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Intervalley Scattering Rates in Tellurium Observed via Time-Resolved Terahertz Spectroscopy

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Abstract

We conducted time-resolved terahertz spectroscopy measurements on the elemental semiconductor tellurium. Pump-probe measurements were used to find the conductivity as a function of time in single crystalline tellurium samples. It was found that the excitation dynamics in tellurium changes for photon energies of 1.03 eV and 1.55 eV. The change in these excitation dynamics was attributed to intervalley scattering effects. A model using intervalley scattering and Auger recombination was fit to the data, giving a value of 2.28 ps for the intervalley scattering time constant in tellurium.

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Cover Page Footnote

The author would like to thank Professor James Heyman for directing him throughout this research project. The author would also like to thank his lab parent Elliot Weiss for helping setup and take measurements and the Massari Research Group at the University of Minnesota for allowing access to the MULE system.

I Introduction

Hot carriers, created by optical excitations or thermal fluctuations in semiconductors, undergo a number of energy loss mechanisms, such as scattering. Measurements of hot carrier dynamics in semiconductors are of importance in understanding the fundamental energy loss and scattering effects in materials. These measurements also give further insights into carrier-carrier interactions, a topic of great interest in semiconductor physics. In order to better understand these scattering interactions, time-resolved terahertz (THz) spectroscopy is used to explore the conductivity of a material. By conducting these measurements on relatively unexplored semiconductors, such as tellurium, we can more rigorously test models for various carrier-carrier interactions.

Time-resolved THz spectroscopy is a non-contact electrical probe that measures the photoconductivity of a semiconductor. The main advantage of these types of measurements is that they allow for measurements of the conductivity as a function of time in what is called a pump-probe measurement. This technique can also be used to measure the carrier dynamics in indirect band gap semiconductors and the frequency-dependent conductivity of a material, which is called a THz scan.

In this experiment, we used pump-probe measurements on a polycrystalline sample and a single crystalline sample of tellurium in order to explore intervalley scattering effects. We observed a difference in the excitation dynamics between photon energies of 1.03 eV and 1.55 eV. This difference is explained by intervalley scattering effects from a secondary minimum to the absolute minimum in the conduction band,

and allows us to extract the time constant for the intervalley scattering process in tellurium.

II Methodology

We used a FemtoLaser XL-500 Ti Sapphire Laser to generate 500 mW of pulsed light at wavelengths of 800 nm. The pulse width is approximately 50 fs with an energy per pulse of 500 nJ. The beam is split into three through the use of beam splitters: the pump beam, the THz beam, and the probe beam. The pump is modulated by a chopper, which temporarily blocks and unblocks the beam, spinning with a frequency of 500 Hz. The modulated beam is then incident on the sample, where it then excites electrons from the valence band to the conduction band. The THz beam is used to produce picosecond pulses of THz radiation at a biased photoconductive antenna made of ZnTe crystal using optical rectification. The THz radiation then reflects from the sample and is detected using a second ZnTe crystal and the polarization of the probe beam, which passes through the THz radiation and the ZnTe crystal, in a process called the electro-optic effect. More information on the electro-optic effect has been previously published [1]. A diagram showing the experimental setup is shown in Figure 1.

Two delay stages can change the path lengths of the pump and THz beams. For a THz scan, the pump beam delay stage is held fixed while the THz beam stage is scanned over a range, allowing for measurements of the electric field from the THz radiation at different times with respect to the probe beam. For a pump-probe scan, the THz delay stage is held fixed while the pump beam stage is scanned over a range,

allowing for measurements of the photoconductivity of a material at different times with respect to the THz radiation. More details on time-resolved THz spectroscopy can be found in previous publications [2].

Measurements were also taken using the Multi-User Laser Experiment (MULE) system in the Massari Lab at the University of Minnesota. This laser system operates in a similar way to the XL-500 laser system, but is setup for transmission measurements and has a tunable pump beam allowing for the energy of the photons incident on the sample to be varied.

