Study on the femtosecond laser-induced refractive index change in a silicate glass by transient lens method

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When a laser pulse is tightly focused inside a transparent solid material, a permanent structural change occurs in the focused region. This phenomena will be very useful to construct many important devices such as integrated optical waveguides, or three-dimensional photonic devices. The mechanism of the light induced structural change inside a glass was investigated in a picosecond–nanosecond region by a transient lens technique with an intense subpicosecond pulsed laser and microscope objective lens. Just after the irradiation of the pump pulse inside the glass, a concave lens was instantaneously created. After this fast response signal, an oscillation with some different frequencies in GHz region was observed. The origin of the oscillation is discussed. [DOI: 10.1063/1.1517744]

I. INTRODUCTION

When intense laser light is focused on a solid surface that has an absorption band at that wavelength, the material will be ejected from the surface.[1] This photoablation effect has been used to fabricate the surface and one of main origins of the ablation is know to be the photothermal effect. On the other hand, when an intense IR laser pulse is focused inside a transparent material, a permanent structural change is frequently observed inside the material.[2,3] In 1996, Hirao's and Mazur's groups showed that a microstructure can be fabricated inside glasses with a femtosecond IR laser to write an optical waveguide[2] or to construct a three-dimensional memory.[3] It has been expected that this technique will open a way to a variety of applications[1,2,4,5] such as the construction of an optical microcomputer system. However, the mechanism of the light-induced structural change or even the rate of the change has not been investigated at all. Here, we report a preliminary trial to elucidate the initial step of the intense light–matter interaction.

The light-induced structural change depends on the laser power (see Fig. 1). When a moderate power of a subpicosecond laser pulse was tightly focused inside a glass, the refractive index at the focused region increases.[2,6,7] However, when the laser power was increased further, a vacant space (void) is created inside the solid material and the refractive index decreases.[3,8] The mechanism of the void structure creation could be explained in terms of the Coulomb explosion or extensive thermal expansion after the multiphoton excitation of the material.[9] It may not be difficult to understand intuitively. On the other hand, the mechanism of increase in the refractive index is less clear. Some groups suggested that a density increase is a main factor of the refractive index increase in the focal region.[5,6,10] However, it is not clear how the density increases. The heating after the laser exposure should lead the thermal expansion and probably result in a less dense structure in the focal region, which is opposite to the observation. To elucidate the light-induced refractive index change, we tried to observe the refractive index change in a fast time subpicosecond (10^{-13} s)–nanosecond (10^{-9} s) region. In order to monitor the refractive index change in a fast time scale in a small spatial area, we used the transient lens (TrL) method with a subpicosecond pulsed laser focused by a microscope objective lens.

II. PRINCIPLE

The principle of the TrL method is similar to that of the traditional thermal lens method.[11,12] When a sample is irradiated by a pump laser pulse having a cylindrical symmetric shape such as a Gaussian shape, the distribution of excited states and the following structural change should have the same spatial shape. The density and absorption change associated with structural change should induce the refractive index change, and the refractive index should have the same cylindrical symmetric distribution, too. If the refractive index at the center becomes higher, this distribution acts as a convex lens. If the light induces the decrease of the refractive index, it creates a concave lens. When a collimated laser beam is passed thorough the lens region, the transmitted beam is expanded or focused depending on the type of the lens. The spatial deformation of the probe beam can be detected as the light intensity change through a pinhole at a far field. Since there are several origins of the refractive index change such as the absorption change, density change, and temperature change, we can monitor the dynamics of these properties.

