SimuLase_TMDC™

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SimuLase_TMDC® for 2D Materials Quick Start Guide

NLCSTR
7562 N. Palm Circle
Tucson, AZ 85704
Phone: +1 (520) 505-4561
Email: info@nlcstr.com
Web: www.nlcstr.com

For information about SimuLase please email simulase@nlcstr.com

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Terminology

SimuLase_TMDC stores all its data in hierarchal structure called a **Database** or **Gain Database**. A Database is composed of three major components:

Structure this file contains all information regarding the layout of the physical device under simulation
Model this file contains all information regarding the numerical model used to simulate the structure
Datasets a collection of parameterized data files containing computed values

For example, an *Absorption Dataset* contains absorption data for all combinations of parameters requested during a single simulation run. At a minimum a complete database contains four primary datasets named, *Absorption*, *Refractive Index*, *Photoluminescence* and *Band structure*. Additional auxiliary datasets may also be generated depending on the particular configuration of SimuLase model settings.

Key features and capabilities of SimuLase_TMDC include:

- Rigorous and predictive a priori computation key material properties, like, absorption, gain and photoluminescence spectra;
- Poisson solver to compute the influences of dielectric environments due the presence of multiple TMDC layers, substrate- or dielectric coating-materials;
- Physics captured within the relativistic Semiconductor Dirac Bloch model;
- Inclusion of higher order many-body correlations;
- Verified band structure inputs derived from first principles DFT calculations and verified against experimental data;
- Output data can be easily imported into other commercial higher level device software simulation tools.

Before you Begin

Installation Requirements

To review complete system requirements and recommendations for your SimuLase software, see the Read Me file included with your software.

Installing the Software

1) Close any other SimuLase applications open on your computer.

2) Launch the installer (SimuLase_TMDC_.msi) application and follow the on-screen instructions.

Activate the Software

SimuLase uses license management technology to ensure compliance with the product license agreement. This technology prompts you to verify the license of your product using a 25 character alphanumeric Product Activation Code (example code: `LMP8M-PSDXB-12345-F9BTC-24680`) during the installation process. This code is unique to each installation of SimuLase and is provided to you when the software is purchased.

Getting to Know SimuLase_TMDC

This Quick Start Guide provides a brief introduction to the key features and capabilities of SimuLase_TMDC as well as a quick tour of its Graphical User Interface. This user interface is designed to reduce clutter, streamline your design process and automats repetitive tasks allowing you to focus on the substance of your design studies. More detailed information, including some work examples, can be found in the companion SimuLase_TMDC User Guide document. Refer to this Users Guide for any feature not outlined in this supplement, or for more detailed information regarding the microscopic modelling of semiconductor devices as implemented in this application.

Using Online Help

The SimuLase_TMDC Quick Start Guide and the SimuLase Users Guide in PDF format can be accessed through its Help-menu on the Main menu bar. Additional information, including the most up-to-date version of all documents, can be downloaded from www.nlcstr.com

1. Design, Simulate, Analyze

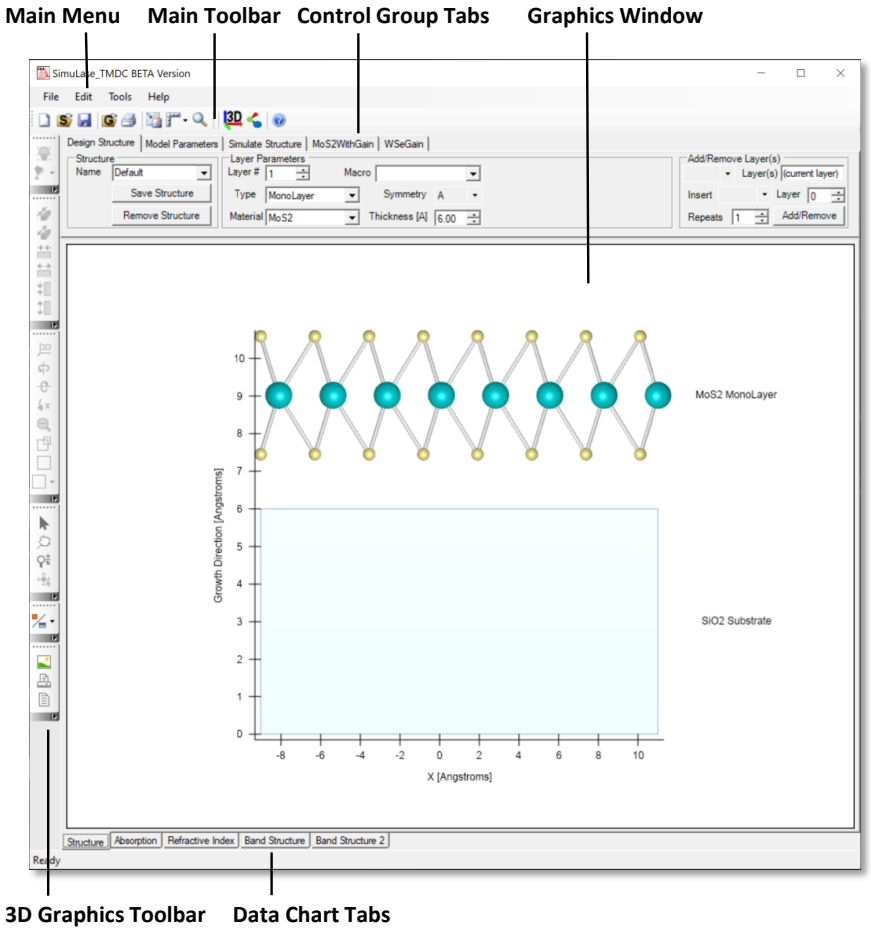


Figure 1 The Main Screen.

SimuLase_TMDC is designed around a three-stage workflow.

1) *Design the structure*: Using the layer designer the user interactively stacks layers of different materials to produce a layered structure device.

2) *Simulate the structure*: The structure is simulated using:
   a. Numerical Model parameters are selected that specify the resolution and complexity of the microscopic many-body model.
   b. Computational parameters that dictate the amount of computational resources allocated, (i.e., number of CPU cores) and parameter ranges over which the structure is simulated – like, e.g., carrier densities or temperatures.

3) *Analyze the structure*: Numerical results generated by the simulation are loaded and rendered in a series of charts. Results from previously executed simulations may also be loaded for comparative analysis. Interactive tools are provided in each chart to facilitate analysis and annotation of data charts. Raw and post processed datasets and publication quality plots may be exported in standard formats.

The SimuLase Graphical User Interface (GUI) implements each stage of this workflow by grouping controls related to each stage in separate tabbed windows. The following is a brief description of the main functionality of the each component of the SimuLase User Interface.

After starting SimuLase_TMDC, various controls are shown at the top of the application window. The controls are organized in three tabs labeled **Design Structure**, **Model Parameters** and **Simulate Structure**. In a typical workflow the user will move from left to right, sequentially clicking each tab and interacting with the controls. Below the control tabs is the main graphics window showing the current device structure. After simulating a structure, a number of additional tabs appear below the main graphics display. These tabs are used to switch between displays of the various datasets generated during the simulation.

1.1 Design a Structure

The controls on the **Design Structure** tab, located above the main graphics window, enable the user to construct a layered device and dynamically view and modify all aspects of the resulting device structure. The **Graphics Window** can display 2D or 3D interactive renderings of the layered device as it is constructed.

Selecting, Adding and Removing Layers

Layers are added or removed from the structure by *left-clicking* the mouse anywhere on the structure. The selected is indicated by adding a dashed black line around the perimeter. A context menu will appear which contains options to remove the selected layer or add a new layer. Adding a new layer will add a new MoS_2 layer to the top of the structure. This newly added layer may be modified by selecting the new layer and using the controls on the Design Structure tab to change the layer Type (Monolayer, HetroBiLayer Bulk or DFT_MDF) and the layer material from the Material pulldown.

The current structure can be cleared by clicking on the **Remove Structure** button on the on the Design Structure tab. This deletes the current structure from SimuLase allowing the user to construct a new structure. The first layer added to a new structure will automatically be chosen to be a Silicon dioxide (SiO_2) substrate layer and the second layer added will be a layer of MoS_2. The material composition of any layer may be modified by the user after it is placed on the canvas.

