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LABORATORY DIRECTED RESEARCH & DEVELOPMENT

Implementing and testing a Kinetic Phase Transition Model

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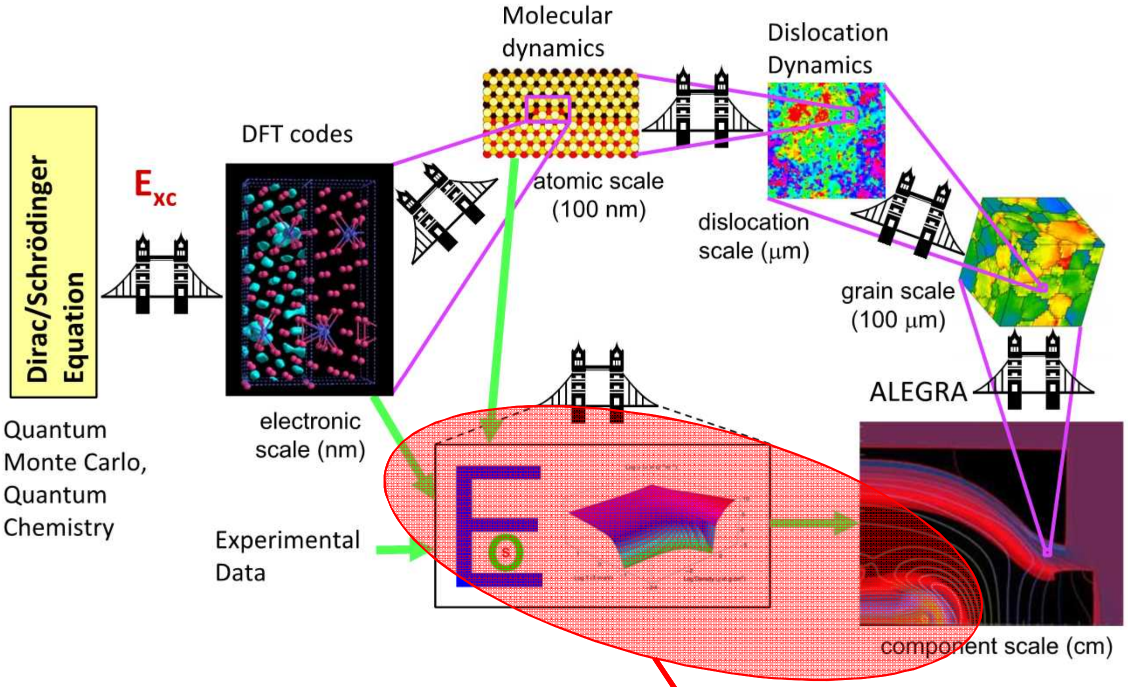
Bridges between Fundamental Law of Nature and Engineering: Kinetics of Phase transitions



Dirac/Schrödinger Equation

E_{xc}

DFT codes

Molecular dynamics

Dislocation Dynamics

atomic scale (100 nm)

dislocation scale (μm)

grain scale (100 μm)

ALEGRA

Quantum Monte Carlo, Quantum Chemistry

electronic scale (nm)

Experimental Data

component scale (cm)

Implementing a kinetic phase transition model into hydro-codes.

- *Developing, documenting and implementing Carl Greeff's (LANL) kinetic phase transition model into Lambda for use in LASLO and ALEGRA for advanced modeling.*

Kinetics of Phase transitions

Only difference between simulations (dashed lines) is how fast the phase transition occur.

Carl Greeff (LANL) has proposed an empirical model for taking this into account and I am implementing it into the codes the scientists at the Z-machine use at Sandia.

Lambda provides subroutines for use in LASLO, ALEGRA, and other codes at Sandia. Lambda can also use these subroutines internally, for testing etc.

