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Femtosecond Pulse Coupling to Near-Field Cantilevered Probes

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A novel nanoprobe for space- and time-resolved nonlinear optical spectroscopy has been developed. The nanoprobe is based on aperture-type SNOM using cantilevered sensors ("SNOM cantilevers"), capable to couple much higher power to the sample as compared to usual tapered fiber probes, along with negligible effect on femtosecond pulse duration, as directly assessed by second-order fringe-resolved autocorrelation in the near field. The use of such nanosource for pump-probe near-field spectroscopy promises to improve both time and space resolution with respect to the current state-of-the-art setups.

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I. INTRODUCTION

A very promising area of research in the physics of nano and mesoscopic structures combines the temporal resolution allowed by femtosecond pump-probe spectroscopy and the spatial resolution enabled by Scanning Near-field Optical Microscopy (SNOM). The most commonly used SNOM configuration is the so-called aperture-SNOM (a-SNOM), employing as the nanoprobe an aperture produced in a metal-coated tapered optical fiber [1]. The main drawback of such SNOM probes is their low throughput and/or total available power for excitation, where as throughput we mean the available near-field power divided by the power incident on the probe. The typical throughput is around 10^{-6} for 50-nm apertures, and the incident power is limited to about 1 mW by thermal damage of the aperture, yielding at the probe output a typical power $P_{max} = 1$ nW. A further constraint is due to the metal skin depth that limits the effective aperture size to about 20 nm in the visible.

In this paper we explore a novel configuration for a-SNOM in combination with femtosecond laser pulses, with the purpose of improving both the spatio-temporal resolution and the intensity in the near field. We use new-generation apertures for a-SNOM, the so-called SNOM cantilevers [2]. They consist of a silicon cantilever with a hollow aluminum-coated pyramid as a tip; the pyramid has a small aperture (50 − 100 nm diameter) drilled at its apex. For such probes, the skin depth resolution limit of 20 nm is still present, but the obtainable throughput can be higher by an order of magnitude compared to fiber probes of the same size, due to the larger taper angle. Furthermore the input power can be about two orders of magnitude higher than with fiber probes, henceforth enabling a series of nonlinear spectroscopies, like femtosecond pump-probe, that up to now were limited to space resolutions of 150 − 200 nm [3]. In addition, SNOM cantilever probes allow the use of tip-sample distance stabilization methods employed in Atomic Force Microscopy (AFM), such as the tapping mode, thus ensuring longer probe lifetimes. A further important advantage of SNOM cantilevers for femtosecond pump-probe spectroscopy is the absence of chirping due to material dispersion in the fiber, that would require suitable precompression and would limit the achievable pulse duration.

In this paper we first describe the setup realized and show preliminary results, including direct measurement of the pulsewidth at the output of the cantilever probe, demonstrating no temporal broadening for pulse durations as short as 30 fs. We then discuss the results in view of the future application with special attention to local femtosecond pump-probe spectroscopy.

II. EXPERIMENT

In order to increase the pulse energy for a given average power at the output of the SNOM tip, we have

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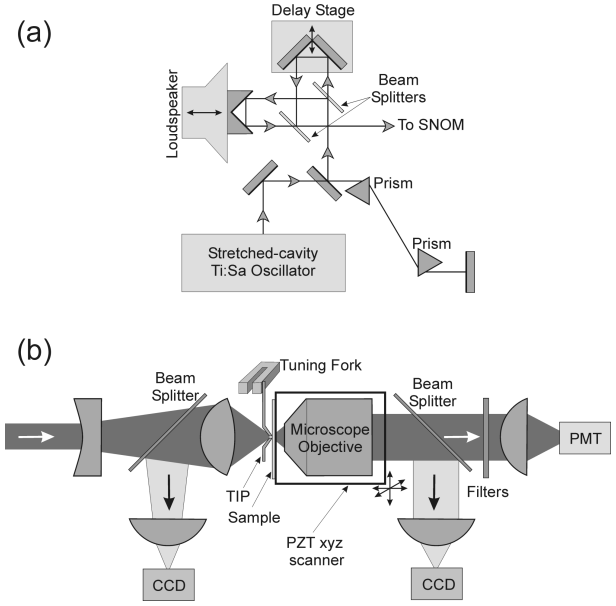

Fig. 1. (a) Sketch of the autocorrelator; (b) Sketch of the aperture-SNOM setup based on SNOM cantilevers.

developed a low repetition rate Kerr-lens mode-locked Ti:Sapphire oscillator, by extending the cavity length of a "standard" oscillator introducing a 1:1 folded intracavity telescope in one arm [4]. The telescope has a unity $ABCD$ matrix so as to keep unchanged the optical stability limits of the cavity. The reduced repetition rate of 26 MHz results in an increase in pulse energy, for a given average power, with significant boosting of induced non-linear effects while maintaining safe power levels. The laser produces $20 - 30$ fs pulses at 800 nm central wavelength with an energy up to 20 nJ. A prism pre-compressor (sketched in Fig. 1(a)) compensates for the pulse broadening as arising from the optical components in the beam path from the laser to the SNOM tip. The pulses are coupled into a balanced Michelson interferometer (see Fig. 1(a)). The end mirrors on one arm of the interferometer are mounted on a precision translation stage allowing performing slow scans of the delay between the two pulse replicas, while those on the other arm are mounted on a loudspeaker enabling fast scans. The collinear pulses at the exit of the interferometer are then fed into the a-SNOM setup shown in Figure 1b. The laser beam is expanded by a diverging lens in order to fill the aspheric focusing lens (NA = 0.5, diameter 10 mm, working distance 6 mm) and obtain on the SNOM cantilever a focused spot size of $\sim$2 μm, much smaller than the base of the pyramid. The reflection is imaged onto a CCD camera for probe alignment. Transmitted light is collected by a microscope objective (40x, NA = 0.65) placed inside an hollow piezotube holding the sample; for a fine adjustment of the coupling, the output beam is split and imaged on a second CCD. Filters can be inserted on the light path towards the detector (a miniaturized photomultiplier, PMT) in order to cut off the pump wavelength in pump-probe experiments.

