Microstructure on Surface of $LiNbO_3$:Fe Induced by a Single Ultra-Short Laser Pulse

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A tightly focused femtosecond Ti:sapphire laser pulse is used to initiate micro-explosions on the surface and internal to an Fe:$LiNbO_3$ crystal. The resulting structure is morphologically different from that induced in an isotropic sample such as fused silica. The regular pyramid and irregular pyramid craters on the surface of the sample are produced at different positions of focal points. The craters suggest vaporization of materials in the process of micro-explosion due to the expansion of high temperature plasma. The embossment pyramids on the surface present the dynamical process of large volume material displacements under high temperature and pressure, and recrystallization of anisotropy crystal materials.

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Ultra-short-laser-pulse-driven micro-explosions provide a unique method for creating microstructures within high bandgap materials with minimal thermal damages. This ability to create three-dimensional objects with sub-micrometre precision may be useful for creating various types of periodic structures, such as diffractive optical elements,[1] photonic bandgap material,[2] pattern gratings in fibres,[3] and three-dimensional data storage.[4,5] The technique is not limited to optically transparent materials, and can also be applied to semiconductors, ceramics,[6] and biological tissues.[7,8]

The micro-fabrication of high-bandgap materials, such as silica glass, diamond, plastics, oxide ceramics, and liquids, usually requires high-intensity light to deposit sufficient energy in the materials, as these materials generally exhibit minimal absorption at low laser intensities. When the incident laser pulse intensity is high enough to induce nonlinear absorption, a very high density of free electron plasma of the order of 10^{18}–10^{20} electrons/cm^{-3} is created. The two mechanisms of multiphoton absorption (dominate for $\tau_p < 100$ fs) and avalanche ionization (dominate for $\tau_p > 1$ ns) are responsible for these laser-induced micro-explosions.[9−11] Since the optical excitation ends before the electrons can transfer their excess energy to the lattice, the temperature of the free electron plasma increases dramatically in a focal volume during optical excitation. Micro-explosions, driven by high pressure and high temperature, can theoretically result in large atomic displacements,[12] densification of materials,[4] and recrystallization[13] within a crystal.

In this Letter, we characterize the structural properties of these micrometre diameter explosions in both fused silica and an Fe:$LiNbO_3$ crystal. In addition, a mechanism is proposed to explain the formation of different structures in the two materials. While the craters formed in fused silica are a relatively featureless empty hole, micro-explosions in the crystal result in the present regular embossed pyramid and irregular cavo-relievo micro-explosions craters on the surface when the focal point is moved from the inner surface to the outer surface. The observed structure strongly suggests that, in contrast to fused silica, micro-explosions in the crystal are highly anisotropic. The displacement and recrystallization of the material occurred in the process of micro-explosions are also highly anisotropic.

Excitation was provided by a home-built chirped pulsed amplified Ti:sapphire laser system[14] (λ = 800 nm, linearly polarized) whose light first passed through a spatial light filter (10 μm-pinhole) to improve the laser beam quality and enlarge its diameter. After being reflected off a dichroic mirror (the 800-nm dichroic mirror permits the light from a light emitting diode to illuminate the sample and to pass through to allow the focus position to be monitored in real time), the beam was focused onto a Z-cut Fe-doped $LiNbO_3$ crystal (0.1 wt.%) by an NA=0.65 objective lens to provide a 0.8-μm diffraction limited spot and with duration of 200 fs and 1 μJ energy of a pulse. The crystal optic axis was aligned parallel to the excitation beam, and perpendicular to the polarization direction of the excitation beam. The crystal was prepared in a cubic shape with six optical surfaces allowing the laser-matter interaction zone to be observed from different orthogonal directions. A computer controlled three-axis translation stage (100 nm resolution at the X direction, 125 nm at the Y direction, and 7 nm at the Z direction) is used to move the sample in each pulse. This is illustrated in Fig. 1. A digital (Nikon)

camera, attached to a conventional phase-contrast optical microscope, was used to observe the crystal after ablation.