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MODELING DYNAMIC PHASE TRANSITIONS IN TI AND ZR

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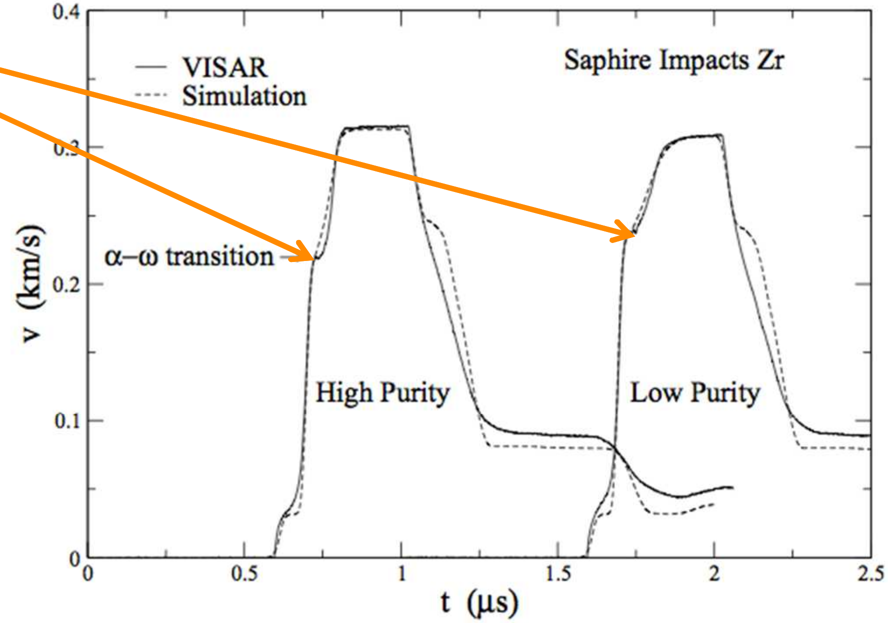

FIGURE 5. Impurity effects on $\alpha - \omega$ transition in Zr. Zr samples impacted by sapphire flyer with sapphire window. Solid curve - VISAR, dashed curve - simulation.

Kinetics of Phase Transitions

- A new variable is needed: the mole fractions of the phases (how much of each phase is present in each element).
- All phases have same temperature and pressure:
  Given input internal energy and density (specific volume) for an element, determine specific volume and internal energy for each of the phases present.
- Given a time step and the model for the kinetics of the phases, update the mole fractions.

Additional requirement: a complete EOS for each of the phases.

All these things are highly non-trivial.

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A complete EOS for each of the phases

Phase transitions are determined by Gibbs Free energy (G), which is also present in any kinetic phase transition model:

$$G = E - T S + P V$$

E: Internal Energy
T: Temperature
S: Entropy
P: Pressure
V: Specific volume

Entropy needed, not only Pressure and Internal Energy as functions of specific volume and temperature as are usually available in EOS models.

The first task: Deriving, and implementing into Lambda, entropy for the Vinet and Mie-Grüneisen models.

Here I call "Mie Grüneisen" the one with a Hugoniot reference curve.
What I call "Vinet" is an extended Vinet formulation with temperature dependence.

Entropy

Both Vinet and Mie-Grüneisen are incomplete EOSs.
They are not derived in a thermodynamically consistent way from a thermodynamic potential, such as the Helmholtz free energy.
There can be more than one, or none, complete EOS consistent with an incomplete EOS.

The assumptions in both Vinet and Mie-Grüneisen are:

- The specific heat at constant volume (C_V) is constant.
- The Grüneisen parameter (Γ) times density (ρ) is constant.
  (Or equivalently, volumetric thermal expansion (α) times isothermal bulk modulus (B) is constant, since $C_V \Gamma \rho = \alpha B$ by definition.)

$$S(V,T) = S_0 + \alpha_0 B_0 \left(V - V_0\right) + C_{V0} \ln \frac{T}{T_{ref}} \, .$$

where S_0 is the entropy at V_0 and T_{ref}.

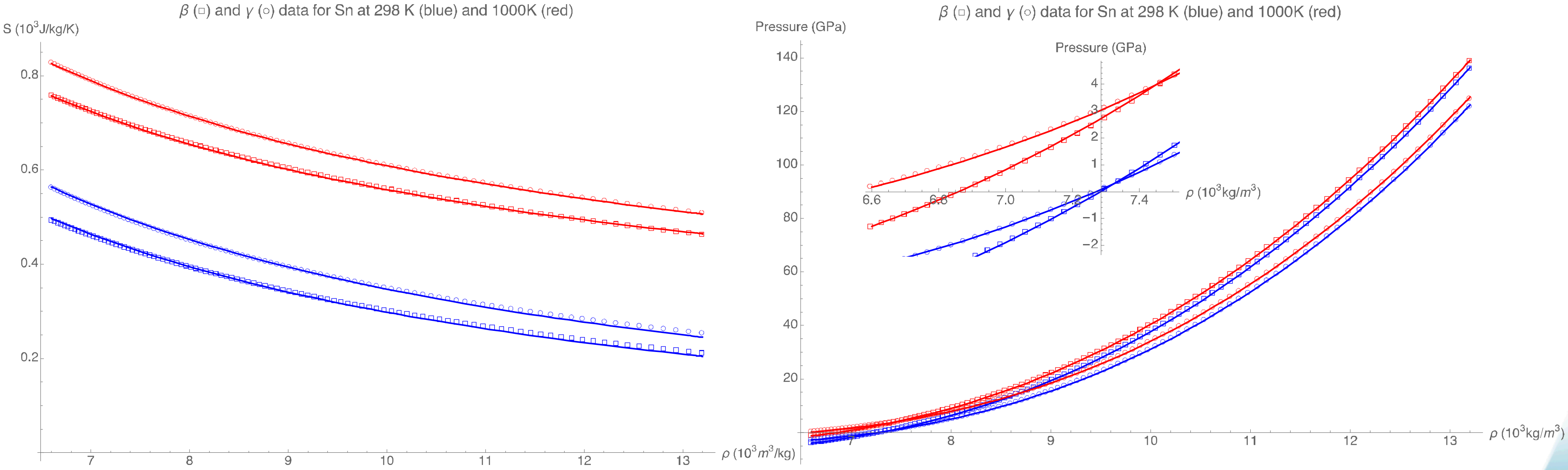
This form violates the **third law of thermodynamics** which states that the entropy of a system at zero temperature is a well-defined constant. Should mainly be used for $T > \approx T_{ref}$.

