Intensity Interference of Ultrashort Pulsed Fluorescence

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We extend the intensity interference experiments to a pulse envelope measurement of ultrashort incoherent optical pulses. The interfering fields can be mutually incoherent light pulses from independent sources. We show that the pulse envelope or width of ultrashort incoherent light pulses can be measured independently from the coherence time by using a combination of interferometer and two-photon coincidence counter. With this method we have measured the envelope autocorrelation of mutually incoherent femtosecond fluorescence from a solution of Nile Blue in dimethylaniline. [S0031-9007(96)00367-5]

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We propose a new scheme to measure independently the coherence time and the envelope of ultrashort incoherent pulses with two-photon interference, and show a measurement of lifetime of ultrashort fluorescence. Since the idea of intensity interference was first introduced by Hanbury Brown and Twiss [1], importance of high-order correlations was realized. Most notably, the quantum theory of optical coherence was formalized using arbitrarily high-order correlations by Glauber [2]. The intensity interference is the fourth order in terms of the amplitudes of the optical field as opposed to the second-order interference such as Young's interference. But most of the studies were concerned with measurements of the coherence of light including the study by Hanbury Brown and Twiss themselves. They focused their attention on the fact that even the intensity interference could measure the coherence of light, which was also measurable with the second-order interference [3]. In principle, the intensity interference should be able to measure not only the coherence time or the beats among the fields but the intensity profile or the pulse envelope of incident light.

In the original intensity interference scheme a time resolution is restricted by the detector response time, so one must use photodetectors having response time faster than the coherence time one wishes to measure. This difficulty was overcome when two-photon intensity was measured by two photodetectors placed at the two output ports of a Mach-Zehnder interferometer [4]. In this scheme the time resolution is determined by the accuracy of the position of the delay line of the interferometer, which can be more precisely determined than the response time of the detectors. With this scheme, two-photon coincidence experiments were performed in quantum mechanical [5] and in classical [6] domains of light. However, all these measurements were carried out on cw light. We applied this scheme to the measurement of the envelope or the width of ultrashort light pulses, such as mode-locked dye laser pulses [7], or scattered light of those from randomly distributed elastic scattering centers [8]. In these measurements the light in the two arms of a Mach-Zehnder (or Michelson) interferometer and in the two outputs from the interferometer ports carried mutual coherence. Therefore, the signals which provided the information of the coherence time and the pulse envelope or width appeared in superposition, and the measurement of these values was sometimes difficult, depending on their ratio. It is hoped that these two signals can be measured separately.

In the present Letter we show that one can measure by interference the intensity profile of mutually incoherent light pulses from independent sources. We also show that the intensity profile or the coherence time of light pulse can be measured separately by intensity interference. If the pulses in two arms of the interferometer have no mutual coherence, the intensity profile can be measured. If those in the two output paths from the interferometer have no mutual coherence, the coherence time can be measured. This new principle enables us to measure the envelope of femtosecond pulses by use of interference, at very low intensity levels in the wide wavelength regions, and with virtually no limit in the time resolution. We apply this scheme to the measurement of a lifetime of ultrashort fluorescence from a dye in the femtosecond regime. In the present study we measured the convolution of fluorescence and excitation-laser envelope functions.

The pulse field is incident upon a beam splitter BS1 of the Mach-Zehnder interferometer in Fig. 1. Two beams meet on the second beam splitter BS2. We perform photon counting with two detectors D1 and D2 located after BS2. We define the coincidence count rate, i.e., the probability that the two detectors detect one photon each, as

$$P_{12}(t,\tau) = K_1 K_2 \langle E^*_{\mathrm{D1}}(t) E^*_{\mathrm{D2}}(t+\tau) E_{\mathrm{D2}}(t+\tau) E_{\mathrm{D1}}(t) \rangle , \tag{1}$$

where E_{D1} and E_{D2} represent the fields at time t and $t + \tau$ at the two detectors, K_1 and K_2 being their quantum efficiencies. The angular brackets denote an average over a statistical ensemble. In an actual experiment, the coincidence count rate is measured for a finite measurement time T_M. The coincidence is defined as two signals received within a resolution time τ_{CR} of the detector circuit, which is usually much larger than the response time of the photodetectors τ_R. With these considerations the coincidence

