I(t)I(t + \tau) \rangle_{\text{min}} + \langle I(t)I(t + \tau) \rangle_{\text{max}}} - 1, \tag{21}
\]

which expresses the electromagnetic degree of coherence \( \gamma(\tau) \) solely in terms of the intensity correlation function and coincides with Eq. (3) when \( g = 2 \).

**Unpolarized superposition beam: lasers with orthogonal polarization states and equal intensities**

For two orthogonally polarized equal-intensity lasers we have \( \langle I_1(t) \rangle = \langle I_2(t) \rangle = \langle I(t) \rangle / 2 \) and since the lasers are independent we further obtain \( \langle I^2(t) \rangle = \langle I(t) \rangle^2 \). Therefore,

\[
\langle I_1(t) \rangle^2 + \langle I_2(t) \rangle^2 = \frac{1}{2} \langle I^2(t) \rangle = \frac{1}{2} \langle I(t)I(t + \tau) \rangle_{\text{max}} = \frac{1}{2} \langle I(t)I(t + \tau) \rangle_{\text{min}}, \tag{22}
\]

where the last equality follows from Eqs. (15) and (16) by noting that \( \text{tr}(\mathbf{J}_1 \mathbf{J}_2) = 0 \). This, in turn, is found by writing the polarization matrices similarly to Eqs. (19) and (20) but with orthogonal polarization states. It is clear that in this case \( \langle I(t)I(t + \tau) \rangle \) is constant, and inserting the results given in Eq. (22) into Eq. (18) one obtains \( \gamma^2(\tau) = 0.5 \). Nonetheless, the quantity \( \gamma^2(\tau) \) for the unpolarized superposition beam may also be written in the form similar to Eq. (21) as

\[
\gamma^2(\tau) = \frac{2\langle I(t)I(t + \tau) \rangle + \langle I(t)I(t + \tau) \rangle_{\text{min}}}{\langle I(t)I(t + \tau) \rangle_{\text{min}} + \langle I(t)I(t + \tau) \rangle_{\text{max}}} - 1, \tag{23}
\]

which is Eq. (3) with \( g = 1 \).

**B. Derivation of Eq. (4)**

The field at the detector is \( \mathbf{E}(t) = \mathbf{E}_a(t) + \mathbf{E}_b(t + \tau) \), where the subscripts \( a \) and \( b \) refer to the interferometer arms and \( \tau \) is the time delay due to the movable mirror in arm \( b \). The instantaneous intensity becomes

\[
I(t) = I_a(t) + I_b(t + \tau) + 2\text{Re}[\mathbf{E}_a^T(t)\mathbf{E}_b^*(t + \tau)]. \tag{24}
\]
where \( I_a(t) \) and \( I_b(t + \tau) \) are the instantaneous intensities of the fields from the two arms. The two-photon absorption signal thus takes on the form

\[
S(\tau) \propto \langle F^2(t) \rangle = \langle I_a^2(t) \rangle + \langle I_b^2(t) \rangle + 2\langle I_a(t)I_b(t + \tau) \rangle \\
+ 4\lfloor \langle I_a(t) + I_b(t + \tau) \rangle \langle \text{Re}[E_a^T(t)E_b^*(t + \tau)] \rangle \rceil \\
+ 4\lfloor \langle \text{Re}[E_a^T(t)E_b^*(t + \tau)] \rangle^2 \rceil,
\]

(25)

where we used the stationarity of \( E_b(t) \). Employing the complex envelope representation \( E_i(t) = A_i(t)e^{-i\omega_0t}, \ i \in \{a,b\} \), where \( \omega_0 \) is an angular frequency within the spectrum, we may write

\[
E_a^T(t)E_b^*(t + \tau) = F(t; \tau)e^{i\omega_0\tau},
\]

(26)

where we have introduced the abbreviation \( F(t; \tau) = A_a^T(t)A_b^*(t + \tau) \). The term in the middle row of Eq. (25) then becomes

\[
4\langle [I_a(t) + I_b(t + \tau)] \langle \text{Re}[E_a^T(t)E_b^*(t + \tau)] \rangle \rangle = \text{Re} \{ F^{(1)}(\tau)e^{i\omega_0\tau} \},
\]

(27)

where \( F^{(1)}(\tau) = 4\langle [I_a(t) + I_b(t + \tau)]F(t; \tau) \rangle \). In terms of Eq. (26), the third row of Eq. (25) can be developed as follows

\[
4\lfloor \langle \text{Re}[E_a^T(t)E_b^*(t + \tau)] \rangle \rfloor^2 = 4\lfloor \langle \text{Re}[F(t; \tau)] \cos(\omega_0\tau) - \text{Im}[F(t; \tau)] \sin(\omega_0\tau) \rangle \rfloor^2,
\]

(28)

which with standard trigonometric identities and complex algebra assumes the form

\[
4\lfloor \langle \text{Re}[E_a^T(t)E_b^*(t + \tau)] \rangle \rfloor^2 = 2\lfloor \langle A_a^T(t)A_b^*(t + \tau) \rangle \rfloor^2 \\
+ \text{Re} \{ F^{(2)}(\tau)e^{i2\omega_0\tau} \},
\]

(29)

where \( F^{(2)}(\tau) = 2\langle F^2(t; \tau) \rangle \). Combining Eqs. (25), (27), and (29) results in Eq. (4).

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