METRICA
METRology Institutional CAtalog

ISTITUTO NAZIONALE DI RICERCA METROLOGICA
Repository Istituzionale

On the existence of nanogratings in commercial oxide glasses

This is the author's accepted version of the contribution published as:

Original
On the existence of nanogratings in commercial oxide glasses / Xie, Qiong; Cavillon, Maxime; Poumellec, Bertrand; Pugliese, Diego; Janner, Davide; Lancry, Matthieu. - (2022), p. BM3A.2. (Intervento presentato al convegno Bragg Gratings, Photosensitivity and Poling in Glass Waveguides and Materials (BGPPM) 2022 tenutosi a Maastricht, Limburg Netherlands nel 24-28 Luglio 2022) [10.1364/BGPPM.2022.BM3A.2].

Availability:
This version is available at: 11696/77378 since:

Publisher:
Optica Publishing Group

Published
DOI:10.1364/BGPPM.2022.BM3A.2

Terms of use:

This article is made available under terms and conditions as specified in the corresponding bibliographic description in the repository

Publisher copyright

(Article begins on next page)

28 April 2024

On the existence of nanogratings in commercial oxide glasses

Qiong Xie[1], Maxime Cavillon[1], Bertrand Poumellec[1], Diego Pugliese[2], Davide Janner[2], and Matthieu Lancry[1]

[1]Institut de Chimie Moléculaire et des Matériaux d'Orsay (ICMMO), Université Paris-Saclay, CNRS, 91405 Orsay, France [2]Department of Applied Science and Technology (DISAT) and RU INSTM, Politecnico di Torino, 10129 Torino, Italy Author e-mail address: qiong.xie@universite-paris-saclay.fr

Abstract: The ability to induce nanogratings using a femtosecond laser in common oxide glasses is investigated experimentally. A simple and general viscosity-based approach is subsequently employed to predict their existence in glass. © 2022 The Author(s)

1. Introduction

Nanogratings, also labeled as "Type II" transformations, were first observed in 2003 [1] after femtosecond (fs) laser irradiation inside silica glass. They are self-organized and sub-wavelength birefringent structures that are formed upon the action of high intensity ultrashort light pulses in the bulk of a transparent material. They have found interest in optics/photonics, microfluidics, optical data storage or again optical sensing applications [2]. However, the ability to successfully imprint 3-dimensional (3D) nanogratings in silicate glasses strongly depends on the glass composition. This work provides insights on the relative difficulty to imprint nanogratings in some of these commercial glasses. The goal is to describe, based on a viscosity approach, the ability for a glass to yield permanent formation of nanogratings. This systematic work allows a direct comparison between the glass samples and their respective nanogratings processing windows. The observed differences are then tentatively linked to viscosity- driven mechanisms, framing the existence of the nanogratings processing window. This work opens the door to future glass viscosity engineering to maximize 3D nanogratings imprinting and related optical properties.

2. Experimental details

Five bulk glasses were selected based on literature results and viscosity profiles: BK7, AF32, Borofloat 33, GeO2, and SiO2 (SuprasilCG). Each glass sample, in the form of a plate was irradiated in similar conditions using a fs laser. In this work, the investigation of nanogratings existence was probed in a pulse energy (Ep, in µJ) – repetition rate (RR, in kHz) landscape. Polarized optical microscopy was used to quantify the birefringence response and characteristics of nanogratings. Additionally, scanning electron microscopy (SEM) was performed on the laser tracks cross sections (x,z plane) to ensure the existence of porous nanolayers and nanopores. The data taken for all commercial glasses investigated were provided by the glass suppliers. And the temperature dependence of glass viscosities ($\eta(T)$) was fitted using the Vogel–Fulcher–Tammann (VTF) equation.

3. Results

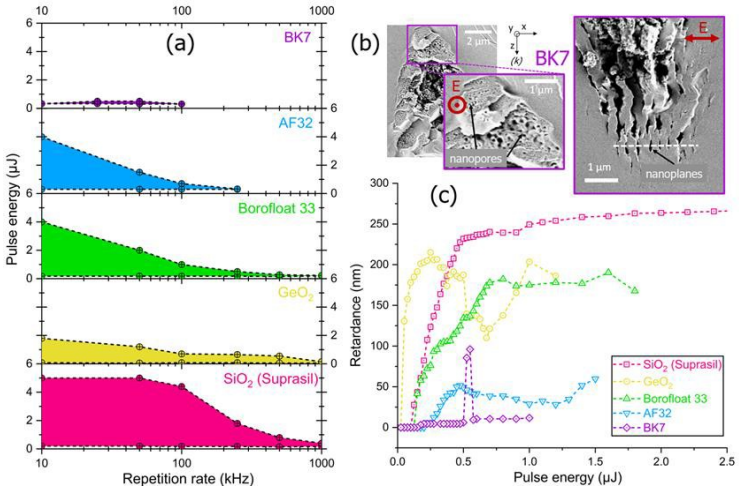

Fig. 1. (a) Determination of observed nanogratings in an energy-repetition rate landscape for five glasses: SiO2 (Suprasil), GeO2, Borofloat 33, AF32, and BK7. (b) Illustration of nanogratings observed in BK7 from SEM analysis; conditions are pulse duration = 800 fs, writing speed = 1 µm/s, RR = 25 kHz, Ep = 0.6 µJ, focal depth = 300 µm. (c) Evolution of retardance as a function of pulse energy (800 fs, 50

kHz, NA = 0.6); each data point has a ± 5 nm uncertainty.