In addition we can obtain negative temperatures in the iterative procedure we need for finding the specific volumes and specific energies for the individual phases. We use: If $T <= 0$ we set $S=0$.

Phase models

For a specific material we need one separate EOS model for each phase.
We (Sandia) do not have those models yet.

Carl Greeff has his own EOS generating code and I got a few tabulated isotherms for the β and γ phases of Tin (Sn) from him that I fitted the Vinet model parameters to reproduce:



Entropy Examples Pressure

Justin has parameterized a few other multi phase tables using this modified extended Vinet formula with good results.

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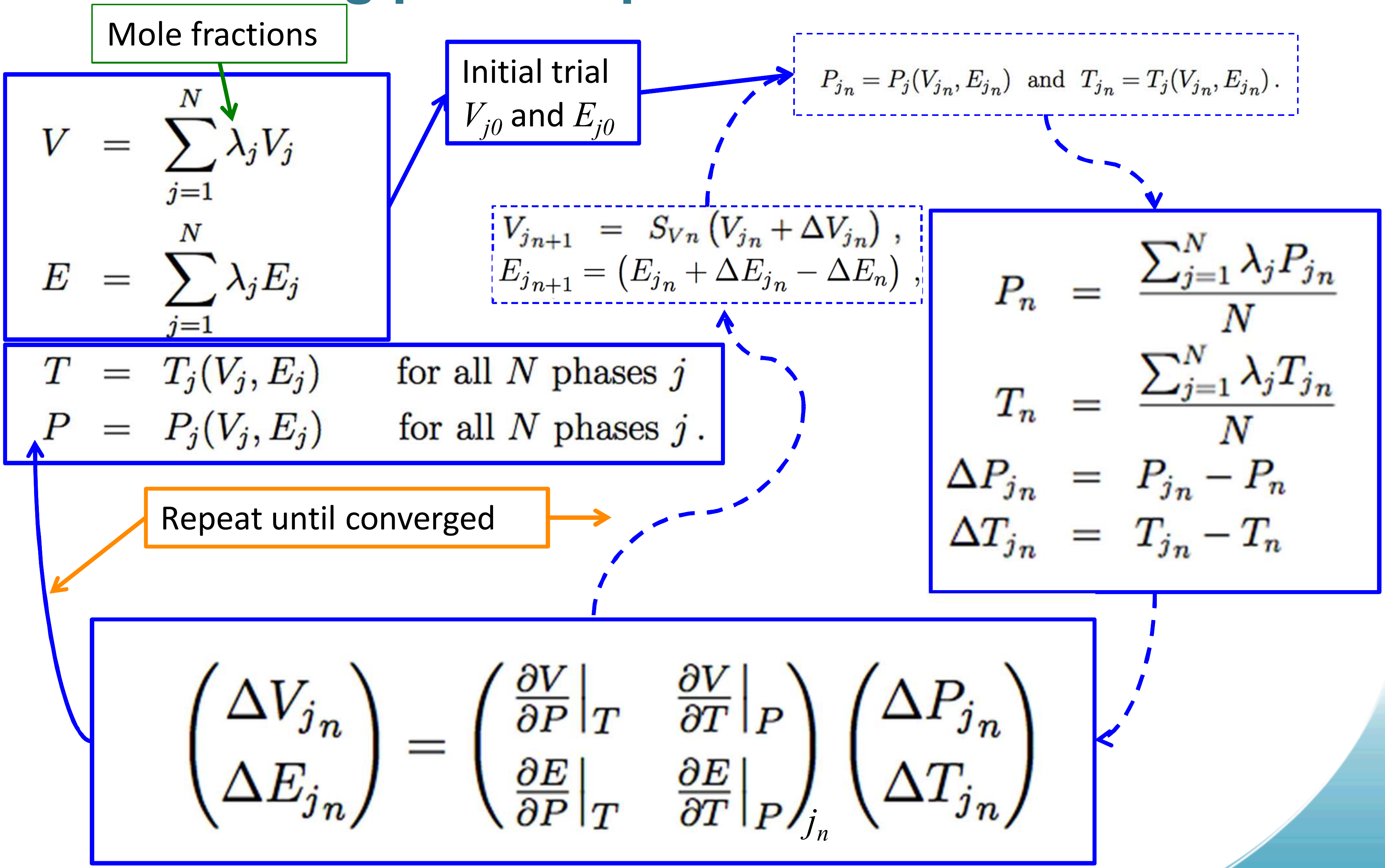
Determining phase equilibrium



