Observation of T2z Optical Phonons and optically induced linear birefringence in epitaxial GeSbTe/GaSb films

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ABSTRACT

Key words: $Ge_2Sb_2Te_5$, Epitaxial, Coherent optical phonons, Raman Modes, Impulsive, Displacive

The phonon spectrum is critical for a full understanding of the phase change process in chalcogenide glasses such as $Ge_2Sb_2Te_5$ (GST) used for data storage applications. The time resolved reflectance (R) and anisotropic reflectance (AR) response of a epitaxial $Ge_2Sb_2Te_5$(15 nm)/GaSb(50-100 nm)/GaSb(001) thin film has been explored in optical pump-probe experiments. A linearly polarized, 800 nm wavelength, 80 fs pump pulse with fluence of 0.42 – 2.77 mJ/cm^2 at close to normal incidence was used to excite the sample. The dynamics of ultrafast hot carriers and stimulated coherent optical phonons (COP) was monitored by a s-polarized probe pulse of 800 nm wavelength, 80 fs pulse with weaker fluence of 0.2 mJ/cm^2 incident at 45°. The time resolved response was measured upto 4 ns delay. The pump polarization and/or orientation of the sample were varied in order to investigate the character of the COPs (amplitude/frequency/phase) and a long-lived transient response. Additional measurements were performed upon a reference substrate (GaSb) without the GST layer in order to understand the origin of the various features observed. For the case that the probe was polarized parallel to the [100] axis and at 45° to the pump, the AR signal revealed an initial peak of 115 fs width due to the specular optical Kerr effect (SOKE), COPs with frequency of 3.4 and 6.7 THz, and a large transient response that decayed on ns timescales. After exposure to fluences in excess of 2.12 mJ/cm^2, COPs were instead observed in AR at frequencies of 4.2 THz in both samples and at 3.1 THz for the GST film. To obtain the best fit to entire data, we employ a fitting function consisting of three independent terms: (i) a Gaussian function (for the initial peak) (ii) some exponentially decaying terms (complicated background) and (iii) two damped oscillator terms of the form $A\exp(-t/\tau)\cos(2\pi ft + \varphi)$. Fitting the raw R and AR data provides the information about important parameters, such as amplitude (A), frequency (f) and initial phase (φ). The data also exhibit a number of relaxation times (τ) that can be ascribed to the interaction of the different populations of electron and phonons. The SOKE peak is commonly observed and is associated with the optical modification of the electronic momentum distribution. The polarization dependence analysis of the amplitude of the signals strongly suggests that the 6.7 THz COP and transient AR are associated with the GaSb substrate, which has the zincblende structure and is consequently piezolelectric. The COP amplitude is fully explained by a combination of transient stimulated Raman scattering (TSRS) described by the Raman tensor for zincblende [1], which may be understood microscopically in terms of the orientation of the pump electric field relative to bonds lying in orthogonal (110) and (-110) planes [2], and an additional component that is independent of the polarization of the pump. The latter component may be understood in terms of an optically-induced

space charge field normal to the substrate [3] that also generates a lateral strain that leads to the transient AR response (tail). The variation of the amplitude of the 3.4 THz mode specific to GST suggests that it has T_{2z} symmetry. This frequency is also observed for c-Sb_2Te_3, suggesting the importance of Sb-Te bonds. The T_{2z} mode is usually believed to be Raman inactive for rock-salt, raising questions about the microscopic excitation mechanism (TSRS) and the detailed crystallographic structure of the epitaxial GST. The ordering of Ge/Sb/vacancies and their displacement from their ideal position may break the inversion symmetry and allow both 1^{st} order Raman scattering and space charge excitation. This may lead to additional zone-centre modes of A and E type.

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Biographies

I am Ashawaraya Shalini, final year "Phd student" in Exeter University, working under the supervision of Prof. Robert J Hicken. My research plan is to study $Ge_2Sb_2Te_5$ (epitaxial/polycrystalline/amorphous) thin films optically and to investigate if non-thermal transition exists in phase switching of phase change materials. I am using time-resolved femtosecond optical pump-probe technique to understand the 'Reflectance' and 'Anisotropic Reflectance' signal of the sample upto few ns delay. The response of the alloy to the polarization of the optical pulse is also explored, since the observation of optically induced birefringence provides a better understanding of the non-thermal nature of the transition.

Prior to phd, I have worked in KLA-Tencor (year 2009), a product-based semiconductor company in bay area. This company builds the inspection and metrology tools to help IC manufacturers to manage yield throughout the entire wafer fabrication process from R&D to final yield analysis. I worked as "Field Application Engineer" to demonstrate the macro-defect detection tools (VIPER 24xx series) capabilities to worldwide customers to help them identify and solve their process issues.

I have received M.Tech degree from IIT Delhi, India (year 2005) with master's project under the joint supervision of Prof M Wuttig (RWTH Aachen, Germany) and Prof S C Kashyap (IIT Delhi, India).