Density Functional Theory

Cs_2Te calculations

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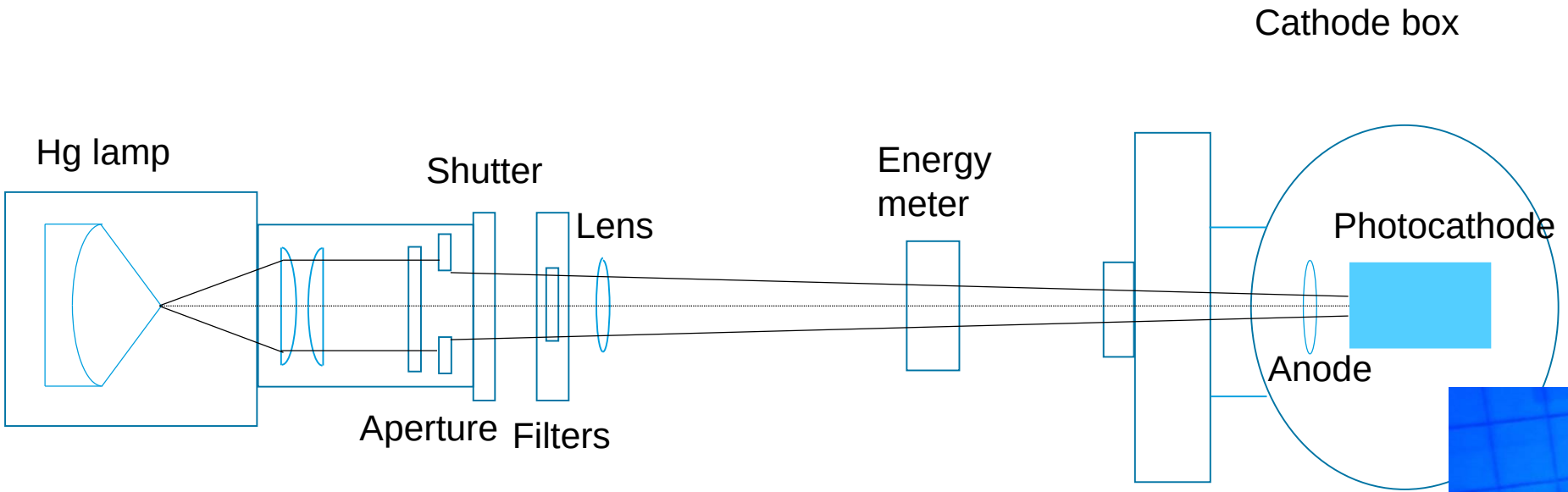
Zeuthen, 22/04/2021

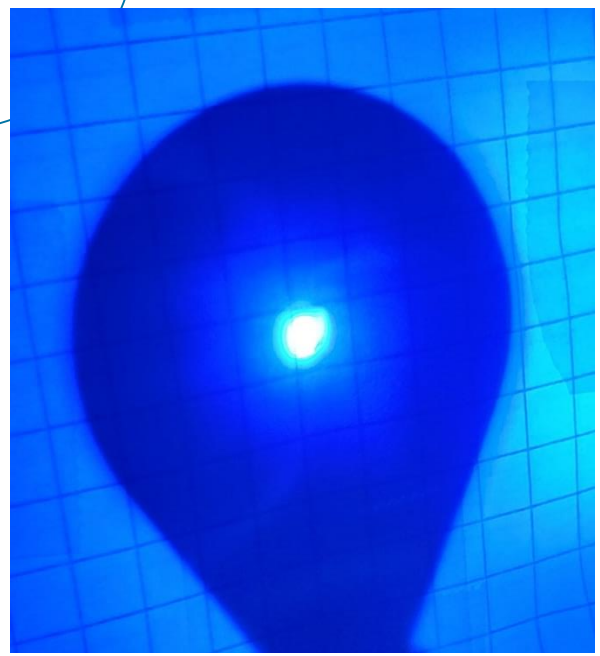
Outline

- Experimental QE measurements
- Density functional theory
  - Finding the minimum energy state $Cs_xTe_{(1-x)}$phase
  - Finding the minimum energy state Cs_2Te crystal structure
  - Band structure calculation Cs_2Te
  - Effective mass calculation Cs_2Te
- Future plans??

Quantum efficiency measurements

The experimental setup 1

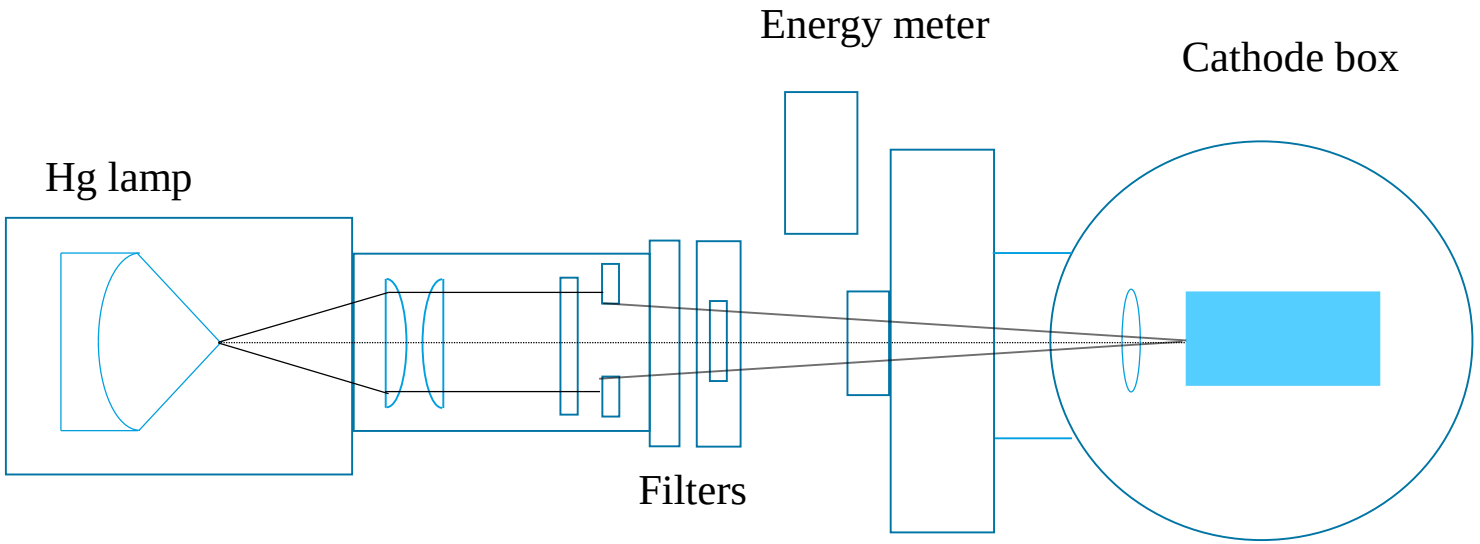




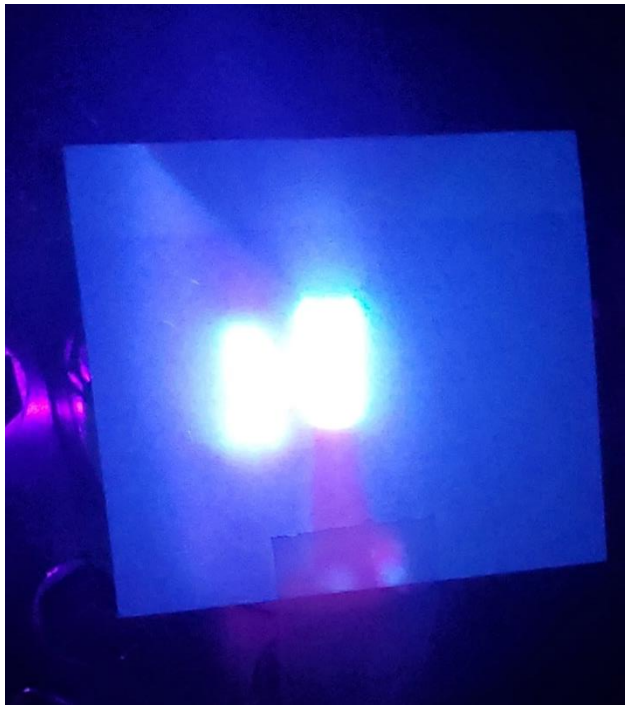
Focused beam on the cathode box window

Quantum efficiency measurements

Experimental setup 2



Inside the tunnel only 40 cm space available near cathode box.