The measurements presented here were conducted on a single crystalline sample and a polycrystalline tellurium sample. The single crystalline tellurium sample was produced by the Princeton Scientific Corporation and has a purity of 99.9999% with a thickness of 1.00 mm and one side polished with a roughness of less than 0.01 μm .

III Results and Discussion

Transient photoconductivity transmission measurements were taken on a polycrystalline tellurium sample using the MULE laser system at photon wavelengths of 1.03 eV and 1.55 eV. The results from these measurements are shown in Figure 2. The excitation behavior between these two measurements exhibit different time constants, as evident by the 1.03 eV measurement reaching its maximum conductivity after about 1 ps whereas the 1.55 eV measurement took about 4 ps to reach its maximum conductivity.

The difference in the excitation behavior observed in Figure 2 can be explained by a change in conductivity from intervalley scattering effects. The conductivity of

the material depends on the number of electrons in the conduction band, or

$$\sigma = ne^2\tau/m^*, \quad (1)$$

where σ is the conductivity, n is the number of electrons, τ is the average time between scattering events, and m^* is the effective mass of the electrons. The effective mass is what mass an electron appears to have to a force from the outside of the material's lattice. The effective mass depends on the second derivative of the electron energy, given by

$$\frac{1}{m^*} = \frac{1}{\hbar^2} \frac{\partial^2 E}{\partial q^2}, \quad (2)$$

where E is the energy and q is the crystal momentum. An interpretation of Equation 2 is that the effective mass of the electron depends on the curvature of the energy band at a certain momentum. This means that the conductivity of a semiconductor is dependent on the location in the conduction band at which the electrons are excited to and can lead to differences in the conductivity behavior in a material based on the excitation energies.

In intervalley scattering, the electrons in a semiconductor are first excited to a secondary minimum in the conduction band. After some time, the electrons undergo scattering and fall down to the absolute minimum of the conduction band, giving their extra energy and momentum to the lattice as a phonon. If the absolute minimum of the conduction band has a higher curvature than the secondary minimum, then the transition increases the conductivity of the material as the electrons move from the secondary minimum to the absolute minimum. Thus, it would take a longer

time for the material to reach its maximum conductivity when the electrons are excited to the secondary minimum than when the electrons are excited directly to the absolute minimum.

We can see that this is the case for the transitions observed in tellurium, as shown in Figure 3, which is modified from [3]. Superimposed on the image, we have shown the approximate transitions corresponding to excitations with 1.03 eV and 1.55 eV photons. As we can see, the transitions corresponding to the 1.03 eV measurements are transitions to the absolute minimum of the conduction band where the curvature of the energy band is high. The 1.55 eV transition corresponds to a transition to a secondary minimum in the conduction band where the curvature, and therefore the conductivity, is lower. Thus, when the electrons undergo intervalley scattering from the secondary minimum to the absolute minimum, the conductivity increases, which explains the behavior we see in Figure 2 well.

We then took transient photoconductivity measurements on a single crystalline tellurium sample at photon energies of 1.55 eV to delays of 1000 ps using the XL-500 laser system. This was done to examine the slower excitation behavior, where intervalley scattering effects were prevalent, in more detail. Since this single crystalline sample was thick, we performed reflectance measurements instead of transmission measurements. The drift in the laser system's power was minimized by taking three separate rounds of measurements: one to a 10 ps delay with a step size of 0.2 ps, one to a 200 ps delay with a step size of 1 ps, and one to a 1000 ps delay with a step size of 5 ps. The results from these measurements are shown in Figures 4 and 5.

Figures 4 and 5 also show the model that we fit to our measurements. To construct

the model, we included terms for the intervalley scattering process and the recombination process. The intervalley scattering model we used is simply an exponential decay. For the recombination effects, we decided to use an Auger recombination model.

