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Fage-Pedersen, Jacob; Kristensen, Martin; Beerman, J.

Published in:
CLEO/Europe

Link to article, DOI:
10.1109/CLEOE.2003.1312274

Publication date:
2003

Document Version
Publisher's PDF, also known as Version of record

Link back to DTU Orbit

Citation (APA):
Fage-Pedersen, J., Kristensen, M., & Beerman, J. (2003). Poling of glass waveguides by a Metal-induced X(3) enhancement. In CLEO/Europe IEEE. DOI: 10.1109/CLEOE.2003.1312274

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Poling of Glass Waveguides by a Metal-Induced $\chi^{(3)}$ Enhancement.

Jacob Fage-Pedersen and Martin Kristensen,
Research Center COM, Technical University of Denmark, Building 345V, DK-2800 Kgs. Lyngby, Denmark.
Jonas Beerman, Institute of Physics, Aalborg University, Pontoppidanstræde 103, DK-9220 Aalborg Ø, Denmark.

While the perspectives of making 2nd-order nonlinearities in optical glasses are exciting, present poling techniques are still inadequate. UV poling has proven difficult to control, and with traditional thermal poling the obtained $\chi^{(3)}$ has remained $< 1$ pm/V. When inducing a permanent field $E_{dc}$ in the glass, the effective 2nd-order susceptibility $\chi^{(2)} = 3\chi_0^{(3)}E_{dc}$ has been limited by the small intrinsic value of $\chi_0^{(3)}$. However, an alternative method consists in utilizing indiffused metal [1]. Here we demonstrate that metal nanoclusters are an attractive means to enhance the $\chi^{(2)}$ value. In combination with a built-in dc-field in channel waveguides, effective $\chi^{(2)}$ values of more than 14 pm/V are obtained.

The channel waveguides were made by UV writing in three-layer structures [SiO$_2$ - Ge:SiON (germanium-doped silicon oxy-nitride) - SiO$_2$] that had been fabricated on Si substrates by plasma-enhanced chemical vapour deposition as in Ref. [1] and then loaded with deuterium. An electrode consisting of a Ag-containing paint was put on each sample and the poling was performed in air by heating the sample while applying $+ 1000$ V to the electrode, keeping the Si substrate on ground potential.

Optical characterisation was performed with second-harmonic scanning optical microscopy where a pump beam at the fundamental (F) wavelength is focused onto the sample and the second-harmonic (SH) signal is measured in the reflection direction while the sample is moved orthogonally to the pump beam. The SH images (examples in Fig. 1, using a 790-nm pump) show that a large $\chi^{(2)}$ nonlinearity is induced throughout the Ge:SiON core layer, peaking at the interfaces of the Ag-modified channel waveguide and being negligible in the SiO$_2$ cladding layers. In particular, the vertical polarisation (parallel to the poling field) of the SH signal is very strong, indicating that a dc field has been built into the core layer. Upon 20-min poling at temperatures of 225, 285, 325, and 375 °C, typical $\chi^{(2)}$ values inside the channels waveguide are 0, 0.6±0.4, 10.3±4.0, and 13.5±5.0 pm/V, respectively, and even higher at the interface. SH images recorded from the end facet of a cleaved sample and from the top surface are in reasonable agreement, confirming that the nonlinearity is large throughout the sample and not merely an artefact of the cleaved surface.

Concerning the poling mechanism, it is a key issue that large concentrations of Ag have indiffused from the electrode to the core layer during poling, as confirmed with secondary-ion mass spectrometry [1]. Moreover, the poling induces an absorption peak at 415 nm (Fig. 2) which is of the width and at the peak position of the surface-plasmon resonance (SPR) of Ag nanoclusters with radius ~ 10 nm in a glass matrix [2]. Since $\chi^{(2)}$ of a metal-cluster embedded dielectric medium is strongly enhanced at the SPR it is therefore concluded that the large $\chi^{(2)}$ observed at $\lambda_{SH} = 395$ nm (near the SPR) is indeed due to a combination of a large $\chi^{(2)}$ value and a built-in field, i.e., $\chi^{(2)} = 3\chi_0^{(3)}E_{dc}$.

We anticipate that an improved poling scheme can be made where the loss is reduced while a large $\chi^{(2)}$ is maintained. This will require lower Ag concentrations and optimisation of $E_{dc}$ and should be combined with waveguides with especially optimised 400-nm transmission. As a further attractive perspective, the shape and dimension of the nanoclusters can potentially be varied in order to tune the width and position of the $\chi^{(3)}$ resonance.

References