Ultrasmall bullets of light—focusing few-cycle light pulses to the diffraction limit

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We demonstrate an essentially dispersion-free and diffraction-limited focusing of few-cycle laser pulses through all-reflective microscope objectives. By transmitting 6-fs-pulses from a Ti:sapphire oscillator through an all-reflective 0.5 NA objective, we reach a focus with a beam diameter of 1.0 μm, preserving the time structure of the pulses. The temporal and spatial pulse profile is recorded simultaneously using a novel tip-enhanced electron emission autocorrelator, indicating a focal volume of these pulses of only 1.8 μm³. We anticipate that the demonstrated technique is of considerable interest for inducing and probing optical nonlinearities of individual nanostructures.

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1. Introduction

The ultrafast optical nonlinearities of metallic nanostructures and metal-dielectric hybrid materials currently attract substantial interest [1–5]. In such systems, optical nonlinearities are often governed by the very strong local enhancement of the electromagnetic field in the vicinity of the metallic nanostructure [6–9]. This field enhancement is so pronounced that higher order optical nonlinearities such as, e.g., higher harmonic generation [10] or field-induced electron emission [11,12], are observed at field intensities which are orders of magnitude higher than typically needed in standard gaseous or bulk materials. Therefore these phenomena can readily be studied in experiments with high repetition-rate laser oscillators [10–12]. It is expected that all these extreme nonlinear optical phenomena depend substantially on the carrier envelope phase (CEP) of the laser [13,14] when being induced...
with few-cycle pulses. CEP-variation may thus be an important parameter for optimizing the efficiency of these nonlinearities and/or to generate high-harmonic or electron pulses with short, possibly attosecond pulse durations [15,16]. Such experiments, which have rarely performed with pulses directly deduced from a laser oscillator [17], inevitably require an essentially dispersion-free focusing of few-cycle laser pulses to the diffraction limit.

This is a rather challenging task when using conventional, high numerical aperture (NA) glass microscope objectives [18–20] because of the rather large spectral bandwidth of such few cycle pulses. Different effects limit the time structure in the focus. Achromatic dispersion effects on the spatial profile are generally already well minimized in high-quality lens systems. The (beam-size averaged) second-order dispersion \( \frac{\partial^2 \varphi}{\partial \varphi \omega^2} \) (where \( \varphi \) is the spectral phase of the pulse and \( \omega \) is frequency) and higher-order dispersion effects induced by such objectives are large (typically of the order of thousand fs\(^2\) [19]) but can in principle readily be pre-compensated by combining prism compressors, grating compressors and/or customized chirped mirrors. A much more severe problem is the essentially unavoidable radial spatial chirp [21] that the pulse experiences since beam rays incident at different distances from the optical axis will necessarily experience different dispersion effects [22]. Radial chirp compensation requires sophisticated dispersion management techniques using, e.g., deformable mirrors [23] or spatial light modulators. So far, to our knowledge, the shortest pulse durations that have been reached in the focus of a high NA (NA = 0.85) glass objective are larger than 9-fs [18]. Such a short pulse duration could only be reached by reducing the entrance aperture diameter by 60%.

In principle, many of these dispersion problems can be overcome by using all-reflective optics, e.g., high NA parabolic mirrors or Schwarzschild objective. Using a combination of deformable mirrors and an off-axis parabolic mirror high-intensity sub-10-fs laser pulses have been focused to a spot size of 1.2 x 1.7 \( \mu \)m\(^2\), without, however, providing a direct measurement of the pulse duration in the focal region [24]. In this paper, we experimentally study the focusing of few-cycle laser pulses through high NA objectives. Using an all-reflective objective, we demonstrate essentially dispersion-free focusing of \( \tau = 6 \) fs-pulses to a beam diameter of \( A = 1.0 \mu \)m\(^2\). The space and time structure of the focused pulse is locally probed using a tip-enhanced electron emission autocorrelator. The measured parameters correspond to a pulse focal volume \( V = c \cdot \tau \cdot A = 1.8 \mu \)m\(^3\), to our knowledge the smallest bullets of light which have been generated and characterized in a linear optical microscope so far.