Mole fractions

$$V = \sum_{j=1}^{N} \lambda_j V_j$$

$$E = \sum_{j=1}^{N} \lambda_j E_j$$

$$\begin{aligned} T &= T_j(V_j, E_j) && \text{for all } N \text{ phases } j \\ P &= P_j(V_j, E_j) && \text{for all } N \text{ phases } j\,. \end{aligned}$$

Initial trial V_{j0} and E_{j0}

$$P_{j_n} = P_j(V_{j_n}, E_{j_n}) \quad \text{and} \quad T_{j_n} = T_j(V_{j_n}, E_{j_n})\,.$$

$$\begin{aligned} P_n &= \frac{\sum_{j=1}^{N} \lambda_j P_{j_n}}{N} \\ T_n &= \frac{\sum_{j=1}^{N} \lambda_j T_{j_n}}{N} \\ \Delta P_{j_n} &= P_{j_n} - P_n \\ \Delta T_{j_n} &= T_{j_n} - T_n \end{aligned}$$

$$\begin{pmatrix} \Delta V_{j_n} \\ \Delta E_{j_n} \end{pmatrix} = \begin{pmatrix} \frac{\partial V}{\partial P}\Big|_T & \frac{\partial V}{\partial T}\Big|_P \\ \frac{\partial E}{\partial P}\Big|_T & \frac{\partial E}{\partial T}\Big|_P \end{pmatrix}_{j_n} \begin{pmatrix} \Delta P_{j_n} \\ \Delta T_{j_n} \end{pmatrix}$$

$$\begin{aligned} V_{j_{n+1}} &= S_{Vn}\left(V_{j_n} + \Delta V_{j_n}\right)\,, \\ E_{j_{n+1}} &= \left(E_{j_n} + \Delta E_{j_n} - \Delta E_n\right)\,, \end{aligned}$$

Repeat until converged

Key to stable implementation

$$\boxed{\begin{pmatrix}\Delta V_{j_n}\\ \Delta E_{j_n}\end{pmatrix} = \boxed{\begin{pmatrix}\left.\frac{\partial V}{\partial P}\right|_T & \left.\frac{\partial V}{\partial T}\right|_P\\ \left.\frac{\partial E}{\partial P}\right|_T & \left.\frac{\partial E}{\partial T}\right|_P\end{pmatrix}_{j_n}}\begin{pmatrix}\Delta P_{j_n}\\ \Delta T_{j_n}\end{pmatrix}}$$

Key to stable implementation:

Analytical derivatives. Thermodynamic derivatives galore…
Hydro-code: Internal Energy E(S,V)
EOS models: Helmholtz free energy F(V,T)
Phase transitions: Gibbs free energy G(P,T)

Example: If we have Helmholtz free energy quantities:

$$\left.\frac{\partial E}{\partial T}\right|_P = \left.\frac{\partial E}{\partial T}\right|_\rho - \frac{\left.\frac{\partial P}{\partial T}\right|_\rho \left.\frac{\partial E}{\partial \rho}\right|_T}{\left.\frac{\partial P}{\partial \rho}\right|_T}$$

Long term plan: Add capability to use multi phase tables in the UTri format, to be able to tabulate these derivatives explicitly to eliminate numerical noise.

Kinetics of Phase Transitions

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- Given a time step and the model for the kinetics of the phases, update the mole fractions.

Additional requirement: a <u>complete</u> EOS for each of the phases.

All these things are highly non-trivial.

Finally: Updating mole fractions

Exponential can give numerical troubles.

$$R_{ij} = \nu_{ij}\,\theta(G_i - G_j)\,\frac{(G_i - G_j)}{B_{ij}}\exp\left((G_i - G_j)^2/B_{ij}^2\right)$$

Calculate and store $log\ R_{ij}$

$$\dot{\lambda}_i = \sum_j \lambda_j R_{ji} - \lambda_i R_{ij}\,.$$

$$\lambda_j^{new} = \lambda_j + \dot{\lambda}_j \Delta t$$

Make sure updated molfractions are not becoming negative by subdividing the time step and updating successively.

Assumption in subdividing time step: Rates do not change substantially over the total time step. Which means Gibbs free energy for a phase should not change substantially during the phase transition.
Main problem: Sign change of G_i-G_j results in numerical slushing between phases. More work needed here.

