

## Thermal poling of glass modified by femtosecond laser irradiation

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Thermal poling of silica glass modified by femtosecond laser irradiation is demonstrated. Increase of second-harmonic generation in the irradiated regions is observed. This enhancement is interpreted in terms of structural modifications in silica glass that make the poling process more efficient. Evidence of a change in the distribution of the electrostatic field frozen in glass during poling is obtained. This technique is used for  $\chi^{(2)}$  grating fabrication. © 2002 American Institute of Physics. [DOI: 10.1063/1.1504181]

Thermally poled silica glass is an attractive material for applications in nonlinear optics. Silica glass is widely used in optoelectronics because of its excellent optical properties, and thermal poling offers a simple and reproducible way to induce permanent second-order nonlinearity in glasses.<sup>1</sup> The poling process relies on the creation of a large electrostatic field ( $E_{dc}$ ) inside the glass when heated ( $\sim 300^\circ\text{C}$ ) with a high voltage applied ( $\sim 4\text{ kV}$ ), and then cooled down to room temperature with the voltage still applied. The effective second-order nonlinearity is induced through a third-order nonlinear optical process:<sup>2</sup>  $\chi^{(2)} = 3\chi^{(3)}E_{dc}$ . The low level of second-order nonlinearity achieved so far in silica glass ( $\chi^{(2)} \sim 1\text{ pm/V}$ ) represents the major drawback of this technology, but efficient quasi-phase-matching (QPM) in periodically poled silica fibres would compensate for this limitation and lead to devices for frequency doubling in lasers, switching and generation of correlated photon pairs by parametric process for quantum cryptography.<sup>3</sup> Obtaining a high contrast and microscale resolution in  $\chi^{(2)}$  gratings are critical issues for QPM. Various techniques have been used for  $\chi^{(2)}$  grating fabrication, including periodical electrodes,<sup>4</sup> erasure by UV irradiation,<sup>5</sup> femtosecond laser irradiation,<sup>6</sup> or electron beam exposure.

Femtosecond laser sources have been recently used in order to directly write microstructures deep within transparent media.<sup>7</sup> Micrometer spatial resolution is obtained while tightly focusing the beam within the bulk of the material. In the volume where the beam is focused a permanent material modification is induced through a multiphoton absorption process. High intensities create damage, such as voids, that are used for data storage applications.<sup>8</sup> A refractive index modification can be also induced, and waveguides or gratings can be fabricated by translating the material with respect to the focal region.<sup>9</sup>

In this letter we propose a technique for  $\chi^{(2)}$  grating fabrication. Instead of periodically erasing a uniform second-order nonlinearity, we obtain a  $\chi^{(2)}$  modulation by first periodically modifying the glass with femtosecond laser irradiation and subsequent poling. A  $\chi^{(2)}$  grating of  $20\text{ }\mu\text{m}$  period is fabricated using this technique, and the irradiation is proven to enhance the second-harmonic generation (SHG).

A refractive index grating of  $20\text{ }\mu\text{m}$  period and  $4\text{ }\mu\text{m}$  line width is written within a fused silica glass plate (Heraasil 1 by Heraeus) by scanning ( $v = 200\text{ }\mu\text{m/s}$ ) with the focused beam produced by a regeneratively-amplified mode-locked Ti:Sapphire laser operating at  $\lambda = 850\text{ nm}$ ,  $150\text{ fs}$  pulse duration, and  $250\text{ kHz}$  repetition rate. The energy is  $1.2\text{ }\mu\text{J}$  per pulse and the polarization is perpendicular to the writing direction, that is, to the grating lines. The grating is written in the bulk of the glass with its plane parallel to the surface of the silica plate. The silica plate is polished until the surface reaches the grating. Thermal poling is then carried out in air for  $15\text{ min}$  at  $280^\circ\text{C}$  with  $4\text{ kV}$  applied. The short poling time, together with the polishing, ensures overlapping between the grating and the nonlinear layer created by the poling, the latter usually being only a few microns thick. After poling, SHG in the poled glass is investigated using a mode-locked and Q-switched Nd:YAG laser ( $\lambda = 1064\text{ nm}$ ), with a focused spot size of  $w_0 = 4.1 \pm 0.4\text{ }\mu\text{m}$ , smaller than the spacing between two grating lines. The polarization of the pump beam is controlled by means of a half-wave plate.

