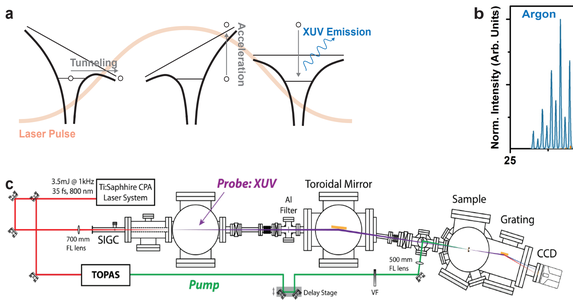
PHOTOEXCITED CARRIERS, PHONONS, and their SCATTERING

Measured by Transient Extreme Ultraviolet Spectroscopy

Scott K. Cushing[1,3], Brett M. Marsh[1], Mihai E. Vaida[1], Stephen R. Leone[1,2,3]

[1]Department of Chemistry, [2]Department of Physics, University of California Berkeley; [3]Chemical Sciences Division, Lawrence Berkeley National Laboratory

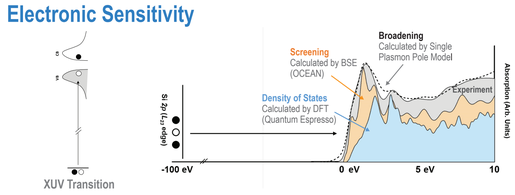
Transient XUV Spectroscopy



(a) High harmonic generation occurs by ionization and acceleration of electrons in the electric field of a laser pulse. (b) The gas determines the harmonic range. (c) Transient extreme ultraviolet (XUV) setup.

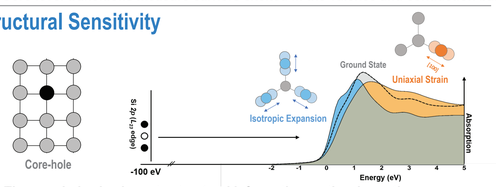
Why X-Rays?

Electronic Sensitivity



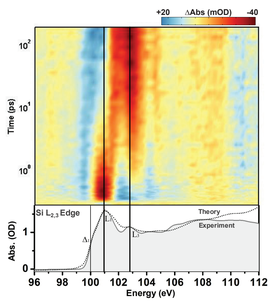
Core-level transitions measure carriers excited in the valence and conduction band

Structural Sensitivity



The core-hole also imparts structural information on the absorption

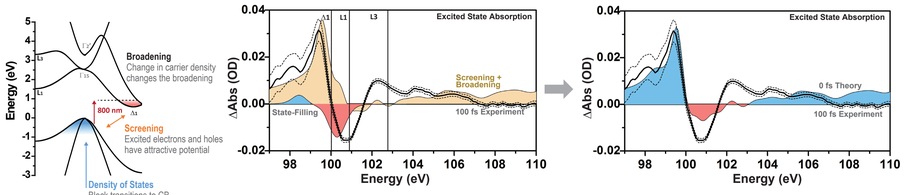
Excited State Absorption



The transient absorption following 800 nm excitation contains a mixture of carrier and phonon contributions

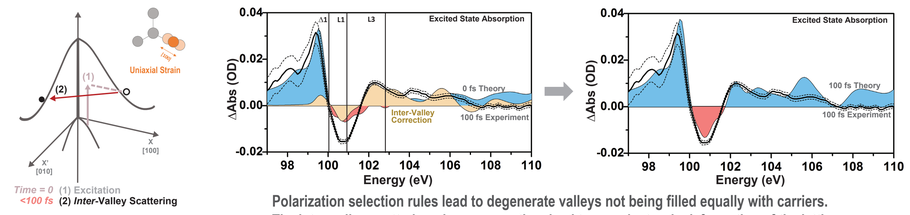
Separating Carriers and Phonons In Transient XUV

Time = 0 fs



The photoexcited change in valence and conduction carrier density modifies the screening, broadening, and possible XUV transitions in the excited state absorption

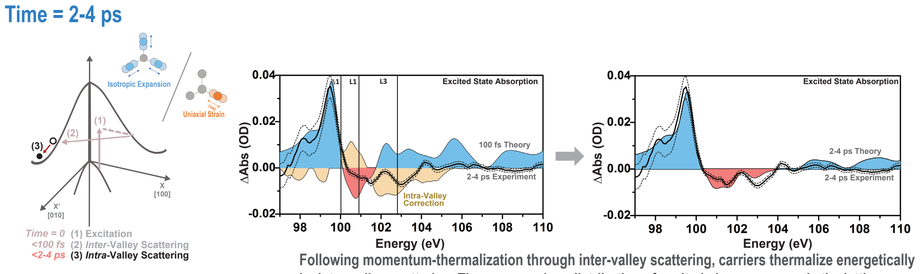
Time = 100 fs



Time = 0 (1) Excitation
<100 fs (2) *Inter*-Valley Scattering

Polarization selection rules lead to degenerate valleys not being filled equally with carriers. The inter-valley scattering phonons can then lead to an anisotropic deformation of the lattice.

