NONLINEAR NANOPROBES FOR CHARACTERIZING ULTRAFAST OPTICAL
NEAR FIELD

A Dissertation in
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by
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ABSTRACT

With the rapid development of ultrafast optics and nanophotonics, it is crucial to measure the spatiotemporal evolution of an ultrafast optical near field in nanometer spatial and femtosecond temporal resolution with minimal perturbation. Although near-field scanning optical microscopy (NSOM) can achieve nanoscale spatial resolution and various ultrashort pulse diagnostic tools can characterize femtosecond laser pulses, yet such capability to non-invasively characterize the nanoscale characteristics of femtosecond pulses in all three spatial dimensions remains elusive.

In this dissertation, we developed different types of nonlinear optical probes to characterize ultrashort optical pulses. The nonlinear optical probe is composed of three parts, a silica fiber taper, a single nanowire bonded to the end of the fiber and nonlinear nanoparticles attached on the tip of the nanowire. The optical fiber taper can be readily mounted on a mechanical stage and served as a macroscopic interface for handling and positioning control. The single nanowire bridges the dimension gap between the nanocrystals and the fiber taper, and is critical for achieving large aspect ratio and hence minimizing optical scattering and perturbation. The nonlinear nanoparticles give rise to its capability to characterize ultrashort optical pulses. The unique fusion of nanoscale and nonlinear features in developed nonlinear optical probes provides the ability of probing ultrafast optical field in complex 3D micro- and nano- structures. The demonstration of such ability is crucial for understanding the interaction of ultrafast optical fields and nanoscale systems. The fabrication processes of the nonlinear optical probes are illustrated in detail and the optical properties of the probes are investigated.

Two different types of nonlinear optical probes, two-photon fluorescent nanoprobe and Second HARmonic nanoProbes (SHARP), are fabricated. Interferometric autocorrelation
measurements near the focal point of an objective through two-photon fluorescent nanoprobes are presented. By replacing the two-photon fluorescent nanoparticles with second harmonic nanocrystals on the tip of optical nanoprobes, more advanced pulse characterization technique, frequency resolved optical gating (FROG), can be applied and detailed pulse profile information, including both amplitude and phase profiles, can be retrieved. The spatial resolution of SHARP is limited by the size of the nanocrystal on the tip. By employing auxiliary focused ion beam (FIB) nanomilling technique, we also fabricated SHARP containing only a single nonlinear nanocrystal. Pulse characterization near the focal point of a high numerical aperture objective by using the SHARP is demonstrated. Furthermore, ultrashort optical pulse characterization results in complex micro- or nano-structures, such as in the core region of a hollow-core photonics crystal fiber, based on collinear second harmonic generation-frequency resolved optical gating (SHG-FROG) measurements are presented. Last but not the least, preliminary ultrashort optical pulse characterization capability of the SHARP in turbid medium (diluted milk) is investigated.
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Chapter 1. Introduction

1.1 Necessity for ultrashort optical pulse measurement

LASER (Light Amplification by Stimulated Emission of Radiation) is one of the greatest inventions in the 20th century. Since its inception, laser has fundamentally shaped the world and changed the way we live or even think. As an important branch of this technology, mode-locked lasers, producing optical pulses on the order of tens of picoseconds ($10^{-12}$) to a few femtoseconds ($10^{-15}$), find a broad range of practical applications in the fields of fabrication industry [1-3], medical imaging [4-6] as well as scientific research and so on. Together with the blooming development of femtosecond laser techniques, from ultrafast optical pulse generation to manipulation, the femtosecond laser pulse characterization methods also significantly advanced at the same time.

In terms of a mode-locked laser system, the pulse width of a laser output is one of the most important parameters for laser scientists and engineers. The pulse measurement methods are not only used to identify the characteristics of the laser system operation, but also to provide detailed ultrashort optical pulse information for increasing application demands. For instance, the application of ultrafast laser for material processing has gained increasing interests in industry [7-13] due to its high precision characteristics. Experimental results demonstrated that ultrashort laser with various optical pulse durations had different
effect on hole drilling [14] and line cutting of materials such as silicon and stainless steel [15, 16].

In the field of femtochemistry, careful control over the generation of the femtosecond pulses and pulse shape are critical to the direct observation of the dynamic change in molecules [17]. A shaped ultrashort pump pulse also can be used to demonstrate the excitation of molecular vibrational coherence via impulsive stimulated Raman scattering [18]. Many other ground-breaking experiments in atomic and molecular level also reveal that complete knowledge of complex ultrashort laser pulse profile is the key to the process [19-24].

In multiphoton microscopy, the incident ultrafast laser beam with different temporal and spectral properties can have significant different signal yields [25]. Also, a tailored femtosecond laser pulse can control the two-photon transitions process. By varying the spectral phase, cancellation of the transitions due to destructive quantum interference was observed [26-28]. In order to control the laser pulses, it is important to develop accurate ultrashort pulse measurement methods.

In addition to ultrashort laser pulses measurement requirement from practical applications and the diagnostics for femtosecond laser system, increasing need for spatially resolved ultrafast optical pulse characterization techniques has also gained more and more research attentions in recent years.
1.2 Development of ultrashort laser pulse measurement techniques

The advancement of ultrafast optical pulse characterization techniques is closely related to the generation of shorter ultrashort laser pulses and increasing demands for accurate measurement of optical pulse profiles in a wide range of practical applications.

In the early stage of the mode-locked laser development, the pulse duration of laser output was in the order of several picoseconds or longer. Together with the pulse spectrum measurement, streak camera was used to characterize the temporal information of the pulses [29]. The inherent speed limitation of the method restricted its application as the duration of ultrashort laser pulses continue to shrink in time.

Owing to the experimental simplicity and ease of result interpretation, the correlation measurement of ultrashort laser pulse is commonly used to extract pulse width information [30-32]. The correlation method is based on the fact that the nonlinear signal intensity (second harmonic signal or two-photon florescent signal) is proportional to the multiplication of two pump intensities. Therefore a measurement of the nonlinear signal as a function of the time delay between two incident pump beams gives an estimate of the pulse duration. One common way to implement the correlation measurement is by using second harmonic bulk medium which can be limited by the non-uniformity of phase-matching condition over wide range of spectrum for ultrashort laser pulse with very narrow pulse duration (broadband spectrum width). However, this can be overcome by using nanocrystal whose phase matching condition is relaxed over broad spectral bandwidth, as the nonlinear medium in the measurement.

Another method to characterize the femtosecond laser pulses is interferometry. The phase
information of a target optical pulse can be extracted by interfering the pulse with a known reference pulse. Spectral Phase Interferometry for Direct Electric-field Reconstruction (SPIDER) [33] is an example for this method.

In order to have a detailed pulse shape of ultrashort laser pulses, both pulse amplitude and phase, more advanced pulse characterization method is required. Second harmonic generation frequency resolved optical gating (FROG) is one of the most popular techniques to fulfill this demand [34-36]. In the measurement, a femtosecond pulse gates a copy of itself in a second harmonic medium and the generated second harmonic signal is then spectrally resolved as a function of the time delay between the two pulses. Retrieval of the pulse amplitude and phase from the FROG trace is accomplished by using a two-dimensional phase-retrieval algorithm [36-38]. The temporally and spatially averaged pulse measurement results from FROG technique are adequate for most of applications. However, the need for the knowledge of spatially-resolved pulse profile rose as the advancement of research and applications. There is an important knowledge gap in the emerging field about how to non-perturbingly map the evolution of an ultrafast optical near field in nano-femto scale spatiotemporal resolution. The work presented in this dissertation is aimed to develop the capability of nano-femto scale spatiotemporal characterization of ultrafast optical near fields in complex micro/nano-structures in all three dimensions. We addressed the challenge by combining the advanced pulse retrieval capability of FROG with well-established high spatial scanning ability of NSOM technique. The demonstration of such ability is crucial for understanding the interaction of ultrafast optical fields and nanoscale systems.
1.3 Dissertation outline

In this dissertation, we developed different types of nonlinear optical nanoprobes, including two-photon fluorescent and second harmonic nanoprobes, for spatiotemporal characterization of various ultrafast optical near field. The unique fusion of nanoscale and ultrafast features in these devices, which consist of fiber taper, nanowire and nanoparticles, gives rise to their capability to characterize ultrashort optical pulses with nanoscale spatial resolution. By employing FIB nanomilling technique, we also fabricated nanoprobes containing only a single nonlinear nanocrystal attached on the end of a carbon nanotube which can in turn potentially lead to even higher spatial resolution.

In chapter two, the basic principle of femtosecond laser pulse generation technique and experimental setup of our femtosecond laser system are described. The detailed theoretical analysis of interferometric autocorrelation and frequency-resolved optical gating methods are discussed.

In chapter three, the fabrication procedure of the nonlinear nanoprobe is illustrated in this section. We described the advantages of proposed nonlinear nanoprobes and the comparison with NSOM technique is made in the chapter.

In chapter four, we have developed a nonlinear nanoprobe, which consists of a fiber taper, a ZnS nanowire attached to the taper, and two-photon fluorescent particles attached to the nanowire. Fabricated two-photon fluorescent nanoprobe was used to characterize femtosecond pulse through interferometric autocorrelation measurement. Two different types of two-photon fluorescent nanoprobes were developed and the measurement results of pulse duration and linear chirping parameter estimation were presented. The proof-of-principle experimental results demonstrated that the technique can be further extended and
led to full nano-femto spatiotemporal characterization of complex ultrafast optical near fields to understand ultrafast dynamics of complex nanostructures.

The ultrashort laser pulse characterization technique by using proposed second order nonlinear nanoprobe is further developed in chapter five. Fabricated SHARP comprises second harmonic nanocrystals attached to a carbon nanotube, which is in turn attached to a silica fiber taper. We demonstrated in situ pulse characterization directly in the air core of a photonic crystal fiber through second harmonic generation-collinear frequency resolved optical gating (SHG-CFROG). Further, it is shown that nanprobes containing a single nanocrystal in the tip of the carbon nanotube can be fabricated by auxiliary focused ion beam nanomilling. The pulse characterization result based on a SHARP containing only one single nanocrystal near foci of a high numerical aperture objective was presented. These results indicate that the proposed nanoprobe can open an avenue for probing the evolution of ultrafast optical fields in complex three-dimensional micro- or nanostructures. At the end of this chapter, observed anomalous power dependence in probes composed of multiwall carbon nanotube (CNT) and BaTiO$_3$ nanocrystals is investigated.

In the future work of the dissertation, preliminary ultrashort laser pulse characterization results in turbid medium (diluted milk) are presented. The advantages of developed nonlinear nanoprobes were discussed and optical probe characterization and main pulse measurement results were summarized. The proposed nanoprobe will significantly advance the ultrashort laser pulse characterization metrology and find increasing applications in the fields of nanotechnology and ultrafast technology.
Chapter 2. Theory of ultrashort laser pulse characterization

2.1 Ultrashort laser representation

An ultrashort laser pulse is basically a burst of electro-magnetic energy which appears in very short time duration, from tens of picoseconds ($10^{-12}$) to a few femtoseconds ($10^{-15}$). The mode-locking technique is the standard method to generate this kind of short pulse. The pulse durations for the output of mode-locked laser are generally in the order of picoseconds ($10^{-12}$ s) to femtoseconds. The basic principle of the mode-locking technique is by enforcing coherence or fixed phase relationship between different cavity modes of the laser resonator, a series of optical pulse train with very short time durations can be produced as the laser pulse oscillated in the cavity.

