ABSTRACT
Improving the resolution and sensitivity in all-optical microscopy and spectroscopy is inevitably one of the most important challenges in contemporary optical and nanoscience. Here, we discuss a novel approach, plasmonic nanofocusing, towards broadband, coherent all-optical microscopy with ultrahigh temporal and spatial resolution. The conceptual idea is to launch radially symmetric surface plasmon polariton modes onto the shaft of a sharp, conical metal taper. While propagating towards the apex of the pointed taper, the spatial extent of the plasmonic mode gradually shrinks, from several microns in diameter to a spot size of less than 10 nm at the pointed apex of the conical taper. Concomitantly, the local field amplitude of the plasmon mode gradually increases, resulting in a pronounced field enhancement at the apex and – thus – a bright and spatially isolated coherent light source with dimensions far below the diffraction limit. In this review, we characterize the optical properties of such three-dimensional conical metal tapers and demonstrate nanofocusing of radially symmetric plasmon modes. We use this nanolight source for coherent light scattering spectroscopy and demonstrate the sensitivity enhancement resulting from the pronounced spatial field confinement. It is shown that such off-resonant plasmonic nanoantennas facilitate the creation of nanofocused light spots with few-cycle time resolution. As a first application of

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this ability to nanolocalize ultrashort plasmon wavepackets, we demonstrate remotely-triggered multiphoton-induced photoemission from the very apex of the taper and implement this novel ultrafast electron gun in a point-projection electron microscope. Our results not only indicate the favourable optical properties of this plasmonic nanolens but also suggest that it may find interesting applications in ultrafast scanning optical spectroscopy and might enable new types of ultrafast electron holography and scanning tunnelling microscopy.

1. Introduction

Optical and electron microscopy have spurred new discoveries over the last centuries, and, driven by urgent questions about the structure and function of (nano) materials, microscopy techniques have been continuously improved to provide the best spatial and temporal resolutions possible. The underlying general idea of optical microscopy, to extract information about a microscopic volume of matter by its interaction with light, means that basically all known spectroscopic methods can be applied and combined with three-dimensional spatially-resolved imaging. Furthermore, adding temporal information by employing ultrafast pump-probe techniques opens up the fourth dimension and thereby enables, in principle, the observation of ultrafast dynamics during technologically relevant processes, such as, e.g. femtosecond chemical reaction dynamics [1,2], optical manipulation of magnetic order [3,4], non-equilibrium dynamics in superconductors [5], optically-induced phase transitions [6], or conversion of sunlight into electrical [7–9] or chemical [10–12] energy. As a specific example from our own work, ultrafast pump-probe spectroscopy has recently revealed the importance of coherent vibronic coupling between electronic and nuclear motion after electronic excitation for charge separation and transfer in a model organic photovoltaic material [13]. Most of those ultrafast experiments are performed with moderate spatial resolution on a large ensemble of emitters. These measurements therefore almost inevitably average over the dynamics of an inhomogeneously broadened ensemble of emitters with wavefunctions that are typically localized on a nanometre length scale. So far, only a fairly limited number of experiments exist that probe the femtosecond dynamics of single quantum emitters, e.g. a single exciton in a semiconductor quantum dot or a molecular light harvesting complex [14–19]. Such studies are challenging and experimental tools to perform them are still at an early stage of their development. Often, they are performed on diluted samples in order to artificially increase the distance between adjacent emitters and to facilitate their spatial isolation. As such, the local environment of the emitter is necessarily different from the native environment and this may significantly affect the ultrafast dynamics of its optical excitations [18–20].

In order to improve our understanding of fundamental light-induced processes at the nanoscale, it would therefore be highly desirable to probe the ultrafast dynamics of single quantum emitters not only in diluted, artificial structures, but also in densely packed nanosystems such as magnetic nanostructures, natural light
harvesting complexes, polymers, organic photovoltaic devices, or metal-molecular hybrid systems. This requires the development of optical nanospectroscopy, combining an ultrahigh temporal resolution with the possibility to localize the illumination to the volume of a single quantum emitter, typically roughly $(5 \text{ nm})^3$ or to the order of $100$ yoctoliters $(10^{-22} \text{ l})$.

Conventional optical far-field microscopy, while offering a plethora of interesting spectroscopic tools, suffers from one serious constraint: its resolution is limited by diffraction to typically half the wavelength of the illuminating light. Recent important advances in far-field microscopy, e.g. stimulated emission depletion microscopy [21,22], photoactivated localization microscopy [23], or stochastic optical reconstruction microscopy [24,25], have allowed for breaking the diffraction limit and for pushing the resolution of far-field microscopy into the 10 nm range. Yet, all of these methods probe the fluorescence of specialized fluorophores that are attached to the structure of interest. This generally makes the combination with femtosecond pump-probe techniques problematic, as the aspired temporal resolution is much below the fluorescence lifetime of the emitter. A possible exception is ground state depletion microscopy [26], where the molecules in the centre node remain in the ground state and should thus be open to time-resolved spectroscopy. Also, even though these far-field super-resolution methods reduce the detection volume effectively down to diameters of several tens of nm, or roughly $10^{-20}$ l, the illuminated volume still remains at its diffraction-limited size.

True nanometre-scale light localization can only be achieved by making use of optical near fields that exist in the vicinity of every type of atomic, molecular or solid-state nanoemitter. Since the spatial extent of these evanescent near fields is dictated by the geometry of the nanoemitter rather than the light wavelength, the localization volume can simply be reduced by decreasing the size of the nanoemitter. As such, even atomic scale all-optical resolution may in principle be reached and this has sparked a tremendous interest in near-field optical microscopy since its first demonstration by Pohl and co-workers [27–29]. Even though, nowadays, near-field microscopes offering a spatial resolution of a few nanometres [30–42] or even below [43] are in operation in a range of laboratories world-wide, such microscopes are still not in widespread use and few instruments are commercially available. This is partly due to the inherent surface sensitivity of the technique, making true three-dimensional optical imaging difficult. Most notably, however, it has proven rather problematic in the past to reproducibly fabricate near-field lenses that allow for funnelling far-field light into a nanometric volume. Initial attempts focused on transmitting light through a nanometre-sized aperture in a metal-coated fibre taper [44]. This approach suffers, however, from the inherently small transmission coefficient of the aperture which decreases sharply with decreasing diameter, the difficulty in manufacturing apertures with diameters of less than 50 nm, and the complex polarization properties of the generated optical near fields. A modern version of this approach is the Campanile tip design [45,46],
which greatly enhances the light throughput by engineering an aperture without cut-off. Yet, so far, the localization diameter is still limited to about 30 nm.

Much improved spatial resolution can be achieved by scattering far-field light off the apex of a sharp metallic or dielectric taper [47,48]. The resulting optical near fields are localized to the apex diameter and hence can be reduced in size to dimensions of 5–10 nm. The interaction of these near fields with the investigated sample is probed by detecting the light that is scattered from the tip-sample region into the far field. Since the taper apex is directly illuminated by far-field light localized to a diffraction limited spot, the recorded scattered light not only contains the desired near-field signal but also a background of scattered far-field light, whose intensity may often exceed the near-field contribution by far [49]. Commonly, a combination of a periodic modulation of the tip-sample distance and an interferometric heterodyne detection scheme is used to filter out the desired near-field signal [50–54]. With this, spatially resolved imaging of optical near fields with spatial resolution in the 10 nm range is now routinely achieved. Also, inelastic light scattering techniques, specifically tip-enhanced Raman spectroscopy and fluorescence spectroscopy, are often used to suppress the scattering background and obtain information about the local optical properties of the investigated sample. Still, elastic near-field scattering spectroscopy and specifically coherent ultrafast optical spectroscopy with nanometre-spatial resolution is largely hampered by this undesired far-field background in conventional apertureless near-field microscopy. Consequently, a large number of near-field probe designs have been introduced that combine the discussed aperture-based and aperture-less approaches [55–58]. Such tip-on-aperture probes are rather difficult to fabricate and so far have not yet found widespread use.

