Characterization of femtosecond light pulses coupled to hollow-pyramid near-field probes: Localization in space and time

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We perform the *in situ* characterization, by second-order optical autocorrelation, of femtosecond pulses transmitted by near-field optical probes. We demonstrate that transmission through hollow pyramid probes with diameter down to 65 nm has negligible effects on the duration of pulses as short as 30 fs. We also show that such probes allow obtaining, at their output, sufficient peak power to perform nonlinear optical experiments in the near field on such a space and time scale. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1852088]

Modern materials science relies heavily on spectroscopy and microscopy. Combining the spatial resolution of near-field scanning optical microscopy (NSOM) and the temporal resolution of femtosecond spectroscopy is of great interest for the study of a wide variety of materials with spatial variations of the optical properties occurring with femtosecond dynamics.[1–6] The possibility of concentrating the peak power of an ultrafast excitation on a nanosystem is currently the object of debate.[7–12] Theoretical investigations indicate temporal broadening and loss of spatial resolution in some conditions, but still predict the feasibility of light localization in space and in time (on the nanometer scale for pulse durations down to 2 fs) in appropriate systems.[9–12] Experimentally a few configurations have been reported, in which the near-field probe consists of a metal-coated tapered optical fiber. The temporal structure of ultrashort pulses transmitted by near-field probes has been characterized in the terahertz region[13,14] as well as in the optical region using photon correlation[15] and crosscorrelation,[16,17] but pulse widths shorter than $\sim$81 fs were never achieved. The pulse peak power at the probe output was, however, insufficient to perform a direct optical autocorrelation measurement, due to the low throughput (typically from 10^{-6} to 10^{-4} for 50 to 150 nm apertures) combined with the low value of the maximum input power which is allowed to avoid thermal damage ($<$1 mW). In practical implementations of time-resolved NSOM using metal-coated fiber probes, the spatial resolution could never be pushed below 100 nm[4] and a temporal resolution of $\sim$150 fs was inferred from the rise time of a pump-probe signal.[2,5]

Recently, a class of near-field aperture probes has been introduced, based on a silicon or silicon nitride cantilever, similar to the ones used in atomic force microscopy (AFM), with a hollow pyramid as the tip.[18–20] The pyramid is metal-coated (usually by aluminium or gold) and a hole is produced at the pyramid apex, with diameter ranging from 40 to 200 nm. As shown in continuous-wave experiments, these cantilevered tips offer several advantages compared to metal-coated tapered optical fibers, namely: (i) the larger taper angle produces higher power throughputs; (ii) the lower absorption allows to couple higher average power before the onset of thermal damage; (iii) tip-sample distance stabilization methods used in AFM, such as the tapping mode, can be employed, thus ensuring longer probe lifetimes. Moreover pulse chirping due to propagation in the fiber is avoided and the pulses traverse a negligible optical thickness.

In this letter we report on the characterization, by second-order optical autocorrelation, of the femtosecond pulses transmitted by this kind of near-field optical probes. We demonstrate that transmission through the NSOM-cantilever probes has negligible effects on pulse duration, with apertures down to 65 nm diameter and pulse durations down to 30 fs, so that it becomes possible to achieve simultaneously high spatial and temporal localization. We also show that this kind of probes allow obtaining an output peak power which is sufficient to perform nonlinear optical experiments in the near field.

