Femtosecond time-resolved dual-angle reflectometry to observe laser-induced dynamics in solids

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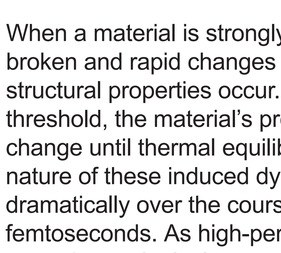
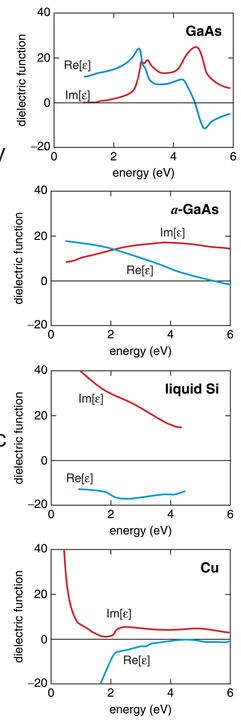
Introduction

When a material is strongly excited, bonds are broken and rapid changes in its electronic and structural properties occur. Below the damage threshold, the material's properties continue to change until thermal equilibrium is reached. The nature of these induced dynamics change dramatically over the course of the first few hundred femtoseconds. As high-performance electronic and opto-electronic devices approach the region where very large, rapidly varying current densities are present due to the miniaturization of features, these effects must be considered for device design. In such devices as high-power laser diodes, excitation can be strong enough to induce semiconductor-to-metal phase changes. Because knowledge of the exact phase of the material at all times is required for accurate device modeling, strong-excitation dynamics must be well understood.



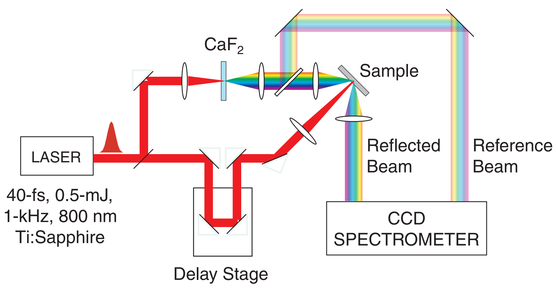
Using femtosecond laser pulses, excitations much greater than those found in any modern device can be easily produced. However, we must somehow probe how the material's properties evolve after pumping.

Because the dielectric function is determined by the band structure and electron occupation, it is a characteristic property of a material and a probe of both the electronic configuration and lattice structure. Therefore, if we measure how the dielectric function changes over time when a sample is excited, we can accurately determine the induced dynamics.

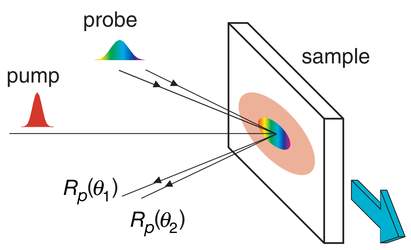


Dual-angle reflectometry technique

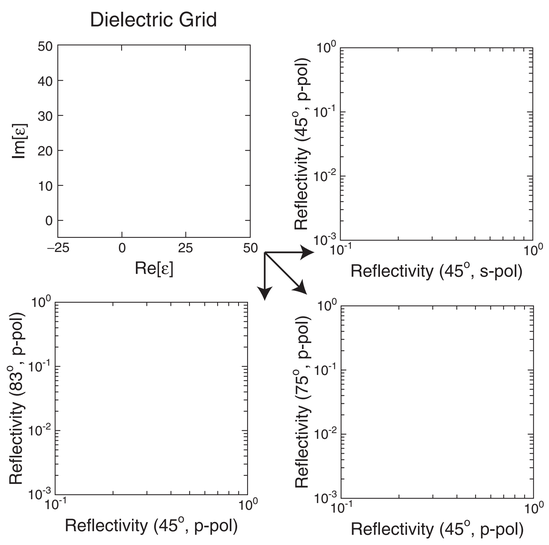
To measure the time-resolved dielectric function, we need a technique that would be far simpler than ellipsometry so that we can repeat the measurement at many different times. We can then vary the time delay between the pump and probe pulses to observe the evolution of the excitation induced by the pump pulse.