Strong nanometre-scale light localization can also be achieved by using metallic antennas that are designed to efficiently transform propagating, far-field light into a non-propagating, strongly localized plasmonic near-field spot [59–66]. For the localization of ultrafast optical pulses, it is important that this antenna structure supports a broad spectral bandwidth or, ideally, is non-resonant. The latter condition is generally fulfilled for sharply tapered, conical waveguides, where optical excitations are transported coherently along the taper in the form of surface plasmon polariton (SPP) waves that are adiabatically transformed into a localized surface plasmon (LSP) of only a few-nm extent at the taper apex [67–69]. Among the different experimentally realized geometries [63,70–73], sharp conical metallic tapers are of particular relevance, because they are easily incorporated into scattering-type scanning near-field optical microscopes (s-SNOM) [42,74,75], where they act as a single, point-dipole-like emitter with nanometre dimensions and well-defined polarization properties. When coupling ultrafast laser pulses to propagating SPPs at the shaft of those tapers, nanometre-localized light spots with pulse durations as short as 10 fs are created at the taper apex with high efficiency [42,76]. Consequently, the optical properties of such antennas have been
studied in considerable detail, both experimentally and theoretically, during the past several years [42,63,64,70,75–91].

Here we briefly review our own recent work on plasmonic nanofocusing using sharply etched metallic nanotapers. After an introduction to the principle of nanofocusing we review tip production, plasmon coupling, plasmon propagation and the properties of the localized plasmon field, specifically its spatial and temporal extent. We show how propagating SPP wavepackets, launched by grating-coupling far-field laser light to the taper surface, are transformed into an LSP at the very apex of the taper. Through nanofocusing, a plasmon field at the apex with a temporal duration of 10 fs and an axial and lateral decay length of <10 nm is achieved, which is the basis for high-resolution, virtually background-free s-SNOM imaging and spectroscopy, as illustrated by different examples. Furthermore, we show that plasmonic nanofocusing of ultrafast optical pulses results in local field intensities that are sufficiently large to introduce pronounced optical nonlinearities at the very apex of the taper. We make use of this field enhancement to induce the emission of photoelectrons from a nanometre-sized region at the very apex of the taper while illuminating the taper at a distance of 50 μm from the apex. Such a remotely triggered photoelectron source has favourable properties for ultrafast electron microscopy, as illustrated in an implementation of point-projection microscopy. Moreover, it may open up a new path towards the realization of ultrafast scanning tunnelling microscopy and form the basis for novel types of ultrafast microscopy, combining both electron and optical imaging with (sub-) nanometre spatial resolution.

2. Plasmonic properties of sharply etched gold nanotapers

2.1. Motivation for plasmonic nanofocusing

Linear response theory shows that the linear optical polarizability of a single, point-like quantum emitter embedded in an isotropic material is given as

\[ \alpha(\omega) = -\frac{|\mathbf{p}|^2}{3\varepsilon_0 \hbar} \left( \frac{1}{\omega - \omega_0 + i\gamma} - \frac{1}{\omega + \omega_0 + i\gamma} \right) \]  

(1)

Here, it is assumed that the optical response can be approximated as that of an electronic two-level system with energy difference \( \hbar \omega_0 \) between its (fully occupied) ground and (empty) excited state. The transition dipole of the two-level system is \( \mathbf{p} \) and \( \gamma = 1/T_2 \) is the dephasing rate of the system. In this Markovian limit, an impulsive optical excitation of the system results in an exponential decay of its electronic polarization with the dephasing time \( T_2 \).

The absorption cross section of the emitter is related to its polarizability as

\[ \sigma(\omega) = \text{Im}(\alpha(\omega)) \cdot \omega/c \]  

(\( c \): speed of light in vacuum) and its radiative damping rate is \( \Gamma_{\text{rad}} = \frac{|\mathbf{p}|^2 \omega}{3\pi\varepsilon_0 \hbar c} \). The on-resonance cross section of a purely radiatively damped (\( \gamma = \Gamma_{\text{rad}}/2 \)) quantum emitter in vacuum is thus given as [92]
with $\lambda_0 = 2\pi c/\omega_0$. Hence, the effective dimension of the emitter, when being probed with a resonant light beam is given by the wavelength $\lambda_0$ of the incident wave, rather than the geometric size of the emitter. Pure radiative damping is typical for atoms in vacuum and the resulting large cross section makes it comparatively easy to maximize light-matter coupling in these systems by matching the optical beam diameter $A$ to $\sigma_{\text{max}}$. The ultimate regime of strong coupling between an atom and a single optical mode of the light field can be reached by placing the atom inside a cavity. The resulting Vacuum Rabi Splitting [93] is one of the hallmarks of cavity quantum electrodynamics [94] and lies at the heart of modern developments in quantum information science.

The situation is fundamentally different for quantum emitters in a solid-state environment at room temperature. Here, pronounced fluctuations of the environment of the emitter result in dephasing rates $\gamma$ which are typically orders of magnitude larger than the radiative damping rate $\Gamma_{\text{rad}}$. Hence, the on-resonance cross section is drastically reduced. Typical values for the dephasing rate are on the order of $(10 \text{ fs})^{-1}$ to $(100 \text{ fs})^{-1}$ while radiative damping rates often range from $(100 \text{ ps})^{-1}$ to $(10 \text{ ns})^{-1}$. Hence, the cross section is reduced to, e.g. $\sim 0.01$ to $0.1 \text{ nm}^2$ for a single dye molecule and a few $\text{nm}^2$ for a typical colloidal quantum dot [95] or J-aggregated supramolecular emitter [96]. This rapid dephasing and the concomitant decrease in cross section have two immediate consequences. First, it is difficult to probe the dynamics of coherent optical excitations of such emitters directly in the time domain as this requires ultrashort optical pulses with durations of less than $T_2$. Second, the probability, $\sigma/A$, to absorb a photon from a monochromatic optical beam with diameter $A$ is usually quite small, making direct (transient) absorption studies of single solid-state quantum emitters at room temperature very challenging. Also, this small cross section has so far prohibited reaching the interesting regime of strong coupling between a single solid-state quantum emitter and a single cavity mode at room temperature. One strategy to overcome these challenges is to spatially confine the optimal beam diameter $A$, preferably to the size of $\sigma_{\text{max}}$ or even below. We will show below that conical metallic tapers constitute a special type of plasmonic nanolens with particularly interesting physical properties that may in fact be very helpful in addressing those challenges.

$$\sigma_{\text{max}} = \frac{3\lambda_0^2}{2\pi}$$

(2)

$$\sigma_{\text{max}} = \frac{3\pi c^2 \Gamma_{\text{rad}}}{\omega_0^2 \gamma}$$

(3)
2.2. Plasmonic eigenmodes of circular wires and tapers

For rationalizing the optical properties of conical metal tapers, it is instructive to first consider the simpler case of a homogeneous circular metal cylinder of radius \( R \) and of infinite length \([62,97–99]\). The dielectric function of the metal is taken as \( \varepsilon_1(\omega) \) and that of the surrounding homogeneous dielectric medium as \( \varepsilon_2(\omega) \). Due to the rotational symmetry of the wire, the electric and magnetic fields inside and outside of the wire can be derived, for each non-radiative plasmon eigenmode, from scalar functions \( \psi_{j,m} \). In cylindrical coordinates these functions can be expressed as \([100]\)

\[
\psi_{j,m}(r, \theta, z) = B_{j,m}(k_j r) \exp \left( i m \theta + i k_z z - i \omega t \right)
\]

(4)

Here, \( m \) is an integer denoting the angular momentum quantum number of the corresponding mode and \( k_z \) is the wave vector component of the mode along the wire axis. \( B_{j,m}(\bullet) \) is an appropriate Bessel function, which is equal to the Bessel functions of the first kind, \( J_m(\bullet) \), and to the Hankel functions of the first kind, \( H_m^{(1)}(\bullet) \), for the regions inside and outside the wire, respectively. The radial component of the wave vector in medium \( j = \{1, 2\} \) is

\[
k_j^2 = \varepsilon_j(\omega/c)^2 - k_z^2,
\]

(5)

as we set the Azimuthal wave vector component to zero. For each of these eigenmodes, the wave vector component \( k_z \) at a given frequency \( \omega \) is obtained as the solution of an eigenvalue equation which may be written as \([97]\)

\[
k_0^2 \left( \frac{J_m'(u)}{u J_m(u)} - \frac{H_m^{(1)'}(v)}{v H_m^{(1)}(v)} \right) \left( \varepsilon_1 J_m'(u) - \varepsilon_2 H_m^{(1)'}(v) \right) - m^2 k_z^2 \left( \frac{1}{v^2} - \frac{1}{u^2} \right)^2 = 0
\]

(6)

Here, \( k = \omega/c, u = k_1 R \), and \( v = k_2 R \). The prime above a cylinder function denotes differentiation with respect to the function argument.

