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Mechanical model of giant photoexpansion in a chalcogenide glass and the role of photofluidity

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ABSTRACT

An analytical model is developed to describe the phenomenon of giant photoexpansion in chalcogenide glasses. The proposed micro-mechanical model is based on the description of photoexpansion as a new type of eigenstrain, i.e. a deformation analogous to thermal expansion induced without external forces. In this framework, it is the viscoelastic flow induced by photofluidity which enable the conversion of the self-equilibrated stress into giant photoexpansion. This simple approach yields good fits to experimental data and demonstrates, for the first time, that the photoinduced viscous flow actually enhances the giant photoexpansion or the giant photocontraction as it has been suggested in the literature. Moreover, it highlights that the shear relaxation time due to photofluidity controls the expansion kinetic. This model is the first step towards describing giant photoexpansion from the point of view of mechanics and it provides the framework for investigating this phenomenon via numerical simulations.

1. Introduction

Chalcogenide glasses cover a wide range of applications [1,2]; first and foremost because of their transparency in the infrared range. This includes thermal imaging cameras [3], generation of new infrared sources [4], chemical and biological sensors [5], optical data storage [6], etc. Chalcogenide glasses are also photosensitive materials. They exhibit a wide range of photoinduced phenomena [7,8] including: photodarkening/bleaching [9], photoinduced fluidity [10,11], photo-induced structural relaxation [12,13], photoinduced expansion [14] or contraction [15], etc. Some of these effects can be scalar or vectorial, i.e.: isotropic or anisotropic respectively, depending on the light polarization [16].

Most photostructural changes can be induced by irradiation with either above or below band-gap light. Some photoinduced effects are much more pronounced in the sub-bandgap range [17], including giant photoexpansion-contraction [14,15], photofluidity [10,11] and photo-induced structural relaxation [18], because, sub-bandgap light permits to illuminate a larger volume. In addition, above bandgap irradiation is associated with high absorption that is sufficient to induce a significant warming and even potential damage, while sub-bandgap irradiation has a negligible thermal contribution under standard irradiation power. Several proofs of the optical origin and athermal nature of photoinduced effects exist including the fact that photofluidity is more pronounced at low temperature [10] and exhibits polarization effects [19] or the formation of wavelength specific Bragg gratings [20]. Nevertheless, it has been recently suggested that the thermal contribution to the photoexpansion itself may not be negligible even for weakly absorbed light [21].

Giant photoexpansion in chalcogenide glasses has many applications including the production of adaptive lenses [22], microlenses arrays [23], optical fiber microlenses [24], structured concave/convex lenses [15] and relief patterning for the production of gratings [19,25,26]. This technique is promising, since complex optical elements can be optically created in a single step. Nevertheless, if the kinetic and the geometry of the expansion cannot be predicted, the development of this technique will be limited because of the large number of tests required to produce the desired lens geometry. Hence a mechanical model describing the photoexpansion would considerably improve the predictability of the expansion geometry and would generally ease the development of photo induced relief patterning techniques. However, the giant photoexpansion is a phenomenon

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which is very difficult to model since its physical origin is still debated, and since other photoinduced effects may contribute to the expansion. Usually, bulk glasses or thin films are irradiated with a laser beam having a diameter much lower than the sample size. This way, the irradiated volume fluidifies and expands while being confined by the quasi-rigid volume surrounding the laser path which remains mostly unchanged. If the irradiated volume can expand easily in the direction of free surfaces, it is stressed in the direction perpendicular to the bean axis: this is where the viscoelastic properties of the glass come into play. This can be easily understood thanks to the phenomenological model proposed by Tanaka et al. [27]. Despite clear indications that the phenomenon is governed by mechanical stress, the mechanical aspect of this phenomenon has been largely ignored or poorly treated. Various models of photoexpansion have been proposed in the literature [17,28], but they all treat the expansion as if it was stress-free. The objective of the present work is to treat the photoexpansion from the point of view of mechanics, by taking into account the combined effect of these stresses and the photoinduced fluidity. We show here, using some simple hypothesis, that the contribution of photofluidity can be treated in a systematic way and that the resulting model gives a good estimation of the kinetic and the strain field of the giant photoexpansion observed experimentally.

2. Micro-mechanical model of photoexpansion via a simple thermodynamical approach

2.1. Eigen strain

Taking into account the physics of the photoexpansion, we assume that viscosity and photoexpansion phenomena induce respectively two so called "eigenstrains": the viscous-strain, $\in^{v}$, and the photoexpansion strain, $\in^{p}$. As suggested by Mura [29], eigenstrain refers to such a category of nonelastic strains such as thermal expansion or phase transitions which can induce a self-equilibrated stress field called "eigenstress fields" without any external force and surface constraint. We will not deal with the structural origin of the eigenstrain due to photoexpansion, but only with the resulting internal stress fields.