A schematic of the experimental setup is shown in Fig. 1. The laser system is a Ti:sapphire oscillator in a standard asymmetric cavity for Kerr-lens mode locking. The cavity length is stretched[21] by a 1:1 telescope (made of two $R=2000$ mm mirrors) decreasing the repetition rate down to 26 MHz. For a given average power, as limited by thermal effects in the near-field probe, this allows an increase of the peak power by a factor of $\sim$4 with respect to a standard 100 MHz cavity, and a corresponding enhancement of nonlinear optical effects. The laser generates pulses at $\sim$800 nm cen-

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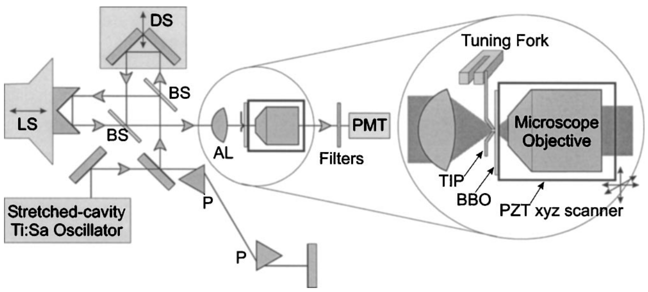

FIG. 1. Experimental setup: DS, delay stage; LS, loudspeaker; BS, beam splitters; *P*, Brewster-cut fused silica prisms; AL, aspheric lens; PMT, photomultiplier; PZT, piezotranslator.

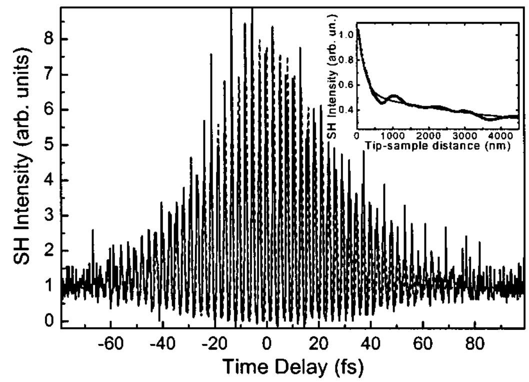

FIG. 2. Solid line: interferometric fringe-resolved autocorrelation of the laser pulses transmitted by a 100 nm NSOM cantilever tip; dashed line, fit. Inset shows an approach curve of the generated SH together with a biexponential fit.

tral wavelength with 10 THz bandwidth (corresponding to a transform-limited pulse duration of 27 fs) and energy up to 20 nJ. The pulses are sent to a compressor consisting of a double pass in a Brewster-cut fused-silica prism pair and then are coupled into a balanced Michelson interferometer. The end mirrors on one arm of the interferometer are mounted on a precision translation stage allowing performing slow scan autocorrelations, while those on the other arm are mounted on a loudspeaker enabling real-time autocorrelations. The collinear pulses at the exit of the interferometer are focused by a 0.5 numerical aperture (NA) aspheric lens onto the NSOM cantilevered tip probe. The lens is completely filled by the beam giving a focused spot size of $\sim 2\ \mu$m, much smaller than the base of the pyramid. This provides a throughput of $\sim 10^{-5}$–$\sim 10^{-3}$, for 50–150 nm apertures respectively, i.e., an order of magnitude higher than the corresponding values obtained from fiber probes. Furthermore, we have observed that the input power could be increased up to tens of milliwatts without damage problems. A nonlinear crystal mounted on a hollow piezoscanner tube allowing *xyz* translation can be brought in the near field of the tip. The cantilever is mounted on a quartz tuning fork dithered at its resonance frequency of about 32 kHz. Tip-crystal distance is stabilized using the AFM tapping mode with an oscillation amplitude of 100 nm peak to peak, which limits the minimum average tip-sample distance to 50 nm. The transmitted light is collected by a 0.6 NA microscope objective and focused onto a photomultiplier tube. The nonlinear crystal used for the pulse characterization experiments was a β-barium borate (BBO) with 100 μm thickness, cut for type I phase matching at 800 nm. Suitable filters allow completely rejecting the fundamental wavelength light and transmitting only the second harmonic (SH) generated in the BBO.

