Versatility of nonlinear optical phenomena induced by infrared pulses: application to pulse characterization, element analysis, and filamentation

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Nonlinear optics is a branch of optics which describes the behavior of light in nonlinear media, where the dielectric polarization responds nonlinearly to the electric field of the light. Techniques based on nonlinear optical phenomena are widely used in many fields, i.e., frequency conversion, optical switching, optical microscopy, spectroscopy, generation and characterization of attosecond pulses, and particle acceleration. Since in the infrared wavelength range, many high power lasers and nonlinear materials with high nonlinear coefficient are available, during most studies on nonlinear optics, the pump laser has the wavelength in this range. In this thesis, we report our experimental study on three different aspects of infrared nonlinear optical phenomena: the characterization of a mid-infrared free-electron laser (FEL), the ablation dynamics during laser ablation of solid sample submerged in liquid by nanosecond near-infrared laser pulses, and the filamentation in liquid induced by femtosecond near-infrared laser pulses.

Chapter 1 Introduction

In this chapter, we introduce the concepts of mid-infrared ultrashort pulses, Laser-Induced Breakdown Spectroscopy (LIBS), and filamentation, as well as their applications particularly those applications related to energy science. We also introduce the background for each topic in the thesis as follows.

- For many applications of the mid-infrared FEL in our institute KU-FEL, it is important to have the information such as beam quality, micropulse duration and wavelength instability. To obtain such information in a normal way, an array-type detector is required. However, such detectors for mid-infrared wavelength range are not available in our institute. Under this condition, we use the knife-edge method to measure the beam quality of KU-FEL beams, and Fringe-Resolved AutoCorrelation (FRAC) to measure the micropulse duration as well as wavelength instability.
- When performing the LIBS measurement of solid samples submerged in liquid, clear spectra can be obtained by using long pulses (>100 ns) as pump pulse. However, the ablation dynamics during long pulse LIBS of solid samples submerged in liquid is not yet

clear. In this thesis, we report the study on the influences of experimental conditions, i.e., the laser focus condition and the ambient liquid temperature on the ablation dynamics during laser ablation of under-liquid solid samples by long pulses.

- Filamentation is a well-known nonlinear optical phenomenon which happens when intense femtosecond pulses propagates in nonlinear media. Due to the interesting behavior of light during filamentation, this topic is widely studied in last two decades. However, most of these studies have used Gaussian beam as incident mode, and filamentation dynamics for other beam modes, such as Bessel beam, are not yet clear. In this thesis, an experimental study is reported to compare the different filamentation dynamics for the cases of Gaussian and Bessel incident beam modes.

Chapter 2 Spatial characterization of mid-infrared free-electron laser pulses by knife-edge method

In Chapter 2, we propose a method of using 2D knife-edge technique to measure the beam quality. Under the assumption that the intensity distribution of the laser beam is separable in two directions, we can reconstruct the 2D beam profiles at different propagation distances by the results of a 2D knife-edge measurement. A numerical test shows this method works well only if the 2D intensity distribution is separable in two directions along which the scanning is performed. After obtaining the beam diameters from the beam profiles at different propagation distances, we can perform a fitting between the beam diameter and the propagation distance, during which the M^2 factor of the beam can be obtained. To reduce the noise possibly introduced by step size jitter during the measurement, data processing such as fitting or smoothing is performed on the raw data before taking the derivative. From the obtained 2D beam profiles, we find the KU-FEL beam has the radially symmetric shape near the focus, while at far field, the beam profiles are quite distorted. Because of this non-Gaussian property of KU-FEL beam, the definition of 4σ beam diameter is applied during the acquisition of the beam diameters from the 2D beam profiles. The M2 factor of KU-FEL beam is measured to be ~1.1 in both vertical and horizontal directions. As a cross check, a burn pattern experiment is also carried out. The burn pattern obtained in this test also shows a round shape near the focus, and asymmetric shape at far field, which is consistent with what we have observed in the knife-edge experiment.

The method proposed in this chapter can be also applied for lasers at other wavelength such as deep ultraviolet where 2D array-type detectors are not easily available.

Chapter 3 Characterization of mid-infrared free-electron laser pulses by auto-correlation method