Updating thermodynamic quantities

Analytical derivatives. Thermodynamic derivatives galore…
Hydro-code: Internal Energy E(S,V)
EOS models: Helmholtz free energy F(V,T)
Phase transitions: Gibbs free energy G(P,T)

Only derivatives along isotherms and isobars are well defined and we need to rewrite other thermodynamic derivatives in terms of these. These also gives a bridge between phase derivatives and total derivatives.
Example:

$$\left.\frac{\partial E}{\partial T}\right|_P = \left.\frac{\partial E}{\partial T}\right|_\rho - \frac{\left.\frac{\partial P}{\partial T}\right|_\rho \left.\frac{\partial E}{\partial \rho}\right|_T}{\left.\frac{\partial P}{\partial \rho}\right|_T} = \sum_{j=1}^{N} \lambda_j \left.\frac{\partial E_j}{\partial T}\right|_{\rho_j} - \sum_{j=1}^{N} \lambda_j \frac{\left.\frac{\partial P}{\partial T}\right|_{\rho_j} \left.\frac{\partial E_j}{\partial \rho_j}\right|_T}{\left.\frac{\partial P}{\partial \rho_j}\right|_T} .$$

Most complicated (in my view): Sound speed:

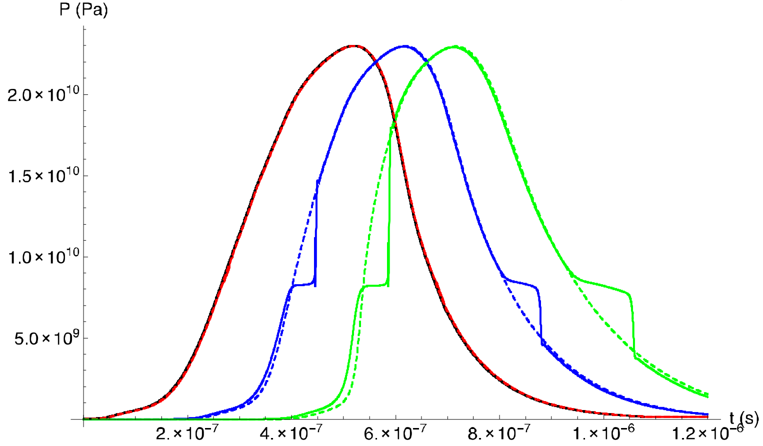
$${C_S}^2 \frac{\left.\frac{\partial E}{\partial T}\right|_\rho}{\left.\frac{\partial P}{\partial \rho}\right|_T} = \sum_{j=1}^{N} \lambda_j C_{S_j}^2 \frac{\left.\frac{\partial E_j}{\partial T}\right|_{\rho_j}}{\left.\frac{\partial P}{\partial \rho_j}\right|_T} .$$

Implementation in Laslo: Sn

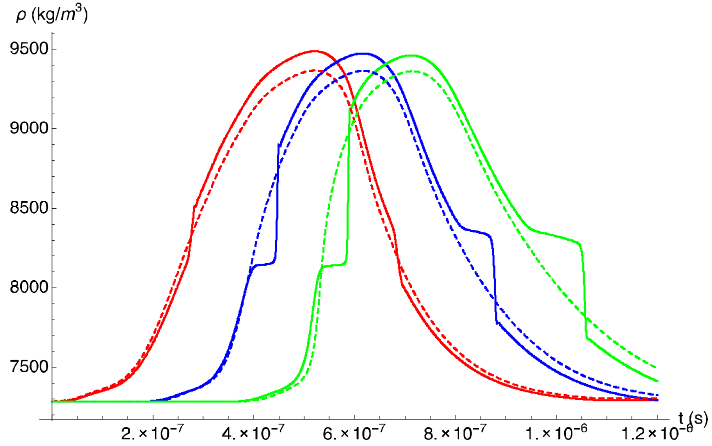
Working with Justin Brown to test it and getting experience on how to use it.

Full: KPT, Dashed: A standard Mie Grüneisen.

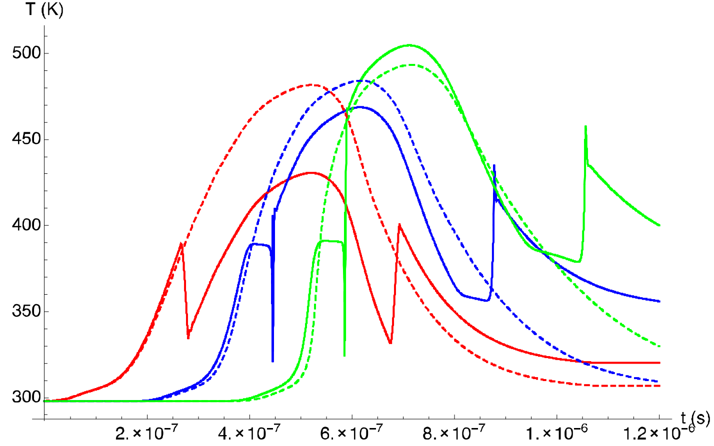
Pressure: Input (black),
at x=0 (red), at x=0.0005 m (blue), and at x=0.001 m (green).