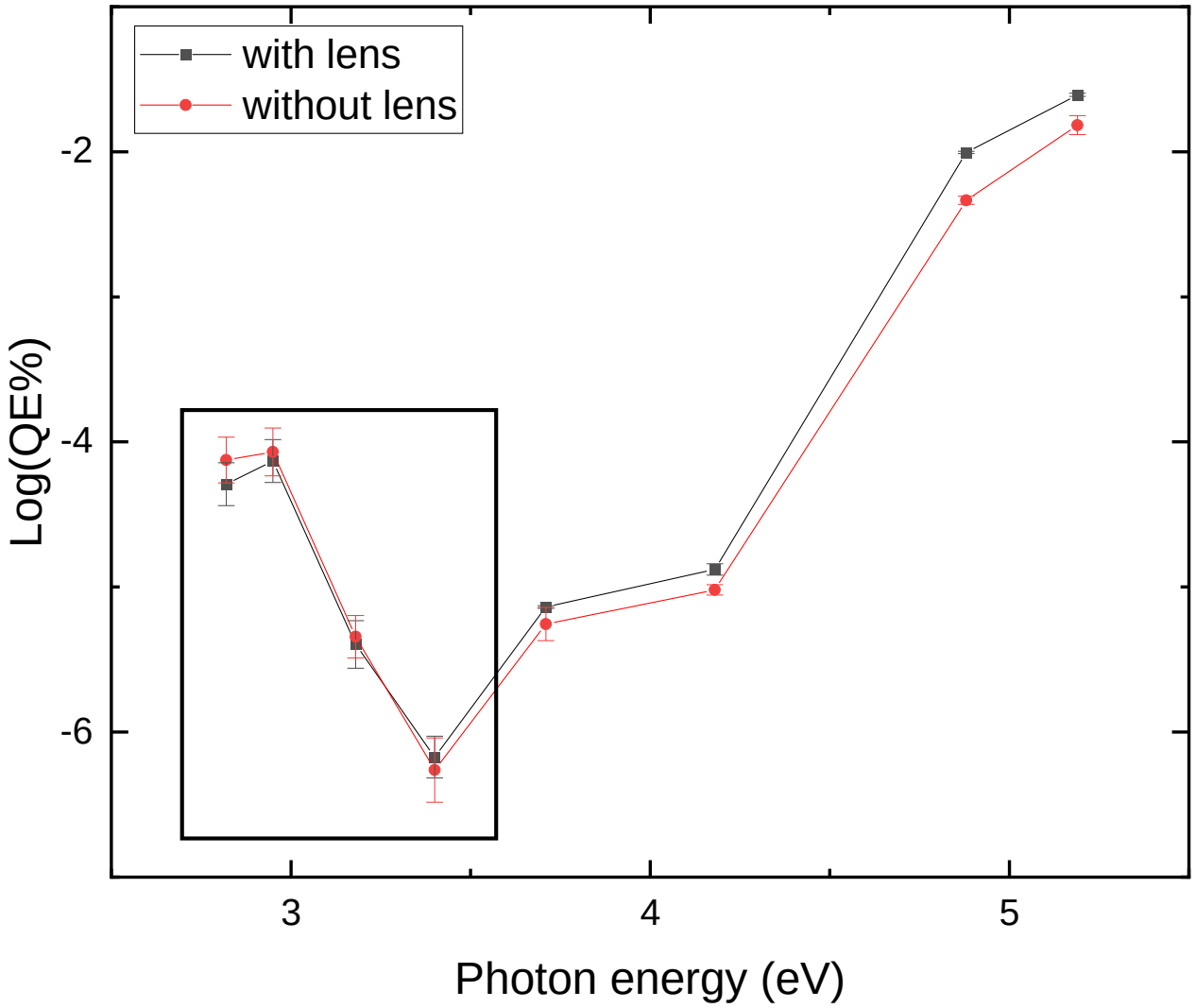
Beam on the cathode box window

QE measurements

17/03/2021

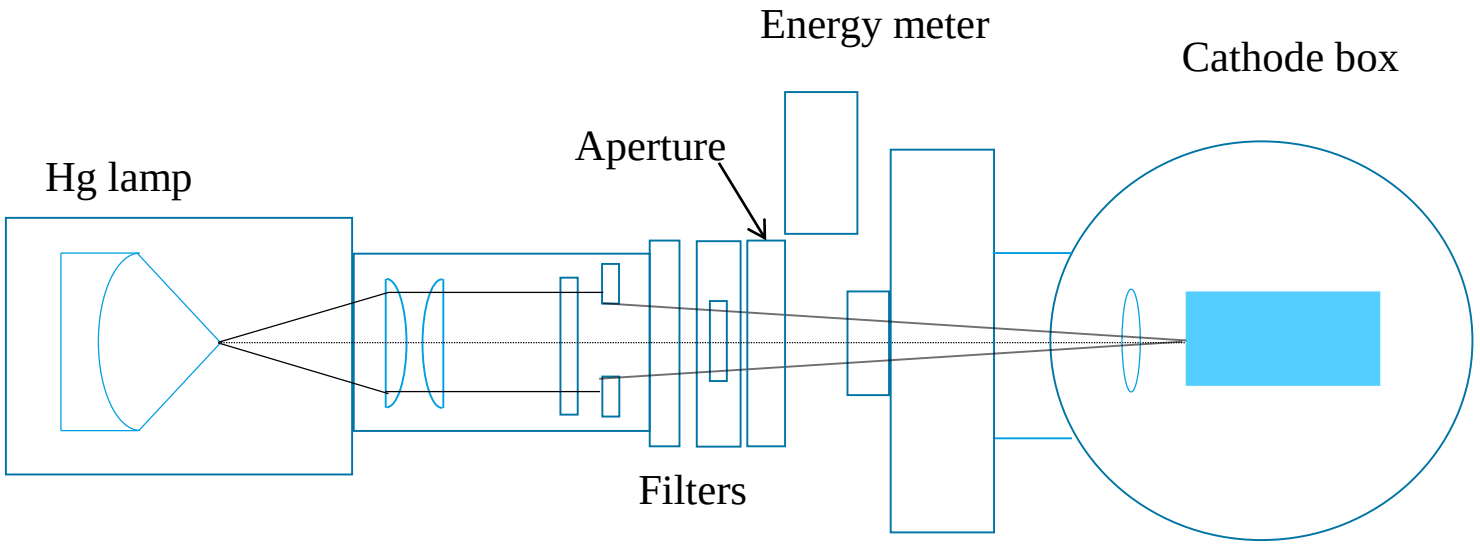
Photon energy (eV)	Current (nA)	Light Power (μW)	QE (%) With the lens	Current (nA)	Light Power (μW)	QE (%) without the lens
5.19	0.0235	0.494	2.4702×10^{-2}	0.021	0.700	1.5270×10^{-2}
4.88	0.0069	0.342	9.8913×10^{-3}	0.00527	0.567	4.6321×10^{-3}
4.18	0.00034	10.8	1.3229×10^{-5}	0.00024	10.4	9.5653×10^{-6}
3.71	0.00017	8.67	7.2843×10^{-6}	0.00012	7.79	5.5536×10^{-6}
3.4	0.00006	30.5	6.7277×10^{-7}	0.00004	24.9	5.4710×10^{-7}
3.18	0.00006	4.89	4.0196×10^{-6}	0.00006	5.596	4.5476×10^{-6}
2.95	0.00006	0.24	7.3966×10^{-5}	0.00007	0.208	8.5377×10^{-5}
2.82	0.00006	0.331	5.1114×10^{-5}	0.00008	0.3004	7.5093×10^{-5}

Comparing experimental setups



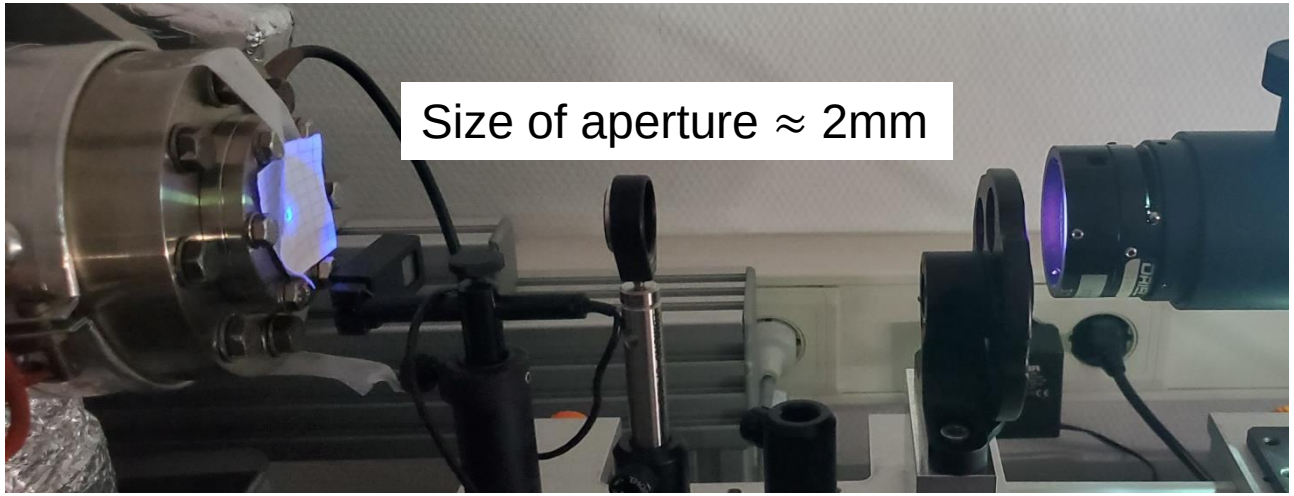
Quantum efficiency measurements

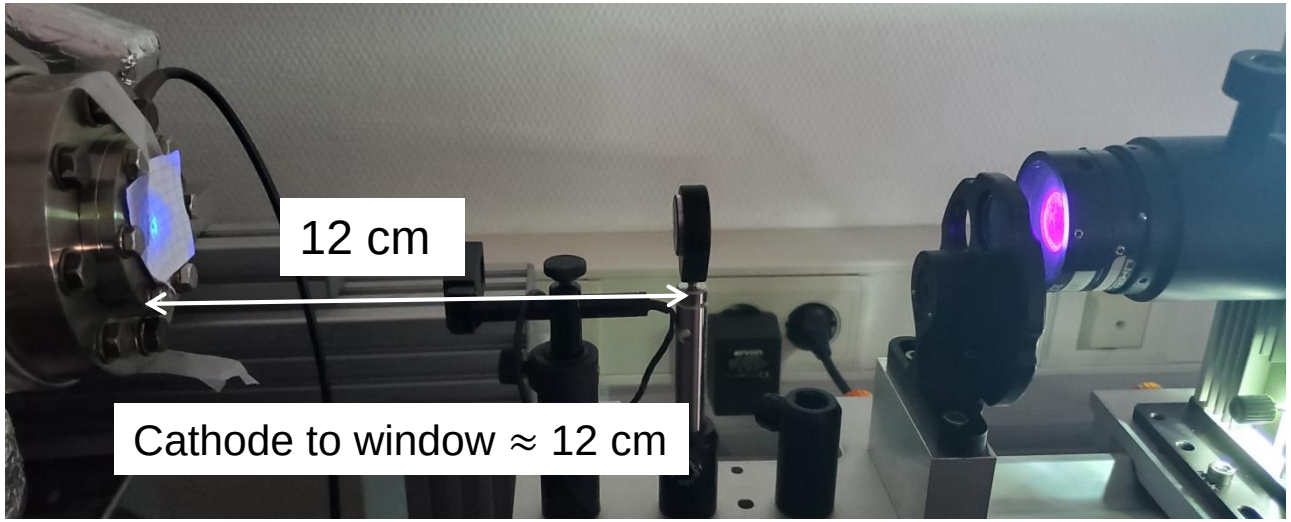
Experimental setup 2



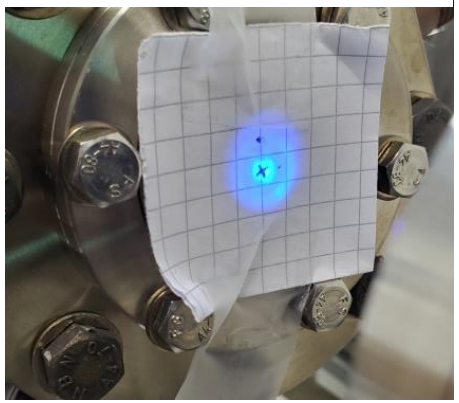
Quantum efficiency measurements

Beam size check

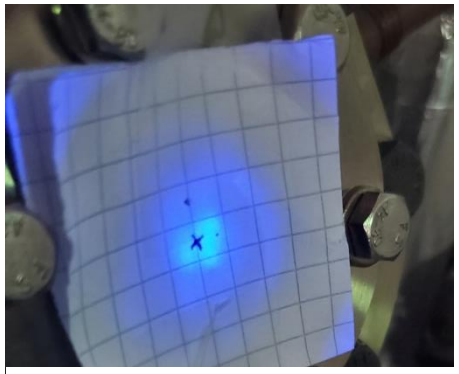




Beam size on the window ≈ 3mm



Beam on the cathode box window



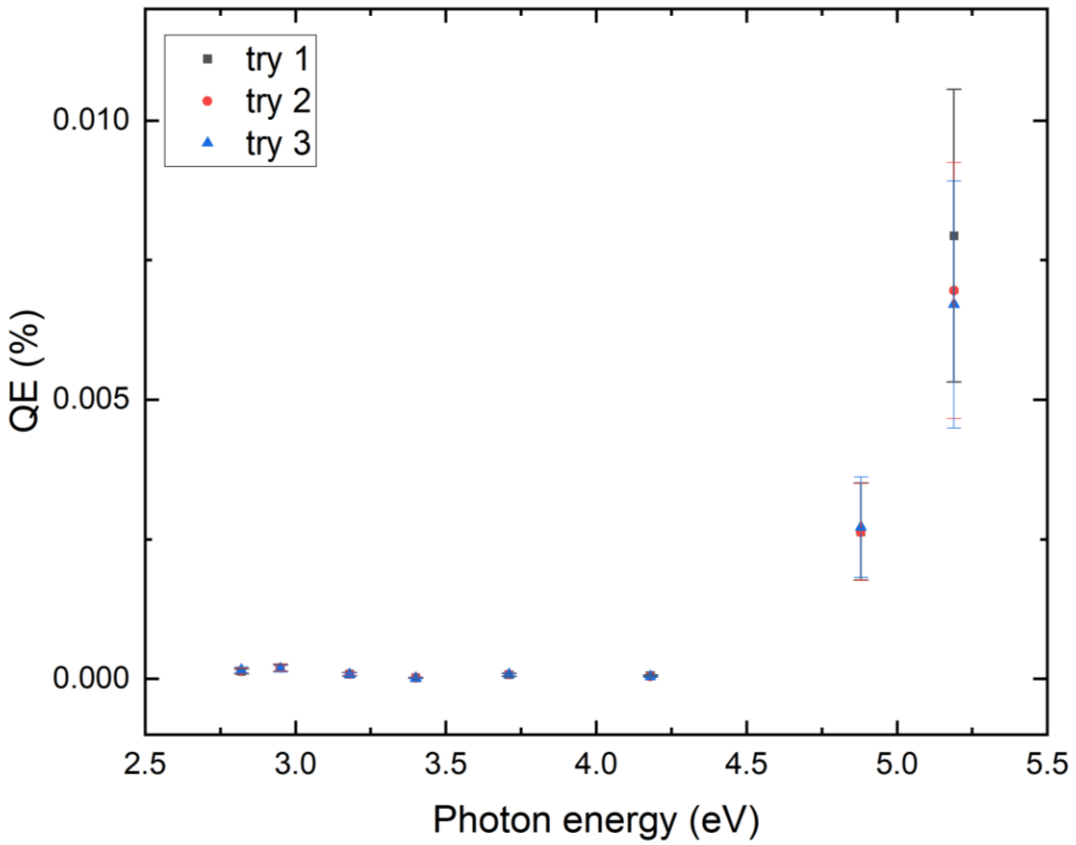
Rough estimation of the beam on the cathode