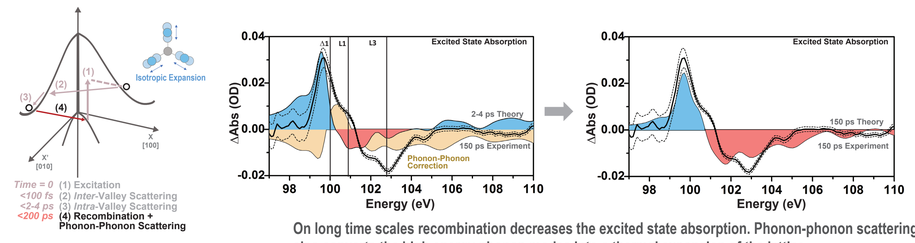
Time = 2-4 ps



Time = 0 (1) Excitation
<100 fs (2) *Inter*-Valley Scattering
<2-4 ps (3) *Intra*-Valley Scattering

Following momentum-thermalization through inter-valley scattering, carriers thermalize energetically by intra-valley scattering. The more random distribution of excited phonons expands the lattice.

Time = 100-200 ps

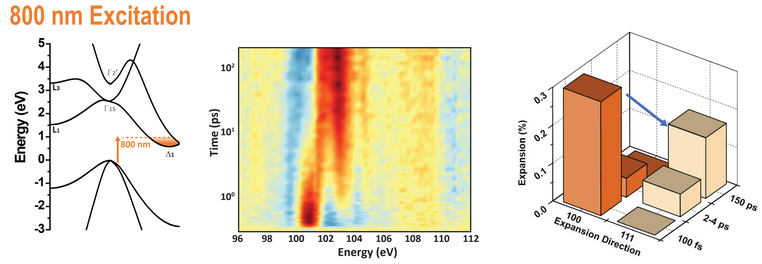


Time = 0 (1) Excitation
<100 fs (2) *Inter*-Valley Scattering
<2-4 ps (3) *Intra*-Valley Scattering
<200 ps (4) Recombination + Phonon-Phonon Scattering

On long time scales recombination decreases the excited state absorption. Phonon-phonon scattering also converts the high energy phonon modes into a thermal expansion of the lattice.

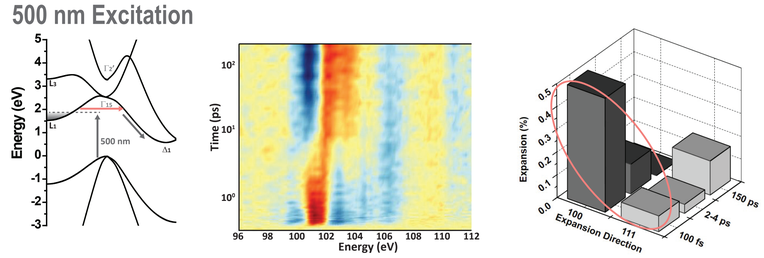
Valley-Specific Electron-Phonon Scattering Pathways in Si(100)

800 nm Excitation



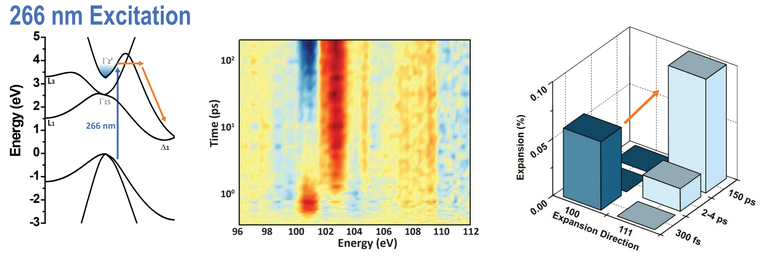
Excitation at 800 nm is followed by inter-valley Δ–Δ' scattering and thermalization, tracked by the measured phonons.

500 nm Excitation



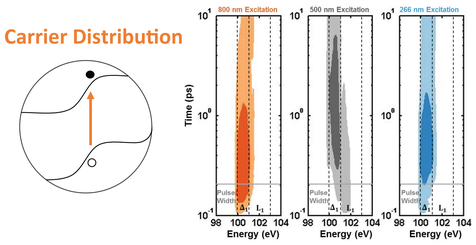
Excitation at 500 nm is followed by inter-valley L-L' and L-X scattering, reflected in the initial phonon distribution.

