ELSEVIER

Ultramicroscopy 57 (1995) 169–172

ultramicroscopy

Characterization of the temporal behavior of ultrashort pulses emerging from a near-field fiber probe

Robert C. Dunn, X. Sunney Xie *

Pacific Northwest Laboratory [1], Molecular Science Research Center, P.O. Box 999, Mailstop K2-14, Richland, WA 99352, USA

Received 1 November 1993; accepted 21 December 1993

Abstract

A photon correlation technique has been used to characterize the temporal behavior of a picosecond pulse train emerging from an aluminum-coated near-field fiber probe with a 100 nm aperture.

1. Introduction

Near-field optical microscopy combines the wealth of optical spectroscopic techniques with an unprecedented spatial resolution [1]. One particular advantage of near-field optical microscopy over other scanning probe microscopies is the high time resolution afforded by optical measurements, allowing the study of fast dynamical processes.

In our effort towards conducting time-resolved molecular spectroscopy at the nanometer dimension, the natural first step is to characterize the temporal behavior of the short light pulses emerging from the aluminum-coated fiber tip. Unfortunately, conventional autocorrelation techniques using second harmonic generation in non-linear crystals is difficult for the low photon flux (10^{10} photons/s) emerging from these tips. While up-conversion gating techniques can be used to characterize the pulse width [2], we have employed a recently developed method based on a photon correlation technique [3,4]. The technique, which does not rely on non-linear crystals, has several advantages, such as a photon counting sensitivity for weak beams and an unlimited time resolution. We have used this technique to evaluate the pulse width of a picosecond pulse train passing through aluminum-coated fiber tips. The fiber tips are fabricated using the standard technique developed by Betzig [5]. Typical fiber tips have a 100 nm aperture and deliver approximately 3×10^{10} photons per second when 1 mW of 633 nm laser power is coupled into the fiber.

2. Experimental principle and design

Fig. 1 shows a schematic of the photon correlation experiment. A high repetition rate picosecond pulse train emerging from the Al-coated fiber tip is collected with a high numerical aper-

* Corresponding author.

[1] Pacific Northwest Laboratory is operated for the US Department of Energy by Battelle Memorial Institute under Contract No. DE-AC06-76RLO 1830.

SSDI 0304-3991(94)00130-8

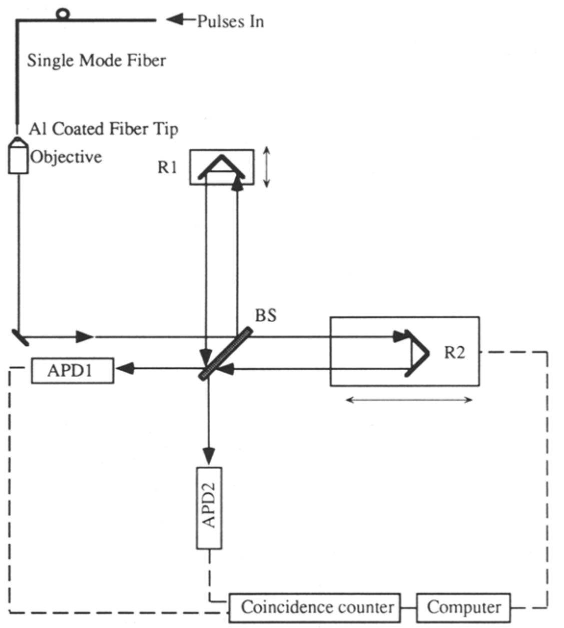

Fig. 1. Schematic of the photon correlation technique for characterization of ultrashort optical pulses. See text for details.

ture (N.A. = 1.3) microscope objective, attenuated with a neutral density filter and split with a 50% beam splitter to two retroreflectors (R1 and R2). R2 is mounted on an optical delay line that changes the path length of one of the two arms of the interferometer. For reasons described below, R1 is mounted on a piezo-transducer driven with a random noise generator. The noise bandwidth is 0.01–1000 Hz and the rms amplitude of R1 travel is several wavelengths. The reflected beams from R1 and R2 are overlapped and split again at the 50% beam splitter, and the two resulting beams are detected with two photon counting avalanche photodiodes (APD1 and APD2, EG& G Model SPCM-200). The output of each APD is sent through a discriminator (Phillips Scientific, Model 710) and into an AND gate (Phillips Scientific, Model 752) which detects the coincidence of counts received at APD1 and APD2. Each coincidence count recorded indicates a simultaneous detection event at the two APDs within a laser pulse. The coincidence counts are recorded as a function of the path length difference between the two arms of the interferometer. It is important to stress that it is the variation in the path lengths which determines the time resolution of the experiment and not the speed of the detectors, analogous to conventional autocorrelation techniques.

