Heat transfer from ultrafast laser heated Bi_2Se_3 film to a sapphire substrate

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1 Introduction

This note describes how to calculate one-dimensional thermal transport from an ultrafast laser excited Bismuth Selenide film into a sapphire substrate. The laser rapidly (≈ 1 ps) raises the temperature of the film uniformly. The film has been depositied on top of a bulk material, which is initially at a uniform colder temperature. The specific experiments considered are 20 nm and 150 nm thick films on top of sapphire (on the optic axis). This treatment neglects the nanoscale properties of the Bismuth Selenide films, which in fact is made of multiple quantum layers.

2 Classical Treatment

2.1 Perfect thermal contact

These results are taken from Example 10.8 of Hahn and Ozisik, *Heat Conduction* (Wiley, 2012). They consider a one-dimensional, two-layer composite slab with a film of thickness L on top of a semi-infinite bulk material. We will identify the film as region 1 and the bulk as region 2. The layers are presumed to be in perfect thermal contact with region 1 initially at a uniform temperature T_0 (caused by rapid laser energy absorption) and region 2 at zero temperature. The problem is easiest stated using a dimensionless temperature $\theta_i(x,t)$ defined as as

$$\theta_i(x,t) = \frac{T_i(x,t)}{T_0} \qquad i = 1,2 \tag{1}$$

where the index $i = 1,2$ refers to the film and bulk, respectively. With this transformation, the heat transfer problem is written

$$\frac{\partial^2\theta_1}{\partial x^2} = \frac{1}{\alpha_1}\frac{\partial\theta_1}{\partial t} \qquad in \qquad 0 < x < L,\ t > 0 \tag{2a}$$

$$\frac{\partial^2\theta_2}{\partial x^2} = \frac{1}{\alpha_2}\frac{\partial\theta_2}{\partial t} \qquad in \qquad x > L,\ t > 0 \tag{2b}$$

subject to the boundary conditions

$$\left.\frac{\partial\theta_1}{\partial x}\right|_{x=0} = 0 \tag{3a}$$

$$\theta_1(x = L, t) = \theta_2(x = L, t) \tag{3b}$$

$$k_1\left.\frac{\partial\theta_1}{\partial x}\right|_{x=L} = k_2\left.\frac{\partial\theta_2}{\partial x}\right|_{x=L} \tag{3c}$$

$$\theta_2(x\to\infty, t) \to 0 \tag{3d}$$

and the initial conditions

$$\theta_1(x, t=0) = 1 \qquad in \qquad 0 < x < L \tag{4a}$$

$$\theta_2(x, t=0) = 0 \qquad in \qquad L < x < \infty \tag{4b}$$

where k_i are the thermal conductivities and α_i are the thermal diffusivities of region 1 (film) and region 2 (bulk). Using Laplace transforms, the solution for the temperature distribution in the two-layer medium is

$$\frac{T_1(x,t)}{T_0} = 1 - \frac{1+\gamma}{2}\sum_{n=0}^{\infty}\gamma^n\left\{\mathrm{erfc}\left[\frac{(2n+1)L-x}{2\sqrt{\alpha_1 t}}\right] + \mathrm{erfc}\left[\frac{(2n+1)L-x}{2\sqrt{\alpha_1 t}}\right]\right\} \tag{5a}$$

$$\frac{T_2(x,t)}{T_0} = \frac{1+\gamma}{2}\sum_{n=0}^{\infty}\gamma^n\left\{\mathrm{erfc}\left[\frac{(2nL+\mu(x-L)}{2\sqrt{\alpha_1 t}}\right] - \mathrm{erfc}\left[\frac{(2n+2)L+\mu(x-L)}{2\sqrt{\alpha_1 t}}\right]\right\} \tag{5b}$$

where the unitless parameters μ and γ are defined by

$$\mu = \sqrt{\frac{\alpha_1}{\alpha_2}} \tag{6a}$$

$$\beta = \frac{k_1}{k_2}\frac{1}{\mu} \tag{6b}$$

$$\gamma = \frac{\beta-1}{\beta+1}. \tag{6c}$$

2.2 Examples of the classical heat conduction result

In addition to calculating the temperature profile evolution in the film and substate, we perform kinematic Time-Resolved X-Ray Diffraction (TRXD) calculations to predict the evolution of diffraction peak angle shift from the average heating of the filmat an absorbed fluence of 0.1 mJ/cm^2. TRXD measurements are all done for the [006] reflection at an x-ray energy of 10 keV. The average centroid shift is calculated using the differential Bragg formula,

$$\Delta\theta = -\frac{\alpha_t\langle T_1\rangle}{cot(\theta_B)} \tag{7}$$

whre α_t is the thermal expansion coefficient, $\langle T_1\rangle$ is the mean temperature of the film, $\Delta\theta$ is the peak centroid shift to be measured by comparission with the rocking curve before the laser excites the sample, and θ_B is the Bragg angle. For these experimental parameters, $\theta_B = 7.7^\circ$ and $\alpha_t = 1.9\times10^{-5}$.

