Femto-Second Laser Pump and Probe Technique and the Two-Temperature Model: Developing a Simulation of Electron-Phonon Heat Exchange

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Abstract

During this project the first task was to gain a basic understanding of the experimental set-up used for electron shadow-imaging and deflectometry, and the two-temperature model (TTM). Gradually, a C++ program which would be able to calculate heat transfer described by the model for the case of a thin gold film was developed. This was done in several steps including: the creation of a program based on a simplified version of TTM, refining to a second more realistic version, revising the program again to increase precision of the calculation. Currently, there is no experimental evidence regarding TTM for sample temperature near and above one eV, only inferential data and theoretical work.

Background: Laser Pump and Probe

A high-powered laser passes through a beam splitter. One line of the light, containing 90% of the laser beam's power, called the pump line is directed to an ultra-high vacuum (UHV) chamber where it is incident normally upon the front surface of a sample. The other beam, the remaining 10% of the laser's power, called the probe line is frequency tripled and converted to an electron pulse via a photocathode in the UHV (electron gun). This stream of electrons then passes parallel to the sample surface (perpendicular to the pump line). The pump line rapidly heats the sample, causing electrons to jump-off the lattice through the processes of multi-photon photoemission (MPE) and thermionic emission (TE). This intense heating temporarily initiates a plasma-like state at a solid density: warm dense matter (WDM). The probe line is used to create images of the charge cloud dynamics over time as the probe electrons interact with the plasma. These electron-shadow images are converted to light images by way of a phosphor screen. The images are intensified before being recorded by a CCD camera.

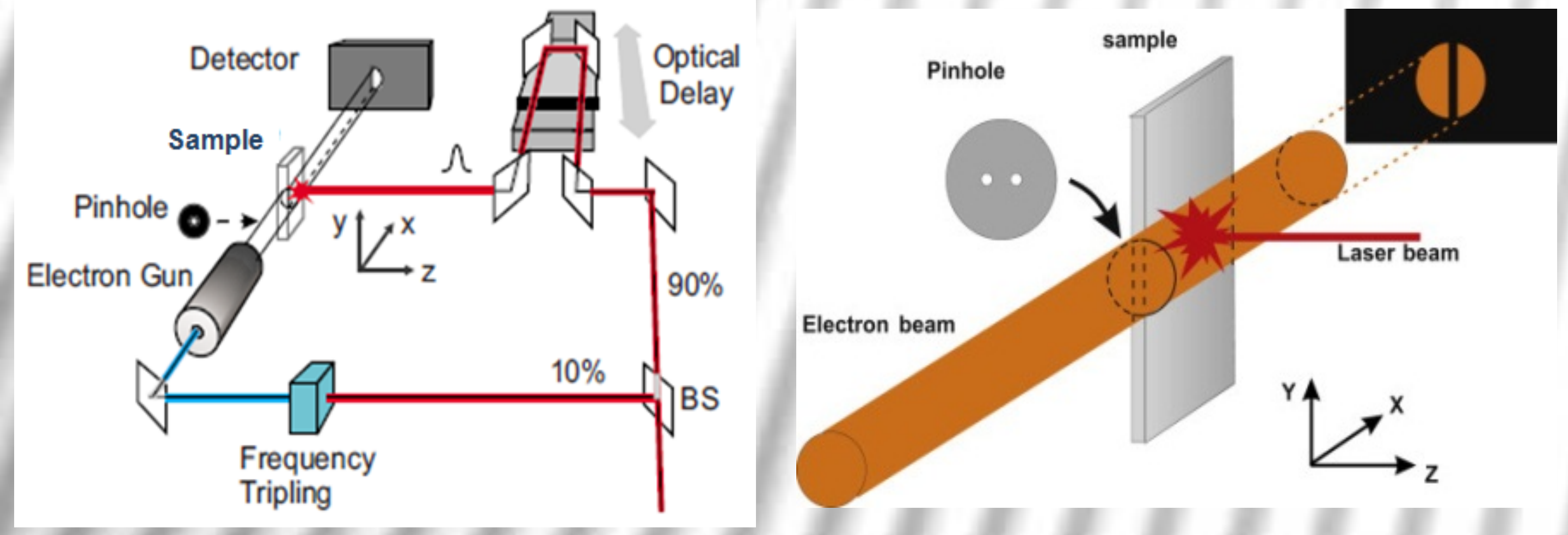


Figure 1. Basic experimental set-up, showing pump and probe lines incident on a sample as well as where the pump and probe pulses hit the sample (deflectometry set-up) (right)