Auger recombination is a model for recombination dynamics that involves three particles: two electrons and one hole. In this model, an electron in the conduction band collides with another electron in the conduction band. One of the electrons then gives its extra energy and momentum to the other electron and falls back down to the valence band to recombine with a hole. The Auger recombination term is therefore given by

$$\frac{dn}{dt} = c_1 n^2 p = c_1 n^3, \quad (3)$$

where n is the number of electrons in the conduction band, p is the number of holes in the valence band, and c_1 is a constant. In this model, the number of holes is equal to the number of electrons, so $n = p$. Then, we can solve Equation 3 using separation of variables. Doing so gives

$$n = \frac{c_2}{\sqrt{1 + \frac{t}{\tau_2}}}, \quad (4)$$

where c_2 is a constant and τ_2 is a time constant. More details on Auger recombination dynamics have previously been published [4].

Our model is then simply given by the product of the intervalley scattering and

the Auger recombination terms, or

$$s = a (1 - e^{-t/\tau_1}) \left(\frac{1}{\sqrt{1 + \frac{t}{\tau_2}}} \right). \quad (5)$$

In this model, s is the measured signal, a is an overall scaling factor, τ_1 is the time constant for the intervalley scattering process, and τ_2 is the time constant for the Auger recombination effects. In order to fit the model to the data, we performed a least squares fit. This fit gave constants of $a = 0.026$, $\tau_1 = 2.28$ ps, and $\tau_2 = 27.86$ ps. For this discussion, the most relevant constant is τ_1 , which gives a time constant of 2.28 ps for the intervalley scattering process in tellurium. This compares favorably to intervalley scattering time constants measured in other materials: GaAs has a time constant of about 2 ps [5] and $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ has a time constant of about 3.1 ps [6].

Our model shows a slight divergence from the data at long delays, where the model rises slightly above the data, and at short delays, where the model is slightly below the data, as evident in Figures 4 and 5. The divergence at short delays is due to the inherit time constant in our electronics. We accounted for this time constant by removing the data where this effect was evident, but this resulted in the data being slightly offset from zero at initial onset. However, our model asserts that the signal is 0 at initial onset, causing the model to be below the data initially. At large delays, the model diverges from the data because the dominant recombination effect is no longer Auger recombination since there are many fewer electrons in the conduction band. At these large delays, it is likely that some other recombination effect becomes

dominant, such as trap-assisted recombination, but this effect is unlikely to affect the model at short delays where the intervalley scattering behavior is evident.

IV Conclusions

In this experiment, we used time-resolved THz spectroscopy to probe the carrier dynamics in the elemental semiconductor tellurium. It was found that tellurium exhibits intervalley scattering for photon energies at 1.55 eV. We fit a model to a measurement on single crystalline tellurium using a simple intervalley scattering model and an Auger recombination model. These models fit the data well, and an intervalley time constant of 2.28 ps was found for the scattering process in tellurium. This compares favorably to other intervalley scattering time constants measured in GaAs and $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$. However, other measurements of the intervalley scattering time constant in tellurium have not been taken, so additional measurements of excitation dynamics in tellurium could help confirm this result.

