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## Ultrafast defect dynamics: A new approach to all optical broadband switching employing amorphous selenium thin films

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Optical switches offer higher switching speeds than electronics, however, in most cases utilizing the interband transitions of the active medium for switching. As a result, the signal suffers heavy losses. In this article, we demonstrate a simple and yet efficient ultrafast broadband all-optical switching on ps timescale in the sub-bandgap region of the a-Se thin film, where the intrinsic absorption is very weak. The optical switching is attributed to short-lived transient defects that form localized states in the bandgap and possess a large electron-phonon coupling. We model these processes through first principles simulation that are in agreement with the experiments. © 2015 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [http://dx.doi.org/10.1063/1.4927543]

Development of next generation high-speed switching and logic circuitry based on conventional silicon semiconductors is limited due to the long diffusion times of the charge carriers. Optoelectronic conversions can help to achieve high bit rates, yet several limitations must be overcome due to complications in the integration of optical and electronic components. An alternative technology to overcome this limitation is an all photonic switching, in which a control light regulates the signal light through a passive non-linear medium. In all-photonic switching, until now, the passive non-linear medium has been based on complex quantum technologies such as plasmonic nanosystems,<sup>1,2</sup> quantum dots,<sup>3,4</sup> nanoscale photonic metamaterials<sup>5,6</sup> and multiple quantum wells,<sup>7</sup> etc. The main shortcomings are their narrow bandwidth and the difficulties in the integration of these complex structures.

Most ultrafast optical switches demonstrated hitherto are based on interband absorption. Consequently, the signal suffers heavy losses in the switching device and hence is difficult to use in a multistage all photonic optical chip, which contain many switches cascaded as that of an electronic chip. Furthermore, it is quite intricate to efficiently couple signal photons between the complex quantum optical switches and waveguides. In this context, amorphous materials are ideal, because of their inherent ability to interact strongly with light<sup>8</sup> and their non-crystalline structure enables hybrid integration. Amorphous chalcogenide glasses (ChGs) have emerged as promising candidates for all-photonic devices due to their high Kerr and third-order optical non-linearities that are two to three orders of magnitude greater than silica.<sup>9,10</sup> Apart from this, ChG also exhibits unique light-induced effects of fundamental interest like photoexpansion,<sup>11</sup> photofluidity,<sup>12</sup> which makes them versatile platforms for optoelectronics, waveguide writing, patterning, and nanofabrication.<sup>13–15</sup> In ChG, Kolomiets *et al.*<sup>16</sup> and Matsuda *et al.*<sup>17</sup> have demonstrated the transient change in photocurrent. Notably, Yumasheve *et al.* showed that these light-induced effects can be attributed to the excitation of electron-hole pairs and thereby enhancing the concentration of localized trap states in the gap.<sup>18</sup> Among ChG, a-Se has attracted much attention in photonics because of its potential

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applications in many diverse fields ranging from high gain avalanche rushing photoconductor in harpicon tube to capture images at extremely low light intensities, x-ray photoconductor for digital radiographic applications in medical diagnosis and photoreceptor in photocopying machines.<sup>19</sup>

In this article, we demonstrate a simple and yet efficient ultrafast broadband optical switching in a-Se prototype with ps timescale in the weakly absorbing regime. The ultrafast switching is attributed to short-lived transient defect dynamics in the optical gap. Concomitantly, we have performed a first-principle calculation of the time resolved imaginary part of the complex dielectric constant (absorption) of the unexcited and photo-excited system and found that the simulation results are in good agreement with the experimental data.