Most interesting are the properties of the lowest order, \( m = 0 \), radially polarized monopole mode (Figure 1(a)). It has electric field components in radial direction, pointing perpendicular to the metal surface, and parallel to the taper axis (taken as the \( z \)-direction) \([100]\). Within the limits of a local, classical electromagnetic theory, it exists as a bound, non-radiative SPP mode even for vanishingly small wire radii. Its wave vector in propagation direction \( k_z \) exceeds the free space wave vector \( k_0 \) and the effective refractive index \( k_z/k_0 \) seen by the propagating modes diverges with decreasing radius as \( k_z/k_0 \propto 1/R \) (Figure 1(b)). As a consequence, the decay length of the evanescent field in the embedding dielectric medium decreases substantially for small wire radii and the local field intensity is enhanced near the wire surface. This is very different for the dipolar, \( m = \pm 1 \) modes. Here, the effective refractive index is slightly larger than unity for radii larger than a certain cut-off radius. For smaller radii, no non-radiative solutions of Equation (6)
with this angular momentum exist. Hence the $m = \pm 1$ modes do not show the field confinement, which is characteristic for the $m = 0$ mode and exist as bound, non-radiative modes only for wire radii exceeding a certain cut-off radius. Beyond this critical radius these evanescent modes are transformed into propagating modes. The respective cut-off radii are readily deduced from the eigenvalue equation, Equation (6). For a vacuum wavelength of 800 nm and a gold wire with dielectric function $\varepsilon_1 = -24.75 + 1.88i$ (corresponding to a vacuum wavelength of 800 nm) and the wire is embedded in air ($\varepsilon_2 = 1$). The refractive index steeply increases for small radii, resulting in a strong reduction in SPP group velocity and thus an increase in field intensity. Simultaneously, the fraction of the SPP mode penetrating the metal and therefore also the propagation losses ($\text{Im}(k_z)$) increase sharply. Effective refractive indices for the dipolar $m = \pm 1$ and the quadrupolar $m = \pm 2$ modes. $\text{Re}(k_z^{(m)})$ approaches $k_0$ for a certain finite cut-off radius ($\sim 70$ nm for $m = \pm 1$ and $\sim 600$ nm for $m = \pm 2$). For smaller wire radii, these modes are no longer non-radiative bound modes of the wire. (c) Eikonal parameter $\delta$ calculated for the $m = 0$ mode. The eikonal parameter remains below unity and approaches $\delta \approx 0.8$ for $R \to 0$.}

Figure 1. (a) Schematic illustration of the lowest order non-radiative surface plasmon polariton eigenmodes of a conical gold taper (from [84]). The monopolar $m = 0$ mode is a bound mode of the taper even for small taper radii. Its field is strongly confined to the taper apex. All higher order modes are only bound if the local taper radius exceeds a certain cut-off radius. Beyond this critical radius these evanescent modes are transformed into propagating modes. (b) Real part of the effective refractive index $\text{Re}(k_z^{(m)}/k_0)$ for the lowest order monopole mode ($m = 0$), propagating along a cylindrical gold wire of radius $R$. The dielectric function of gold is $\varepsilon_1 = -24.75 + 1.88i$ (corresponding to a vacuum wavelength of 800 nm) and the wire is embedded in air ($\varepsilon_2 = 1$). SPP propagation along conical tapers has first been studied theoretically by Babadjanyan et al. [69]. They analysed the solutions of Equation (6) for the lowest order monopole mode ($m = 0$), propagating along a cylindrical gold wire of radius $R$. The dielectric function of gold is $\varepsilon_1 = -24.75 + 1.88i$ (corresponding to a vacuum wavelength of 800 nm) and the wire is embedded in air ($\varepsilon_2 = 1$). SPP propagation along conical tapers has first been studied theoretically by Babadjanyan et al. [69]. They analysed the solutions of Equation (6) for the lowest order monopole mode ($m = 0$), propagating along a cylindrical gold wire of radius $R$. The dielectric function of gold is $\varepsilon_1 = -24.75 + 1.88i$ (corresponding to a vacuum wavelength of 800 nm) and the wire is embedded in air ($\varepsilon_2 = 1$).
idealized conical taper with infinitely sharp, point-like apex and showed that for sufficiently small cone opening angles, the energy that is stored in the propagating SPP field is essentially completely localized at the apex, at least if Ohmic losses during propagation are weak. Such adiabatic plasmon nanofocusing with negligible internal reflections and radiative damping of the propagating SPP requires that the eikonal parameter or adiabaticity parameter, defined as

\[ \delta = \left| R' d(k_0 n) / dR \right| \]  

is sufficiently small, i.e. \( \delta < < 1 \). Here, \( R' = dR/dz \) and \( n = \text{Re}(k_z/k_0) \) is the local effective refractive index, which increases sharply with decreasing distance from the taper apex. For small opening angles, the adiabaticity condition is fulfilled even at the tip extremity and this results in a pronounced enhancement of the local electric field energy density \( W(r) = \{ d(\omega e(r, \omega))/d\omega \} |E(r)|^2 \) at the apex by about three orders of magnitude with respect to that in the region with \( n \approx 1 \). For this idealized structure, the eikonal approximation predicts that a SPP wave-packet, i.e. a coherent superposition of \( m = 0 \) modes with different colours, is adiabatically slowed down and eventually stopped as it propagates towards the apex. A second important consequence of the local increase in refractive index is a pronounced decrease in the decay length of the evanescent SPP field in the surrounding dielectric. Hence, the SPP field is greatly enhanced in the focus of the plasmonic nanolens, i.e. at the taper apex. In real tapers, even if they could be fabricated with arbitrary precision, the predicted divergence of the local refractive index is of course suppressed due to the wave nature of the electronic excitations at the apex and the resulting non-locality of the dielectric function [101]. As such, also a finite amount of radiative damping of the localized SP field is expected, even though this may indeed be negligibly small for very sharp tapers [102].

As a measure for the radiative damping we expect in our gold nanotapers, we have determined the eikonal parameter as defined in Equation (7) from the mode analysis. The tips used in our experiments typically have an opening angle of 20°, yielding a wire grading of \( R' = dR/dz = 0.18 \). The real part of the refractive index of the \( m = 0 \) mode, \( n_{10}^{(0)}(R) = \text{Re}(k_{10}^{(0)}/k_0) \), was obtained from the eigenvalue Equation (6) as described above and plotted in Figure 1(b). The resulting eikonal parameter remains below unity even for very small apex radii and approaches a finite value of \( \delta \approx 0.8 \) as the wire radius approaches zero (Figure 1(c)), as required for adiabatic plasmon nanofocusing.

In practice it is technologically challenging to fabricate metal tapers with a geometry that closely matches the idealized conical shape used in the above simulations. Even when using advanced etching and ion-beam or electron-beam lithography techniques the smallest apex radii that have been produced so far are of the order of 5 nm [42]. This finite apex radius necessarily limits the expected increase in local refractive index, yet still gives rise to pronounced SP localization and field enhancement at the taper apex.
To directly probe this local field enhancement for conical gold tapers in a very broad spectral range, we have used electron energy loss spectroscopy (EELS) [86] and energy-filtered transmission electron microscopy (EFTEM) [103]. In these measurements, a monochromatic beam of relativistic electrons at 200 keV kinetic energy propagates parallel to the $x$-axis, perpendicular to the taper axis, which coincides with the $z$-axis. The electron beam focus lies in the $y$–$z$-plane at $x = 0$. The transmitted beam is energy-filtered and the detected EELS signal is recorded as a function of the electron impact parameter $(y_0, z_0)$ and as a function of loss energy.