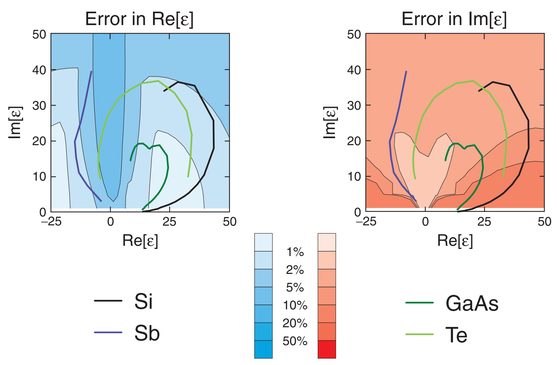
To obtain the dielectric function from this setup, we take reflectivity data at two different angles of incidence. We then use this data, combined with knowledge of sample geometry (e.g. probe angle of incidence relative to crystal axes) to invert the Fresnel formulae to obtain the real and imaginary parts of the dielectric function.



To ensure spatial homogeneity of the probed region, the pump spot size is about four times larger than the probe spot. For pump fluences above the damage threshold, the sample is translated between shots.



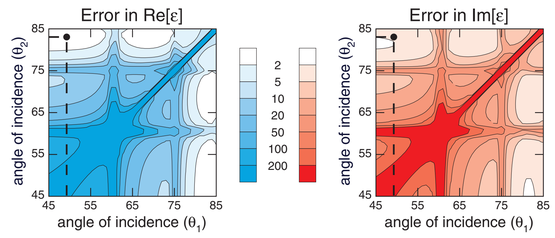
The two angles of incidence should not be chosen randomly. For angles of incidence that are too similar, the reflectivity data does not uniquely determine the dielectric function. This can be seen where the dielectric grid collapses to a line when projected into reflectivity space at close angles.



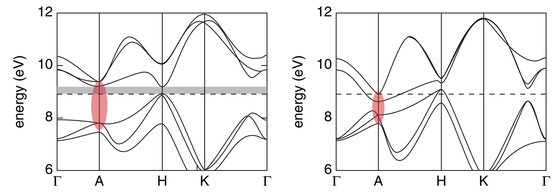
The plots show the error in Re[ε] and Im[ε] given a 1% accuracy in the measurement of *p*-polarized reflectivity at 45° and 83° angles of incidence. Solid lines indicate the dielectric properties of various solids from the near-IR to the near-UV (1.5 to 3.5 eV).

Results in tellurium

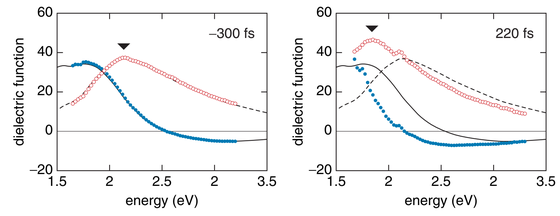
Tellurium forms a uniaxial crystal with a relatively small band gap of 0.33-eV. In this experiment we observe coherent phonons induced by intense femtosecond laser pulses in tellurium. We cannot excite the tellurium enough to observe a semicondutor-to-metal phase change without causing permanent damage.



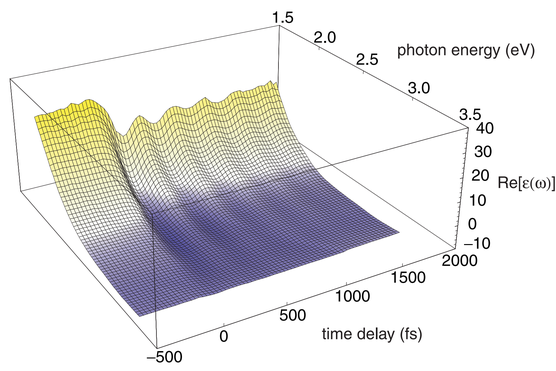
The uncertainty in the dielectric function of tellurium is minimized for p polarized light when the chosen angles of incidence are 49.7° and 83.5°.