The tip/sample distance stabilization in our a-SNOM is achieved by non-optical methods in order not to introduce spurious light sources deleterious for spectroscopy experiments. Namely, quartz tuning forks are used to vibrate the tip perpendicularly to the sample surface to realize dynamic force microscopy stabilization of amplitude [5] or frequency [6] by controlling the tip-sample distance. As in AFM, such adjustment tracks the topography reliefs upon lateral sample scanning. The SNOM cantilevers are glued to one arm of the tuning fork (similarly to Ref. [7]) in such a way to have enough clearance for both tip illumination and sample approach.

III. RESULTS AND DISCUSSION

As a first application of the time-resolved a-SNOM, we measured the pulse duration of the femtosecond laser after propagating through the aperture. To this purpose we have used as a sample a nonlinear β-barium borate (BBO) crystal for generating the second harmonic (SH): by sweeping the time delay using the loudspeaker and visualizing the SH signal with a digitizing oscilloscope, we could obtain a collinear Fringe-Resolved Auto-Correlation (FRAC).

First we removed the tip from the beam path. 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$150-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. Next the laser was coupled into a 100-nm tip, made of silicon covered with aluminium [8], kept in contact with the crystal surface. A maximum total input

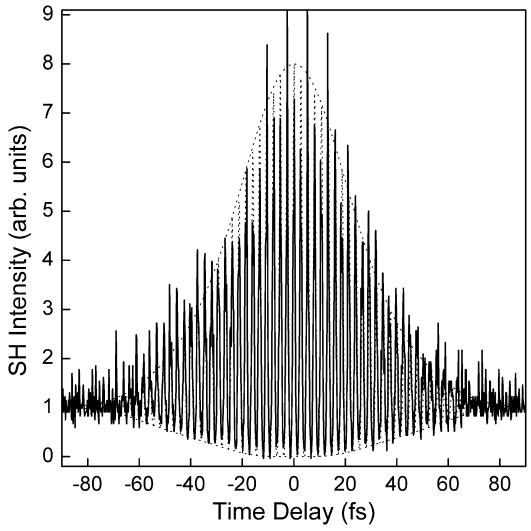

Fig. 2. Second-order FRAC of a 35-fs pulse propagating through a 100-nm aperture.

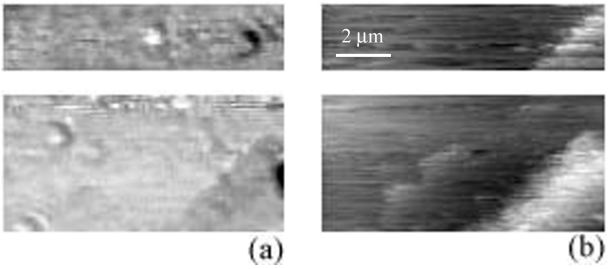

Fig. 3. Two maps of the SH emission (a) from the surface of a BBO crystal when scanned by the tip of our aperture-SNOM with an aperture of about 200 nm, and (b) corresponding topography.

power of 20 mW was used, observing no tip degradation; the measured throughput for this tip was 4×10^{-5}. 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 150-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.

To demonstrate the operation of our a-SNOM, we have mapped the local SH generation on the surface of the BBO crystal upon excitation by the localized fs-pulses. By using a somehow larger aperture ($\sim$ 200 nm), due to wear of the probe after many scans, we have obtained a SH level high enough to allow imaging with a remarkable S/N ratio in typical AFM scanning times. In Fig. 3 we show a couple of SH maps of the BBO surface (a), and the corresponding topographic reliefs (b) acquired simultaneously. Features of the order of 500 nm are clearly visible in the SH map, even on flat areas. The same image acquired with the 100-nm tip before being worn off revealed a uniform SH signal map within the noise. We believe that the features observed in Fig. 3 are given by buried regions that are not reachable by the near field extension of the smaller probe, while the larger apertures have considerable far-field emission and thus permit imaging of deeper structures, although with limited lateral resolution.

IV. CONCLUSION

In this work we presented an experimental apparatus coupling femtosecond light pulses to an a-SNOM based on hollow-tip cantilevers. This setup allows to boost the near-field power by nearly three orders of magnitude compared to standard metal-coated fiber probes; in addition, much better temporal resolutions can be achieved. As a demonstration of the high peak power available at the probe output, we were able to perform a direct *in situ* characterization of the pulsewidth by second-order autocorrelation. We also proved that propagation through the tip does not significantly lengthen the pulses for durations down to $\approx$ 30 fs. High contrast maps of second-harmonic generation in a nonlinear crystal have been shown.

As a perspective, besides time and space resolved spectroscopy, we can also envisage other different applications for our a-SNOM. The high peak powers should enable local two-photon PhotoLuminescence (PL) which, up to now, has been performed only with aperturless-type SNOM [9]. Moreover femtosecond pulses are widely used for material micromachining, because of rapid energy deposition and fast ablation without heat and shock affected zones; in combination with a SNOM, the size of the micromachined structures can be brought down to the sub-micron range. While previous experiments reported feature sizes of the order of 200 nm [10], with our instrument we can expect to push feature size below 100 nm.

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