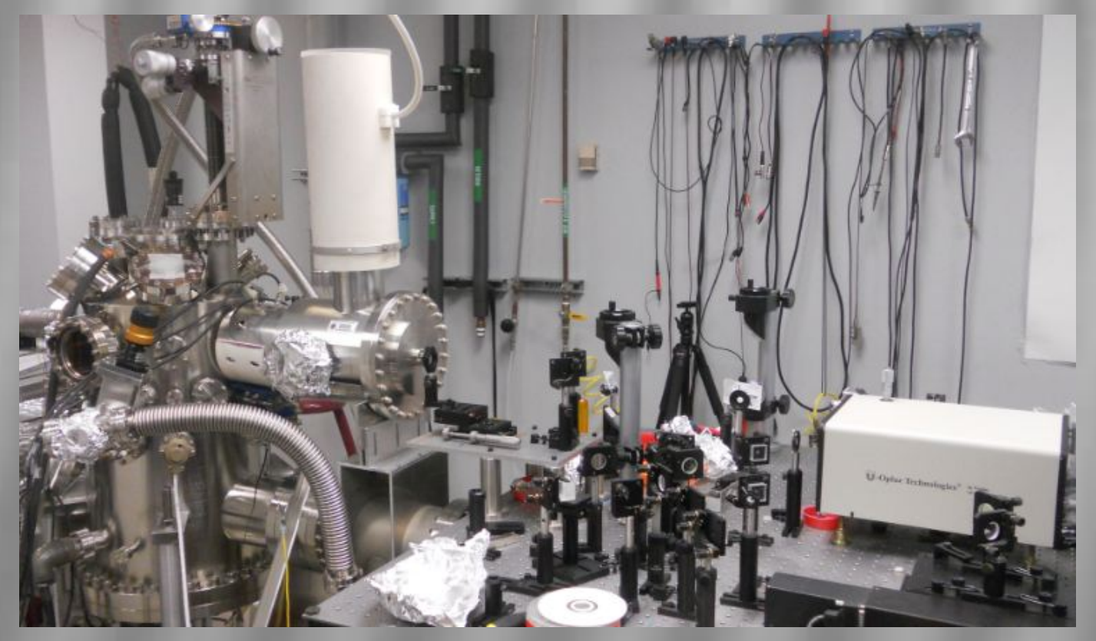
TTM describes the kinetics of heat transfer between electrons and lattice in solids initiated by an ultra-short laser pulse excitation. It is assumed that the electrons which absorb all the laser pulse energy will instantaneously come to an internal thermal equilibrium (at a temperature T_e) via electron-electron collisions. Then the energy of electrons is transferred to the lattice (represented by a temperature T_l) via electron-phonon scattering, leading a temperature rising of the lattice and simultaneously the cooling of electrons. Eventually, electrons and lattice will reach a new thermal equilibrium. This process is modeled by coupled partial differential equations. The electron-phonon interaction determines the rate of energy flow from the electrons to the lattice, as well as the related plasma dynamics.

Simulating TTM: Writing a C++ Routine

In writing the code to simulate TTM, the first step was to create a C++ program which could perform a simplified version of the calculation which does not account for the laser heat source S(x, t) or for electron transport (neglected because the film is so thin). Quantitatively, the functions describing the temperatures of the electrons and the lattice are as follows:

$$\frac{\partial T_e}{\partial t} = f(t, T_e, T_l) = \left(\frac{G}{C_e}\right)(T_l - T_e)$$

$$\frac{\partial T_l}{\partial t} = g(t, T_e, T_l) = \left(\frac{G}{C_l}\right)(T_e - T_l)$$



In this simplified case where the power of the laser is relatively low, G is the electron-phonon coupling 'constant' (approximately constant and) equal to 3x10^{16}Wm^{-3}K^{-1}. C_l is the specific heat of the lattice and equals 3.5x10^{6}Jm^{-3}K^{-1}. The parameter t is simply the time. The specific heat of the electrons, C_e, depends upon T_e such that:

$$C_e = \gamma T_e$$

Where γ is a constant equal to 71.5Jm^{-3}K^{-2}.

