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Double fitting of Maker fringes to characterize near-surface and bulk second-order nonlinearities in poled silica

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An experimental analysis of the distribution and thickness of the bulk nonlinearity induced in poled silica is reported. The second-order susceptibility decreases exponentially from the anodic interface. Maker fringe patterns showing a double structure are interpreted in relation to the presence of two nonlinear profiles, one concentrated near the anodic surface and another extending into the bulk of the sample. The Maker fringe theory is properly generalized and a double fitting technique reproducing well the experimental results is used to characterize the induced nonlinearities. The dependence of the second-harmonic signal on the poling temperature is given, which is different from that of sol-gel silica. © *2000 American Institute of Physics.* [S0003-6951(00)02923-5]

Glass is a main material used in optics and optical communications and so progress in the knowledge of its properties produces important applications. Particular attention is devoted to the possibility of inducing large and permanent second-order nonlinearities and one of the investigated techniques is thermal poling in silica-based glasses.[1–4] The observed second-harmonic generation (SHG) is usually attributed to a second-order susceptibility located in a thin near-surface region of about 10 μm at the anodic side of the poled plate (near-surface effect).[1–3] Evidence of nonlinear regions on both the anodic and cathodic surfaces without contribution of the bulk of the sample has also been reported for similar conditions.[5] In other cases, SHG in poled silica has been associated with a bulk nonlinearity extending across the sample thickness (bulk effect)[6,7] and its coexistence with the near-surface effect has been pointed out.[7] Different models have been introduced to explain the near-surface effect on the basis of depletion,[1,8] screening,[5] and accumulation[9] of charges and there is one explanation about the volume effect by the re-orientation of defects in silica.[7]

The Maker fringe analysis[10] is commonly used to determine the active thickness and the nonlinear coefficient in poled silica. The technique has been recently improved for measuring the nonlinearity in a thin layer.[11] In this letter we consider situations in which both near-surface and bulk effects are significant. The Maker fringe theory for nonuniform nonlinearity profiles[11] is properly generalized and a double fitting procedure is used to determine the thicknesses and the ratio between the nonlinear coefficients of both profiles. The distribution of induced bulk nonlinearity is estimated from poled multipiece samples and it is found to be exponential.

We use ultraviolet (UV) grade synthesed silica from Castech, China, in 20 mm diameter plates either 1 or 0.4 mm thick, polished on both sides. The samples include a number of different compositions but specified with less than 2 ppm of Na and K and 1 ppm of Ca. The sample is poled in an oven at 3.5 kV and at a temperature from 130 to 400 °C for 30–60 min. The detecting configuration is similar to Refs. 1–4, with a Q-switched Nd:yttrium–aluminum–garnet (YAG) laser emitting light at 1.06 μm in 10 ns pulses with 10 Hz repetition rate and energy between 0.1 and 0.9 mJ. The laser beam polarized parallel to the incident plane is focussed with a 15 cm focal lens onto the sample.

Figure 1(a) shows experimental Maker fringes obtained by transmission in a 1 mm sample poled at 200 °C and 3.5