Since the demonstration of mode-locking technique [39], the ultrafast optical technology has found its remarkable application in many different fields. Passive mode-locking technique, which does not require any external modulation signal applied to the laser, has been widely used to produce ultrashort laser pulses whose bandwidth approaches the limits of the gain medium. One of such common devices is by using saturable absorber to produce short pulses. As the laser beam oscillates in the cavity, the saturable absorber will selectively absorb low-intensity light, and transmit pulses with sufficiently high intensity [40-41]. When the process continues, the leading and trailing edges of the pulse will be attenuated and high intensity part of the pulse, e.g. the pulse peak, will survive. After many trips, the pulse gets
shorter and shorter and eventually stabilizes. Another phenomenon that is widely used in the mode-locking technique is kerr-lensing effect. In the most popular laser sources of femtosecond laser pulses, the Ti:sapphire laser, this nonlinear optical effect is utilized to produce short pulses [42]. In this nonlinear process, the induced refractive index change of the gain medium is proportional to the intensity of the beam. Due to the non-uniform intensity distribution of Gaussian beam in laser cavity, by combining the self focusing effect with a hard or soft aperture, effective saturable absorption can be realized [43, 44].

The ultrashort laser pulse is an electro-magnetic wave as a function of space and time, i.e. \( \hat{E}(x, y, z, t) \). As we are mainly interested in its temporal behavior and treat it as a linearly polarized field, the complex field of the ultrashort laser pulse can be expressed as [36]:

\[
E(t) = \sqrt{I(t)} e^{i\omega_0 t} e^{-i\phi(t)}
\]

(2-1)

Where, \( I(t) \) and \( \phi(t) \) are the pulse intensity and pulse phase as function of time (in temporal domain), \( \omega_0 \) is the carrier angular frequency of the pulse. In ultrashort laser pulse measurement, we will focus on the temporal intensity and temporal phase of the pulse, \( I(t) \) and \( \phi(t) \). On the other hand, the ultrashort laser pulse can also be defined in the frequency domain as:

\[
E(\omega) = F[E(t)] = \int_{-\infty}^{\infty} E(t) e^{-i\omega t} dt
\]

(2-2)

\( E(\omega) \) is the Fourier Transform of complex field of the pulse \( E(t) \) and also can be expressed into:
\[ E(\omega) = \sqrt{S(\omega)}e^{-i\varphi(\omega)} \quad (2-3) \]

Similar to the definition in the time domain, \( S(\omega) \) and \( \varphi(\omega) \) are the spectral density and spectral phase of the optical pulse. From the definition above, \( I(t) \) will determine the time duration of the pulse and \( S(\omega) \) is the spectrum of the pulse.

### 2.2 Femtosecond laser generation

The most popular mode-locked laser system to generate ultrashort laser pulses is solid-state mode-locked Ti:sapphire laser [45,46]. Ti:sapphire is an attractive gain medium material for ultrafast laser operation. The broad gain bandwidth can support femtosecond ultrashort pulse generation and wide tuning spectral range at near-infrared region. A picture of the Ti:sapphire femtosecond laser system used in our experiments is shown in figure 2.1.
The pump laser for the Ti:sapphire laser system is a Verdi™ diode-pumped single-frequency 5W continuous-wave green (532nm) laser from Coherent Inc. A water cooling system is used to maintain the temperature of Ti:sapphire crystal gain medium. The Ti:sapphire laser kit system, which is shown in figure 2.2 is from Kapteyn-Murnane Laboratories L. L. C.
The mode-locking mechanism used in this system is Kerr lens mode-locking phenomena. The pump laser beam with an average power of 4.5 W is focused onto Ti:sapphire crystal through a lens in order to have a better overlap with the cavity mode in the crystal and to achieve a desired pump power density. The Ti:sapphire crystal is placed in the center of the cavity. The main cavity mirrors are used to support broad fluorescent bandwidth from gain medium and provide positive feedback for laser operation. A folded mirror is used to reduce the foot-print of the system. A pair of intro-cavity prism is utilized to compensate for the dispersion. A homemade mechanical slit mounted on a one-dimensional translational stage is inserted between the prism pair. The central wavelength of the femtosecond laser system can be adjusted from 750nm to 830nm by selectively blocking part of the spectrum through slit. Another pair of prism is placed outside of the laser cavity to further compensate for the dispersion. A transform-limited ultrashort laser pulse can be achieved by optimizing these two pairs of intra-cavity and outside cavity prisms. The femtosecond laser system will not automatically operate under mode-locked condition as we apply pump to the gain medium. A small and instant mechanical disturbance, by translating cavity mirror 2 back and forth quickly, on the laser cavity will initiate laser to jump to mode-locking oscillation from continuous wave operation.

Under 4.5W 532nm continuous wave pump condition, the average output power of the Ti:sapphire mode-locked laser system is around 500mw and the repetition rate is around 85MHz. The pulse duration for the transform-limited femtosecond pulse is around 50fs at full width at half maximum (FWHM). The spectral tuning range is from ~750nm to ~830nm (central wavelength), two typical spectrum of the ultrafast laser system are shown in figure 2.3.
After dispersion compensation, the transform-limited femtosecond laser pulse was measured by a commercial autocorrelator. The autocorrelation trace and intensity autocorrelation are shown in figure 2.4. The central wavelength of laser system for the measurement is around 810nm and calculated pulse duration is about 50fs at FWHM.
2.3 Interferometric autocorrelation

One of the popular tools to measure the pulse duration of femtosecond laser pulses is autocorrelator or any other instruments based on the principles of autocorrelation. Compared with intensity autocorrelation method, interferometric autocorrelations (based on second harmonic generation or two-photon absorption) are sensitive to the pulse width as well as the chirp parameters of the pulse [30, 47].

A standard setup for the realization of interferometric autocorrelation is a Michelson interferometer as shown in figure 2.5.

![Figure 2.5 An experimental diagram for interferometric autocorrelation](image-url)
A laser beam enters a Michelson interferometer and is separated into two identical copies by a beam splitter. Two replicas of the optical field collinearly propagate and are focused into a nonlinear medium (e.g. second harmonic crystals). The recorded signal by the detector is an interferometric autocorrelation as one arm of the interferometer is scanned. The detected signal is proportional to the function of [48]:

$$ G_2(\tau) = \int_{-\infty}^{\infty} \left| E(t) + E(t - \tau) \right|^2 dt \quad (2-4) $$

In which E(t) is the complex electric field, $\tau$ is the time delay between two arms of interferometer, and $G_2(\tau)$ is the interferometric autocorrelation. Replacing the field E(t) in the equation with:

$$ E(t) = \sqrt{I(t)} e^{i\omega t} e^{-i\phi} \quad (2-5) $$

Then we yield [48]:

$$ G_2(\tau) = A_0(\tau) + \text{Re}[A_1(\tau)e^{-i\omega\tau}] + \text{Re}[A_2(\tau)e^{-2i\omega\tau}] \quad (2-6) $$

In which,

$$ A_0(\tau) = \int_{-\infty}^{\infty} \left[ I^2(t - \tau) + I^2(t) + 4I(t - \tau)I(t) \right] dt \quad (2-7) $$

$$ A_1(\tau) = 4 \int_{-\infty}^{\infty} \sqrt{I(t - \tau)} \sqrt{I(t)} \left[ I(t - \tau) + I(t) \right] e^{-i[\phi(t - \tau) - \phi(t)]} dt \quad (2-8) $$

$$ A_2(\tau) = 2 \int_{-\infty}^{\infty} I(t - \tau)I(t) e^{-2i[\phi(t - \tau) - \phi(t)]} dt \quad (2-9) $$

Several unique properties can be found from the interferometric autocorrelation. A peak to background ratio of the interferometric autocorrelation of 8 to 1 can be found. The d.c. term of the function, $A_0(\tau)$, has a peak to background ratio of 3 to 1 instead. The term of $A_0(\tau)$ contains pulse duration information. From the FWHM of $A_0(\tau)$, we can estimate of the pulse width of the ultrafast optical pulses. The linear chirp parameter can also be quantitatively
determined from interferometric autocorrelation trace. By taking the Fourier transform of the $G_2(\tau)$ function, different frequency components will be separated in frequency domain, e.g. d.c. term, $\omega$ term and $2\omega$ term. Through identifying, filtering out the $2\omega$ component and performing inverse Fourier transforms, the linear chirp parameter can be estimated by assuming a particular pulse profile, e.g. Gaussian [48].

### 2.4 Frequency-resolved optical gating

In order to have more detailed information about ultrashort laser pulses including intensity and phase, advanced characterization technique should be introduced. One of the most successful tools is FROG [34-36] technique, which is a spectrally resolved correlation. The target femtosecond pulse was gated by a copy of itself in a nonlinear optical medium and the generated signal is then spectrally resolved as a function of the time delay between the two pulses. Compared with interferometric autocorrelation, the FROG measures spectrogram of the pulse, which is a measurement in the time-frequency domain. A general expression for FROG trace is [36]:

$$I_{FROG}(\omega, \tau) = \left| \int_{-\infty}^{+\infty} E_{\text{sig}}(t, \tau) e^{-i\omega t} dt \right|^2$$  \hspace{1cm} (2-10)

$I_{FROG}(\omega, \tau)$ is the recorded spectrogram by a spectrometer, $E_{\text{sig}}(t, \tau)$ is the signal field as a function of time and delay. For various FROG realizations, we have different mathematical expressions for signal field $E_{\text{sig}}(t, \tau)$ [36]. Two very important implemental geometries
involved in this dissertation are Second-Harmonic-Generation FROG (SHG-FROG) and Second-Harmonic-Generation Collinear FROG (SHG-CFROG). A simple representation of the difference between these two geometries is shown in figure 2.6.

