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BOOK OF ABSTRACTS

SEVENTH INTERNATIONAL CONFERENCE ON RADIATION IN VARIOUS FIELDS OF RESEARCH

June 10-14, 2019
Herceg Novi
Montenegro

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30 years

RAD 7 SEVENTH INTERNATIONAL CONFERENCE ON RADIATION IN VARIOUS FIELDS OF RESEARCH

Transient optical effects in spin-coated chalcogenide glass thin films induced by UV radiation

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Thermally evaporated thin films of chalcogenide glasses are known to be highly photosensitive to bandgap and super-bandgap radiation. However, their practical application is limited because of long-term structural relaxation accompanied the photoinduced changes. Spin-coating is one of the alternative methods of thin film preparation which mostly eliminates photosensitivity to bandgap radiation and removes metastable component of photoinduced changes caused by super-band gap (mostly UV) light. Such films are usually manufactured through chemical dissolution of bulk glasses in different amine-based solvents with subsequent spin-coating of the liquid onto silica substrate and appropriate one-stage or multi-stage thermal treatment. Such photostability can be very useful, especially for the nonlinear optical applications, which require high transparency in IR spectral region and minimum sensitivity to the high energy light radiation.

Structure of thermally stabilized spin-coated films can, in general, be considered as consisting of fragments of bulk glass connected through the residual units of organic solvents. However, increase of annealing temperature promotes direct connections between the glass fragments (through chalcogen atom or newly formed appropriate structural units). The decrease in photosensitivity relatively to the evaporated thin films is linked to the lack of excessive concentration of homopolar bonds in the film structure. At the same time, noticeable transient red shift of optical absorption edge (photodarkening) is still observed at irradiation of the spin-coated thin films with super-bandgap UV light. It is shown that by changing post-synthesis annealing conditions it is possible to control the transient optical switching effect.

Kinetics of the transient photoinduced optical changes demonstrates significant deviation from the exponential behavior. Mechanisms of the transient effects are debated based on parameters of fitting of the kinetic curves with stretched exponential function. Possible applications of the observed effects are discussed.

Acknowledgments: APSU team acknowledges financial support from NSF grant DMR-1409160.

On the phenomenological identity of radiation-induced effects in glassy chalcogenides under a prism of unified configuration-enthalpy model

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Unified configuration-enthalpy model evolving conjugated configuration-coordinate and thermodynamic-enthalpy diagrams is developed to describe optical response in metastability of chalcogenide glasses (ChG) caused by combined physical ageing and high-energy irradiation. Within this approach, these glasses are supposed be stabilized in the ground state and temporary excited state, the former being presented by tightly interconnected metastable wells (i.e. rejuvenation-induced, irradiation-induced, physically-aged and deep crystalline-like ones) linked by thermally-activated over-barrier and tunneling through-barrier transitions.

Effect of high-energy γ-irradiation on ChG such as glassy As-S is reflected in vertical transitions of atomic sites into excited state followed by spontaneous non-radiative relaxation into irradiation-induced ground-glass sub-state. Thermodynamic enthalpy diagram linked with configuration-coordinate one allows complete parameterization of corresponding optical responses related to these states, defined in blue (bleaching) or red (darkening) shifts in the fundamental optical absorptions edge of As-S glasses. Thus, the phenomenological identity of “pure” radiation-induced optical changes can be revealed in ChG of multinary chemical compositions obeying competitive changes from many supplemented influences.

PUBLISHER: RAD Centre, Niš, Serbia
Bulevar Nikole Tesle 17/12, 18000 Niš, Serbia
www.rad-centre.org

FOR THE PUBLISHER: Prof. Dr. Goran Ristić

YEAR OF PUBLISHING: 2019

EDITOR: Prof. Dr. Goran Ristić

COVER DESIGN: Vladan Nikolić, PhD

TECHNICAL EDITING: Saša Trenčić, MA

PROOF-READING: Saša Trenčić, MA and Mila Trenčić, MA

CD BURNING AND COPYING: RAD Centre, Niš, Serbia

PRINT RUN: Electronic edition - 350 CDs (CD-R)

ISBN: 978-86-901150-0-6

The Seventh International Conference on Radiation in Various Fields of Research (RAD 2019) was financially supported by the Central European Initiative (CEI).

CIP - Каталогизација у публикацији
Народна библиотека Србије, Београд

539.16(048)(0.034.2)
57+61(048)(0.034.2)

INTERNATIONAL Conference on Radiation in Various Fields of Research (7 ; 2019 ; Herceg Novi)

Book of abstracts [Elektronski izvor] / Seventh International Conference on Radiation in Various Fields of Research, RAD 7, [RAD 2019], 10-14.06.2019 Herceg Novi, Montenegro ; [editor Goran Ristić]. - Niš : RAD Centre, 2019 (Niš : RAD Centre). - 1 elektronski optički disk (CD-ROM) ; 12 cm

Sistemski zahtevi: Nisu navedeni. - Nasl. sa naslovne strane dokumenta. - Tiraž 350.

ISBN 978-86-901150-0-6

а) Јонизујуће зрачење -- Дозиметрија -- Апстракти б) Биомедицина – Апстракти

COBISS.SR-ID 277775116

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