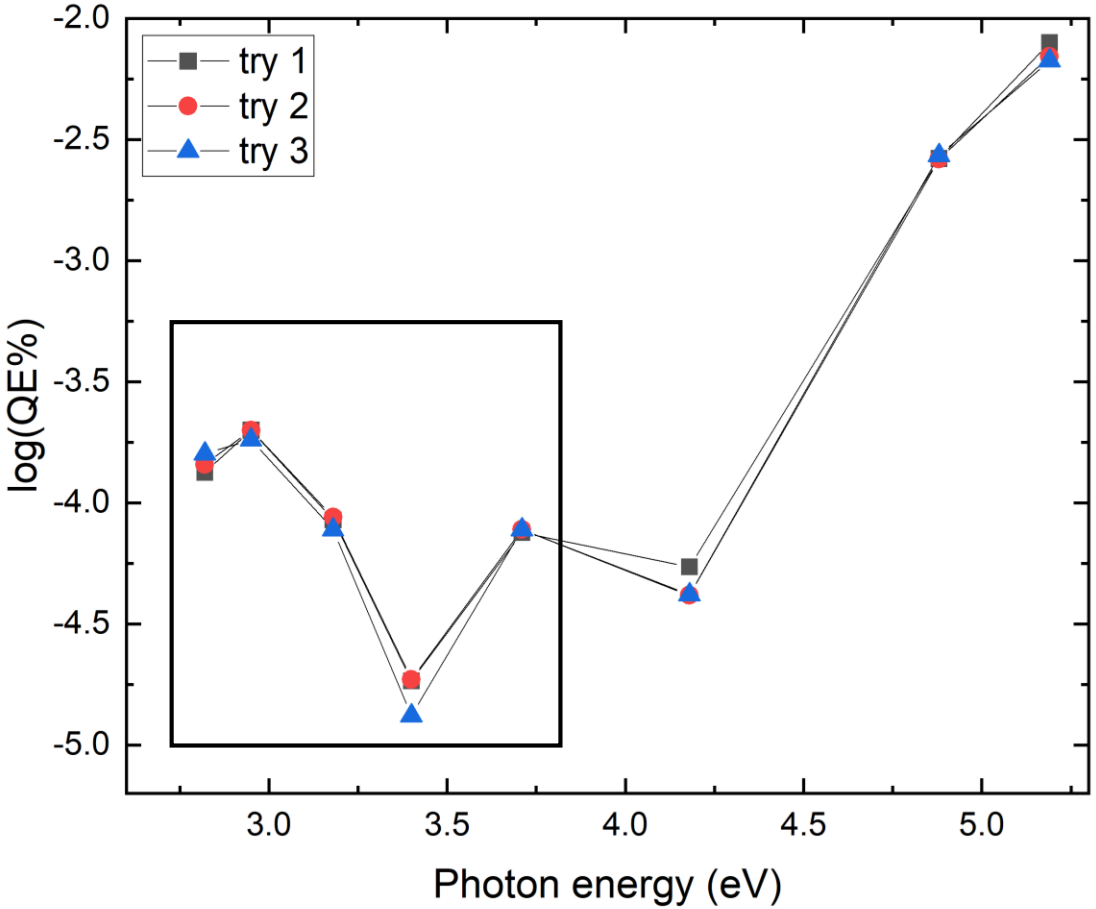
Beam size on the cathode ≈ 5mm

Comparing experimental setups

Photon energy (eV)	QE (%) Measurement 1	error	QE (%) measurement 2	error	QE (%) measurement 3	error
5.19	7.9342E-03	2.618E-3	6.9544E-03	2.295E-3	6.7050E-03	2.213E-3
4.88	2.6382E-03	8.706E-4	2.6289E-03	8.675E-4	2.7204E-03	8.977E-4
4.18	5.4458E-05	1.797E-5	4.1475E-05	1.369E-5	4.1764E-05	1.378E-5
3.71	7.5249E-05	2.483E-5	7.7620E-05	2.561E-5	7.7635E-05	2.562E-5
3.4	1.8385E-05	6.067E-6	1.8636E-05	6.150E-6	1.3232E-05	4.367E-6
3.18	8.5076E-05	2.808E-5	8.7425E-05	2.885E-5	7.7629E-05	2.562E-5
2.95	1.9958E-04	6.400E-5	1.9938E-04	6.419E-5	1.8196E-04	5.934E-5
2.82	1.3371E-04	4.412E-5	1.4412E-04	4.756E-5	1.5946E-04	5.262E-5

Comparing experimental setups





Density Functional Theory

Introduction

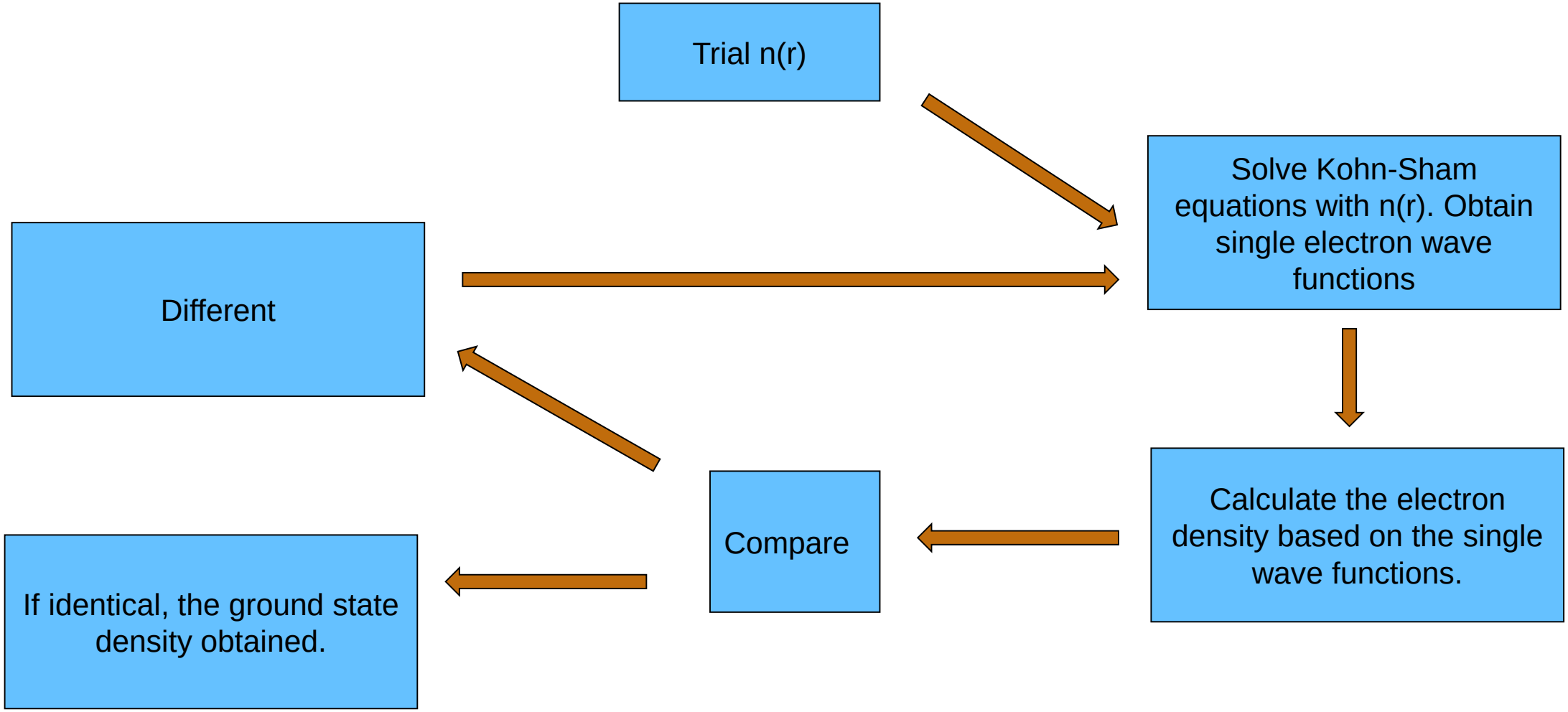
- To solve many body problems by Schrödinger’s equation.
- Only up to one electron problem we can solve Schrödinger's equation exactly.
- It is very hectic to solve the Schrödinger's equation for a N- body system.
- We must involve some approximation to solve the problem a method to obtain an approximate solution to the Schrödinger's equation of a many body system is DFT.
- DFT is a computational quantum mechanical modelling method used in physics ,chemistry, & material science to investigate the electronic structure (ground state) of many body systems .
- Using this theory the properties of many- electron system can be determined by using FUNCTIONALS.
- In DFT instead of considering wave function we considered density functional . DFT : work in terms of density

$$E=E[n(r)]$$

$$\varphi^2=n(r)$$

Density Functional Theory

Self- Consistency Scheme



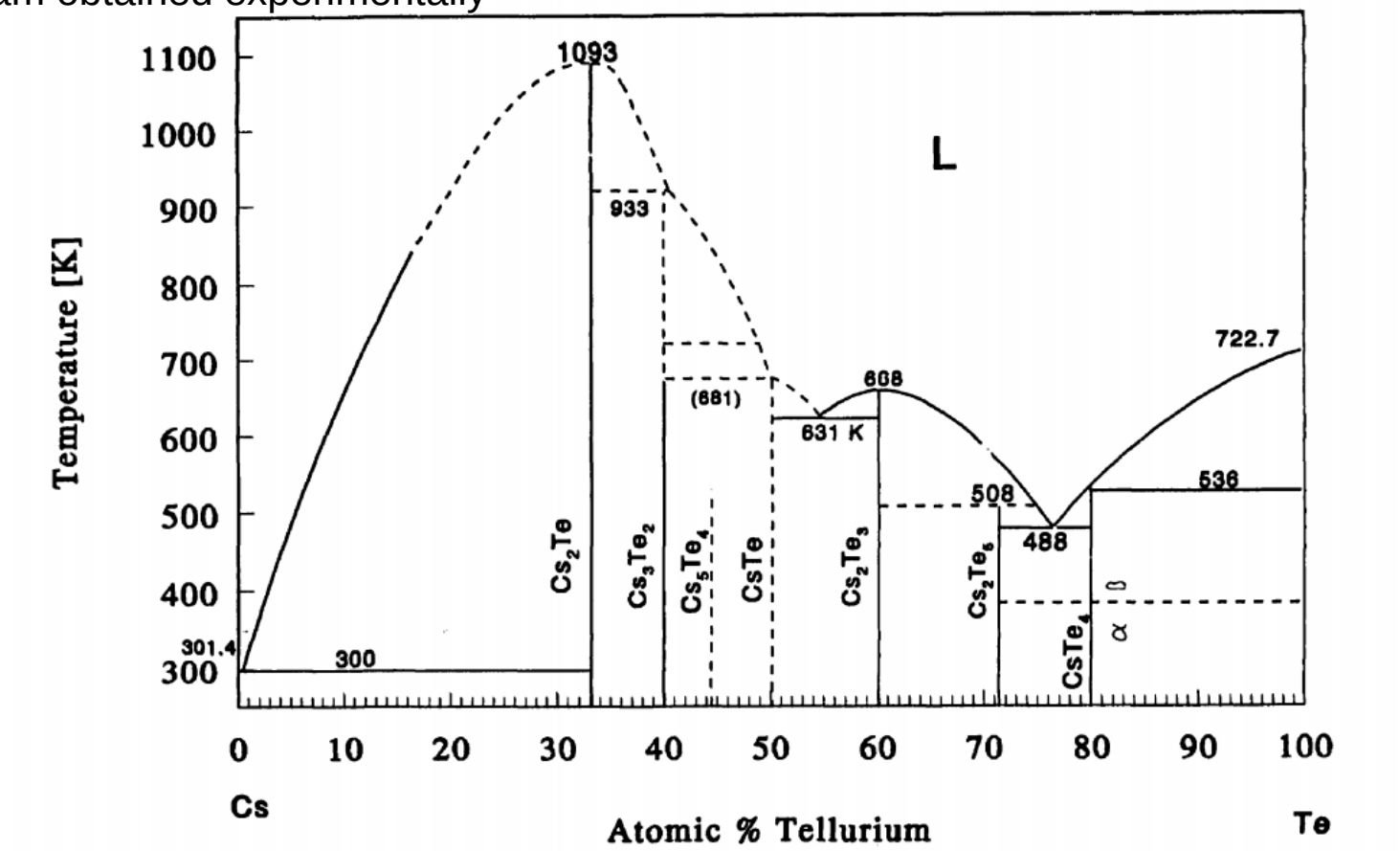
Density Functional Theory

- The PWscf code of the Quantum ESPRESSO suite
- Ultra-soft pseudopotentials (USPP) with the generalized gradient approximation (GGA)
- Relativistic, spin-orbit coupling, approximations are included for the 4d, 5d and 6p transition metals
- The convergence tests performed to achieve minimum total energy configuration.
- Formation energy is used instead of total energy to find the minimum energy state for different phases

$$E_f = \sum E_{Cs_xTe_{1-x}} - (E_{Cs} \times no\ of\ Cs\ atoms) - (E_{Te} \times no\ of\ Te\ atoms)$$

