

Second-order nonlinearity generation in alkali-glass-based optical fibers

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Abstract—We analyzed the mechanism of alkali-ions migration taking part in obtaining the frozen ionic potential responsible for the creation of second-order nonlinearity in optical fibers made of silica and other oxide-based glasses. We confirmed that the presence of alkali ions in the oxide glass, along with a high $\chi^{(3)}$ value, enables the generation of ionic potential, even though these ions are structural elements of the glass rather than impurities. Creation of second-order susceptibility in glass is possible and, depending on alkali-ion concentration, $\chi^{(2)}$ can be higher than for a silica-based fiber of the same geometry. Calculated $\chi^{(2)}$ reached 0.58 pm/V, and the maximum value obtained for silica-based fiber for the same thermal poling conditions was only 0.33 pm/V.

The induction of second-order nonlinearity in optical fibres, typically composed of centrosymmetric materials, remains a compelling research area in the pursuit of novel nonlinear fiber-based devices. One of the oldest methods for second-order susceptibility $\chi^{(2)}$ creation in glass is thermal poling of fiber glass [1], which allows one to obtain $\chi^{(2)}$ of the order of a fraction or single pm/V [2–5], e.g., for silica-based fibers, it is typically 0.3 pm/V. This phenomenon observed in glasses, and mostly in silica-based optical fibers, is well described in the literature [2–4], and it is based on the relation of third-order nonlinearity $\chi^{(3)}$ of the fiber and frozen-in electric field E_{DC} in the core area, accordingly to the formula:

$$\chi^{(2)} = 3\chi^{(3)}E_{DC}. \quad (1)$$

For centrosymmetric materials, $\chi^{(2)}$ equals zero, whereas $\chi^{(3)}$ has a non-zero value, e.g., for silica-based fibers it is $\chi^{(3)}_{SiO_2} \sim 2 \times 10^{-22} \text{ m}^2\text{V}^{-2}$ [6]. Thus, the most interesting aspect is how large a potential of the electric field can be created in the fiber's core and how $\chi^{(2)}$ can be enhanced when the fiber is fabricated from glasses with high $\chi^{(3)}$.

Silica glass is usually contaminated with various impurities: Na, Li, K. Raising the temperature to an appropriate level leads to the formation of corresponding

ions: Na^+ , Li^+ , K^+ . It is assumed that the positive ions are uniformly distributed throughout the glass volume and, as a result of the applied voltage, migrate toward the cathode.

It is also assumed that water molecules are deposited on the electrode surface. Under the influence of the applied voltage, these molecules may dissociate into various ions. It is most commonly assumed that H_3O^+ ions are formed. These ions are injected into the glass volume from the anode. We assumed the most realistic model of injection rate $V_{H_3O^+}$ proportional to the electric potential ϕ at the anode surface:

$$V_{H_3O^+} = \sigma_2 \phi, \quad (2)$$

where $\sigma_2 = 5 \times 10^{12} \text{ m}^{-2}\text{V}^{-1}\text{s}^{-1}$ is a proportional constant determined by fitting the simulation to the experimental results [7].

All positive ions that reach the cathode are completely absorbed – no accumulation occurs. It is also assumed that negative ions, specifically non-bridging oxygen ions (NBO^-), are immobile and present in sufficient quantities to balance the positive charge in the glass before the external voltage is applied. Decreasing the temperature results in ‘fixing’ the last ion distribution, thus obtaining a frozen-in non-uniform electric field E_{DC} .

Under the influence of an external electric field, ions move. Accordingly to the model presented in [8] the measure of this movement is the ionic flux:

$$J_i = -D_i \nabla c_i - \mu_i z_i F c_i \nabla \phi, \quad (3)$$

where: J_i – ionic flux of the type i ions [$\text{mol}/(\text{m}^2\text{s})$], D_i – diffusion coefficient for the ions of type i [m^2/s], c_i – i -type ions concentration [mol/m^3], μ_i – i -type ions mobility [$\text{m}^2\text{V}^{-1}\text{s}^{-1}$], z_i – ions charge (+, -), F – Faraday constant [C/mol], ϕ – electrical potential [V/m].

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If no chemical reactions occur, the law of mass conservation applies:

$$\nabla J_i = 0. \quad (4)$$

When an external voltage is applied, the distribution of the electric field is governed by Poisson's equation:

$$\nabla(-\varepsilon\nabla\phi) = -\Delta(\varepsilon\phi) = \rho, \quad (5)$$

After switching off the external field, the charge density ρ depends on the sign and concentration of the individual ions:

$$\rho = F \sum_i z_i c_i. \quad (6)$$

Finally, the potential, which determines the distribution of the field (E_{DC}), is calculated from the following equation:

$$E_{DC} = -\nabla\phi \approx \frac{\rho r}{\varepsilon}, \quad (7)$$

where an approximation due to the small distance r between the centre of the core and the anode was used, and $\varepsilon = \varepsilon_r \varepsilon_0$ stands for absolute material permittivity and includes vacuum permittivity and relative permittivity of dielectric, e.g., for silica glass it is $\varepsilon_{rA} = 3.8$ [9] and for borosilicate glasses it equals $\varepsilon_{rB} = 4.7$ [10].