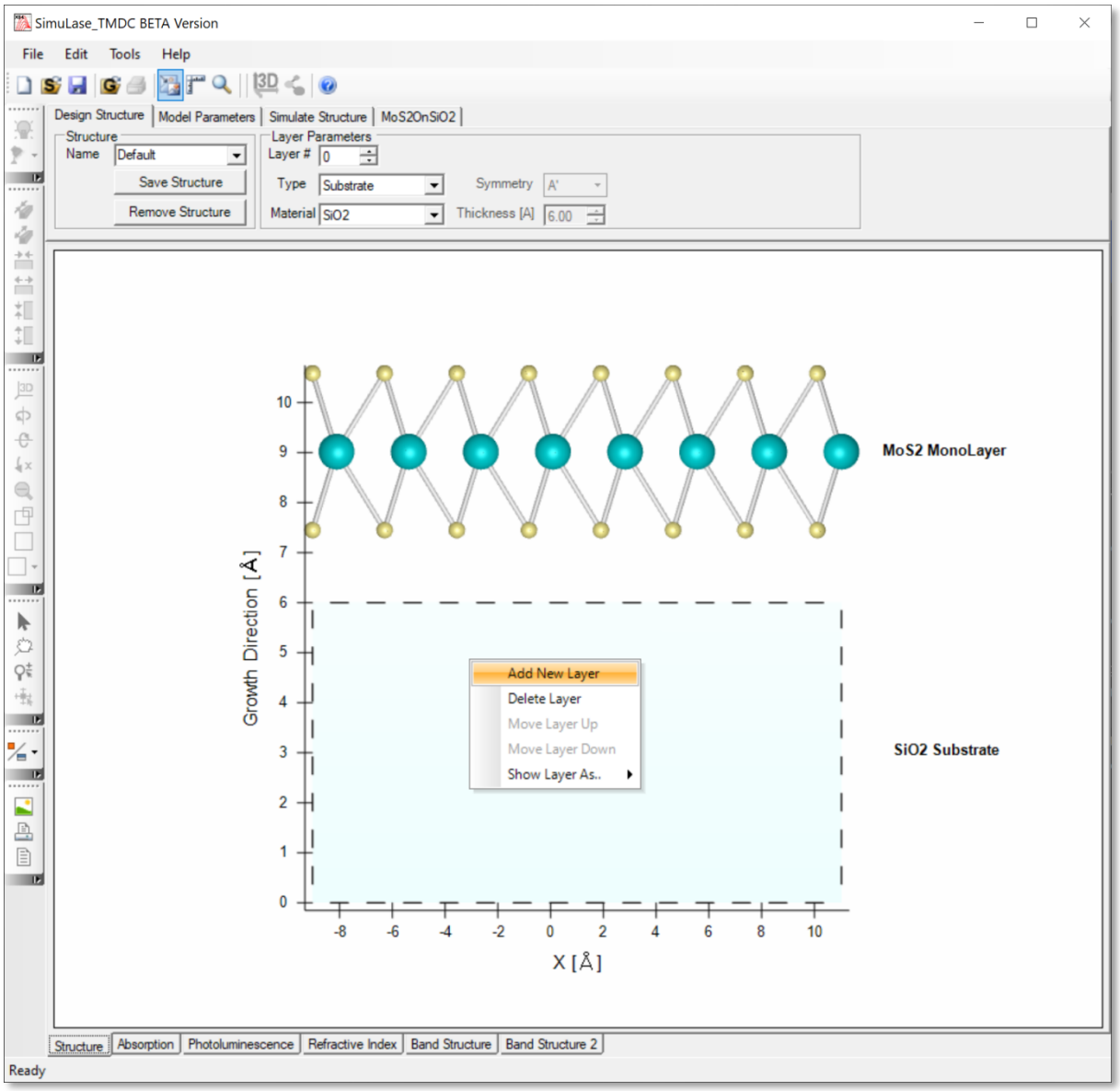


Figure 2 Selecting and Adding or Removing layers using the mouse context menu

The **Design Structure** tab is the default tab that appears when SimuLase is started. The default graphics window tab displays a basic 2-layer structure consisting of a Molybdenum disulfide (MoS_2) monolayer on a Silicon dioxide (SiO_2) substrate layer. The default structure may be replaced by selecting **File | Open Structure...** from the main menu to load a previously stored structure from disk.

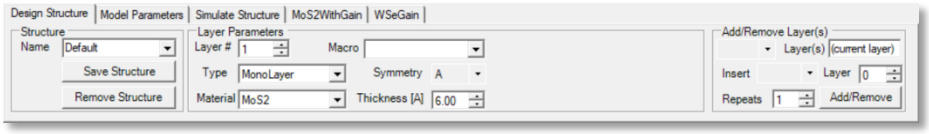


Figure 3 Design Structure.

The controls on the **Design Structure** tab are arranged into two functional groups that provide the following functionality:

Structure controls for naming, Saving and Removing the entire structure;
Layer Parameters controls for viewing and editing of the properties of a single layer;

The individual controls in each of these functional groups are detailed in the following sections.

Structure

The controls in this group relate to the overall structure. They are used to attach a unique identifying name to the structure, save the structure to disk or remove the entire structure from memory.

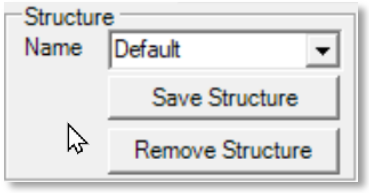


Figure 4 The Structure Controls.

Name is a string use to describe the entire structure. The structure name is initially assigned the name "Default" but should be changed by the user for each new structure. It is used throughout SimuLase to uniquely identify the structure. The name is prepended to all data files that are generated when the structure is simulated. When multiple Databases are loaded into SimuLase, for data analysis and comparison, all loaded structures are listed in the **Name** pulldown, the user may select which structure is rendered in the Structure window be selecting the appropriate name from the list.

Save Structure Information describing all aspects of the current structure is saved in a *SimuLase Structure File* (*.sls) using the structure name as the filename and the three letter extension **.sls**. The file is saved in ASCII using standard human readable xml-format. These files can be read using an external viewer or editor application such as, Microsoft Word, Microsoft Excel, Notepad or any Web Browser. While it is possible to edit this file in an

external editor, great care should be taken as it may result in an invalid file when read back into the SimuLase GUI.

Remove Structure Deletes the currently loaded structure from SimuLase allowing the user to construct a new structure.

Layer Parameters

The various controls in this group are used to display and edit the physical properties of the currently selected layer. Changing any of the displayed values for the current layer will cause the corresponding layer in the structure to be updated.

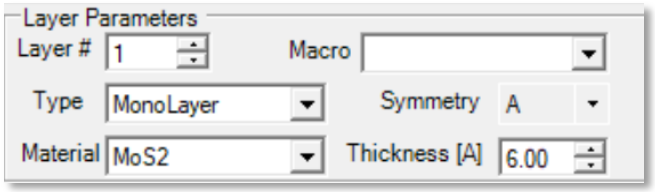


Figure 5 The Layer Parameters Controls.

Layer# A single layer is selected by changing the number field in this control (or using the mouse to select the layer by clicking anywhere on the layer shown in the graphics window). Layers are numbered sequentially starting with layer 0. The graphics window highlights the currently selected layer by surrounding the layer with a thick dashed line around the layer perimiter.

Type A descriptor of the type of layer in the structure. A layer can be one of five types, *Substrate*, *MonoLayer*, *Hetero-Bi-Layer*, *Bulk or DFT_MDF*.