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counts become

$$C_{12}(\delta\tau) = \int_{-T_M/2}^{T_M/2} dt \int_{-\tau_{CR}/2}^{\tau_{CR}/2} d\tau\, P_{12}(t,\tau). \quad (2)$$

We use an ultrashort repetitive pulse train of a mode-locked laser with the pulse cycle T_0 and pulse width τ_p as incident light to excite fluorescence. Then the following relation holds:

$$T_M \gg T_0 \gg \tau_{CR} \gg \tau_R \gg \tau_p. \quad (3)$$

In Fig. 1, delays by one pulse cycle are included in one of the two arms of the interferometer and in one of the paths between the interferometer and a time-to-amplitude converter (TAC). In a standard measurement, however, these delays are not used, and two arms and two paths are equal in length. We explain the case of standard measurement first. The fields at the detectors D1 and D2 at time t are

$$E_{D1}(t) = \sqrt{T}\,E_1(t) + i\sqrt{R}\,E_2(t+\delta\tau), \quad (4)$$

$$E_{D2}(t) = \sqrt{T}\,E_2(t+\delta\tau) + i\sqrt{R}\,E_1(t), \quad (5)$$

where T and R are the transmittance and reflectivity of the beam splitter; $c\delta\tau$ is the path difference between the two arms of the interferometer introduced by translating a corner cube. We obtain sixteen terms by substituting Eqs. (4) and (5) into Eq. (1). By taking the average over t, ten of them reduce to zero if E_1 and E_2 are mutually incoherent or if a random phase modulator is inserted in one of the interferometer arms, and the next six terms remain:

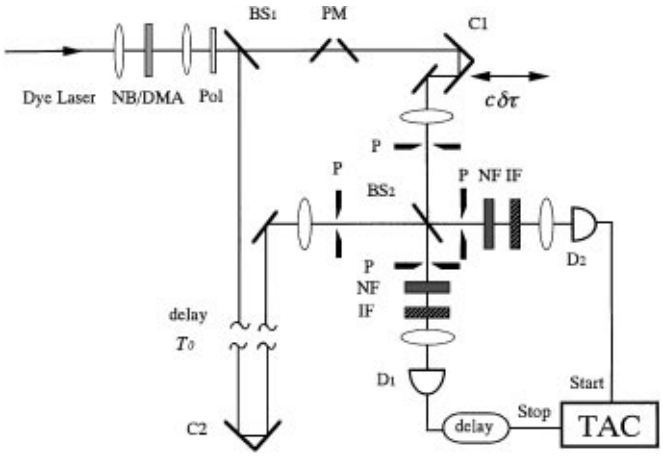

FIG. 1. Schematic diagram of the experimental setup for measuring coincidence of fluorescence photons between adjoining pulses. One arm of the interferometer or the output path is stretched by one pulse cycle of the mode-locked laser. NB, Nile Blue/DMA solution; P, pinholes; Pol, polarizer; BS1 and BS2, beam splitters; PM, phase modulator; C1 and C2, corner cubes; NF, notch filters; IF, interference filters; D_1 and D_2, photodetectors; TAC, time-to-amplitude converter.

$$\begin{aligned} P_{12}(t,\tau) = {} & K_1K_2\langle T^2|E_1(t)E_2(t+\tau+\delta\tau)|^2 + R^2|E_2(t+\delta\tau)E_1(t+\tau)|^2 + RT|E_1(t)E_1(t+\tau)|^2 \\ & + RT|E_2(t+\delta\tau)E_2(t+\tau+\delta\tau)|^2 - RTE_1^*(t)E_2^*(t+\tau+\delta\tau)E_1(t+\tau)E_2(t+\delta\tau) \\ & - RTE_2^*(t+\delta\tau)E_1^*(t+\tau)E_2(t+\tau+\delta\tau)E_1(t)\rangle. \end{aligned} \quad (6)$$

The fifth and sixth terms are interference terms, and give a dip in the correlation signal as a function of $\delta\tau$. The first four terms give constant contributions independent of $\delta\tau$ after the integration by τ.