Cs-Te crystallization phases

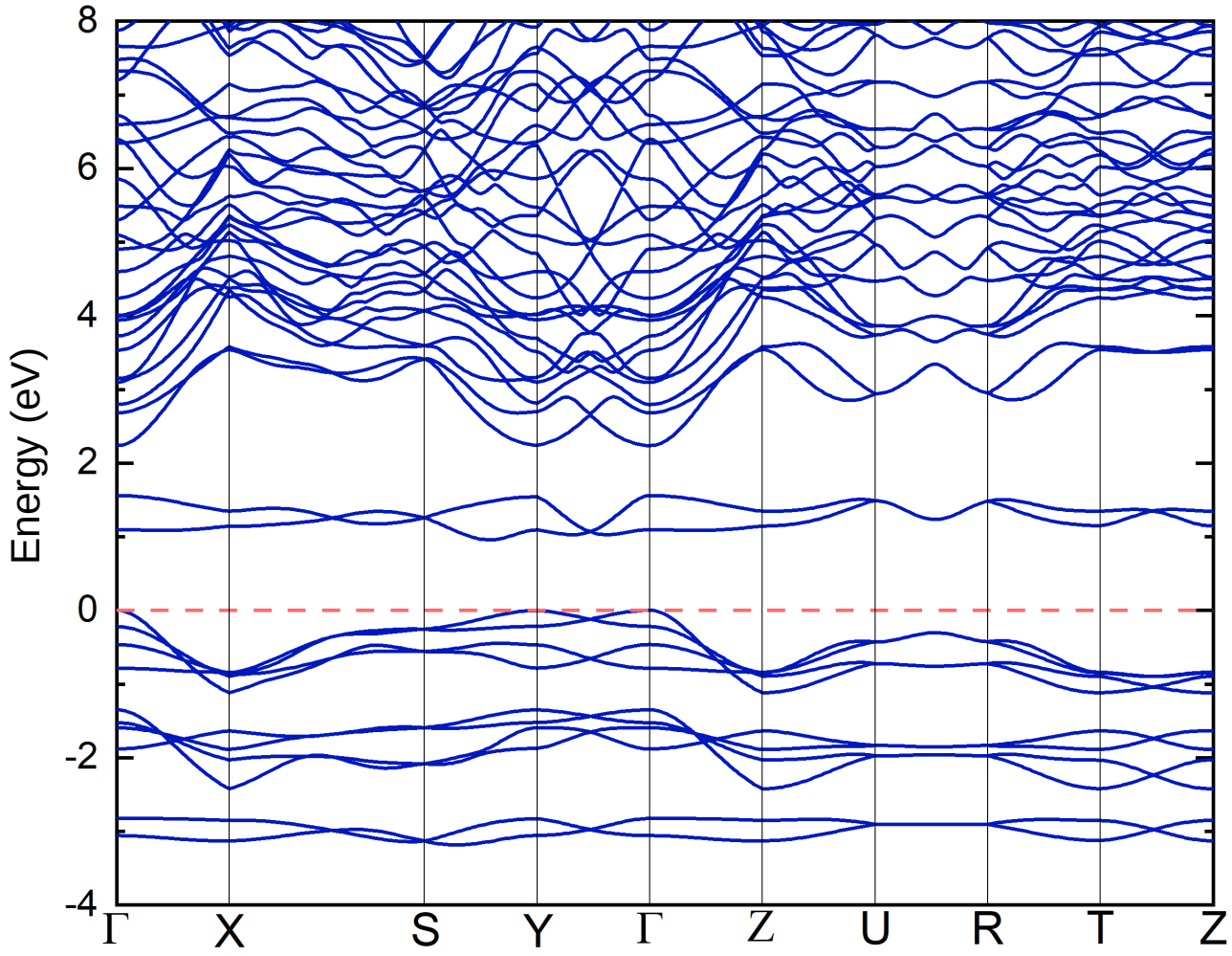
Phase diagram obtained experimentally

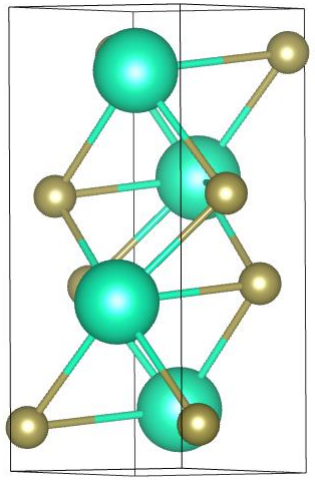


CsTe	Cs_2Te_3	Cs_2Te_5	Cs_2Te	$CsTe_4$
Orthorhombic pbam(55)	Orthorhombic cmc21(36)	Orthorhombic cmcm(63)	Orthorhombic pnma(62)	Orthorhombic pbam(55)