2.2.1 20 nm film

A 20 nm thick film gives a temperature rise of 38.6 K. After 9.9 ns, the average film temperature rise is only 0.7 deg C. The maximum bulk temperature rise is only 3.4 deg C.

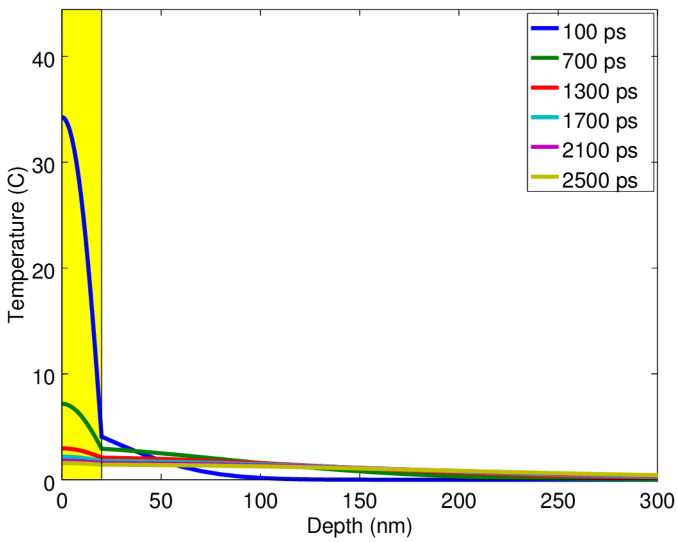

Figure 1: Classical thermal transport result for 20 nm Bi_2Se_3 on Sapphire with an absorbed laser fluence of 0.1 mJ/cm^2.

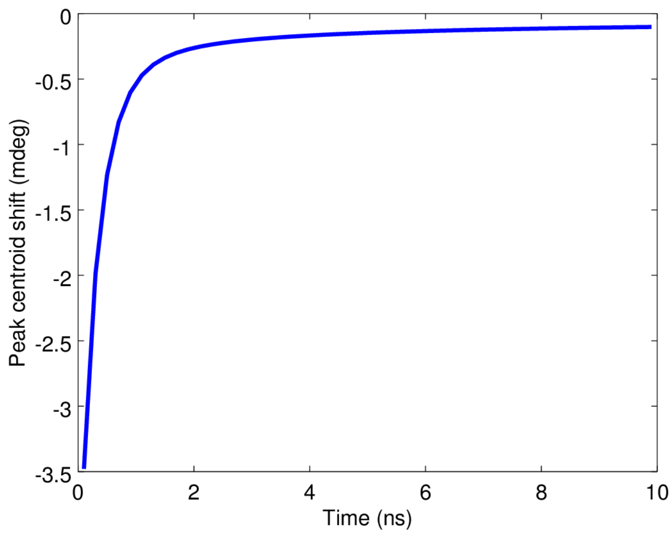

Figure 2: TRXD calculations corresponding to classical thermal transport result for 20 nm Bi_2Se_3 on Sapphire. This is a kinematic diffraction calculation, and the centroid shift is measured relative to before the laser excites the sample.

2.2.2 150 nm film

A 150 nm thick film gives a temperature rise of 5.1 K. After 9.9 ns, the average film temperature rise is only 2.5 deg C. The maximum bulk temperature rise is only 0.5 deg C.