The experimental setup is described in Fig. 1. The silica plate containing the grating is mounted on a rotation stage and placed in the focus of the Nd:YAG laser beam. A step-motor translation stage is used in order to scan the beam across the grating. The second-harmonic (SH) light generated in the poled region is detected by a photomultiplier (PMT) and is recorded by an oscilloscope. Simultaneously, by means of a beam splitter placed in front of the PMT, the SH light spot is imaged on a CCD camera connected to a computer for analysis. A white light beam collinear with the laser beam is also used in order to overlap, in the plane of the CCD camera, the image of the grating with the image of the SH light spot, thus allowing one to assess whether the SH light is generated outside the grating, on a grating line or in between two lines. We probe the sample with either the grating lines parallel or perpendicular to the plane of incidence and with either  $s$ -polarized or  $p$ -polarized pump beam (see Fig. 2).

First, before poling, the sample is tested to characterize structural modifications induced by femtosecond laser irradiation.<sup>10,11</sup> The photoluminescence (PL) spectrum is recorded by pumping with an He-Ne laser ( $\lambda = 632.8\text{ nm}$ ), while scanning the beam perpendicular to the grating lines.

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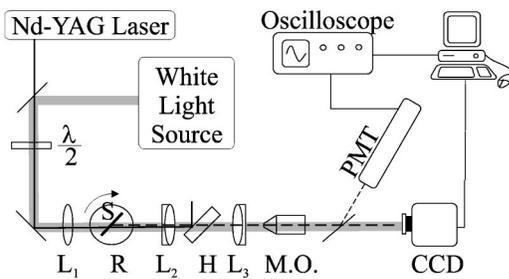


FIG. 1. Experimental setup for measuring and imaging the SH light generated from the sample (S). Paths of the pump beam (1064 nm) and the SH light are indicated by thin and dashed lines, respectively. White light illumination used for imaging the sample is indicated by a shaded gray stripe.  $L_1$ ,  $L_2$ ,  $L_3$ : lenses;  $R$ : rotation stage;  $H$ : harmonic separator;  $M.O.$ : microscope objective;  $PMT$ : photomultiplier tube;  $CCD$ : charge coupled device.

The PL intensity peak at 1.9 eV (655 nm) is associated with nonbridging oxygen hole centers (NBOHC).<sup>12</sup> The extremely strong PL signal observed on the grating lines (Fig. 3, top) indicates that a large number of these defects is induced by irradiation. No SH light is detected before poling.

After poling, a well-defined modulation of the SH light, having the same period as the grating, is observed, while scanning the  $p$ -polarized pump beam across the grating lines oriented parallel to the plane of incidence (Fig. 3, bottom). In the region well away from the grating SH light is generated, as expected for uniformly poled regions, with a well-defined Gaussian spot [Fig. 4(a)]. A significant enhancement in the SH intensity is observed when the pump beam is focused on the grating lines [Figs. 4(b) and 4(d)]. The average enhancement is of a factor of 2 compared to the poled region well away from the grating. Remarkably, the SH intensity generated when the pump beam is focused between two lines [Fig. 4(c)] has the same level as outside the grating. With the  $s$ -polarized pump beam focused on a grating line no SH light is generated on that line, but, quite surprisingly, two lobes are observed, one on each side of the line [Fig. 4(e)]. The diffraction pattern in the far field, although not shown here, presents a gap in the center, suggesting that the two lobes are  $\pi$ -out of phase. Testing the sample in a different geometry, that is, by scanning across the grating with the plane of in-