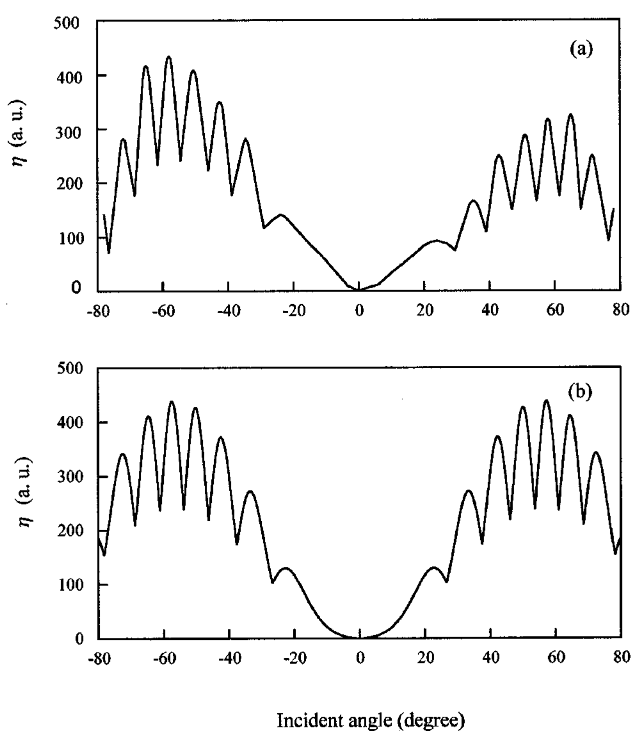

FIG. 1. Experimental Maker fringe pattern (a) and the doubly fitted one (b) for a poled silica sample in which both the near-surface and bulk nonlinearities are significant. The fitting gives the ratio $d_{33v}/d_{33s}=0.38$.

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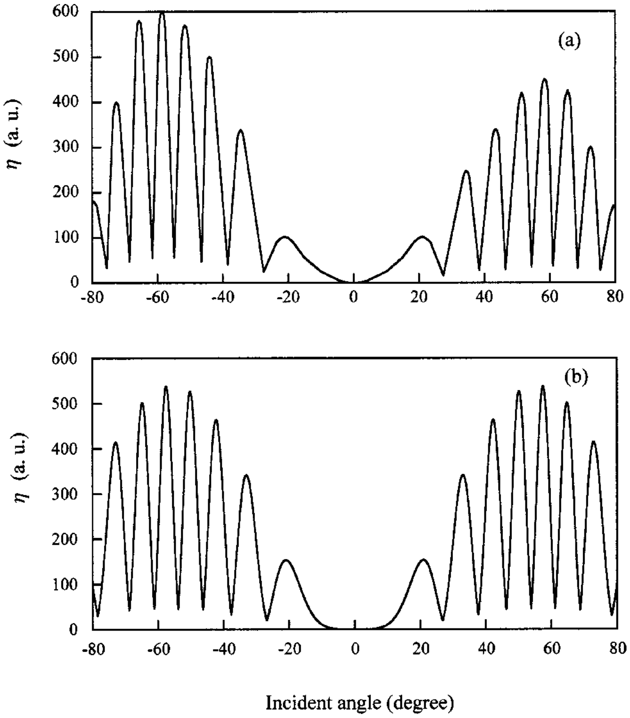

FIG. 2. Experimental Maker fringe pattern (a) and the doubly fitted one (b) for a poled silica sample in which the bulk nonlinearity is the main one.

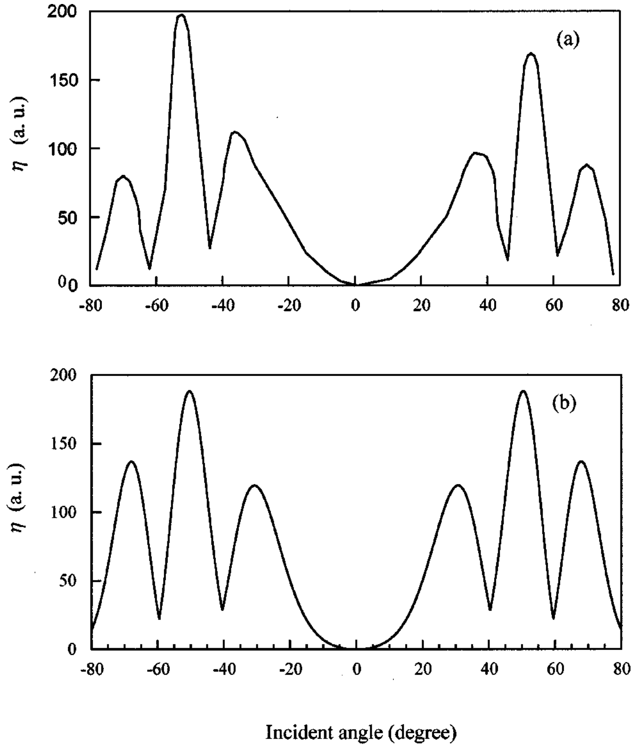

FIG. 3. Experimental Maker fringe pattern (a) and the doubly fitted one (b) for a 0.4-mm-thick sample poled together with other three plates between the electrodes. The results correspond to the plate near the anode.

kV for 30 min. As discussed later, our analysis associates the short-period structure with bulk effect SHG and the slowly varying modulation of the fringe minima with near-surface effect SHG. Figure 2(a) corresponds to a sample poled at 280 °C, where the near-surface effect becomes small. The bulk effect seems strongly sensitive to uncontrolled effects during poling but it is always induced in more or less degree.