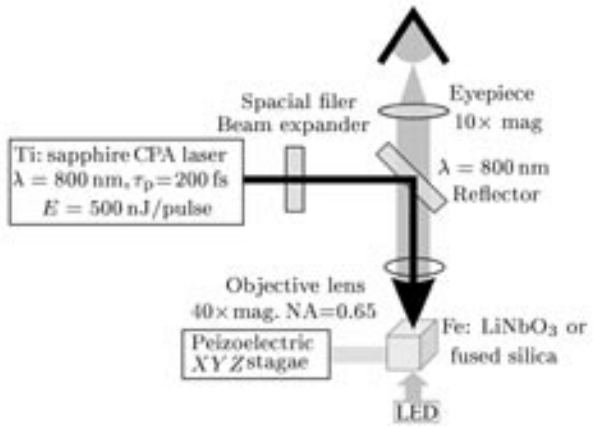

Fig. 1. Schematic diagram of the experimental set-up. The 800-nm dichroic mirror permits the light from a light emitting diode to illuminate the sample and to pass through to allow the focus position to be monitored in real time.

Figure 2 presents a phase-contrast image of the surface of fused silica after a series of micro-explosions have been initialized on the surface. Each bit is the result of a single micro-explosion. There is no debris surrounding the cavity, demonstrating that there is no fluid phase in the process of these micro-explosions. This is because increasing temperature is rapid. The craters are symmetrically distributed in the sample surface. The three-dimensional shapes of the crater are a cone, which is in agreement with Ref. [4].

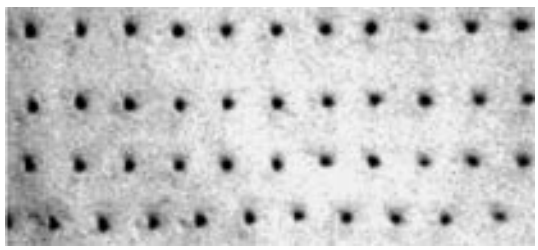

Fig. 2. Craters on the surface of fused silica viewed from the direction of excitation with spacings of 5 μm. The uniform distribution of the micro-explosions are clear in contrast to Fe:$LiNbO_3$.

For comparison, we present the morphologies of micro-explosions on the surface of the Fe:$LiNbO_3$ crystal in Figs. 3 and 4. In Fig. 3, the focus of excitation beam is within the sample ($\sim$3 μm beneath the surface). Similar to the case of fused silica, there is no debris surrounding the craters. However, the cross-section of the cavity perpendicular to the excitation light is not clearly circular. In Fig. 4, the beam is focused outside the sample ($\sim$3 μm). The embossed pyramid presents very clear image of material displacement and recrystallization during the femtosecond-pulse-driven micro-explosions. The ablation craters on the surface of the $LiNbO_3$ crystal are similar to the configuration of a pyramid, which is associated with the trigonal system of the 3-m point group in which $LiNbO_3$ is included.

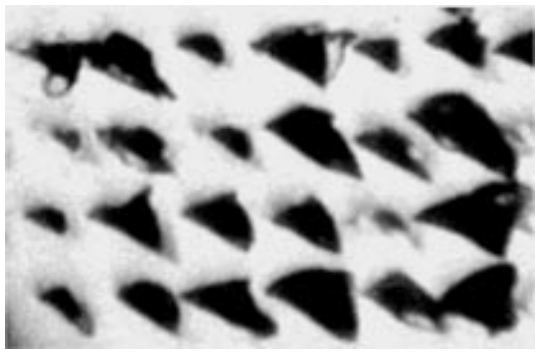

Fig. 3. Craters on the surface of an Fe:$LiNbO_3$ crystal when the laser focal point is within the sample (3 μm below the surface, and the beam waist $w_0 = 0.8$ m and Raleigh focal range $Z_R = 5.4\,\mu$m) as viewed from the direction of excitation pulse. Each crater is located at 5 μm away from its nearest neighbours. The incident energy is 1 μJ. Laser instability is responsible for the non-uniform size of craters. The area of refractive index change is clearly anisotropic.

Fig. 4. Image of the recrystallized material found on the surface of Fe:$LiNbO_3$ when the laser is focused in the air (3 μm above the sample) as viewed from the direction of excitation.