Let us first consider the simple geometry of SHG-FROG in figure 2.6 (a) (off-axis second harmonic generation). The signal field for the case is given by:

\[ E_{\text{sig}}(t, \tau) = E(t)E(t-\tau) \quad (2-11) \]

Spectral resolving yields the Fourier transform of the signal with respect to time and detector is sensitive to the square of the magnitude, so, the SHG-FROG trace can be expressed by:

\[ I^{\text{SHG}}_{\text{FROG}}(\omega, \tau) = \left| \int_{-\infty}^{+\infty} E(t)E(t-\tau)e^{-i\omega t} dt \right|^2 \quad (2-12) \]

By considering \( E_{\text{sig}}(t, \tau) \) to be a one-dimensional Fourier transform with respect to \( \tau \), we can rewrite \( E_{\text{sig}}(t, \tau) \) as:
\[ E_{\text{sig}}(t, \tau) = \int_{-\infty}^{+\infty} E_{\text{sig}}(t, \Omega)e^{-i\Omega \tau} \, d\Omega \]  \hspace{1cm} (2-13)

Through substituting the equation for \( E_{\text{sig}}(t, \tau) \) into the expression of SHG-FROG trace, we have:

\[ I^{\text{SHG}}_{\text{FROG}}(\omega, \tau) = \left| \int_{-\infty}^{+\infty} E_{\text{sig}}(t, \Omega)e^{-i\omega t} e^{-i\Omega \tau} \, dt \, d\Omega \right|^2 \]  \hspace{1cm} (2-14)

From equation (2.14), the SHG-FROG trace is the square of the magnitude of the two-dimensional Fourier transform of \( E_{\text{sig}}(t, \Omega) \). We will show how to solve for the optical field \( E(t) \) in the later part of this section. Briefly \( E_{\text{sig}}(t, \tau) \) can be obtained from two-dimensional phase retrieval. From equation (2.11), we clearly see that, when \( t=\tau \),

\[ E(t) = E_{\text{sig}}(t, t) / E(0) \]  \hspace{1cm} (2-15)

Before we continue to the phase-retrieval problem, let us consider the geometry of SHG-CFROG (collinear second harmonic generation) in figure 2.6 (b) [49]. The signal field is given by:

\[ E_{\text{sig}}(t, \tau) = [E(t) + E(t - \tau)]^2 \]  \hspace{1cm} (2-16)

According to the new signal field relation, SHG-CFROG trance can be expressed as:

\[ I^{\text{SHG}}_{\text{C-FROG}}(\omega, \tau) = \left| \int_{-\infty}^{+\infty} [E(t) + E(t - \tau)]^2 \exp(-i\omega t) \, dt \right|^2 \]  \hspace{1cm} (2-17)

In order to establish the relations between SHG-FROG and SHG-CFROG, we expand the equation (2.17), do the Fourier transform and yield:

\[ I^{\text{SHG}}_{\text{C-FROG}}(\omega, \tau) \propto 2I^{\text{SHG}}_{\text{FROG}}(\omega) + 4I^{\text{SHG}}_{\text{FROG}}(\omega, \tau) + 2I^{\text{SHG}}(\omega) \cos(2\omega_0 + \omega) \tau \\
+ 4 \text{Re}\{E_{\text{SHG}}^*(\omega)E^{\text{SHG}}_{\text{FROG}}(\omega, \tau)[\exp(-i\omega_0 \tau) + \exp(i(\omega_0 + \omega) \tau)]\} \]  \hspace{1cm} (2-18)
Where, \( \omega_0 \) the carrier frequency of optical field \( E(t) \),

\[
I_{SHG}(\omega) \propto |E_{SHG}(\omega)|^2 \quad \text{and} \quad I_{SHG-FROG}^{SHG}(\omega, \tau) \propto |E_{SHG-FROG}^{SHG}(\omega, \tau)|^2.
\]

\[
E_{SHG}^{SHG}(\omega, \tau) \propto \int_{-\infty}^{+\infty} E(t)E(t-\tau)\exp(-i\omega t)dt \quad \text{(2-19)}
\]

\[
E_{SHG}(\omega) \propto \int_{-\infty}^{+\infty} E^2(t)\exp(-i\omega t)dt \quad \text{(2-20)}
\]

From the Fourier analysis of the equation (2.17) and (2.18), the first and second term of equation (2.18) are the d.c. part of in its Fourier domain. The third and fourth term of the Eq. (2.18) are well separated from d.c. part and can be removed by a digital low pass filter [49]. The second term of the Eq. (2.18), a part of the d.c. term, is exactly the same as the SHG-FROG (non-collinear second harmonic generation). This clearly shows that the SHG-CFROG contains all the information for the case of SHG-FROG. After filtering out the higher frequency component, the SHG-FROG can be obtained from SHG-CFROG by subtracting the first term of Eq. (2.18). After we have the SHG-FROG, the phase information can be retrieved by employing two-dimensional phase retrieval algorithm.

The two-dimensional phase retrieval algorithm can be implemented by the following steps [36]:

First, we start with a initial guess for the field \( E(t) \), including amplitude and phase, and generate a signal field \( E_{\text{sig}}(t, \tau) \) as shown in equation (2-11). Then, we perform one dimensional Fourier transform to the signal field with respect to \( t \) to have \( E_{\text{SHG-FROG}}(\omega, \tau) \) in frequency domain.

Second, the amplitude of the current guess will be replaced with the square root of the intensity of the experimental measured FROG trace \( I_{\text{SHG-FROG}}(\omega, \tau) \). The phase of current
guess will remain unchanged, which yields new \( E'_{\text{SHG-FROG}}(\omega, \tau) \). Then, we perform one dimensional inverse Fourier transform and yield \( E_{\text{sig}'}(t, \tau) \) in time domain.

Third, a new guess of \( E(t) \) can be achieved from \( E_{\text{sig}'}(t, \tau) \) by using equation (2-15). A calculated FROG trace based on new guess of \( E(t) \) can be constructed from equation (2-12). A FROG error \( G_0 \) is introduced to serve as a convergence measure as well as an acceptable error factor for iteration. If the calculated G error between experimental measured FROG trace and the calculated FROG trace is smaller than \( G_0 \), then, the new guess of \( E(t) \) will be accepted as final field.

Finally, if the G error is larger than \( G_0 \), the algorithm will go back to the first step with the new guess of \( E(t) \) and proceed the algorithm and iteration again. The last guess of \( E(t) \) with \( G < G_0 \) will be accepted as the final result of the field.

In this dissertation, the developed nonlinear nanoprobe is integrated with two-photon fluorescence and FROG technique to characterize ultrashort laser pulses in complex micro- or nano-structures with 3-D scanning capability. Femtosecond pulse measurement results based on interferometric autocorrelation and collinear second harmonic FROG techniques are demonstrated.
Chapter 3. Fabrication of nonlinear optical probes

3.1 Introduction of nonlinear optical probes

With the continuing development of nanophotonics where both nanodevices and ultrashort lasers have been playing central roles, it has become increasingly important to characterize optical fields at nano-femto spatiotemporal scale in order to gain fundamental understanding of the interaction between ultrashort optical pulses and nanoscale materials and devices [50-54]. Nanotechnology and ultrafast technology have also led to many innovative applications and created exciting scientific frontiers such as biophotonics and nanophotonics. However, there still exists a fundamental knowledge gap at the intersection of nano- and ultrafast technology. On one hand, the well-established near-field scanning optical microscopy (NSOM) technique can be used to achieve near field imaging with nanoscale spatial resolution, and on the other hand, various ultrashort pulse measurement techniques have been applied to characterize femtosecond laser pulses. However, currently it is still difficult to perform non-perturbative measurement of an ultrafast optical near field with nano-femto scale spatiotemporal resolutions.

In this dissertation we developed nonlinear optical probes for nano-femto scale characterization. The proposed nanoprobe consists of a silica fiber taper, a single nanowire (ZnS nanowire or Carbon nanotube) attached to the tip of the taper, and nonlinear particles (i.e., two-photon fluorescent micro- or nano-spheres, or BaTiO$_3$ nanocrystals, or LiNbO$_3$
nanocrystals) attached to the nanowire. The illustration of the nonlinear optical probe is shown in figure 3.1.

![Illustration of a nonlinear optical probe](image)

**Figure 3.1 Illustration of a nonlinear optical probe [Rendered by using POV-Ray]**

The nano- feature of the probe comes from the size of the nanowire and nanoparticles. The nanowire will ensure optical near field measurement with minimal perturbation and the size of nanoparticles determines the spatial resolution of the probe. The femto- feature of the probe comes from the nonlinear nanoparticles on the tip of the nanowire. The ultrashort laser pulse characterization measurement can be achieved through dynamic interactions between ultrafast optical field and nonlinear nanoparticles. The optical fiber is very easy to handle and readily to be integrated with near field scanning capability of current NSOM technique. So, by using developed optical probes, ultrashort pulse characterization in complex structures with nanoscale spatial resolution can be potentially achieved.
3.2 Nonlinear nanoprobe fabrication

3.2.1 Fiber taper fabrication

Corresponding to the three parts of the optical probe, the fabrication of the proposed nonlinear optical probe is carried out in three steps.

The first step of the fabrication is to prepare a fiber taper through regular optical fiber. There have been different techniques such as hydrofluoric (HF) etching available for fabricating fiber tapers from regular silica optical fibers [55-57]. In our experiments, we have used a fiber fusion splicer (Ericsson FSU-975) to fabricate fiber tapers by programming the discharging current and fusion time. A picture of the splicer is shown in figure 3.2.

![Figure 3.2 A photo of fusion splicer](image)

Figure 3.3 (a)-(c) shows a few snapshots of the fusion splicer screen illustrating the fiber taper fabrication process. Figure 3.3 (a) shows an optical fiber before being tapered. The
fiber taper fabricated by the fusion splicer is shown in Figure 3.3 (b). The fiber taper was then purposely broken in the middle into two by applying a slight perturbation as shown in Figure 3.3 (c). The diameter of the obtained taper tips is typically around 1–2 μm.

![Figure 3.3 Snapshots of the fiber fusion splicer screen illustrating the fiber taper fabrication process](image)

Figure 3.3 Snapshots of a fiber fusion splicer screen illustrating the fiber taper fabrication process. (a) an optical fiber before being tapered. The fiber taper fabricated by the fusion splicer is shown in (b). The fiber taper was then purposely broken in the middle by a slight perturbation as shown in (c) and ready to use.

### 3.2.2 Single nanowire attachment

The second step of optical probe fabrication is to prepare the nanowire and to attach a single nanowire to the tip of fiber taper which is fabricated in the first step.

We have two different kind of nanowires available for the experiments, Zinc Sulfide (ZnS) nanowire and Multi-Wall Carbon Nanotube (Nanolab, PD100L520). The ZnS nanowires
were synthesized [58-60] by late professor Eklund’s lab. The typical diameter of a ZnS nanowire is found using SEM to be 100 ~ 200 nm and the lengths is typically in the range ~ 20-30 μm. The carbon nanotubes are commercial available. Due to the excellent mechanical strength and hardness as well as large length to diameter ratio, carbon nanotubes are also good candidate for our application [61-64]. The length of a carbon nanotube can be as long as ~ 40-50 μm and the diameter is around 200nm. The nanowires were first dispersed on a glass substrate and then placed under a home-modified microscope system as shown in figure 3.4 (a).

![Figure 3.4](image)

*Figure 3.4 (a) a picture of home-modified microscope system and (b) the diagram of the setup*

The nanowire suspension (ZnS or Carbon nanotube) is dispersed on a glass substrate, the thickness of which is about 0.15mm. After drying in the air, it is placed under a home-modified microscope system. The glass substrate is mounted such that the side with the nanowires is facing down and the top can be accessed by an objective lens (including oil-immersion objective for higher resolution) as shown in figure 3.4 (b). A fiber taper is mounted on a three-dimensional linear translational stage. Before attaching the nanowire, a
small amount of optical adhesive (Norland Optical Adhesive 60) will be applied through a separate fiber taper to the side of glass substrate where nanowire resides. Since the nanowires are quite long, they can be easily observed under a microscope. A single nanowire can then be selected and glued to the fiber taper. Figure 3.5 shows a sequence of images illustrating this process. A ZnS nanowire can be seen in Figure 3.5 (a). A fiber taper (already with a small amount of optical adhesive applied to it) was moved toward the nanowire and successfully attached to it as shown in Figure 3.5 (b) – (f).