The two functions were used in conjunction with Euler's equations in order to calculate the temperatures of electrons and lattice over time given some smaller time interval h (in this case h=5fs). Euler's equations follow:

$$y_{n+1} = y_n + hf(t, T_e, T_l)$$

$$z_{n+1} = z_n + hg(t, T_e, T_l)$$

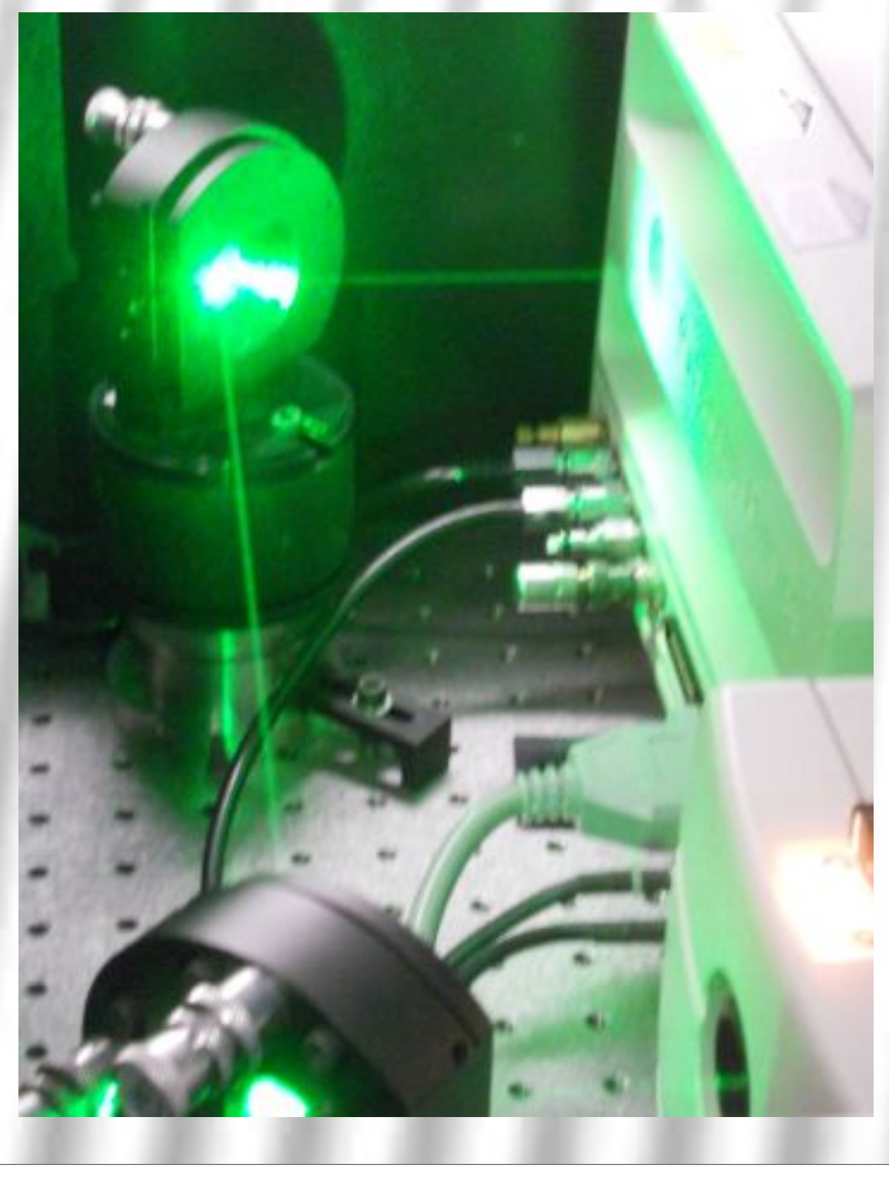
In this case, y and z are simply the values of T_e and T_l respectively. The next step in modifying the program was to account for effects due to the laser heat source, S(x, t).