2.2. Model basis

2.2.1. Athermal nature

The photoexpansion is assumed to proceed continuously and quasi-statically under permanent irradiation; the kinetic of this evolution is described by a single time parameter (τ_{A0}). The eigenstrain saturates at long time, and this saturation level is taken to be proportional to the local light intensity. In this work, we will neglect the thermal aspect of the photoexpansion and we will not sort out the question of the physical origin of the corresponding eigenstrain. In fact, we will show here quantitatively that thermo-viscoelastic coupling and heat conduction can be disregarded, as a first approximation. We also assume that photoexpansion occurs in a viscoelastic medium. Indeed, under irradiation, chalcogenide glasses undergo a large and athermal viscosity decrease named photofluidity or photoinduced fluidity [11]. The viscosity under irradiation is inversely proportional to the number of photons absorbed per second per atom [30,31] and does not depend on the stress [32]: this further justifies the assumption that irradiated glasses undergo a linear viscoelastic behavior. The glass structure is also assumed to be isotropic. For elasticity, the values of the bulk modulus k and of the Poisson's coefficient ν given from [33]:

$$k = 10.2 \pm 0.2\ \text{GPa} \quad \nu = 00.307 \pm 0.01 \tag{1}$$

will be adopted for this study.

2.2.2. Self-focusing

Since the glass undergoes photoinduced refractive index changes, the glass sample may focus the laser beam along the thickness through a photoinduced self-focusing process [14]. This effect will be neglected in the present model. It was shown that the self-focusing is prominent when the ratio sample thickness/beam radius is much larger that 10 (Ref. [14]). In our experiment, this ratio is 9.3. The divergence of the laser beam due to the photoexpansion will also be neglected. Indeed, during the expansion, the average divergence of the beam is lower than 2°.

2.3. Model approach

A detailed mathematical description of the proposed model is presented in Supplemental Material section. Here we describe the general approach adopted to quantify the mechanical response of the glass under irradiation. First the free energy per unit volume is expressed within the framework of the Generalized Standard Model [34] using two distinct tensors for photoexpansion and viscous flow which are treated as uncoupled. Dissipation due to viscous strain rate is also accounted for using a dissipation potential. These yield a state law for the stress which provides the stress-strain response under irradiation. Photoexpansion has only spherical contribution to straining and viscosity plays a deviatoric role, i.e. only shear viscosity comes into play. The problem is treated as rotationally symmetrical around the z-axis of the beam.

3. Experimental

3.1. Glass synthesis

The giant photoexpansion has been investigated on $GeSe_9$ glasses. The glass production procedure is detailed in Ref. [35]. A glass rod was sliced and cut to the desired testing specimen geometry using a diamond saw. The surfaces of the specimens were mirror polished using SiC paper and alumina suspension with 0.25 μm particle size. The sample thickness was 1.5 ± 0.02 mm. The glass transition temperature of $GeSe_9$ is 92 °C (Ref. [30]).

3.2. Irradiation

A tunable Ti-sapphire laser 3900S from Spectra Physics was used in continuous mode for all the experiments. The Ti-sapphire was pumped with a 5 W, 532 nm laser. The Ti-sapphire laser was tuned to 790 nm so that the excitation wavelength corresponds to the Urbach edge of the $GeSe_9$ sample. The photon energy is 1.57 eV, far below the bandgap Eg=1.95 eV (Ref. [30]) measured as the photon energy corresponding to an absorption of 1000 cm^{-1}. At this photon energy, the absorption coefficient is lower than 10 cm^{-1} even if the photodarkening is taken into account [30]. The collimated laser beam was perpendicular to the surface and was not focused on the sample. The samples were irradiated for various periods of time, and the profile of the permanent photoexpansion was collected using a stylus profilometer Dektak 6M with a 12.5 μm radius diamond tip a few minutes after the irradiation periods. The samples were glued on a metallic support with a hole in the middle, so that no reflection of the laser beam occurred on the metallic support. The power of the laser beam was set at P=33.8 mW using 2.0 and 0.2 neutral density filters with a rotationally symmetrical intensity profile. The laser beam profile was characterized and is plotted on Fig. 1. This experimental profile is modeled by a Gaussian fit according to Eq. (2).

$$I(r) = I_0 exp\left[-\frac{2 \bullet r^2}{R^2}\right] \tag{2}$$

where I0=2P/πR2=82.45 W cm^{-2} is the maximum intensity and R=1.615×10^{-2} cm the radius of the beam.

The saturation level of the eigenstrain corresponding to photoexpansion is then taken to be proportional to the incident intensity and