![Images of nanowire attachment process](image)

**Figure 3.5** A sequence of images showing the process of attaching a single ZnS nanowire to a fiber taper. (a) and (b): a fiber taper with a small amount of glue already applied to it was moved towards a ZnS nanowire; (c)-(f): the ZnS nanowire was successfully glued to the fiber taper and lifted off the substrate as the taper was translated.
After the nanowire was attached on a fiber taper, approximately half an hour of ultraviolet light exposure was then followed to cure the adhesive to ensure good bonding between the nanowire and the fiber taper.

### 3.2.3 Nonlinear particle attachment

The third step of fabrication is to attach nanoparticles or microsphere to the very end of the nanowire.

The microsphere or nanoparticles attachment is very similar to that of nanowire. The microsphere or nanoparticles suspension (fluorescent microsphere or BaTiO$_3$ nanocrystals) is dispersed on a glass substrate, air dried and also placed under the home-modified microscope system as shown in figure 3.6. The nanoparticles are facing down and the top can be accessed by a high numerical objective lens. A fiber taper with a single nanowire attached on the tip is mounted on a three-dimensional linear translational stage.
A fluorescent microsphere can be seen in Figure 3.7 (a). A fiber taper with a ZnS nanowire attached on the tip (already with a small amount of optical adhesive applied to it) was moved toward the microsphere and successfully attached to it as shown in Figure 3.7 (b) – (i). In figure 3.7 (c)-(e), the microsphere was pressed by the nanowire against the glass substrate and held for a while. From figure 3.7 (f) to figure 3.7 (i), the nanowire was gradually lifted up with microsphere attached on the end of nanowire and moved away from the surface of substrate.
The microscope images of nonlinear optical probes are shown in figure 3.8. A fluorescent microsphere was attached on the tip of a single ZnS nanowire on the nonlinear optical probes.

![Microscope images of nonlinear optical probes](image1)

**Figure 3.8 Two microscope images of nonlinear optical probes with single microsphere**

A Field Emission Scanning Electron Microscopy (FE-SEM) image of a nonlinear optical probe with a microsphere attached on the tip is shown in figure 3.9.

![FE-SEM image of nonlinear optical probe](image2)

**Figure 3.9 A field emission scanning electron microscope (FESEM) image of a nonlinear optical probe**
The procedure for attaching second harmonic nanocrystals is similar to that of attaching microsphere. Nanocrystal cluster with diameter smaller than 1 μm (e.g., from multiple, a few, to sometimes a single nanocrystal) can be obtained depending on the size and concentration of the dispersed nanocrystals on glass substrate. The attachment of nanocrystal clusters can also be achieved by first dispersing some nanoparticles on a separate fiber taper and then gently rubbing the nanowire on the nanoparticles. Nanocrystal clusters with various sizes in diameter are shown in figure 3.10.

Figure 3.10 Scanning electron microscope images of nonlinear probes functionalized with BaTiO$_3$ nanocrystals (a) nanocrystal cluster, (b) a few nanocrystals and (c) a single nanocrystal were attached to a single CNT
3.2.4 Focused ion beam refinement

The spatial resolution of the nonlinear optical probe is mainly limited by the size and distribution of the nanocrystals on the tip of the nanowire. Depending on the different applications, the fabricated nonlinear optical probes as previously discussed may be suitable or may require further refinement. In order to precisely control the size and position of the nonlinear nanocrystal(s) on the nanowire, an additional fine-fabrication procedure by using Focused Ion Beam (FIB) nano-milling can be utilized, which can remove the excess nanocrystals and trim the nanowire. Figure 3.11 shows two examples of nonlinear probes before and after the FIB nano-milling. A Quanta 200 3D Dual Beam FIB system was utilized in the experiment, during which a Ga\(^+\) ion beam (20 nm, 30 kV) was used to mill along a line.

![Figure 3.11 Fabrication of nonlinear probes by utilizing focused ion beam nanomilling. Two probes before (a, c) and after (b, d) the FIB nanomilling are shown respectively. (a) and](image)
(b): a small section of the carbon nanotube and a protruding nanocrystal were selectively removed by the FIB; (c) and (d): a small part of the carbon nanotube was removed by the FIB to result in a single nanocrystal located at the tip of the nanotube.

Now, we have developed a systematic approach to fabricate a variety of nonlinear optical probes. Particles of different sizes, ranging from microsphere to nanosphere, can be attached to the tip of a single nanowire, which is in turn attached on the end of fiber taper. Advanced nanofabrication technique, Focused Ion Beam (FIB) was used to further modify the probe to precisely control the size and position of the nonlinear nanocrystal(s) on the nanowire.

3.3 Comparison between nonlinear optical probe and NSOM

It is necessary to classify the differences between the developed nonlinear optical probes with existing Near-field scanning optical microscope (NSOM) technique [65-71]. They have fundamental differences not only on the operational principles but also in the fields of practical applications.

High resolution imaging (sub-wavelength) is an important topic in the field of optics. Near-field scanning optical microscope is a well-established technique for sub-wavelength imaging that beats the far-field diffraction limit in traditional optical microscopy. The resolution in classical microscopy is given by:

\[ D \sim \frac{\lambda}{2N.A.} \]

Where, \( \lambda \) is the wavelength of light and N. A. is the numerical aperture of the optical system.
Sub-wavelength resolution can be achieved by placing a probe very close to the surface of the specimen. In this way, the resolution of the image is only determined by the aperture size of the probe. Another critical factor of the NSOM technique is the distance control between the probe and the surface of specimen. With all those precise feedback systems available, AFM and NSOM become powerful techniques for surface investigation with sub-wavelength resolution. An illustration picture of the NSOM probe is shown in figure 3.12.

![NSOM probe diagram](image)

**Figure 3.12 A diagram for NSOM operation**

One of the popular operation conditions for NSOM probes is aperture-mode [72, 73]. A typical NSOM probe operated under this mode consists of a fiber taper and a layer of metal coating outside of the fiber taper. The aperture (without coating) at the tip of the probe will serve as a signal-collection entrance of the system as the probe scanned through the surface of the specimen. The collected signal can also be delivered through the fiber. So, the resolution of the system, which is determined by the size of scanning aperture, can be very high and beats the optical diffraction limit in traditional microscopy.

From a structure point of view, the developed nonlinear optical probes consist of three parts and the main difference is the active nonlinear optical elements on the tip of the probes. However, the NSOM probes are passive devices without any active elements involved. The
high resolution scanning capability of the NSOM is one of the most important features of the technique. Our proposed nonlinear optical probes are also made of silica fiber taper. So, they are readily to be integrated into current NSOM system and can take advantage of the well-established high resolution scanning capability.

From the function point of view, in most applications of NSOM probes, linear processes are involved and the techniques are based on measuring intensity of locally generated optical signal on the surface of specimen. The optical fiber taper will serve as a delivery optical path for excitation light and the signal will be collected through the tip of the taper. However, in our proposed nonlinear optical probes, the optical fiber taper is mainly served as a mechanical handing of the probe. The presence of nanowire extends the reach of fiber tapers and bridges the gap between macroscopic handling of fiber taper and precisely control over nanoscale particles on the tip of the nanowire. Especially for the introduction of carbon nanotubes (CNT), the large aspect ratio is very attractive for near-field optical characterization of complex micro- or even nanostructures. Another major difference between the nonlinear optical probe and NSOM probe is that there are various nonlinear optical processes involved in our probes. Due to all different kind of active nonlinear optical elements available on the tip of the probes, we can exploit the dynamical interaction between optical field and nanoparticles. Detailed information about the optical field in complex structures can be extracted from these interactions. As a demonstration of the application of the probes, femtosecond laser pulse characterization results were achieved by using two kind of nonlinear optical probes near the focal point of a high N.A. objective and a photonic crystal fiber. The pulse measurement will be illustrated in detail in the rest of the dissertation.
Chapter 4. Interferometric autocorrelation based pulse measurement

4.1 Two-photon excited fluorescence

Two-photon absorption is one of the most important nonlinear optical processes. It was first theoretically predicted in 1931 by Maria Goeppert-Mayer in her thesis [74] and experimentally demonstrated in the early 1960s [75, 76]. Two-photon absorption is a process of the simultaneous absorption of two photons to excite an atom from its ground state to an excited state. An illustration for the process is shown in figure 4.1.

![Energy diagram for two photon absorption process](image)

Figure 4.1 Energy diagram for two photon absorption process
The energy difference between the ground state and excited state of the atomic system is connected by simultaneously absorbing two identical laser photons. It is a third order nonlinear optical process, which requires high pump laser intensity to observe it.

The absorption cross section $\sigma$ is given by [77]:

$$\sigma = \sigma^{(2)} I$$  \hspace{1cm} (4-1)

where $\sigma^{(2)}$ is a coefficient which describes the efficiency of two-photon absorption.

Compared with the absorption cross section, which is a constant, in linear absorption, the absorption cross section in two-photon absorption process is proportional to the laser intensity.

The strength of absorption depends on the square of the laser intensity as [77]:

$$R = \frac{\sigma^{(2)} I^2}{\hbar \omega}$$  \hspace{1cm} (4-2)

Where $R$ is the atomic transition rate.

One example of the two-photon absorption process is the two-photon excited fluorescence technique [75], or in short two-photon fluorescence. Two pump laser photons with longer wavelength (lower photon energy) are simultaneously absorbed by a fluorophore. After the molecule system getting excited, a fluorescence photon will be emitted subsequently. The probability for the fluorescence signal emission is proportional to the probability of absorbing two pump photons simultaneously, which is proportional to the square of pump intensity. The wavelength of the fluorescent signal is usually shorter than that of the pump and fluorescent signal will have higher energy than either of the pump photon. So, in a typical two-photon fluorescence system, a pump laser beam with relatively longer wavelength (in near infrared region) is focused onto the specimen and the generated
fluorescence signal with relative shorter wavelength (in visible region) is emitted. Under exact the same excitation wavelength, the fluorescence emission may be different for the different types of fluorophore. Even for the same type of fluorophore, the fluorescence signal may also be different under various excitation wavelengths. A blue fluorescent microspheres spectrum (Duke Scientific Corp.) for different excitation conditions is shown in figure 4.2 (adapted from product specification).

Figure 4.2 Fluorescence spectrum of the blue fluorescent microspheres from Duke Scientific Corp. [adapted from product specification from Duke Scientific Corp.]

For the same kind of fluorescent microspheres, when the excitation comes from the wavelength of 365nm, the emission spectrum will be relatively smooth over the spectrum region from 400nm to 520nm. One main peak at 445nm is observed. If the excitation spectrum is shifted to a longer wavelength at 412nm, the emission spectrum will also change
significantly. The fluorescence spectrum region is located from 430nm to 520nm and two main peaks are observed at 445nm and 473nm.

In our following experiment, we used the same kind of blue fluorescent microsphere as shown in figure 4.2, but under different excitation wavelength via two-photon absorption. As a result, different emission spectral distribution was expected in the experiment.

