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A Density Function Investigation of Excited-State Effects due to Ultrafast Excitation in $Ge_2Sb_2Te_5$ Epitaxial Films

Paul Fons[1,4], Kirill Mitrofanov[1,4], Kotaro Makino[1], Ryo Terashima[2], Alexander V. Kolobov[1], Junji Tominaga[1], Alessandro Giussani[3], Raffella Calarco[3], Henning Riechert[3], Tetsuo Katayama[5], Takahiro Sato[4], Kanade Ogawa[4], Tadashi Togashi[5], Makina Yabashi[4] and Muneaki Hase[1,2,4]

[1]Nanoelectronics Research Institute, AIST, Tsukuba Central 4, Higashi 1-1-1,Tsukuba 305-8562, Japan.
[2]Institute of Applied Physics, University of Tsukuba, 1-1-1 Tennodai, Tsukuba 305-8573, Japan.
[3]Paul-Drude-Institut fur Festkorperelektronik, Hausvogteiplatz 5-7, 10117 Berlin.
[4]RIKEN SPring-8, XFEL Research and Development Division, 1-1-1 Kouto, Sayo-cho, Hyogo 679-5148, Japan.
[5]XFEL Project Head Office, Japan Synchrotron Radiation Research Institute,
1-1-1 Kouto, Sayo-cho, Hyogo 679-5148, Japan.
E-mail: paul-fons@aist.go.jp.

1. Introduction

As phase-change materials such as $Ge_2Sb_2Te_5$ (GST) undergo dramatic changes in material properties due to a fundamentally local change in bonding behavior, the ultimate speed limits of the atomic switching process offer hope both for ultrafast switching as well as non-adiabatic energy efficient routes for memory applications [1, 2]. To this end, we have carried out experiments using the free-electron laser SACLA to explore sub-picosecond lattice dynamics in epitaxial GST (111) in response to 30 fs optical pump excitation. The SACLA x-ray beam was used to probe the lattice dynamics using a symmetric diffraction geometry.

2. Results and discussion

A set of a rocking curves for time delays close to time zero was obtained to determine the crystal strain evolution data associated with the propagation of a coherent acoustic phonon (CAP). A dynamic strain model indicates the need for large differential stress inconsistent with that provided by thermal effects alone due to the lack of observed decrease in rocking curve intensity for short delays. It strongly suggests that non-thermal excited state effects are responsible for the generation of the CAP. The presence of p-electron dominated bonding with only 3 e^-/atom (so-called *resonant* bonding) makes the system unusually susceptible to excited-state effects. We have calculated the effects on both cell size, the associated bonding changes, and the modified lattice dynamics using constrained density-functional theory and the ab initio code VASP. We have used GeTe as an approximant for the more complicated GST structure as pump-probe measurements on GeTe show a similar behavior. The results suggest that excitation of even a small percentage of valence electrons leads to significant changes in cell volume leading to a displacive induced generation of coherent phonon-like mechanism. Lattice dynamics also demonstrate significant softening for comparable excitation densities consistent with optical coherent phonon measurements suggesting that the thermalization of the hot carriers may be prolonged due to a reduction in the vibrational density of states at the zone center.

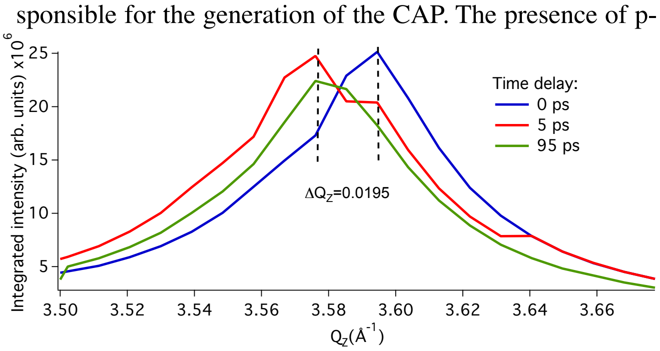

Fig. 1. A set of rocking curves for GST sample at different time delays. Laser pulse fluence is 13.4 mJ/cm^2.

3. Conclusions

Density-functional calculations suggest that the effects of ultrashort pulse excitation on epitaxial GST have pronounced effects on cell volume and potential softening effects as well. These softening effects may be effective in dynamically reducing the activation barrier between the two end states allowing ultrafast switching.

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