Optical characteristics of the undamaged and laser damaged K9 glass in terahertz band

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Abstract
K9 glass is an important optical element of the high energy laser system. In an attempt to investigate the variation of optical properties of K9 glass before and after laser induced damage, based on the transmission terahertz time-domain spectral system, the undamaged and the damaged K9 glasses induced by laser were tested to obtain the terahertz time-domain and frequency-domain spectra and the refractive index and absorption coefficient of the samples were calculated in terahertz band for further analysis. Results show that with the increase of laser energy, from the undamaged to the damaged K9 glasses to different extent, the peak to peak value in time domain and the amplitude value in frequency domain present a decreasing trend in varying degrees, and in the 0.4 ~ 0.5 THz band, the absorption coefficient increases continuously with a slight change of refractive index, which indicates that K9 glass not only appears the melting and fracture in the macroscopic morphology after laser-induced damage, but also forms non-bridged oxygen atoms in its microstructure with more ion bonds and free electrons and the change of ion polarization. In this regard, this paper provides a technical basis for exploring the micro characteristics of K9 glass after laser-induced damage, and lays a good foundation for the application of terahertz time-domain spectroscopy to laser-induced damage analysis.

1. Introduction
In the high-power system, there exist a lot of optical elements, whose interaction with laser has always been the concern of public [1, 2]. As an important optical element, the performance of K9 glass determines the output power and beam quality [3, 4]. In the studies of its interaction with laser, great attention has been paid to the damage mechanism and process [5–7], in the damage mechanism, it is mostly studied by focusing on the optical parameters before and after damage [8–10]. Optical parameters, the link between the macroscopic properties and microscopic states of materials, not only represent the macroscopic physical properties but present the microscopic properties and mechanism of materials [11], therefore it is of great significance. At present, it is rarely reported to investigate the variation of optical parameters of K9 glass before and after laser-induced damage, and lays a good foundation for the application of terahertz time-domain spectroscopy to laser-induced damage analysis.

Terahertz time-domain spectroscopy is a newly emerging spectrum measurement technology. The optical parameters in the terahertz band can be calculated based on the measurement of amplitude and phase changes of the terahertz wave after passing through the samples. Compared with the traditional Fourier transform infrared spectroscopy, terahertz time-domain spectroscopy technology contributes to the rapid and efficient calculation of optical parameters without Kramers-Kronig transformation [12, 13]. Naftaly et al employ THz time-domain spectroscopy to obtain the refractive index and absorption coefficient of quartz glass and BK7 glass and analyze the properties of materials [14, 15]. Parrott et al according to terahertz time-domain spectroscopy technology, investigate the relationship between the content of sodium with different proportions and the atomic charge distribution in sodium silicate glass [13]. Yatongchai et al probe the refractive index and dielectric constant to
study the hydroxyapatite-glass composites [16]. On the basis of terahertz time-domain spectroscopy, Ravagli A and Kang SB et al inquire into the relationship between the dielectric properties of chalcogenide glasses with different components and their structural composition and polarizability [17, 18]. Zalkovskij et al study the optical properties of chalcogenide glass within the 0.2–18 THz band [19]. Yang J et al carry an elaborate analysis of the important characteristic of glass—boson peak in the terahertz band [20–22]. In view of the studies above, the optical parameters of glass are obtained based on the terahertz time-domain spectroscopy technology for further investigation of the properties of glass. However, few studies touch upon the optical properties of glass before and after laser-induced damage. Therefore, the present paper probes into the optical parameters of K9 glass before and after laser-induced damage in the terahertz band to further investigate the effect of laser-induced damage on the microscopic characteristics, which also provides a technical foundation for studying the influence of laser-induced damage on the optical characteristics of optical elements.