- *Substrate* is either a metal or dielectric material. Its thickness is assumed to be infinite below or above the active TMDC material. Only the dielectric properties of the material are taken into account in evaluating the screening of the Coulomb interaction of the TMDC material. NLCSTR will add additonal Substrate materials on demand at no cost to the user.
- *Monolayer* is a single layer of TMDC material. Several *Monolayers* of the same material can be stacked to form a homo-multi-layer structure.
- *Hetero-Bi-Layer* is a set of two monolayers of different TMDC materials stacked on top of each other. In contrast to a homo-multi-layer structure, this configuration requires additional material paramters specifying the band offsets between the two materials.
- *Bulk* is bulk TMDC material. For this case potentially present *Substrate* layers are not taken into account in the calculation. The calculation is performed using the same

material parameters as for a monolayer but replacing the dielecrtic evironment by that of the bulk TMDC material.

- *DFT_MDF* is a user-defined material for which the user provides all relevant material parameters as obtained through DFT calculations. The determination of material parameters from the output of VASP calculations is aided by the **DFT_to_MDF** tool under the **Tool** pull-down menu. For a description of the details of the implementation of a user define DFT material see the corresponding section of the user manual.

Material The material composing the layer.

Texture The Texture pull-down provides a list of jpg pictures that can be used to represent the TMDC layer inn the absence of a ball-and-stick representation that is generated from a VASP POSCAR file. This can be used, e.g., to represent user defined **DFT_MDF** materials.

Symmmertry This field is currently disabled. In future versions of SimuLase_TMDC, this field will allow to select between A- and B-type stacking-orders of TMDC monolayers.

Thickness [A] The thickness of the current layer in Angstroms. This field is currently disabled. In future versions of SimuLase_TMDC this field will allow to specify the thickness of *Substrate*-type layers, like, e.g., air or dielectric coatings, which can be located between layers of TMDC material.

The Graphics Window

The **Graphics Window** is used to display the current structure or line plots of the datasets produced after executing a simulation of the structure. A series of tabs along the bottom drawing area are used to select between the various types of data that can be rendered on the drawing canvas.

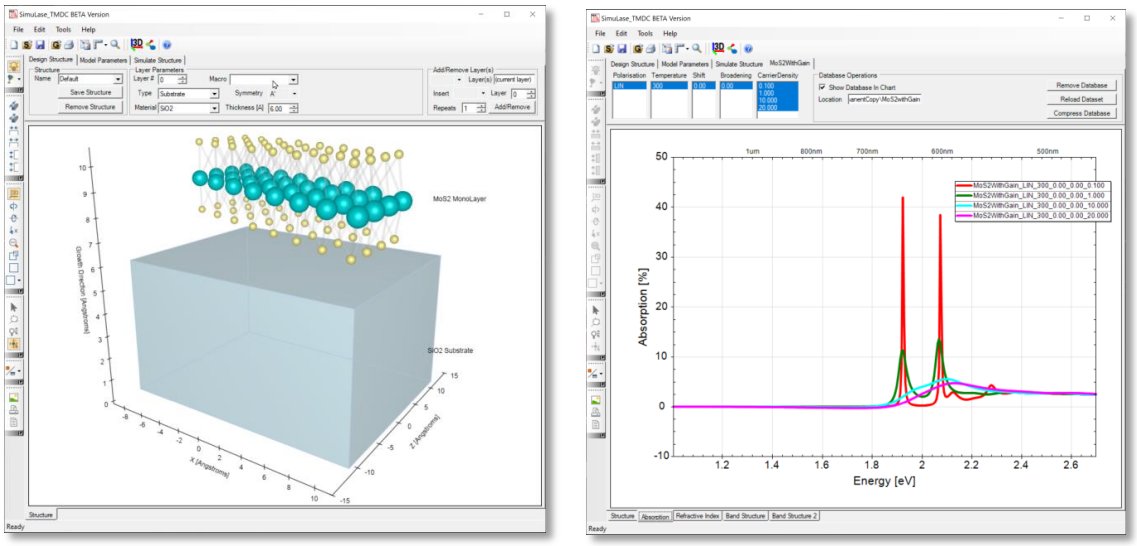


Figure 6 The Graphics Window.

As shown in Figure 6 (left), when the **Structure** tab at the bottom of the graphics window is selected a 2D or 3D interactive rendering of the current layer device is displayed. After simulating a structure, or loading in precomputed results from a previous simulation, a number of new tabs are added to the bottom of the graphics window. Selecting one of the tabs causes the relevant data to be rendered to the graphics canvas. For example Figure 6 (right) shows line plots of the Absorption data for the Molybdenum disulfide (MoS_2) monolayer on a Silicon dioxide (SiO_2) substrate layer structure shown in Figure 6 (left).

1.2 Simulate a Structure

Simulating a structure involves setting controls on the tabs **Model Parameters** and **Simulate Structure**. The Model parameters tab contains controls that specify the resolution and complexity of the model.

The Simulate Structure tab contains controls that allow the user to specify a range of parameters, such as polarizations, temperatures, carrier densities for which the structure is simulated. It also shows the available computational resources and allows the user to dictate the number of CPU cores allocated to the simulation.

Model Parameters

The controls on the **Model Parameters** tab are arranged into four functional groups that provide the following functionality:

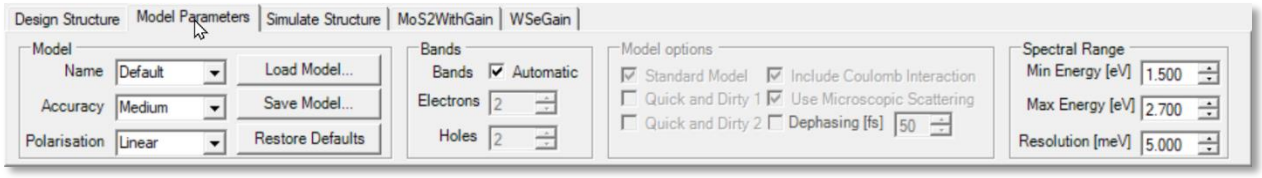


Figure 7 Model Parameters.

Model controls that deal with the model such as a name for the model and saving and loading pre-defined models.

Model Options controls the microscopic model for simulating structures. The default is the

Standard Model which includes all Coulomb effects and microscopically calculated electron-electron and electron-phonon scatterings.

Quick and Dirty 1 includes excitonic effects on the Hartree-Fock level but replaces the dephasing causing scatterings by a simple dephasing time, T_2.

Quick and Dirty 2 neglects all Coulomb effects and performs a simple single-particle calculation on the level of Fermis Golden Rule.

Spectral Range controls the spectral range for which spectra shall be calculated. Default values that cover the spectral range of highest interest are set automatically for all materials that are implemented in SimuLase_TMDC. The user can use these controls to refine the resolution, expand or reduce the spectral range and to set these parameters for user defined DFT_MDF materials.

The individual controls in each of these functional groups are detailed in the following sections.

Model

The controls in this group relate to the overall structure. They are used to attach a unique identifying name to the structure, save the structure to disk or remove the entire structure from memory.

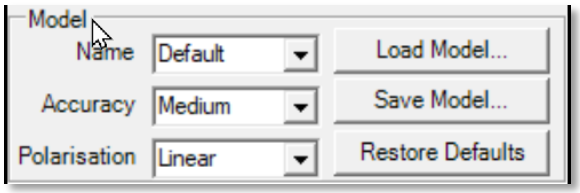


Figure 8 Models Panel.

Name is a string use to describe the collection of model parameters. The model name is initially assigned the name "Default" but should be changed by the user for each new model configuration created. It is used in SimuLase to uniquely identify the collection of model parameters. The name is employed when the same structure is modeled using a different set of parameters.

Accuracy accuracy can be set to *High*, *Medium* or *Low* and determines the number of momentum grid points used in the simulation and, thus, the accuracy of the calculations. The calculation time and CPU-memory requirement scale strongly with the accuracy level. Typicaly, the calculation time increases by about a factor of four when going to a higher accuracy level. By default the accuracy-level is set to *Medium* which we recommend for typical calculations.

Polarization sets the light-polarization for which the spectra will be calculated to Linear, Circular -L (left circular) or Circular-R (right circular). The calculation time is independent of the choice of polarization.

Save Model Information describing all aspects of the current model is saved in a *SimuLase Model File* (*.slm) using the model name as the filename and the three letter extension **.slm**. The model parameters file is saved in ASCII using standard human readable xml-format.