Ultrashort fluorescence field can be expressed by the envelope function of the fluorescence $C(t)$ and a random fluctuating function $A(t)$ as follows:

$$E(t) = C(t)A(t), \quad (7)$$

$$C(t) = C\exp(-t/\tau_L), \quad (8)$$

$$A(t) = \sum_k A_k \exp(-i\omega_k t), \quad (9)$$

where τ_L is the decay time of the fluorescence, and ω_k and A_k are a mode frequency and a random fluctuating complex amplitude of the kth mode. The ultrashort fluorescence has a very wide spectral range, so we use interference filters in front of the detectors to reduce the spectral width and to make the ratio of the coherence time to the lifetime not too small. This is advantageous to detect a part of the signal which indicates the lifetime of fluorescence. (See the discussion below.) We measured the transmission spectra of the interference filters and found that the distribution of $\langle|A_k|^2\rangle$ was approximately Lorentzian. So, we may write

$$\langle A^*(t)A(t+\tau)\rangle = A^2\exp(-i\omega_0\tau)\exp(-|\tau|/\tau_c), \quad (10)$$

where τ_c is the coherence time defined as the inverse of the pass band of the interference filter, and A^2 is the time averaged power of $A(t)$. We will further assume the field $A(t)$ to obey complex Gaussian statistics. Therefore, the following theoretical analysis is possible when the number of random fluctuating modes is not very small.

The fifth term of Eq. (6) which gives the dip then becomes

$$\begin{aligned} [P_{12}]_5 = {} & -K_1K_2RTC_1(t)C_2(t+\tau+\delta\tau)C_1(t+\tau)C_2(t+\delta\tau) \\ & \times\{\langle A_1^*(t)A_2(t+\delta\tau)\rangle\langle A_2^*(t+\tau+\delta\tau)A_1(t+\tau)\rangle + \langle A_1^*(t)A_1(t+\tau)\rangle\langle A_2^*(t+\tau+\delta\tau)A_2(t+\delta\tau)\rangle\}. \end{aligned} \quad (11)$$

The sixth term is the complex conjugate of this. If we look carefully at the two terms of Eq. (11), we see that the first term is caused by a second-order interference effect between the fields in different arms expressed by $\langle A_1^*(t_1)A_2(t_1 + \delta\tau)\rangle$ and $\langle A_2^*(t_2 + \delta\tau)A_1(t_2)\rangle$ with $t_2 = t_1 + \tau$. This term exists when the two fields A_1 and A_2 are mutually coherent and $|\delta\tau|$ is smaller than the coherence time τ_c. Duration of this interference with respect to $t_2 - t_1$ is limited only by the pulse width τ_p. The second term, corresponding to the product of the correlation functions $\langle A_1^*(t_1)A_1(t_2)\rangle$ and $\langle A_2^*(t_2 + \delta\tau)A_2(t_1 + \delta\tau)\rangle$, arises even when A_1 and A_2 are mutually incoherent, since the fields at the two detectors are correlated when $|t_2 - t_1| < \tau_c$. This means that the fields from different output ports, but from the same arm, interfere within the coherence time τ_c. Therefore the duration of this interference with respect to $\delta\tau$ is limited only by the pulse width τ_p, hence, by measuring coincidence, we can obtain the envelope function of the pulses from this term. Observation of this second type of interference requires the use of two photodetectors, and it is intrinsically a fourth-order effect. In addition, ordinary interferometer stability of the paths is not required.

The coincident counts for one pulse cycle T_0 are calculated from Eqs. (6) and (2) as

$$C_{12}(\delta\tau) = 4H\left\{\left(1 - \frac{1}{2}\exp(-2|\delta\tau|/\tau_c)\right) + \frac{K}{H}\left(1 - \frac{1}{2}\exp(-2|\delta\tau|/\tau_L)\right)\right\} \tag{12}$$

under the assumption of Eqs. (7)–(10) and by replacing T_M with T_0 in Eq. (2). Here, $H = \frac{1}{4}\tau_L^2, K = \frac{1}{8}\tau_L\tau_c, K/H = \tau_c/2\tau_L$. Equation (12) shows a dip at $\delta\tau = 0$ that consists of two components, i.e., a narrow deep component with the width τ_c and a wide shallow component with the width τ_L. These two components come from the two terms of Eq. (11).