The first experiments were performed removing the tip from the beam path: in this case the setup easily allowed obtaining the fringe-resolved autocorrelation (FRAC) by real-time observation on a digital oscilloscope while vibrating the loudspeaker. We compared experimental FRAC with numerical FRACs of pulses obtained from the experimental pulse spectrum adding a residual second order dispersion. The best fits were obtained for a $\sim 100\ \mathrm{fs}^2$ residual group delay dispersion (GDD), and the corresponding pulse has a duration of 30 fs, which is close to the transform-limited value of 27 fs. This residual chirp can be ascribed to nonperfect optimization of the precompressor and to residual third-order dispersion; nonetheless, this indicates that the prism compressor is able to satisfactorily compensate for the phase distortions introduced by the output coupler, the beam splitters and the focusing optics.

Next the laser was coupled into a 100 nm tip, made of silicon covered with aluminium,[22] kept in contact with the crystal surface. A maximum total power of 20 mW was used, observing no tip degradation. The SH signal at the detector was still sufficient to perform a real-time FRAC measurement. These data are shown in Fig. 2 together with the best fit obtained again adding a $100\ \mathrm{fs}^2$ GDD. The agreement with the fit and the good fringe visibility out to the tails of the FRAC indicates that no significant temporal distortion has occurred to the pulse upon transmission through the tip.

Withdrawal of the BBO crystal from the tip caused a steep drop of the generated SH, as shown by the approach curve reported in the inset of Fig. 2. The short distance steep decay has a characteristic length of 180 nm, while a longer distance decay due to far field components occurs with decay length of 5.5 μm. The longer decay component is very close to the one obtained without the tip (6.0 μm). We note that the concept of phase matching, commonly used for SH generation, does not apply to the near-field region[23] because of broad angular spectrum of **k** vectors and the short spatial extension of the near-field region with respect to the coherence length of the nonlinear material.

Finally, the laser beam was coupled into a 65 nm diameter tip, made of silicon nitride covered with gold.[19,20] Given the strong dependence of the near-field power P on the tip aperture diameter $d (P \propto d^6)$ the SH signal actually dropped to a level preventing its direct detection on an oscilloscope. However, upon modulating the input beam power and using lock-in detection, it was still possible to measure the SH signal. By slowly scanning the translation stage and low-pass filtering the fringes (by a fast but small dithering of the loudspeaker arm), we could perform a collinear intensity autocorrelation. One of such measurements is shown in Fig. 3, together with a numerical fit; also in this case the best fit was obtained by considering a residual GDD of $\approx 100\ \mathrm{fs}^2$. This result shows that even the 65 nm tip does not introduce any significant perturbation in the pulse temporal structure. It is worthwhile noting that the insertion of the NSOM aperture, acting as a spatial filter on the incident Gaussian beams, optimized the overlap of the beams.

In conclusion, we performed the characterization of femtosecond pulses transmitted by near-field optical probes by second-order autocorrelation measurements. We demonstrate

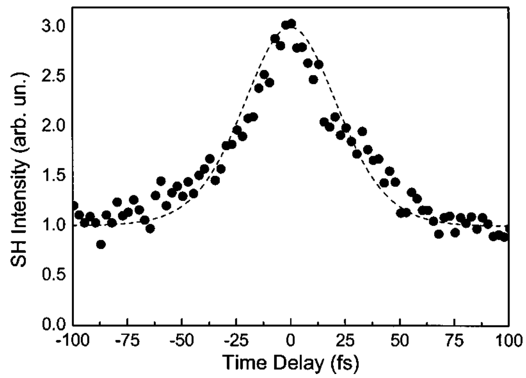

FIG. 3. Dots: collinear intensity autocorrelation of the laser pulses transmitted by a 65 nm NSOM cantilever tip; dashed line, fit.

that transmission through probes with diameter down to 65 nm has negligible effects on pulses with duration around 30 fs, thus allowing simultaneous strong localization both in space and time. We also show that cantilevered tip probes allow obtaining, at their output, sufficient peak power to perform nonlinear optical experiments in the near field, combining high spatial and temporal resolution. Work is in progress to extend this characterization to aperture sizes <50 nm and pulse durations <20 fs.

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