One advantage of the two-photon fluorescence technique is the well separation between the excitation wavelength and the signal spectrum. For example, in our experiment, the excitation wavelength is around 810nm and the fluorescent signal is around 450nm which is well separated from each other. The two-photon fluorescence is a third order nonlinear process and its signal yield is proportional to the square of the pump intensity. It is very sensitive to the intensity of the excitation beams. Based on this property, another advantage of the technique is it can be used to characterize the femtosecond laser pulses near the focal point of high numerical objective lens. Ultrafast laser system normally has output pulses with short time duration and high intensity. After being focused by an objective, the pump intensity near the foci is extremely efficient to excite the two-photon process.

By taking advantage of the properties of fluorescent microspheres and integrating with our nonlinear optical probe technique, the femtosecond pulse characterizations by measuring the interferometric autocorrelation through the two-photon fluorescence from such nonlinear nanoprobe are demonstrated in this chapter. Our results indicate the proposed nanoprobe can lead to optical near-field measurement with nano-femto scale spatiotemporal resolutions.
4.2 Experimental demonstration of femtosecond laser characterization

Different kinds of femtosecond laser pulse characterization methods are available for measuring and characterizing optical pulses in different levels. The most popular methods are autocorrelation [30-32], FROG [34-36] and SPIDER [33]. It is often implicitly assumed that the spatial propagation and the temporal evolution of the ultrashort pulses are largely independent. Spatiotemporal characterization has been previously investigated to study spatiotemporal pulse distortions [78, 79]. With the continuing expansion of the application of ultrafast lasers (e.g., nonlinear microscopy), it also becomes important to characterize ultrashort pulses near the focal point of a lens, where the spatial and temporal evolution of the optical pulses is often coupled together and can be highly complex.

Lots of excellent works have been done to characterize ultrafast optical pulses near the focal point of a high numerical objective. A technique called SEA TADPOLE based on spectral interferometry method was reported [80, 81]. Authors used an optical fiber to scan the ultrafast optical field at different locations near the focal point of a lens. Spatially resolved pulse profile can then be obtained by interfering the sampled field with the field of a reference pulse. It has been shown that this method can achieve submicron spatial resolution by using NSOM fiber probes. Earlier efforts to characterize ultrashort pulses near the foci also include interferometric autocorrelation [82, 83] and collinear second harmonic FROG [84] based methods implemented by directly placing a thin layer of two-photon fluorescent or $\chi^{(2)}$ nonlinear medium in the focal plane of an objective. Recently, a nano-FROG technique was developed in which individual second harmonic nanocrystals were dispersed on a substrate and used as a probe to characterize ultrashort pulse profile through
frequency resolved optical gating [37]. This approach represents significant progress toward nanoscale spatial resolution. However, the requirement of substrate attachment may limit the applicability of the probe to characterizing the near fields of nanostructures possessing complex three-dimensional geometries.

We developed a new kind of nonlinear nanoprobe for nano-femto scale characterization. The proposed nanoprobe consists of a silica fiber taper, a ZnS nanowire attached to the tip of the taper, and two-photon fluorescent micro- or nano-spheres attached to the ZnS nanowire. We demonstrated proof-of-principle femtosecond pulse characterization by interferometric autocorrelation measurement through two-photon fluorescence from fabricated nonlinear nanoprobes.

The proposed nonlinear nanoprobe can be easily integrated into current NSOM and atomic force microscope technologies to characterize the spatiotemporal properties of ultrashort pulses at nano-femto scale. Furthermore, the nonlinear nanoprobes with different functional nanoparticles can be used to study the light-matter interaction at the nanoscale level, understand ultrafast dynamics of complex nanostructures, investigate nonlinear optics in nanoscale plasmonic structures, and characterize spatiotemporal evolution of ultrashort pulses in nanoscale structures and waveguides.

We fabricated two types of nanoprobes by attaching fluorescent micro- and nano-spheres (commercially available from Duke Scientific Corp.) at the tip of the ZnS nanowire. The first type (type I) consists of a single fluorescent microsphere with a 1 μm nominal diameter attached to the ZnS nanowire. The second type (type II) of nanoprobe has multiple fluorescent nanospheres (~ 100 nm in diameter) attached near the end of the ZnS nanowire.
The fabrication procedures for these two kinds of nanoprobes have been described in detail in the previous chapter. The field emission scanning electron microscope images of two nanoprobes (type I – Figure 4.3 (a); and type II – Figure 4.3 (b) and (c)) are shown in Figure 4.3 (a) and (b). Figure 4.3 (c) is a zoomed-in view of the tip of the type II nanoprobe shown in Figure 4.3 (b).

![Field Emission Scanning Electron Microscope Images](image)

Figure 4.3 (a) and (b): Field emission scanning electron microscope images of a nanoprobe attached with a single fluorescent microsphere (~ 1 μm diameter) and a nanoprobe attached with multiple fluorescent nanospheres (~ 100 nm diameter), respectively (c): zoomed-in view of the tip of the two photon nanoprobe shown in (b).

The optical properties of the nanoprobes were confirmed by the spectral and power dependence investigation. Two-photon excited fluorescence can be observed from the micro- or nano-spheres when excited by ultrashort pulses centered at around wavelength of 808 nm from a Ti:Sapphire laser (KMLabs). To verify that the observed two-photon fluorescence was from the attached fluorescent spheres, we measured two-photon fluorescence spectra.
from nano-probes and spectra from ZnS nanowires only. Within the sensitivity of our
detection system, possible background contributions from the nanowire, the optical adhesive,
and the fiber taper were not observed. Strong two-photon fluorescence signal from the
fluorescent micro- and nanospheres in the wavelength range from about 450 nm to 500 nm
was observed as shown in Figure 4.4.

Figure 4.4 (a) two-photon fluorescence spectrum of a type I nano-probe attached with
a single 1μm sphere (solid blue curve) and spectrum of ZnS nanowire only (dotted red curve),
(b) two-photon fluorescence spectrum of a type II nano-probe attached with multiple
fluorescent nanospheres (~ 100nm diameter) (solid blue curve) and spectrum of ZnS nanowire
only (dotted red curve).

Figure 4.5 the power dependence measurement of a two photon fluorescent nanoprobe
To characterize the power dependence property of the two-photon fluorescent nonlinear nanoprobes, a femtosecond laser beam was first focused by an objective lens. A type II nanoprobe with multiple nanospheres attached on the tip was mounted on a three dimensional translational stage and placed at the focal point of the focusing lens. The generated two-photon fluorescent signal was collimated by the same focusing objective in the backward direction, filtered by a filter and detected by a spectrograph with a liquid nitrogen cooled CCD camera. The power dependence of the generated two-photon fluorescence signal on the average power of the fundamental beam was measured as the input pump beam was attenuated at different levels. A measured power dependence relationship curve is shown in figure 4.5. Although the fitting equation of the curve is slightly off the expected quadratic relation of 2, it is still in acceptable range. One possible reason is that environmental perturbation during the measurement may change the focusing condition of the nanoprobe, which in turn may decrease the two-photon fluorescence yields. The spectral and power dependence investigations of two-photon nanoprobes confirm that they will retain the optical properties of the blue fluorescent micro- and nanospheres. As a preliminary demonstration, we used the two-photon nanoprobes to characterize the pulse width of femtosecond laser pulses near the focal point of an objective lens.
A photo of our experimental setup is shown in figure 4.6 and the schematic diagram of the setup is shown in figure 4.7 (a). Femtosecond beam from the Ti:Sapphire laser first passed through a long-pass filter (Chroma Technology). A chopper (not shown) was inserted into the beam path, and was used in conjunction with a lock-in amplifier to improve the detection sensitivity. The beam then entered a Michelson interferometer to produce two copies of the incoming pulses. Their relative time delay can be controlled by translating a mirror in one arm of the interferometer. The output beam from the interferometer was subsequently focused by an objective lens (60 x, numerical aperture: 0.85). A nanoprobe was placed in the focal point of the objective. Two-photon excited fluorescence signal from the nanoprobe was then gathered by the same objective lens, passed through a dichroic filter and then a combination of short-pass filters (Chroma Technology), and detected by a photomultiplier tube (HAMAMATSU H7827-011). The detected signal was further amplified with a lock-in amplifier. Interferometric autocorrelation traces can then be obtained by measuring the two-
photon fluorescence signal as the delay time is scanned. In order to reduce the scanning time and improve the stability of the measurement, the scanning mirror was translated continuously at a constant speed of 0.8 μm/s rather than by an incremental step each time [38]. The fluorescence signal was then recorded. We first used a type I nanoprobe. The measured interferometric autocorrelation curve is shown in Figure 4.7 (b) (blue curve). We also performed measurement by using a type II nanoprobe shown in Figure 4.3 (b). The measured interferometric autocorrelation trace is shown in Figure 4.7 (c) (blue curve).

![Schematic diagram of the experimental system](image)

**Figure 4.7** (a) Schematic diagram of the experimental system (b) Measured interferometric autocorrelation trace by using a type I nano-probe (blue) and theoretical curve (properly scaled and shifted) calculated by assuming a Gaussian pulse profile with a pulse width of 120fs (full width at half maximum) and a linear chirp parameter α of 1.9 (red) (c) Interferometric autocorrelation trace obtained by using a type II nano-probe (blue) and theoretical curve (properly scaled and shifted) calculated by assuming a Gaussian profile with a pulse width of 127fs (full width at half maximum) and a linear chirp parameter α of 2.5 (red)
4.3 Result interpretation and data processing

The pulse width estimation from recorded interferometric autocorrelation trace in figure 4.7 (b) and (c) was accomplished by following the data processing procedure [48] in chapter 2.

First, we take Fourier transforms of the interferometric autocorrelation trace in figure 4.7 (b). The spectral components of the trace are clearly shown in figure 4.8. The d.c. part of the components contains the pulse duration information. Then, we will filter out the middle part of the spectrum as shown in figure 4.9.
We then do the inverse Fourier transforms. If we assume a Gaussian pulse profile, the resulting full width at half maximum (FWHM) of the intensity autocorrelation is about 1.414 times of the actual pulse intensity profile. The estimated pulse width from this interferometric autocorrelation trace is about 120 fs.

The linear chirping parameter can also be estimated by assuming a Gaussian pulse profile, $E(t)=\exp[-(1+i\alpha)(t/\tau_G)^2]$. We first filtered out the $2\omega$ spectral component in figure 4.8 and did inverse Fourier transform. Proper coefficient will be given to fit the resulting curve and a chirp parameter $\alpha$ around 1.9 is estimated. The theoretical curve (properly scaled and shifted) obtained by using these parameters is also shown for comparison in figure 4.7 (b) (red curve). It should be noted that the baseline of the interferometric autocorrelation curve is not even on both sides. This is likely due to slight instability of the probe during the experiment, which may cause the background to change during the course of the measurement. Compared with the thin ZnS nanowire, the 1 μm fluorescent sphere is rather large and is likely more susceptible to environmental perturbation. This problem can be alleviated in the type II nanoprobe. Compared to the response of the type I nanoprobe, the
signal strength is reduced. By following the same data retrieval procedures for interferometric autocorrelation trace in figure 4.7 (c) (blue curve), the retrieved pulse width is about 127 fs (full width at half maximum) and a chirp parameter $\alpha \sim 2.5$ is estimated.