III. EXPERIMENT

The schematic experimental setup is shown in Fig. 2. The fundamental pulse (775 nm and ~150 fs) of the integrated Ti:Sapphire amplified laser system (Clark-MXR, Inc., CPA-2001) was used for a pump beam. The excitation beam was focused in a sample (silicate glass) by a 20× microscope

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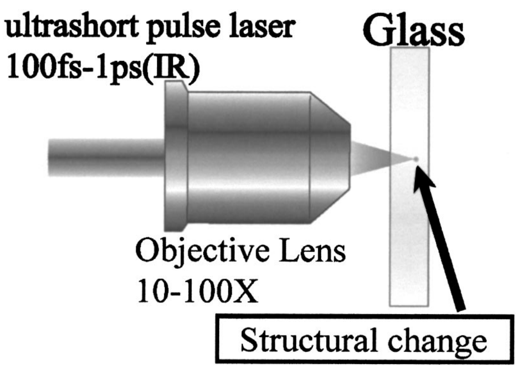

FIG. 1. The structural change inside a glass induced by tightly focused ultrashort IR pulsed laser light.

objective lens. A part of the fundamental light was used to produce the light of the second harmonic (388 nm) by a BBO crystal, and used as a probe beam. The UV light was temporally delayed by an optical delay line and put into the objective lens collinearly with the pump beam. A He–Ne laser was used for a purpose of alignment of the laser beams and the samples. The probe beam extended by an objective lens was collimated by a lens ($f=60$ mm), and the center of the probe beam was passed through an aperture of about 1 mm diameter. After separating the transmitted probe and pump beams by a prism, only the probe beam was detected by a photomultiplier. The detected signal was averaged by a boxcar integrator and recorded by a computer. The sample was moved by a stepping motor driven *XZ* stage with a speed of 1 cm/s to prevent multiirradiation at the same spot.

IV. RESULTS AND DISCUSSION

The TrL signals, after focusing the subpicosecond laser pulse into a silica glass, are shown [in Fig. 3(a), short-time range; Fig. 3(b), long-time range]. The increase and decrease of the probe light intensity indicate the focusing and expansion, respectively, of the probe beam due to the pump laser-induced lens effect. Since the pump and the probe beams were focused by the same objective lens, the expansion of the probe beam does not necessarily mean the creation of the concave lens. The spatial deformation of the probe beam depends on the type of the lens as well as the relative focal position of the pump and probe beams inside the sample. We examine the type of lens by monitoring the probe beam profile sufficiently after the pump pulse irradiation based on the fact that, under the present condition, it is known that the created permanent lens is a convex lens.[6] We found that the beam expansion corresponds to the concave lens creation.

In a short-time range [Fig. 3(a)], the signal decreases after irradiation, i.e., the pump pulse creates the concave lens inside the glass. The temporal shape of this component is nearly the same as that of the pump pulse, which means that the response of this component is very fast (<500 fs). After this signal, an oscillation with a frequency of a few GHz appears [Fig. 3(b)]. Initially light intensity decreases indicating the creating of a concave lens, and the convex lens be-

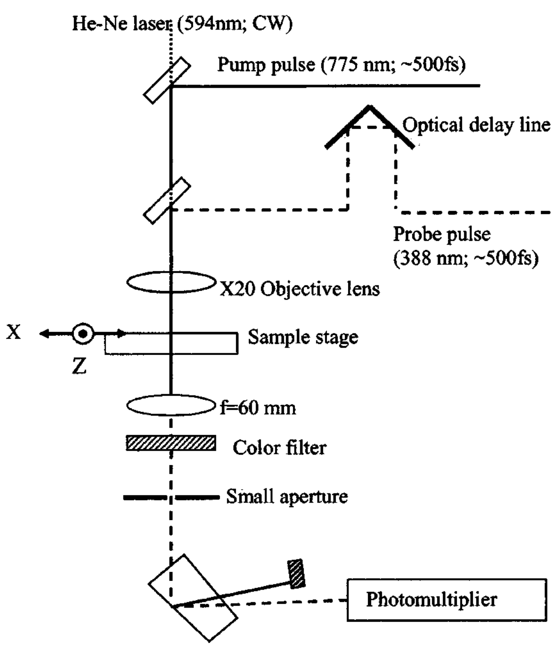

FIG. 2. Schematic illustration of the experimental setup of the TrL method. The pump and probe beams are put into an objective lens collinearly. The pump beam transmitted through the sample was blocked by a color filter and separated from the probe beam by a prism. Only the center of the probe beam is passed through an aperture and detected by a photomultiplier. A He–Ne laser was used for laser alignment purpose.