Optical switching requires that a control beam substantially changes the signal amplitude. In our scheme, this is achieved by induced absorption (IA) by fs laser. To observe the results experimentally, we started with  $\sim 0.4 \ \mu m$  thick a-Se thin film deposited on a quartz substrate by the conventional thermal evaporation technique. The substrate was kept at room temperature. To study the IA, we employed a pump-probe optical absorption spectrometer. In our experimental setup, 120 fs pulses centered at 800 nm with a repetition rate of 1 kHz, was split into two beams to generate the pump and the probe beams. The first beam was passed through an optical parametric amplifier to generate the pump beam. The second beam was delayed with a computer-controlled motion controller and then focused into a  $CaF_2$  plate to generate a white light continuum (450-850 nm) and was used as the probe beam. The pump and probe beams are spatially overlapped on the sample and the IA of the probe beam  $\Delta \alpha = \Delta A/d = (-1/d)\log[I_{ex}(p)/I_0(p)] - \log[I_{ex}(r)/I_0(r)]$ was calculated. The symbols p and r correspond to the probe and the reference, respectively, d,  $I_{ex}$ and  $I_0$  are the thickness of the sample, transmitted intensities of the sequential probe pulses after a delay time  $\tau$  following excitation by the pump beam and in the ground state respectively. The time resolution of our experiment is 130 fs as determined by the solvent two photon ( $hv_{pump} + hv_{probe}$ ) absorption by using cross-correlation of pump and probe pulses.<sup>20</sup> The kinetic traces have been chirp corrected using a procedure reported elsewhere<sup>20</sup> using ethanol as the solvent.

To study the defect dynamics, we have recorded the transmission spectrum of the sample and is shown in Fig. 1. The bandgap of the sample is calculated as 2.1 eV (590 nm) from the Tauc plot (inset Fig. 1). For this, we have chosen the region of the transmission spectrum where the absorption coefficient is more than  $10^4$  cm<sup>-1</sup>. To probe the defect dynamics with bandgap excitation, we excited the sample with 560 nm, 120 fs pulses with an average fluence of 500 µJ/cm<sup>2</sup>. Fig. 2(a) shows the contour plot of IA at different time scales and for additional insight, several cross sections of the contour, that is, IA at different time delays are plotted in Fig. 2(b). As can be seen in these figures, the most prominent features of IA are: (1) strong and fast decaying IA at sub-bandgap region (above 650 nm) and (2) weak and slow decaying IA at the bandgap region (560-650 nm).



FIG. 1. Optical transmission spectrum and inset Tauc plot to determine bandgap.

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FIG. 2. (a) Contour plot of  $\Delta \alpha$  followed by the pump beam excitation at 560 nm and (b)  $\Delta \alpha$  at various time delays between pump and probe beams. (c) Contour plot of  $\Delta \alpha$  for 400 nm excitation and (d)  $\Delta \alpha$  at various time delays. Temporal evolution of IA for (e) 590 nm and (f) 730 nm.

To study the wavelength dependence of IA, we have excited the sample with above bandgap light of wavelength 400 nm, with average fluence and pulse width same as that of bandgap excitation. As can be seen in the Fig. 2(c) & 2(d), the spectral characteristics are similar to that of bandgap excitation— IA is dominant at the sub-bandgap region, and as we move towards bandgap region, it get weakens. Strikingly, with above bandgap excitation, IA is  $\approx 3$  times stronger than bandgap excitation, although the spectral features remain the same, the kinetics appears to be slower.

To get new insights on the kinetics of TA, temporal evolution of IA at two different probe wavelengths; one at the TD region (730 nm) and the other at the bandgap region (590 nm) are shown in Fig. 2(e) & 2(f). At these wavelengths, a single exponential decay was unable to fit the experimental data and so we used the following equation<sup>21,22</sup>

$$\Delta \alpha = \frac{i(t)}{d} \otimes \sum_{l=1}^{n_{comp}} A_l(\lambda) . \exp(-t/\tau_l)$$
(1)

where i(t),  $A_l(\lambda)$ ,  $\tau_l$  and  $\otimes$  are instrument response function, amplitude of the exponential decay, decay time constant ( $A_l(\lambda)$  to decay to 1/e of its original value), and the convolution operator respectively. For a-Se, l = 2 model, gives the best fit to the experimental data and the respective fitting parameters are listed in Table I. It can be seen from the table that TA at 730 nm decays faster than 590 nm. This is because the electronic states formed by the TD states at 730 nm are highly localized, and hence they strongly couple to the lattice vibrations and relax faster than the 590 nm states, where such effects are less in the bandgap region.