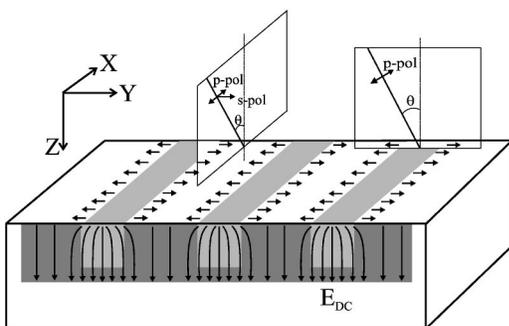


FIG. 2. Schematic of the sample and geometry used for SHG tests. The regions of the glass modified by femtosecond laser irradiation before poling (i.e., the grating lines) are indicated in light gray. The poled region, lying underneath the anode side (top surface), is indicated in dark gray. In the poled region the field lines of the electrostatic field frozen in glass during poling ( $E_{dc}$ ) are sketched. The frozen-in field has components along  $z$  and  $y$  directions (the latter is symbolized by arrows on the top surface). For SHG tests, the plane of incidence is either parallel or perpendicular to the grating lines and the pump beam is either  $p$ -polarized or  $s$ -polarized.

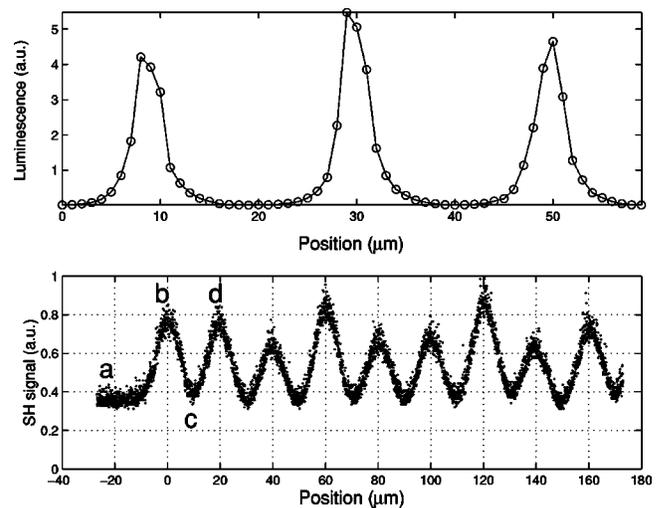


FIG. 3. Upper graph: PL (1.9 eV) intensity recorded before poling while scanning a He-Ne beam across the sample. The intensity peak occurs when the beam position is on a grating line. Lower graph: SH signal recorded after poling while scanning the Nd:YAG pump beam (1064 nm) across the sample. Labels indicate beam positions: outside the grating (a), on a grating line (b) and (d), in between two lines (c).

cidence perpendicular to the grating lines and with a  $p$ -polarized pump beam, a bright SH spot is visible on the left side of each line if the incident angle is positive, whereas the spot is on the right side if the incident angle is negative (Fig. 5).

The enhancement of SHG when the pump beam is upon a grating line gives evidence that the poling process is more efficient due to the structural modifications in the irradiated glass. There are indications that  $E_{dc}$  is higher in the irradiated regions. The presence of a large number of defects such as NBOHC could be responsible for the higher frozen-in electrostatic field<sup>13</sup> and moreover these defects could be oriented and contribute to the enhancement of SHG. There are also indications that the spatial distribution of  $E_{dc}$  is changed by the femtosecond laser irradiation treatment. Assuming the field lines of  $E_{dc}$  are those sketched in Fig. 2, our experimental results can be explained. In the first geometry, with the plane of incidence parallel to the grating lines and with  $p$ -polarized pump beam, the component of  $E_{dc}$  perpendicular to the surface ( $z$ -axis) is probed. Possible distortions of the electrostatic field distribution are expected to lie in the  $yz$