Possible Phases Cs_xTe_{1-x}

CsTe

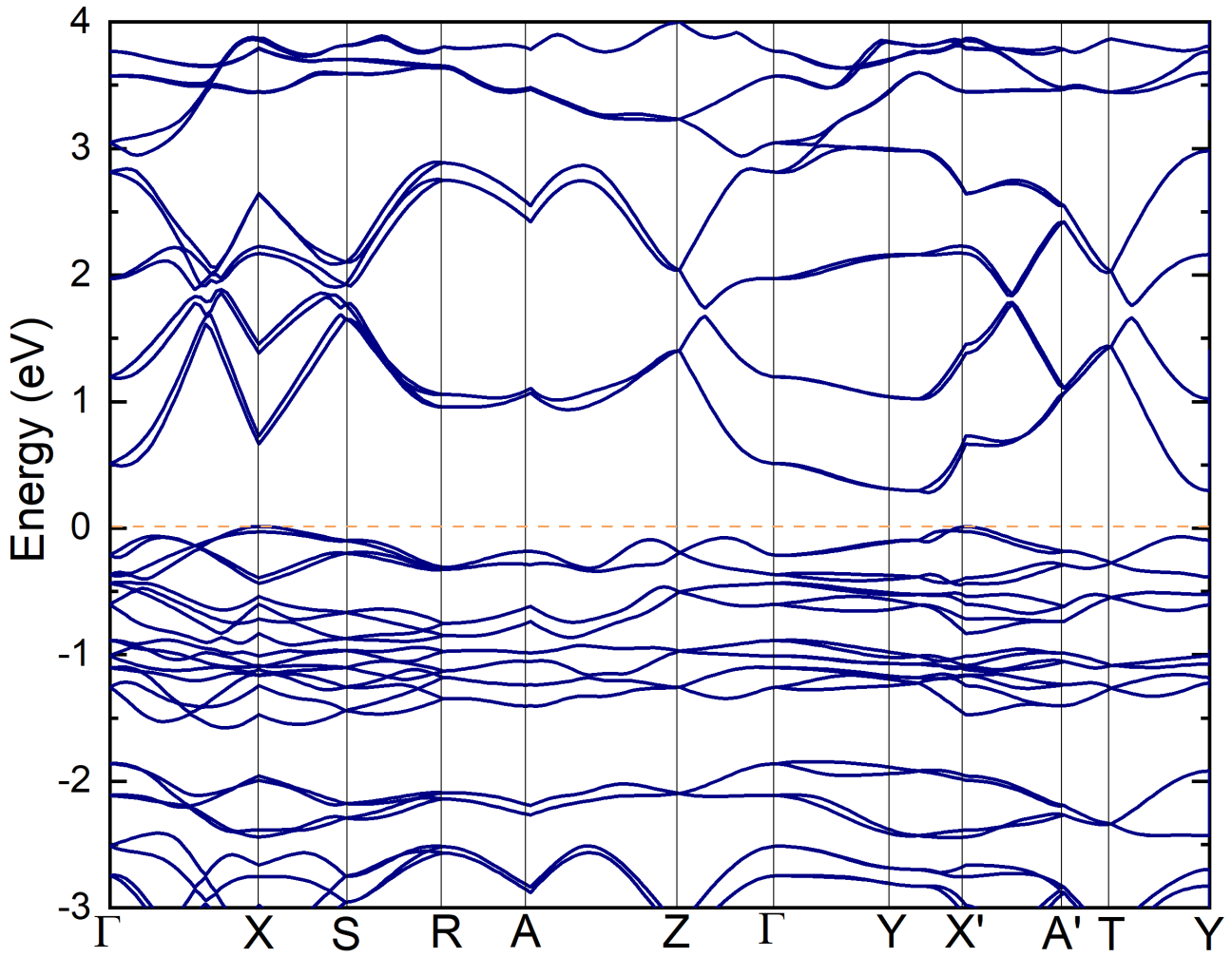


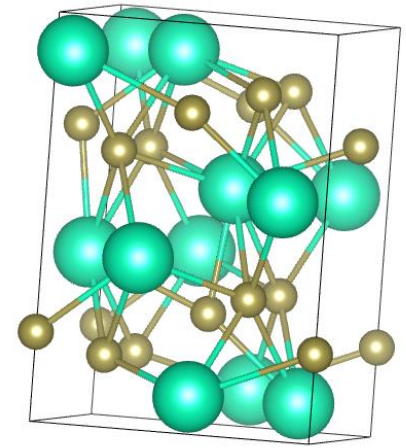


Orthorhombic crystal structure
Pbam (55)
a = 5.049 Å
b = 6.287 Å
c = 11.847 Å

Possible Phases Cs - Te

Cs_2Te_3

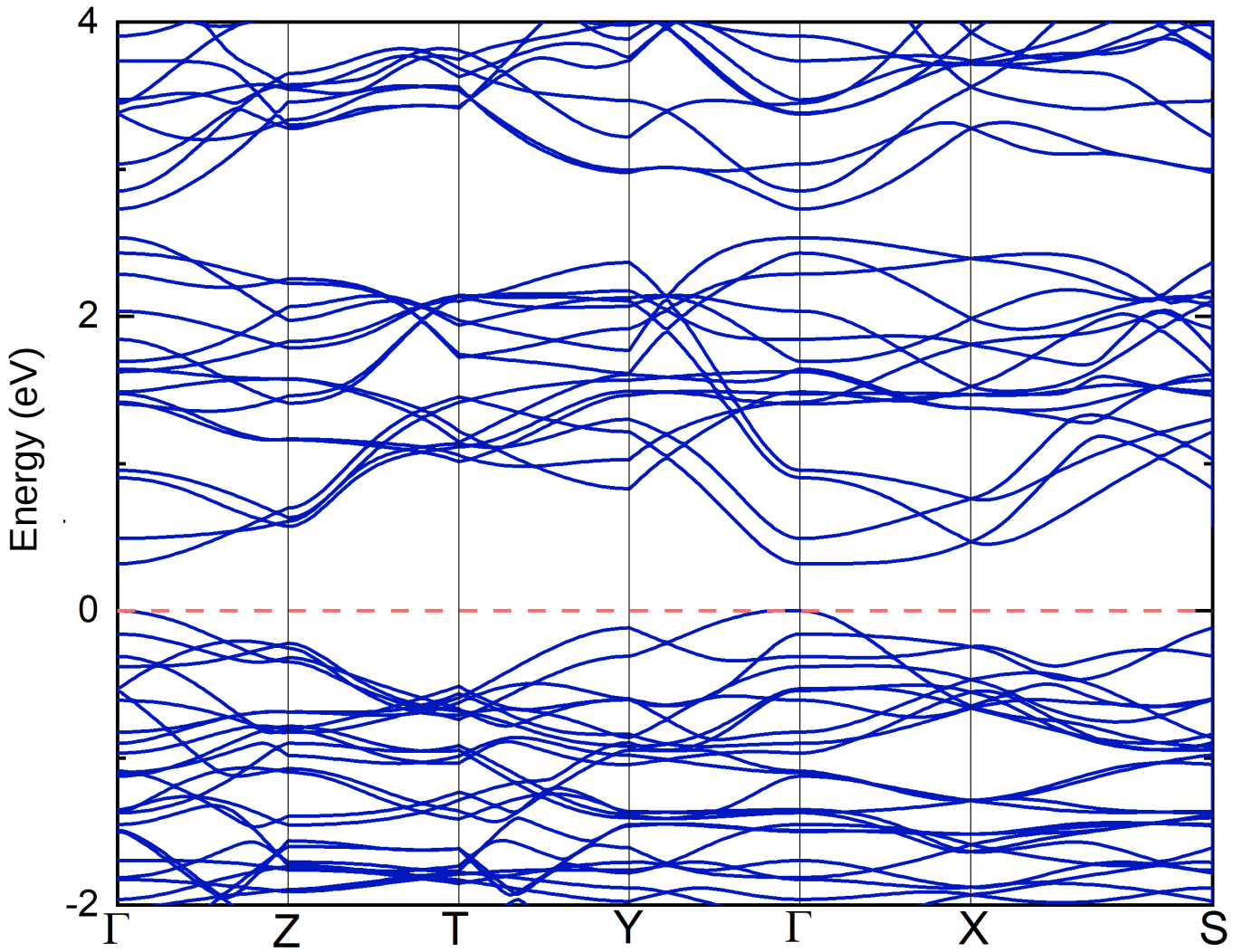


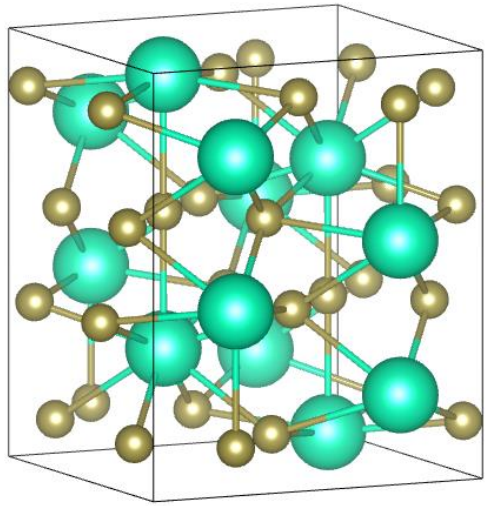


Orthorhombic crystal structure
Cmc21 (36)
a = 8.689 Å
b = 12.452 Å
c = 8.7047 Å

Possible Phases Cs - Te

Cs_2Te_5

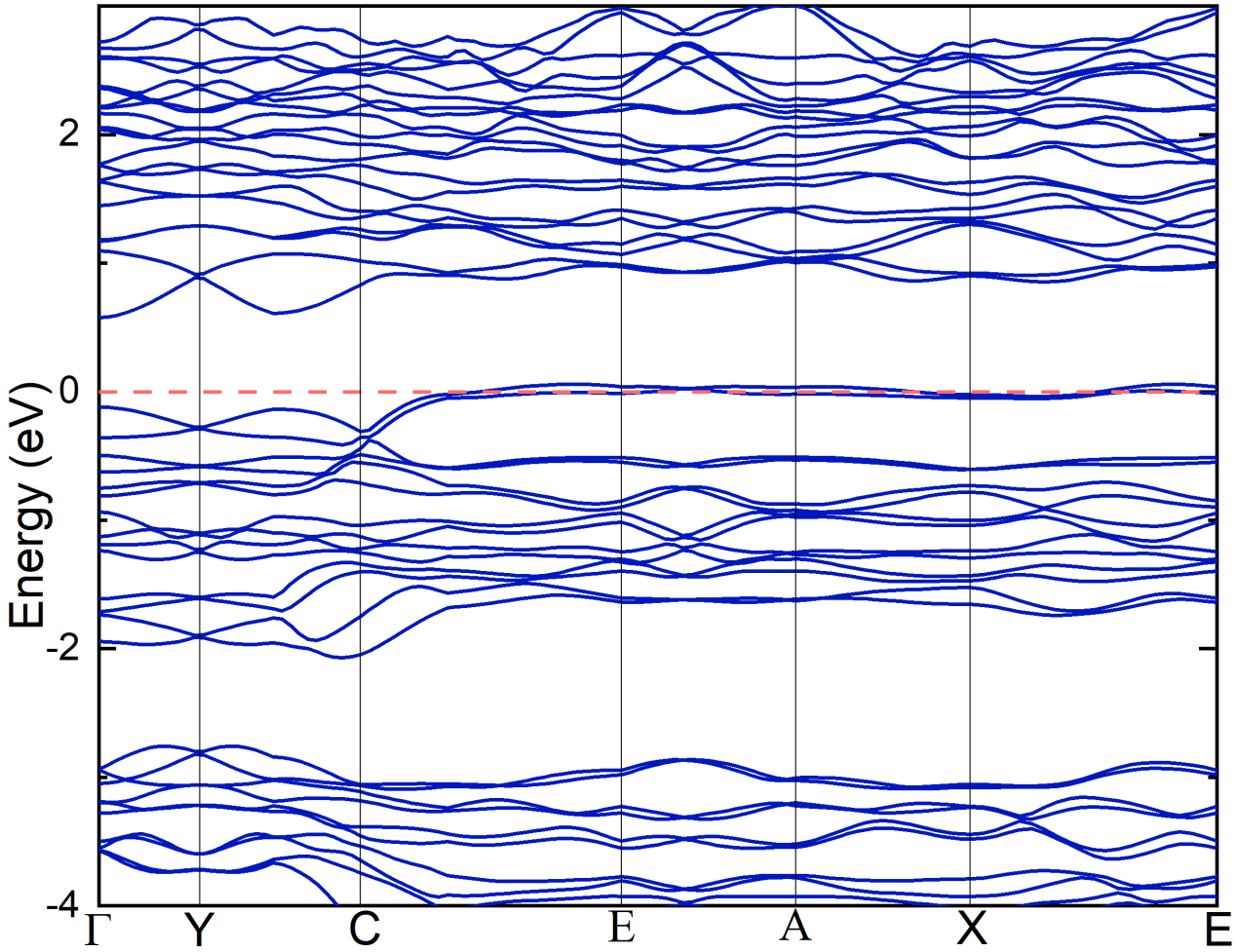


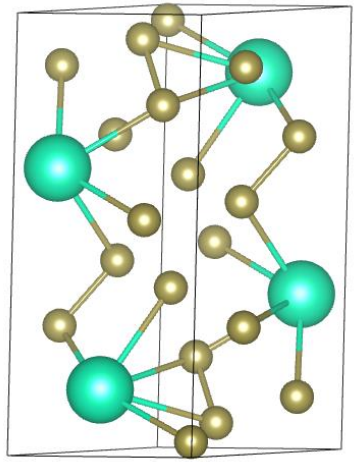


Orthorhombic crystal structure
Cmcm (63)
a = 9.657 Å
b = 12.324 Å
c = 10.340 Å

Possible Phases Cs - Te

$CsTe_4$

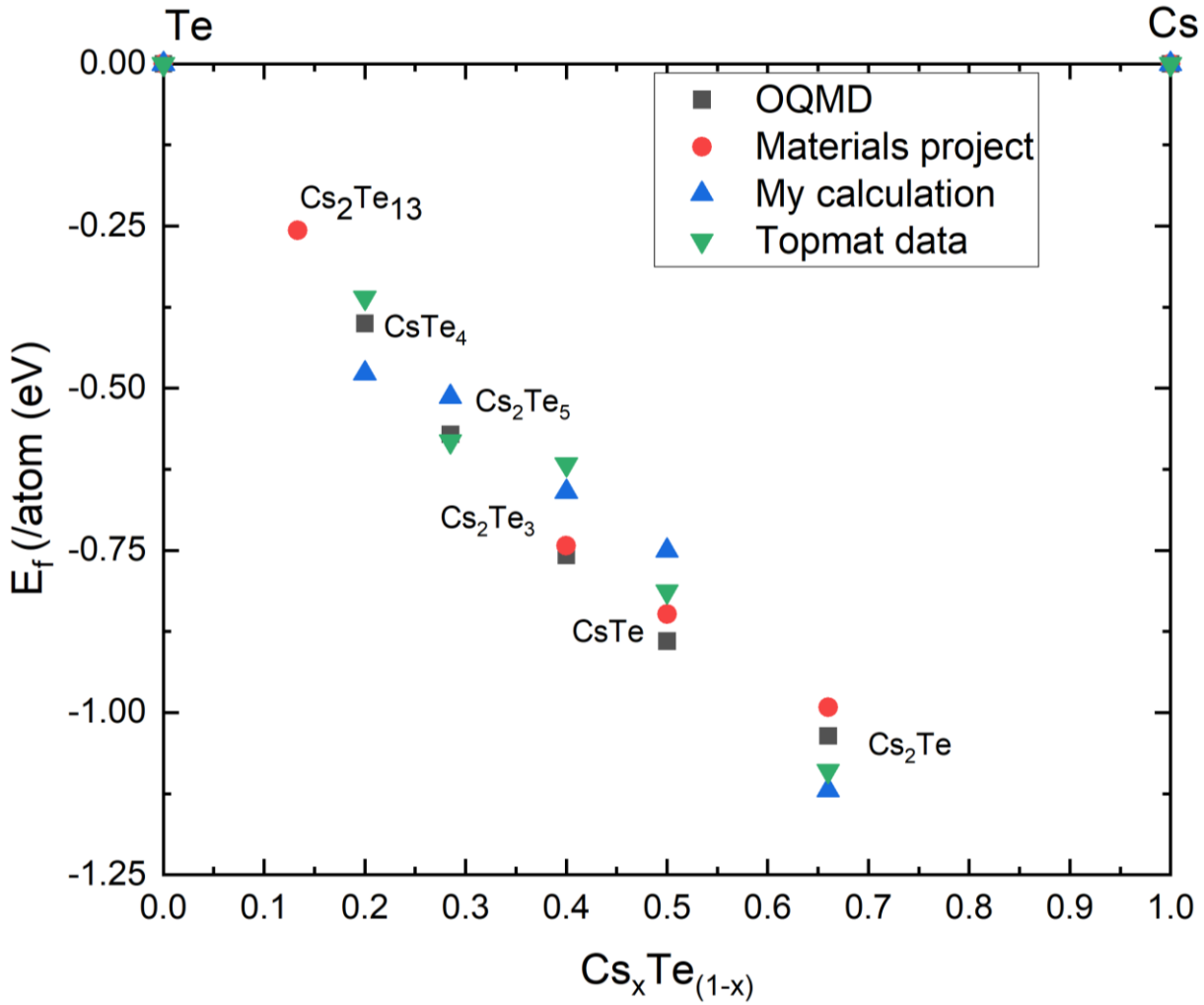