2. Experiment

2.1. Experiment apparatus

The terahertz time-domain spectroscopy system generally consists of femtosecond laser, terahertz generation device, terahertz detection device, delay device and sample stage. In the present experiment, the transmission terahertz time-domain spectroscopy system is shown in figure 1, where MaiTai laser is adopted with the center wavelength of 800 nm, pulse width of 60 fs, repetition frequency of 80 MHz and output power of 950 mW [23]. The femtosecond laser pulse, emitted by the laser, is divided into two beams by the beam splitter, one as pump light and the other as detection light. With the time delay system, the pump light focuses on GaAs photoconductive antenna, which contributes to the generation of terahertz wave. After collimating and focusing on the sample by the off-axis paraboloid mirror, the terahertz pulse including the sample information and the detection light pass through the ZnTe electro-optic crystal at the same time. Since the THz electric field changes the refractive coefficient of ZnTe crystal, the transmitted detection light pulse is transformed into the elliptically polarized light. With the treatment of Wallaston prism, the voltage is output by the balance detector, which reflects the electric field intensity of terahertz wave passing through the sample. Based on the point-to-point delay, the whole time-domain spectrum of terahertz pulse can be obtained through the ellipsometry change of
the detection light. During the experiment, the whole system is set in the air environment, with the temperature of 23 °C, the relative humidity of 13% and the signal-to-noise ratio of 1008 dB.

2.2. Experimental samples
K9 glass of 630 × 2 mm is selected as the experimental samples and N-on-1 mode is adopted in laser-induced experiment, that is, multi-pulse laser induction. In addition, Nd:YAG solid-state laser is employed with the output wavelength of 1064 nm, the pulse width of 10 ns and the maximum output energy of 400 mJ. The output laser beam after the attenuating and focusing irradiates on the surface of K9 glass with the laser energy density of 12.66 J cm⁻², 19.9 J cm⁻², 25.88 J cm⁻² and 48.68 J cm⁻² respectively. With the same irradiation time, the damaged area of the sample, observed by microscope, is shown in figure 2 and the diameters of the damage spot in figures 2(a)–(d) are about 1 mm, 2 mm, 3 mm and 3.5 mm respectively.

In the case of the laser energy density of 12.66 J cm⁻², damage occurs on the surface of glass with the severely ablated irradiation centers and obvious annular cracks in the periphery. When the laser energy density is 19.9 J cm⁻² and 25.88 J cm⁻², severe ablation is in the irradiation center, more obvious ring-shaped crack in the periphery, the damage area is enlarging and the damage extends from surface to interior with the increasing of damage depth; Given the laser energy density of 48.68 J cm⁻², severe ablation and pitting appears in the irradiation center of glass with circular cracks in the periphery. Besides, there are also ablation cracks along the radial direction, and two obvious fracture areas appear in the lower right part of figure 2(d).

2.3. Extraction model of optical parameters
The optical parameters of sample in the terahertz band can be calculated according to the physical model proposed by Dorney and Duvillaret [24, 25]. The THz signal through air is taken as the reference signal and that through the sample is taken as the sample signal. The reference signal $E_a(\omega)$ and the sample signal $E_s(\omega)$ in frequency domain can be obtained by Fourier transformation. The transfer function $H(\omega)$ of THz wave through the sample is expressed as:

$$H(\omega) = \frac{E_s(\omega)}{E_a(\omega)} = \frac{4\tilde{n}_s(\omega)}{[1 + \tilde{n}_s(\omega)]} \exp \left[ -\frac{j\delta(\tilde{n}_s(\omega) - 1)\omega}{c} \right] = \rho(\omega)e^{-j\omega}$$ (1)
where \( c \) is the propagation speed of THz wave in vacuum; \( d \) is the sample thickness; \( \rho(\omega) \) and \( \phi(\omega) \) are the amplitude ratio and phase difference of sample signal and reference signal respectively; \( \tilde{n}_i(\omega) \) is the complex refractive index of the sample, which is written as

\[
\tilde{n}_i(\omega) = n_i(\omega) - ik(\omega)
\]

where \( n_i(\omega) \) is the real refractive index, presenting the dispersion characteristics of the sample; \( k(\omega) \) is the extinction coefficient, describing the absorption characteristics of the sample; Since the absorption coefficient \( \alpha(\omega) \) is expressed as

\[
\alpha(\omega) = \frac{2\omega k(\omega)}{c}
\]

the refractive index and absorption coefficient of the sample, based on the substitution of Formula (2) and Formula (3) into Formula (1), are presented as follows.

\[
n_i(\omega) = \frac{c\phi(\omega)}{\omega d} + 1
\]

\[
\alpha(\omega) = -\frac{2}{d} \ln \left( \frac{n_i(\omega) + 1}{4n_i(\omega)} \right)^\rho(\omega)
\]

In Formula (5), the absorption coefficient of the sample includes the absorption of the incident THz waves by the sample itself and the attenuation caused by the scattering and reflection [26–28].