2. Experimental methods

The experimental setup used in this work is schematically shown in Fig. 1. Few-cycle laser pulses with an energy of 2.6 nJ are generated in a commercial Ti:sapphire oscillator (Femtolasers Rainbow) operating at a repetition rate of 82 MHz. Its spectrum extends approximately from 650 to 1050 nm (Fig. 2(a), inset) and a bandwidth-limited pulse duration of 5.9 fs is deduced from interferometric autocorrelation (IAC) measurements (Fig. 2(a)). The dotted line in Fig. 2 shows a simulation of the IAC trace expected for chirp-free, bandwidth-limited pulses with a spectrum shown in the inset. The pulse dispersion is controlled by a pair of chirped mirrors with a group delay dispersion (GDD) of \( \sim 45 \) fs\(^2\)/bounce (Femtolasers GSM014). A pair of wedges (Femtolasers, Suprasil, 2°48') is used to fine-tune the dispersion. Appropriately pre-compensated pulses (< \( \sim 200 \) fs\(^2\)) enter a dispersion-balanced Michelson interferometer with low-dispersion broadband dielectric beamsplitters. In this interferometer, a collinearly propagating pair of pulses with variable time delay \( \tau \) is generated.

The pulse delay is controlled using a hardware-linearized single-axis piezo scanner (Physik Instrumente P-621.1CD PI Hera). Fluctuations of \( \tau \) due to mechanical vibrations of the interferometer and the finite precision of the piezo scanner are less than 50 as. The pulse...
Fig. 1. Schematic of the experimental set-up. Few-cycle pulses with 5.9-fs-duration and a spectrum extending from 650 to 1050 nm are generated in a Ti:sapphire oscillator. Pulse dispersion is controlled by a set of chirped mirrors and a pair of wedges. A pair of collinearly propagating pulses is generated in a dispersion-balanced Michelson interferometer, expanded in beam size in a reflective telescope and focused through different microscope objectives. Their temporal intensity profile is measured by an interferometric autocorrelator before and after focusing. The spatial intensity profile is probed by scanning an aluminum-coated near-field optical fiber, mounted on a piezo stage, through the focus.

The pulse pair is expanded to a beam size of 20 mm in an all-reflective Kepler telescope. We investigate the focusing properties of two different objectives, (i) an extra long working distance 60x glass microscope objective (Nikon, MPlan 60, ELWD 0.7) with a numerical aperture (NA) of 0.7 and a working distance of 4.9 mm and (ii) an all-reflective, aluminum-coated 36x Schwarzschild (Davin Optronics, 5004-000) microscope objective with a NA of 0.5, 36x magnification and a working distance of 8.6 mm. The spatial intensity profile of the focused spot, we locally collect the laser light through an aluminum-coated near-field scanning optical (NSOM) fiber probe (VEECO) with an aperture diameter of ~300 nm, fabricated by focused ion beam milling. The tip is mounted on a hardware-linearized three-axis piezo stage (Physik Instrumente NanoCube) with a positioning accuracy of better than 10 nm. The intensity of the collected laser light is detected with a photomultiplier tube (PMT) while scanning the tip through the focus. To record the time structure of the focused pulses, the tip is replaced with a 10 µm thick BBO crystal and IAC traces are recorded in the laser focus. The spatio-temporal profile of the laser pulses is measured by mounting the objectives inside a high vacuum chamber \((p = 5 \times 10^{-7} \text{ mbar})\) and by focusing the laser pulses onto a sharp chemically-etched gold tip with an aperture radius of less than 30 nm. When such tips are illuminated with short light pulses, optical field enhancement results in intense electron emission by a four-photon photoemission process \([11,12,25]\). Due to the strong field enhancement and high-order optical nonlinearity, the electron emission is localized in space to less than the tip diameter \([11,25]\). A measurement of the electron flux using a microchannel plate detector (MCP) as a function of tip position therefore allows one to resolve the spatial intensity profile of the laser with deep sub-wavelength resolution. When illuminating the tip with a phase-locked pair of laser pulses, a measurement of the electron yield as a function of time delay \(\tau\) provides simultaneous information about the time structure of the laser pulses.

