DEVELOPMENT OF A PUMP-PROBE SYSTEM USING A NON-COATED ZnSe BEAM SPLITTER CUBE FOR AN MIR-FEL

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Abstract

A reliable pump-probe technique is essential to gain a proper understanding of the interaction of lasers with tissue and similar materials. Our pump-probe system divides an incident s-polarized MIR-FEL (Mid-Infrared Free Electron Laser) into two beams of equal intensity, and simultaneously crosses the two beams. One beam acts as a pump beam, while the other as a probe beam with an additional time delay. Time-resolved absorption spectroscopy using this technique provides us with information about the vibrational dynamics of molecules. We have developed this system using a non-coated ZnSe beam splitter cube. The beam splitter cube is composed of two ZnSe prisms in the shape of a trapezoid. Two pulses with equal intensity are generated due to Fresnel reflection/transmission at the boundary between the two prisms, which then simultaneously illuminate the same position around the point of intersection of the two divided beams. We have conducted a proof-of-concept experiment for this system using the MIR-FEL. We showed that this system requires no complicated or time-consuming optical alignment, and that it is applicable for a broad MIR waveband (5~11 μm).

INTRODUCTION

In order to properly understand the interaction of lasers with tissue and other materials, a pump-probe technique is required. Conventional pump-probe techniques using a Michelson interferometer split an incoming pulse into two beams; a pump beam and a probe beam with a variable time delay relative to the pump beam. In this technique the two beams must simultaneously illuminate the same position, so it requires complicated and time-consuming optical alignment. Additionally, optical alignment for invisible mid-infrared (MIR) light is technically difficult.

From the above point of view, we have proposed and designed a new pump-probe system for a tunable MIR-FEL (Free Electron Laser) using a non-coated ZnSe beam splitter cube. Since one beam splitter cube divides an incoming pulse into two pulses with equal intensity, complicated and time-consuming optical alignment is no longer requied. We adopted the use of Fresnel reflection/transmission and total internal reflection to equally divide the incident MIR-FEL pulse into the two pulses. Since AR coating severely limits the applicable waveband range, no AR coating was used. In this article, we describe the principles and features of this pump-probe system. In order to confirm the performance of a trial pump-probe system, we have conducted its proof-of-concept experiment using an MIR-FEL. Since the FEL user facility at Osaka Univ. can provide an MIR-FEL within the range of λ = 5~12 μm [1], we designed the ZnSe beam splitter cube for λ = 7.5 μm. The refractive index of ZnSe is 2.421 at λ = 7.5 μm.

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PRINCIPLES

Figure 1(a) shows the principles of our pump-probe technique involving the ZnSe splitter cube. The ZnSe splitter cube consists of two non-coated ZnSe prisms in a trapezoidal configuration. An air layer with a thickness of 50 μm exists between the two ZnSe prisms. The incident MIR-FEL beam is s-polarized. The incoming MIR-FEL pulse divides the two pulses (Beams A and B) at the boundary between the ZnSe prisms (point C). The reflected and transmitted beams are reflected due to total internal reflection, and then each beam crosses simultaneously outside the beam splitter at point D. In order to generate two pulses A1 and B1 with equal intensity, the angle θ_3 must be set at 20.4 deg, leading to reflectance R = 0.382, as shown in Fig. 1(b). Multiple Fresnel reflection occurs at point C. If we let the intensity

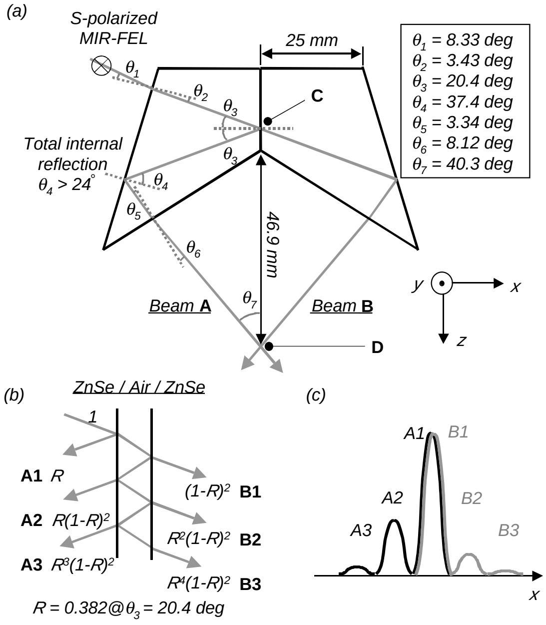

Figure 1: Principles of the pump-probe system using a non-coated ZnSe beam splitter cube. (a) Configuration of this system. The definitions of the x-, y-, and z-axes are also shown. (b) Generation of Beams A1 and B1 with equal intensity at the boundary between the two prisms (point C). (c) Typical profiles of Beams A and B along the x-axis at the point of intersection (point D).