To theoretically simulate the defect dynamics in the tails and middle of the gap, we began with a 216 atom model of a-Se by Zhang and Drabold<sup>23</sup> and relaxed it to its local minimum using conjugate gradient algorithm<sup>24,25</sup> as implemented by the Vienna Ab-Initio Software Package, VASP.<sup>26,27</sup>

Excitation (nm)	Probe wavelength (nm)	$\Delta\alpha_1(\lambda)(cm^{-1})$	$\Delta \alpha 2(\lambda)(cm^{-1})$	$ au_1(ps)$	$\tau_2(ps)$
560	590	17.5	7.3	15	269
	730	100	25	6.6	318
400	590	50	34	15	8100
	730	325	52	6.8	1800

TABLE I. Fitting parameters calculated from Equ. (1).

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FIG. 3. (a) The EDS (black curve) and IPR (green lines) calculated for our model. The red line marks the position of Fermi energy. (b) Evolution of states around the gaps after the electron is excited from VB to CB. Only states neighboring the gap are shown. (c)  $\Delta A$  as a function of wavelength at different times. The inset shows  $\Delta A$  as a function of time at different wavelengths.

The relaxed model is quite realistic and appears to correctly reproduce structural, electronic and vibrational properties of a-Se.<sup>22</sup> Topologically, it consists of two-fold Se-atoms forming chains and has two coordination defects, a 3-fold Se-atom adjacent to a 1-fold Se, making it a suitable model to follow the light-induced defect dynamics. The electronic density of states calculated for our model accurately predicts the band structure reported earlier<sup>28</sup> (Fig. 3(a)). Besides, it registers gap states originating from the defects, which have been predicted by the work of Vanderbilt and Joannopoulos<sup>29</sup> and many others. We probe the localization of these states by calculating inverse participation ratio (IPR) (Fig. 3(a)) and observe highly localized band tail and gap states. All these clearly indicate that what we simulate is a realistic computer model of a-Se.

A full non-adiabatic simulation of photo-excitation of the amorphous model we created is indeed a very challenging problem, and hence we treat that here in an approximate way. In our scheme, we view the light as inducing promotions from occupied to unoccupied states. If the system is initially at equilibrium, it will not be after the promotion since there will be Hellman-Feynman forces associated with the newly occupied state. While this change of occupation is a proxy for what is fundamentally a very complex process, it has the key advantage of offering a reasonable prediction of light-induced structural changes since the electron-phonon coupling is realistic.<sup>30,23</sup> Here, we add an electron from an occupied state to an unoccupied state and let the system evolve freely. We monitored the eigenstates (Kohn-Sham orbitals) of our system around the Fermi-energy and observed the closing of the gap as the lowest unoccupied molecular orbital (LUMO) level descends. The levels we explore are initially above the Fermi level and, therefore, unoccupied. The additional electron occupies the level, and the response of this state to drop toward the valence edge (due to the change in occupation to neutral or negative if one or two electrons are added). The relaxation is very rapid, of order 1 ps (Fig. 3(b)). The relaxation times are also in close agreement with previous molecular dynamic simulations by Hoshino *et al.*<sup>31</sup> which shows the stabilization of antibonding LUMO state on the basis of light-induced bond breaking at 500K which also formed the theoretical basis of semiconductor-metal transition.

A detailed mechanism leading from electronic excitations to defect creation is elusive. As our first principle simulation suggests that excited carriers are trapped in its own lattice deformation the lattice is not stable upon creation of the electron-hole pair. Such a self-trapping process occurs in a-Se since the non-bonding lone pair orbital possesses strong electron-phonon coupling, which strongly favors the formation of low energy defect pairs.<sup>32</sup> This process can be understood in the following way. When illuminated with bandgap light, the electrons, and holes are separated by a distance (R) which is lower than the critical distance R<sub>C</sub>, known as the Coulomb capture (Onsager radius), defined as