3. Results and discussion

The test results of K9 glass before and after laser-induced damage using the transmission THz-TDS system are shown in figure 3. As the figures indicate, the peak to peak amplitude of THz wave passing through the undamaged K9 glass is 10.28% of the reference signal in figure 3(a), and the delay time is 10.19 ps. The former is caused by the absorption, scattering and surface reflection of THz wave by K9 glass, and the latter is that the refractive index of K9 glass is higher than that of air. It can be seen from figure 3(b) that compared with the undamaged region, the peak-to-peak signal of THz wave passing through the damaged K9 glass presents an attenuation trend, with the peak-to-peak amplitude of damage area decreasing by 3%, 11.28%, 16.14% respectively given the laser energy density of 12.66 J cm\(^{-2}\), 19.9 J cm\(^{-2}\), 25.88 J cm\(^{-2}\) and 31.66% given the laser energy density of 48.68 J cm\(^{-2}\). The main reason is that when the laser acts on the K9 glass surface, the energy will diffuse in the glass body in the way of thermal mechanical coupling, which contributes to the generation of the temperature field and stress field and makes the glass transit from no damage to surface damage and then to three-dimensional damage. Furthermore, surface melting, internal fracture and rough damage area are caused, thus increasing the absorption and scattering of THz wave energy and weakening the signal strength of THz wave. As the laser energy density increases, the damage area and depth present an upward trend with more obvious three-dimensional damage. The surface and inside show stronger absorption and scattering ability for terahertz wave, and the signal amplitude of terahertz wave decreases significantly.

All the acquired time domain signals are treated with Fourier transform to obtain THz spectrum of 0.35 ~ 0.5 THz, as shown in figure 4. Since the measurement is carried out in the air environment, the obvious absorption peaks of water vapor are shown in the figure [29, 30]. Moreover, the amplitude of the damaged area is lower than that of the undamaged area at each frequency. For the damaged area induced by laser with energy density of 12.66 J cm\(^{-2}\), 19.9 J cm\(^{-2}\), 25.88 J cm\(^{-2}\), with the increase of the laser energy density, there is a gradual declination in the amplitude, while the amplitude value decreases sharply at the laser energy density of 48.68 J cm\(^{-2}\), consistent with the peak to peak value variation of the corresponding time domain signal, since the morphology of K9 glass changes in varying degrees under laser irradiation. When the laser energy density is 12.66 J cm\(^{-2}\), the surface damage, the main form of damage morphology, plays an important role in the absorption and scattering of THz waves at different frequencies. As the laser energy density increases to 19.9 J cm\(^{-2}\) and 25.88 J cm\(^{-2}\), the damage area enlarges gradually and the damage depth increases slowly, the absorption and scattering of THz waves with different frequencies occurs on the surface and within a certain depth, the amplitude declines slowly. As for the laser energy density of 48.68 J cm\(^{-2}\), the damage area is larger and the damage depth becomes longer quickly, which results in the stronger absorption and scattering of THz waves, so the amplitude decreases obviously.

The refractive index and absorption coefficient of undamaged and damaged area of K9 glass can be calculated according to Formula (4) and Formula (5), as shown in figures 5 and 6. As figure 5 indicates, the refractive index of K9 glass in the undamaged region falls within 2.55–2.61 in the 0.4–0.5 THz band, which is consistent with the value given in [15]. In terms of the damaged area, the refractive index has changed a little, mainly owing to the close correlation of refractive index with the ion polarization. Under the action of strong
Figure 3. The time domain waveform obtained using the transmission THz-TDS system with an aperture of 1 mm at the focus position of THz wave (a) without sample and (b) through K9 glass without damage and with damage induced by laser on condition of the laser energy density of 12.66 J cm$^{-2}$, 19.9 J cm$^{-2}$, 25.88 J cm$^{-2}$ and 48.68 J cm$^{-2}$.