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Figures

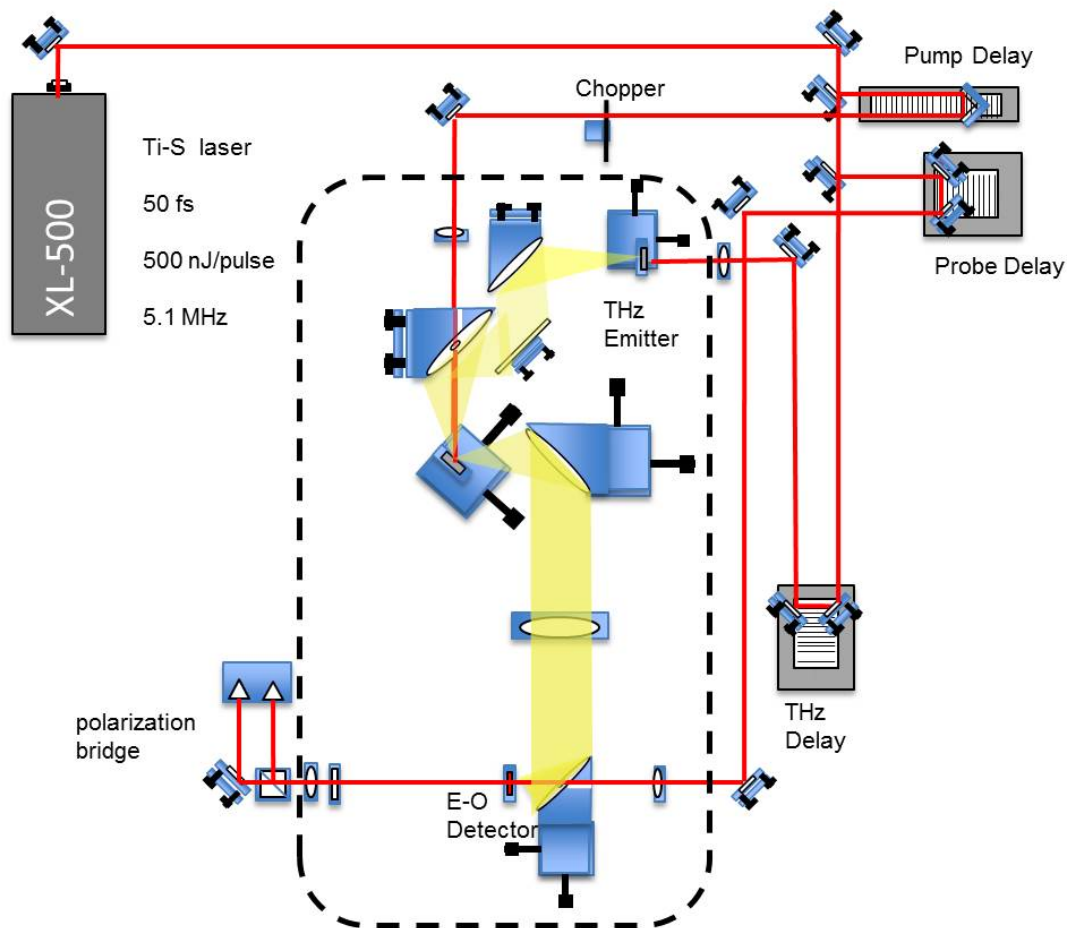


Figure 1: This diagram shows the experimental setup for the photoconductivity reflectance measurements taken with the XL-500 laser system on the single crystalline tellurium sample. The laser beam leaves the Ti:S laser and reaches two different beam splitters which split the beam into three separate beams. The pump beam excites the electrons in the sample, the THz beam creates THz radiation incident on the sample, and the probe beam measures the polarization of the THz radiation after it reflects from the sample. The pump delay is a stage with a pair of mirrors mounted on it that moves back and forth, allowing us to change the time that the pump beam is incident on the sample with respect to the THz radiation. This allows us to measure the conductivity of the sample at different points during its excitation.

