

Characterization of Ultrashort Pulse Laser by Using KNbO_3 Nanoneedles Based Frequency-resolved Optical Gating (FROG)

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Abstract— We demonstrate a measurement of Second Harmonic Generation (SHG) FROG using potassium niobate (KN) nanoneedle clusters, instead of bulk crystal, as nonlinear medium. Using this excellent nonlinear nano-material, we characterize the pulse time/frequency-dependent intensity of a 1040 nm picoseconds pulse laser. Due to the nanometer scale of this nonlinear medium, the phase matching constraints can be relaxed, and the measurement system is significantly simplified and thus more accessible for common users.

1. INTRODUCTION

Ultrafast pulse laser have been widely used in many areas because of their excellent properties, especially in nonlinear excitation. There are many methods to measure both the amplitude and the phase structure of an ultrafast pulse [1–3]. FROG is one of the most common approaches for this kind of characterization. With a piece of nonlinear crystal, a FROG system based on different nonlinear effects can be implemented, such as SHG-FROG and third-harmonic-generation (THG) FROG. However, being an essential element in FROG technique, the bulk nonlinear crystal always needs specially cutting for different wavelength band before usage. Moreover, when the pulse laser alters slightly, complicated modulations have to be operated to meet the altered phase matching condition.

Many efforts have been made to simplify the complexity of the conventional FROG system, and maintain the performance as well [3, 4]. Some research has reported using single nanoparticle instead of bulk crystal as nonlinear medium [5, 6], which avoid the complexity the of using bulk crystal and achieve a better spatial resolution than ever. However, since the signal generated from the single nanoparticle is weak, the acquisition time has to be extremely long. It will badly affect the measurement efficiency and signal to noise ratio (SNR) apparently.

In this work, we propose KN nanoneedle clusters based FROG approach to overcome the drawback of the low SNR in the previous literatures, without renouncing the performance of the FROG technique. By employing the nanoneedle clusters in a conventional noncollinear FROG system, the optical properties of a 1040 nm picoseconds pulse laser are characterized in both temporal and spectral domain.

2. METHODS

2.1. Experimental Set-up

As illustrated in Figure 1, a 2.5 ps Yb doped large-mode-area (LMA) photonic crystal fiber (PCF) laser is employed, with center wavelength at 1040 nm, ~ 50 nm width, and repetition rate of ~ 45 MHz. The output of the laser is split at beam splitter (BS) into the two arms of a homemade interferometer. A delay stage (DS) with 10 μm /step resolution, corresponding to 33.3 fs in time domain, is used to induce a difference between the optical paths. After being reflected by two retro-reflector (RR1 and RR2) respectively, the two beam are recombined at BS but with a separation. They are then non-collinearly focused on to the KN nanoneedles by a lens ($L1$). The signal is collected by another lens ($L2$) and detected by a spectrometer (Ocean optics, QE65000). No optical filter is required in this non-collinear system.

2.2. KN Nanoneedle

The KN nanoneedles used in this work are synthesized by a hydrothermal method [7]. The length of single KN nanoneedle varies around 1.6 μm , and the diameter decreases uniformly from 60 nm for bottom to 20 nm for top approximately [8]. The TEM image of individual KN nanoneedle and SEM images of nanoneedle clusters are provided in Figure 2. The clusters used for SHG are obtained by dropping suspension of nanoneedles in deionized water onto a coverslip and dried in the air. The

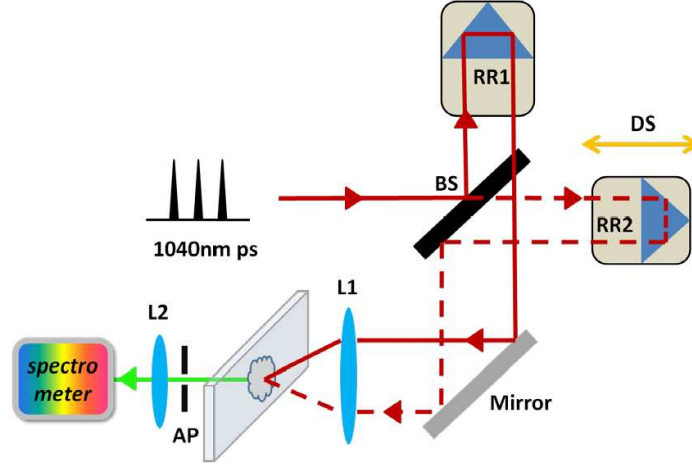


Figure 1: Schematic diagram of pump-probe system in the FROG measurement. BS: beam splitter; RR1 and RR2: retro-reflectors; DS: delay stage; L1 and L2: lens; AP: aperture.