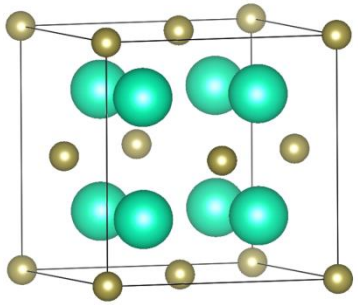
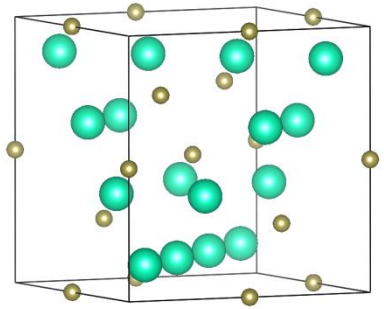
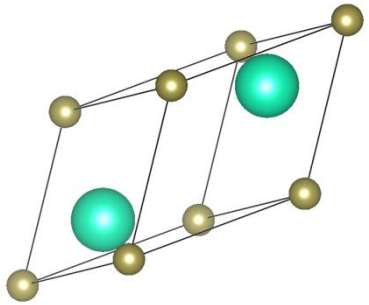
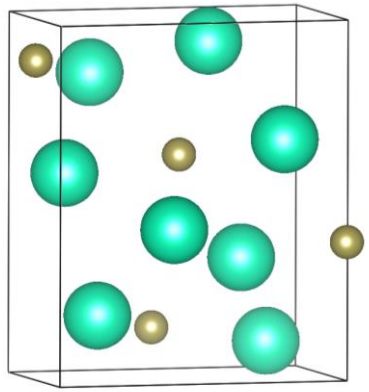
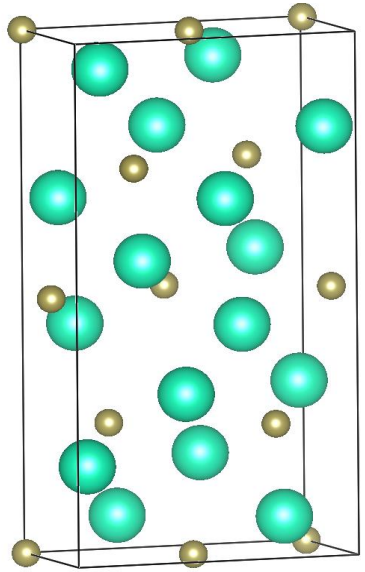
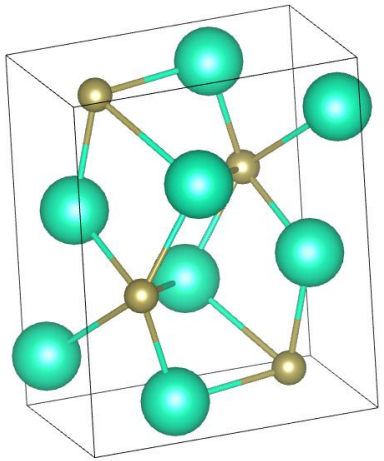
Orthorhombic crystal structure
Pbam (55)
a = 8.140 Å
b = 7.354 Å
c = 14.229 Å

Possible Phases Cs - Te

Cs_xTe_{1-x}

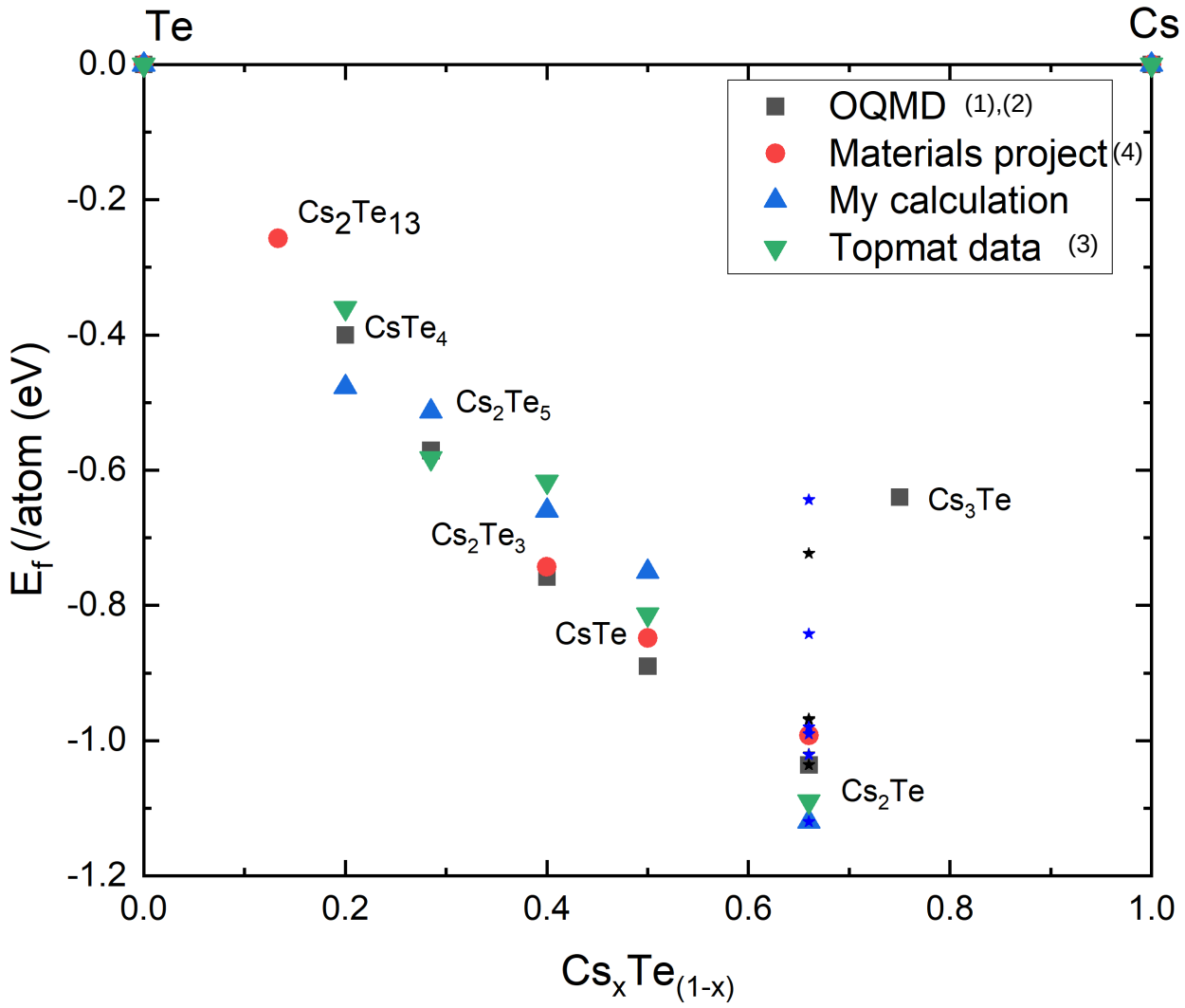


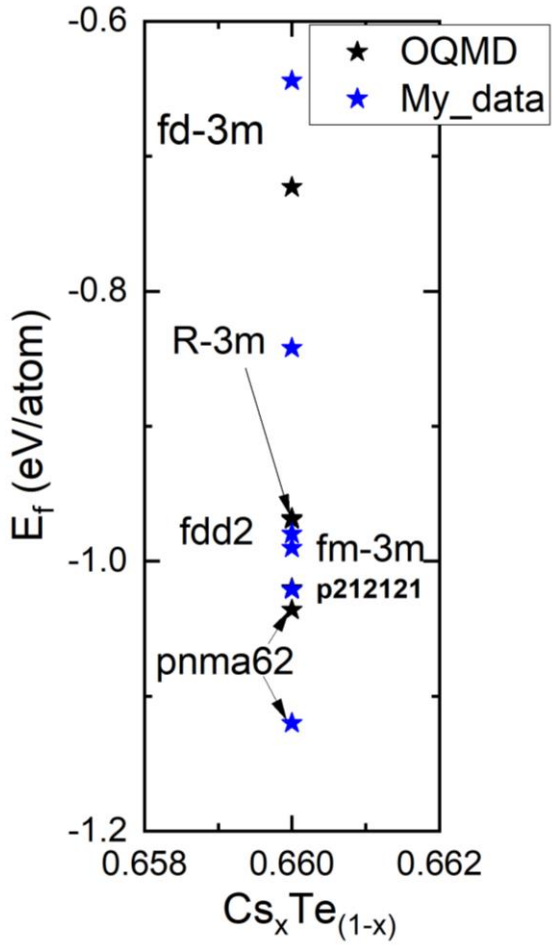
Cs_2Te - Phases

Cubic Fm-3m (225)	Cubic Fd-3m (227)	Trigonal R-3m (166)	Orthorhombic P212121 (19)	Orthorhombic Fdd2 (13)	Orthorhombic Pnma (62)
					
-1.036	-0.723	-0.968	-1.020	-0.969	-1.036
-0.990	-0.644	-0.842	-1.021	-0.979	-1.119