The resulting images can be approximated as [86]

$$\Gamma_{\text{EELS}}(y_0, z_0, \omega) \propto \left( \frac{-qV}{\pi \hbar \omega} \right) \mathcal{R} \int_{-\infty}^{+\infty} dx \, E_x(x, y = y_0, z = z_0, \omega) e^{-i\frac{q}{\hbar}V x}, \quad (8)$$

and provide a map of the $x$-component of local electric field at frequency $\omega$ projected onto the wave function of the monochromatic energy beam, propagating with velocity $V$. Near the taper apex, the oscillatory term is essentially constant, and the images probe the vector component of the local electric near field that is pointing along the propagation direction of the incident electron beam.

The results of such transmission electron microscopy studies are shown in Figure 2 for two selected conical gold tapers with radii of curvature of ~10 nm and opening angles of ~45° (a) and ~15° (b), respectively. All images recorded at loss energies of less than 2 eV reveal a pronounced, spatially localized loss signal at the very apex of the taper. The spatial extent of this signal is increasing with decreasing loss energy, indicating that its size is given by the spatial resolution of the microscope and does not reflect the extent of the localized near field. At

![Figure 2](image_url)

**Figure 2.** Bright-field and energy-filtered transmission electron images of conical gold tapers with an opening angle of (a) 45° in the energy-loss interval from 1.3 to 2.3 eV and of (b) 15° in the energy-loss interval from 1.2 to 2.2 eV. The colour bar on the right symbolizes the energy-loss probability which is a measure of the overlap between the monochromatic electron wavefunction and the local electric near field in the vicinity of the taper apex. From [104].
energies above 2 eV, the local signal at the apex is drastically reduced. At these energies, interband excitation of d-band electrons has a strong influence on the dielectric function, and the resulting large imaginary part of the dielectric function suppresses the field enhancement. In summary, these measurements reveal pronounced local field enhancement at the taper apex in a very broad energy range, extending from 0.5 eV, a lower limit imposed by the energy resolution of the microscope, to ~2 eV, a value set by the onset of interband absorption. The EFTEM images of the taper region are readily understood by assuming that the electron beam indeed couples mostly to the \( m = 0 \) monopole mode of the taper.

Recent EELS measurements and simulations [86, 89, 91] confirm the broadband, non-resonant character of the field enhancement at the taper apex. The observation of oscillations in the spectra recorded at larger distances from the apex was interpreted as a characteristic signature of the excitation of higher order angular momentum modes [86]. The formation of standing waves due to the back reflection of SPP modes near the apex may also give rise to spectral oscillations for certain taper geometries, as evidenced in [89, 91].

To analyse the SPP modes that these nanotapers can support at optimum conditions, we performed numerical finite-difference time domain simulations for a three-dimensional conical gold taper. The taper was modelled as a hyperboloid with an opening angle of 19° and an apex radius of 12 nm and excitation was by a linearly polarized, spatially homogeneous 5 fs laser pulse, centred at 1.55 eV, with the electric field along the taper axis. The resulting local field distribution near the apex is shown in Figure 3 at an energy of 1.55 eV, below the onset of interband absorption in the gold taper. This evidences the formation of a dipolar field oriented along the taper axis and strongly localized at the apex. To further study the coupling of optical near fields to this taper, we placed a point-like dipole with a broad band emission spectrum, polarized along the taper axis, at a distance of a few nanometre from the apex [105, 106]. From the simulations, we determined

![Figure 3](image)

**Figure 3.** Finite-difference time domain (FDTD) simulation of the vectorial field around a gold nanotaper. A hyperboloid gold taper is optically excited by illumination with a femtosecond laser pulse with the electric field vector along the taper axis. The plots display the electric field component (a) \( E_x \) (along the taper axis) and (b) \( E_y \) (perpendicular), (c) shows the absolute value of the electric field with the vector orientation indicated by arrows.
the field that is transformed into a propagating plasmon and transported from the
apex along the taper in the reverse direction. The results of such simulations are
presented in Figure 4. The taper is assumed as a cone with an opening angle of 45°
and with an apex radius of 12 nm. The oscillating dipole efficiently excites SPPs at a
wide range of energies, which propagate along the taper away from the apex (Figure
4(a)). The induced field directly at the taper apex is enhanced by about a factor of
10 with respect to the incident dipole field (Figure 4(b)). This is in good agreement
with experimental observations, where the taper apex was directly illuminated by
focused laser light, and the field strength at the apex was determined with respect
to the field strength when the taper was absent [107–110]. For plasmonic nanofo-
cusing with a grating, however, it is more helpful to know the enhancement of the
electric field strength at the taper apex with respect to the field strength incident on
the grating. Measurements of photoelectron emission yield induced by plasmonic
nanofocusing indicate an apex field which is 12 times larger than the incident field.

Figure 4(c) shows a marked spatial variation of the induced electric field
amplitude $|E_x|$ along the taper shaft, calculated for a photon energy of 1.0 eV. The
coupling efficiency is calculated as the ratio of power emitted by the dipole that
is converted into a propagating plasmon wave. The SPP power is obtained by

![Figure 4. FDTD simulation: SPP excitation on conical gold tapers by coupling to a point-like dipole emitter placed at a distance of 3 nm from the taper apex. (a) Maps of the local field amplitude $|E|/|E_0|$ normalized to the amplitude $|E_0|$ of the incident field at the taper apex, for four different photon energies of the SPP field. The scale bar is 20 nm. (b) Spectral intensity of the incident point-dipole field (dashed red curve) and of the induced electric field (blue curve) at the taper apex. A field enhancement factor of 10 is observed which is essentially independent on the photon energy for energies of up to 2 eV. (c) Spatial variation of the induced electric field amplitude $E_x$. The excitation of propagating SPP fields at the taper shaft is apparent. (d) Coupling efficiency of a gold taper, i.e. the ratio between the power $P_S$ converted into propagating plasmons and that emitted by the dipole, as a function of excitation energy. The graph demonstrates a high coupling efficiency over a broad bandwidth, up to photon energies of about 2 eV.](image-url)
integrating the $x$-component of the Poynting vector along a plane positioned at a distance of 1.5 μm from the apex. The coupling efficiency (Figure 4(d)) remains above several tens of per cent over a broad bandwidth, up to the gold interband absorption at a photon energy of about 2 eV. This again demonstrates that these gold nanotapers are indeed, in very good approximation, very broadband, essentially non-resonant antennas.

An interesting aspect of these simulations is that they provide a time-domain perspective of the plasmon dynamics in the apex region. They indicate that it merely takes ~1 fs for the field to be transformed from an LSP at the apex into a propagating SPP modes at the taper shaft. This time should be compared to the typical lifetime of an LSP on a gold nanoparticle of ~ 10 nm radius. This lifetime (~5–10 fs) is indeed much longer than the 1 fs transformation time. Hence non-radiative damping of the localized apex mode due to Ohmic losses is less efficient than mode transformation into propagating SPPs. This suggests that the conversion efficiency can be further optimized by tailoring the apex geometry.

2.3. Fabrication of conical gold tapers and grating coupling to plasmons

Gold tapers are produced from commercially available gold wire with a diameter of 125 μm, which is annealed prior to etching to yield a single-crystalline structure (see [42] for details on tip production). The annealed wire is then electrochemically etched in HCl (aq. 37%) with a rectangular pulsed voltage applied between the wire and a platinum ring electrode. The taper shape is inspected by scanning electron microscopy. This etching method typically yields tapers with a very smooth and defect-free surface, with a tip opening angle between 20° and 30°, and with a radius of curvature at the taper apex well below 30 nm and reaching down to below 10 nm with reasonable yield (see Figure 5(b)). The sharpest tapers with apex radii around 10 nm are selected and a line grating is milled onto their taper shaft at a distance of several tens of μm from the apex with a focused ion beam (FEI Helios 600i). Here, we use a simple equidistant line grating as coupler with a line spacing of about 1–1.5 μm, slit width of ~400 nm and a groove depth of ~200 nm. With such gratings, we can excite SPP radiation on the taper shaft with a spectral bandwidth of ~200 nm [42,75,110]. This bandwidth may in principle be further enhanced by using more sophisticated grating designs such as fan gratings [111]. The coupling efficiency of the grating can be tailored by using non-rectangular slit shapes or chirped gratings [81] or by adapting the wavefront of the laser radiation that is incident on the grating [85].