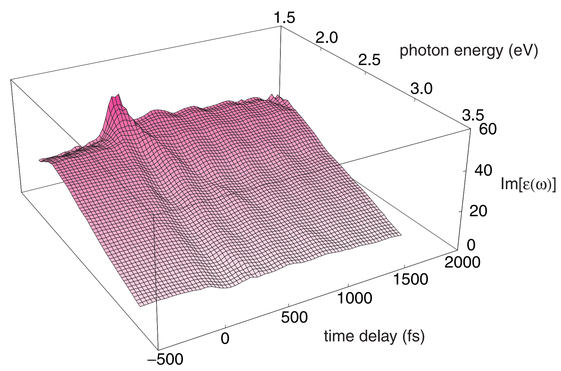


Tangney and Fahy, Phys. Rev. B **65**, 054302 (2002)



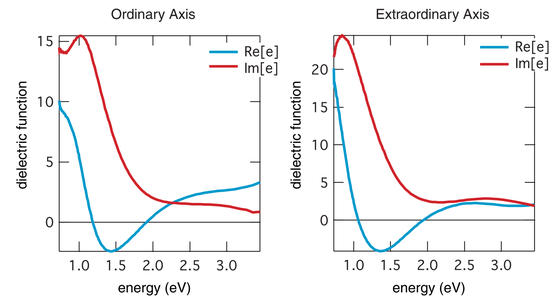
The red shift of the dielectric function of tellurium under photoexcitation corresponds to a narrowing of the semiconductor bandgap. Under a sufficiently intense pulse, a band collapse would cause a semiconductor-to-metal transition.

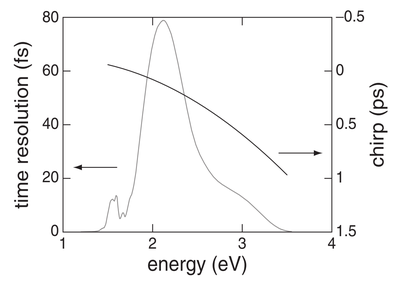




Moving to Ti_2O_3 and improving white-light generation

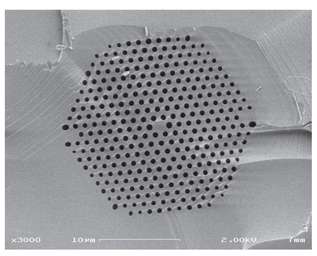
We are currently performing femtosecond dual-angle reflectometry on Ti_2O_3 in the hope of observing a semiconductor-to-metal phase change. We believe that we will be able to observe this transition because Ti_2O_3 has a smaller band gap and a higher damage threshold than tellurium. However, since the band gap is much smaller, the peak of the dielectric function appears much further into the near-infrared.





Material dispersion from the white-light continuum generation for the probe beam broadens the pulse from 40-fs to the picosecond scale. The increased duration of the white-light pulse limits the time resolution, because it extends the period of time that any given spectral component is incident on the sample. The relationship between the spectrum, chirp and the calculated time resolution for the CaF_2 is pictured at the left.

To observe the spectral region of interest for Ti_2O_3, we use a highly-nonlinear photonic crystal fiber to generate a supercontinuum extending from 500- to 1600-nm. A scanning electron microscope image of the end-face of this fiber is shown at the right. Because the probe pulse must travel through over a meter of fiber to generate the supercontinuum, the pulse is broadened considerably in time. To achieve better time resolution, we are planning on compressing our probe pulse. Since the photonic crystal fiber generates the continuum with a highly nonlinear chirp that is symmetric around the fiber zero dispersion wavelength, a more advanced compression scheme than a traditional single grating must be devised.



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