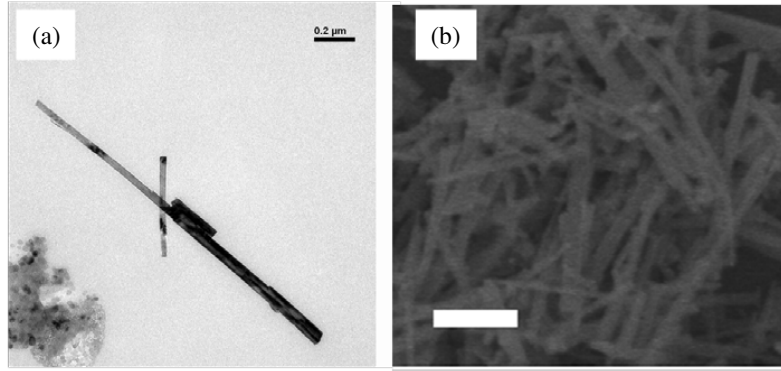


Figure 2: (a) TEM image of single KN nanoneedle. Scale bar: 200 nm. (b) SEM image of KN nanoneedle clusters. Scale bar: 500 nm.

randomly distributed clusters will not introduce disturbance into the pulse characterization as long as the same ensemble of clusters is excited in the process of pulse scanning.

As reported previously [9], the KN nanoneedles have the ability of wide-band frequency conversion, and the SHG efficiency is about 19.6 pm/V at 1004 nm, which is relative large among similar nano-materials [10].

2.3. Theory

Briefly, the FROG measurement can be described as follows. Define the complex electric fields of each pulse as

$$\hat{E}(t) = E(t) \exp(j2\pi f_0 t),$$

where the $E(t)$ is complex amplitude and f_0 is the carrier frequency.

The measured intensity of non-collinear FROG signal can then be written as

$$I_{\text{FROG}}^{\text{SHG}}(\tau, f) \propto \left| \int_{-\infty}^{\infty} \hat{E}(t) \hat{E}(t - \tau) \exp(-j2\pi f t) dt \right|^2,$$

with τ is the relative delay between the pulses, and $\hat{E}(t - \tau)$ is a variable-delay gate function. The spectrogram is the set of spectra of all gated chunks of $\hat{E}(t)$ as the delay is varied. Knowledge of the spectrogram of $E(t)$ is sufficient to completely determine $E(t)$ [11].

3. RESULTS AND DISCUSSION

Figure 3(a) shows the result of our measurement. The power of the LMA-PCF laser output was about 800 mW, and before the focused lens was about 10 mW for each arm. The non-collinear

FROG spectrogram can be obtained by measuring the spectrum of the SHG signal as the delay time between the two pump beams is scanned. The delay step of the FROG trace was fixed at $\Delta\tau = 33.3$ fs. The integration time of each spectrum was 100 ms, and the total acquisition time is 5 min, most of which is cost in the delay stage step motion.