3. Results and discussion

3.1. Time structure of the focused pulses

In order to characterize the effect of the microscope on the time structure of the focused pulses, we first take a conventional interferometric autocorrelation (IAC) measurement of the
pulses in front of the focusing objective. In Fig. 2(a), the IAC trace of the unfocused laser pulses \( I_{\text{IAC}}(\tau) \propto \int_{-\infty}^{\infty} [E(t) + E(t-\tau)]^2 \, dt \) is shown in red. Here, \( E(t) \) denotes the time-dependent electric field of one of the two pulses delayed by \( \tau \). This IAC trace is compared with a chirp-free IAC trace (black dotted line) that is calculated using the laser spectrum extending from 650 to 1050 nm as shown in the inset of Fig. 2(a). At first sight, no major differences are observed and a pulse duration of \( \Delta t = 5.9 \) fs (full width at half-maximum, FWHM, of \( I(t) = |E(t)|^2 \)) is deduced. A closer inspection shows that a small amount of residual higher order chirp leads to a slight change in the fringe spacing at time delays of about 10 fs. This is confirmed by independent SPIDER measurements [26].

- **Fig. 2.** Interferometric autocorrelation (IAC) traces of different pulses: (a) IAC of the incident laser pulse (red line) and a simulated IAC trace (black dotted line) expected for chirp-free, bandwidth-limited pulses with the measured laser spectrum shown in the inset. (b) IAC trace in the focus of a conventional Nikon extra long-working distance 60x microscope objective with a numerical aperture of \( NA = 0.7 \) (solid green line). The pulse is broadened to \( \Delta t = 810 \) fs due to the overall dispersion of the objective. The orange line shows a simulation with \( \varphi_i = 1800 \) fs\(^2\). (c) IAC in the focus of an all-reflective Schwarzschild objective with a \( NA \) of 0.5. The time structure of the pulse remains essentially undistorted.

The IAC trace behind the Nikon glass objective, shown as a solid green curve in Fig. 2(b), is then recorded by positioning the BBO crystal in the focus of the laser beam. As expected, the pulse is severely stretched due to the large chirp of the Nikon glass objective. The IAC trace consists of a short, oscillatory part with a duration of approximately 20 fs at the center of the IAC trace and a low-intensity tail persisting for nearly a picosecond. The measured IAC trace has been simulated by using the laser spectrum from Fig. 2(a) and by introducing a finite group delay dispersion (GDD) of \( \frac{\partial^2 \varphi(\omega)}{\partial \omega^2} = 1800 \) fs\(^2\) for the glass objective. The reconstructed IAC (orange line in Fig. 2(b)) shows almost the same oscillatory structure near zero delay as the experimental IAC trace and almost the same the low-intensity, long persisting background at longer time delays. A temporal duration of the reconstructed pulse in the focus of \( \Delta t = 810 \) fs is deduced. Slight deviations between experimentally recorded and
simulated IAC trace are seen at shorter delays $|\tau| < 200\text{fs}$. The inclusion of higher order chirp in the simulations did not allow us to achieve much better agreement between experiment and simulation. We therefore conclude that the remaining deviations mainly reflect a finite amount of radial chirp introduced by the objective. No attempt has been made to precompensate for the dispersion of the glass objective.

We then replaced the glass objective by an all-reflective Schwarzschild objective. As illustrated in the inset of Fig. 2(c), focusing is achieved by reflecting the incident beam off a small convex inner mirror and collecting the reflected light with a second curved outer mirror. Precise adjustment of the relative positioning of the two mirrors is critical for achieving good spatial and temporal beam profiles. An IAC trace recorded in the focal plane of the Schwarzschild objective is shown in blue in Fig. 2(c). The IAC trace is nearly indistinguishable from that recorded in front of the objective (Fig. 2(a)). Evidently, the time structure of the incident pulse is preserved and the dispersion of the objective, once it is well aligned, is negligible. A detailed inspection reveals a slight increase in pulse duration from 5.9 fs to 6.0 fs. We attribute this slight pulse broadening mainly to a small residual amount of radial chirp introduced by the objective, as discussed in the next section.

3.2. Spatial beam profile of the focused pulses

To characterize the spatial intensity profile $I(\vec{r}) = \int |E(\vec{r}, t)|^2 \, dt$ of these broadband laser pulses in or near the focus of the two objectives, we have scanned an aluminum-coated near field optical fiber with an aperture diameter of 300 nm through the focus and have recorded the intensity $I(\vec{r})$ collected by the fiber probe as a function of its position $\vec{r}$. The movie (Media 1) in Fig. 3 displays cross sections $I(x, y, z_0)$ of the intensity distribution in the focal region of the Schwarzschild objective. These cross sections are taken over a scan range of 5 x 5 μm$^2$, moving the tip in steps of 0.25 μm. The scan range in the z-direction is 9.5 μm with a step size of 0.25 μm along the propagation direction of the laser beam.