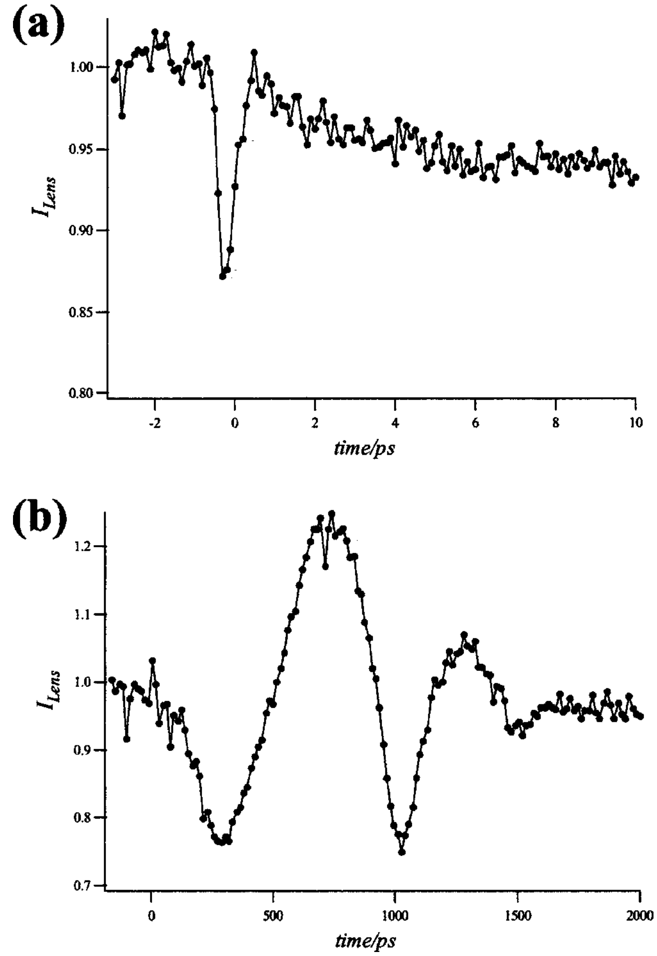

FIG. 3. The TrL signals after irradiation of focused fs-laser pulse inside a silicate glass, (a) in a short-time range, and (b) in a long-time range. The power of the pump beam was 1.45 μJ/pulse, and the polarization of the pump and probe was perpendicular. The signals were normalized by the light intensity without the pump pulse. Upper direction indicates the creation of the convex lens.

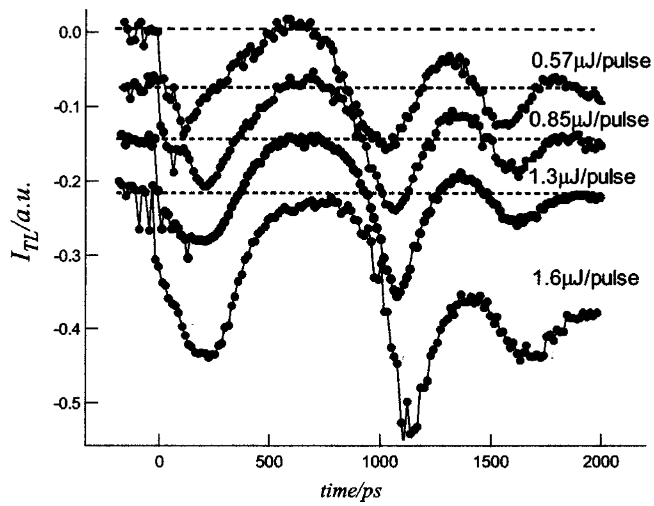

FIG. 4. The TrL signals with various pump power. The peaks of the signals are delayed as the pump power increases. Baseline of the signal is shifted vertically to prevent overlap of the signals.

comes apparent at around 600 ps after excitation under this experimental condition. The oscillation is not a single frequency oscillation but it contains several frequencies in the GHz region.