Model files can be read using an external viewer or editor application such as, Microsoft Word, Microsoft Excel, Notepad or any Web Browser. While it is possible to edit this file in an external editor, great care should be taken as it may result in an invalid file when read back into the SimuLase_TMDC GUI. A .slm file is also created by each simulation with the prefix as specified for the database.

Load Model Information describing all aspects of the current model is loaded from a preexisting *SimuLase Model File* (*.slm). All existing model parameter values in the GUI are erased and replaced with the corresponding values in the loaded model file.

Restore Defaults Model All existing model parameter values in the GUI are erased and replaced with default values.

Model Options

The controls in this group specific the global behavior of the backend numerical calculation.

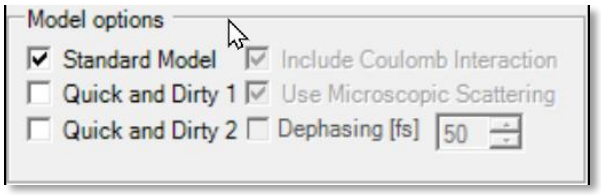


Figure 9 Model Options Panel.

Standard Model by checking this option, the recommended fully microscopic model for calculating reliable Gain Databases is selected. This is the most extensive model available, including all Coulomb effects and microscopically calculated electron-electron and electron-phonon scatterings that are required for correct lineshapes and amplitudes.

Quick and Dirty 1 by checking this option the GainDatabase is set up using a model that includes everything the *Standard Model* (above) takes into account except for microscopic scatterings and higher order excitonic correlations which are source terms for the PL. Since the scatterings are not used to describe the dephasing of the optical polarization, a dephasing time *Dephasing* has to be used. Calculation times within this model are much shorter than when using the the *Standard Model* However, the resulting lineshapes, spectral positions and amplitudes will have significant errors. The calculations lose their quantitatively predictive quality that the *Standard Model* provides. The resulting radiative carrier lifetimes are typically wrong by a factor of five or more.

Quick and Dirty 2 by checking this option the least computationaly intensive model will be used in the simulations. In addition to the simplifcations used by option 'Quick and Dirty 1', this model also neglects all Coulomb effects, including excitons and bandgap renormalizations. It requires the user to specify a value for Dephasing time. With these simplifications the calculation time for one carrier density and one temperature usually reduces to a few seconds. This option can be used to quickly get some very rough estimates or to check the influence of Coulomb effects by comparing to the more sophisticated models.

Include Coulomb Interaction if this option is not checked, all Coulomb effects are neglected, including all microscopic scatterings and higher order excitonic correlations. Unchecking this option reduces the model to the level of *Quick and Dirty 2* described above. If un-checked, a dephasing time T_2 has to be specified.

Use Microscopic Scattering if un-checked, the dephasing of the optical polarization will be described by a dephasing time, T_2, rather than by calculating the underlying electron-electron and electron-phonon scattering processes.

Dephasing Time if checked, the specified dephasing time will be used for the dephasing of the optical polarizations. This usually leads to significant errors in the lineshapes, amplitudes and spectral positions. Even if microscopic scatterings are taken into account, this dephasing time is added into the description of the dephasing of the polarizations unless this field is un-checked or deactivated.

Spectral Range

The controls in this group set the spectral range for which spectra shall be calculated. By default these values are set to a 'reasonable' estimate according to the bandstructure of the TMDC materials.

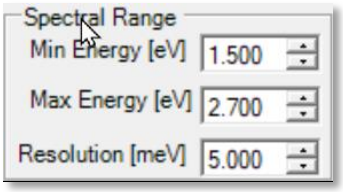


Figure 10 Spectral Range.

Min Energy sets the lowest transition energy, in electron Volts, for which spectra shall be calculated. By default, this is set to be about 0.5 eV below the A-bandgap.

Max Energy sets the highest transition energy, in electron Volts, for which spectra shall be calculated. By default, this is set to be about 0.5 eV above the B-bandgap.

Resolution sets the spectral resolution, in meV . By default this is set to 5 meV. For spectra with sharp features this should be reduced. The calculation time scales about linearly with the inverse of the resolution.

Simulate Structure

The controls on the **Simulate Structure** tab are arranged into two functional groups that provide the following functionality:

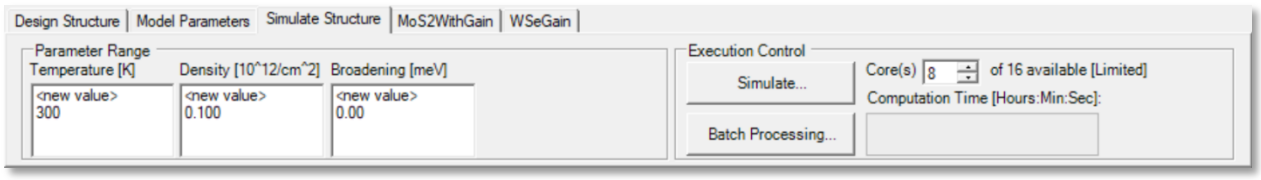


Figure 11 Simulate Structure.

Parameter Range used to specify the parameters for which the backend numerical simulator will produce datasets;
Execution Control controls relating to the launching and execution of structure simulations;

The individual controls in each of these functional groups are detailed in the following sections.

Parameter Range

The controls in this group relate to the overall structure. To enter a value to the list double-click on *'<new value>'*, replace the string by a number and hit return to confirm. To delete an entry double click on it, hit the *'backspace'* or *'delete'* button and then *'return'* to confirm. Data will be calculated for all possible combinations of temperatures, densities and inhomogeneous broadenings. The calculation time and amount of produced data scales with the number of entries in the 'Temperature'-field times the number of entries in the 'Density'-field. The number of broadenings has virtually no influence on the calculation time. However, the amount of produced data increases linearly with the number of broadenings which might have be considered if very large number of datasets are to be created.

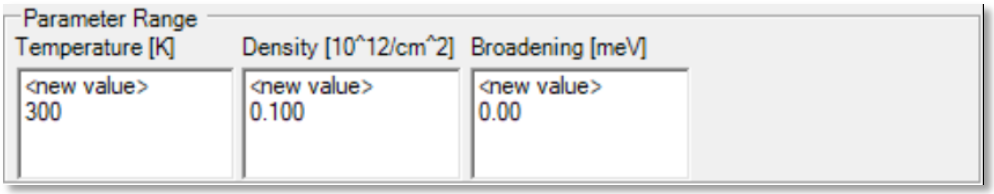


Figure 12 Parameter Ranges.

Temperature [K]: sets up a list of temperatures in Kelvin for which the database shall be created. The temperature as entered here is used as lattice and carrier temperature. The calculation effort increases with decreasing temperature as higher momentum grid resolution is required due to increased spectral sharpness at lower temperature.

Density sets up a list of electron sheet carrier densities in units of [10^{12}/cm^2]. It is assumed that the carriers are in thermal equilibrium. Currently SimuLase_TMDC assumes an equal electron and hole carrier density and each is given by the values as specified here.

Broadening Sets up a list of inhomogeneous broadenings in units of [meV] (FWHM). Besides the spectra that include only the homogeneous broadening due to electron-electron and electron-phonon scattering, inhomogeneously broadened copies of the spectra will be generated.

Execution Control

The controls in this group relate to the launching of a calculation of the structure using to the model parameters that have been set in the Parameter Range control group. After the *Simulate* button is clicked a file manager window opens to specify a name for the database and the directory into which it shall be written. In case of computations that require significant CPU memory your computer will be mostly unusable for other purposes. Using the computer for other purposes while a database is created can cause problems to the calculation. On a multi-processor machine one should leave at least one processor open for alternative work.

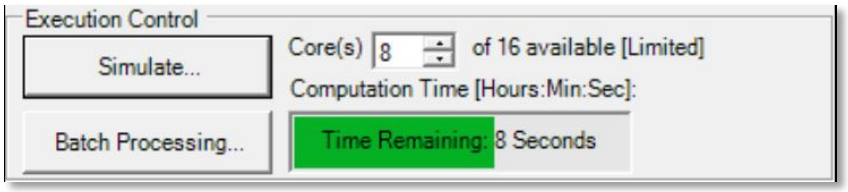


Figure 13 Execution Control.