Possible Phases Cs - Te

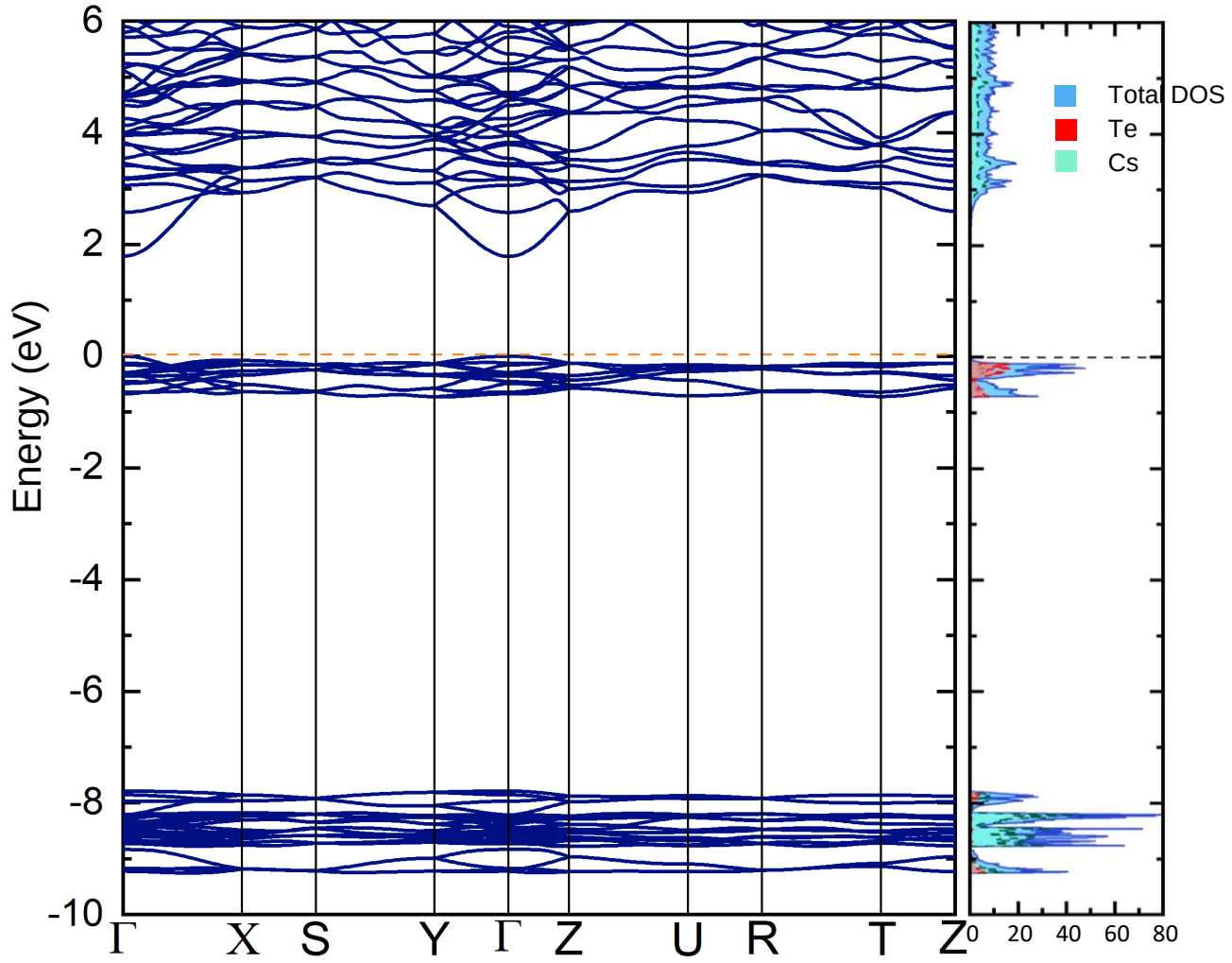
Cs_xTe_{1-x}

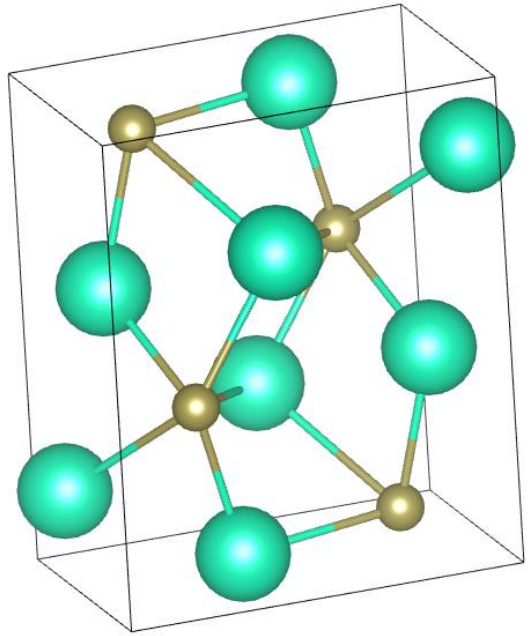




Orthorhombic crystal structure

Space group pnma (62) - Most common structure

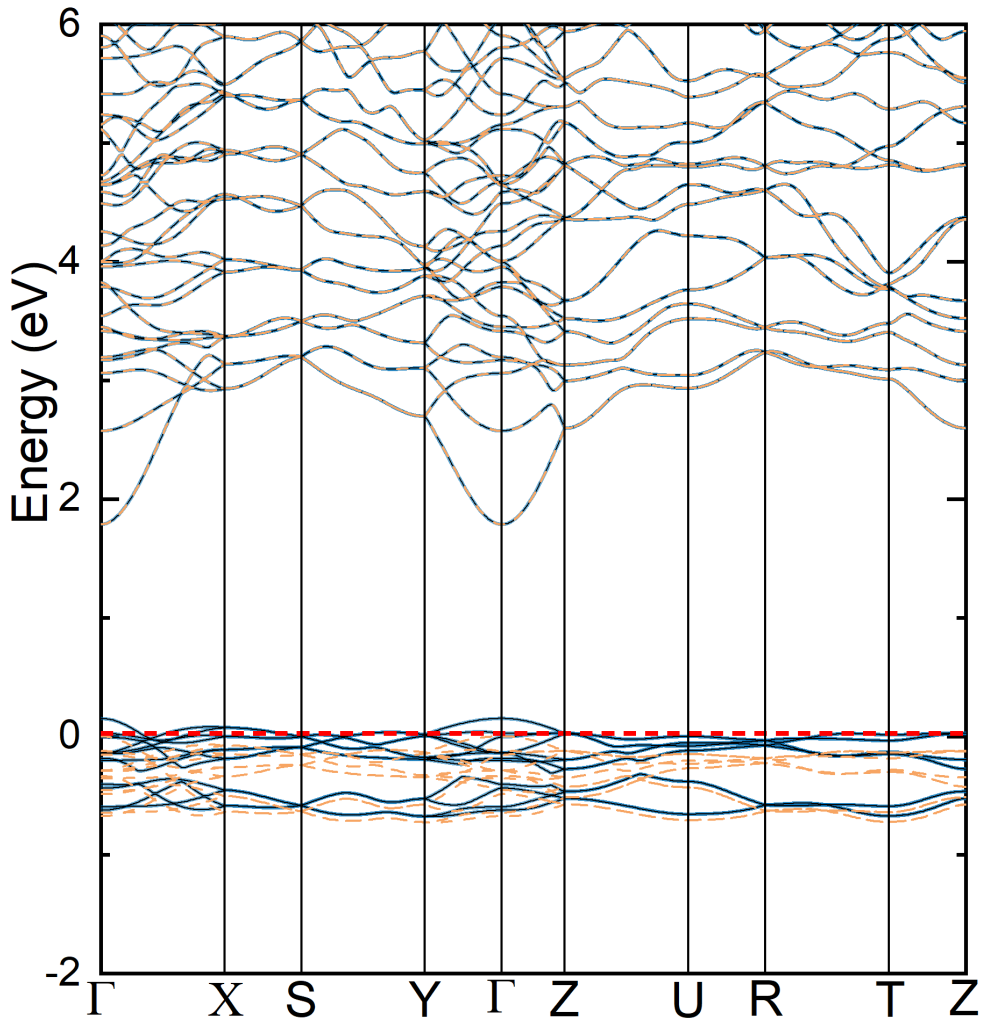




	DFT	Ref (6)	EXP (5)
a (Å)	5.844	5.845	5.871
b (Å)	9.415	9.542	9.109
c (Å)	11.663	11.591	11.474

Cs_2Te band structure

With relativistic and spin orbit coupling



Method	E_{gap}	E_{gap}+SOC (5)	Experimental E_{gap} (7)
PBE	1.78	1.63	
reference	1.76	1.58	3.3

Effective mass calculation

Conduction bands

- I used two different methods to calculate effective mass for different bands.
- Old method uses a parabolic fit to the band and then calculate the effective mass using the curvature of the fit.
- The effective mass at the Fermi energy using

$$m^* = \hbar^2 \left({d^2E} \Big/ {dk^2|_{k=k_f}} \right)^{-1}$$

- New method takes point by point double derivative to calculate the effective mass.

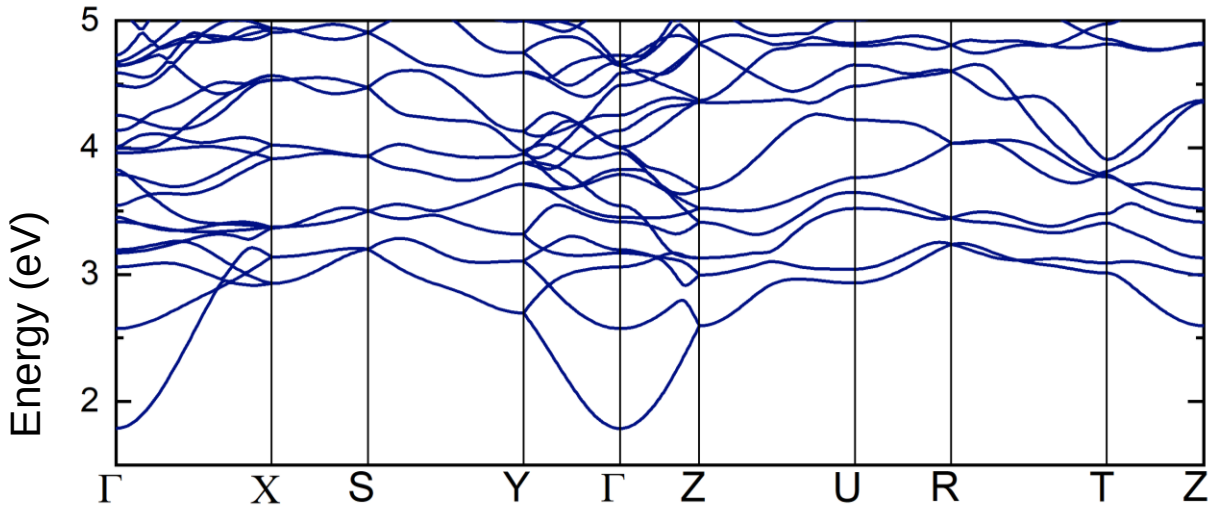
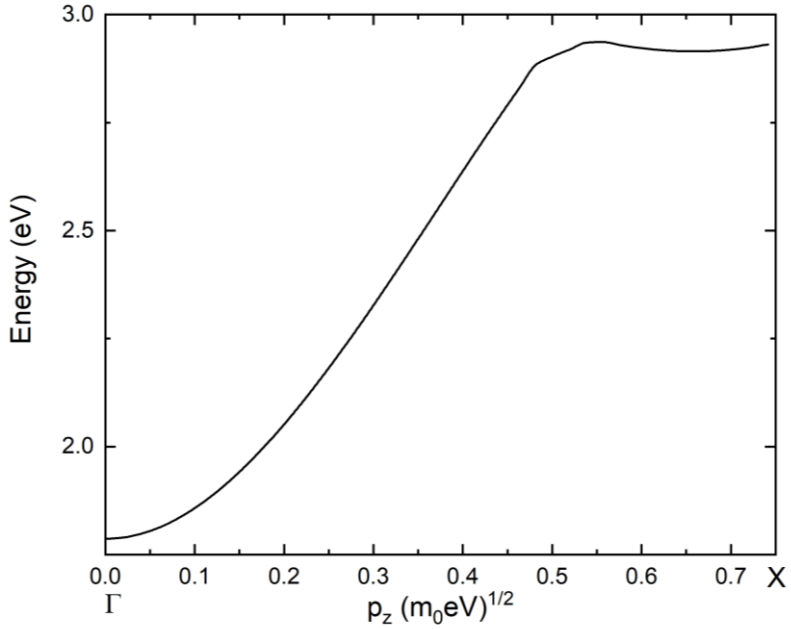


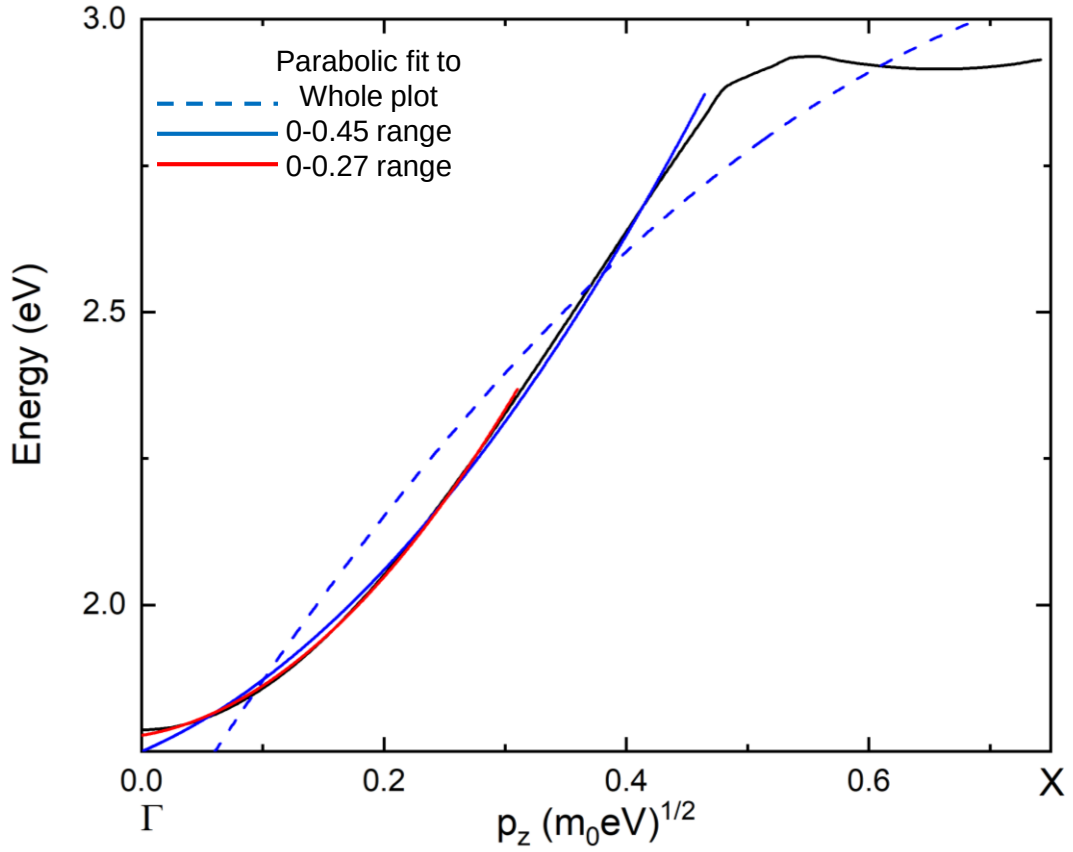
Figure represents the zoomed conduction band plot.