The exact origin of this oscillation is not clear at present. However, we may suggest that the oscillation indicates a pressure wave created in the focused region. If we assume that the sound velocity in the glass is 5×10^3 m/s, the 700 ps period corresponds to the sound wave traveling distance of about 3.5 μm. Let us estimate the approximate temperature rise at the focal region. If half of the input photon energy is absorbed by the multiphoton excitation in the focal volume, the temperature increase is estimated about 6000 K.[13] Considering that the glass transition temperature of the silica is not more than 1500 K,[13] the material at the focal region should be melted or at least turned to be very soft. The concave lens creation at the initial part of the signal could suggest, that initially, the material in the focal region thermally expands and the pressure wave propagates to the outside from the center. The pressure wave may be reflected back on the interface between the heated liquidlike region and solid region. This reflection may create the observed oscillation.

The pump power dependence of the TrL signal in the long-time range is shown in Fig. 4. The frequency of the oscillation decreases as the pump laser power increases. Based on the this interpretation, the power dependence of the frequency may be explained by the fact that the flexible region around the focal point becomes larger when the pump power becomes high. This vibration feature suggests that the mechanism of the fs-laser-induced structural change inside solid materials is difficult to explain from that of the laser ablation at the surface or UV-induced structural change. In order to elucidate the mechanism of the light-induced structural change in more detail, other experiments should be conducted using various glasses under various experimental conditions.

In summary, we measured the TrL signal after focusing the subpicosecond intense laser pulse inside a glass for the first time, and observed an oscillating signal with GHz frequencies. We tentatively attributed the oscillation of the lens to bounding back and forth of the acoustic wave created by the local heating at the light focused region.

[1] B. C. Stuart, M. D. Feit, A. M. Rubenchik, B. W. Shore, and M. D. Perry, Phys. Rev. Lett. **74**, 2248 (1995).
[2] K. M. Davis, K. Miura, N. Sugimoto, and K. Hirao, Opt. Lett. **21**, 1729 (1996).
[3] E. N. Glezer, M. Milisavljevic, L. Huang, R. J. Finlay, T. H. Her, J. P. Callan, and E. Mazur, Opt. Lett. **21**, 2023 (1996).
[4] H. B. Sun, V. Mizeikis, Y. Xu, S. Juodkazis, J. Y. Ye, S. Matsuo, and H. Misawa, Appl. Phys. Lett. **79**, 1 (2001).
[5] K. Miura, J. Qiu, T. Mitsuyu, and K. Hirao, Opt. Lett. **25**, 408 (2000).
[6] K. Hirao and K. Miura, J. Non-Cryst. Solids **239**, 91 (1998).
[7] K. Minoshima, A. M. Kowalevicz, I. Hartl, E. P. Ippen, and J. G. Fujimoto, Opt. Lett. **26**, 1516 (2001).
[8] W. Watanabe, T. Toma, K. Yamada, J. Nishi, K. Hayashi, and K. Itoh, Opt. Lett. **25**, 1669 (2000).
[9] C. B. Schaffer, A. Brodeur, J. F. Garcia, and E. Mazur, Opt. Lett. **26**, 93 (2001).
[10] J. W. Chan, T. Huser, S. Risbud, and D. M. Krol, Opt. Lett. **26**, 1726 (2001).
[11] M. Terazima, Opt. Lett. **20**, 25 (1995).
[12] M. Terazima, J. Chem. Phys. **105**, 6587 (1996).
[13] R. Bruckner, J. Res. Natl. Bur. Stand., Sect. C **5**, 123 (1970).