Lehigh University
Center for Optical Technologies

*Electronic Origin of Photoinduced Reversibility in a-As_xSe_{1-x} Glasses**

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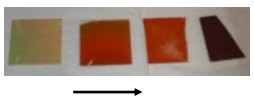
OBJECTIVE

Understand the origin of reversible and anisotropic effects in arsenic selenide glasses using synchrotron XPS

BACKGROUND

Chalcogenide Glasses:

Compounds of S, Se and Te. Most common examples are Se or the selenides and sulfides of As, Ge and Sb.

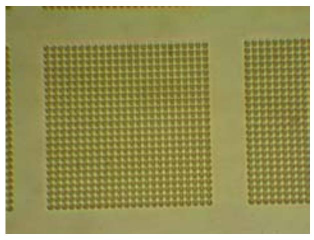


$As_x(Se/S)_{1-x}$ glass films on quartz substrate.

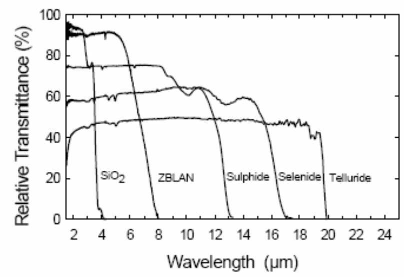
Applications:

These glasses have many optical **applications** because of their high refractive index (~ 1.75 – 2.5), high nonlinearity and uncommon photosensitivity.

Phase-change optical memories e.g., DVDs etc.



Microlenses

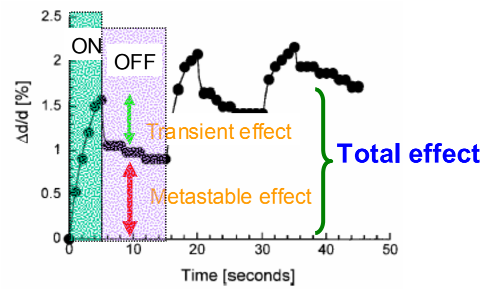


IR detectors, optical fibers
Transmission in mid-IR region

Properties:

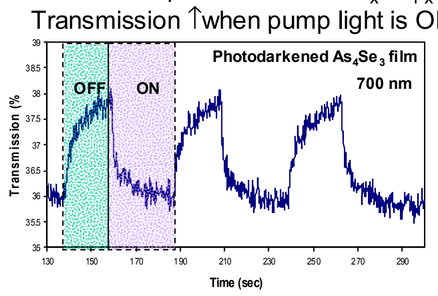
Illumination with light of energy ≥ Eg, → Reversible and permanent changes in volume, refractive index, optical band gap, etc.

Light-induced changes in film thickness with time (after A. Ganjoo et al., Appl. Phys. Lett., **74**, 2119 (1999) *)

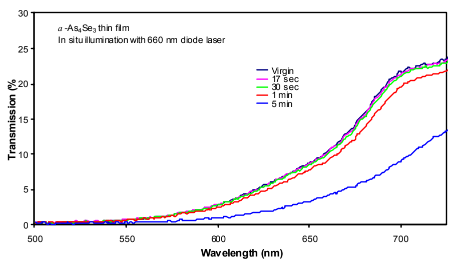


Optical switching!

Observed in photodarkened As_xSe_{1-x} film
Transmission ↑when pump light is OFF



Photodarkening – a permanent change as marked by a shift in the absorption edge towards higher wavelength.



The optical band gap of the material has decreased.

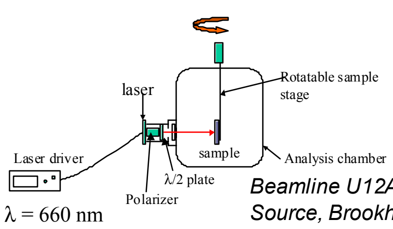
NOTE: For As_xSe_{1-x}, the magnitude of the decrease in the optical band gap is very large ~ 0.26 eV

XPS

Photoinduced effects in the ***optical properties*** of this class of materials are mainly due to the changes in their electronic structure!

X-ray photoelectron spectroscopy (XPS) gives quantitative information on (i) the elemental composition of a surface, (ii) the local "chemical environment" of the atoms comprising the surface and (iii) the valence band structure that determines the optical response. From this information one can infer identity of:
the elemental electrons that contribute to the different types of effects observed
→ the formal oxidation states from the observed chemical shifts

EXPERIMENTAL



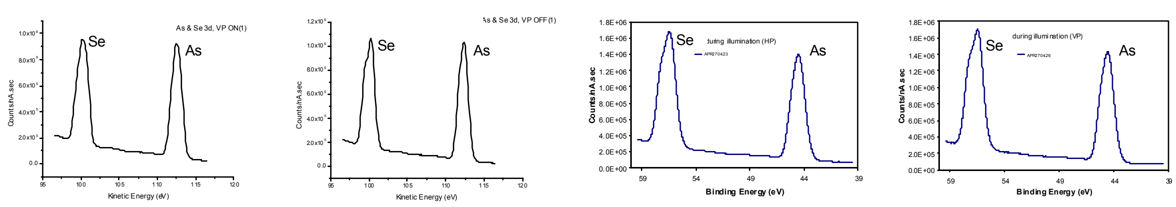
For in situ measurements of photoinduced effects, including the reversible component of photodarkening and photoinduced optical anisotropy

Beamline U12A at National Synchrotron Light Source, Brookhaven National Laboratory

RESULTS

In situ reversibility experiments

In situ anisotropic measurements



Laser ON, VP | Laser OFF, VP | HP | VP

Obvious light-induced changes are observed around the Se atom. In the reversibility measurements, the FWHM of the Se peak increases for the laser ON condition, and decreases for the laser OFF condition. In the anisotropic measurements, again the changes occur around the Se peak – the peak is broader for the light in horizontal polarization. These results indicate the importance of the chalcogen atom in the light-induced effects that we observe on a macroscopic scale.

CONCLUSIONS

1. XPS can be used to identify the atoms that are responsible for the light-induced effects that are characteristic of chalcogenide glasses
2. It appears that it is the electrons around the Se atom (the chalcogen atom) rather than the electrons around the arsenic atom that are responsible for the light-induced effect.