Simulate clicking this button starts the calculation of the structure using to the model parameters.

Core(s): defines the number of processors that will be used for the calculations. Creating the data for each temperature-density combination is done sequentially but the calculation is parallelized over the number of processors as defined in the '*Core(s)*' control.

Progress Bar this control displays information regarding the progress of the simulation and the aproximate time remaining.

Batch Processing this control is currently disabled. In future versions of SimuLase_TMDC it will allow to batch-process variations of parameters other than those as specified on the field *Parameter Ranges*, like dielectric layer widths or MDF parameters of user defined *DFT_MDF* materials.

1.3 Analyze Results

Numerical results generated by the simulation are loaded and rendered in a series of line charts displayed in the Graphics Window. Results from previously executed simulations may also be loaded for comparative analysis. Interactive tools are provided in each chart to facilitate analysis and annotation of data charts. Raw and post processed datasets and publication quality plots may be exported in standard graphics file formats.

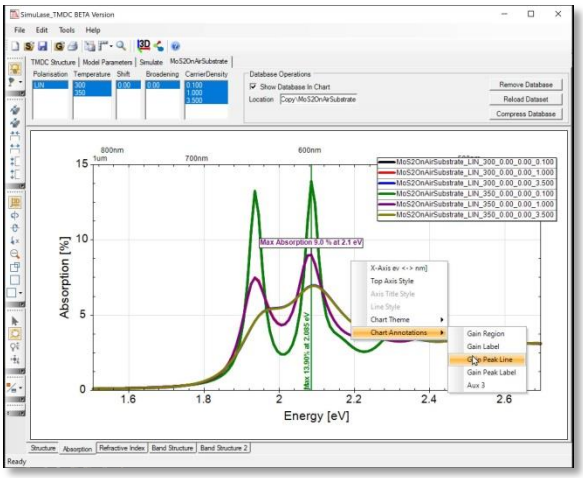


Figure 14 A typical view of SimuLase_TMDC while analyzing data.

Loading and Interacting with Databases

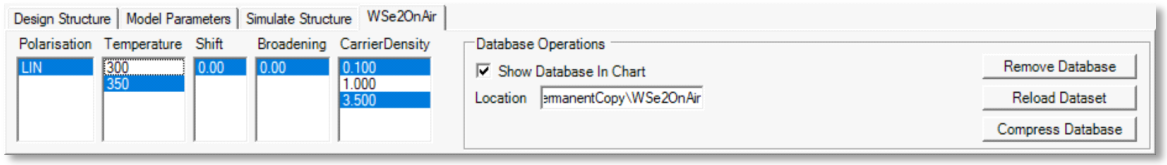
Multiple databases may be loaded into SimuLase_TMDC. After running a simulation, the newly created database is automatically loaded. A preexisting database can be loaded into SimuLase by selecting **File | Open Gain Database...** or using the Open Database icon .

After loading a database, a new tab containing the database name is added to the Control Group Tabs near the top of the screen. A new chart tab for each of the primary datasets (Absorption, Photoluminescence etc.,) is added to the chart tabs along the bottom of the graphics window. All currently loaded databases are plotted in same chart, for example the Absorption Chart can display absorption data from all currently loaded databases. This enables comparative analysis of datasets from different simulation databases.

Dataset plots are accessible via a series of tabs along the bottom of the Graphics Window. By selecting the appropriate tab, a user can view line plots of the main simulation result datasets i.e. 'Absorption', 'Photoluminescence' (i.e. Spontaneous Emission) and 'Refractive Index'.

Interacting with Datasets

When a database is loaded, a new tab bearing the name of the database is added to the Control Group Tabs above the Graphics Window. The controls on this tab select which data series from the database are plotted in the various charts, and perform various operations on the database.



The tab shows lists of all parameters contained in the database. The user may select a subset of parameters by left-clicking the mouse on the entries in the parameter lists. At least one entry in each parameter list must be selected. A single mouse click within the list will select that single parameter value. Multiple disjoint parameters may be selected by pressing the CTRL key while clicking entries in a parameter list. A continuous range of parameters may be selected by clicking on a value, and then pressing the SHIFT key while clicking on another parameter in the same list, all parameter values between the two clicked values will be selected. In all cases the data displayed in all charts will be immediately updated to display line plots of the selected parameter combinations.

Database Operations

The controls in this group are used to perform various operations on a whole database.

Show Database In Chart If this control is checked, the selected parameter sets will be plotted in the chart window. This is primarily of use when multiple databases have been loaded for comparative analysis. The user may dynamically Show or Hide line plots from the database in the chart window.

Location Shows the location of the database on disk. If the full pathname to the database is too long to fit in the location window, moving the mouse pointer into the next window will cause a temporary panel to pop showing the entire pathname.

Remove Database This will unload the current database from the GUI.

Reload Database This will reload the current database from the disk in case data has been externally modified or a calculation has been re-run with different model options.

Compress Database This enables the user to save a compressed copy of the database to a new location on disk.

Interacting with Charts

Charts in SimuLase are configurable in terms of their graphical presentation such as label size, number format, background shading etc. The user may also select and interact with line plots to measure quantities or attach various annotations to data series within the chart. Most of these operations are accessed via a context menu that appears when the

user right clicks the mouse anywhere within the chart window. Alternatively, measurement tools may be selected from the main menu or main toolbar.

In general, interacting with charts and data series fall into three main categories:

Customizing chart visuals involves right clicking anywhere on the chart background and changing visual attributes such as access to logarithmic scale, background color, numerical accuracy of legends and axis labels etc.

Measuring data sets this involves selecting a measuring tool from main menu and dragging the mouse across the chart. Measured values are dynamically extracted from the data series and displayed in a pop-up window as the mouse is dragged across the chart. Measurements can be restricted to a subset of the data series by pre-selecting the desired data series before applying the measurement tool.

Annotating data series involves selecting one or more data sets from the chart and right clicking anywhere on the chart background and selecting an annotation to be applied to the chart data set. Usually annotations operate in toggle mode, in that, re-applying an annotation to a chart series will remove the existing annotation.

Mouse Interaction Modes

Three icons on the main toolbar control how the mouse interacts with the chart. Mouse interaction with a chart can be in one (or none) of the following modes: selection, measure, or zoom.

Select Chart Datasets, **,** enables the selection of one or more data sets in the chart window. Selected datasets are highlighted, and subsequent dataset operations are applied to all selected datasets. Generally, if no datasets are selected then dataset operations are applied to **ALL** displayed datasets.

Measure, **,** interactively measure and display raw dataset values or computed quantities derived from the dataset directly on the chart.

Zoom, **,** zooms into regions of chart data sets adjusting the axes as appropriate. Clicking the left mouse button when the mouse is over the chart and dragging the mouse from the top left-hand corner towards the bottom right-hand corner will select a region highlighted in blue which represents the zoomed region. To un-zoom a chart, hold the left mouse button and drag the mouse from the bottom right-hand corner towards the top left.

Selecting Datasets

Left-clicking the mouse on a displayed line series will select that line series. Multiple line series may be simultaneously selected within a chart. A selected series is indicated by overlaying square symbols on top-of-the-line plot. Measurement interactions usually apply to all line series on the chart, but maybe restricted if one or more line series are specifically selected. Annotation interactions usually require that at least one line series be selected.

