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Valley-dependent Carrier and Lattice Dynamics in Silicon measured by Transient XUV Spectroscopy

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Abstract: Transient XUV core level spectroscopy is used to resolve photoexcited electron and hole distributions, as well as carrier-phonon and phonon-phonon scattering times, in the Γ , L, and X valleys of silicon.

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1. Introduction

The tracking of hot carrier distributions and the determination of carrier-phonon and phonon-phonon scattering times is vital to the modeling of electronic devices, especially for high-field transistors and photo-excited devices. The possible scattering processes are well characterized for benchmark materials like Si and Ge, where they have been collectively determined by a range of characterization techniques spanning from time-resolved transient absorption and Raman experiments to temperature-resolved transport measurements. Unfortunately, the myriad characterization techniques required is not conducive to rapidly obtaining the thermalization and scattering-processes in new materials, especially in higher-energy bands not accessible by ground state transport measurements.

Transient extreme ultraviolet (XUV) spectroscopy is a powerful new method that allows energy-resolved measurement of photoexcited electron and hole distributions. Furthermore, the response of the XUV absorption edge is characteristic of the optical and acoustic phonons involved in the carrier thermalization process, allowing the electron-phonon and phonon-phonon scattering times to be determined. By repeating this measurement with multiple pump wavelengths, the carrier and lattice dynamics are separated for the different valleys of Si.

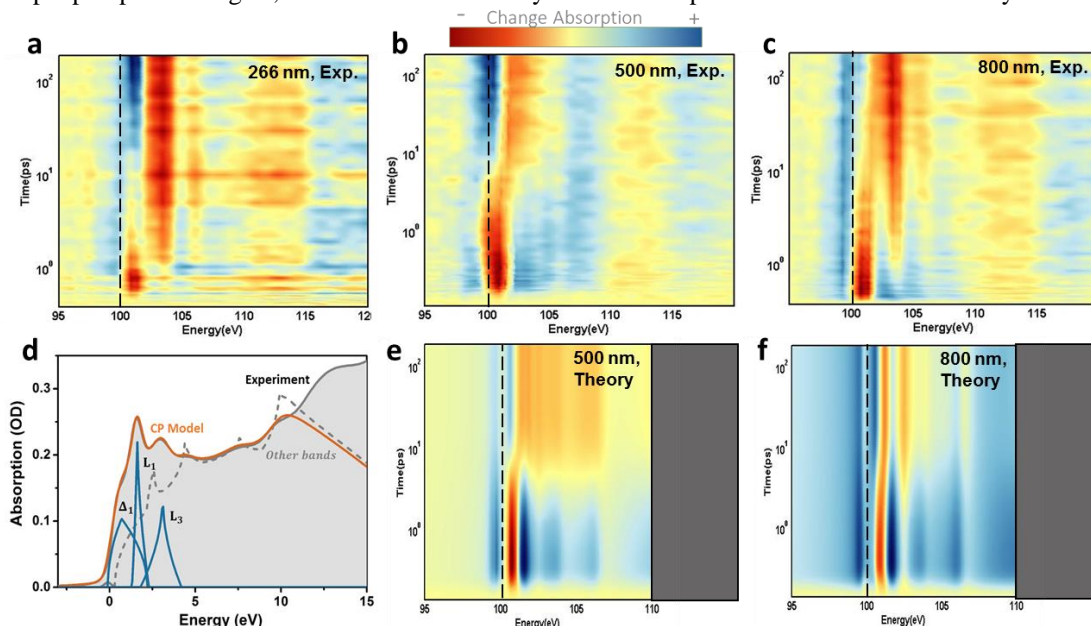


Figure 1. Differential transient XUV absorption for (a) 266 nm, (b) 500 nm, and (c) 800 nm excitation (edge onset shown as dashed line). d, Ground state Si $L_{2,3}$ absorption and critical point model. Theoretical model of differential absorption including broadening, state-filling, and renormalization effects for (e) 500 nm and (f) 800 nm excitation.