We notice that there is slight difference between the two retrieved pulse widths and chirping parameters. This is likely due to the noise in the measurement as well as to slight changes in the laser cavity between the two experiments. The chirping is primarily caused by the dispersive propagation of the laser pulses in a relatively thick cube beam splitter used in the interferometer in this preliminary study.

In conclusion, we have proposed and fabricated a new type of nonlinear nanoprobe for characterization of ultrashort optical pulses. Proof-of-principle Interferometric autocorrelation measurements near the focal point of an objective through two-photon fluorescence from such nonlinear nanoprobes were demonstrated. We believe that our technique can be further improved to incorporate other types of nonlinear nanoparticles and led to full nano-femto spatiotemporal characterization of complex ultrafast optical near fields, which can find important applications in ultrafast nanophotonics.
Chapter 5. Ultrashort optical pulse characterization using second-order nonlinear nanoprobes based on FROG technique

5.1 Introduction

We have developed capability for characterizing ultrafast optical pulses based on interferometric autocorrelation measurements. By replacing the two-photon fluorescent nanoparticles with second harmonic nanocrystals on the tip of optical nanoprobes, more advanced pulse characterization technique, FROG, can be applied and detailed pulse profile information, including both amplitude and phase, can be retrieved. The developed second harmonic nanoprobes can be used to fill in the important knowledge gap in the field of nano-femto optics, i.e., how to non-perturbingly measure the spatiotemporal evolution of an ultrafast optical near field in nano-femto scale.

Lots works [37, 80, 81] have been done to fully characterize the femtosecond laser pulses in the vicinity of the focal point of a lens. However, the existing methods, when extended to probing ultrafast optical near field in complex three-dimensional (3D) micro- or nano-structures (such as photonic crystal cavities or metamaterials), either require the conversion of local field into propagating waveguide modes (which, as a result, can introduce perturbation) or have limited accessibility due to limited aspect ratio.
In this chapter we demonstrated a Second HARmonic nano-Probe (SHARP). It consists of a silica fiber taper to provide an interface for macroscopic handling and positional control, a single carbon nanotube attached to the taper in order to achieve large aspect ratio, and nanocrystal(s) possessing second order nonlinearity attached to the nanotube. The nonlinearity of the second harmonic nanocrystal(s) is exploited for femtoscale temporal characterization while nanoscale spatial resolution can be achieved due to the nanoscale dimensions of the nanoprobe. Since SHARP has a large aspect ratio and is built from nanocrystals attached to a nanowire, optical scattering can be minimized and measurements with minimal perturbation can be performed in complex three-dimensional structures.

**5.2 Pulse characterization near the focal point of an objective with SHARP**

The fabrication of the second harmonic nanoprobes follows the procedures described in previous chapters. Briefly, a fiber taper with a typical tip diameter of about 1-2 μm was first fabricated from a silica fiber by using a fiber fusion splicer. A single multiwall carbon nanotube was then attached to the taper using UV curable optical adhesive. A single or multiple second-harmonic Barium Titanate [85] (tetragonal phase) nanocrystals were attached to the tip of the carbon nanotube. Owing to the small size of nanocrystals, phase
matching condition is relaxed and therefore our SHARP is suitable for characterizing ultrashort pulses with a large bandwidth.

We fabricated SHARPs with various number of nanocrystals attached on the tip of carbon nanotube by controlling the concentration of nanocrystals suspended in water. The number of the nanocrystals on the SHARP ranges from several tens to a few. The spatial resolution of SHARP is limited by the size of the active nonlinear component. In order to accomplish true nanoscale resolution, SHARP with a single nanocrystal is required. As aforementioned, we developed a systematic approach to retain only a single nanocrystal and to precisely define its position relative to the nanotube by exploiting FIB nano-milling as an additional nanofabrication step.

5.2.1 Pulse characterization near the focal point of an objective with SHARP with multiple nanocrystals

We carried out the ultrafast optical pulses characterization experiments with different SHARPs. We first measured the femtosecond laser pulses near the focal point of a high numerical aperture objective by using the SHARP with multiple nanocrystals attached on the tip. The microscope and FESEM images of the probe are shown in figure 5.1 (a) and (b).
The schematic diagram for the experimental setup is shown in figure 5.2. The femtosecond laser beam first entered a Michelson interferometer, which was formed by an ultrafast beam-splitter and a pair of mirrors. The relative time delay between the resulted two copies of the incoming pulse can be controlled by translating the mirror in one of the arms of the interferometer. The output beams from the interferometer collinearly propagated and were subsequently focused by a high numerical objective lens (60 x, numerical aperture: 0.85). The SHARP as shown in figure 5.1 was mounted on a three dimensional stage and translated to the location of focal point of the objective. The generated second harmonic signal from the probe was then collected and collimated in the forward direction by a long working distance objective (Mitutoyo 50X M Plan Apo, numerical aperture: 0.55). After passing through some appropriate filters, the second harmonic signal from BaTiO$_3$ was detected by a spectrograph with a liquid nitrogen cooled CCD camera (Princeton Acton, SP2500i). The FROG trace was obtained as the delay was scanned at step interval of 200nm or time delay of about 0.67fs.
The recorded second harmonic collinear frequency resolved optical gating (SHG-CFROG) trace is a two dimensional spectrogram of the pulse as shown in figure 5.3 (a). The corresponding interferometric autocorrelation trace is a simple and good preliminary validation for the spectrogram. By adding up all the spectral values and substracting the background noise along each delay step, the interferometric autocorrelation trace is shown in figure 5.3 (b). The peak to background ratio is about 6.7 to 1. In theroy, the ratio is 8 to 1. Considering the possible perturbation and experimental alignment, this trace is good enough for pulse retrieval.
The Collinear-FROG pulse retrieval algorithm coding work was done by Dr. Zhe Zhang, who was a visiting scholar in our lab, and the pulse retrieval results from SHG-CFROG traces were based on his codes.

As we discussed in chapter 2, the obtained collinear FROG trace (SHG-CFROG) contains the information of off-axis SHG-FROG trace [37, 38, 49, 84, 86]. We first do the Fourier transforms to the spectrogram in figure 5.3 (a). Then, the d.c. term on frequency domain will be well separated from other higher frequency components. After applying a low pass band filter and subtracting the background, the remain part will be filtered out as shown in figure 5.4.
It contains the exact same information as SHG-FROG (non-collinear second harmonic generation). After we have the SHG-FROG, the phase information of the pulses can be achieved by employing two-dimensional phase retrieval algorithm [36] as discussed in chapter 2. Starting with an initial guess for the field E(t), including amplitude and phase, and constructing the calculated FROG trace by squaring the magnitude of E(\omega,\tau), we will do the iteration until the FROG error G is converged as shown in figure 5.5.
The retrieved pulse intensity and phase profiles of the femtosecond laser pulses near the focal point of the objective are shown in figure 5.6 (a). The retrieved pulse width (full width at half maximum) is about 59 fs. The phase profile is nearly flat over the pulses. This indicates that there are no significant chirping effects on the pulses. In terms of this no chirping effect, the retrieved phase profile is consistent with the experimental observation from interferometric autocorrelation trace in figure 5.3 (b), where the upper and lower envelopes of the interference pattern split evenly from the background level. In order to further check the reliability of our retrieved results, we measured the laser spectrum and compared it with the calculated power spectrum of the retrieved pulses (i.e., the square of the absolute value of the Fourier transform of the retrieved pulses). The measured laser spectrum and the calculated power spectrum matched each other very well as shown in figure 5.6 (b). It is a good indication for high-fidelity pulse retrieval.
5.2.2 Pulse characterization near the focal point of an objective with SHARP with a few nanocrystals
Based on previously successful femtosecond laser pulses characterization by using SHARP with multiple BaTiO$_3$ nanocrystals, we further improved the spatial resolution of the system by employing a SHARP with only a few nanocrystals on the tip. The microscope and FESEM images of the fabricated SHARP with several nanoparticles are shown in figure 5.7.

![Microscope and FESEM images of the SHARP](image)

**Figure 5.7** (a): microscope picture of the SHARP with only a few nanocrystals on the tip. (b): FESEM image of the SHARP. (c): Zoomed-in view of the SHARP shown in (b).

By following the same experimental procedures and setup as shown in figure 5.2, the obtained SHG-CFROG trace and the interferometric autocorrelation trace are shown in figure 5.8.
The retrieved pulse intensity and phase profiles of the femtosecond laser pulses near the focal point of the objective by using the SHARP in figure 5.7 are shown in figure 5.9 (a). The retrieved pulse width (full width at half maximum) is about 56 fs. Considering the possible measurement errors and slightly changes of the condition of laser cavity between two experiments, the pulse duration result is in excellent agreement with our previous measurement. There is also no chirping effect observed over the pulse region in phase
retrieval results. The measured laser spectrum and the calculated power spectrum of the retrieved pulses are also consistent with each other as shown in figure 5.9 (b).

Figure 5.9 (a) retrieved temporal profile of the pulse (solid line: temporal profile, dotted line: phase) and (b) pulse spectrum (solid line: retrieved pulse spectrum, dots: measured spectrum)
5.2.3 Pulse characterization near the focal point of an objective with SHARP with a single nanocrystal

The spatial resolution of the SHARP is mainly limited by the size and distribution of the nanocrystals on the tip of the carbon nanotube. From previously study, the fabricated SHARPs are powerful and reliable tools to characterize the ultrafast optical pulses near the focal point of an objective with high spatial resolution. In order to precisely control the size and position of the nonlinear nanocrystal(s) on the carbon nanotube, an additional fine-fabrication procedure by using FIB nano-milling were utilized to remove the excess nanocrystals and trim the carbon nanotube. Figure 5.10 shows two examples of SHARPs before and after the FIB nano-milling.

![Figure 5.10 Fabrication of SHARP by utilizing focused ion beam nanomilling. Two SHARP’s before (a, c) and after (b, d) FIB nanomilling are shown respectively. (a) and (b): a small section of the carbon nanotube and a protruding nanocrystal were selectively removed by](image)
FIB; (c) and (d): a small part of the carbon nanotube was removed by FIB to result in a single nanocrystal located at the tip of the nanotube.

Fig. 5.10 (a) and (b) show that a small section of the carbon nanotube and a protruding nanocrystal were selectively removed by FIB while in Fig. 5.10 (c) and (d) a small part of the carbon nanotube was removed by FIB to result in a single nanocrystal located at the tip of the nanotube.

We used the SHARP shown in Fig. 5.10 (d) to probe the pulse profile near the focal point of an objective as illustrated in Fig. 5.11. The SHARP has only one single BaTiO$_3$ nanocrystal located on the tip of carbon naotube. So, we can now achieve sub-wavelength spatial resolution in this characterization. The experimental setup is the same as shown in figure 5.2. The pump power of each beam measured before the objective was about 0.2 mW. An integration time of 5 s was used in our experiment. During the experiment, the center wavelength of the laser was tuned at around 775 nm.