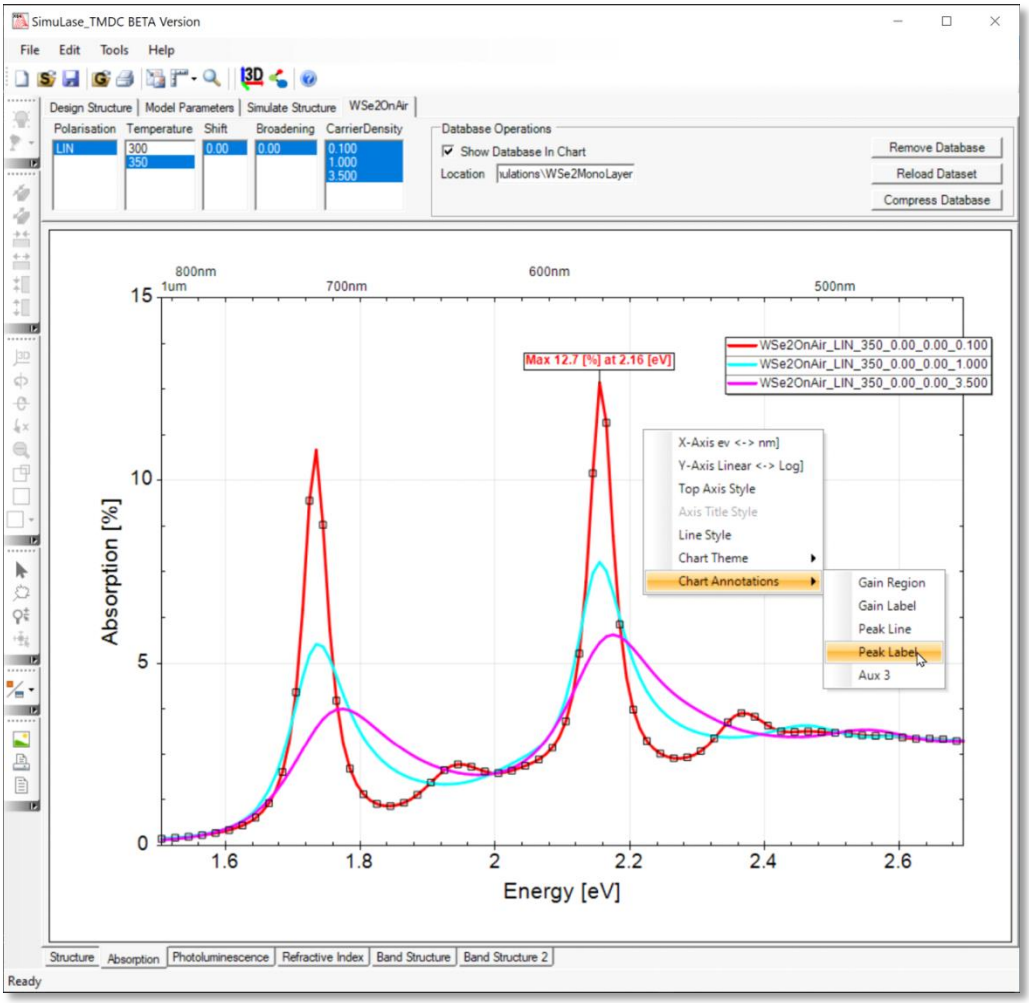


Figure 15 Selecting and annotating datasets

Chart Context Menu

Various operations may be applied to the whole chart or to one or more line series by right-clicking the mouse anywhere on the chart.

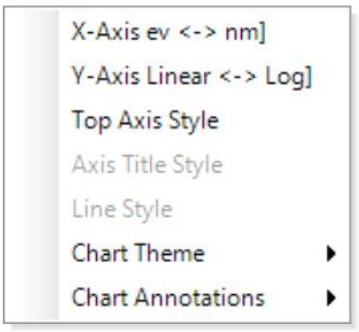


Figure 16 The main chart context menu.

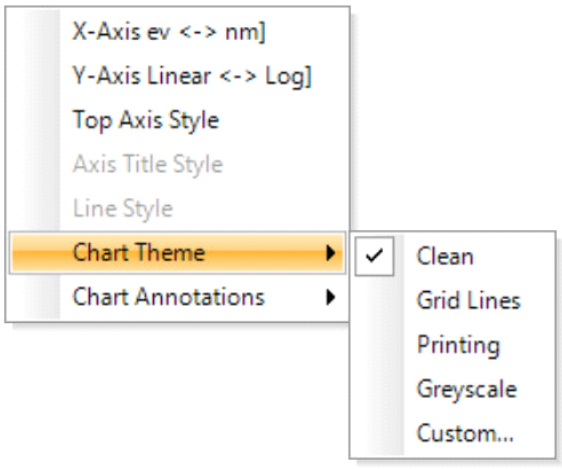
X-Axis eV <-> nm This will toggle the units on X axis from Energy in electron Volts to Wavelength in nanometers. The secondary X axis on top of the chart will toggle accordingly.

Y-Axis Linear <-> Log This will toggle the Y axis from a linear scale to a logarithmic scale

Line Style This opens a dialog that allows to change drawing style, color and line thickness of the selected data series.

Chart Themes

Chart themes enable the user to quickly change the appearance of a chart in order to make your data presentation look exactly the way you like. SimuLase_TMDC provides 4 predefined chart themes and also allows the user access to fully customize all graphical attributes of the chart.



Clean This provides an uncluttered presentation suitable for on screen viewing and analysis of datasets. This is the default setting when SimuLase is started.

Grid Lines In addition to the major gridlines, this theme adds dotted horizontal and vertical gridlines at the minor axis scale tick marks.

Printing This provides a chart presentation suitable for publishing or screen presentation where the chart size is reduced. It improves chart legibility by increasing the font size on all axes titles and axis scales relative to the chart area and reducing the number of major tick marks and the number of decimal places in tick labels.

Custom... SimuLase_TMDC makes it easy to create custom user interface themes based on any of the existing ones. Selecting this option will cause a property control to appear as shown below.

Custom Chart Themes

Selecting the **Custom...** option from the Chart Theme context menu will display the Chart Theme property Grid as shown

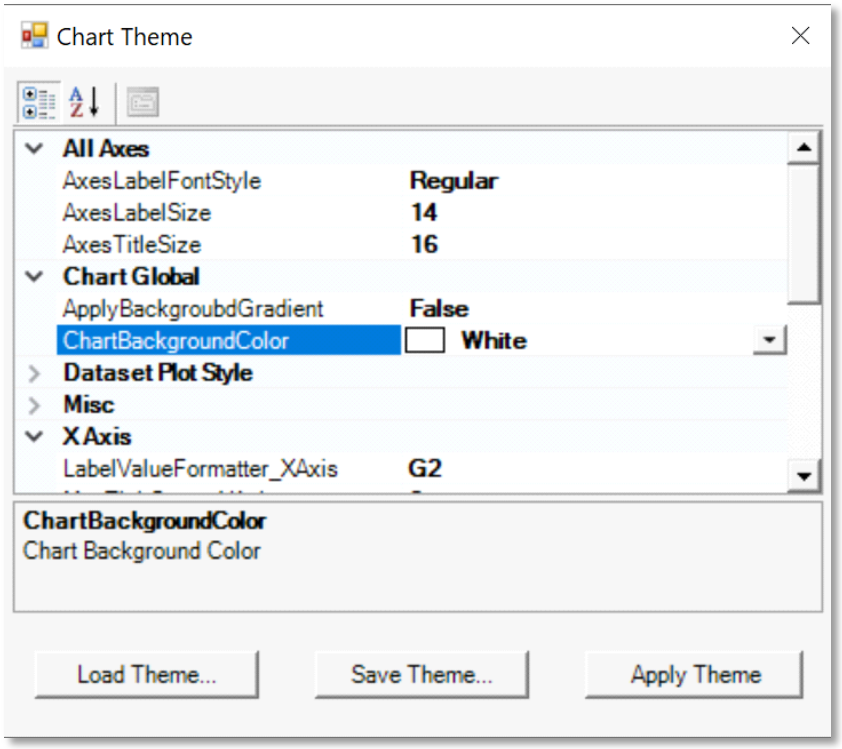


Figure 17 The Chart Theme Property Grid.

The Chart Theme Property Grid control displays all the user customizable properties of a chart in a user-friendly way, enabling end users to edit the visual properties as desired to construct a custom chart. Properties are organized into named categories that group related properties according to their function. At the top is the toolbar, which allows the

properties to be classified by category or listed alphabetically by property name. The central window lists properties in a <name-value> format. At the bottom is a help text that provides a brief description of the property that the user has selected.

Most basic types of properties are simple text that the user may edit directly to change the value of type of the displayed string or numeric value in general.