$$e^2/4\pi\varepsilon\varepsilon_0 R_C = kT_g \tag{2}$$

where e,  $T_g$ ,  $\varepsilon$ ,  $\varepsilon_0$  and k are the electronic charge, glass transition temperature, dielectric constant of the sample, dielectric constant of the air and the Boltzmann constant respectively. For a-Se,  $R_C$  according to equ. (2) is ~ 83Å ( $\varepsilon \approx 6$  and  $T_g \approx 313$  K). On the other hand, the effective mass approximation model predicts electron-hole separation in the ground state is ~3 Å<sup>33,34</sup> and upon excitation it increases by a factor  $\sqrt{\beta t}$ , where  $\beta$  is the diffusion constant and *t* is thermalization time. Taking  $\beta \approx 1 \times 10^{-3}$  cm<sup>2</sup>/sec<sup>35,36</sup> and *t* is of the order of few ps (the fast decay constant in our experimental data) we find  $\sqrt{\beta t} \approx 7$  Å. Thus R increases to  $\approx 10$  Å and since R < R<sub>c</sub>, the excited electron-hole pairs form excitons. Based on the calculations,<sup>34</sup> it can be seen that R < R<sub>c</sub>, self-trapped exciton mechanism is effective in both excitations. Since the formation energy of coordination defects are significantly lower than the bandgap and the exciton energies, they are highly unstable and interact with the lattice to form TD, which exactly corresponds to self-trapping of excitons. Electronic states formed by the TD states in the gap are highly localized and can be seen from our simulated IPR values (Fig. 3(a)). Since the TDs are unstable and strongly couple to the lattice vibrations, they relax faster than the bandgap states. Besides, pump beam illumination creates many TDs, which significantly increases the density of states in the gap. This in turn will generate more pronounced  $\Delta \alpha$  in the sub-bandgap region than there at the bandgap region (Fig. 2 & 3). Though comparable changes are generated at both the regions by light, however in the bandgap region, the changes are much smaller due to the already existing high density of states.

For more insights of the lifetime of TD, we have computed the temporal evolution of the imaginary part of the complex dielectric function of a-Se during and after photo-excitation. Such calculations will give direct evidence of IA as in that of our experiment. For this, we have computed the frequency dependent absorption (complex dielectric function,  $Im(\varepsilon)$ ) by summing over the probabilities of transition from occupied states to unoccupied states in accordance with the model derived in Ref. 37. After that, we calculated the IA spectrum by calculating the difference in imaginary dielectric function before and after excitation ( $Im(\varepsilon)_{excited} - Im(\varepsilon)_{ground}$ ). We show in Fig. 3(c), IA at different times followed by photo-excitation and in the inset the same at selected wavelengths. As can be seen in these figures a broad IA extending from bandgap to sub-bandgap. Clearly, the most prominent features are: (1) fast decaying strong IA band at sub-bandgap region (above 680 nm) and (2) slow decaying weak IA band in the bandgap region (613 nm). Our theoretical and experimental results concur that the fast processes are dominant at sub-bandgap wavelengths whereas the slow component of decay increases as we move towards bandgap wavelengths.

The concept of short-lived defect states can be extended toward achieving very high-speed optical switching, which is otherwise difficult to achieve in bandgap regions. In our scheme, optical switching is visualized through ultrafast transient defect dynamics induced by fs control beam. Strikingly, our results reveal that fast processes are dominant near the sub-bandgap probe wavelengths whereas the slow component of decay increases as we move towards resonant probe wavelengths. As shown in Fig. 4, the excited state (when the IA is high) is termed as 'off' state and the ground state as 'on' state. From Fig 4(a), it can be seen that the IA of 730 nm probe signal switches from 'on' to 'off' state  $\approx$  instantaneously and 'off' to 'on' state within 16 ps, following the pump beam excitation. Fig. 4(b) shows the schematic of the probe beam transition in our switching scheme. Thus, we demonstrate an all-optical switching at ultrafast time scales and can be translated into broader wavelength range.



FIG. 4. Ultrafast all-optical switching scheme. (a) Growth and decay kinetics of IA at 730 nm following the pump excitation. (b) Schematic of switching. The optical switching (on - off state) of the signal beam (red) by control beam (blue) is shown. The signal beam is switched off (not transmitted) instantaneously by the control beam and reinstated within 16 ps.

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In conclusion, we have realized a novel ultrafast broadband all-optical switching on ps timescale in the sub-bandgap region of the a-Se thin film, where the intrinsic absorption is very weak. In simple terms, optical switching in our scheme is understood through ultrafast transient defect dynamics induced by a fs control beam. A first principle calculation shows that the TA maximum at sub-bandgap wavelengths, which is fully reversible in a few ps similar to our experimental results. It, therefore, becomes credible that the concept of short-lived defect states may be extended towards achieving high-speed optical switching.

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