For imaging, the grating is illuminated by light from a titanium sapphire laser, with a spectral bandwidth of 10 nm centred at 800 nm. We typically apply about 300 μW average power at a repetition rate of 80 MHz. Figure 5(c) shows a wide-field image of the illuminated taper, recorded in back-scattering geometry. Light scattering from the illuminated grating is spatially well separated from that from the apex region. This evidences efficient excitation transport by propagating SPPs,
which travel to the apex along the smooth taper surface without much scattering. Due to finite spatial resolution of the collecting microscope, emission from the $m = 0$ and $m = \pm 1$ modes cannot be distinguished.

3. Plasmon nanofocusing for optical imaging

3.1. Demonstration of plasmonic nanofocusing

In order to characterize the nanofocused light spot and to isolate the emission from the $m = 0$ mode, we performed angle-resolved studies of the light transmission through a 30 nm thick gold film deposited on a glass cover slip. The purpose of the semi-transparent gold film is to reduce the transmission of propagating light fields while enhancing that of evanescent fields in the tip-sample region. The transmitted light was collected using a high-numerical-aperture oil immersion (NA = 1.3) microscope objective (MO) and recorded as a function of tip-sample distance, controlled via a tuning-fork-based force sensor in a non-contact mode atomic force microscope (AFM, see [75,84] for details). The $k$-vector distribution of the collected light, i.e. the intensity distribution at the back focal plane of the MO was imaged onto a CCD camera. Light that is associated with in-plane $k$-vectors $k_0 < k_{||} \leq 1.3 \cdot k_0$, i.e. larger than $k_0 = \omega / c_0$ and up to the largest $k$-vectors supported by the MO, is found in a ring-shaped distribution on the CCD camera (see [84] for details). This geometry allows us to selectively image forbidden light, originating from evanescent fields at the taper apex [113].
Polarization-resolved $k$-space images of the light transmitted through this gold film are shown in Figure 6 for an intermediate tip-sample spacing of 120 nm. A polarizer was placed in front of the CCD detector with its orientation as indicated by the blue arrows. For clarity, the central part of the screen (with $k_{//} < k_0$) is covered, such that only the light associated with the evanescent near fields is shown. For each polarizer setting, a dipolar emission pattern is found, with its main axis aligned with the polarizer axis. This is a distinct signature of a radially polarized light field. A quantitative analysis reveals a degree of radial polarization of >93%. This indicates that the forbidden light indeed stems from the lowest order, radially symmetric $m = 0$ mode.

To demonstrate plasmonic nanofocusing of the $m = 0$ mode to the very apex of our tapers, we studied light-scattering off a nanostructured metallic slit grating (Figure 7). For this, the grating coupler on the taper shaft was illuminated with light from the titanium: sapphire laser, and the signal scattered from the apex region was collected from the side, using a MO with 0.35 NA. As the sample, we prepared a 30 nm thick gold film on a glass substrate and milled 30 nm deep grooves by focused Ga-ion beam lithography (Figure 7(b), inset). When approaching the tip above a gold-covered section and detecting the scattered signal as a function of tip-sample distance, we find a strong increase in the signal for distances of less than 10 nm (Figure 7(a)). This increase has a very short decay length of only 8 nm. The increase in scattering signal is readily understood on the basis of a classical image dipole model [49,50,114]. Plasmonic nanofocusing to the very apex of the taper results in a dipolar emission pattern emerging from a spatially highly localized source at the tip apex. This dipole field is back-reflected from the surface resulting in a modification of the local field at the taper apex and, thus, a change in the light

**Figure 6.** Polarization-resolved $k$-space imaging of light emitted from the apex of a sharp conical gold taper (Figure 5(b)). Incident far-field light (800 nm wavelength) is coupled to the apex by plasmonic nanofocusing and collected in forward direction after passing through a 30-nm thick gold layer deposited on a glass substrate. Only leaky evanescent light with lateral wave vectors $k_0 < k_{//} \leq 1.3 \cdot k_0$ is collected and imaged onto a CCD camera through a linear polarizer. Polarization-resolved images are shown for a distance between tip and gold surface of 120 nm for polarizer settings of (a) 0°, (b) 45°, and (c) 90°, as indicated by the blue arrows. A clear, dipolar emission pattern is observed at all angles with an intensity distribution, which is independent on the chosen angle. This demonstrates that apex emission is dominated by the emission from the radially polarized $m = 0$ mode. Reprinted with permission from [112]. Copyright 2016 American Chemical Society.
scattering intensity. Due to the conical shape of the taper and the radially polarized field of the incident $m = 0$ mode the effective point-dipole is preferentially oriented along the taper axis. Both the observed enhancement in scattering intensity by a factor of 2.6 as well as the decay length of $\sim 8$ nm match reasonably well with the expectations from a simple model of a $z$-polarized point dipole positioned close to a spatially homogeneous surface [49,50]. Such a model predicts a somewhat shorter decay length of 4 nm as a characteristic of the point dipole field, but is known to underestimate the decay length of a more realistic apex field distributions by about a factor of two [115]. The pronounced signal enhancement and its short decay length are quite remarkable, in particular since these measurements have been recorded without the use of any demodulation technique by simply collecting the scattered light from the apex region. The data thus indicate a very efficient nanofocusing of the $m = 0$ mode to the very apex of the taper.

Typical power levels of the scattered light amount to few tens to hundreds of nW, roughly 0.1% of the laser power focused onto the grating coupler. The drastic enhancement in scattering intensity within the last 10 nm suggests that almost all the detected scattered light emerges from the focus in the apex region, generating a bright and essentially background-free isolated light spot with a diameter of less than 10 nm. This makes such tapers almost ideal plasmonic nanolenses, transforming a micron-sized laser focus into a tiny, nanometre-sized and spatially well-defined LSP spot.

When repeating this approach measurement for a number of positions across the grating, the enhancement in scattering intensity varies strongly with lateral signal, revealing intensity maxima in the centre of the wider gold stripes (Figure 7).

**Figure 7.** (a) Scattered light intensity as a function of tip-sample distance at fixed lateral sample position, revealing an exponential decay of the scattering enhancement with a decay length of $L_0 \sim 8$ nm. The increase in light scattering provides a map of the local field enhancement at the apex due to scattering off the metal surface. (b) Plasmonic nanofocusing light scattering images recorded from a periodic slit grating milled into a 30 nm thick polycrystalline gold film deposited on a glass surface. The back-scattered light intensity is recorded as a function of the tip-sample distance $d$ and the lateral sample position $x$. A pronounced local enhancement in scattering intensity is seen when the tip is approached to within 10 nm of the sample surface. Inset: Scanning electron microscopy image of the grating. Reprinted with permission from [112]. Copyright 2016 American Chemical Society.
This enhancement in scattering intensity thus provides a measure of the locally induced dipole moments in the sample and thus, the near-field mode profile or the partial local optical density of states, projected along the tip axis [116–119] at the frequency of the incident laser.

### 3.2. Plasmonic nanofocusing microscopy of single metal nanoparticles

The demonstrated plasmonic nanofocusing capabilities of our microscope make it particularly interesting for probing local optical properties of nanostructures with high spatial and spectral resolution. As a first application, we use it to study off-resonant light scattering of small gold nanoparticles with 10 nm diameter. As is well known [120], such particles have well-defined dipolar particle-plasmon resonances at around 510 nm with typical on-resonance extinction cross sections of few tens of nm². For off-resonance excitation at around 800 nm, Equation (1) dictates that the extinction cross section is largely reduced, to a few tenths of nm². This small cross section makes it exceedingly difficult to sense the optical fields of a single nanoparticle, using either far-field or near-field methods. A plasmonic nanofocusing image of such particles is reported in Figure 8. For this, we illuminate a grating coupler on a nanofocusing taper with spectrally broad band laser light from a coherent white light source and simply collect the light that is backscattered from the taper apex. This light is then spectrally dispersed in a monochromator and detected with a CCD camera. When scanning sample position relative to the spatially fixed nanofocusing taper, local light-scattering spectra are recorded. Data, recorded at a fixed tip-sample distance of 2 nm, are shown in Figure 8. Here, simultaneously recorded topography images (Figure 8(a)) and optical images (Figure 8(b)) at a laser wavelength of 815 (+/− 5) nm are compared. Despite their small extinction cross section, the nanoparticles are clearly visible in the optical images, showing a local reduction in light scattering intensity by about 5–8%. A cut through both images (Figures 8(c) and (d)) shows anti-correlated topography and optical signals. The particles appear with a diameter of about 25 nm both in the optical and topographic images. This diameter is roughly a factor of two larger than the nominal diameter of the spheres, and is given by a morphological convolution of the particle shape with the taper apex of finite radius. Cross sections indicate a spatial resolution of better than 15 nm, when applying a 10/90 criterion for the rise in signal intensity at the edges of the particle. Measurements of larger nanoparticles show that even higher resolution of well below 10 nm can be reached [42].

