Design rules for chalcogenide thin films toward on-chip highly nonlinear optical components in the Mid-Infrared

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ABSTRACT

Some chalcogenide glasses (ChGs) are showing a large transparency window in the infrared coupled with outstanding optical nonlinearities offering tremendous opportunities for achievement of innovative mid-infrared (MIR) on-chip components. By means of industrial co-sputtering deposition technique, we study the amorphous structure and the nonlinear optical properties of As-free amorphous $GeSb_wS_xSe_yTe_z$ chalcogenide thin films. The nonlinear refractive Kerr index (n_2) of the films were evaluated by means of modeling of spectroscopic ellipsometry data validated experimentally for some compositions by means of advanced nonlinear optical characterizations in waveguides. State-of-the-art and higher n_2 values were obtained. Depending on the composition of the $GeSb_wS_xSe_yTe_z$, n_2 can vary of more than one order of magnitude. Finally, Fourier-Transform Infrared (FTIR), Raman and X-ray Absorption (XAS) spectroscopies analysis of the amorphous structure of some ChGs prototypical compositions in relation with their nonlinear optical properties was used as a basis for ab initio molecular dynamics (AIMD) simulations. Thus, the intimal link between local atomic configurations and optical nonlinearities is proposed giving unique clues to control optical nonlinearities of chalcogenide materials.

Key words: chalcogenide glasses; mid infrared; nonlinear optics; co-sputtering; AIMD; photonics

1. INTRODUCTION

Among their unique portfolio of properties, some chalcogenide glasses (ChGs) offer also unique opportunities for innovative mid-infrared (MIR) components thanks to their outstanding trade-off between a high transparency window and large optical nonlinearities in the infrared range. However, the REACH European recommendation and the World Health Organization have both identified Arsenic as one of the ten most harmful chemicals for human health. In that context, we study here how to tailor the linear and nonlinear optical properties of As-free amorphous $GeSb_wS_xSe_yTe_z$ chalcogenide thin films with a particular emphasis on their compatibility with CMOS technologies for future realization of on-chip MIR components.

2. EXPERIMENTS

The $GeSb_wS_xSe_yTe_z$ thin films were obtained by means of magnetron co-sputtering in an industrial 200 mm deposition cluster tool. Modeling of spectroscopic ellipsometry data acquired on the as-deposited films allowed to determine their optical constants (refractive index n, extinction coefficient k and optical bandgap E_g^{opt}) as well as evaluating their nonlinear refractive Kerr indices (n_2). Advanced optical characterizations of nonlinearities of chosen prototypical CGs compositions in waveguides were also performed in order to validate modeling of 3rd order nonlinear refractive indices. The link between amorphous structure and enhanced optical nonlinearities was probed by means of FTIR, Raman and XAS spectroscopies on some chosen compositions. On the basis of all experimental data, AIMD simulations permitted to calculate the local electronic polarizability depending on the local structural motifs in the different prototypical glasses evidencing the non-obvious link between some peculiar structural motifs and the material's enhanced macroscopic optical nonlinearity.

3. RESULTS & DISCUSSION

In **Fig. 1a** are plotted the n_2 Kerr refractive indices as a function of the bandgap of the studied $GeSb_wS_xSe_yTe_z$ thin films. Depending on the composition, the amorphous $GeSb_wS_xSe_yTe_z$ films exhibit a wide range of n_2 values reaching up to 500 times that of silicon dioxide. We emphasize that these results are well supported by previous study of similar $GeSb_xSe_y$ compounds in the literature[1]. Kerr index shows a huge dependence on the ChG's composition. In order to relate optical nonlinearities and structure, four prototypical compositions differing by their n_2 values were selected and studied experimentally as well as by AIMD simulations.

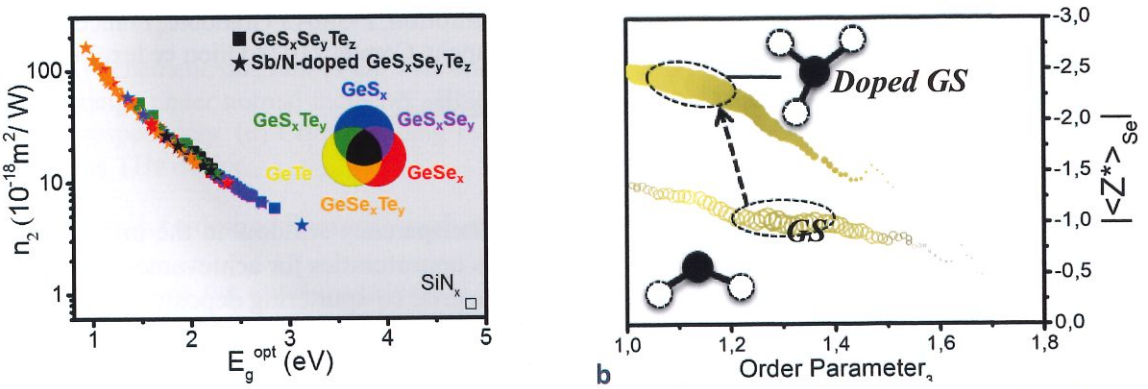

Figure 1: (a) 3rd order nonlinear refractive indices at 0,534 x E_g^{opt} depending on optical band gap energy of (co-)sputtered $GeSb_wS_xSe_yTe_z$ films. (b) Born effective charges Z extracted from AIMD calculations for GS and doped GS materials (open and close symbols). Dots' size is proportional to number of Se-based structural motifs distinguished by means of the order parameter. A significant increase of Z* and thus electronic polarizability is observed for GS upon doping.*

From the calculation of the atomic Born effective charges, one could relate the origin of the local enhancement of electronic polarizability in the amorphous network to the enhanced optical nonlinearities of the material. We show that this unique local mechanism is somehow reminiscent to the unique bonding mechanism behind optical and electrical contrast between amorphous and crystalline state of phase-change materials as revealed very recently[2,3].

4. CONCLUSION

The outstanding linear and nonlinear optical properties of some chalcogenide glasses at origin of their promising potential of applications for MIR on-chip components is shown to result from formation of specific bonds and peculiar structural motifs. For instance, incorporation of some element increases dramatically the local electronic polarizability by inducing structural motifs supporting newly-introduced metavalent bonding mechanism arising in some local crystal-like motifs[4]. These results pave the way to control and further enhancement of the unique optical properties of chalcogenide glasses and thin films.

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Biographies

Jean-Baptiste Dory obtained an engineer diploma in materials science in 2015. He joined CEA Leti French laboratory in 2016 as a Ph.D. student in physics from Bourgogne Franche-Comté University. During his thesis he developed skills in optics and materials science to design innovative amorphous chalcogenide thin films for nonlinear photonics in the mid-infrared. His field of expertise covers co-sputtering deposition of chalcogenide materials in industrial clean room, thin films structural/optical advanced characterization, waveguiding devices design by means of finite element simulations and supervision of 200 mm technological nano-fabrication of photonic components.