

Competition Between Thermal and Non-Thermal Processes During Femtosecond Switching of Phase Change Materials

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Abstract: We use ultrafast optics and electron diffraction to measure irreversible amorphization of crystalline Ge₂Sb₂Te₅ phase change films. We find that light directly modifies the local bonding environment leading to a hardening of bonds.

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1. Introduction

Phase change materials (PCMs) have metallic-crystalline and insulating-amorphous states, both of which are stable at room temperature. Unusually, the two phases exhibit vastly different electronic properties from dc to optical frequencies, make PCMs useful in optical data storage. The phase transition can be driven incredibly rapidly, with crystallization occurring after a 500 ps current burst [1]. This pushes the switch rates to new extremes and makes PCMs promising candidates for universal memories, where high speed data operations can be performed on the same element used to store the data [2]. In this paper we examine the ultrafast switching of PCMs by femtosecond light pulses and high-light the speed limits for this transformation.

1.1. Resonant bonding and phase transformation

The crystalline phase of PCMs is resonantly bonded with ordered *p*-orbitals [3]. This network is only weakly held together and is particularly susceptible to lattice distortions. This is in contrast to the strong, short-range bonds of the amorphous phase, in which the ions exhibit a different bonding coordination. Transformation between the crystalline to the amorphous phase is typically achieved by heating the PCM above the melting temperature and then rapidly quenching into the amorphous phase by either an electrical or optical pulse. We examine how the transition occurs when ultrashort pulses are used and non-thermal dynamics may be important. By using a combination of single-shot, and very-low-repetition-rate multi-shot optical and electron diffraction experiments, we show that photoexcitation directly destabilizes the crystalline structure. Photo-excitation perturbs the resonant bonding network leading to bond stiffening. Below a critical fluence, the system recovers; however, further excitation can destabilize the lattice triggering a collapse to an amorphous phase.

2. Results

We study 30 nm thin films of the prototypical phase change material Ge₂Sb₂Te₅ on fused silica substrates with a 20 nm capping layer of Si₃N₄. We use a single 800 nm 35 fs laser pulse with a 2 mm diameter focus to measure the permanently induced changes. Fig. 1a shows reflection images of the sample after excitation by a single laser pulse. Three regimes were observed: Region I, pump fluences, $F < 11 \text{ mJ cm}^{-2}$ the system recovers and no permanent change occurs; Region II, $11 < F < 30 \text{ mJ cm}^{-2}$, the system undergoes a phase transformation which is more transparent and less reflective, consistent with the properties of the amorphous phase; Region III, $F > 30 \text{ mJ cm}^{-2}$, the system ablates.

In order to understand the dynamics of these processes, we use a 10-ps-long linearly-chirped probe pulse and spectral encoding to map the temporal dynamics onto the frequency domain of our probe pulse. This enables us to measure 5 ps transients with a single laser pulse with femtosecond time resolution. Fig. 1b shows the time dependent change in the transmission and reflection arising from a single pump pulse. In region I, the dynamics are linear and exhibit large

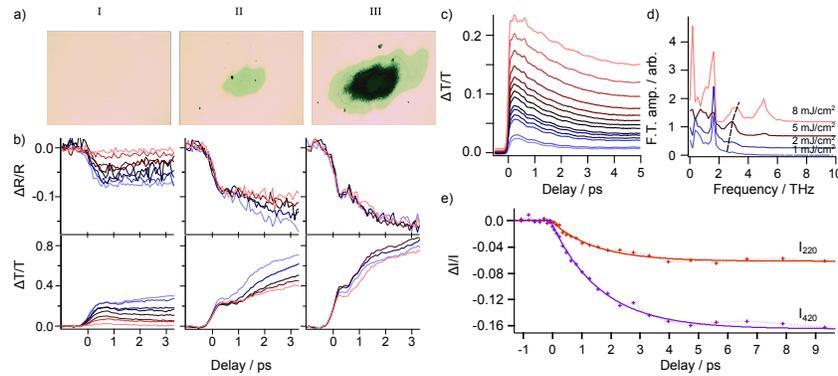


Fig. 1. a. Reflection images of the sample after excitation with single laser shot showing three regimes: I. no change, II amorphization, III ablation. b. Single-shot dynamics of the reflectivity and transmission during the transition process. c. multi-shot time resolved transients in region I. from 1 to 8 mJ cm^{-2} . d. Fourier transform of c after background subtraction. Dashed line indicated mode that hardens under excitation. e. Electron diffraction data showing a loss in crystalline lattice peak intensity for the (220) and (420) Bragg peaks in region I

and rapid changes followed by slow recoveries. In region II, we observe a marked difference in the evolution of the properties, with changes saturating during the first picosecond but continuing to evolve on longer timescales. Region III shows that the signal at later delays saturates sooner for increasing pump power.

To investigate the nature of the phase transition, we also perform multi-shot optical and electron diffraction experiments to improve our signal to noise close to the single shot transition threshold. In order to access the region close to the single-shot transition threshold, we operate at 40 Hz repetition rate to avoid accumulative heating, and no permanent change is induced. Fig. 1c shows that photoexcitation generates coherent phonons on top of an exponential decay. Fitting this decay gives a time constant of approximately 2 ps. After subtracting the slow background and performing a Fourier transform we see that there are three phonon modes, as shown in Fig. 1d. The intensities of these modes show a non-linear power dependence and we observe one mode stiffen with increasing photoexcitation. In addition, we perform electron diffraction measurements, shown in Fig. 1e, which show that the 2 ps time constant is related to the reduction of the Bragg peak intensities and the rapid thermalization of the electrons with the lattice.

3. Conclusions

These results demonstrate how femtosecond optical excitation drives the phase transition. On the ultrafast timescale photoexcitation modifies the lattice potential resulting in harder bonds. These bonds prefer an amorphous phase coordination and at sufficient fluence trigger the phase transition. Similar hardening is observed in highly excited gold [4] and phase transformations based on a change in internal unit cell potential are seen in VO_2 [5, 6]. However, although this lattice potential modification happens on the ultrafast timescale, electron diffraction indicates that electron-lattice thermalization occurs over a few picoseconds. As a result, thermal processes ultimately dictate which final state is adopted, thus placing important restraints on ultrafast switching in PCMs.

References

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