More complex entries provide sophisticated default editors for value types or select from a list of predefined available values. For example, a property of type Color has a drop-down list that lets you select any available color as shown.

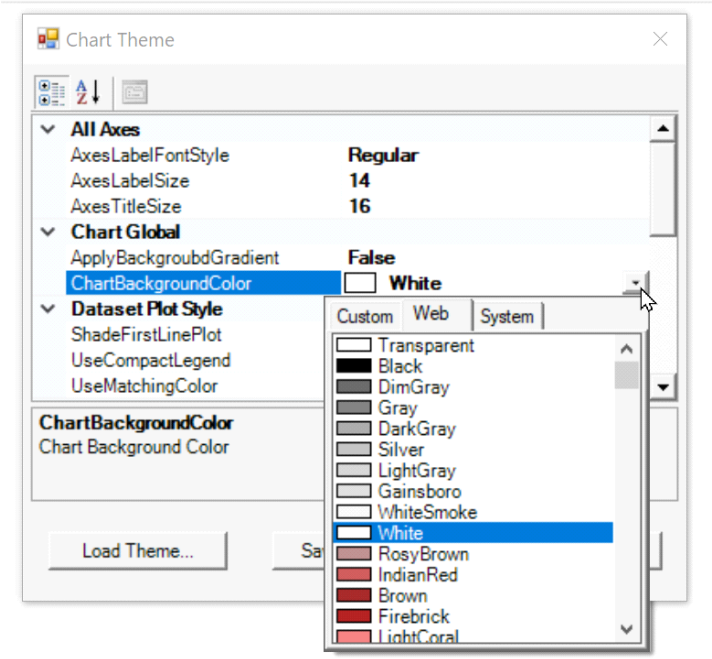
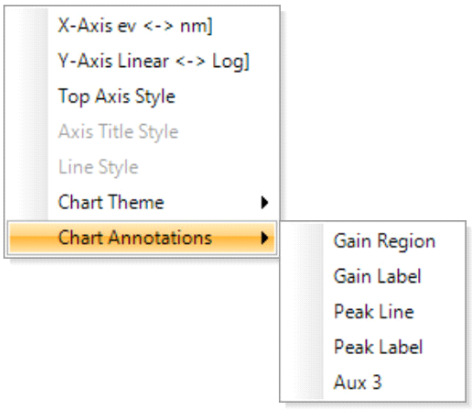


Figure 18 The color selector drop-down in the Chart Theme Property Grid

Chart Annotations

Selecting the Chart Annotations on the context menu allows the user to attach annotations that highlight certain aspects, for example the location and peak value of a data set, or the gain region on an Absorption chart.



Gain Region highlights the region over which the absorption exhibits gain by using a band of transparent color that matches the color of select data series line plot. Multiple overlapping gain regions may be drawn on the chart.

Gain Label highlights the region over which the absorption exhibits gain (negative absorption) – if such a region exists - by adding an additional label to the X axis scale. Multiple overlapping Gain Labels may be added to the chart axes.

Peak Line finds the maximum numeric value in the data set that has been selected using the Select Chart Dataset, , option and inserts a color matching vertical marker line and text detailing the X axis location and numeric value of the peak.

Peak Label finds the maximum numeric value in the data set that has been selected using the Select Chart Dataset, , option and inserts a color matching text box above the peak detailing the X axis location and numeric value of the peak.

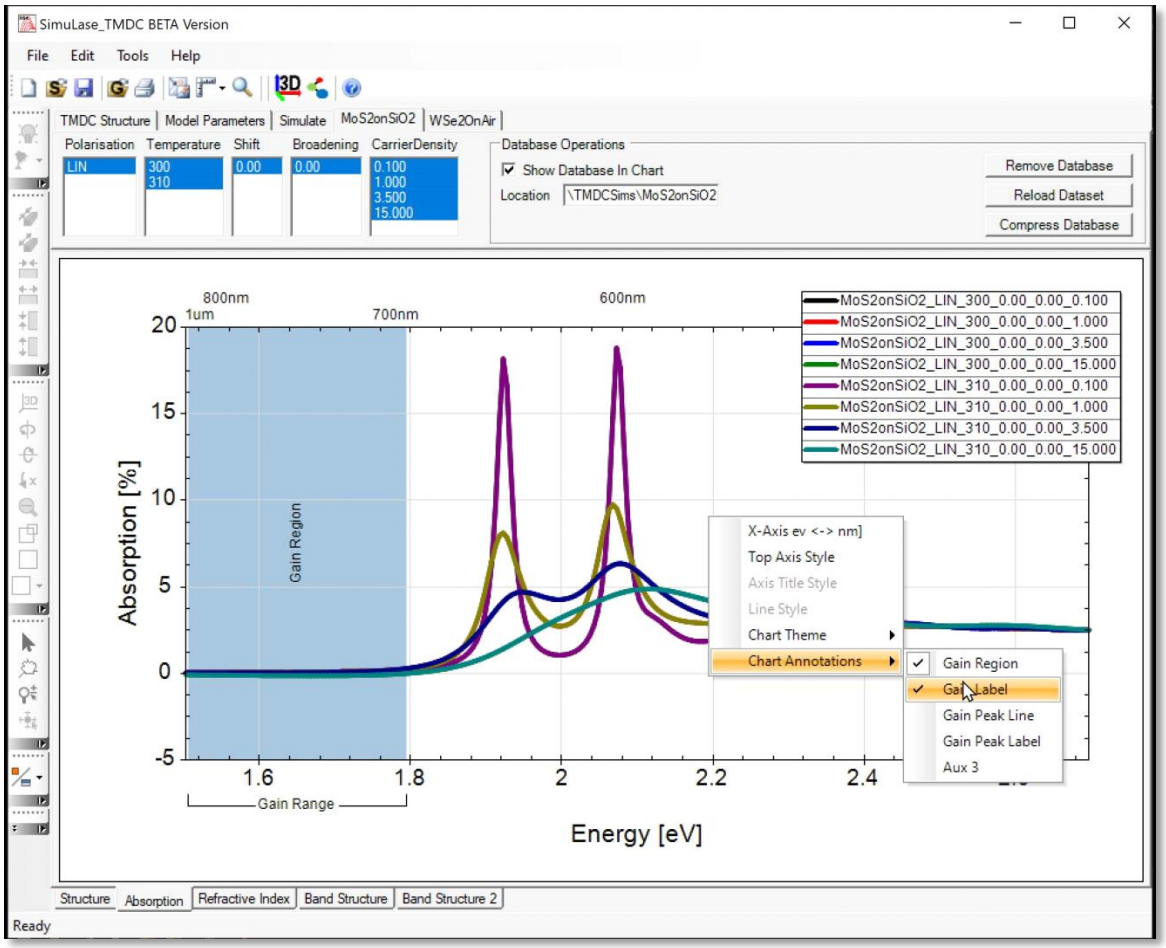


Figure 19 Chart showing both Gain Region and Gain Label annotations.