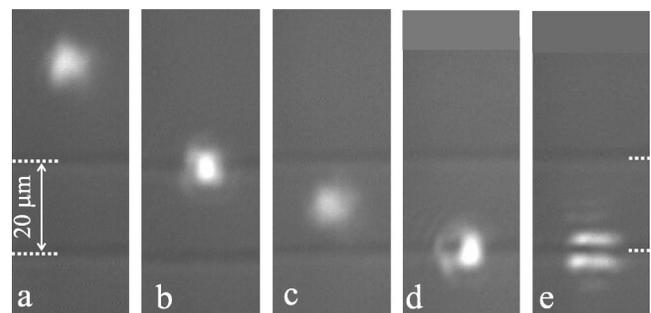


FIG. 4. Images of the SH light spot recorded while probing the sample with the grating lines parallel to the plane of incidence, with  $p$ -polarized pump (a to d) and  $s$ -polarized pump (e). The incident angle is about  $40^\circ$ . The position of the pump beam in images (a) to (d) corresponds to labels (a) to (d) in the lower graph of Fig. 3. The position of the pump in image (e) is the same as in (d). Two grating lines are visible (dashed line).

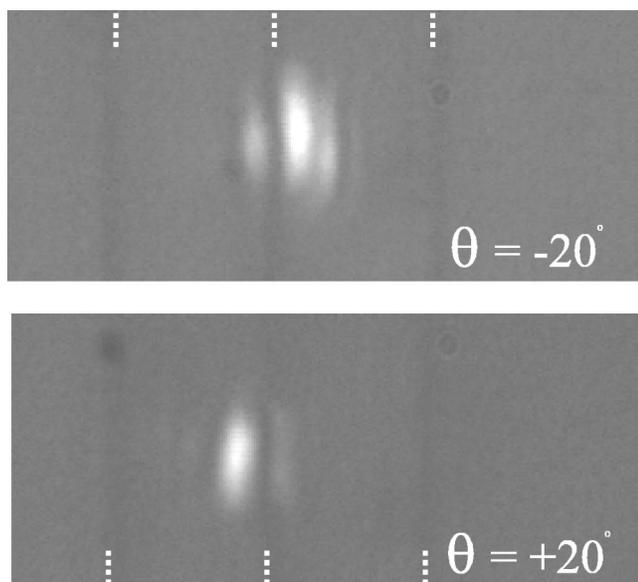


FIG. 5. Images of the SH light spot recorded while probing the sample with the grating lines perpendicular to the plane of incidence and with  $p$ -polarized pump, at incident angles of  $+20^\circ$  and  $-20^\circ$ . Three grating lines are visible (dashed lines).

plane from symmetry considerations and therefore cannot give any contribution to SHG in this geometry. The observed enhancement of the SHG when the beam is focused on a grating line is due to a higher electrostatic field in the irradiated regions [Figs. 4(b),4(d)]. In the same geometry, but with  $s$ -polarized pump beam, the  $Z$ -component of  $E_{dc}$  does not give any contribution to SHG and there is no SHG on a line, as observed [Fig. 4(e)]. On the other hand, distortions of the field lines introduce a component of  $E_{dc}$  along the  $Y$ -direction, which contributes to SHG and accounts for the observed two-lobe pattern [Fig. 4(e)]. The fact that the two lobes are  $\pi$ -out of phase, as indicated by the far-field distribution, reveals that  $E_{dc}$  field lines have opposite directions at the two sides of the grating line. Such a conclusion is reinforced by the measurement in the second geometry, that is, with the plane of incidence perpendicular to the grating lines and  $p$ -polarized pump beam. In this geometry, while probing the grating at an angle, the field lines are more aligned with the direction of the pump polarization on one side of the

grating line than on the opposite side. Therefore, a brighter SH spot is expected on the side where the field lines are more aligned. The opposite situation is expected when reversing the angle of incidence (Fig. 5).

In conclusion, we have demonstrated a technique for  $\chi^{(2)}$  grating fabrication that involves femtosecond laser irradiation of the sample with subsequent poling. Exposure of the glass to the focused beam of the femtosecond laser makes it possible to achieve structural modification in the glass with microscale resolution. The technique has been used for the fabrication of a  $\chi^{(2)}$  grating with a  $20\ \mu\text{m}$  period. Enhancement of the SHG has been observed in the irradiated region. Further experiments to clarify the mechanism are in progress. This technique may find applications in QPM devices where microscale periodic structuring are required.

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