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Electric surface potential and frozen-in field direct measurements in thermally poled silica

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We report measurements of the electric surface potential (SP) and its temporal evolution in thermally poled silica samples, thus, providing a direct quantitative evidence of the frozen-in voltage. The SP value is found to be directly related to the voltage across the depletion layer and there is a clear link between frozen-in electric field and nonlinear optical coefficient, inferred from SP and second harmonic generation measurements respectively. Our studies also show the presence of screening surface charge layers. We have calculated the space charge distribution making evident that the thinner the sample, the larger the screening surface charge density. © 2008 American Institute of Physics. [DOI: 10.1063/1.2827175]

Thermal poling of silica glass is an efficient technique for inducing a second order optical nonlinearity¹ (SON) in an otherwise centrosymmetric material and is of great interest for wavelength conversion and electro-optic modulation in silica fibers. Several works²⁻⁵ have concentrated on the actual process lying behind thermal poling, showing how two charged layers form inside the sample and give rise to a frozen-in voltage (V). Efforts have also been made to establish the values of V and of the internal electric field (E_{in}). The latter is directly proportional to V through the relation $V = E_{in}L$, where L is the depletion layer or space charge region thickness in which most of E_{in} is concentrated. The value and optimization of E_{in} in the depletion layer are crucial to maximize the SON. In fact, it is now widely accepted that the nonlinear optical coefficient (d_{eff}) is directly proportional to E_{in} via the third order nonlinear susceptibility ($\chi^{(3)}$),

$$d_{eff} = 3/2\chi^{(3)}E_{in}. \quad (1)$$

So far, the values of V and E_{in} in poled silica have never been directly measured but always inferred through indirect means, such as second harmonic generation (SHG),⁶ electric-field induced SHG,⁷ and differential etching measurements.⁸ In this letter, we investigate the temporal evolution of the surface potential (SP) and its dependence on sample thickness in thermally poled glass. The measured values of SP are close to the external applied voltages during poling, thus, providing a direct evidence of the fact that most of the applied voltage is frozen-in across the depletion layer after thermal poling. Through SP measurements on both anodic

and cathodic sides and taking into account depletion thickness L measured through SHG,^{6,9} we calculate the space charge distribution using a finite element method (FEM). From calculation, we find out that additional screening surface charge layers need to be taken into account to explain the experimental results. In addition, the value of E_{in} and its evolution with poling time are calculated and compared with those obtained from SHG measurement. Despite being large, E_{in} is still about three times smaller than what is required to justify d_{eff} from SHG. This is probably due to a nonperfect "evaporation" of screening charges during reheating, thus, effectively reducing the measured SP and the inferred E_{in} . This effect being larger for thinner samples.

Samples of fused silica¹⁰ of different thicknesses (0.1, 0.5, and 1 mm) were thermally poled over a 1 cm² area in air at 270 °C, 4 kV, and for various poling times: 2, 5, 10, 20, 30, 45, and 90 min. Surface potential measurements were carried out using an earthed field meter (JCI 140CF, John Chubb instrumentation) calibrated against a metal plate of same dimensions (1 cm² area) to which a known potential was applied, as shown as an inset in Fig. 1. The Maker fringes analysis of similar samples has been reported in Refs. 6 and 11. Those previous results will be used for comparison with SP measurements.

We observed a high SP (~1 kV) from all samples in the first minutes after thermal poling. While the SP was still being measured, the sample was cooled down. We then observed a slow decrease in the SP down to negligible values in about 10 min. All samples showed very similar behavior. We believe that this is due to the formation of screening charge layers of atmospheric particles (mainly surface H₂O, i.e.,

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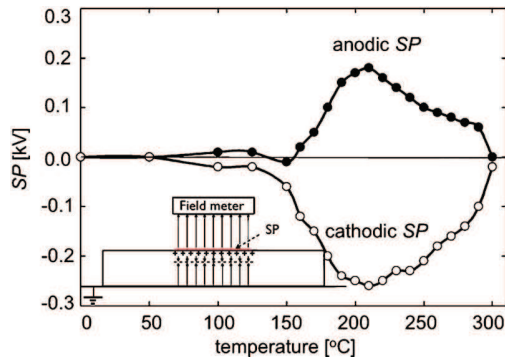


FIG. 1. (Color online) Measured anodic and cathodic surface potentials for a silica slide thermally poled at 270 °C, 4 kV, and 10 min using a field meter. The corresponding setup and geometry for measurements from the anodic side are shown in inset.