Fig. 3. Single-frame excerpt at $z_0 = 0\mu m$ from the movie sequence (Media 1) showing the spatial intensity profile $I(x, y, z_0)$ of 6-fs laser pulses near the focus of a Schwarzschild-objective with a NA of 0.5. $I(x, y, z_0)$ is recorded by scanning an aluminum-coated near field optical fiber with an opening of 300 nm through the focus. The light collected by the fiber is measured by a photomultiplier. The scan-range in z-direction, i.e., along the axis of the objective is 9.5 μm with a step size of 0.25 μm between each cross section. Cross sections $I(x, y, z_0)$, taken with a pixel size of 0.25 μm, are shown in a scan range of 5 x 5 μm$^2$. 

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All images show a pronounced central spot with a dimension of about 1 µm, the diffraction limit of the focused pulses, and clear Airy fringes which increase in intensity with increasing distance $|z_0|$ from the focus. Different features in these movies are interesting. The diameter of the Airy rings decreases continuously while moving the fiber probe away from the objective, from $z_0 = -4.75$ µm to $z_0 = +4.75$ µm. This change in fringe diameter reflects a small amount of uncorrected spherical dispersion of the objective, as discussed below. It is connected with an asymmetric variation of the intensity maximum of the beam profile $I(0,0,z_0)$ while scanning along the z-direction. The movie (Media 1) in Fig. 3 shows that $I(0,0,z_0 > 0)$ decreases more slowly with increasing distance from the focus than in case of positioning the fiber probe in front of the focal plane ($z_0 < 0$). It is also seen that the intensity distribution of the individual Airy rings is not fully uniform and does not show complete circular symmetry. The homogeneity of the fringe pattern depends sensitively on the relative alignment of inner and outer mirror of the objective. Its symmetry can be improved by careful adjustment of the objective. By testing several objectives, we found it difficult to improve the fringe uniformity beyond that shown in Fig. 3.

Fig. 4. (a) Spatial intensity profile $I(x,y,0)$ of a 6-fs-laser pulse in the focal plane of a 36x Schwarzschild objective with a NA of 0.5. $I(x,y,0)$ is probed by scanning an aluminum coated near-field optical fiber probe with an aperture of 300 nm through the focus. (b)-(d) Simulations of $I(x,0,0)$, i.e., a cross section along the red line in (a), based on a wave equation model. (b) Simulations of $I(x,0,0)$ for different NA and a wave vector spectrum extending from $k_{\text{min}} = 0.4 \cdot 2\pi \cdot NA / \lambda$ to $k_{\text{max}} = 2\pi \cdot NA / \lambda$. (c) $I(x,0,0)$ for NA = 0.4 and different $k_{\text{min}}$. (d) Experimental intensity profile $I(x,0,0)$ (red open circles) and optimized fit obtained for NA = 0.4 and $k_{\text{min}} = 0.6 \cdot k_{\text{max}}$ (black solid line). The blue dotted line shows a fit using the nominal NA = 0.7 and a restricted Gaussian $k_x$-spectrum.
The spatial intensity profile \( I(x, y, 0) \) in the focal plane of the all-reflective Schwarzschild-objective is shown in Fig. 4(a). The central spot with a full width at half maximum of 1.0 \( \mu \text{m} \) is surrounded by two clearly visible Airy rings with intensities of about 10 and 5\% of the maximum \( I(0, 0, 0) \). These Airy rings are much more pronounced than in conventional glass objectives since in the reflective objective the central part of the angular spectrum of the focused pulses is blocked by the inner mirror. This is directly seen by comparing the data in Fig. 4 to similar measurements of the intensity profile \( I(x, y, 0) \) in the focal plane of a glass objective with NA of 0.7 (Fig. 5). Also these measurements have been performed with 6-fs-pulses from the Ti:sapphire oscillator with a spectrum shown in Fig. 2(a). Here, the width of the central spot is reduced to 0.76 \( \mu \text{m} \) (FWHM) and the intensity of the first Airy fringe, hardly visible in the two-dimensional cross section, is reduced to less than 0.04.