An extremely intense femtosecond laser pulse can ionize atoms and can generate a high density free electron plasma. After formation of a plasma by electronic excitation inside the medium, energy transformation from the plasma to the lattice of the solid material leads to a melting and vaporization.[15] The evaporation of Fe:$LiNbO_3$ on the surface can form the ablation craters and can drive micro-explosions in the material interior.

When the focus is beneath the surface of Fe:$LiNbO_3$, micro-explosions only occur at the inner surface of the crystal. High temperature of the free electron plasma leads to the material expansion within the focal volume. This expansion produces a pressure of 10^{10} Pa, which leads to 20–50% increase in density to silica glass[16,17] around the area of the microcav-

ity. When these micro-explosions occur near the sample surface (within $\sim 3\,\mu$m), a shock wave from this high pressure pushes a thin layer of the solid material towards the surface along the excitation light beam. Due to the interaction time is so short that the gaseous materials moving to the surface solidifies and the solid material recrystallizes instantaneously on the surface. A structure is generated on the surface as shown in Fig. 3. Trachenko *et al.*[12] also discussed theoretically inducement by high pressure of structural relaxation, atomic displacements, and densification in silica glass.

In Fe:$LiNbO_3$, the craters are not conical hole, unlike those induced by the same laser source on the surface of fused silica, but irregular pyramid craters. The shape of micro-explosions depends on the confined plasma channel in isotropic materials such as fused silica. If the same model fits for anisotropic crystals, one would expect that the distribution of electron density in the plasma would be ellipsoidal, implying the distribution of laser intensity at the focal point. This indicates that the model needs to be modified for anisotropic materials. We suggest that two mechanisms are responsible for the pyramid craters. One may be anisotropic photoelectron and absorption of photons. This is due to the angular distribution of photoelectrons excited by polarization light, which is similar to the isotropic solid state materials discussed by Kazansky *et al.*[18] The electron density moving in the polarization direction of the incident laser pulses is higher than that in the perpendicular direction, and that in the negative electronic-field direction is higher than that in the positive direction. The fact that the absorption is related to the electron density also leads to the anisotropic absorption. Furthermore, anisotropic absorption deteriorates the nonuniform of electron density. As a result, the cross section of refractive-index change presents an elliptical configuration in isotropic media such as silica glass, after being, etched by hydrofluoric acid.[19] However, it is difficult to see clearly anisotropy in fused silica without acid, etch. In contrast, we use the same laser source to perform the same experiment in fused silica, craters on the surface is regular cones, as shown in Fig. 2.

The second mechanism may be related to the large bulk converse piezoelectric effect associated with the asymmetry of the $LiNbO_3$:Fe crystal lattice and the polar character of trigonal system in the $3\,m$ point group of the crystal. When exposed to high-intensity laser pulses, the intense laser electronic field leads to a converse piezoelectric effect in the focal volume. The elements of the strain tensor are linearly related to the electric field components and associated with the third-rank tensor of the $3\,m$ point group of the crystal.

The exact focal position of the beam with respect to the sample surface influences dramatically the size of pyramids. As the focal point is moved away from the sample, embossed pyramid and irregular pyramid craters come out on the surface of the $LiNbO_3$:Fe crystal. In addition, the size of the structure observed becomes smaller. When the focal point is placed within the crystal ($> 10\,\mu$m from the surface), a long filament is formed (as previously reported in Ref. [12]). It should be noted that, in our system, the stability of laser is responsible for non-uniform of size of pyramids observed as our XYZ translation stage has a Z-resolution of 7 nm. Good quality materials and a high stability femtosecond laser pulse source should result in uniformly sized pyramids on the surface of the samples.

In conclusion, we have observed that tightly focused 200-fs laser pulses initiate micro-explosions on the surface of an Fe:$LiNbO_3$ crystal. In contrast to isotropic materials such as fused silica, a distinct structure is seen in the cavities and recrystallization areas in the crystal. Embossment pyramid and anisotropy micro-explosions created craters are formed on the surface of the $LiNbO_3$:Fe crystal. The exact structure of the surface depends on the focal position of the beam relative to the sample surface. Micro-explosions beneath the sample surface with several micrometres present a micrometre-order material displacement and recrystallization of the material. The craters suggest that an explosive laser ablation on a crystal may be characterized by anisotropy dependent on the group of the crystal.

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