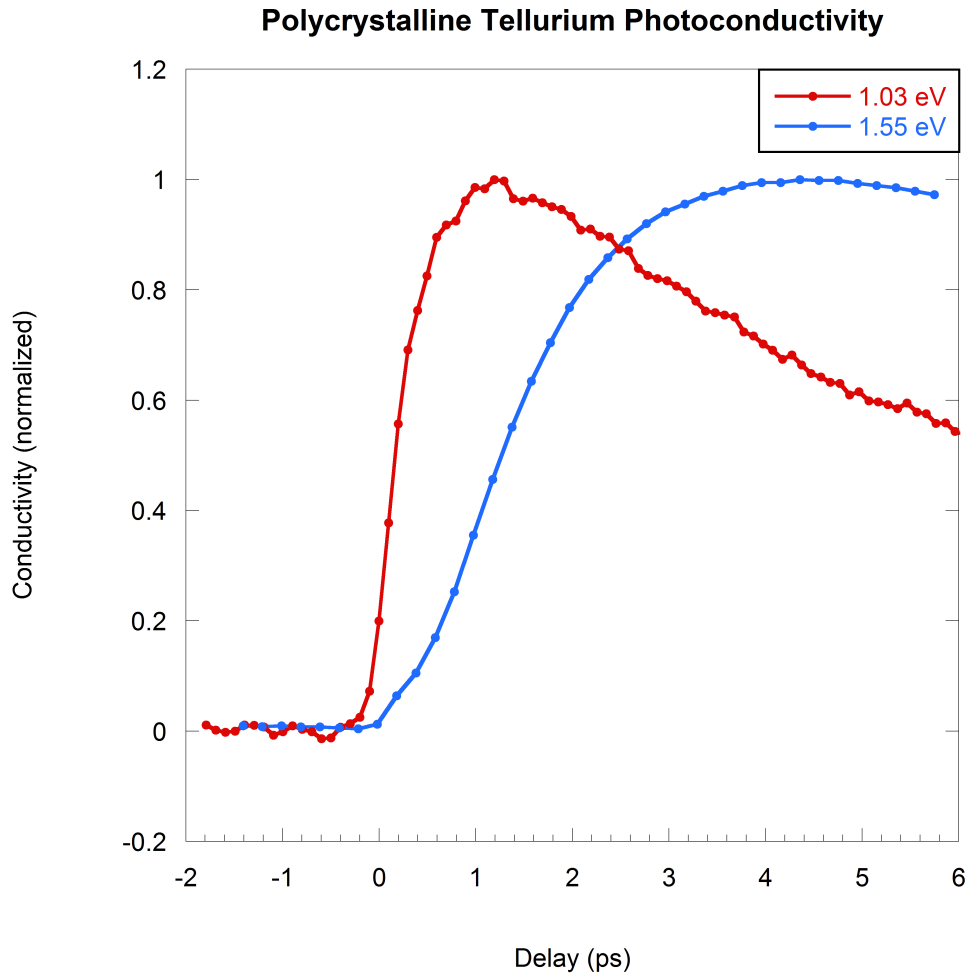


Figure 2: Transient photoconductivity measurements on a polycrystalline tellurium sample taken at photon energies of 1.55 eV and 1.03 eV. The y-axis is the normalized conductivity of the samples in order to better compare the results between the two measurements and the x-axis is the delay in picoseconds between the pump beam and the THz beam.