![Figure 5.11 an illustration picture of the pulse characterization near the focal point of an objective](image-url)

The recorded spectrogram is shown in figure 5.12. The second harmonic signal is centered at 388 nm, which is half the wavelength of our fundamental beam.
The retrieved pulse intensity and phase profiles are shown in Fig. 5.13 (a), which are obtained by using the same retrieval algorithm discussed earlier. A pulse width (FWHM) of 62 fs was obtained. This pulse duration measurement is in well consistent with our previous characterization (59fs and 56fs). The pulse is slightly chirped. The experimental setup layout and the focusing lens used in the experiments are all the same in these three measurements. So, consistent results were achieved as expected. Considering the possible slight changes of laser cavity, the three groups of pulse characterization results are in good agreement to each other.
Figure 5.13 (a) retrieved temporal profile of the pulse (solid line: temporal profile, dotted line: phase) and (b) pulse spectrum (solid line: retrieved pulse spectrum, dots: measured laser spectrum)

By now, we have demonstrated the ultrafast optical pulse characterization capability near the focal point of a high numerical objective with sub-wavelength spatial resolution by using our developed SHARP. The spatial resolution of SHARP is limited by the size of the nanocrystal on the tip. By employing auxiliary FIB nanomilling technique, we fabricated SHARP containing only a single active nonlinear nanocrystal.

The unique fusion of nanoscale and nonlinear active features in this device not only gives rise to its capability to characterize both the amplitude and phase profiles of ultrashort optical pulses, but also provides the ability of probing ultrafast optical field in complex 3D micro- and nano-structures, such as in the hole region of hollow-core photonic crystal fiber. We will continue to investigating the application of nonlinear nanoprobes in this field.

5.3 Pulse characterization in the core region of a photonic crystal fiber with SHARP

Another unique feature of SHARP is that it is capable of probing ultrashort optical field in complex micro- or nano-structures due to its high aspect ratio. In this section, we will
demonstrate in-situ ultrashort pulse characterization directly in the air core region of a hollow-core photonic crystal fiber.

The schematic diagram for the experimental setup is shown in figure 5.14. It is very similar to our previous measurement setup. A femtosecond laser beam entered a Michelson interferometer to produce two identical copies of the incoming ultrafast optical pulses and their relative time delay was controlled by translating a mirror in one of the arm. The two collinearly propagating beams at the output of the interferometer were then coupled into a section of hollow core photonic crystal fiber through a 20x objective on one end. A SHARP was mounted on a three dimensional stage and translated into the air core region of the hollow core PCF on the other end. The relative position between the tip of the SHARP and the PCF can be precisely controlled by monitoring two imaging systems for side and cross section views. Some typical snapshots for these views are shown in figure 5.15. The air-core diameter of the hollow core PCF is around 6 μm and a zero dispersion wavelength is at 753 nm. The femtosecond laser was pre-chipped using a prism pair after the output of the laser system and the center wavelength was tuned to 754 nm in order to match the minimum dispersion region of the PCF. The generated second harmonic signal from the probe was then collected and collimated in the forward direction by a long working distance objective (Mitutoyo 50X M Plan Apo, numerical aperture: 0.55) and detected by a spectrograph with a liquid nitrogen cooled CCD camera (Princeton Acton, SP2500i). The FROG trace can be obtained as the delay was scanned.
Figure 5.14 diagram for the experimental setup

Figure 5.15 cross section and side views of inserting SHARP into the core region of the PCF under microscope system. (a) and (c): cross section and side view of SHARP before inserting into PCF. (b) and (d): cross section and side view of SHARP after inserting into PCF.
To characterize the ultrafast optical pulses in the core region of the PCF, a SHARP probe was inserted into the hollow core region of the PCF. Typical FESEM pictures of the probe are shown in figure 5.16.

![Figure 5.16 Field emission scanning electron microscope image of a SHARP; (b) Close up view of the attached BaTiO$_3$ nanocrystals](image)

In order to ensure the signal strength level of second harmonic signal and retain reasonable characterization spatial resolution in the air-core region of PCF, the sizes of nanocrystal cluster are in the order of 1-2 μm in diameter in the experiments. Then, the SHARP will be able to be inserted into PCF as shown in figure 5.15. Two snapshots pictures in figure 5.15 (a) and (b) demonstrate the cross section images of hollow core PCF. As the SHARP approaching into the PCF, the scattered fundamental beam from nanocrystals appeared as a bright spot on the imaging camera. When the probe was far away from the air core of PCF, the SHARP was out of focus and cannot be observed in figure 5.15 (a). Two side view pictures were shown in figure 5.15 (c) and (d). It is clearly shown in figure 5.15 (b) that the SHARP was successfully inserted into the PCF. By combining and monitoring these two views, the position of the probe was precisely controlled and tracked in the measurements. An illustration picture for probing the pulse profiles in the core region of PCF is shown in figure 5.17.
During our experiment, a delay incremental step of 0.67 fs was used. The pump power of each beam measured before the coupling lens (placed in front of the fiber) was around 3 mW. The integration time of the spectragraph was 600 ms. The total elapse time for acquiring a FROG spectrogram was about 15 minutes.

The side-view micrographs of a SHARP probe are shown in figure 5.18. After it was inserted into the air core of the PCF as shown in figure 5.18 (b), the collinear SHG FROG
spectrogram was recorded as the delay of two fundamental beams is scanned. The obtained collinear FROG trace is shown in figure 5.19.

Figure 5.19 the collinear SHG-FROG trace obtained in the core region of PCF

Both the amplitude and the phase profile of the ultrashort pulses at the air core of PCF can be retrieved from the trace. By following the same pulse retrieval procedure as we employed before, pulse intensity and phase profiles are shown in figure 5.20 (a). The retrieved pulse width (full width at half maximum) is about 150 fs. The pulse is chirped as indicated by a nearly parabolic phase profile. The measured laser spectrum and the calculated power spectrum of the retrieved pulses are consistent with each other as shown in figure 5.20 (b).

Our results have shown clearly that SHARP has the unique capability to probe the ultrafast optical near field in complex three-dimensional micro- and nano-structures. Furthermore, by employing auxiliary FIB nanomilling technique, it is possible to characterize the nano-femto
scale spatiotemporal evolution of ultrafast optical field in complex 3D micro- and nanostructures with fabricated SHARP containing only a single active nonlinear nanocrystal.

Figure 5.20 (a) retrieved temporal profile of the pulse (solid line: temporal profile, dotted line: phase) and (b) pulse spectrum (solid line: retrieved pulse spectrum, dots: measured spectrum)

We believe that the development of SHARPs will be a step forward for fulfilling the goal of having ultimate control of photons and developing capability to engineer arbitrary ultrafast optical near fields.
5.4 Phase transition temperature measurement of BaTiO$_3$ nanoparticles

To ensure accurate measurement of ultrashort pulses, a key requirement is that SHARP must effectively retain the nonlinear property of the second harmonic nanocrystals. Figure 5.21 (a) and (b) show two CFROG spectrograms measured at the focal point of an objective by using a SHARP comprising a multiwall carbon nanotube and BaTiO$_3$ nanocrystals. At low pump power (Figure 5.21 a) a typical spectrogram was obtained, which, as expected, shows increased signal level in the central part where the two copies of pulses overlapped in time. However, at relatively high pump power (Figure 5.21 b) an abnormal spectrogram was observed which surprisingly shows suppressed second harmonic signal in the central part of the spectrogram. This anomalous phenomenon can also be clearly seen in the interferometric autocorrelation traces, which can be obtained by integrating the spectral intensity. Figure 5.21 (c) shows the autocorrelation trace at low pump power that has a peak-to-background-ratio of about 6.7:1 (theoretical ratio: 8:1) whiles the trace at high power shows significant distortion. Obviously, large error can be resulted if the abnormal CFROG spectrogram is used for pulse characterization.
Figure 5.21 Collinear frequency-resolved optical gating spectrograms and interferometric autocorrelation traces obtained at the focal point of an objective. (a) and (c): measured at low pump power; (b) and (d): measured at relatively high pump power.

To better understand the origin of the abnormal spectrogram, in the following we examine SHARPs fabricated from two types of nanowires (multiwall carbon nanotube (CNT), Nanolab, PD100L520, and Zinc Sulfide (ZnS)) and two types of micro- or nano-crystal clusters (Barium Titanate (BaTiO$_3$), NanoAmor Inc. APS: 200nm, and Lithium Niobate (LiNbO$_3$) [87]). A total of four different combinations, i.e., CNT-BaTiO$_3$, CNT-LiNbO$_3$, ZnS-BaTiO$_3$, and ZnS-LiNbO$_3$ are studied. Since our goal is to perform optical characterization of the fabricated nonlinear probes to confirm whether the functionalized probe retains the same optical nonlinear response of individual nanocrystal, for simplicity and to ensure enough signal, only nonlinear probes attached with micro- or nano-crystal clusters were used in the experiments. The auxiliary FIB fine-fabrication was not employed. Figure 5.22 shows the typical field emission scanning electron microscope images of the four different kinds of nonlinear probes under this study.
To characterize these nonlinear probes, a femtosecond laser beam was first focused by an objective lens. A second harmonic nonlinear optical probe, which was mounted on a three dimensional translational stage, was then placed at the focal point of the focusing lens. The generated second harmonic signal was collimated by a long working distance objective (Mitutoyo 50X M Plan Apo, numerical aperture: 0.55) in the forward direction, filtered by a band-pass filter (D400/70, Chroma Technology), and detected by a spectragraph with a liquid nitrogen cooled CCD camera (Princeton Acton, SP2500i). The power dependence of the generated second harmonic signal on the average power of the fundamental beam is measured as the input pump beam was attenuated to different levels. The schematic diagram of the experimental setup is shown in figure 5.23.
The measured power dependence relation for the different types of probes is plotted in figure 5.24 in logarithmic scale. Ideally, a quadratic relationship is expected, which corresponds to a slope of 2 in the logarithmic plots. The three types of probes, CNT-LiNbO$_3$, ZnS-BaTiO$_3$, ZnS-LiNbO$_3$ all exhibit quadratic power dependence under our experimental conditions. However, the CNT-BaTiO$_3$ probe shows quadratic dependence only under low-power regime. As the power of the fundamental beam continues to increase, the measured second harmonic power starts to decrease and then maintains at a very low signal level, which significantly deviates from the expected quadratic relation. As a comparison, the power dependence for BaTiO$_3$ nanocrystals directly dispersed on a glass slide was also shown, which does not exhibit similar behavior. This phenomenon was not observed in the other three kinds of probes.
We believe that this anomalous second harmonic power dependence was likely caused due to the local heating of the attached BaTiO$_3$ nanocrystals by CNT probe, where the tightly focused femtosecond laser beam can significantly increase the local temperature of the multiwall CNT. As the laser power was increased, the temperature of the BaTiO$_3$ nanocrystals rose from room temperature to a critical value, at which the crystal structure of some BaTiO$_3$ nanocrystals underwent a transition from the tetragonal to the cubic phase. As a result, second harmonic generation vanishes (under dipole approximation) due to the occurrence of inversion symmetry [88]. This phase transition behavior is likely responsible for the observed deviation of the second harmonic signal from a quadratic relationship in the CNT-BaTiO$_3$ nonlinear probe, and hence the anomalous CFROG spectrogram. Since ZnS has much less absorption at the laser wavelength than CNT, thermal effect was not apparent in the ZnS-BaTiO$_3$ nonlinear probe and a normal quadratic relationship was observed under
our experimental conditions. Similarly, BaTiO$_3$ nanocrystals directly on a glass substrate also manifest a normal quadratic power relationship. Note that compared with BaTiO$_3$ (tetragonal to cubic phase transition temperature: ~ 130 °C), LiNbO$_3$ (bulk Curie temperature: ~ 1142 °C) has a much higher phase transition temperature [89-91], which explains the normal power dependence observed in the probes comprising CNT and LiNbO$_3$ micro-crystals.