Before considering the Maker fringe in detail, it is convenient to analyze previously the depth profile of the induced nonlinearity. This has been made by poling together a set of silica plates and then measuring the SH signal from each piece separately. We used sets of four pieces, each of 0.4 mm thick, poled at 280 °C and 3.5 kV for 40 min. The Maker fringe shown in Fig. 3(a) correspond to the plate in contact with the anode during poling and thus they are due to both near-surface and bulk effects. In contrast, the SHG from the other plates is mostly due to bulk effect with similar patterns, and the SH signal decreases with distance and the ratios between fringe maxima for the successive pairs of consecutive plates are 2.5, 2.1, and 1.9, respectively. By assuming a bulk nonlinearity with exponential profile across the set of plates and considering the SH signal of a given plate as proportional to the square of the nonlinear coefficient, the observed decrease for the SHG of successive plates means that the profile has a $1/e$ thickness of 1.15 mm. On the other hand, by applying differential chemical etching at the anodic side of a sample with noticeable near-surface effect, we also estimate an exponential fall off for the near-surface nonlinearity, in agreement with previous works.[1,4]

The Maker fringe theory for nonlinear crystals[10] has been recently extended for nonuniform nonlinear profiles[11] and we further develop it here to make it suitable to include both bulk and near-surface effects. The external conversion efficiency is written as follows:

$$\eta=\frac{P_2}{P_1{}^2}$$

$$=A\,\frac{\sin^2\theta_1}{\cos^2\theta_2}\,\frac{T_2(\theta_2)}{T_1{}^2(\theta_1)w^2(\theta_1)}\,(f_b+f_s+2\,f_b{}^{1/2}f_s{}^{1/2}),\quad (1)$$

where subscripts 1 and 2 denote the fundamental and SH waves, respectively, P_j and θ_j are the power and internal propagation angle of beam j,T_1 is the power-transmission Fresnel coefficient of the fundamental wave through the front surface, and T_2 is the corresponding coefficient for the SH wave at the backsurface. A is a constant factor, w^2 describes the effective cross section of the fundamental beam and the subscripts s and b are associated with the anodic surface and bulk nonlinearities, respectively. The functions f_b and f_s enclosed in the brackets describe the phase mismatch accumulated by the two waves across the sample and have the following integral expression:

$$f_j=\left|\int_0^L d_{33j}(z)e^{i\Delta kz}dz\right|^2,\qquad (2)$$

where the subscript j stands either for the subscript b or s, $d_{33j}(z)$ is the nonlinear coefficient and Δk is the phase mismatch between the fundamental and SH waves in the nonlinear medium. By assuming exponential profiles for both nonlinear coefficients $d_{33s}(z)$ and $d_{33b}(z)$, with $1/e$-thicknesses L_s and L_b, respectively, we obtain

$$f_j=\frac{d_{33j}{}^2}{L_j{}^{-2}+\Delta k^2}\left[(1-e^{L/L_j})^2+4e^{L/L_j}\sin^2\frac{\Delta kL_j}{2}\right],\qquad (3)$$

where d_{33j} denotes the corresponding coefficient at $z=0$, L is the sample thickness, and the phase mismatch depends on the propagation angles as follows:

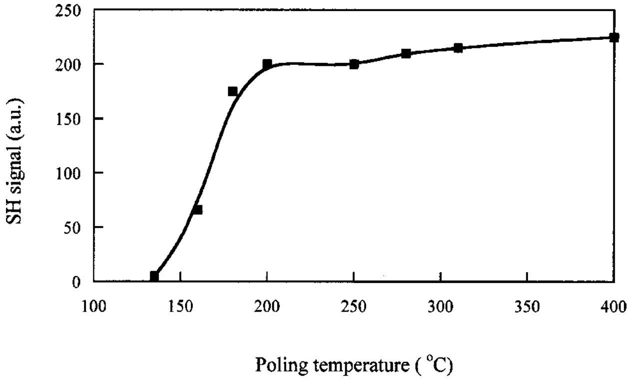

FIG. 4. Dependence of the SH signal on the poling temperature for 1-mm-thick silica samples poled at 3.5 kV for 30 min.

$$\Delta k = \frac{4\pi}{\lambda}(n_1 \cos\theta_1 - n_2 \cos\theta_2), \tag{4}$$

where n_j is the refractive index for wave j.

By fitting the experimental results of Figs. 1(a), 2(a), and 3(a) with Eqs. (1)–(4), with $n_1 = 1.449\,63$ and $n_2 = 1.460\,71$, we obtain the fringe patterns represented in part (b) of each figure, and the values $L_s = 10\,\mu$m, $L_b = 0.955$ mm, and $r = d_{33v}/d_{33s} = 0.94$ for the case of Fig. 1(b), $L_s = 15\,\mu$m, $L_b = 0.955$ mm, and $r = 2.6$ for Fig. 2(b) and $L_s = 21\,\mu$m, $L_b = 0.38$ mm, and $r = 1.6$ for Fig. 3(b). The L_b value is close to the sample thickness in the three cases. The very different depth of the two nonlinear profiles allows us to associate the short-period fringes with the bulk effect. The rising of the envelope of the fringe minima is due to the near-surface effect as well as the exponential distribution of the bulk nonlinearity. If uniform profiles with depth thicknesses L_s and L_b are assumed instead of the exponential distributions, we then have

$$f_j = \frac{d^2{}_{33j}}{\Delta k^2}\left[4 \sin^2 \frac{\Delta k L_j}{2}\right] \tag{5}$$

instead of Eq. (3). Equations (1) and (5) reproduce the experimental Maker fringes as well and in this case the fringe minima envelop would be due to the near-surface effect exclusively.

Figure 4 indicates the dependence of the SH signal on the poling temperature for 1 mm thick silica samples poled at 3.5 kV for 30 min. The curve presents significant differences from that by poling sol-gel silica.[6] The SHG begins at 130 °C, instead of the 150 °C for sol-gel silica, while the pronounced increase up to 200 °C is a common feature, the almost flat dependence for higher temperatures up to 400 °C is in clear contrast with the strong decrease observed in sol-gel silica for temperatures higher than 200 °C.

Electron paramagnetic resonance spectroscopy (Bruker ESP 300 E) has been used to analyze the presence of defect sites in the silica samples, with technical parameters similar to those of Ref. 7 and at temperatures of 300 and 143.4 K, but no evidence of defects has been found. Hence, we have to conclude that the bulk nonlinearity induced in our poled silica is not related to defect sites. Possibly it may be caused by the electric field from charges in the bulk silica, which may also have the layer structure just like those in the near-surface effect.[3,12] The bulk distribution of charges may be strongly dependent on the poling and cooling conditions and so it can explain the high sensitivity to small variation on those conditions.

In conclusion, the SHG in poled silica glasses may exhibit Maker fringe patterns with a double structure and, by using the Maker fringe theory properly generalized to describe the presence of two superposed nonlinear profiles, a really good fitting of the fringe structure is achieved. By poling multiple silica plates together and by using selective chemical etching, we have found decaying profiles with roughly exponential forms for the bulk and near surface effect, respectively. By assuming exponential profiles we have obtained the $1/e$ thicknesses of the two profiles and the ratio between the two second-order nonlinearities at $z = 0$. The double fitting associates the short period with a bulk effect extending across the sample and the rising up of the fringe minima with both near-surface effects at the anodic side of the poled plate and the exponential distribution of second-order nonlinearity. The bulk second-order nonlinearity may be tentatively related to the electric field produced by charges, which may have a layer structure. The sensitivity of this charge distribution to different effects during poling could explain the variable results obtained for about similar conditions. The dependence of the SH signal on the poling temperature for silica is given.

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