From the above consideration we find that the two terms of Eq. (11) can be measured separately. If the fields at the two arms 1 and 2 are mutually incoherent, the first term of Eq. (11) disappears, and the envelope function can be measured, i.e., the wide component of the dip alone can be measured. On the other hand, if the fields at the two detectors are mutually incoherent, the second term disappears, and the field correlation function can be measured, i.e., the narrow component alone can be measured. In order to obtain two incoherent pulses at the arms 1 and 2, either one can place a fluorescence sample in each of the two arms or, more conveniently, one can use adjoining pulses of fluorescence pulse train excited by cw mode-locked laser. These fluorescence pulses completely lose the coherence of the excitation pulses, but have a common envelope function with each other.

We adopt the second method in the present study, and add a delay cT_0 to the second arm in order to measure the envelope function as shown in Fig. 1. In Eq. (11), $\delta\tau$ is then replaced by $\delta\tau - T_0$. [If, on the other hand, one wishes to measure the field correlation function, one can add a delay to one of the paths between BS2 and TAC or, more conveniently, to one of the output cables from the detectors as shown in Fig. 1. In Eq. (11), τ would then be replaced by $\tau - T_0$.] Equation (11) now becomes

$$\begin{aligned}[P_{12}]_5 = &- K_1K_2RTC_1(t)C_2(t + \tau + \delta\tau)C_1(t + \tau)C_2(t + \delta\tau)\\ &\times \{\langle A_1^*(t)A_2(t - T_0 + \delta\tau)\rangle\langle A_2^*(t + \tau - T_0 + \delta\tau)A_1(t + \tau)\rangle\\ &+ \langle A_1^*(t)A_1(t + \tau)\rangle\langle A_2^*(t + \tau - T_0 + \delta\tau)A_2(t - T_0 + \delta\tau)\rangle\}.\end{aligned} \tag{13}$$

It is clear that the first term of Eq. (13) disappears, and the second term survives when $\tau < \tau_c$ and $\delta\tau < \tau_p$ as before, and gives us the intensity correlation function approximately of the form $C_1^2(t)C_2^2(t + \delta\tau)$. The final form of the coincidence counts for two independent sources becomes

$$C_{12}(\delta\tau) = 4H\left\{1 + \frac{K}{2H}\left(1 - \exp(-2|\delta\tau|/\tau_L)\right)\right\}. \tag{14}$$

From Eq. (14) we see that it is better to make the ratio, $K/2H$ (i.e., proportional to τ_c/τ_L), large. Therefore we limit the spectra of the fluorescence with the interference filters to make the coherence time τ_c long.

We measured the femtosecond fluorescence of Nile Blue (NB) dissolved in neat N-dimethylaniline (DMA), which acts as a weakly polar electron-donating solvent. In this system, electron donors and acceptors are in direct contact, and electron transfer is not limited by translational diffusion, so the decay time is very short and is about 100 fs [9]. In Fig. 1 the dye solution was excited by a synchronously pumped cw mode-locked dye laser of the wavelength at 580 nm and the repetition rate of 82 MHz. In order to obtain the interference of the field from an incoherent source or sources, we have to select a spatially coherent part of the fluorescence light which is as intense as possible. For this purpose, we focused the fluorescence on a small pinhole of a diameter 100 μm and selected out the central disk of diffraction at the position of the detector, about 25 cm from the pinhole. There is no need to operate the phase modulator, since two fields in two arms of the interferometer are

mutually incoherent in this measurement. In addition, a single polarization was selected using a polarizer. Laser light unintentionally transmitted through the dye cell was blocked by super-notch filters (transmissivity, 10^{-6}). The interference filters had center wavelength, 690 nm, and a full width at half maximum, 1.4 nm. The time intervals between signals from D1 and D2 were measured, and cases where they were shorter than a certain coincidence resolution time τ_{CR} (10 ns) were counted as coincident.