We applied the model presented above [8] to calculate the electrical potential in Fiber A, made of silica glass, and in Fiber B, made of in-house borosilicate glass with a high $\chi^{(3)}$ value. Analysis was performed at an applied voltage of 5 kV, with the temperature elevated to 265 °C. We consider the same geometry for both fibers, with side air holes with electrodes and an off-center core to allow E_{DC} to reach the core area (see Fig. 1). Fiber A has a core and cladding made of fused silica, whereas for Fiber B, we used a pair of borosilicate glasses labelled as SK222 for the cladding and ZR3 for the core [11]. Third-order susceptibilities for in-house glasses are as follows: $\chi^{(3)}_{SK222} \sim 4.34 \times 10^{-22} \text{ m}^2 \text{V}^{-2}$ and $\chi^{(3)}_{ZR3} \sim 8.87 \times 10^{-22} \text{ m}^2 \text{V}^{-2}$, respectively [12].

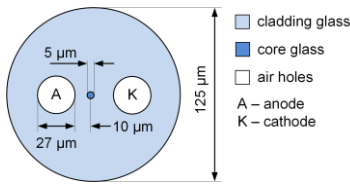


Fig. 1. Scheme of the fiber used for simulations.

For Fiber A, we assumed migration of Na^+ and Li^+ ions only. In the case of Fiber B, only Na^+ and K^+ ions were taken into consideration. For both fibers, we postulated immobility of NBO^- balancing ions and the injection of H_3O^+ ions, with the concentration proportional to the applied electric field. The parameters used for the simulations are listed in Table 1, including the concentrations of K^+ , Na^+ , and Li^+ ions in the considered glasses for Fiber A and Fiber B.

We show the maximum concentration c of ions for Fiber B; however, we also analyzed the generated E_{DC} and $\chi^{(2)}$ for lower concentrations. To compare results for both

fibers, we defined normalized concentration k , which equals 0.1 for 1 ppm and $k=1$ for 12.3 ppm. Since alkali ions are presented only as impurities in silica glass, we considered Fiber A only for $k=0.1$.

Results of simulations show that E_{DC} increases gradually only for several seconds after applying a voltage (external electric field) and saturates. The highest value is obtained for silica and reaches $5.48 \times 10^8 \text{ V/m}$ (Fig. 2a, series with bold black line).

Table 1. Concentration c , charge z , mobility μ and diffusion D values used in simulations for Fiber A and Fiber B.

Fiber	A		B		A		B	
	c		z		μ		D	
Ion	ppm				$\text{m}^2/(\text{Vs})$		m^2/s	
Na^+	1	12.3	+1	+1	2×10^{-15}		9.0101×10^{-17}	
Li^+	1	0	+1	n/a	$0.15 \mu_{\text{Na}}$	n/a	$0.15 D_{\text{Na}}$	n/a
K^+	0	0.7	n/a	2	n/a	$0.15 \mu_{\text{Na}}$	n/a	$0.15 D_{\text{Na}}$
H_3O^+	$\sigma_2 E$		+1	+1	$0.001 \mu_{\text{Na}}$		$0.001 D_{\text{Na}}$	
NBO^-	2	13	-1	-1	0		0	

For Fiber B, the maximum E_{DC} equals $4.41 \times 10^8 \text{ V/m}$ and is obtained for the lowest concentration of alkali-ions $k=0.1$, similar to one considered for Fiber A. We suppose that the reason for this behaviour is the high-alkali ions concentration, which hinders the migration of the positive ions by shielding the electric field. This means that the ions do not move as far away from the anode, so that the maximum potential is in the core region (Fig. 2b).

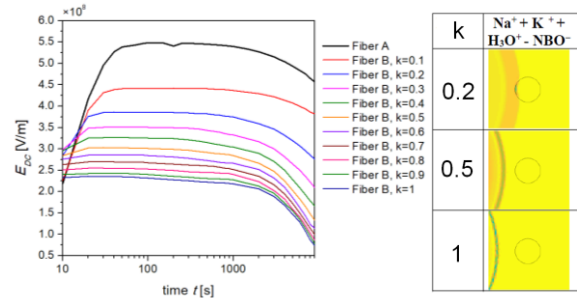


Fig. 2. Electric field E_{DC} : a) induced in the core area as a function of thermal poling duration. The black series in bold represents Fiber A, while the color series represents Fiber B with different concentrations of alkali ions. Distribution of E_{DC} b) in the vicinity of the core of Fiber B (circle indicates core location).