The physical origin of the reduction in the scattering intensity at the particle position is readily understood by analysing light-scattering images as a function of the distance between tip and sample. Experimental results from a gold particle with a diameter of 30 nm are shown in Figure 9(a). For such weakly scattering objects, these images reveal a more pronounced far-field background than in Figure 7, which increases with increasing distance by roughly a factor of two.
for distances of up to 100 nm [84,112]. This background is independent on the sample position (Figures 9(b) and (c)). When approaching the tip to the glass
substrate supporting the gold particles, an enhancement in light scattering intensity is seen within the last 10 nm, as in Figure 7(a). The enhancement is much weaker than for a gold surface, and amounts to only ~10%, in contrast to ~100% for the gold surface. This is readily understood on the basis of the image dipole model [50] as a consequence of the different dielectric functions and hence the different amplitudes of the image dipoles induced in a planar gold and glass surface, respectively. When approaching the tip to the centre of the gold nanoparticle, however, the enhancement of the light scattering is negligible. As a consequence of the off-resonant excitation of the nanoparticle, the local scattering cross section and thus the projected local density of optical states is small and, hence, no near-field enhancement in light scattering intensity can be detected. The contrast in Figure 9(b) is thus a mixture of the distance-dependent variation in background signal and the local enhancement in near-field scattering, probing the partial local optical density of states. Both contributions are readily distinguished by recording distance-dependent light scattering images, as shown in Figures 7 and 9. Alternatively, plasmonic nanofocusing offers the attractive possibility to record spatially resolved light scattering spectra, allowing to separate near-field signal and far-field background by means of their different spectral characteristics. Such an analysis is currently underway.

3.3. Plasmonic nanofocusing of ultrashort light pulses

The results in the previous section demonstrate the creation of nanometre-sized light spots by plasmonic nanofocusing on conical gold tapers and discuss their use in imaging and spectroscopy. One of the unique properties of these conically tapered waveguides is their enormous spectral bandwidth. They support, transport and nanolocalize radially polarized $m = 0$ SPP modes in a broad spectral range, from the visible to the infrared. As such they seem ideally suited for nanofocusing of ultrashort optical pulses. This opens up the possibility to generate spatially isolated, nanometre-sized light spots with few cycle time duration and high intensity at the very apex of such a tip. Such controlled, ultrafast light localization seems attractive for a quite a number of applications, ranging from time-resolved ultrafast spectroscopy of single nanoparticles, to spatially-resolved nonlinear optical spectroscopy or ultrafast scanning tunnelling microscopy. So far, only few of these applications have actually been pursued, most likely due to the remaining technological challenge of fabricating high quality tapers [42,111].

An important prerequisite for such work is to demonstrate plasmonic nanofocusing of few cycle pulses and to directly measure the temporal duration of the localized plasmon field at the apex (Figure 10). For this, the grating coupler of a conical taper was illuminated with a sequence of two replicas of a 6 fs laser pulse with finely adjustable delay derived from a titanium:sapphire laser (Femtolasers, Rainbow), and the second harmonic (SH) radiation generated directly at the apex was recorded with a spectrometer as a function of the delay. This technique is
termed interferometric frequency-resolved autocorrelation (IFRAC) and yields the complete temporal waveform of the electric field at the apex (see [42] for more information about the measurement and [121,122] for general information on IFRAC). Prior to this measurement, the temporal waveform of the incident 6 fs pulses was determined by replacing the taper with a β-barium borate crystal, yielding the spectrum and a nearly flat spectral phase of the incident pulse (shown in Figure 10(b)). When inserting the conical taper, we ensured that nanofocusing was so efficient that SH radiation was only emitted from the taper apex. Simultaneously recorded light scattering images of single gold particles demonstrated light localization to a spot size of less than 10 nm.

IFRAC measurements using the SH from the apex, in comparison, yield a slightly longer pulse duration of about 10 fs. This slight pulse stretching is due to two effects: firstly, the intensity spectrum that is scattered from the apex is somewhat narrower than the incident spectrum. Specifically, short wavelengths are suppressed, which is due to the finite bandwidth of the grating coupler. This bandwidth may further be increased by using improved grating coupler designs, e.g. fan gratings [111]. Secondly, the spectral phase has acquired a slight second-order chirp of 25 fs². This chirp arises from the spectral dispersion of the $m = 0$ mode. It is so small that it can easily be compensated by conventional pulse compression using, e.g. chirped mirrors. Overall, the measurements give an upper boundary for the dispersion of the SPP wavepacket during propagation over 30 μm from the grating coupler to the apex and demonstrate nanofocusing of 10 fs ultrafast light pulses to the very apex of a conical plasmonic nanolens. Experimentally, we find that we can reach pulse energies of a few tens of fJ at the apex without thermal

Figure 10. Temporal duration of spatially localized plasmon wavepackets generated by plasmonic nanofocusing. (a) Time profiles of the electric field of the laser pulse incident on the grating coupler and of the electric field scattered from the apex, both determined by interferometric frequency-resolved autocorrelation measurements. (b) Spectrum and the spectral phase of the incident laser pulse support a pulse duration as short as 6 fs. (c) Spectrum and spectral phase as measured by light scattering from the taper apex. The spectral bandwidth is slightly reduced and a slightly quadratic phase is observed, reflecting a second-order dispersion of 25 fs². This results in an increase in pulse duration to 10 fs. Reprinted with permission from [42]. Copyright 2012 American Chemical Society.
damage. Even though this corresponds to only $10^5$ photons per pulse, the field intensities that are reached at the apex already exceed 10 TW/cm², making it possible to not only induce a broad range of nonlinear optical phenomena near the taper apex but to even reach the non-perturbative regime of strong-field optical nonlinearities in these solid-state nanostructures [107,123,124].

4. Plasmon nanofocusing for ultrafast electron microscopy

As a first application of the demonstrated ability to localize few-cycle optical pulses to nanometre spot sizes by plasmonic nanofocusing, we discuss and demonstrate its use to trigger multiphoton-photoemission from the apex of a conical gold taper. We implement this novel ultrafast nanoscale electron source as an emitter in a point projection microscope (PPM) and present first results obtained with this new instrument.

Metallic nanotips have recently served as an important model system for probing the interactions of solid-state nanostructures with strong laser fields with amplitudes close to the intra-atomic field strength. In atomic and molecular systems, such strong field interactions are known to enable a wealth of important phenomena such as high harmonic generation, attosecond streaking, and the generation of attosecond electron bunches for tomographic reconstruction of atomic orbitals [125–130]. Strong laser fields have opened the door to observing, analysing, and using the motion of electron wavepackets on the attosecond time scale [131–134]. During the last five years, strongly related phenomena have also been discovered for metallic nanotips. When illuminating the taper apex with ultrafast laser pulses, electron emission is observed by perturbative, multiphoton ionization [110,135], above-threshold ionization [123,136], and, for higher intensities, by non-perturbative strong-field tunnel emission [108,109,124,137]. In the strong-field regime, ponderomotive electron acceleration in the near field of the taper apex and carrier-envelope phase effects have been observed [107,108,123,124,138–141]. Very much like in atomic systems, the transition between weak and strong-field emission is observed when the Keldysh parameter [142],

$$\gamma = \frac{\omega \sqrt{2\Phi}}{e\cdot f \cdot E_0},$$

reaches a value of about unity. Here, $E_0$ is the peak electric field amplitude at the centre frequency of the laser, $\omega$. The electron charge is $e$, $m$ the mass of the electron, and $\Phi$ is the work function of gold. Importantly, the field enhancement factor $f$ at the apex can be quite large. For far-field illumination of sharp gold tapers such as those shown in Figure 5, values of up to nine could be demonstrated [107]. This implies that optical field emission phenomena from sharp metal tips can already be observed for rather small peak field intensities in the GW/cm² range. Such intensities can easily be reached with high-repetition rate few-cycle laser oscillators, making it comparatively easy to study strong-field nanophysics experimentally.