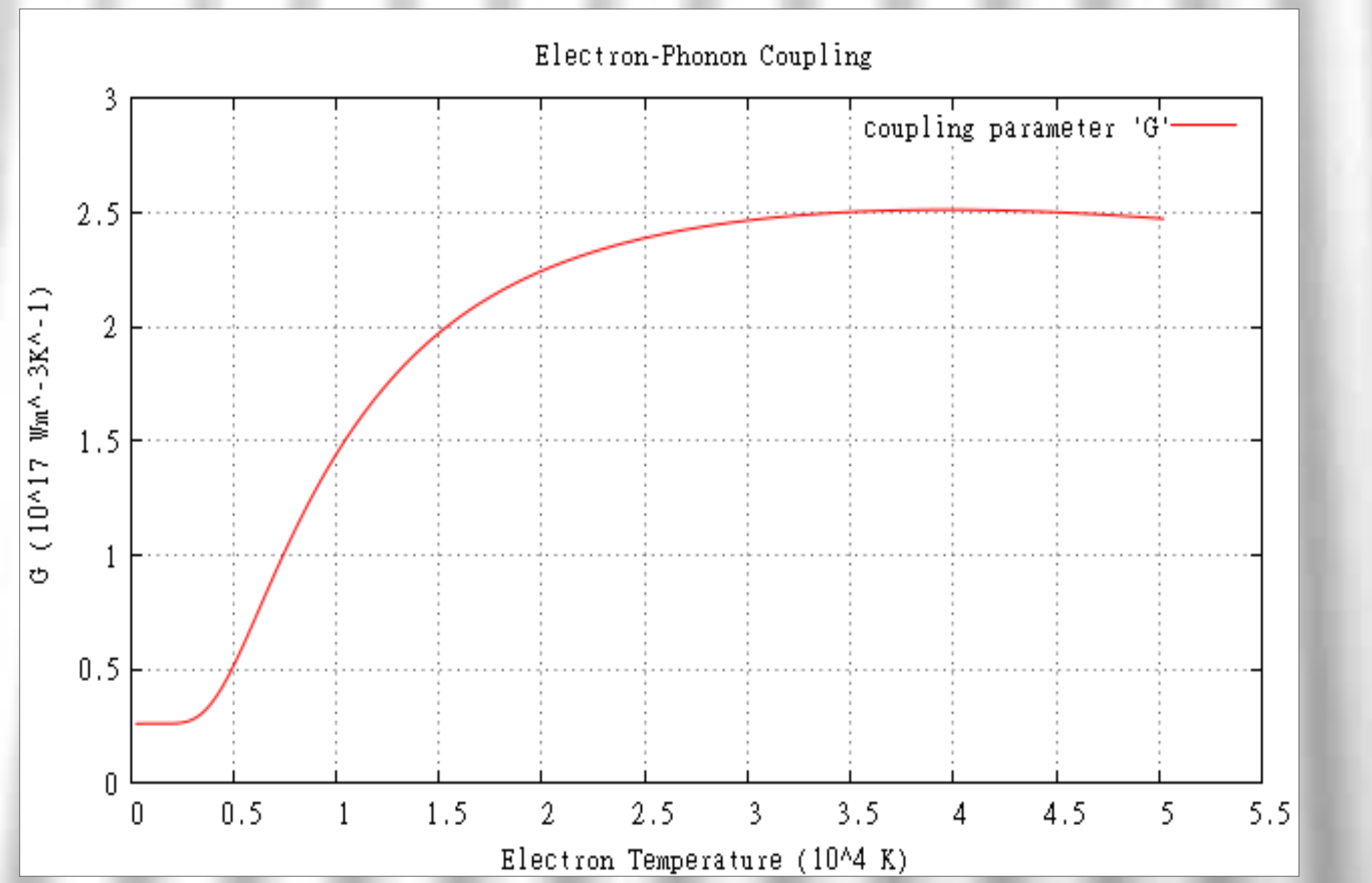
$$S(x,t) = \sqrt{\frac{\beta}{\pi}}\left(\frac{1-R}{t_p\delta_s}\right)\phi * \exp\left[-\left(\frac{x}{\delta_s}\right) - \beta\left(\frac{t-2t_p}{t_p}\right)^2\right]$$