Figure 4. The amplitude spectra in frequency domain of the reference signal and the sample signals obtained by terahertz wave passing through the undamaged area and the damaged area in K9 glass with the laser energy density of 12.66 J cm$^{-2}$, 19.9 J cm$^{-2}$, 25.88 J cm$^{-2}$ and 48.68 J cm$^{-2}$. 
light field, a polarization component of the polarization intensity, exists in relation to the higher-order power of the applied electric field. In other words, there is a nonlinear refractive index. In this regard, the refractive index of optical glass presents the characteristics related to the intensity \[31\]. Furthermore, the polarization intensity is closely related to the microstructure of materials. With the strong laser irradiation, the Si–O bond and B–O bond in the glass network structure are broken, non-bridged oxygen atoms form and more ion bonds appear, which causes the change of glass refractive index \[32, 33\].

The absorption coefficient curves in the 0.4 ~ 0.5 THz band are shown in figure 6. The absorption coefficients in both undamaged and damaged regions increase firstly and then decrease with the change of frequency as a result of the influence of water vapor absorption in terahertz band. As the laser energy density increases, the absorption coefficient also increases at each frequency, since in the interaction of laser with the glass, the irradiation center melts and breaks with a large amount of heat deposition, and the crack spreads from the center to the surrounding. With the increase of laser energy, the damage area also presents an increasing trend, and the crack depth gradually deepens until the glass breaks \[32\]. After cooling, the solidification of splashes, the deepening of melting coloring concentration and the roughness of glass surface increase the absorption of THz wave energy. At the same time, the change of macro-morphology also corresponds to the change of microstructure and properties. For the damaged area, the band gap narrows down and non-bridged oxygen hole color centers and E′ color centers form, as well as more ion bonds and free electrons, which increases the optical absorption of glass \[32, 34\]. The absorption coefficient further shows that the amplitude of

![Figure 5. Refractive index curves of the undamaged area and the damaged area in K9 glass with the laser energy density of 12.66 J cm$^{-2}$, 19.9 J cm$^{-2}$, 25.88 J cm$^{-2}$ and 48.68 J cm$^{-2}$.](image)

![Figure 6. Absorption coefficient curves of the undamaged area and the damaged area in K9 glass with the laser energy density of 12.66 J cm$^{-2}$, 19.9 J cm$^{-2}$, 25.88 J cm$^{-2}$ and 48.68 J cm$^{-2}$.](image)
terahertz wave decreases when it transmits through the damaged area of K9 glass which is induced by laser with different energy densities as a result of the stronger absorption of THz wave at the damaged area. Due to the limitation of experimental conditions, the terahertz spot diameter is about 2 mm, and the energy is at micro watt level. In this regard, the measurement can only be performed for the damage points with the diameter larger than 0.6 mm. For smaller damage points, smaller test beam and higher power are required.

4. Conclusion

Based on the transmission terahertz time-domain spectral system, for K9 glass, both the undamaged region and the damaged regions induced by laser with the energy density of 12.66 J cm\(^{-2}\), 19.9 J cm\(^{-2}\), 25.88 J cm\(^{-2}\) and 48.68 J cm\(^{-2}\) respectively are tested to obtain the corresponding time-domain and frequency-domain amplitude curves and further calculate the refractive index and absorption coefficient at 0.4–0.5 THz. The results show that there is a significant decrease of the peak to peak value in time domain and the amplitude value in frequency domain by the comparison of the damaged area with the undamaged area, and the higher the laser energy density, the larger the amplitude attenuation. In the 0.4–0.5 THz band, the refractive index of the damaged area changes slightly, and the nonlinear refractive index occurs with the change of glass microstructure. Furthermore, the absorption coefficient of the damaged area becomes large with the increase of laser energy density, which indicates the increasing absorption of terahertz wave energy and the narrowing band gap of corresponding microstructure with more ion bonds and free electrons produced. In this regard, THz time-domain spectroscopy technology serves as a useful tool for the analysis of optical properties of K9 glass before and after laser-induced damage, and the optical parameters of the undamaged and damaged area of K9 glass can be extracted at the same time and be compared, to analyze the effect of laser-induced damage on the optical property and microstructure of K9 glass, which will provide technical basis for studying the mechanism of laser-induced damage of optical elements.

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