In order to qualitatively describe these experimental results and to characterize a possible spatio-temporal chirp of the focused few-cycle pulses, we use a simplified three-dimensional vectorial wave equation model. We write the electric field distribution near the focal region as a superposition of plane waves propagating in air with frequency \( \omega = \frac{c}{k} \) (\( c \), speed of light in vacuum) and wave vector \( \vec{k} = \left( k_x, k_y, k_z \right) = \left( \vec{k}_h, k_z \right) \):

\[
\vec{E}(\vec{r}, t) = \sum_{k_x} \sum_{k_y} \sum_{\omega} \vec{E}(k_x, k_y, \omega) \cdot e^{\left(k \cdot \vec{r} - \omega t\right)}.
\] (1)

We take the \( z \)-axis as the optical axis and choose an equidistant grid of wave vector components in \( x \)- and \( y \)-direction. The wave vector component \( k_z = \sqrt{k^2 - k_x^2 - k_y^2} \). In Eq. (1), \( \vec{E}(\vec{k}_h, \omega) \) denotes the complex amplitude of the electric field vector of the corresponding plane wave.

We thus implicitly assume that the employed apertured fiber probes the local electric field of light. In earlier experiments, sensing plasmon fields in metallic nanostructures [27,28], we found good agreement with fully vectorial solutions of Maxwell's equations by assuming that the in-plane components of the electric field vector (\( E_x, E_y \)) are predominantly detected in this type of collection-mode near-field microscopy. These conclusions have been confirmed in subsequent detailed investigations [29,30]. Yet, it is known that metalized fiber probes with specific geometries may also be sensitive to the magnetic field of light [31–33]. This sensitivity is most pronounced for small-aperture probes [31] and probes with reduced symmetries [32]. For symmetric probes with rather large apertures, as used in this work, the coupling to magnetic fields is reduced and we hence restrict the discussion to the electric field vector. Based on the work in [27,30] we assume that we collect only in-plane components of the electric field vector. Since we used linearly (\( x \)-)polarized incident light, this allows us to use a scalar spectral amplitude

\[
\vec{E}(\vec{k}_h) \approx \tilde{A}(\vec{k}_h, \omega) e^{\varphi(\vec{k}_h, \omega)} \hat{e}_x
\] (2)

to model the experimental data. Here \( \hat{e}_x \) is the unit vector along \( x \), \( A(\vec{k}_h, \omega) \) a real amplitude and \( \varphi(\vec{k}_h, \omega) \) the spectral phase of the corresponding plane wave. The finite spatial resolution of the aperture is then taking into account by convoluting \( \vec{E}(\vec{r}, t) \) with a Gaussian point spread function.
We now use Eqs. (1) and (2) to analyze the spatial images recorded in the focal plane of the two objectives (Figs. 4 and 5). To a first approximation, we neglect variations in the spectral phase \( \phi(\vec{k}_\parallel, \omega) \) by putting all phase terms to zero. Evidently, an important geometric difference between the two objectives is the blocking of the central part of the focused beam by the inner mirror of the Schwarzschild objective. We incorporate this in the model by restricting the \( k \)-vector spectrum \( \tilde{A}(\vec{k}_\parallel, \omega) \) to the range \( k_{\text{min}} \leq |\vec{k}_\parallel| \leq k_{\text{max}} \). While \( k_{\text{max}} = NA \cdot \omega/c \) is given by the numerical aperture of the objective, \( k_{\text{min}} \) is taken as a free parameter. For simplicity, we assume a homogeneous illumination of the objective \( \tilde{A}(\vec{k}_\parallel, \omega) = \tilde{A}_0(\omega) \) in this spatial frequency range with \( \tilde{A}_0(\omega) \) as the measured laser spectrum.