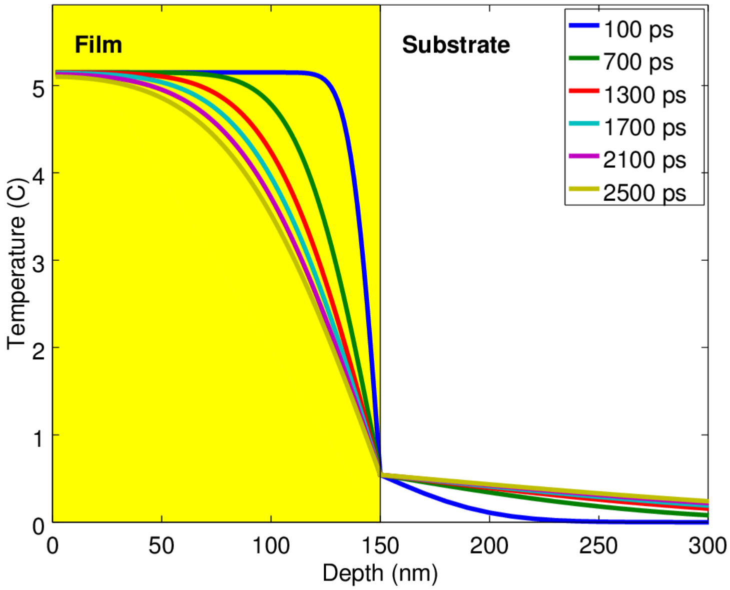

Figure 3: Classical thermal transport result for 150 nm Bi_2Se_3 on Sapphire with an absorbed laser fluence of 0.1 mJ/cm^2.

Appendix: Parameters

parameter	Bi_2Se_3	Sapphire	unit
Specific Heat	189.83	761.00	J/(kg K)
Density	6820	3980	kg/m^3
Thermal Conductivity	0.75	23.1	W/(m K)
Thermal Expansion	1.9×10^{-5}	-	1/K
Bragg Angle	7.7	-	degrees

Table 1: Material properties used in these calculations.

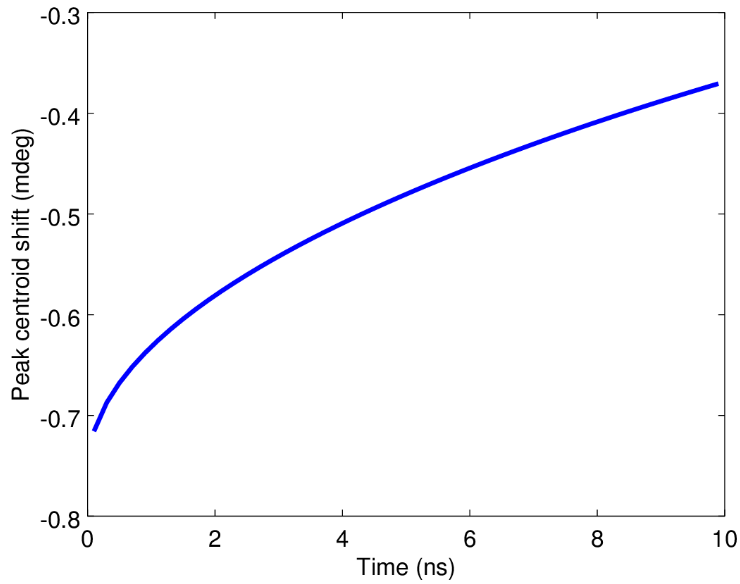

Figure 4: TRXD calculations corresponding to classical thermal transport result for 150 nm Bi_2Se_3 on Sapphire. This is a kinematic diffraction calculation, and the centroid shift is measured relative to before the laser excites the sample.

