Voltage-assisted cooling:
a new route to enhance $\chi^{(2)}$ during thermal poling

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Abstract:

Modifying the standard constant-voltage poling procedure significantly enhances the in-built electric field strength (and consequently $\chi^{(2)}$) and leads to control of the nonlinear region evolution, both crucial parameters to integrate nonlinearity in waveguiding regions.

Second-order susceptibility induced by the formation of a space-charge region is widely accepted as the macroscopic effect of standard thermal poling, which consists of inducing ionic migration by high static electric field applied across a pre-heated amorphous material and subsequently freezing it with the same voltage still applied [1]. While several studies have investigated how the dynamic SHG behaves in respect to different poling parameters (such as time [2], voltage [3], atmosphere [4]), none to date has been performed when the voltage is increased during the cooling phase. By adopting such a voltage-assisted treatment, we dramatically improve the second harmonic (SH) signal compared to the standard poling procedure, for identical conditions at the start of it; most importantly, the second order susceptibility $\chi^{(2)}$ is doubled when the initially constant voltage is raised up to higher voltage during cooling, as a combined result of both a dampen increase in the depth of the nonlinear region and enhanced frozen-in electric field.

In this work, we report a comparative study between voltage-assisted and standard thermal poling carried out in air on two sets of Herasil1 samples, with different thickness of S=0.2mm and 1.0mm. In order to isolate how applying the voltage differently affects second-order nonlinearities (SONs), both poling time and temperature are set at t_{pol}=10min and T=280°C, respectively. Two pressed-contact silicon plates are used to apply an initial constant voltage (in the range between 2 and 5kV) across the samples. After t_{pol}, either the same or higher voltages are delivered to the electrodes during the cooling time, until the temperature contribution to poling can be considered significant (T>200°C). However, regardless of which mechanism is responsible for creation of the nonlinearity (nonlinear dipole orientation and/or permanent space charge field [5]), increasing voltage during standard poling has led to increase the width (depth) of the nonlinear region though [3] and the nonlinear efficiency could be improved only for optimum poling temperature, as high as 400°C [6].

Firstly, we investigate the spatially resolved evolution of the depletion region when both standard constant-voltage (e.g., 3kV) and voltage-assisted cooling (e.g., from 3kV ($V_{initial}$) up to $V_{cooling}$=4kV) are performed.

Table 1. Nonlinear depth evolution under constant voltage (e.g., 3kV) and voltage-assisted cooling (e.g., 3-4kV).

sample #	voltage (kV)	nonlinear depth (±0.5μm)	
		S=0.2mm	S=1.0mm
1	3	11.2	9.3
2	3 - 4	12.3	10.4
3	3 - 5	13.9	11.8
4	3 - 6	15.1	13.0
5	4	15.7	13.5
6	4 - 5	17.5	15.8
7	5	18.7	16.4
8	2 - 5	12.7	10.6

Table 1 lists the depth of the nonlinear boundary below the anode surface measured by microscope inspection after cross sectional etching in 48% HF for 1min. The ridge observed reflects the edge of the depletion region, where the etching rate is reduced by local field distribution [2]. When standard poling is performed (e.g., 3kV continuously applied), increases in the voltage induce a monotonic increase in the nonlinear region width (40% bigger for 5kV), as expected [3]; the trend is even confirmed in thinner samples, whose deeper nonlinear thicknesses are consistent with the increase of the applied electric field across them [7]. However, if the voltage is raised after t_{pol}=10min (e.g., from $V_{initial}$=3kV up to 5kV during cooling), the nonlinear depth evolves tending to increase less compared to the 5kV constant-voltage. Based on this experimental evidence, we speculate that increasing voltage when the ion mobility decreases during cooling can allow control of the space charge region evolution and thickness, which appeals when referred to waveguiding devices.

