Probing Ultrafast Optically-Induced Structural Changes in $Ge_2Sb_2Te_5$ using a Free Electron Laser

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ABSTRACT

Recently, low-energy non-thermal switching based upon optically-induced coherent phonons generated by ultrafast femtosecond optical pulses in interfacial phase-change memory has been reported. While the state-change measured by reflectivity was permanent, the underlying mechanism can only be speculated upon. To understand the collective atomic motion that occurs in response to ultrafast optical pulses and its relationship to phase-change, we have initiated pump-probe studies using an optical pump and ultrafast optical pulses from a free-electron laser. We use diffraction to understand the dynamics changes of the lattice in response to ultrafast optical pulses. Here we report on our initial results of lattice dynamics

Key words: ultrafast, coherent phonon generation, time-resolved structural dynamics

Makino et al. recently reported upon optically-induced crystallization of interfacial phase-change memory using coherent phonons generated by 800 nm ultrafast laser pulses from a Ti-sapphire laser system [1]. Although the crystallization temperature of $Ge_2Sb_2Te_5$ (GST) is known to be approximately 150°C, crystallization was achieved using coherent phonons with an average temperature rise of less than a 1°C. The switching was achieved by selective excitation of a single optical phonon mode with a period of 270 fs. This suggest the presence of an athermal pathway between SET and RESET states that may allow the fabrication of more energy-efficient and potentially faster switching devices. The presence of a non-thermodynamic pathway between SET and RESET states has also been demonstrated theoretically in GeTe where selective motion reminicent of an optical phonon-like displacement lead to the spontaneous switching of the SET into the reset state without heating [2]. The same report also emphasized the importance of the bond energy hiearchy present in phase-change materials lying along the GeTe-Sb_2Te_3 pseudobinary tie-line in enabling the non-thermodynamic pathway to exist. Finally, earlier optical pump-x-ray probe work observing structural changes in GST induced by 500 ps laser pulses reported that the RESET state could be achieved without passing through the molten state [3]. As the time resolution of this experiment was on the order of 500 ps, the experiment could not provide details on processes occuring on time scales less than 150 ps.

In the current paper, we report on a new series of optical pump/x-ray probe experiments to observe atomic motion in phase-change materials with a time-resolution as low as 30 fs enabling the tracking of the atomic displacements of individual phonons. A thin film epitaxial layer of GST grown on a Si (111) substrate was optically pumped by a 30 fs 800 nm optical pulse generated by a chirped optical amplifier for a variety of different fluences. Epitaxial layers were used in light of the unexpected observation that epitaxial layers switched into the amorphous state by ultrafast pulses could be recrystallized in the the epitaxial SET state by a few ultrafast optical pulses of lower power [4]. In contrast, although recrystallization by ultrafast pulses is possible for polycrystalline GST films as well, hundreds of pulses are required suggesting a different, possibly thermal, recrystallation mechanism. In the current experiment,

the induced structural changes were probed by 12 keV (1.03 Å) x-ray pulses produced by the free-electron laser facility SACLA at SPring-8. The duration of the x-ray pulses was 10 fs allowing probing of collective atomic motion with a resolution sufficient to monitor the strain field of coherent phonon induced displacements for even the highest frequency modes by monitoring changes in the diffraction pattern. In these initial experiments, the laser and x-ray pulses were directed in a nearly coaxial arrangement and the 111 and 222 reflections were monitored. The laser was triggered using the RF signal from the linac synchronously and the relative delay between the optical pump and the x-ray probe could be varied using an optical delay line. The changes in the diffraction signal position and intensity were monitored by a CCD system and diffraction data for each pump-probe pair was read out and individually saved. Here we report on observations of the strain field detected for acoustic phonons induced by the optical pump signal with a time-resolution of 2 picoseconds. We have also carried out optical pump-probe measurements on the same epitaxial layers and have observed both acoustic and optical coherent phonon modes. The period of the acoustic mode observed was approxiately 15 ps (0.067 THz). We have modeled the strain field induced in the GST layer by the optical-pump pulses by solving the normal mode problem in one dimension and have used the computed strain field to compute the expected diffraction signal by numerical solution of the Takagi-Taupin dynamical x-ray equation. The predicted time-dependent change in diffraction intensity and position was in good agreement with the observed time-dependence of the x-ray diffraction signal. We plan to further increase the time-resolution of these experiments to allow observation of individual optical phonon modes in the near future.

REFERENCES

[1] K. Makino *et al.*, Opt. Express **19** (2011) 1260.
[2] A. V. Kolobov *et al.*, Nature Chem. **3** (2011) 311.
[3] P. Fons *et al.*, Phys. Rev. B **82** (2010) 041203.
[4] P. Rodenbach *et al.*, Appl. Phys. Lett., **101** (2012) 061903 .

Biography

Kirill Mitrofanov graduated from Ryazan State Radio Engineering Institute in 2008. In 2012 he defended his thesis on chalcogenide semiconductors phase change process parameters. From the August of 2012 he began work as a postdoctoral fellow in the functional materials group at the National Institute of Advanced Industrial Science & Technology (AIST) located in Tsukuba, Japan. He is current studying coherent phonons in phase-change materials using ultra-fast time-resolved diffraction and a hard x-ray free electron laser.

Paul Fons graduated from the University of Illinois at Urbana-Champaign in 1990 writing his thesis on the growth and electronic structure of metastable semiconductor alloys. Subsequently, he became a research fellow at the University of Tsukuba in Japan. In 1993, he joined the National Institute of Advanced Industrial Science & Technology (AIST) also located in Tsukuba. While at AIST, he has carried out extensive work in molecular beam epitaxy, photoluminescence, and structural measurements using synchrotron radiation including diffraction, x-ray absorption, and high-pressure, and ultra-fast time-resolved synchrotron studies as well as density-functional theory calculations. He is currently working in the functional materials group at AIST on phase-change materials.