In this chapter, we describe a measurement of the duration as well as the wavelength instability of KU-FEL micropulses by FRAC method. The idea can be simply described as follows. The FRAC signal consists of two parts, the envelopes and the oscillation. The envelopes (upper and lower) are determined by the temporal pulse shape and the chirps, while the period of the oscillation is one optical cycle of the central wavelength. The FRAC signals of two pulses which are exactly the same except for the central wavelength will have the same envelopes, but oscillate at different frequencies. If we add them together, the fringes at large delays will fade away because of the different oscillation frequencies, and the width of the envelopes will become smaller. Taking the FRAC signal of a macropulse which consists of thousands of micropulses with different central wavelength is somehow similar with the process we mentioned above. So we expect the wavelength instability can be known from the shapes, or widths of the FRAC envelopes. By a numerical experiment taking different kinds of instability and chirps into consideration, we clarify this idea, and point out that the wavelength instability can be obtained by fitting the FRAC envelopes. However, the shapes of FRAC envelopes are also influenced by other parameters such as pulse duration and chirps, and the strong shot-to-shot instability of KU-FEL gives a bad quality to the measured FRAC signal. For this case, a direct fitting does not work well. To solve this problem, we should know the micropulse duration before the fitting. Fortunately, we find that although the shape of FRAC envelopes change with many parameters, the integration of the oscillations is proportional to nothing but the micropulse duration. Note that no matter how narrow the envelopes are, the value of this integration is determined only by the pulse width. This means from a FRAC signal, we can extract the pulse duration even without knowing anything in advance. By the method we mentioned above, the micropulse duration of KU-FEL is measured to be 0.58 ps, and the wavelength instability is around 1.3%. As a cross check, an Intensity AutoCorrelation (IAC) measurement is also carried out to measure the micropulse duration of KU-FEL. The IAC result suggests a micropulse duration of 0.66 ps, which is similar to what we obtained from the FRAC signal.

The method we propose in this chapter can greatly improve the performance of FRAC when it is used for the characterization of ultrashort pulses.

Chapter 4 Ablation dynamics of a solid target in liquid by nanosecond near-infrared laser pulses

In the first part of this chapter, we report our experimental study on the influence of laser focus position on the ablation of a solid target in liquid by long nanosecond pulses. During this experiment, we observe the change of plasma plume, emission intensity, and emission spectra with focus position at several different incident pulse energy. From the result, we find the proper laser intensity near the sample surface is the key point to obtain result with good quality in such LIBS measurement. Too low laser intensity results in a small plasma plume, and weak emission, while at too high laser intensity, the water above the sample frequently get broken down, which causes a distortion in the spectra. We also confirm this breakdown happens due to the impurity of the water near the sample which is caused by precedent laser pulses. As a conclusion, we find during long pulse LIBS of under-liquid solid samples, to obtain the data with good quality, we can either focus above or below the sample, as long as the laser intensity near the sample is in some certain range.

As we know, the ablation dynamics during under-liquid LIBS is closely related to the evolution of laser-induced bubble. And the bubble dynamics is related to some properties of the solvent, such as viscosity and vapor pressure which may be sensitive to ambient temperature. In the second part of this chapter, we report our experimental study on the influence of solvent temperature on ablation dynamics during under-liquid LIBS of solid samples. In this experiment, we measure the plasma image, emission intensity, emission spectra, and bubble lifetime during ablation of a Cu sample in water, ethylene glycol (EG), and polyethylene glycol 300 (PEG) at different temperatures of 22 and 60 ^{o}C. We find that at higher temperature the plasma emission is brighter. And from the result of spectral measurement, we find the signal enhancement for Cu lines is not obvious, while the spectral intensity of the components related to the solvent (C_2 emission in the cases of EG and PEG) increases a lot. At the same time, the bubble lifetime also becomes longer at higher ambient temperature for all three kinds of liquid. One of the possible reasons for the observations is that the vapor pressure is higher at higher temperature. As we know, during long pulse LIBS, just around 100 ns after the peak of the laser pulse hits the sample, the size of laser-induced bubble becomes larger than the plasma plume, and the plasma plume expands inside the bubble. When the bubble expands to the size of hundreds of micrometers, the pressure in the bubble is quite low, and liquid molecules will enter the bubble in the form of vapor. If vapor pressure becomes higher, more molecules will get vaporized, and the density of solvent molecules increases in the bubble. As a result, the spectral intensity of the components related to the solvent increases. And from Rayleigh-Plesset equation, we find the bubble lifetime becomes longer when the pressure in the bubble becomes higher. At higher temperature, the pressure in the bubble becomes higher because more molecules enter the bubble in the form of vapor. This may explain the increase of bubble lifetime in the case of water. For the cases of EG and PEG, however, the increase of the vapor pressure is insufficient to cause an obvious increase of the bubble lifetime. For these cases, one possible reason for the bubble lifetime increase is that more solvent molecules decompose into smaller molecules such as C_2 at higher temperature. This decomposition increases the pressure in the bubble, which results in a longer bubble lifetime at higher temperature.

Chapter 5 Nonlinear propagation through liquid media of femtosecond near-infrared pulses in Gaussian and Bessel modes

In the first part of this chapter, we introduce the properties of Gaussian and Bessel beams during linear propagation. In this part, we also assume several kinds of misalignments during generation of Bessel beam using axicon, and study their influence on the quality of created Bessel beams by numerical simulation.

In the second part, we report a comparison study of filamentation dynamics for Gaussian and Bessel incident beams. In this experiment, we observe the beam profiles and spectra changes during nonlinear propagation in methanol for Gaussian and Bessel beams with different incident energy. For both incident beams, we can clearly observe the starting and stopping positions of self-focusing and Self Phase Modulation (SPM). We find that compared with the case of Gaussian incident beam, when the incident beam is Bessel, nonlinear phenomena such as self-focusing and SPM starts and ends at much higher on-axial laser intensity.

Chapter 6 Summary and outlook

In this chapter, we summarized the work we reported in previous chapters, and suggest some prospective experiments which we can do in future.