Figure 1 maps the depth of the nonlinear region as the standard/voltage-assisted procedures are concerned. The experimental data are reported as closed circle with an estimated error bar of ±0.5µm. For the sake of clarity, the data points on the graph have been connected by solid line as guide to the eye, only.

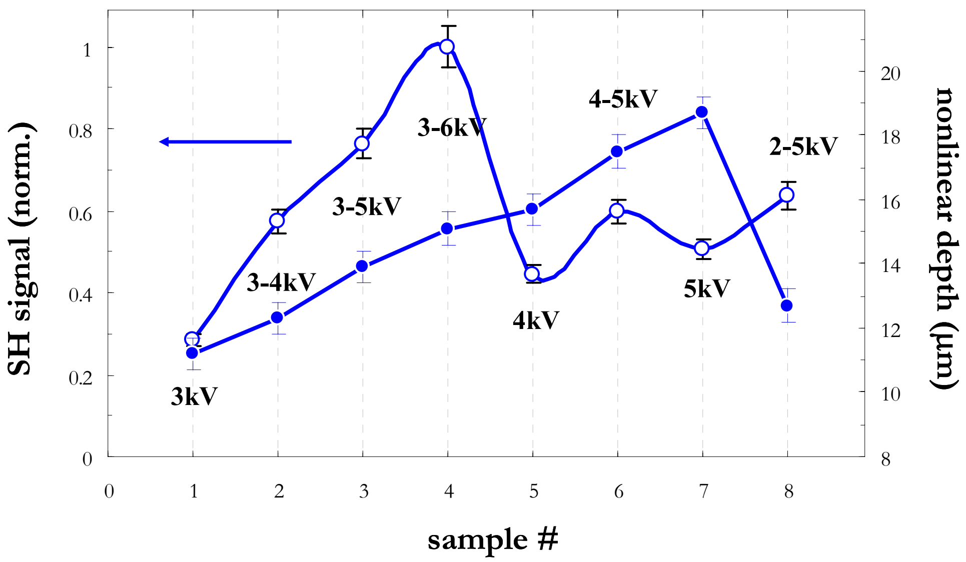


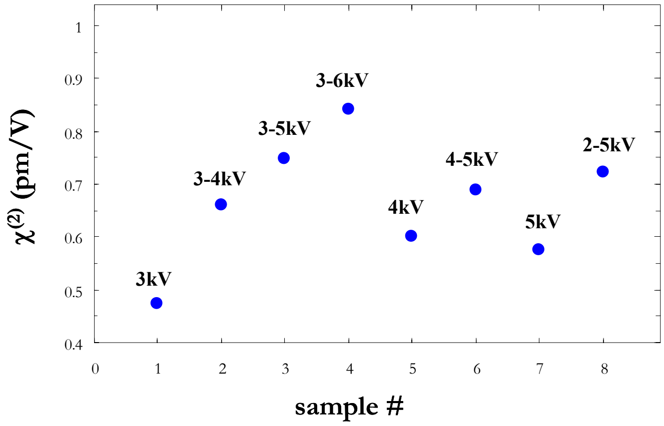
Fig. 1) Poling procedure dependence of SH generation (open circle) and depletion edges (closed circle). Standard poled samples # 1, 5 and 7 are used as a reference.

Solid line in Figure 1 also shows the trend of the normalized SH signal (open circle), which is homogeneously generated by scanning across the sample the p-polarized 1.064µm pump from a mode-locked and Q-switched Nd-YAG laser. During this measurement, the sample is mounted on the standard Maker Fringe apparatus at the incident angle which maximizes the SH generation. It is striking to observe how substantially the SH signal ($P_{2\omega}$) increases with increased voltage during cooling; the same behavior is even confirmed when the initial voltage is as low as 2kV (point 2-5kV in Figure 1).