Given the parameters: β=4ln(2), t_p=100fs, φ=13.4J/m^2, δ_s=15.3nm, and R the reflectivity; and evaluating across the depth (x) of the film from its front surface, the expression for the initial value of S (which will be added into the function 'f') simplifies. Thus, the additional term in the expression for f(t, T_e, T_l) is S/C_e such that (substituting for C_e and simplifying):

$$f(t, T_e, T_l) = \left(\frac{G}{C_e}\right)(T_l - T_e) + \left(\frac{8.803 * 10^{18}}{T_e}\right) * \exp\left[-\beta\left(\frac{t-2t_p}{t_p}\right)^2\right]$$

The final step in writing the program was to consider that the electron-phonon coupling 'constant' is not in fact a constant at (higher laser power and therefore) higher temperatures. Therefore, data from the University of Virginia regarding this value (G) and also the value of C_e, which does not follow the simple proportion (shown above) at higher temperatures, with respect to temperature was saved in two files. Gnuplot generated graphs of these data are shown.





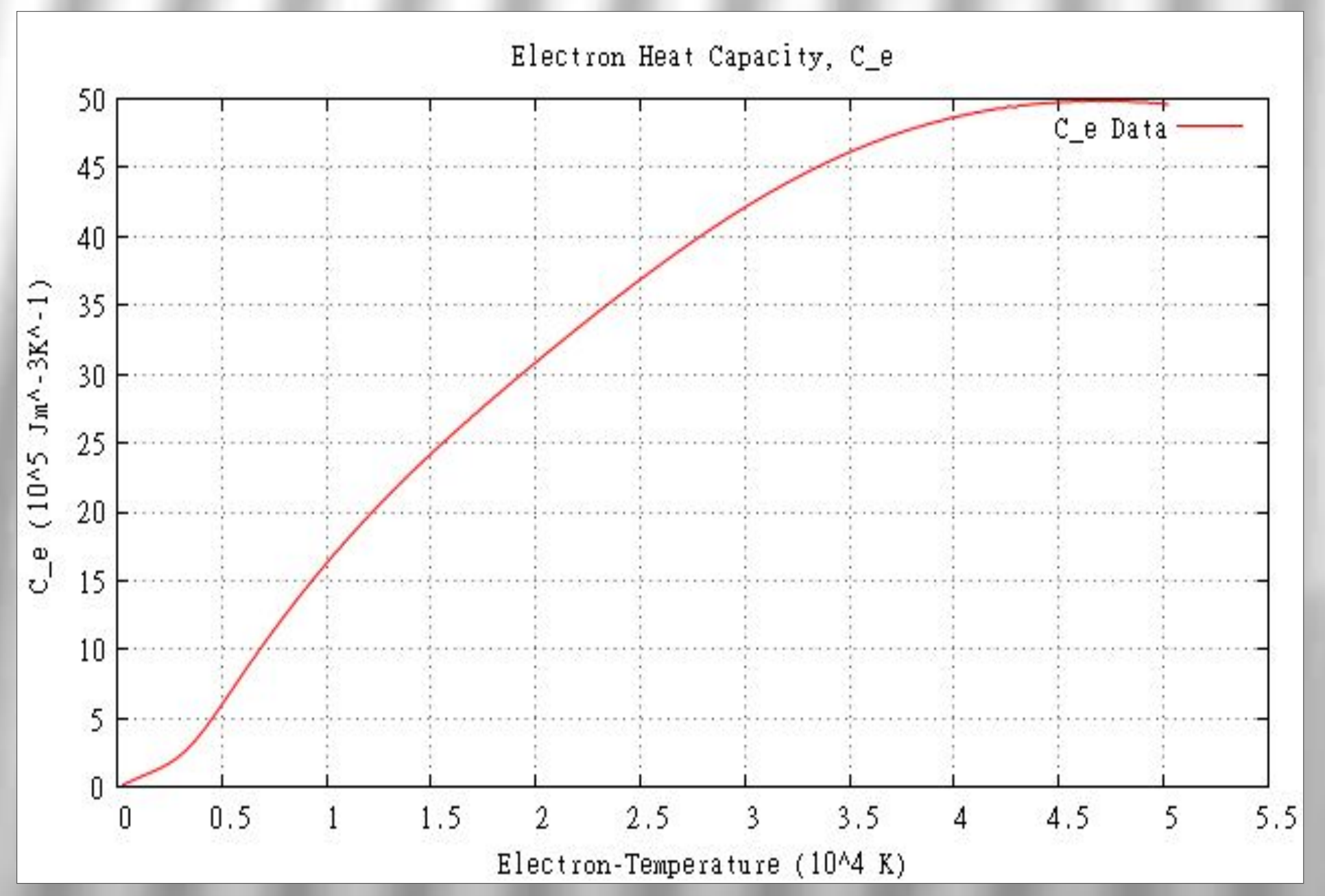


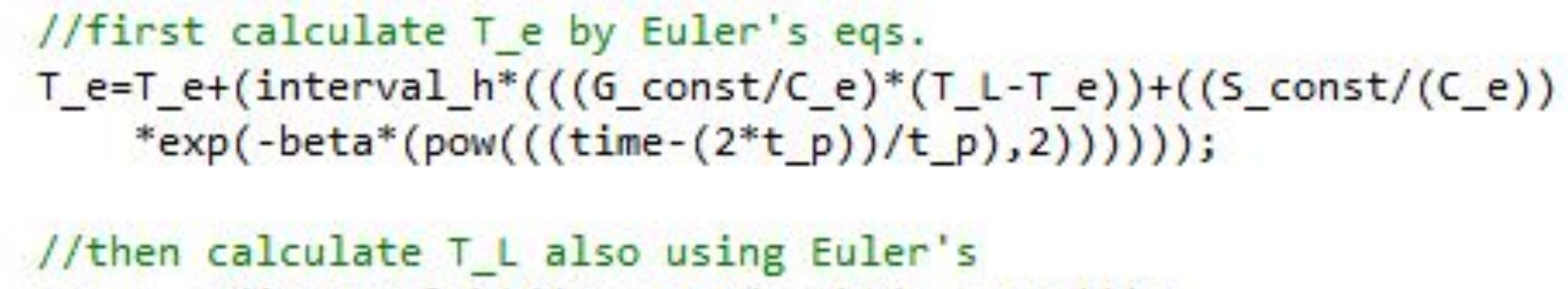
Figure 2. (Top Graph) Graph showing how the electron-phonon coupling parameter varies with electron temperature**. Figure 3**. (Bottom Graph) Graph showing electron heat capacity with respect to electron-temperature.
Picture 1. (left) Shows UHV chamber, electron gun, part of optical bench, and frequency tripler.
Picture 2. (above) Shows green continuous-wave laser (source laser) used in the lab.

The program utilizes the initial electron temperature and converts it to an index in order to look-up these two values. Originally, if no exact value was found, a linear interpolation method between the two closest points was assumed. Later, this method was exchanged for a Hermite cubic-spline interpolation which is a more precise estimate of the two desired parameters G and C_e. The next two values of T_e and T_l are then calculated using Euler's equations and the process is repeated until a given maximum time is reached. The final expressions for the functions f and g used in the program follow (next column).

$$f(t, T_e, T_l) = \left(\frac{G}{C_e}\right)(T_l - T_e) + \left(\frac{6.29 * 10^{20}}{C_e}\right) * \exp\left[-\beta\left(\frac{t-2t_p}{t_p}\right)^2\right]$$

$$g(t, T_e, T_l) = \left(\frac{G}{C_l}\right)(T_e - T_l)$$

The following shows the final C++ script of Euler's equations used for calculating the temperatures of electrons and lattice respectively:



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//first calculate T_e by Euler's eqs.
T_e=T_e+(interval_h*(((G_const/C_e)*(T_L-T_e))+((S_const/(C_e))
    *exp(-beta*(pow(((time-(2*t_p))/t_p),2))))));

//then calculate T_L also using Euler's
T_L=T_L+(interval_h*((G_const/C_L)*(T_e-T_L)));
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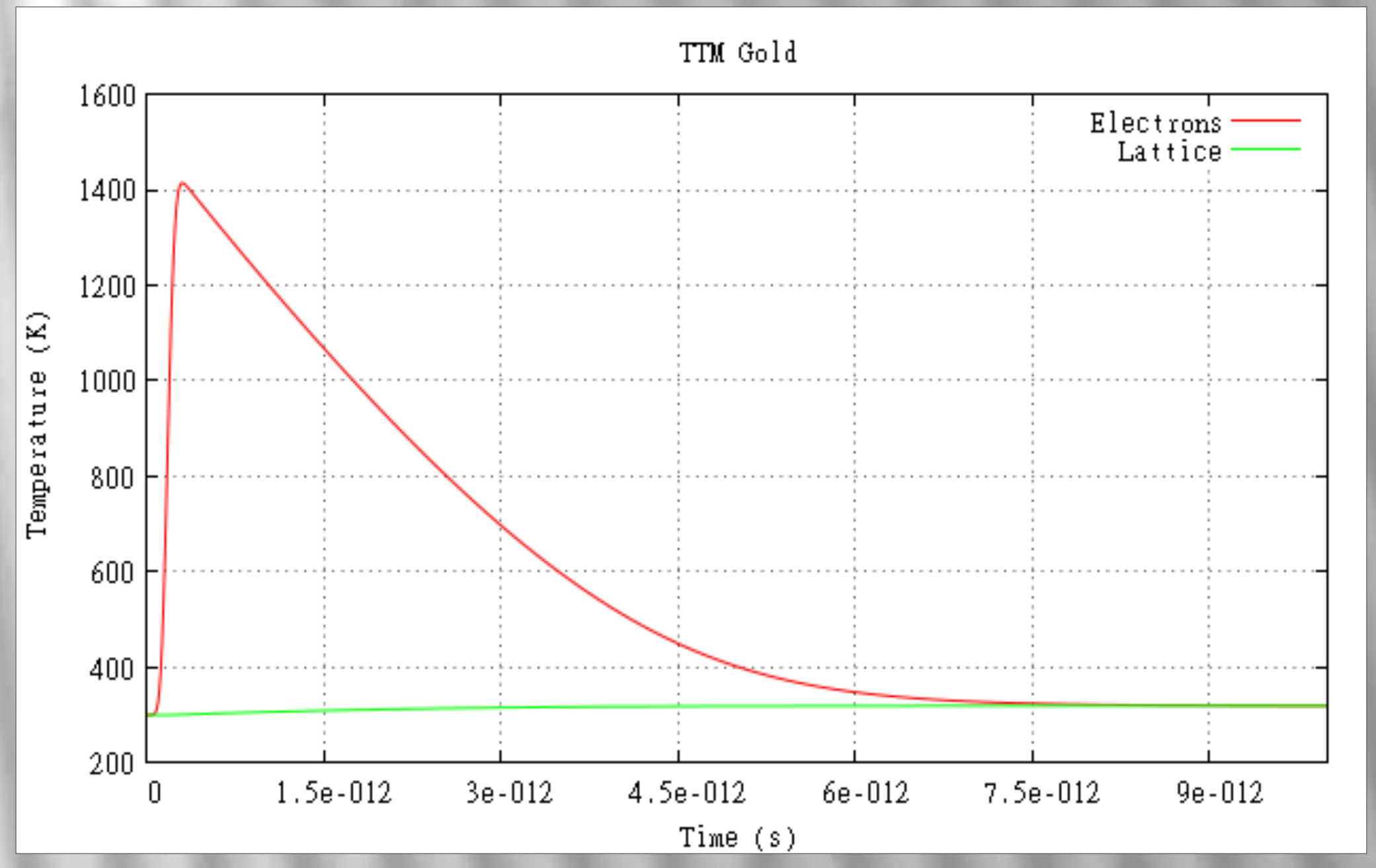
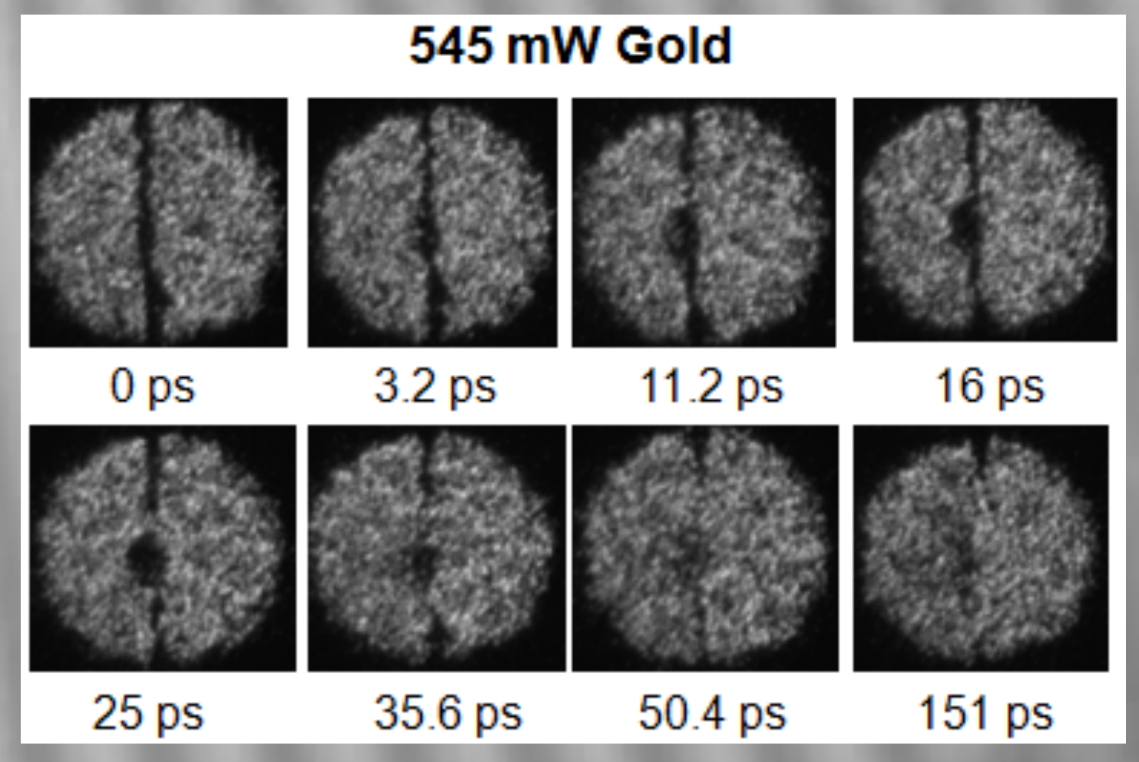
Figure 4. The graph illustrates the calculations produced by the simulation with both lattice and electrons having an initial temperature of 300K, and allowing the simulation to run up to a maximum time of 10ps.



Conclusion

Over the course of this project, a C++ program to mathematically simulate TTM was developed for the case of a thin gold foil and an 800nm laser (pump line). The temperature transfer predicted by TTM cannot yet be experimentally verified due to the minuscule timescale in which it occurs. Therefore, a method must be developed and in the mean time different theoretical models should be compared against each other, and in light of electron shadow imaging and deflectometry measurements. In the future, this program could be used to model heat flow in different metals including Nickel and Silver, for which there are already plans of further investigation.

Figure 5. Electron-shadow images of a gold nano-film with 545μJ pump energy at various time delays. **Picture 3**. Laboratory optical bench and laser.



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