The formation of nanogratings in the Ep-RR landscape was investigated. The domain of nanogratings existence for each glass is shown in Fig. 1(a). The glass material strongly influences the ability to imprint nanogratings from the "self-organized" plasma. For BK7, which presents the smallest nanogratings window, porous nanogratings could be observed, as shown in Fig. 1(b), but for very specific conditions (<100 kHz, and for typ. 50,000 pulses/μm). Additionally, and for all glasses, an example of measured retardance values (i.e., birefringence x nanogratings length along z-axis) with respect to energy and at a constant RR = 50 kHz is provided in Fig. 1(c). The nanogratings window is the largest for SiO_2, intermediate for GeO_2, Borofloat 33, and AF32, and is extremely reduced for BK7 (an alkali "rich" borosilicate). Correspondingly, much higher values of retardance are found in silica glass compared to AF32 and BK7, which agrees with the tendency to form nanogratings more easily in SiO_2 or GeO_2 are strong network formers. The principal aim of this paper is to highlight the link between nanogratings window and glass viscosity behavior with temperature. The viscosity as a function of temperature for multiple oxide glasses (in addition to the ones discussed in this paper) is reported [3-4] (Fig. 2(a)). An estimated domain of nanogratings existence is provided in Fig. 2(b), by taking the temperature difference between *Tmax* (set as $\eta = 10^{3.0}$ Pa·s) and *Tsoft* ($\eta = 10^{6.6}$ Pa·s), respectively upper and lower bounds. The lower limit corresponds to a temperature at which the viscosity is ~$10^{6.6}$ Pa·s, where occurs nanocavitation of the glass, forming the nanopores that compose the nanogratings. An upper temperature limit, set for a viscosity value of ~$10^{3.0}$ Pa·s, relates to either collapse or growth of the nanopores, resulting in the erasure of the nanopores, hence the nanogratings. The experimental results agree with the predictions made by this viscosity approach and literature data [5].

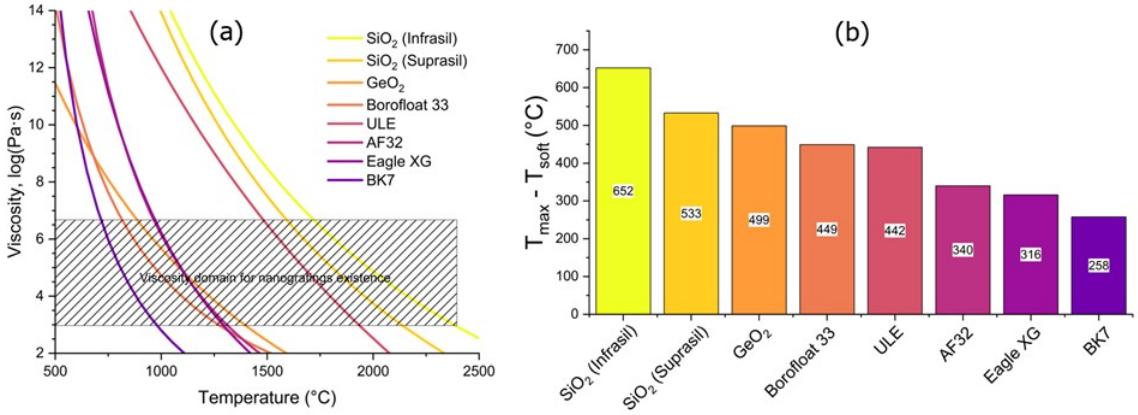

Fig. 2. (a) Viscosity as a function of temperature for a variety of commercial and typical glasses, along with an estimated domain of nanogratings existence from *Tsoft* ($\eta = 10^{6.6}$ Pa·s) to *Tmax* ($\eta = 10^{3.0}$ Pa·s). (b) Temperature difference (*Tmax* – *Tsoft*) as a function of glass composition. A larger value suggests a wider processing window (with respect to temperature) to form nanogratings.

4. Conclusions and perspectives

Understanding the formation of nanogratings inside silicate and germanate glasses is an attractive research field, since these sub-wavelength and self-organized structures enable miniaturized functionalization and unique optical properties including the form of birefringence, stress-birefringence engineering and even optical chirality. This work addresses the challenges to imprint, or not, nanogratings, based on a viscosity approach. These predictions are validated by experimental work performed on five commercial glasses.

3. References

[1] Y. Shimotsuma, P. G. Kazansky, J. Qiu, and K. Hirao, "Self-Organized Nanogratings in Glass Irradiated by Ultrashort Light Pulses," Phys. Rev. Lett., **91** (24), 247405(1)-247405(4) (2003).

[2] R. Stoian, "Volume photoinscription of glasses: three-dimensional micro- and nanostructuring with ultrashort laser pulses," Appl. Phys. A Mater. Sci. Process., **126**(6), 1–30(2020).

[3] A. Rudenko, J. P. Colombier, and T. E. Itina, "Nanopore-mediated ultrashort laser-induced formation and erasure of volume nanogratings in glass," Phys. Chem. Chem. Phys., **20**(8), 5887–5899(2018).

[4] Y. Wang, M. Cavillon, N. Ollier, B. Poumellec, and M. Lancry, "An Overview of the Thermal Erasure Mechanisms of Femtosecond Laser- Induced Nanogratings in Silica Glass," Phys. Status Solidi Appl. Mater. Sci., **218**(12), 2100023(1)-2100023(15) (2021).

[5] E. O. Kissi and Y. Bellouard, "Self-organized nanostructures forming under high-repetition rate femtosecond laser bulk-heating of fused silica," Opt. Express, **26**(11), 14024(2018).