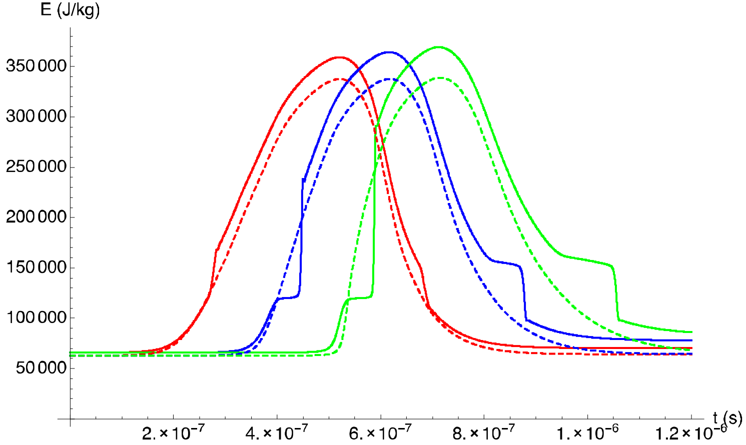
Density:
at x=0 (red), at x=0.0005 m (blue), and at x=0.001 m (green).



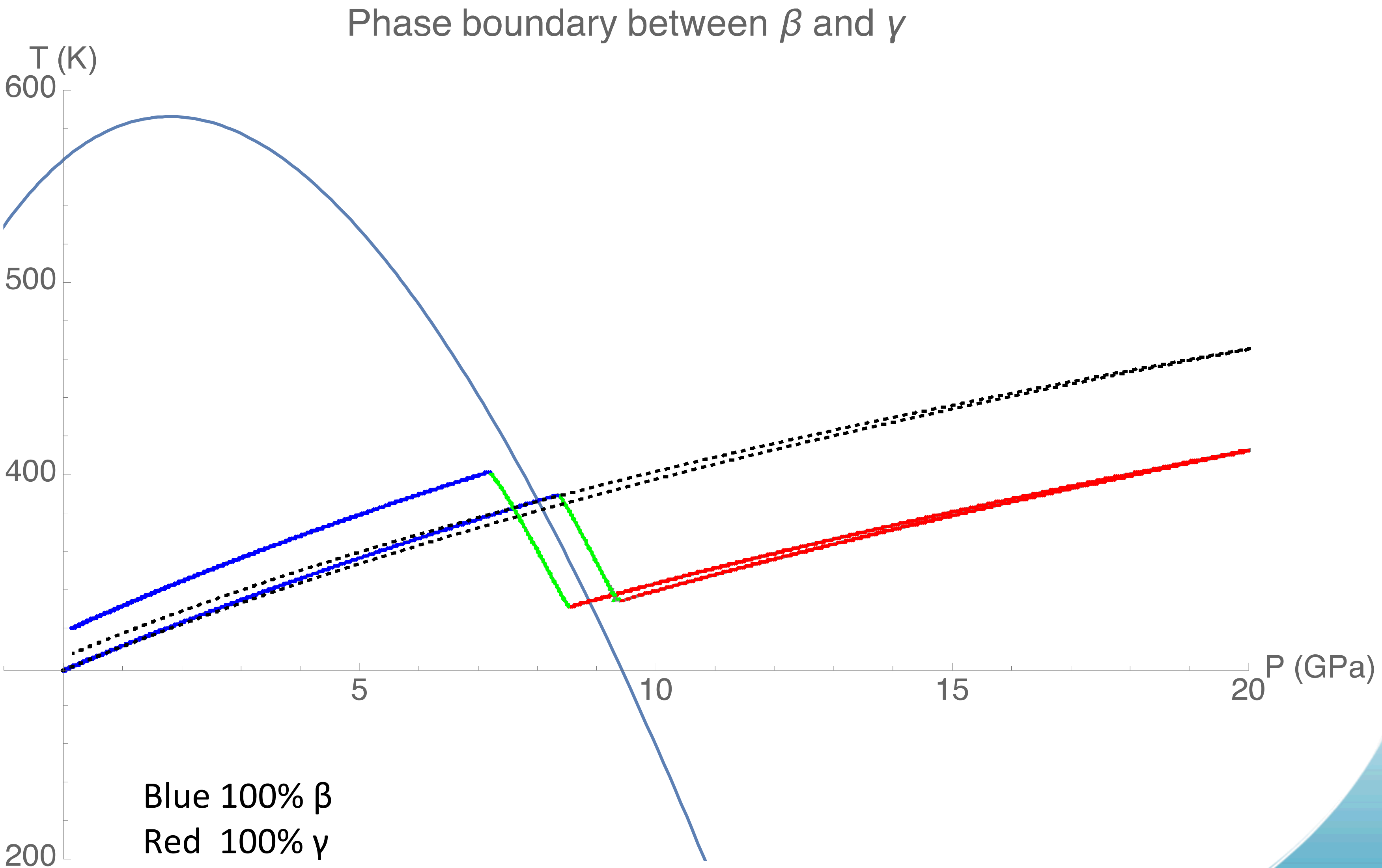
Temperature:
at x=0 (red), at x=0.0005 m (blue), and at x=0.001 m (green).