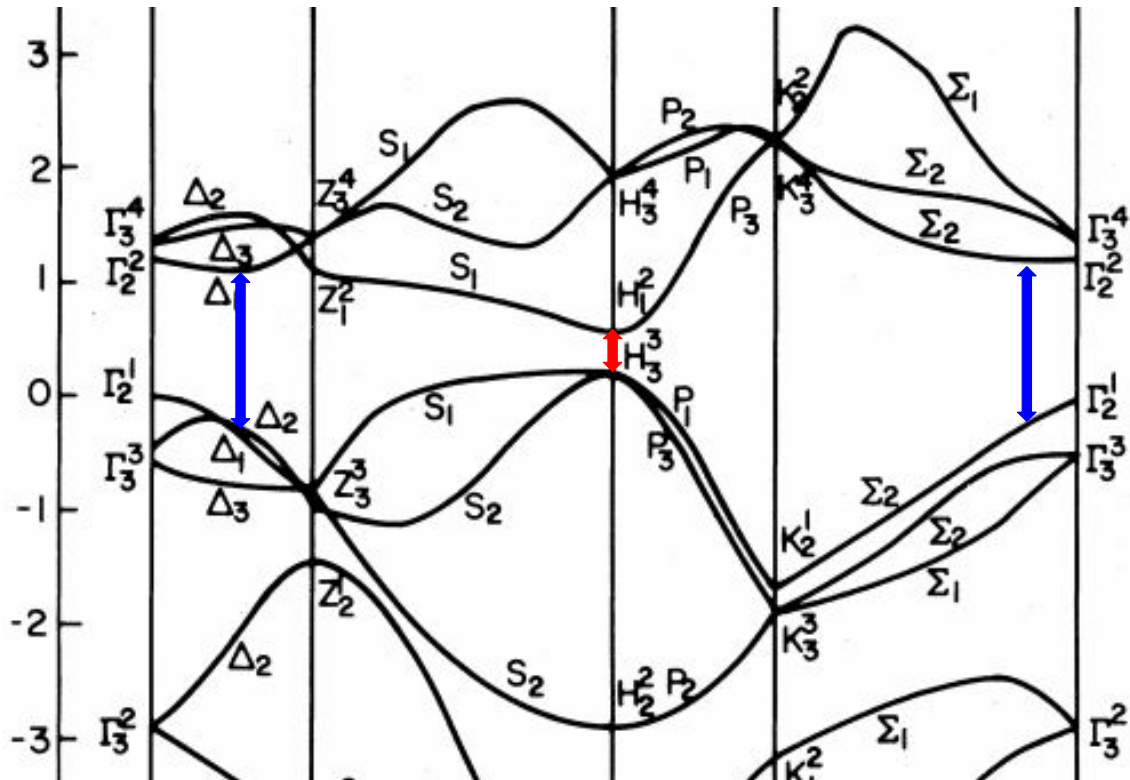


Figure 3: This is the energy bands of tellurium, modified from [3]. The y-axis is the electron energy and the x-axis is the crystal momentum. The energy bands on the top of the diagram represent the conduction band and the bands on the bottom of the diagram represent the valence band. The dips in the conduction band are local minima, where the lowest peak is the absolute minimum. Superimposed on the image, we have shown the transitions corresponding to excitations with 1.03 eV (red arrow) photons to the absolute minimum and 1.55 eV (blue arrow) photons to a local minimum.