Then, the choice of \( k_{\text{max}} \) defines the width of the central peak of \( I(x,y,0) \) while \( k_{\text{min}} \) mainly affects the amplitude of the Airy fringes. Good agreement with the experimental data in Fig. 4 is found for \( NA = 0.4 \) and \( k_{\text{min}} = 0.6 \cdot k_{\text{max}} \). The same model also reproduces the intensity distribution in the focus of the glass objective when putting \( NA = 0.6 \) and \( k_{\text{min}} = 0 \). Evidently, the low spatial frequency components greatly reduce the intensity of the Airy fringes. The deduced values for the NA (0.4 and 0.6) are slightly smaller than those given by the manufacturers (0.5 and 0.7). We anticipate that slightly better match can be achieved by further increasing the diameter of the entrance beam and/or by choosing a more realistic representation of the amplitude spectrum \( \tilde{A}(\vec{k}_\parallel, \omega) \). Indeed, reasonable agreement between experiment and simulations based on the nominal NA of the objectives (0.5 and 0.7) when using a Gaussian-shaped \( \vec{k}_\parallel \) distribution with a finite width of the k-spectrum (blue dotted lines in Figs. 4(d) and 6(b)). Different physical effects contribute to this apparent reduction in width of the k-spectrum. First, the finite geometric size of the incident laser beam reduces the intensity of beams with large in-plane \( k \)-vectors. Also, the polarization dependent collection-efficiency of the NSOM probe [34] contributes. A full quantitative modeling of the intensity profiles in the focal plane should take both effects into account.

We also noticed that the beam obscuration by the inner mirror of the Schwarzschild objective of about \( (k_{\text{min}}/k_{\text{max}})^2 = 0.36 \) is larger than the specified value. This may slightly be improved by optimizing the illumination of the entrance pupil. The simulations show that the finite optical resolution of the fiber probe of 300 nm has little effect on the measured intensity profiles, apart from a very slight increase in width of the central peak and a slight reduction of the modulation of the Airy fringes.

While these chirp-free simulations can reproduce the focal plane images rather well, they cannot account for the temporal pulse broadening in the glass objective or the position-dependent diameter of the Airy rings in the Schwarzschild objective. To explain these observations the effect of the objective on the spectral phase \( \phi(\vec{k}_\parallel, \omega) \) must be considered. We express the in-plane wave vector in spherical coordinates \( \vec{k}_\parallel = (k \sin(\theta) \cos(\phi), k \sin(\theta) \sin(\phi)) \). Neglecting a possible dependence of \( \phi(\vec{k}_\parallel, \omega) \) on the azimuthal angle \( \phi(\vec{k}_\parallel, \omega) = \phi(\theta, \omega) \), a Taylor-series expansion of the spectral phase gives...
\[
\varphi(\theta, \omega) = \varphi(0, \omega_0) + \frac{\partial \varphi}{\partial \omega}\bigg|_{\omega_0} (\omega - \omega_0) + \frac{1}{2} \frac{\partial^2 \varphi}{\partial \omega^2}\bigg|_{\omega_0} (\omega - \omega_0)^2 + \\
\frac{\partial \varphi}{\partial \theta}\bigg|_{\omega_0} \theta + \frac{1}{2} \frac{\partial^2 \varphi}{\partial \theta^2}\bigg|_{\omega_0} \theta^2 + \frac{1}{2} \frac{\partial^2 \varphi}{\partial \theta \partial \omega}\bigg|_{\omega_0} \theta(\omega - \omega_0) + \ldots,
\]

(3)

where \(\omega_0\) denotes the center frequency of the laser pulse. The physical significance of the different terms in this expansion is readily transparent. \(\varphi(0, \omega_0)\) denotes the absolute phase of the pulses, important for higher-order nonlinear phenomena. The group delay \(\partial \varphi/\partial \omega\) results in a mere time shift and the second order chirp or group delay dispersion (GDD) \(\partial^2 \varphi/\partial \omega^2\) stretches the pulse in the time domain without significantly affecting the spatial focusing. This can readily be seen in the experiments performed with the conventional glass objective. Even though the pulse is stretched to more than 800 fs (Fig. 2b), corresponding to a GDD of 1800 fs\(^2\), the beam diameter (Fig. 5) is close to its diffraction limit. The measured GDD parameter agrees well with earlier results [19].

In similarity to the effect of the group delay, the angular dispersion \(\partial \varphi/\partial \theta\) mainly leads to a spatial displacement of the beam in the focal plane while leaving the beam diameter and its time structure almost unaffected. The dominant effect of the second order angular dispersion \(\partial^2 \varphi/\partial \theta^2\) is to enlarge the beam diameter in the focal plane. As its counterpart GDD, it also has a small, not very pronounced but non-negligible effect of the time structure of the focused pulses. It effectively describes a delay of those parts of the pulse propagating at larger inclination angles \(\theta\) with respect of those propagating close to the optical axis. As such it results in a slight delay (advancement) of the central part of the beam focus with respect to the outer region of the spot. When blocking the rays with small inclination angles \(\theta\), as in the case of the focusing with the Schwarzschild objective, this gives rise to a change in the diameter of the Airy rings along the optical axis, as observed in the movie shown in Fig. 3. From these experiments, we deduce a second order angular dispersion of \((20^\circ)^2\). This dispersion can be controlled by varying the distance between inner and outer mirror.