According to Ref. [3] for a Gaussian pulse

$$E(t) = C \exp\left(-t^2/\tau_p^2\right) \exp(-i\omega t), \tag{1}$$

the coincidence counts are given by

$$K\left[1 - \tfrac{1}{2} \exp\left(-\tau^2/\tau_p^2\right)\right], \tag{2}$$

where τ is the difference between path lengths of the two arms, τ_p is the pulse width, and C and K are proportionality constants. The coincidence count rate decreases by half at $\tau = 0$ (pulses overlapped), and the width of the dip in coincidence measures the pulse width. This phenomena is the result of an interference effect that can be rationalized as follows.

First, consider the case where we are no longer detecting coincidence and are just recording the counts from a single detector (APD1 or APD2). For this experiment, the piezo connected to R1 would be turned off so that R1 remained fixed. Fig. 2a shows the expected counts at the detector (APD1 or APD2) as a function of τ. An envelope

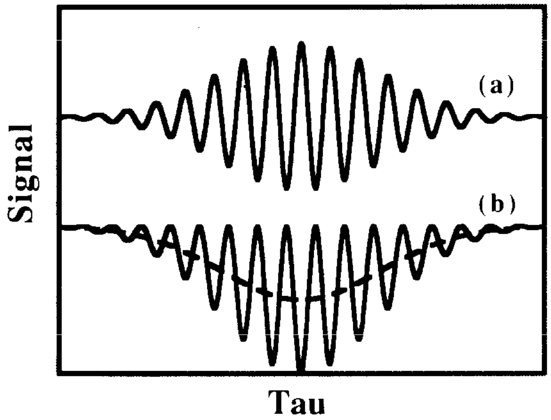

Fig. 2. (a) Calculated signal at APD1 (or APD2) as a function of the optical path difference of the two interferometric arms. (b) Calculated coincidence signal of APD1 and APD2 as a function of the optical path length difference. The dashed line is the signal when a small random phase delay is added into one arm of the interferometer.

of fringes due to constructive and destructive interferences are observed as the delay line is scanned. Although the envelope of the fringes can be used to determine the pulse width, this method would require long data acquisition times and interferometric accuracy in the scanning of the delay line. Next, consider the case where the coincidence between events at APD1 and APD2 is recorded. For the moment, the piezo connected to R1 remains off. Fig. 2b (solid line) shows the coincidence counts between APD1 and APD2 as a function of τ. Now, instead of the bipolar envelope observed in Fig. 2a, there is a drop in signal when the pulses overlap that can be explained as follows. When one photon from R1 and another photon from R2 are not temporally overlapped, each can get to the two detectors with the same 50% probability. However, when they are temporally overlapped at the beam splitter they interfere with each other. Both constructive and destructive interference results in both photons going to only one detector (APD1 or APD2), decreasing the number of coincidence events. The virtue of coincidence detection is that the coincidence counts always tend to decrease when the pulses overlap, rather than oscillating around a fixed value as in Fig. 2a. Finally, a small random motion provided by the noise driven piezo is applied to R1 to average through the fringes in the coincidence counts, making interferometric accuracy unnecessary. This results in the dashed curve in Fig. 2b which is identical to the intensity autocorrelation of the pulses.

A Coherent 702 dye laser operating with rhodamine 6G was synchronously pumped by a frequency-doubled Nd:YAG (Coherent Antares). With a three plate birefringent filter inside the dye laser cavity, the center wavelength was tuned to 590 nm. The pulse width of the dye laser output was measured with a conventional autocorrelator (InRad) giving a FWHM of 10 ps. The 76 MHz pulse train was attenuated down to 1 mW average power before being coupled into the 1 m long single mode fiber (633 nm, Corning). The light emerging from the 100 nm aluminum-coated fiber tip on the other end was attenuated so that each APD received 3×10^5 counts/s. The coincidence counts with unequal arms was approximately 500 counts/s. A dip in the coincidence of 40%–45% was usually observed when the pulses overlapped ($\tau = 0$). The fact that it is smaller than the theoretical limit of 50% probably reflects an imperfect beamsplitter and alignment. P polarization was used for these experiments.