H_3O^+ and OH^- , and electrons, depositing on both sample sides). This is also supported by the fact that, while SP died off during cooling, the SHG signal remained constant, thus, indicating that V was not changing. This surface charge neutralization is a well known mechanism, for example in ferroelectrics, such as LiNbO_3 and LiTaO_3 .

In order to be able to remeasure SP, we tried heating up the samples. We increased the temperature in 10 °C steps and repeated the SP measurement for both the anodic and cathodic faces at each step. All samples showed the same behavior. An example is reported in Fig. 1. Below 160 °C, the SP remains zero but above this temperature it begins to increase due to a likely evaporation of the screening surface charge layers. A maximum value was reached for 210 °C. Then SP started to decrease once again, suddenly returning to zero as the poling temperature (270 °C) was reached. Furthermore, the SH, measured at each step, remained constant and only started to decrease above 210 °C, thus, indicating that we started erasing E_{in} , i.e., the internal voltage V . Note that, as expected, positive values of SP are measured from the anodic face and negative values from the cathodic face.

Figure 2 shows that the SP values are functions of both sample thickness and poling time. In fact, values of 1.3 kV for SP have been measured, meaning that a significant fraction of the voltage (4 kV) applied during poling is actually stored inside the sample. The difference between SP and applied voltage is probably due to a nonperfect evaporation of screening charges during reheating as well as to fringe field effects, thus effectively reducing the measured SP. The reduction due to fringe field effects was confirmed by measure-

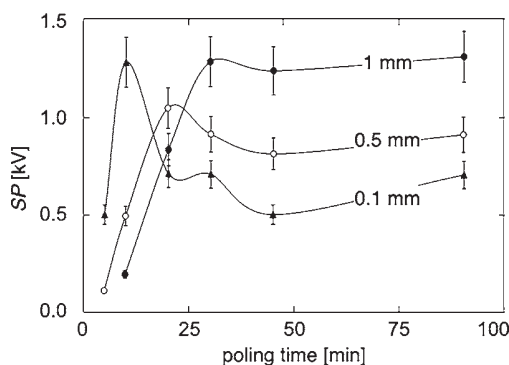


FIG. 2. Anodic SP evolution with poling time for samples of different thickness. The lines are only a guide for the eye.

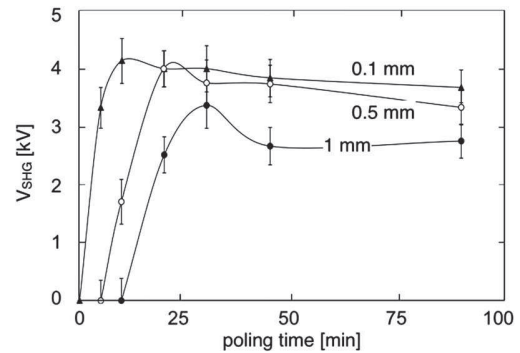


FIG. 3. Calculated V from SHG and its evolution with poling time for samples of different thickness. The lines are only a guide for the eye.

ments performed on samples with a much larger poled area (about 10 cm^2) where the SP value detected was 30% higher (about 1.7 kV).

Figure 3 shows the evolution for the frozen-in voltage V as a function of poling time and thickness measured through SHG Maker's fringes technique (which will be referred to as V_{SHG}). Contrary to an intuitive picture, if one compares SP and V_{SHG} evolutions (Figs. 2 and 3), some clear differences exist, in particular, a reverse trend with respect to sample thickness for poling times larger than 30 min. However, one should bear in mind that SP is measured with samples at a certain temperature while V_{SHG} comes from room temperature SHG measurements. The two situations might, thus, present a different surface charge screening which depends on temperature and poling thickness. Note that in terms of internal field, the SHG measurements provide values up to 1 $\text{kV}/\mu\text{m}$ assuming a $\chi^{(3)} \approx 4 \times 10^{-22} \text{ m}^2/\text{V}^2$, very close to those in Ref. 12 while the SP measurements provide corresponding values up to 0.35 $\text{kV}/\mu\text{m}$.

In order to investigate the differences explained above, we have calculated the charge distribution σ by means of FEM numerical calculation. In this model, the scalar potential function $\phi(x, y)$ is governed by the Poisson's equation as given below

$$\partial^2 \phi(x, y) / \partial x^2 + \partial^2 \phi(x, y) / \partial y^2 = \sigma(x, y) / (\epsilon_0 \epsilon_r). \quad (2)$$

ϵ_r is the relative dielectric constant of either air or silica, equal to 1 or 3.8, respectively, and ϵ_0 is the permittivity of vacuum.