Fig. 5. (a) Spatial intensity profile \(I(x, y, 0)\) of a 6-fs-laser pulse in the focal plane of a 60x extra long working distance microscope objective with a NA of 0.7. (b) Experimental intensity profile \(I(x, 0, 0)\) (red open circles, taken across the red line in (a)) and optimized fit obtained for NA = 0.6. (black solid line). The blue dotted line shows a fit using the nominal NA = 0.7 and a restricted Gaussian \(k_i\)-spectrum.
The most critical term for focusing of short laser pulses is \( \frac{\partial^2 \varphi(\theta, \omega)}{\partial \theta \partial \omega} \), which may be understood as a first order radial chirp. It results in a stretching of the pulse in both space and time. Our experiments with the Schwarzschild objective (Figs. 2, 3, and 4) indicate that the radial chirp induced by the objective has negligible effect on the experimental data, once the objective is well aligned. Somewhat surprisingly also the data taken with the glass objective show little evidence for radial chirp despite using very broadband excitation pulses. This suggests that it should be possible to remove much of the temporal chirp by appropriate precompression schemes. Such experiments are currently underway.

3.3 Spatio-temporal beam profile in the focal plane

So far, we have discussed separate measurements of the temporal and spatial profile of the focused pulses. Ideal measurements should allow for a complete reconstruction of \( \hat{E}(\vec{r}, t) \). To our knowledge, such a fully vectorial reconstruction of a few-cycle light spot has not yet been achieved. Recently, SEA-TADPOLE has been introduced as technique for directly measuring the electric field of focused ultrashort pulses [35]. When being combined with high-spatial resolution detection schemes such as, e.g., a NSOM fiber probe, electric field measurements can be performed with subwavelength resolution [36]. So far, the time resolution in these measurements has been limited to a few tens of fs. We therefore adopt a slightly different technique, replacing the NSOM fiber with sharp, chemically etched gold taper with a tip diameter of less than 30 nm (Fig. 6(a)). When illuminating such tips with intense laser pulses polarized along the tip axis, field enhancement results in the emission of electron pulses from the end of the tip [11,12,25,37]. For moderately laser peak intensities below \( 10^{11} \) W/cm\(^2\), the electron emission is due to multiphoton absorption while for higher intensities optical field emission sets in [35]. Due to the local field, the enhancement is localized in space to much less than the tip diameter [11,25], probing the local field intensity with deep-subwavelength resolution.

Experimentally, it was shown, that for sufficiently low intensity, electrons are emitted from the tip by multiphoton excitation of electrons above the vacuum level. In the absence of a bias voltage and for excitation wavelengths around 800 nm a 4-photon excitation process is dominant with an electron yield \( I_d(\vec{r}) \propto \int \hat{E}(\vec{r}, t)^{2n} \, dt \) with \( n \approx 4 \) denoting the order of the optical nonlinearity [12,25]. With increasing bias voltage \( n \) decreases towards unity [12,37]. Hence, the electron yield probes in a highly nonlinear fashion the local electric field at the very apex of the tip. This strong nonlinearity makes it interesting to use such a tip as a new kind of autocorrelator with high temporal and spatial resolution [12].

The ultimate time resolution of this autocorrelator depends on the lifetime (\( T_1 \)) time of the surface plasmon resonance of the tip. For larger tips, \( T_1 \) times of the order of 10 fs have been measured [38]. For very sharp tips considerably shorter \( T_1 \) times are expected. Here, we experimentally explore the use of the electron emission from such tips for probing the time structure of focused few cycle light pulses.