The result of the coincidence count between adjoining pulses with a stretched arm by $T_0 = 12.2$ ns is shown by dots in Fig. 2. One dot represents the counts during a measurement time of 500 s. During this time, the number of the coincidence counts was about 2180 at the largest delay of the corner cube ($c\delta\tau = \pm 2000$ μm), while the numbers of the separate counts by two detectors were 1.08×10^7 and 1.07×10^7. At each point of the delay we divided the former number by the product of the latter. In Fig. 2 the data are then shown normalized so that their values at the largest delay may be equal to 1.

In Eq. (12) we assumed that the excitation laser pulse was δ function and the dye fluorescence decayed exponentially. Actually, the pulse width of the incident laser pulse is 650 fs, which is longer than the decay time 100 fs of the dye fluorescence, therefore, the detected coincidence curve of the fluorescence should be the convolution of the laser and fluorescence envelopes. We approximate the envelope of the laser pulse to be Gaussian. In calculating the convolution, we divide the excitation laser pulse into short time meshes and assume each part as δ function excitation. Fluorescence pulse is the sum of the slightly time-shifted field components which follow Eq. (14). We determined the parameter τ_L by the least squares fitting to the experimental data. We thus get a deconvolved τ_L of 90 fs, which gives the minimum variance to the fitting of the curve to the data. This value coincides well with the decay time 100 fs of NB/DMA fluorescence reported by Kobayashi *et al.* [9].

The result of the calculation is shown by the curve in Fig. 2. The full width at half maximum of the dip of the curve is about 730 fs. It clearly shows that this present scheme can measure the pulse width of ultrashort incoherent light pulse. In contrast to the above case, when we measured coincidence count between adjoining pulses by stretching the distance from BS2 to the TAC by the delay cable, we found that the dip of the pulse width disappeared, but the dip of the coherence time $\tau_c = 370$ fs was observed. These results will be published elsewhere [10].

In conclusion, we showed that one can measure separately the ultrashort coherence time and the pulse envelope or width of pulse field with two-photon interference. We showed especially that, by taking correlation between adjoining pulses, one can measure the intensity profile or the lifetime of incoherent fluorescence from independent sources. This is the first observation of the fourth-order interference separated from the second-order interference, to the knowledge of the authors. This method is expected to be useful for ultrashort pulse measurement in the uv or x-ray regions.

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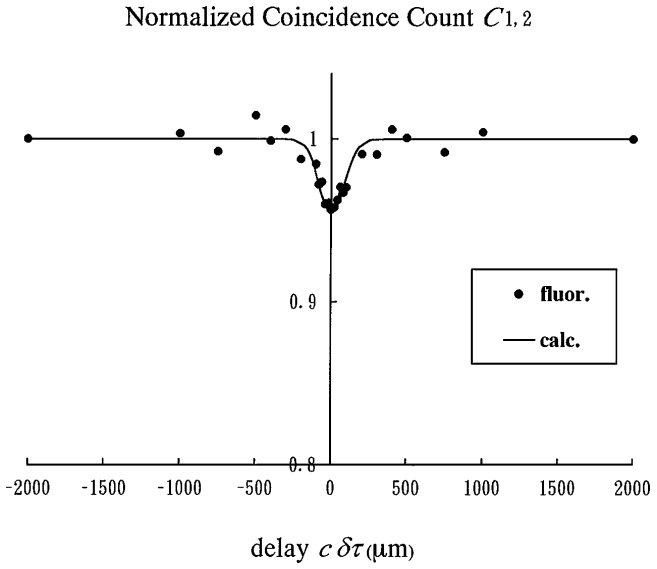

FIG. 2. Plot of normalized coincidence counts for the ultrashort fluorescence pulse (dots) and the calculated curve versus the delay of the corner cube. The dip shows an autocorrelation function of the fluorescence envelope separated from the term which gives the coherence time.

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