In the Maker Fringe technique, where the nonlinear thickness w is smaller than the coherence length:

$$\chi^{(2)} \propto \frac{\sqrt{P_{2\omega}}}{w}$$

which implies that the $\chi^{(2)}$ strength is clearly increasing when our modified poling procedure is adopted, since the nonlinear region depth does not change significantly though. By assuming a nonlinearity profile as recently retrieved with an inverse Fourier transform Maker Fringe technique [8] and assuming a nonlinear coefficient peak at about 1µm below the anode surface, we can estimate the nonlinear susceptibility $\chi^{(2)}$ for an effective width of the nonlinear layer when the induced second order nonlinearity in the bulk of the sample is considered negligible.



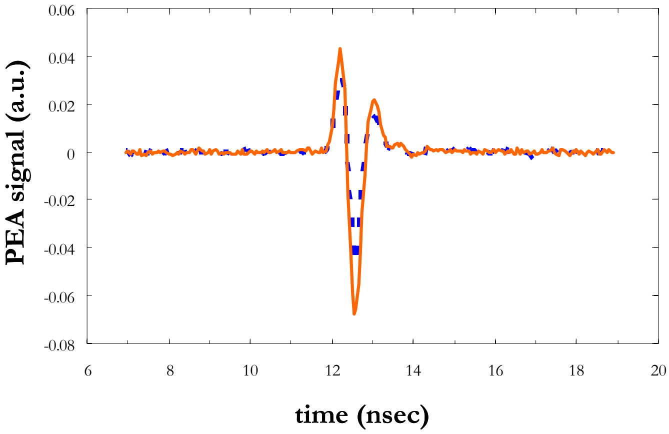


Fig. 2a) Second order susceptibility evaluated by measuring MF curve on differently poled samples. Standard poled samples # 1, 5 and 7 as reference. Fig. 2b) Inferred charge distribution (and electric field strength) by PEA measurements on standard poled sample #1 (dashed line) and voltage-assisted poled sample #3 (solid line)

Figure 2a) shows the calculated $\chi^{(2)}$ values as a function of different poling procedure. For samples poled under a continuous constant-voltage (#1,5,7) the $\chi^{(2)}$ distribution is consistent with previous results [6], whilst for voltage-assisted poled samples a linear trend of SON with the final voltage applied is shown. Furthermore, for identical ΔV ($V_{cooling} - V_{initial}$), the nonlinear coefficient is sensitively influenced by which voltage value ($V_{initial}$) is kept applied during the first t_p=10min of poling.

Indeed, under higher voltages, the interaction among much less mobile carries (H^+, K^+ …) injected at the anode surface into the high field depleted region might concretely modify its charge distribution and improve the in-built electric field strength. Most importantly, as the dielectric breakdown point of the sample increases during cooling, the assisted-voltage is believed to give dramatic benefit to the recorded electric field inside the nonlinear region.

As a confirmation, the space-charge field strength is determined by Pulsed Electro Acoustic (PEA) method. These preliminary measurements are carried out on the sample poled at a constant 3kV (dashed line in Figure 2b) and on sample #3 (solid line in Figure 2b), poled under increased voltage during cooling – from 3 up to 5kV. As a result of applying a high voltage electric pulse of ultrashort duration (2ns) to the poled sample, the space charge inside it is stimulated and experiences a pulse force that travels as an acoustic wave through the sample. By detecting the resultant pressure pulse arriving at a piezoelectric transducer in close contact with one of the electrodes, the space charge distribution in the poled samples can be obtained from the evolution of the output voltage of the transducer itself. The sign of the PEA signal is the same as the sign of the charge. Although the spatial resolution of the PEA system is limited to ~10μm due to the speed of the acoustic wave in silica being equal to 5.95×10^3 m/sec, the PEA method reveals a high potential of exploitation in the study of poling-induce space charge evolution.

In conclusion, voltage-assisted cooling during poling significantly enhances the second order nonlinearity compared to the standard poling procedure. Moreover, we show experimental evidence of control on the nonlinear region depth, which will be exploited to optimize the overlap between nonlinear region and core in periodically poled fibre.

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