Experimentally, a small DC bias voltage of 200 V has been applied between tip and MCP to accelerate the electrons towards the detector. It has been observed before [12,25] in experiments performed under similar conditions that this reduces the order of the optical nonlinearity from 4 (at zero bias) to slightly less than 4. A spatial map of the electron count in the focal plane of the Schwarzschild objective for illumination with the 6-fs pulses characterized before is shown in Fig. 6(b). Compared to Fig. 3, the spot size of the image is reduced to 500 nm (FWHM) and the Airy rings are practically absent. This is shown in Fig. 7 in more detail. Here, the cross section of the focus scan of the linear (Fig. 7b)) and the
Fig. 6. Simultaneous measurement of the spatial and temporal profile of a 6-fs laser pulse in the focus of an all-reflective Schwarzschild objective. (a) The laser is focused to the apex of a sharp gold tip. Optical field enhancement results in multi-photon induced emission of electrons from the tip apex. The electrons are accelerated towards a microchannel plate (MCP)-detector by applying a small bias voltage. Their count-rate is detected as a function of the relative position \( r \) of tip and focus. A Photodiode (PD) is used to facilitate tip positioning. (b) Electron count-rate \( I(x, y, 0) \) in the focal plane of the objective. The incident laser is polarized parallel to the tip axis. (c) IAC trace recorded by measuring the electron emission rate \( I(\tau = 0, \Delta \tau) \) as a function of time delay \( \Delta \tau \) between a pair of 6-fs pulses focused onto the tip apex.

Nonlinear measurement (Fig. 7a)) are compared and one sees clearly that the width of the electron signal is reduced by a factor of \( \sqrt{n} \approx 2 \). This reduction directly results from the strong optical nonlinearity of the electron generation process.

For locally probing the time structure, we use a phase-locked pair of incident pulses and record the electron signal as a function of pulse delay \( \tau \) (Fig. 6(c)). In comparison with Fig. 2(c), the width of the interferometric autocorrelation trace \( I_{el, IAC}(\tau) \) is further reduced and the modulation of the individual fringes clearly enhanced. Also the enhancement factor \( \eta = I_{el, IAC}(\tau = 0) / \lim_{\tau \rightarrow \infty} I_{el, IAC}(\tau) \), i.e., the ratio of the electron count at zero delay and that at long delays increases to a value of 78. The strong optical nonlinearity of the imaging process accounts for most of these effects. As the IAC enhancement is given as \( \eta = 2^{2n-1} \), the measurements suggest an order of the nonlinearity of \( n_{el} = 3.7 \), which is close to what has been observed earlier for similar bias voltages [12, 25]. Also, the temporal modulation of the fringes is consistent with such a value. A detailed simulation of \( I_{IAC}(\tau) \) based on the known laser spectrum and \( n_{el} = 3.7 \) is shown as a dashed line in Fig. 6(c). Reasonable agreement between measurement between experiment and simulation is only reached by assuming that the pulse duration of the incident pulse has increased to \( \Delta \tau = 7.5 \) fs. Such an increase in time...
resolution is not seen in conventional IAC measurements recorded by using a thin BBO crystal (Fig. 2(c)). We therefore expect that this increase in time resolution reflects the finite lifetime of the surface plasmon polariton resonance of the gold tip and hence the frequency dependency of its local field enhancement factor at the tip apex. Our results thus indicate that such photoelectron emission autocorrelation traces are sensitive probes of the real-time dynamics of surface plasmon polariton excitations in metallic nanostructures.

Fig. 7. Comparison of the nonlinear (shown left) and the linear measurement (right) of the focus area using the Schwarzschild objective. While for the linear measurement, the diameter has been determined to about 1.0 μm, the nonlinear experiment with the electron yield as the intensity signal results to a focus diameter of 0.5 μm. The reduction in beam diameter by a factor of two can be well explained by the nonlinear imaging process of the electron generation.

4. Summary and conclusion

In summary, we have studied the spatio-temporal focusing of few-cycle laser pulses by conventional glass and all-reflective metal objectives. By using different interferometric autocorrelation techniques, including a novel tip-enhanced electron emission autocorrelator, we have demonstrated an essentially dispersion-free and diffraction-limited focusing of few-cycle laser pulses by a well-aligned, Schwarzschild microscope objective. By focusing 5.9-fs laser pulses through an all-reflective microscope objective with a numerical aperture of 0.5, we reached a focus diameter of 1.0 μm, while preserving the time structure. This corresponds to a focal volume of these light bullets of only 1.8 μm³. In the focal plane of such an objective pulses from a conventional high-repetition laser oscillator with an average power of 100 mW reach peak intensities of the order of 10^{13} W/cm². We therefore assume that the presented technique is of considerable interest for inducing and probing optical nonlinearities of individual nanostructure with few-cycle time resolution.