Initially, we have assumed two charge layers, one negative (σ^-) located below the anodic surface at a depth L and one positive (σ^+) on the anodic surface, L being obtained from SHG measurement. From high temperature (hot) SP values on both anodic and cathodic sides, we have calculated the evolution of both σ_{hot}^- and σ_{hot}^+ for different sample thickness and poling time. The value of σ_{hot}^+ oscillates between 10^{-9} and $10^{-8} \text{ C}/\text{mm}^2$ depending on poling time and sample thickness, as reported in Fig. 4. σ_{hot}^- was found to be of the same order of magnitude.

To be consistent with SP measurements performed at room temperature (cold), we have then recalculated σ^+ (σ_{cold}^+) while keeping the same value for σ^- ($\sigma_{\text{cold}}^- = \sigma_{\text{hot}}^-$). In order to get negligible SP on both faces at room temperature, in agreement with what was experimentally obtained, we also had to introduce a very small positive surface charge with a value close to $(\sigma_{\text{hot}}^+) - (\sigma_{\text{cold}}^+)$ on the anodic side (back side), deduced from calculation. Figure 4 shows that the val-

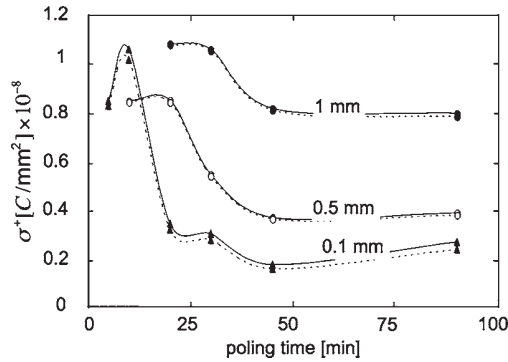


FIG. 4. Evolution of positive charge (σ^+) on anodic surface for different sample thickness and poling time with solid and dashed line corresponding to σ_{hot}^+ and σ_{cold}^+ , respectively.

ues of σ_{cold}^+ are smaller than those of σ_{hot}^+ which indicates the presence of a screening surface charge (σ_s), $\sigma_s \equiv \sigma_{\text{hot}}^+ - \sigma_{\text{cold}}^+$, as mentioned previously. In addition, it may be noted that σ_s is higher for thinner sample thicknesses, as shown in Fig. 5. To be more specific, for 1 mm thick samples, σ_s is $(4-7) \times 10^{-11}$ C/mm², while for 0.1 mm samples the difference is increased about ten times. Consequently, there are more charges that need to evaporate to reveal the frozen-in voltage in terms of SP for thinner samples. This is probably the reason why SP for thin samples, measured in the hot situation, as presented in Fig. 2, are usually smaller than those for thick samples, although the frozen-in voltage measured from SHG in cold situation has an opposite trend (Fig. 3).

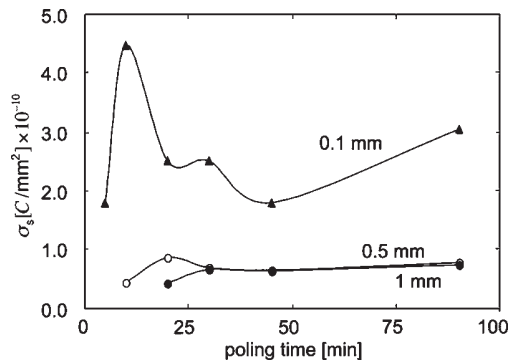


FIG. 5. Evolution of screening charge (σ_s) for different sample thickness and different poling time.

From the room temperature (cold) charge distribution of all samples for different poling time, we have calculated the internal field (E_{in}) in the depletion region. We obtain that the values of E_{in} are ≤ 0.35 kV/ μm , hence, corresponding to a nonlinear optical coefficient d_{eff} , calculated using Eq. (1), smaller than 0.1 pm/V. These values are about three times smaller than the corresponding ones measured through SHG.^{6,11}

In conclusion, direct measurements of surface potential in thermally poled silica samples have been carried out. The inferred internal electric field ($E_{\text{in}} \leq 0.35$ kV/ μm) in the depletion region and the corresponding nonlinear coefficient ($d_{\text{eff}} \leq 0.1$ pm/V) follow the behavior predicted by SHG measurements, provided that screening surface charges are taken into account. The difference in values of E_{in} and d_{eff} between SP and SHG measurements (values from SP about three times smaller than those from SHG) is mainly due to an underestimate of SP as a consequence of nonperfect evaporation of screening charges and partly fringe field effects. This is also in agreement with the opposite trend of SP and V_{SHG} with respect to sample thickness if one bears in mind that the thinner the sample, the higher the quantity of charge that needs to evaporate to reveal the frozen-in voltage in terms of SP.

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