of the incident beam at point C be unity, then the intensities for the respective divided beams are as follows: $I_{A1} = 3.82\times10^{-1}$, $I_{A2} = 1.46\times10^{-1}$, $I_{A3} = 2.13\times10^{-2}$, $I_{B1} = 3.82\times10^{-1}$, $I_{B2} = 5.57\times10^{-2}$, and $I_{B3} = 8.13\times10^{-3}$. Here we neglected the forth reflection/transmission and higher. As shown in Fig. 1(c), we can simultaneously illuminate a target at point D by using the two pulses (Beams A1 and B1) of equal intensity. Beams A1 and B1 can be used as either a pump or a probe beam. Note that the intensities for Beams A1 and B1 are ~0.3 (=0.382×(1–0.3)) times the incident MIR-FEL, since the total reflectance loss at the first and final boundaries is ~0.3.

Figure 2 shows the configuration of a typical pump-probe experimental set-up. The sample is located at point D. In this case, an optical window, whose material has good transmission characteristics for MIR-FEL, is inserted to the beam line for Beam B1, which is used as a probe beam. The optical window (such as CaF_2 and MgF) acts as a time delay relative to Beam A1, which acts as the pump beam. The time delay can be arbitrarily varied within the range 1~20 ps by changing the thickness of the optical window. The intensity of the transmitted probe beam is time-resolved by an MCT detector. This time-resolved signal, $S(t)$, gives us information about the vibrational dynamics of molecules during MIR-FEL irradiation.

The main features of this pump-probe technique are as follows.

(1) Since one beam splitter cube divides two pulses with equal intensity and crosses them over, optical alignment of this system is simple and not time-consuming.

(2) ZnSe prisms have good transmission characteristics

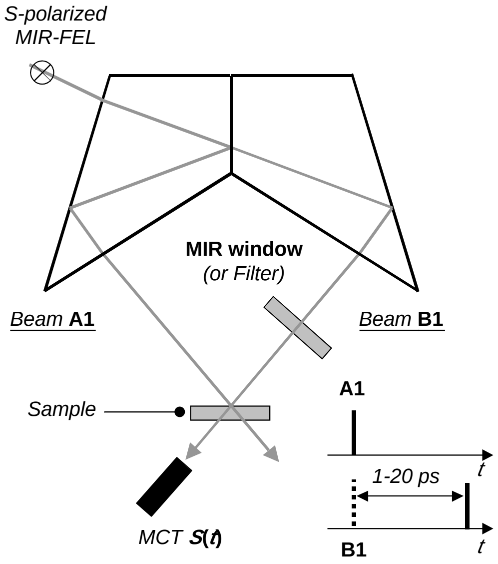

Figure 2: Typical configuration of the pump-probe experimental set-up. In this case, Beam A1 is the pump beam, while Beam B1 is the probe beam. The MIR window acts as an optical delay.

over both the MIR and the visible spectrum, which enables us to use He-Ne lasers as the guide laser for optical alignment.

(3) The variation of the refractive index for ZnSe is less than 1% over a broad waveband of 2~11 μm [2]. In addition, non-coated ZnSe prisms are used. This means that tunable lasers can be utilized.

(4) This system is composed of transmittive optics. The use of ultra-fast pulse lasers is not possible due to group velocity dispersion within the system. This system is applicable for picosecond MIR-FELs, since the effects on the group velocity dispersion become significant in the femtosecond range.

(5) The time delay is constrained by the thickness of the MIR windows and is less than a few tens of picoseconds.

(6) The reflectance of the MIR windows is about 3~5%, leading to a slight difference in intensity between the two pulses.

PROOF-OF-CONCEPT EXPERIMENT

We have conducted a proof-of-concept experiment using a tunable MIR-FEL in order to confirm the following performances of this system: (1) the division into two pulses (A1 and B1) with equal intensity over λ = 5~11 μm and (2) the focusability of the two pulses onto the same position.

Division

When the intensity of Beam A1 is exactly equal to that of Beam B1, the intensity ratio of Beam A to Beam B is 1.23 (as shown in Fig. 1(b)), where Beam A (B) generally terms multiple beams A1~A3 (B1~B3). The intensity ratio can be controlled by the incident angle of the MIR-FEL, θ_1. We measured the average powers for Beams A and B while adjusting θ_1. As a result, we confirmed that the intensity ratio can be set at 1.23 with a possible error of ±10% over λ = 5~11 μm by only adjusting θ_1. Therefore, we can easily generate two pulses (A1 and B1) with equal intensity within an error of ±10%. This error causes due to the uncertainty of the measurement of an average power.