Interestingly, the ponderomotive acceleration of the photoemitted electrons by the driving laser field is quite different from that in atomic and molecular systems.
In the latter systems, the instantaneous electric field amplitude in the vicinity of the atom is spatially uniform and the effects of spatial field gradients on the quiver motion of the electron can generally be neglected. In contrast, such optical near-field gradients are particularly large in the vicinity of the apex of the metal tip. Here, the optical near field decays over a distance of only a few nm, and the ponderomotive acceleration of electrons is profoundly affected by these strong field gradients. As a consequence, the usual quiver motion of an electron in the oscillating laser field can be suppressed if the near-field decay length $l_F$ is shorter than the electron quiver amplitude $l_q = \frac{|e|fE_0}{mc^2}$. The spatial adiabaticity parameter $\delta = \frac{l_F}{l_q}$ is thus an important characteristic for the motion of electrons in the near field around nanostructures [124,143,144]. Near-field gradient acceleration hence provides an interesting experimental concept for controlling the trajectories and kinetic energy distributions of electron pulses generated by strong-field photoemission from metallic nanotapers.

Importantly, not only the fundamental optical properties of such metallic nanotapers have been studied in considerable detail. Metallic nanotips have also recently been employed as miniaturized electron sources in different time-resolved electron microscope set-ups [88,145,146]. The extremely small size of the emitter region results in an exceptionally high beam brightness and excellent coherence properties, which are critical for emerging applications in ultrafast electron diffraction [135]. When being implemented in ultrafast transmission electron microscopes (TEM), spot sizes of the focused electron beam of a few nanometres could already be demonstrated [132,147–149]. Currently, however, the time resolution of such microscopes is limited to a few hundred femtoseconds, both by kinetic energy spread of the photoemitted electron beam and by Coulomb repulsion in multi-electron beams. Conceptually, a straightforward approach to improve the temporal resolution of such microscopes to the much-desired few femtosecond or even attosecond regime would rely on a consequent miniaturization of the microscope and, in particular, a drastic reduction of the emitter-sample distance. This may, in principle, be achieved, for example in point projection microscopy set-ups [148] by placing the sample under investigation directly behind the electron emitter and by imaging a highly magnified shadow diffraction image of the sample onto an electron detector. Theoretical simulations suggest that it is necessary to reduce the emitter-sample distance to less than 10 μm for bringing the time resolution to less than 10 fs [150]. When using far-field illumination of the nanotip emitter apex, this almost inevitably results in simultaneous sample illumination and, thus, in undesired sample heating or photoemission [150].

One approach towards overcoming this challenge and avoiding unwanted sample illumination is to apply plasmon nanofocusing for electron photoemission, thereby removing the excitation laser spot from the apex and the sample. For this, we have implemented a modified PPM set-up [87], using a sharp conical gold taper for photoemission equipped with a grating for plasmon coupling at a distance of 50 μm from the apex. The grating with a period of 2.0 μm is designed to couple
light with a centre wavelength of 1.6 μm and with a broad spectral bandwidth to support few-cycle femtosecond laser pulses. The near-infrared wavelength is chosen because, besides scattering and absorption being small, the photon energy \( \hbar \omega = 0.77 \) eV is much lower than the work function of gold, \( \Phi = 5.5 \) eV. This means that a seven-photon process is required for photoemission of an electron from the Fermi level, and such a high nonlinearity restricts electron emission to regions of high local electric field strength, i.e. to the apex of the taper. The near-infrared light pulses are supplied by a home-built system of noncollinear optical parametric amplifiers and difference frequency generation, which creates pulses with a duration as short as 16 fs or three optical cycles, a pulse energy of up to 110 nJ, and at a repetition rate of 5 kHz [151].

We investigated photoelectron emission from nanofocusing tapers by spatially resolved electron detection on a microchannel plate detector (MCP) followed by a phosphor screen and a camera, which are placed perpendicular to the taper axis and at a distance of 15 mm from the apex (see Figure 11(a)). When raster-scanning the taper through the laser focus, which is formed by an off-axis parabolic mirror and has a diameter of about 6 μm, electrons are detected when the laser focus is positioned either on the grating or on the apex. Figure 11(c) shows the electron count as a function of the laser focus position, as was verified by monitoring the shadow image of the taper (not shown, compare [87]). One can see that for direct apex illumination, electron emission is rather low, while it is increased more than 50-fold to about 0.6 electrons per pulse, when the same intensity is incident on the grating. When illuminating the grating on gold tapers with a sharp apex of about 10 nm radius, electron emission is observed from the apex region at incident

**Figure 11.** (a) Schematic of the set-up for plasmon-driven electron emission for point-projection microscopy. Few-cycle near-infrared light pulses are grating-coupled to a sharply etched gold taper. The nanofocused SPP wavepacket leads to a strong field concentration and to plasmon-induced electron emission. Electrons interact with a sample placed at a small distance in front of the apex and are afterwards detected using a microchannel plate equipped with a phosphor screen. (b) Scanning electron microscopy image of the gold nanotaper with grating coupler, and (c), electron emission from the gold nanotaper as a function of the position of the 6 μm-diameter laser focus on the taper. Electron emission is detected when placing the focus either on the grating or on the apex of the taper. The weak electron emission for apex illumination is increased by a factor of >50 when the focus lies on the grating. Reprinted from [87].
pulse energies of around 160 pJ. This is evidenced by a homogeneous, circular electron distribution on the MCP. For comparison, when illuminating the grating of a blunt taper, with apex radius of curvature around 50 nm, electron emission sets in at much higher pulse energies of around 4 nJ. A heavily distorted electron distribution on the MCP is clear evidence that in this case electrons are emitted from the grating region rather than from the apex.

We can now use these data to characterize the plasmon-induced electron emission process. The incident pulse parameters correspond to a peak electric field strength of only about 0.5 V/nm, at the position of the grating. This field strength is too weak to induce significant nonlinearities and hence this prevents unwanted electron emission from the taper shaft. We estimate that plasmonic nanofocusing increases this field strength to around 6.0 V/nm at the apex, i.e. it leads to an enhancement of 12 with respect to the incident field strength on the grating. Due to the pronounced spatial field confinement at the apex such a high field strength is already reached with 10 fs pulses with moderate pulse energy of only a few hundred fJ. As a result, plasmon-induced electron emission from the apex is not only possible, but it is also induced already at very low pulse energies, during the short duration of few-cycle femtosecond pulses and from a spatially isolated volume of about 500 nm³.

We have demonstrated the advantageous microscopic imaging properties of this plasmon-induced electron source by implementing the taper in a PPM set-up. As a first test sample, we have dispersed single silver nanowires with a diameter of nominally 100 nm on a TEM grid, which was placed at a distance of a few tens of μm in front of the taper. Electrons were emitted by focusing femtosecond laser pulses on the taper grating, and shadow electron images of the wires were recorded on the MCP. Three such images are shown in Figure 12, for taper-sample distances of 60 μm (resulting in a low magnification of M~220), of 24 μm (M~540) and of 15 μm (M~830). At the lowest magnification the grid appears as a dark shadow, but when increasing the magnification, the wire and its branching become discernible, until finally, at the highest magnification even the edges of the wire are clearly resolved. The cross section of the wire (Figure 12(d)) shows a rectangular profile, similar to what has been observed in PPM before [152,153]. The inverted contrast and the increased width are readily explained by the well-known biprism effect [154], but the edge steepness of 110 nm gives an estimate of the spatial resolution in this first demonstration of PPM using plasmon-induced electron emission. It is in our current set-up limited by its sensitivity to weak static electric fields and to small variations of the potential at the wire surface. Our ongoing work aims at improving the spatial resolution by better shielding of static fields, as well as utilizing the short temporal duration of electron emission for time-resolved pump-probe experiments. Since our first implementation of such a plasmon nanofocusing electron source [87], related work has also been reported in Refs. [88,90].
5. Summary and conclusion

5.1. Summary over the presented work

This review treats our own recent work on plasmonic nanofocusing using sharply etched conical metallic nanotapers. We show that such tapers represent a very special type of nanoplasmonic lens with exceptional nanofocusing capabilities. By coupling far-field light to a few-micron spot size onto a grating coupler separated by a few tens of microns from the taper apex, SPPs are launched on the taper shaft. Radially polarized fields of the lowest order eigenmode of the taper are nanofocused into a localized plasmon mode at the very apex of the taper where it is partially scattered and converted into far-field radiation. During this nanofocusing, the spatial extent of the propagating plasmon mode gradually shrinks until it reaches a spot size solely limited by the geometric size of the taper apex. With present-day nanofabrication, plasmon spots with diameters of less than 10 nm are readily created and this spot size may in principle be reduced even further by sculpting the geometry of the nanotaper. As such, the apex of these conical tapers very much acts like a grey hole for light, capturing all the radiation from the radially symmetric taper mode before releasing it partially into the far field. Hence, these optically excited tapers serve as bright and coherent light sources with dimensions far below the diffraction limit and high potential for a variety of applications in nanooptics.