2. Results

The differential transient XUV absorption on the Si $L_{2,3}$ edge near 100 eV is shown in Figure 1a through Figure 1c for excitation wavelengths of 266 nm, 500 nm, and 800 nm. These excitation wavelengths correspond to transitions into the Γ , L, and X/ Δ valleys of silicon, respectively. In the differential absorption plot, red corresponds to decreased absorption and blue corresponds to increased absorption following optical excitation. The onset of the silicon $L_{2,3}$ edge at 99.5 eV is shown as a dashed line [1]. The static absorption is shown in Figure 1d.

After excitation, distinct changes in the differential absorption are visible on the time scales of carrier-phonon (<1 ps) and phonon-phonon scattering (>1 ps) for each excitation wavelength. The changes in the differential XUV absorption with optical excitation results from three sources. First, excitation of carriers from the valence to conduction band changes the electron loss function, broadening the excited state spectrum [2]. Second, state-filling effects from the photoexcited carriers lead to an increased absorption for holes in the valence band and a decreased absorption for electrons in the conduction band [3]. Finally, various renormalizations of the electronic structure occur including carrier-carrier interactions, carrier-phonon scattering, and thermal expansion of the lattice due to the decay of hot phonons into lower energy acoustic phonons [3,4]. In each case, the valence and conduction bands shift in response to the optically-induced perturbation, shifting the XUV absorption spectrum and resulting in changes to the differential absorption.

The critical points of the Si band structure can be used to model the static and differential absorption [1]. The critical point density-of-states for the dipole-allowed transitions from the 2p core level to the Δ_1 , L_1 , and L_3 valleys is shown in Figure 1d. The energies of the critical points are shifted to match the core-hole renormalization of the $3s/3p$ hybridized Δ_1 and L_1 points. The background component is modeled by inspection of the remaining critical points in the band structure. Broadening is included to the critical point density of states using a single-plasmon pole model with the plasmon frequency at 16.8 eV, corresponding to the valence carrier density of Si [2]. The 100 meV instrumental resolution is further included by a Gaussian broadening. After excitation, electrons are promoted to the CB, modifying the valence plasmon frequency and blocking relaxation pathways of the photoexcited electrons. These state-filling effects adjust the loss function as well as the conduction and valence band absorption, increasing the broadening of the XUV peaks and blocking or allowing new XUV transitions. After the initial excitation, hot carriers thermalize by scattering off of acoustic and optical phonons in intravalley and intervalley processes [3,4]. The dominant phonon-scattering mode is determined by the valley that the excited carriers reside in, with the emission of a phonon by an excited carrier leading to a corresponding expansion of the lattice. This non-equilibrium strain acts to distort the lattice, changing the energy of and splitting of the otherwise degenerate critical points. This can be modeled using mode Gruneisen parameters and the known relationships between strain and band structure in silicon [5]. Carrier-based renormalization of the band gap occurs on a time-scale shorter than the pulse width, and it was not found necessary to describe the longer time data here. On time scales greater than a picosecond, the hot optical and acoustic phonons thermalize by scattering into low energy, low momentum acoustic phonons. These phonons act to heat the lattice, with the band gap decreasing due to thermal expansion [4].

The extent of broadening, state-blocking, and renormalization contributions to the differential absorption can be related to the temperature of the carrier, optical phonon, and acoustic phonon baths following excitation. A three-temperature model using the reported carrier-phonon and phonon-phonon scattering times for silicon can therefore capture the time-dependent change in the transient XUV spectrum, as demonstrated for 500 nm excitation in Figure 1e. Good agreement is found around the modeled critical points, with the discrepancies increasing at higher and lower energy. Similar agreement is seen for 800 nm excitation, Figure 1f, with the model taking into account the different state-blocking energy and carrier-phonon scattering involved in the Δ versus L valley. The discrepancies that do exist occur because the critical point model and strain-induced band structure changes are only valid in the 100 to 105 eV range and do not capture the change in density of states with strain. The model is improved upon by using a Bethe Salpeter Equation (BSE) calculation (Quantum Espresso / OCEAN) to directly predict the core-hole modified joint density of states and its modification with a non-equilibrium strain and thermal expansion. The three-temperature model can also be replaced by an ensemble Monte-Carlo method to fully describe the k-space dependent carrier populations and phonon scattering. These results allow the photoexcited carrier relaxation and resulting lattice dynamics to be quantified in the major valleys of silicon using one experimental technique, demonstrating the potential of transient XUV spectroscopy for hot carrier dynamics.

3. References

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