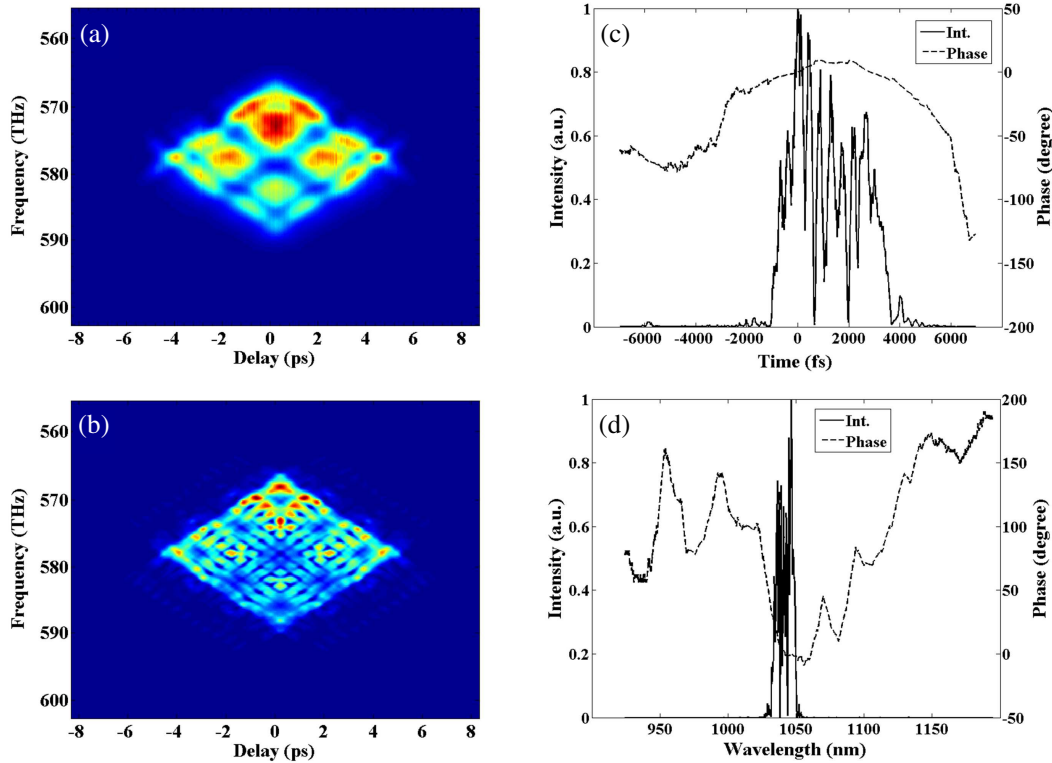


Figure 3: (a) Experimental FROG trace. (b) Retrieved FROG trace. Electric field intensity (solid line) and phase (dashed line) as a function (c) of time and (d) of wavelength.

The retrieval result shown in Figure 3(b) is obtained through a method given in Ref. [12]. Using the standard FROG algorithm, the complex pulse profile can be retrieved from the measured non-collinear SHG-FROG trace. The retrieved pulse intensity and phase profiles in temporal and spectral are also reported in Figures 3(c) and (d), respectively. The measurement can be further improved by employing a delay stage with finer step.

In our previous report, the KN nanoneedles can realize a wide-band second and third harmonic generation [9]. Combining the aforementioned ability to relax phase match constraints, they can be a potential replacement of bulk nonlinear crystal in ultrafast pulse characterization for SHG-FROG but THG-FROG. With respect to the bulk crystal, the nanoneedle clusters almost require no adjustment for different laser, which is more accessible for common users. Furthermore, by placing the nanocrystals on the focal plane, the pulse laser's properties on the sample can be reflected from the measurement, which is regarded as very valuable information for *in situ* bio-imaging.

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REFERENCES

1. Müller, M., J. Squier, and G. J. Brakenhoff, "Measurement of femtosecond pulses in the focal point of a high-numerical-aperture lens by two-photon absorption," *Optics Letters*, Vol. 20, No. 9, 1038–1040, 1995.
2. Iaconis, C. and I. A. Walmsley, "Self-referencing spectral interferometry for measuring ultra-short optical pulses," *IEEE Journal of Quantum Electronics*, Vol. 35, No. 4, 501–509, 1999.

3. Bowlan, P., et al., “Measuring the spatiotemporal electric field of ultrashort pulses with high spatial and spectral resolution,” *J. Opt. Soc. Am. B*, Vol. 25, No. 6, A81–A92, 2008.
4. Amat-Roldán, I., et al., “Starch-based second-harmonic-generated collinear frequency-resolved optical gating pulse characterization at the focal plane of a high-numerical-aperture lens,” *Optics Letters*, Vol. 29, No. 19, 2282–2284, 2004.
5. Li, H., et al., “Characterizing ultrashort optical pulses using second-order nonlinear nanoprobe,” *Applied Physics Letters*, Vol. 97, No. 26, 261108, 2010.
6. Extermann, J., et al., “Nano-FROG: Frequency resolved optical gating by a nanometric object,” *Optics Express*, Vol. 16, No. 14, 10405–10411, 2008.
7. Santos, I. C. M. S., et al., “Studies on the hydrothermal synthesis of niobium oxides,” *Polyhedron*, Vol. 21, No. 20, 2009–2015, 2002.
8. Wang, Y., et al., “Synthesis and second harmonic generation response of KNbO_3 nanoneedles,” *Journal of Crystal Growth*, Vol. 341, No. 1, 42–45, 2012.
9. Cai, F., et al., “Tunable SHG/THG from KNbO_3 nano-needles and their applications in finding optimal wavelength for deep-tissue NLO imaging,” unpublished.
10. Nakayama, Y., et al., “Tunable nanowire nonlinear optical probe,” *Nature*, Vol. 447, No. 7148, 1098–1101, 2007.
11. Trebino, R., et al., “Measuring ultrashort laser pulses in the time-frequency domain using frequency-resolved optical gating,” *Review of Scientific Instruments*, Vol. 68, No. 9, 3277–3295, 1997.
12. Trebino, R., *Frequency-resolved Optical Gating: The Measurement of Ultrashort Laser Pulses*, 1st Edition, Klumer Academic, Boston, MA, 2002.