266 nm Excitation



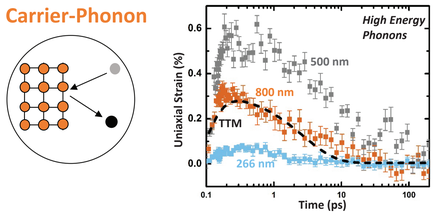
Excitation at 266 nm is followed by inter-valley Γ-X scattering followed by a long thermalization period due to excess pump energy, as seen by comparing the amplitude of the initial to final phonon modes to 800 nm excitation

Carrier Distribution



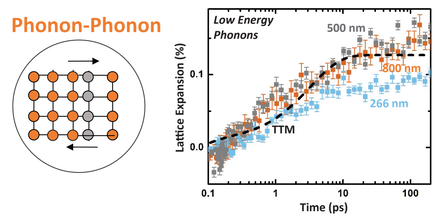
The carrier distribution relaxes from the L to Δ valley after 500 nm excitation. For 266 nm excitation, only slow filling of Δ is measured as Γ is dipole forbidden.

Carrier-Phonon



The phonon population from inter-valley scattering has a distinct rise time at each excitation wavelength due to the possible scattering pathways.

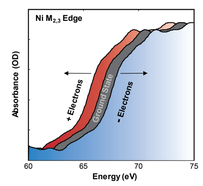
Phonon-Phonon



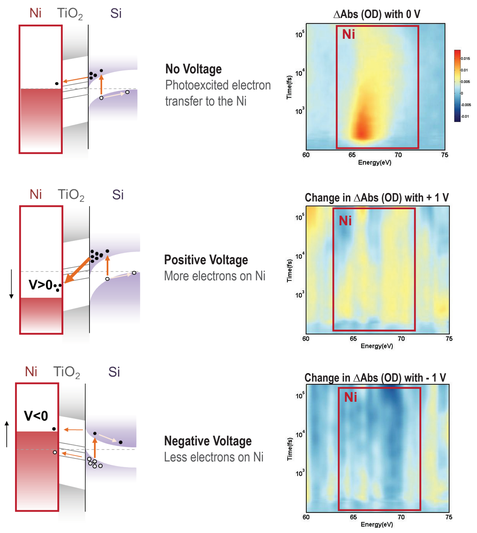
The phonon population from intra-valley and phonon-phonon scattering is less dependent on excitation wavelength since the process is more isotropic

Charge and Heat Transfer in a Metal-Insulator-Semiconductor Junction

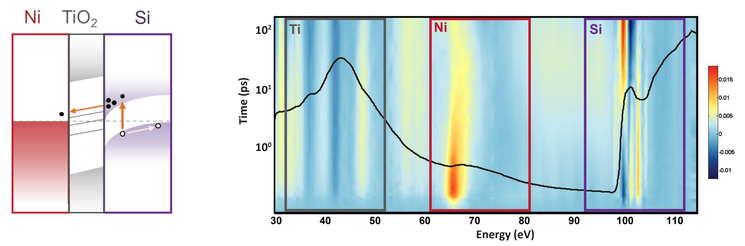
Tracking Charge on Transition Metals



The charge accumulated can be tracked by the energy of the metal edge

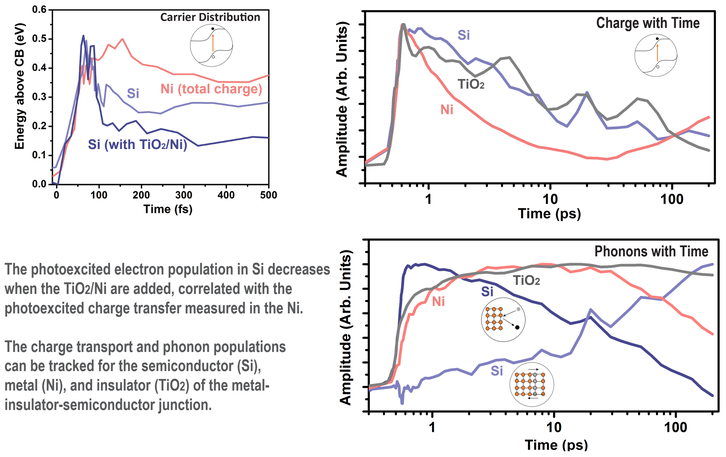


Photoexcited Charge Transfer in Si/TiO2/Ni



Photoexcitation results in charge transfer from the Si, through the insulating TiO2 layer, and into the Ni metal.

Carriers and Phonons in each Component



The photoexcited electron population in Si decreases when the TiO2/Ni are added, correlated with the photoexcited charge transfer measured in the Ni.

The charge transport and phonon populations can be tracked for the semiconductor (Si), metal (Ni), and insulator (TiO_2) of the metal-insulator-semiconductor junction.

Other Materials:

1. **Ultrafast Charge Trapping by Polarons in Fe_2O_3**
S.K. Cushing, et. al Nature Materials, Just Accepted 2017

2. **Electrons and holes in Ge and Si:Ge**
M. Zurch Nature Communications 8 (2017), 15734;
M. Zurch Structural Dynamics, 4 (2017), 044029.

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