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Light-induced Vector Changes in the Local Structure of As-Se Glasses

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Beamline: X18B

Introduction: Many properties of semiconducting glasses based on chalcogens are known to be sensitive to the light of energy $h\nu \approx Eg$, where ν is the frequency of light and Eg is the bandgap energy. Among such changes, the light-induced mass transport [1], photo-crystallization [2], and an opto-mechanical effect [3], which imply a change in the atomic structure that depends also on the ***polarization*** of light, are particularly intriguing. However, the atomistic origin of these vector effects is not clear.

Methods and Materials: We have been investigating the atomistic origin of these so-called *vector* effects (because they depend on the direction of the electric field vector of light, E_L) by extended X-ray absorption fine structure (EXAFS) analysis around Se as well as As atoms in As_xSe_{1-x} (x=0.4, 0.5, 0.57) films. For observing a vector (or anisotropic) property the probe should also be anisotropic. The plane polarized synchrotron radiation (the electric field (E_X) of the X-rays is parallel to the ground plane) has allowed the identification of the direction of changes in local structure. Since the photo-induced changes can be temporary and/or permanent, we determined the structure under *in situ* conditions such that the laser and the X-ray beam overlapped on the sample [4].

Results: The results of data analysis for a-$As_{40}Se_{60}$ film are shown in Fig. 1, where the sample stage AP means as-prepared film, ON means laser is turned on, and OFF means laser is turned off. Fig. 1(a) shows the results of nearest neighbor (NN) bond distance for Se atoms under different polarized light illumination. Here HP means $\mathbf{E_L}$ is parallel to the ground (the polarization plane of the x-rays), and VP means $\mathbf{E_L} \perp$ the ground. Fig. 1(b) shows the change in local disorder (MSRD) around Se atoms. The error bars for these parameters represent the actually observed range of maximum scattering within six consecutive scans during the AP stage. The unusually high accuracy of the data is due to the very high signal to noise ratio within the transmission mode and the fact that there was no physical change (in sample configuration) within a given set of experiment except for the change of polarization of light beam. From fig. 1(a) we can clearly see that the NN bond distance increases during light illumination, and it remains the same after the laser is turned off. It is remarkable that the increase in R also depends upon the polarization of the light: HP light creates greater expansion than the VP light does. There is also expansion around As atoms, but its polarization dependece is not discernable within the experimental error (data not shown). From Fig. 1(b), we find that the light creates transient disorder around Se atoms, and it is almost completely recovered after the light is turned off. The MSRD change around Se atoms is apparently polarization dependent.

Conclusions: In conclusion, we have found the light polarization-dependent local expansion around Se atoms in a-$As_{40}Se_{60}$ film. The effect is very similar to what we found earlier in a-$As_{50}Se_{50}$ films. Anisotropic local expansion around Se atoms occurs as bandgap light reorients Se electron lone pairs, and helps form anisotropic As-Se bonds. Such polarization dependent change in structure is not discernable for as-prepared a-$As_{57}Se_{43}$ and annealed a-$As_{40}se_{60}$ films that have higher local order than the as-prepared $As_{40}Se_{60}$ and $As_{50}Se_{50}$ films. Our experiments provide the first atomistic view of photo-induced vector effects in chalcogenide glasses.

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References:

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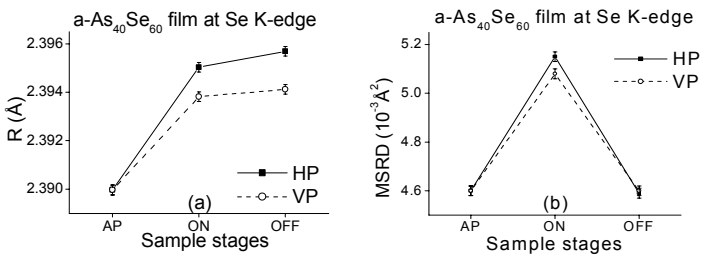

Figure 1: (a): Se NN bond distance and (b) Se NN MSRD for different sample stages and different polarizations of the laser light. AP: as-prepared; ON: laser on; OFF: laser off; HP: horizontal polarization; VP: vertical polarization.