In order to further investigate and verify the phase transition phenomena, we designed another controlled experiment. A picture of experimental setup is shown in figure 5.25.

![Figure 5.25 A picture of experimental setup for phase transition temperature measurement](image)

A femtosecond laser beam was focused onto a BaTiO$_3$ nanocrystal cluster (~10 µm in diameter) located at the backside of a glass slide (~0.15 mm thickness) through a focusing lens. A thermistor (YSI 55004) was co-located on the backside of the glass slide within a few millimeters of the nanocrystal cluster. In order to ensure good thermal transfer from the
glass slide to the resistor, a layer of heat-sink compound (RadioShack 276-1372 A) was applied between the glass slide and the thermistor. A solder iron was placed in extremely close proximity to the front side of glass slide (without surface contact) and served as a localized heater for BaTiO$_3$ nanocrystal cluster. A close-up view of the setup is shown in figure 5.26.

![Figure 5.26 A close-up view of the experimental setup](image)

As the temperature of the solder iron increased, the generated second harmonic signal from BaTiO$_3$ nanocrystal cluster on the backside of the glass slide was detected by a spectragraph with a liquid-nitrogen-cooled CCD camera. Afterwards, the thermistor was translated to the previous location of the nanocrystal cluster. This was done to obtain a good temperature measurement of the location where the nanocrystal cluster previously resided. The experimental results are shown in the figure 5.27.
The relationship between the solder iron temperature and the intensity of second harmonic signal from BaTiO$_3$ nanocrystals was first measured. Afterwards, we translated the thermistor to the place where BaTiO$_3$ nanocrystal cluster originally resided. The relationship between the voltage of thermistor and the iron temperature was given in figure 5.27 (a). Provided by the manufacture of the thermistor, the product data sheet was used to calibrate the thermistor voltage and temperature, as shown in figure 5.27 (b).
The experimental results are shown in the Fig. 5.28. It demonstrates that when the temperature was below 90 °C, the generated second harmonic signal remained at a relatively stable level. The signal fluctuation was caused by environmental perturbations such as small movement of the glass slide during the measurement. As the temperature increased to ~110 °C, the signal dropped significantly. The decrease of the signal indicates that the phase transition of the BaTiO$_3$ from tetragonal to cubic occurred as previously discussed. When the temperature increased to ~120 °C, the transition process was complete and there was almost no observable second harmonic signal generated from the cubic nanocrystal structure. The second harmonic signal power was restored to the original level when the temperature was lowered back to room temperature. In our experiment, the measured phase transition temperature differs from previously published reports [91]. One possible reason is that the heat transfer from the cover glass slide to the thermistor is not efficient enough. Nevertheless, this controlled experiment clearly suggests that the phase transition behavior of the BaTiO$_3$ nanocrystal is responsible for the anomalous relationship of the second harmonic signal observed in the CNT-BaTiO$_3$ nonlinear probe.
We have developed and fabricated different types of nonlinear optical probes. The optical spectral and power dependence characterization shows that functionalized nonlinear probe retains the nonlinear properties of the second harmonic nanocrystals. However, we also observed anomalous power dependence relationship in the nonlinear probe comprising CNT and BaTiO$_3$, which is likely caused by the phase transition of BaTiO$_3$ nanocrystal from the tetragonal to the cubic phase. At relatively high pump power level, this type of nonlinear probe should be avoided. However, similar behavior was not observed in the other three types of probes under our experimental conditions. The nonlinear probes discussed here can be used to characterize ultrashort optical pulses through frequency resolved optical gating by exploiting the nonlinear interaction between the probe and the field. It can potentially allow for optical near field characterization in complex three-dimensional micro- or nanostructures with nano-femto spatiotemporal resolution.

5.5 Summary

In this chapter, we fabricated several types of nonlinear optical nanoprobes to investigate various ultrafast optical near field. In order to have detailed ultrafast optical pulses information, more advanced ultrashort pulse characterization methods have been performed by using SHARPs. By employing auxiliary focused ion beam (FIB) nanomilling technique, we also fabricated a nanoprobe containing only a single active nonlinear nanocrystal attached on the end of carbon nanotube which can in turn push the spatial resolution even higher. Exciting in situ characterization measurement results directly in the air core of a photonic crystal fiber and near the focal point of an objective with high spatial resolution.
have been demonstrated.

The experimental results indicated that the developed nanoprobes can open a new way for probing the evolution of ultrafast optical fields in complex 3D micro- or nanostructures and the presented research works will be helpful in understanding the fundamental problems and find its applications in the fields of nonlinear optical microscopy and nanophotonics.
Chapter 6. Future work and conclusions

6.1 Preliminary ultrashort pulse characterization in turbid medium

Imaging through turbid medium [92-96] is important for the applications in the field of biology and medical research. Focusing the laser beam through the target turbid medium is a general requirement in many imaging applications. However, due to the inhomogeneous properties of the biological tissue, the laser beam passing through the medium will experience seriously scattering (low absorption medium) and the pulse will be broadened and distorted. It is necessary and helpful to characterize the optical fields in different locations of turbid medium. The measurement result will provide insight of optical pulse profiles and serve as a feedback signal to achieve desirable pulse profile at the location of focusing in the medium.

By taking advantage of the proven ultrafast optical pulses characterization capability and flexibility of probing the field in complex structures or liquid medium, we have started to investigate the possibility of utilizing SHARP to characterize the femtosecond laser pulses in turbid medium. In our experiments, diluted milk was used as a scattering medium.
Figure 6.1 diagram of experimental setup for the pulse characterization in turbid medium

As a preliminary exploration of SHARP in turbid medium, the experimental setup is shown in figure 6.1. The photo of a part of setup is shown in figure 6.2.

Figure 6.2 a photo for the experimental setup
A femtosecond laser beam was focused into the turbid medium (diluted milk) and a SHARP will be translated to the focal point of the focusing lens in the medium. A microscope picture of the SHARP is shown in figure 6.3.

![Microscope Image of SHARP](image)

**Figure 6.3 a microscope image of the SHARP**

As aforementioned, when the time delay between two arms of interferometer was scanned, a collinear SHG-FROG trace can be obtained. The optical density of the scattering medium is given by:

\[
OD = \log_{10}(I/I_0)
\]

Where, \(I_0\) is the input laser power and I is the measured laser power after passing through the scattering medium.

So, the optical density of the medium can be increased by adding un-diluted milk into diluted milk solution in the cell container again and again. So, different spectrograms can be recorded at various optical densities of the diluted milk in experiments. A typical spectrogram for optical density of 0.77 (total transmission of 17%) is shown in figure 6.4 (a). The retrieved femtosecond pulse profile and the spectrum from the trace are shown in figure 6.4 (b) and (c).
The retrieved pulse width (full width at half maximum) is about 132 fs. We characterized the femtosecond laser pulses under different optical density (optical transmission) conditions of the diluted milk. The relationship between the optical density and retrieved pulse duration is shown in figure 6.5.
The curve shows that when the optical density of the diluted milk increased from 0.08 to 0.43, the pulse duration also broadened from ~80fs to ~130fs. However, as the optical density continued increasing, the optical pulse width will fluctuate from ~120fs to ~150fs. Because of the weak second harmonic signal, the data for the turbid medium with optical density larger than 1 is not available in our experiments. We did not observe significant broadening of the femtosecond laser pulse width over the range of optical density studied in our experiments.

One possible explanation of the phenomena [97] is that, after passing through the turbid medium, the optical pulse profile consists of three parts of photon. One is Ballistic photons, which experienced no scattering at all. The second is snake photon, which experienced part of scattering from the particles in the medium. The last is diffused photon, which experienced lots of scattering from the medium. Due to the scattering from the medium (diluted milk), the polarization of the diffused photon will change randomly and the pulse
phase will also be different shot by shot as it passes through the medium. However, in our characterizations, a scalar optical field was assumed and we also assumed the optical pulse will maintain relatively stable phase for different shots. The optical pulses broadening effect in turbid medium was caused mainly by the presence of diffused photon. So, under the assumption of our current pulse characterization technique, we can only mainly measure the coherent part of the pulse, the Ballistic photons. The diffused photons cannot be characterized at this time.

6.2 Conclusions and future work

In the dissertation, we developed two types of nonlinear optical nanoprobes, two-photon fluorescent nanoprobes and second harmonic nanoprobes, to investigate spatiotemporal characteristics of the ultrafast optical near fields.

The nanoprobes consist mainly of three parts, a silica fiber taper with one to a few micrometers in diameter on the tip, a single nanowire bonded to the end of the fiber taper and functional nonlinear nanoparticles attached to the tip of the nanowire. The optical fiber taper can be easily mounted on a translation stage and served as an interface between experimental handling and optical near field probing in complex nano-scale structures. The single nanowire is a spatial extension of the fiber taper. The ready availability and excellent inherent mechanical and optical properties of different nanowires make them ideal carriers for the nanoparticles as well as indispensable bridges for uniting the probe. The nonlinear optical characteristics of the probe come from the nanocrystals on the tip. The nonlinearity of the nanoparticle enables temporal characterization through autocorrelation or frequency resolved optical gating measurements while the nanoscale spatial resolution can be
potentially achieved through near field scanning of the nonlinear nanoparticle. Because of the relaxed phase matching condition, the nanocrystals are also ideal materials for broadband optical source characterizations. The spatial resolution of the probe is mainly determined by the size and distribution of the nanoparticles on nanowire. By employing FIB nanomilling technique, we fabricated nanoprobes containing only a single nanocrystal on the end of nanowire.

In preliminary development of the technique, interferometric autocorrelation measurements near the focal point of a high numerical aperture objective through two-photon fluorescence from nanoprobes were demonstrated. The estimated femtosecond pulse width and linear chirp parameters were obtained.

Detailed pulse characterizations of ultrashort optical pulses have been demonstrated by using SHARPs. Exciting in situ characterization measurement results directly in the air core of a photonic crystal fiber and near the focal point of an objective with high spatial resolution have been obtained which indicate that the proposed nanoprobe can open a new way for probing the evolution of ultrafast optical fields in complex 3D micro- or nanostructures.

Furthermore, by using the unique capabilities of the proposed nanoprobes, we have also started to investigate the application for characterizing the ultrashort optical pulses in scattering medium (diluted milk).

Our research works have presented the capability of pulse characterization of ultrafast optical near fields in complex micro/nano-structures. The demonstration of such ability is crucial for understanding the interaction of ultrafast optical fields and nanoscale systems.

In the future studies, we will develop the nano-scale scanning capability of the technique and integrated with our current pulse characterization ability of nonlinear nanoprobes to achieve the nano-femto spatiotemporal characterization of ultrafast optical near field.
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