Appendix: Code

The most recent and up to date code should be obtained from `https://github.com/elandahl/Bi2Se3_thermal`. This code listing is for reference only.

```
% thermalFilm.m
% Classical thermal calc for Bismuth Selenide (Bi2Se3) film on sapphire substrate
% Presumes that the temperature rise on the film is uniform and instantaneous
% and that the thermal contact is perfect
% A more sophisticated model with better Physics (e.g. AMM, DMM) will be needed!
% Also, does not include nanoscale phenomena (e.g. phonon mean free path)
% Also, no acoustic propogation is included.  This could be called later.
% Model: see p. 429 (Sec. 10.7) of Hahn, "Heat Conduction" 3rd edition
% Material properties are hard coded
% Based on thermalFilm.m, First written by Eric Landahl, 12.28.2016
% Revised by EL 2.12.2017
%
%% INPUTS:
%   L           Bi2Se3 film thickness in meters (sapphire substrate is infinite)
%   fluence     absorbed laser fluence in mJ/cm^2
%   time        a vector of times to be calculated in seconds
%   max_depth   maximum depth calculated into sapphire, typ. 5 * extinction depth
%% OUTPUTS:
%   T1         temperature of film, size = length(time) x length(z1)
%   T2         temperature of substrate, size = length(time) x length(z2)
%   z1         a vector of film depths in meters
```

```
%   z2         a vector of substrate depths in meters
%
%% TYPICAL USAGE
%
% [T1 T2 z1 z2] = Bi2Se3_thermal (150e-9, 1, (1e-10:2e-10:1e-8), 1e-5);
%
function [T1 T2 z1 z2]=Bi2Se3_thermal (L,fluence,time,max_depth)

% Bi2Se3 film properties.  "1" refers to the film
  C1 = 124.3*1000/654.8; %Specific heat of film in J/(kg K)
  rho1 = 6.82e3; % Film density in kg/m^3
  k1 = 0.75; % Film thermal conductivity in W/(m K)
  %alpha1 = 0.312e-4 % Film thermal diffusivity in m^2/s
  alpha1 = k1/(rho1 * C1); % Film thermal diffusivity in m^2/s

% Sahhpire substrate properties.  "2" referes to the substrate
  C2 = 761; % Specific heat of sapphire in J/(kg K)
  rho2 = 3.98e3; % Sapphire density in kg/m^3
  k2 = 23.1; % Sapphire thermal conductivity in W/(m K) parallel to optic axis
  alpha2 = k2/(rho2 * C2); % Sapphire thermal diffusivity in m^2/s

  %% Temporary for troubleshooting: make the sampe all semiconductor
%  rho1 = rho2;
%  k1 = k2;
%  C1 = C2;
%  alpha1 = alpha2;
[k1 k2]
[C1 C2]
[rho1 rho2]

% Calculate initial temperature rise
  fluence = fluence*10; % Convert from mJ/cm^2 to J/m^2
  T0 = fluence/(L * C1 * rho1); % Initial temperature rise in film
  fprintf('A %d nm thick Bi2Se3 film gives a temperature rise of %.1f K.\n',L*1e9,T0)

% Unitless parameters (see Hahn, "Thermal Conductivity", Eqs. 10-135 and 10-138)
  mu = sqrt(alpha1/alpha2);
  beta = (k1/k2)/mu;
  gamma = (beta - 1)/(beta + 1);

% Spatial grid
  num_depths = 1000;  % number of depth points z to be calculated
  dz = max_depth/num_depths;
  z = dz:dz:max_depth;

% Meshgrid for calculation speed & ease
  [Time Z] = meshgrid(time,z); % Time and Z are 2D, time and z are 1D

% Calculate temperature profile in bulk
```

```
  max_n = 100; % number of terms in series expansion, default 100
  T2a = 0.*Time.*Z; % each term gets added to this, starts at zero
  for n = 0: max_n  % Series expansion solution of heat equation
    T2b = erfc((2*n*L + mu*Z)./(2*(sqrt(alpha1*Time)))); % temporary
    T2c = erfc(((2*n + 2)*L + mu*Z)./(2*sqrt(alpha1*Time))); % temporary
    T2a = T2a + (gamma^n) * (T2b - T2c); % temporary, adding up
  end
  T2 = T0 * (1/2) * (1 + gamma) * T2a; % Temperature at all z and time

% Film calculations
% Calculate temperature profile in film.  zz and ZZ are the film depths
  dzz = L/100; % Choose 100 depth points in the film by default
  zz = dzz:dzz:L;
  [Time ZZ] = meshgrid(time,zz);
  T1a = 0.*Time.*ZZ;
  for n = 0:max_n
    T1b = erfc(((2*n + 1)*L - ZZ)./(2*sqrt(alpha1*Time)));
    T1c = erfc(((2*n + 1)*L + ZZ)./(2*sqrt(alpha1*Time)));
    T1a = T1a + (gamma^n) * (T1b + T1c);
  end
  T1 = T0 - T0 * (1/2) * (1 - gamma) * T1a; % Temperature in film
  T1_end = mean(T1(:,end)); % Average temperature at final timepoint
  fprintf('After %.1f ns, the average film temperature rise is only %.1f deg C.\n', ...
  time(end)*1e9,T1_end);
  fprintf('The maximum bulk temperature rise is %.1f deg C.\n', max(max(T2)));

  z1 = zz; % for output
  z2 = z; % for output

% Calculate heat in film
% Q = integral dzz of rho*C*T
%  Q1 = trapz(ZZ,T1*rho1*C1);
%  Q2 = trapz(Z,T2*rho2*C2);
%  Q1 = Q1/10; % convert from J/m^2 to mJ/cm^2
%  Q2 = Q2/10; % convert from J/m^2 to mJ/cm^2

% Outputs not needed for TRXD

save thermalFilmOut.m; % Save all variables for future use

  end
```