3. Results

Fig. 3 shows the photon correlation data for (a) an uncoated fiber tip and (b) an aluminum-coated fiber tip. Both traces show the same FWHM of 10 ps, which is identical to that of the direct autocorrelation of the dye laser output. Furthermore, this width does not change as the tip–sample distance is changed using a sheer force feed-back mechanism. [5] These results demonstrate the temporal intensity profile of the picosecond pulses coming out from the near-field fiber probe remains unchanged. In particular, non-linear processes such as self phase modula-

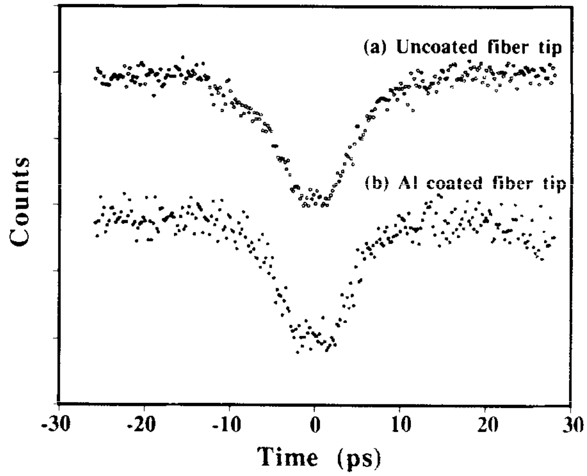

Fig. 3. (a) Experimental data on an uncoated fiber tip (1 m long). (b) Experimental data on an aluminum coated fiber tip. The FWHM of both traces are 10 ps, which is identical to that of the input laser pulses measured with a conventional autocorrelation technique.

tion inside the fiber does not affect the spectral and temporal behavior of the picosecond pulses.

It is worth pointing out that, compared to conventional second harmonic autocorrelation techniques, this photon correlation technique is not particularly sensitive to chirping [3]. This is less problematic for picosecond pulses than femtosecond pulses. However, we are currently implementing the wavelength-resolved version of this photon correlation technique in a fashion similar to previous wavelength-resolved correlation techniques [6,7]. This will allow direct determination of the amplitude and phase of femtosecond light pulses with photon counting sensitivity and high time resolution. We believe that this and related photon correlation techniques are particularly suitable for near-field optics.

4. Conclusions

We have used a novel photon correlation technique to examine the temporal behavior of a picosecond pulse train emerging from a near-field fiber probe. This technique does not require non-linear crystals and has photon counting sensitivity particularly suitable for near-field applications. We conclude that the aluminum-coated near-field fiber tip does not change the temporal behavior of picosecond pulses.

Acknowledgments

We thank Gordon Anderson for assistance on electronic design, Errol V. Allen for assistance on fiber tip fabrication, Gary Holtom for many helpful discussions, and Alan Joly and Don Friedrich for the loan of the dye laser used in this experiment. This work was supported by the Chemical Sciences Division in the Office of Basic Energy Sciences of the US Department of Energy at Pacific Northwest Laboratory under Contract No. DE-AC06-76RLO 1830.

References

[1] E. Betzig and J.K. Trautman, Science 257 (1992) 189.
[2] S. Smith, B.G. Orr, R. Kopelman and T. Norris, Ultramicroscopy 57 (1995) 173.
[3] Y. Miyamoto, T. Kuga, M. Baba and M. Matsuoka, Opt. Lett. 18 (1993) 900.
[4] Z.Y. Ou, E.C. Gage, B.E. Magill and L. Mandel, J. Opt. Soc. Am. B 6 (1989) 100.
[5] E. Betzig, P.L. Finn and J.S. Weiner, Appl. Phys. Lett. 60 (1992) 2486.
[6] J.L.A. Chilla and O.E. Martinez, Opt. Lett. 16 (1991) 39.
[7] C. Yan and J. Driels, J. Opt. Soc. Am. B 8 (1991) 1259.