Focusability

We examined the focusability of the two pulses onto the same position using heat sensitive papers, which were located at the point of intersection (point D). We obtained burn patterns on the papers by MIR irradiation while moving the target along the *z* direction. Note that this experiment was carried out after adjustment of the intensity ratio as described previously.

Figure 3 shows the burn patterns on the paper observed with an optical microscope after MIR-FEL irradiation. The wavelength and average power of the incident MIR-FEL were 9.4 μm and ~25 mW, respectively. An increase in *z* means a backward movement away from the beam splitter. We can see three burn patterns for Beams A1, A2, and B1. Patterns for the other beams were not observed because of their weaker intensities. As *z* increases, the burn patterns of A1 and A2 gradually become closer to that of B1. Around z = 0.5 mm, patterns A1 and B1 overlap with each other, and then they separate again with an additional increase in z.

From Fig. 3, we can obtain data for the optical paths of Beams A1 and B1. When z is changed from 0 to 0.2 mm, the x-position of the center of Beam B1 shifts laterally by Δx = ~150 μm. We first experimentally estimated θ_7' from these data using the expression: $\theta_7' = \tan^{-1}(\Delta x/\Delta z)$, resulting in θ_7' = 36.9 deg. The specification of θ_7 is 40.3 deg (Fig. 1 (a)). This disagreement gives us the difference in path length between Beams A1 and B1. This difference in path length, ΔL, can be roughly evaluated by using the equation: $\Delta L = L_{\mathrm{total}}(1-\cos(|\theta_7-\theta_7'|))$, resulting in ΔL = 0.206 mm. This difference leads to the time difference between the both beams, Δt (< 1 ps in this case). Δt is considerably shorter than the pulse duration of our MIR-FEL and is negligible. Here L_{total} is the total path length from the first ZnSe surface to point D, and was about 117 mm.

Secondly, we estimated the positional difference between both beams in the y direction. The distance between the centers of the both beams in the y direction is seen to be ~100 μm from the topmost image in Fig. 3. A positional difference should be reduced down to less than one-tenth the spot size of each beam at point D.

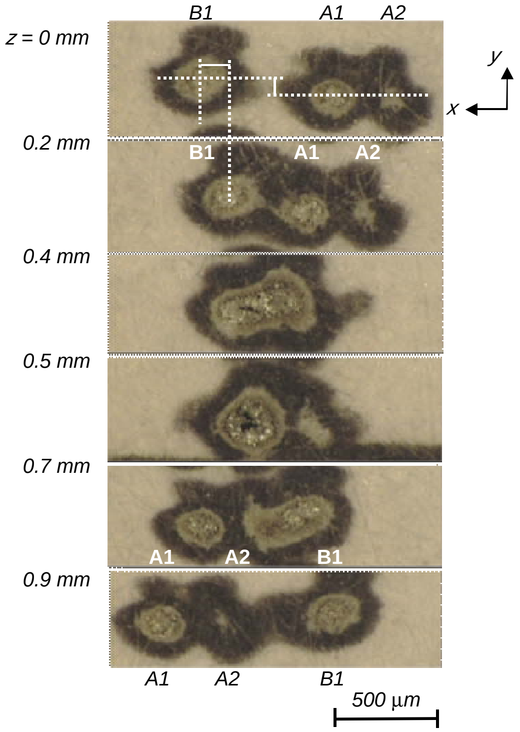

Figure 3: Observed burn patterns by MIR-FEL irradiation as a function of z. The definitions of the x-, y-, and z-axes are similar to those used in Fig. 1(a).

From these observations, the following can be derived.

(1) The pump-probe technique using the ZnSe beam splitter cube does not require complicated and time-consuming optical alignment and is applicable for a tunable MIR-FEL system. Especially, we can divide two pulses with equal intensity by only adjusting the incident angle of the MIR-FEL even over a broad waveband (λ = 5~11 μm).

(2) The time difference between both pulses was less than 1 ps and can be neglected.

(3) The positional difference in the y direction was ~100 μm, meaning that the spot size of the beams should be set at 1000 μm or more at point D.

CONCLUSIONS

In order to establish a useful pump-probe technique using a ZnSe beam splitter cube, we have designed it and have carried out its proof-of-concept experiment using an MIR-FEL. We demonstrated that this pump-probe technique is a wavelength-independent system and is applicable for a broad MIR waveband.

We will further measure the dynamic absorption properties [4] of soft tissue using this pump-probe technique. The dynamic absorption properties are those during laser irradiation and strongly dominate the extent of the heating, coagulation, and cutting of tissue. For non-invasive and predictable laser surgery, a knowledge of the dynamic absorption coefficients is essential.

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