Experimentally, we demonstrate that our gold nanotapers resemble non-resonant antennas for a broad range of optical frequencies from the mid-infrared.
to the visible region, limited only by the onset of interband absorption in the metal. We directly demonstrate the rotational symmetry of the radially polarized plasmon mode and show that the near field generated by the localized plasmon at the apex is equivalent to that of a dipole oscillating along the taper axis. When approaching such tapers to metallic surfaces, the coupling between the optical near field at the taper apex and the local polarizability of the sample results in a pronounced increase in light scattering from the apex for tip-sample distances of only a few nm. The short decay length of the near-field interaction (~8 nm) reflects the small geometric size of the nanofocused light spot, as directly measured by spatially and spectrally resolved coherent light scattering spectroscopy of single, off-resonant gold nanoparticles, demonstrating a spatial resolution of this plasmonic nanofocusing microscope of <15 nm.

As a first application of the nanotapers’ ability to nanofocus ultrashort light pulses, we have used the high local field strength at the very apex of the taper to induce the emission of photoelectrons from a nanometre-sized region while remotely illuminating the taper at a distance of 50 μm from the apex. Due to the pronounced field enhancement at the apex, electrons can be efficiently released by highly nonlinear multiphoton emission even with low, femt joule pulse energies, greatly reducing undesired local heating effects. The favourable properties of the remotely triggered photoelectron source for ultrafast electron microscopy are demonstrated by point-projection microscopy imaging of a single nanowire. Remote electron emission removes laser illumination from the taper apex, and this facilitates electron microscopy and diffraction with short taper to sample distances of only a few μm or below. Over such short propagation distances a temporal spread of the released electron bunch is basically negligible and this may be a key to reducing the time resolution in ultrafast electron microscopy to 10 fs or even less.

### 5.2. Properties and potential of plasmon nanofocusing

It is the aim of this final section to briefly put our findings into perspective and to provide a personal view on the major prospects but also on the current limitations of plasmonic nanofocusing microscopy. First and foremost, the nearly loss-free propagation over mesoscopic distances and the pronounced spatial localization of the plasmon field at the apex is the basis for the taper’s potential for principally background-free all-optical imaging. In contrast to more commonly employed optical near-field microscopy schemes, where a near-field probe is brought into a diffraction-limited laser focus [47,48], undesired background scattering is avoided and an essentially point-like and isolated nanoscopic light and/or electron source is generated. This source can easily be employed for different types of surface sensitive scanning probe microscopy. Of course imaging and spectroscopy with full, three-dimensional spatial resolution is more challenging, but may in principle also be achieved by implementing different types of holographic imaging techniques, using either electrons or light as excitation sources.
We have shown that the spot size of the nanolocalized light source at the very apex of the taper can reach dimensions well below the diffraction limit and hence enables optical imaging with extremely high spatial resolution. So far, a lateral resolution on the order of 5 nm has been achieved and can possibly be improved towards 1 nm or even below by making use of gap plasmon localization \([43,112]\). Plasmonic nanofocusing microscopy thus offers optical imaging with unsurpassed lateral resolution. This high resolution is obviously linked to the ability to fabricate tapers with sufficiently small radius of curvature at the apex. Currently, these tapers can only be individually hand-crafted by chemical etching. The fabrication is therefore not fully reproducible and requires careful inspection and selection. Clearly, the ability to fabricate such tapers by batch processing \([155]\) would be an enormous breakthrough.

An interesting and important aspect of plasmonic nanofocusing is the well-defined radial polarization of the plasmonic mode transporting the energy to the taper apex. As such the vectorial properties of the electric field profile at the apex are well defined: it closely resembles that of a point dipole oriented along the taper axis. This is of key importance for a quantitative analysis of the near-field contrast. At the same time, however, this may also be considered a drawback since this makes it not so easy to freely control the polarization state of the optical near field. In principle, polarization control may be achieved by attaching nanoresonators with defined geometry or by using phase-locked pairs of excitation pulses, as illustrated, e.g. in \([81]\). Alternatively, conventional, dielectric scanning probe tapers may of course be used to generate optical near fields with arbitrary polarization state. In this case, however, the suppression of scattering background, e.g. by tip modulation techniques, is important.

As is well known, near-field microscopy is an invasive technique. The coupling between the near field of the taper and the optical resonances of the nanostructure will necessarily perturb the optical response of the nanostructure and this is often considered to be a drawback of optical near field spectroscopy. In the case of plasmonic nanofocusing we believe, however, that this is a strength rather than a nuisance. The optical near field of the taper is well-controlled and the near-field coupling between taper and nanostructure can be measured with high precision by recording local scattering spectra. As such, we trust that the application of this method to nanospectroscopy will provide unique new insight into the optical properties of various nanostructures and – in particular – into vectorial near field couplings between nanostructures. Of course this near-field coupling between taper and nanostructure may be greatly reduced by placing an off-resonant dielectric nanoresonator at the very apex of the taper.

One of the most interesting and so far basically unexploited features of these plasmonic nanolenses is the non-resonant and broadband nature of plasmonic nanofocusing. Such conical tapers support surface plasmon polariton modes in a broad, more than octave-spanning spectral range, which makes them ideally suited for ultrafast, coherent light scattering spectroscopy of a variety of quantum
systems in their natural, dense environment. To our knowledge, this is a unique feature of conical nanotapers. Currently, plasmonic nanofocusing has mostly been studied for gold nanotapers. Besides gold being a rather soft material, which limits the durability of the tips, this also restricts the spectral range of nanofocusing in practice to wavelengths above roughly 700 nm due to the onset of interband absorption. In order to extend the spectral range towards shorter wavelengths and in order to obtain more durable tips it is of high interest to examine different materials for tip production or for coating the tips while maintaining the small focus size.

The broad bandwidth supported by the nanofocusing tapers enables the generation of nanofocused light spots with ultrashort, few-cycle time duration, when launching coherent plasmon wavepackets with state-of-the-art femtosecond lasers. As such they represent an interesting platform to perform time-resolved, nonlinear optical spectroscopy in a nanometric volume. In combination with multiple pulse sequences this allows for pump-probe measurements and coherent 2D spectroscopy with nanometre-spatial resolution. Specifically, sharply etched metal tapers offer an interesting opportunity to explore the ultimate regime of strong coupling between a single solid-state quantum emitter and a localized light field at room temperature.

With pulses of few-femtosecond duration, a local electric field strength of 20 V/nm or a local peak intensity of 40 TW/cm² can easily be reached. This enables the utilization of nonlinear optical effects for nonlinear nanospectroscopy. In particular, using second or third harmonic spectroscopy can extend the spectral range of gold nanotapers to wavelengths below the threshold for interband absorption. Furthermore, such high local electric field strength can be utilized, e.g. for the study of strong-field effects in solid-state matter and for ultrafast electron generation. We firmly believe that plasmonic nanofocusing will be a key enabling technology for ultrafast electron microscopy. Our initial result suggest that this novel ultrafast electron gun may open up a new path towards the realization of ultrafast scanning tunnelling microscopy, ultrafast electron holography, and also novel, related types of scanning probe microscopy. Plasmonic nanofocusing along sharply etched gold nanotapers may in the future form the basis for novel types of ultrafast microscopy, combining both electron and optical imaging with (sub-)nanometre spatial resolution and hence is likely to find key applications in nanoscience.

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**Disclosure statement**

No potential conflict of interest was reported by the authors.

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