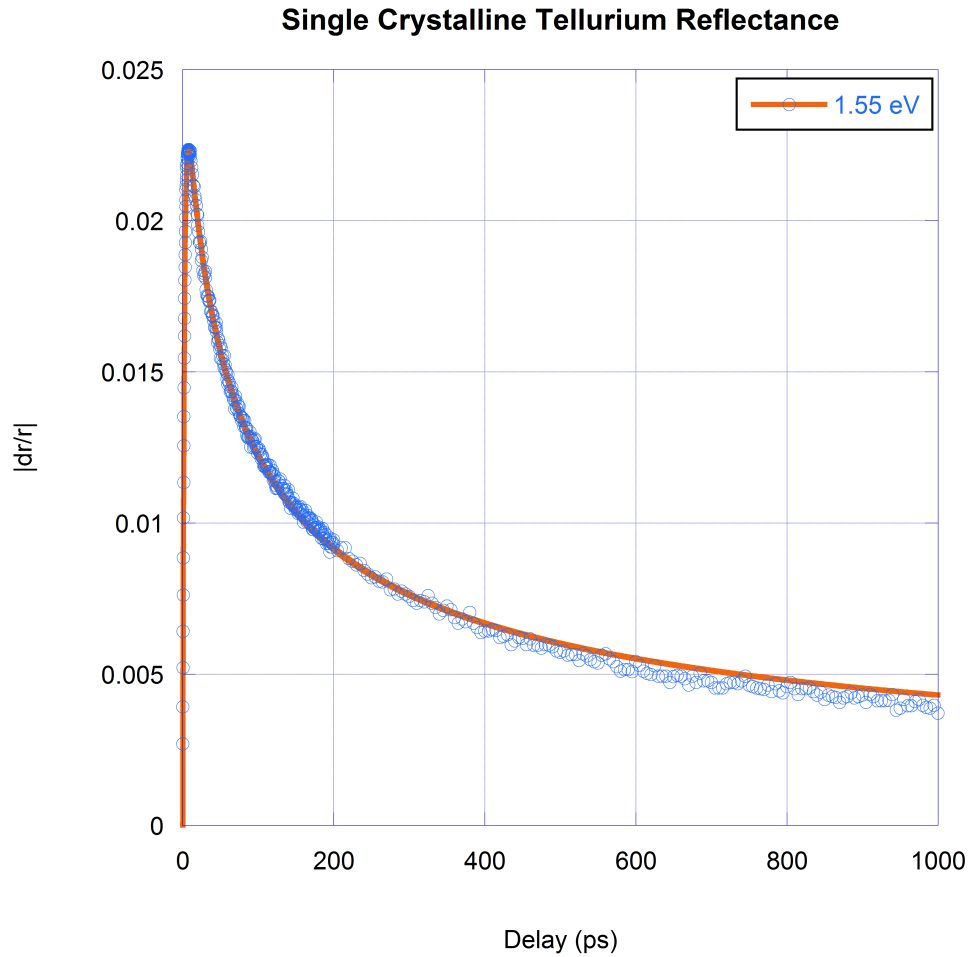


Figure 4: A transient photoconductivity measurement of our single crystalline tellurium sample taken with photon energies of 1.55 eV to pump delays of 1000 ps. The y-axis is the absolute value of the change in the reflectance divided by the reflectance, which is proportional to the conductivity. A model was fit to the data using intervalley scattering and Auger recombination dynamics, which is shown in orange.

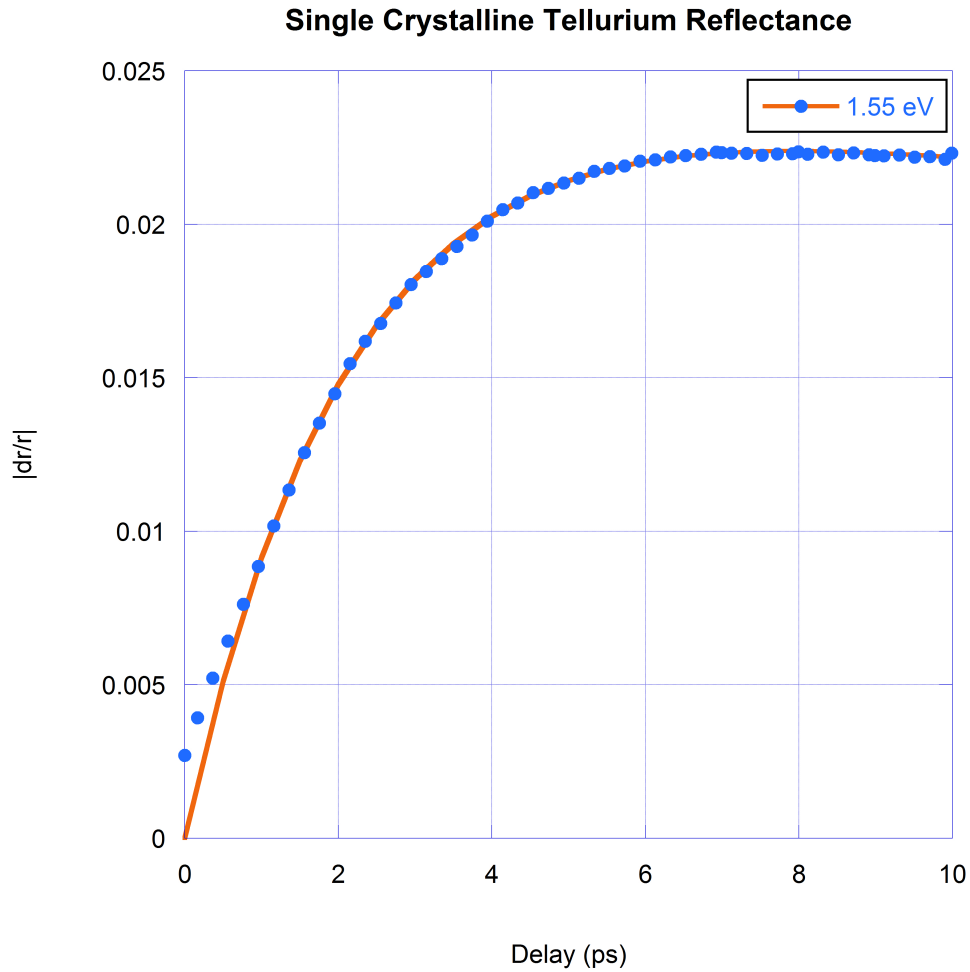


Figure 5: The results from the same transient photoconductivity measurement as in Figure 4, but only showing data out to delays of 10 ps in order to better display the excitation behavior in the single crystalline tellurium sample. The same model as in Figure 4 is also shown.