Internal Energy:
at x=0 (red), at x=0.0005 m (blue), and at x=0.001 m (green).

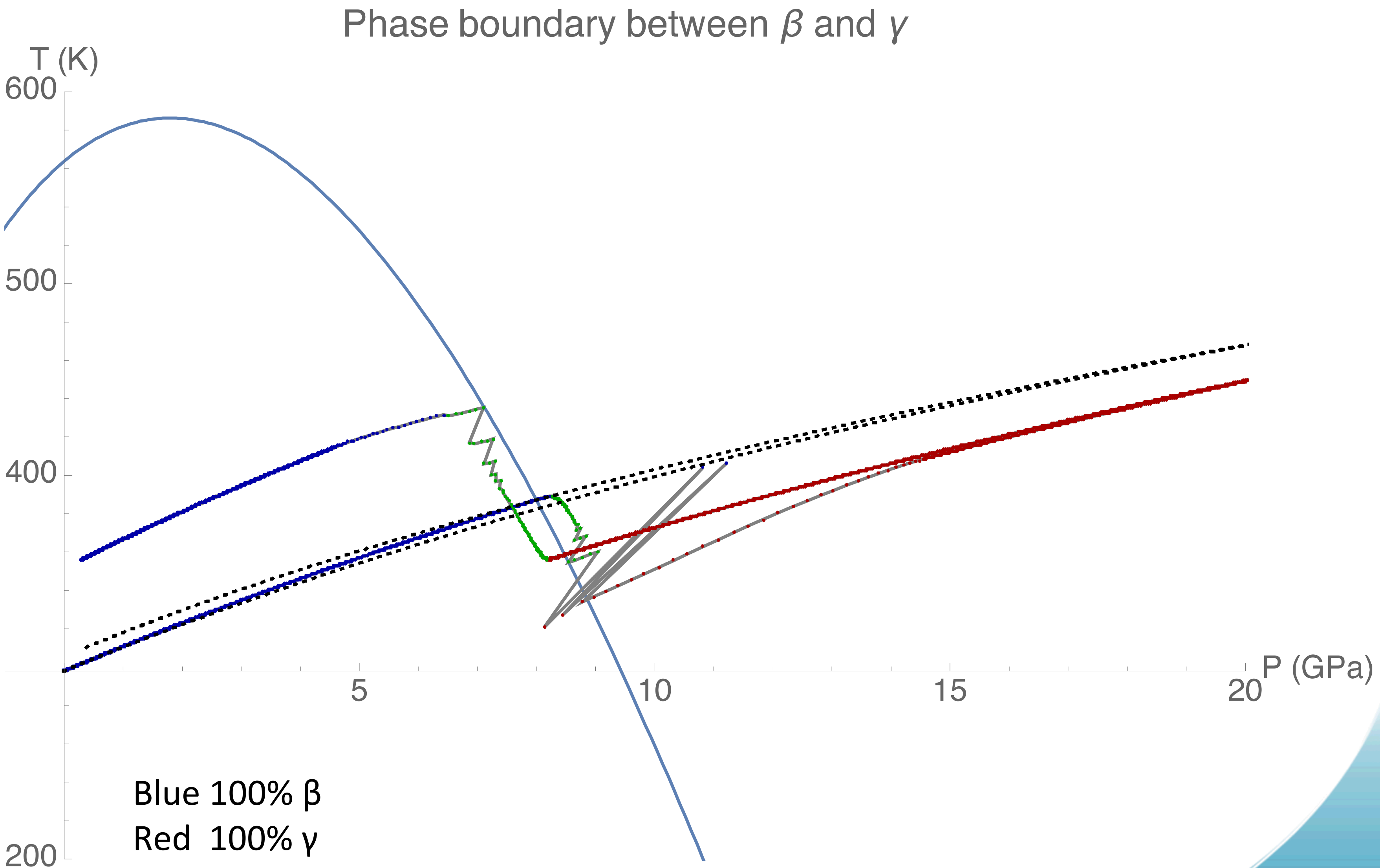


Implementation in Laslo: Sn



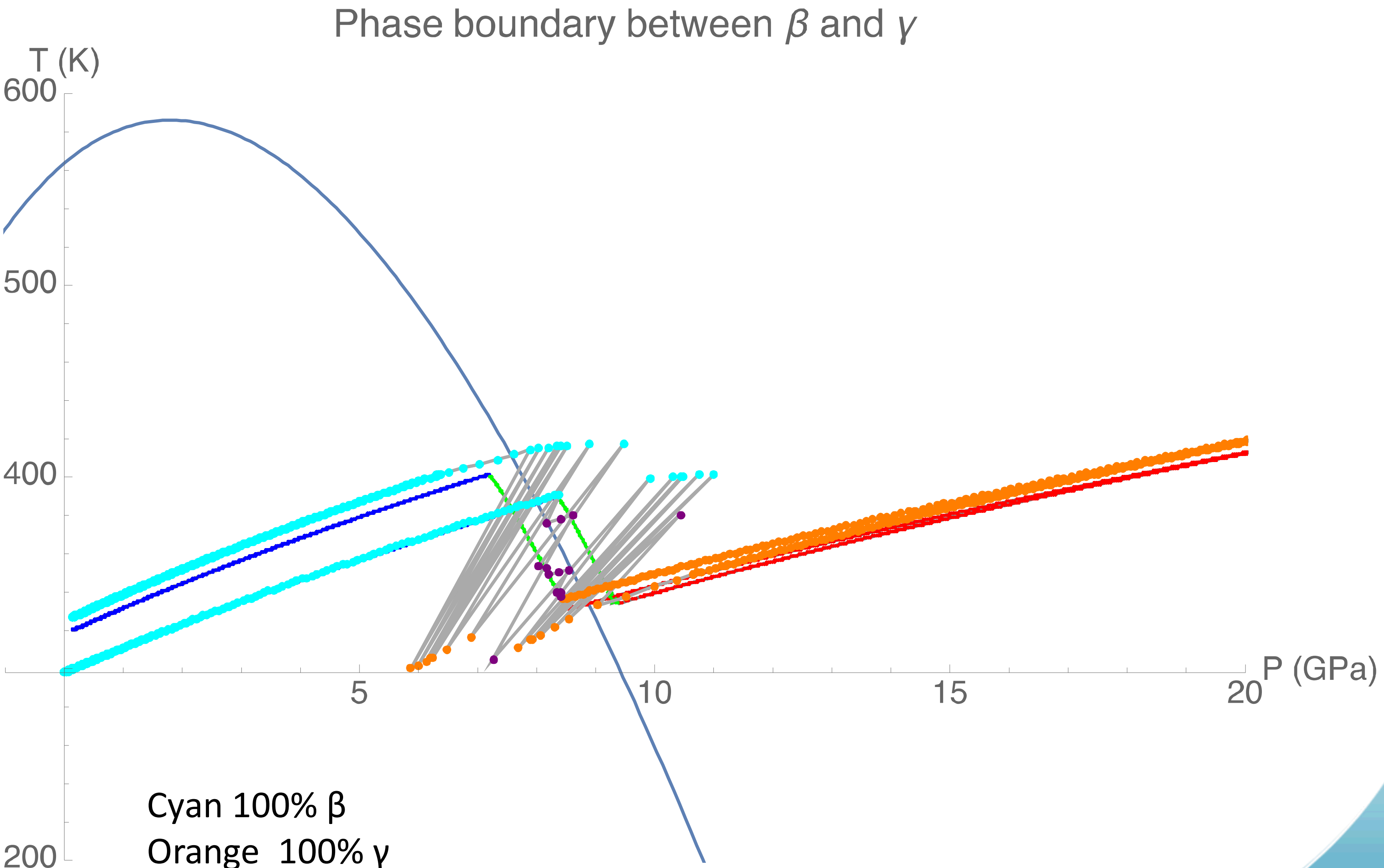
Blue 100% β
Red 100% γ
Green mixed
Dotted MG

Implementation in Laslo: Sn



Blue 100% β
Red 100% γ
Green mixed
Dotted MG

Implementation in Laslo: Sn



Cyan 100% β
Orange 100% γ
Purple mixed

Summary

- We have subroutines for Carl Greeff’s kinetic phase transition model in Lambda.
- We have used them for a good implementation in Laslo.
- We will use them also for an implementation in ALEGRA.

- The subroutines in Lambda can use the extended Vinet and the Mie Grünesien EOSs as phase models.
- We will add the capability of using Utri tables.

- We have several ideas about how to address the “time step problem”.