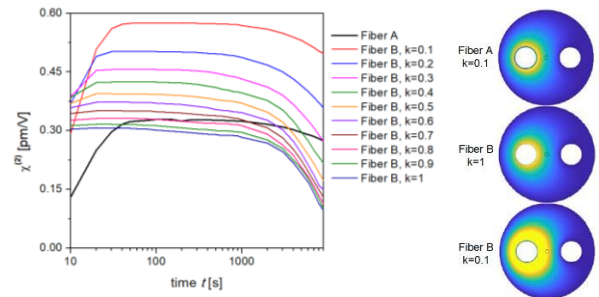


Fig. 3. Second-order susceptibility $\chi^{(2)}$ a) induced in the core area as a function of thermal poling duration. The Black series in bold is for Fiber A, while the color series is for Fiber B with different concentrations of alkali ions. Distribution of $\chi^{(2)}$ b) for Fiber A and Fiber B for thermal poling duration of $t=1000 \text{ s}$.

Summarizing, the obtained E_{DC} values for Fiber B are smaller than for Fiber A and decrease together with the rise of alkali-ions concentration. However, this is not the case for created $\chi^{(2)}$. Fiber B is primarily composed of SK222 glass; therefore, for the calculation of $\chi^{(2)}$, according to Eq. (1), we used $\chi^{(3)}_{SK222}$, which is more than twice as high as that of silica glass. As a result, the induced susceptibility of the second order of Fiber B can be higher than for Fiber A (Fig. 3). For the same concentration of alkali-ions, we obtained $\chi^{(2)}$ equal to 0.33 pm/V and 0.58 pm/V, respectively for Fiber A and Fiber B.

Another question concerns the initiation of the thermal poling process and the activation temperature for ion mobility. We can ask if it is possible to induce and erase $\chi^{(2)}$ in a relatively short time, i.e., a several seconds? Or is it possible to perform the poling process without elevating the temperature?

To answer those questions, we compared created $\chi^{(2)}$ for Fiber A and Fiber B, for $k=0.1$, changing only temperature of poling (Fig. 4). At room temperature $\chi^{(2)}$ is about three times higher in time range between 10 – 20 s, and saturates at higher level, reaching maximum 0.61 pm/V in the case of Fiber B. For Fiber A, we observe the same behaviour. However, the difference is smaller at the beginning of the process and for saturation, as well, reaching a value of 0.35 pm/V. It is also evident that saturation occurs faster at room temperature.

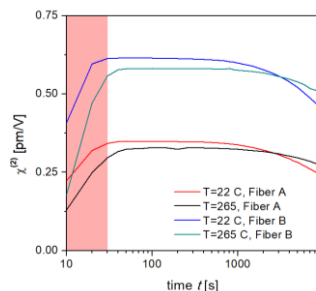


Fig. 4. Second-order susceptibility $\chi^{(2)}$ induced in the core area of Fiber A and Fiber B as a function of thermal poling duration for two temperatures: 22 °C and 265 °C. The red rectangle indicates the first 30 s of the process.

Another aspect is the stability of the induced electric potential and nonlinearity. It is well known that potential E_{DC} can be frozen in the glass when high temperature, i.e., $T=265$ °C, of poling is decreased for biased voltage. It means that the mobility of ions is drastically reduced, and they cannot return to their initial position. At room temperature, the mobility of ions remains constant; therefore, when the bias voltage is turned off, there is no force to hold the ions in their positions. We expect that the ions can slowly return to equilibrium. This fact, along with much higher $\chi^{(2)}$ values achieved after 10÷20 s of poling at room temperature, may be useful for dynamic induction and deletion of second-order susceptibility.

Summarizing, we numerically analysed the mechanism of inducing second-order susceptibility in optical fibers of

two types: Fiber A made of silica glass with alkali impurities and Fiber B made of borosilicate glasses with a high concentration of alkali-ions, being a glass structural elements. We have shown that, for typical thermal poling conditions ($T=265$ °C and $U=5$ kV) for Fiber B with high alkali-ions concentration $k=1$, obtained maximum value of $\chi^{(2)}$ is lower than for silica-based Fiber A with $k=0.1$. This is related to the fact that the potential range in Fiber B is reduced due to the impeded migration of too many alkali ions. However, for a lower concentration of alkali-ions of Fiber B, i.e., $k=0.1$, the electrical potential range and value are high enough to achieve $\chi^{(2)}=0.58$ pm/V, almost twice as high as for Fiber A.

A particularly interesting case is the thermal poling process conducted at room temperature ($T = 22$ °C). We have shown that for Fiber A and Fiber B with the same concentration of alkali ions, $k = 0.1$, after the first 15 seconds of poling, $\chi^{(2)}$ created is significantly higher than for poling with $T = 265$ °C, and the process is faster for Fiber B. Since mobility of the ions is the same during poling, induced E_{DC} won't be frozen in the fiber, which allows to consider a generation of reversible $\chi^{(2)}$ nonlinearity in glasses.

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