Effective mass calculation

ΓX *direction example*



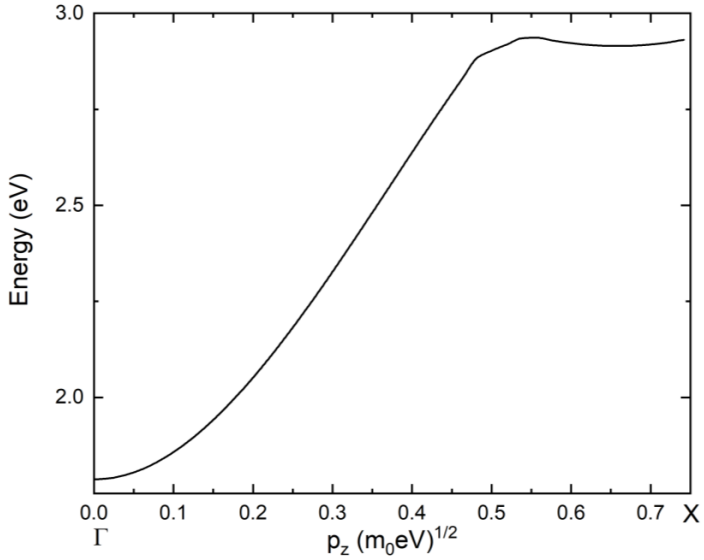
Parabolic fits



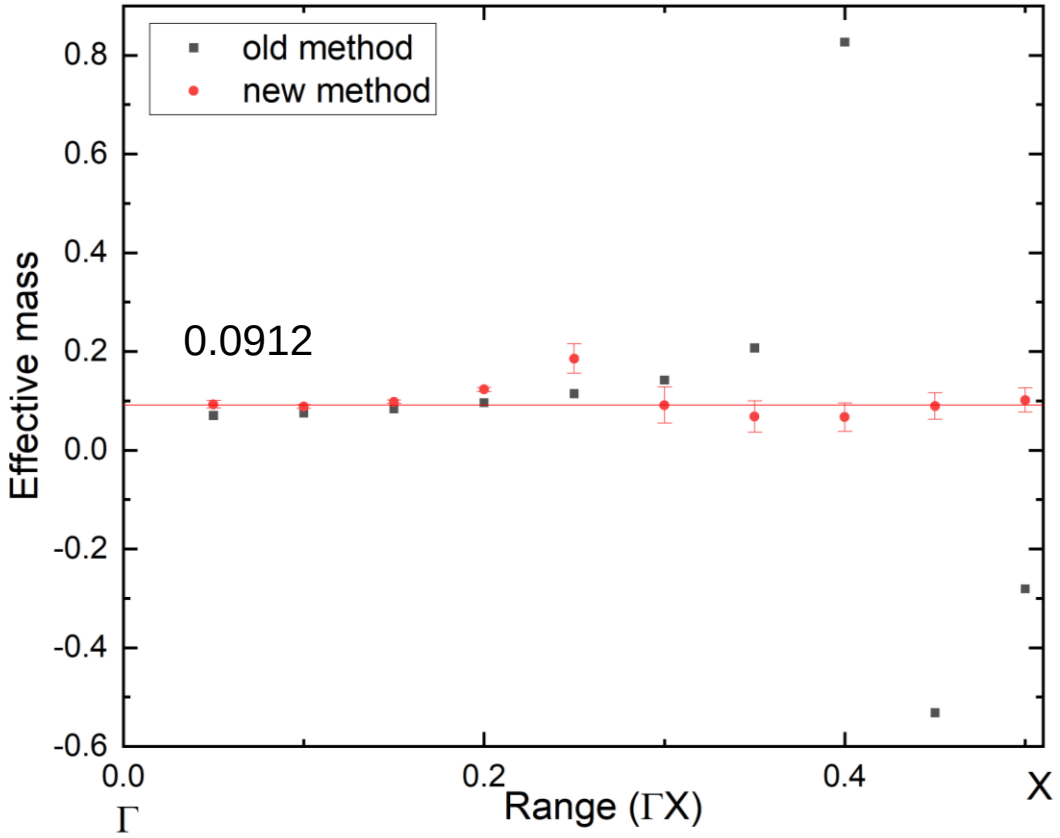
This figure represents the lowest conduction band in the ΓX direction. All three parabolic fits for different regions are not good fits to the band.

Effective mass calculation

ΓX *direction as an example*



- Above figure represents the minimum conduction band in the $\boldsymbol{\Gamma X}$.
- Since it is not a perfect parabolic band, I consider small k-point value regions starting from 0 such as (0-0.05), (0-0.1), (0-0.15) … and finally full range (0-0.5).
- I used a parabolic fit and point by point derivative method to obtain effective mass for each region.
- The right figure represents the obtained effective mass plot against the range.

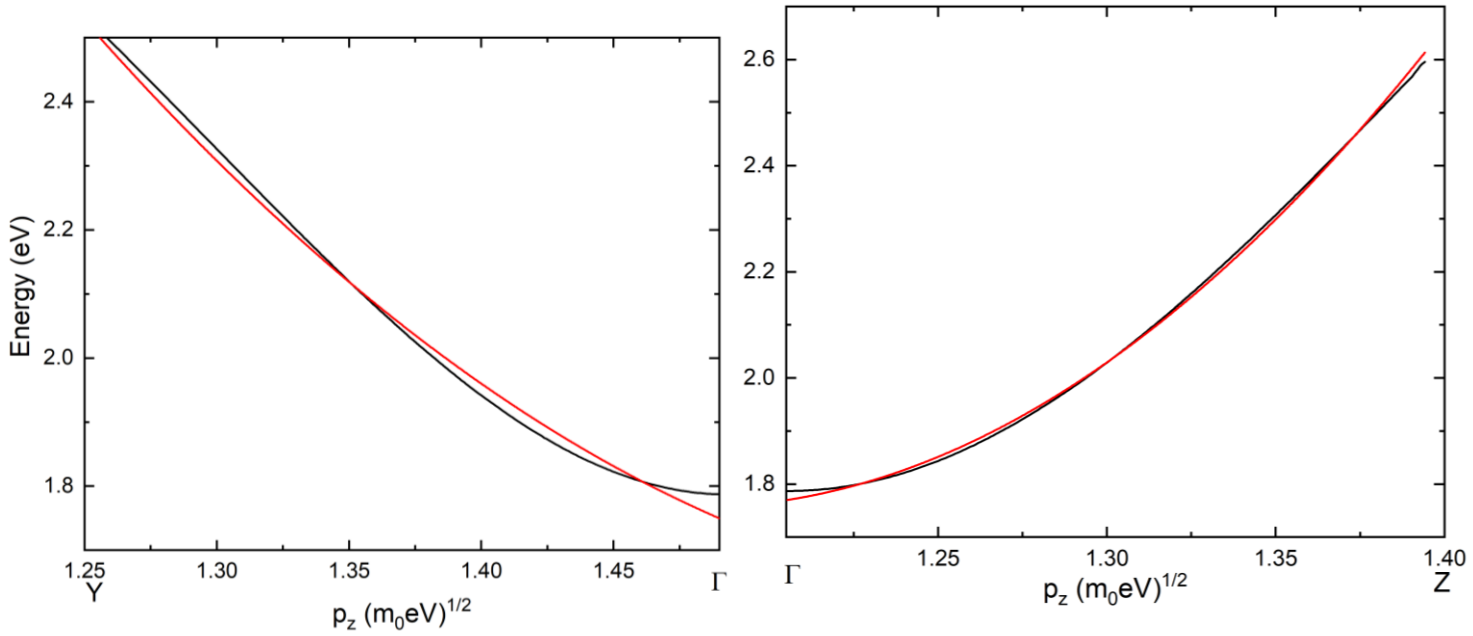


Red solid line represents the average of red values (0.0912) which is almost within the error bars.

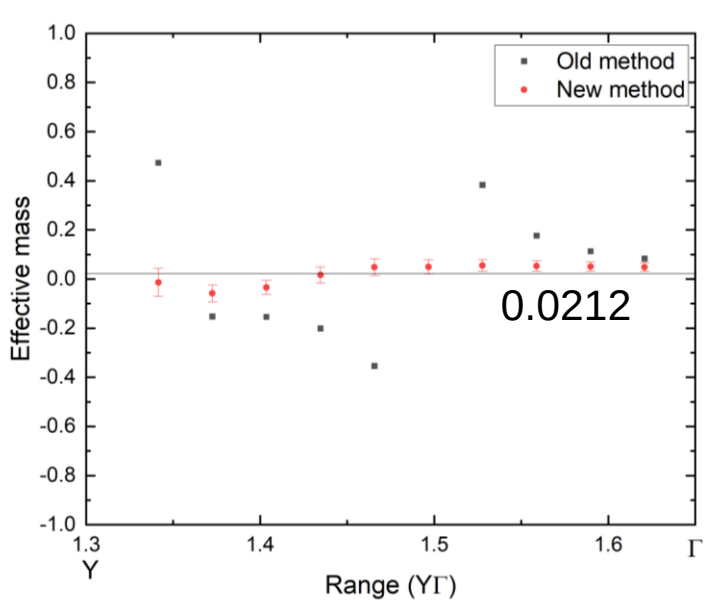
Effective mass calculation

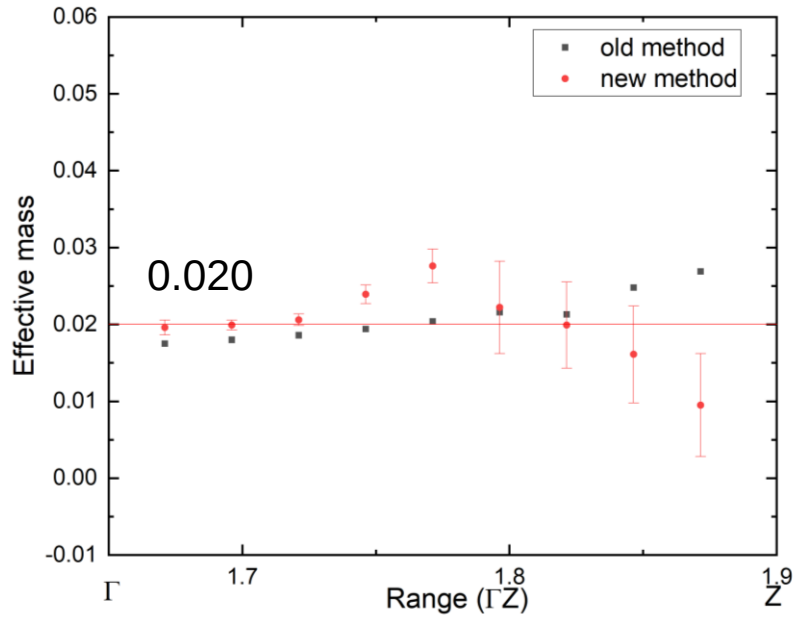
YΓ and ΓZ direction

- These top figures represent the lowest conduction band in the YΓ and ΓZ directions.
- Red solid lines represent the parabolic fits to relevant bands.



- These below figures represent the calculated effective masses similar to the ΓX direction.





Future plans???

- Which valence bands contribute to the photoemission?
- Which conduction bands are populated by the incident photon energy?
- Find the absorption coefficient to evaluate the UV absorption depth (how far electrons need to go to get out into the vacuum).
- Find the significance of phonon scattering (using the electron phase velocity).
- Three step analysis for Cs_2Te.

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Thank you

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Most recent QE measurements

17/03/2021

Photon energy (eV)	Current (nA)	Light Power (μW)	QE (%) With the lens	Log (QE%)	Current (nA)	Light Power (μW)	QE (%) without the lens	Log (QE%)
5.19	0.0235	0.494	2.4702×10^{-2}	-1.6073	0.021	0.700	1.5270×10^{-2}	-1.8162
4.88	0.0069	0.342	9.8913×10^{-3}	-2.0048	0.00527	0.567	4.6321×10^{-3}	-2.3342
4.18	0.00034	10.8	1.3229×10^{-5}	-4.8785	0.00024	10.4	9.5653×10^{-6}	-5.0193
3.71	0.00017	8.67	7.2843×10^{-6}	-5.1376	0.00012	7.79	5.5536×10^{-6}	-5.2554
3.4	0.00006	30.5	6.7277×10^{-7}	-6.1721	0.00004	24.9	5.4710×10^{-7}	-6.2619
3.18	0.00006	4.89	4.0196×10^{-6}	-5.3958	0.00006	5.596	4.5476×10^{-6}	-5.3422
2.95	0.00006	0.24	7.3966×10^{-5}	-4.1309	0.00007	0.208	8.5377×10^{-5}	-4.0687
2.82	0.00006	0.331	5.1114×10^{-5}	-4.2915	0.00008	0.3004	7.5093×10^{-5}	-4.1244