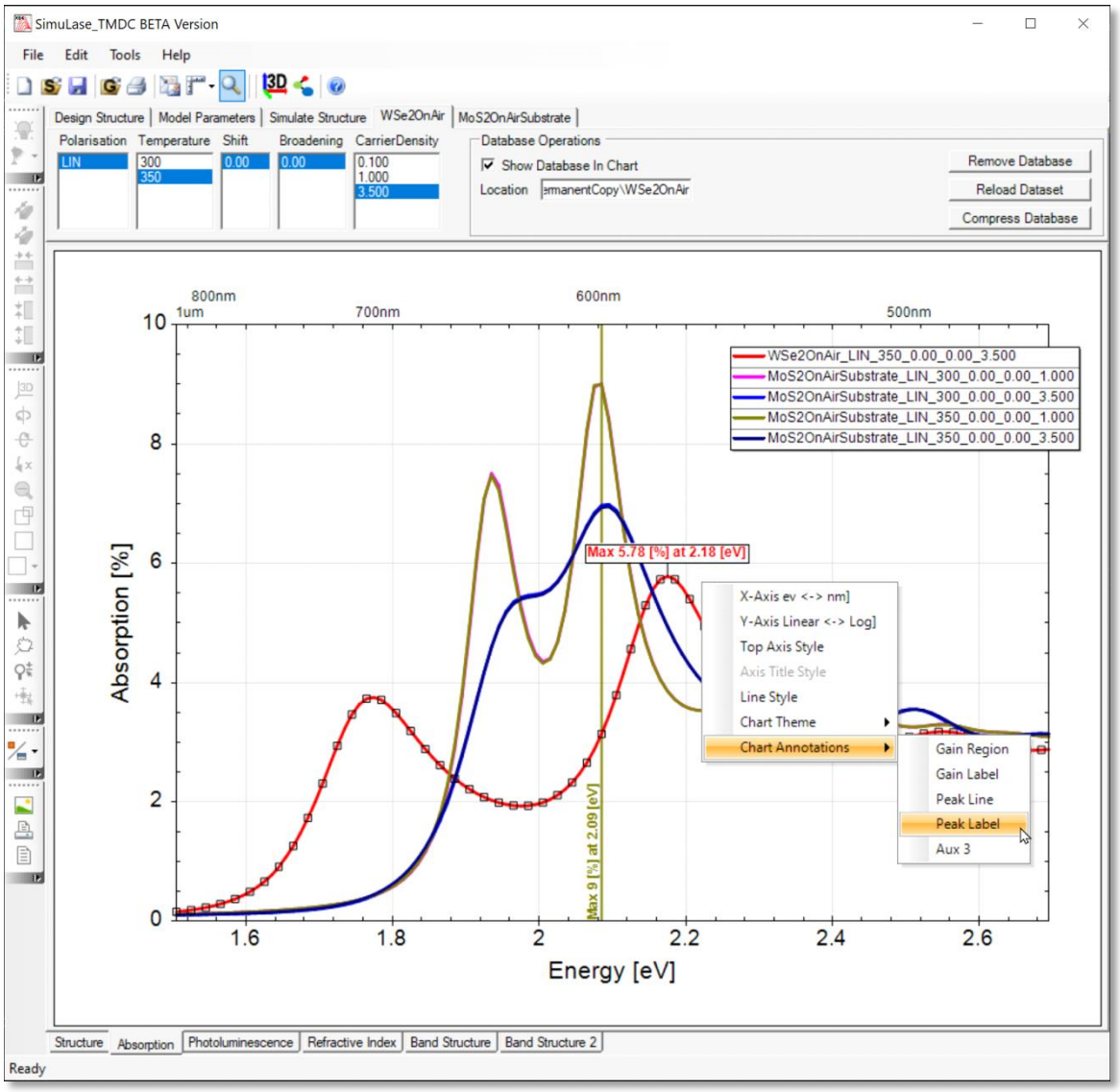


Figure 20 Chart showing both Peak Line and Peak Label annotations.

Measure Dataset Value

Figure 21 Measurement Tools drop-down on main tool bar

In addition to attaching visual annotations to data series it is also possible to interactively measure and display a subset of raw or derived quantities from the loaded dataset. The measurement results displayed depend on the selected tool and the type of data contained in the chart.

The Measure Dataset Value tool is activated by selecting from the main tool bar and clicking into the graph. This will change the mouse pointer to a crosshair cursor that displays the exact X-value and a list of Y-values read from each of the selected data series. If no data series is currently selected, then the values are presented from all displayed data series.

The following image shows the Measure Dataset Value tool as used in an Absorption chart. The floating panel shows the current Energy value in the header and a list of Absorption values for all displayed data series. If a data series exhibits gain (negative absorption) then that value is highlighted in yellow.

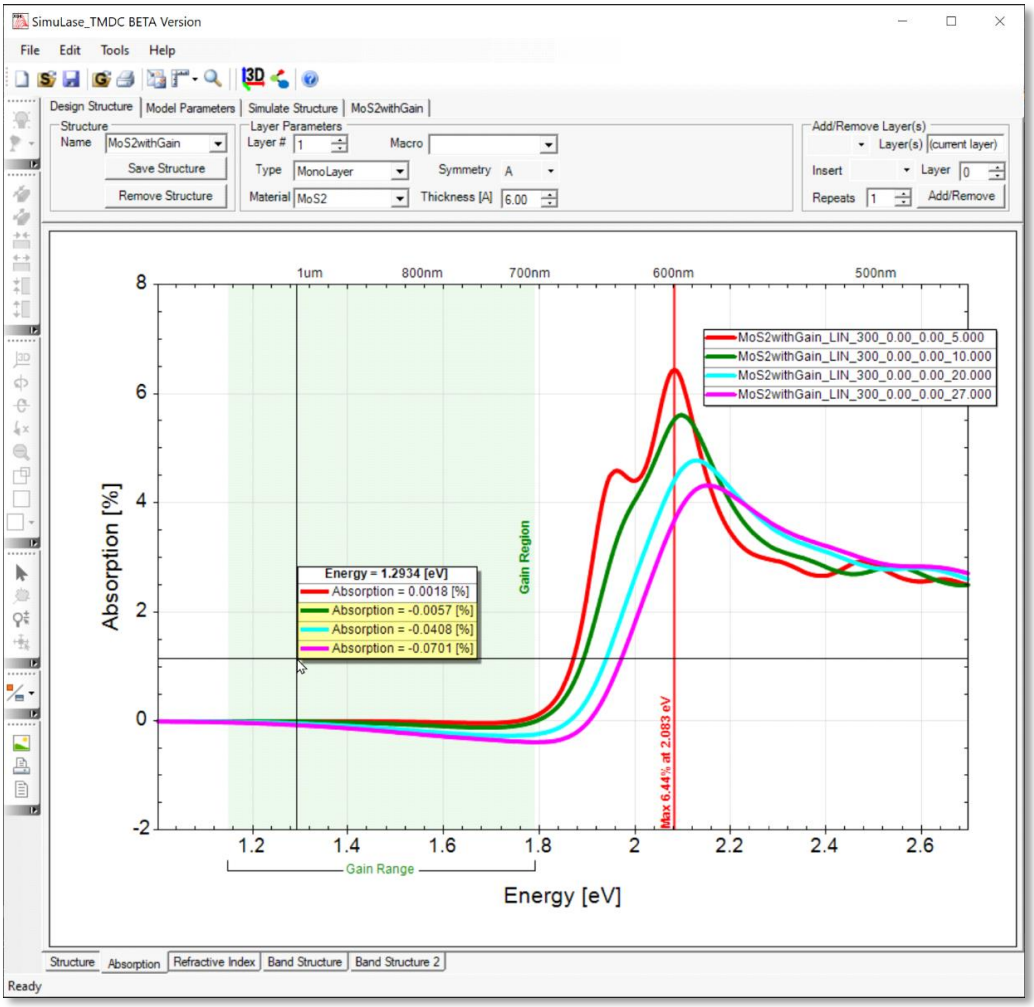


Figure 22 The Measure Dataset Value tool in an Absorption chart

Band Structure Chart

The Band Structure chart displays the density and temperature dependent conduction ad valence bands.

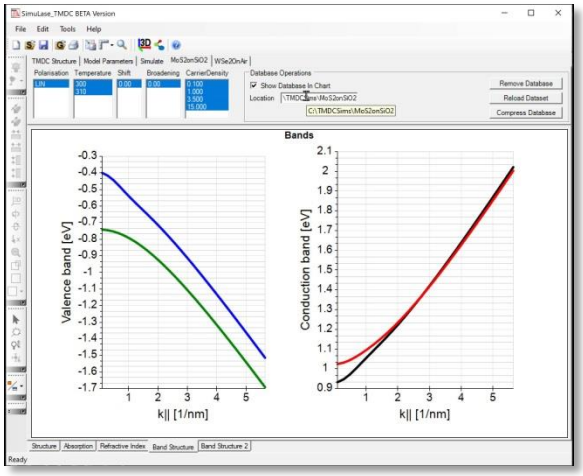


Figure 24 Band Structure chart

Band Structure 2

The Bands Structure 2 chart displays the bands of a database in an alternative form to that of the Band Structure chart.

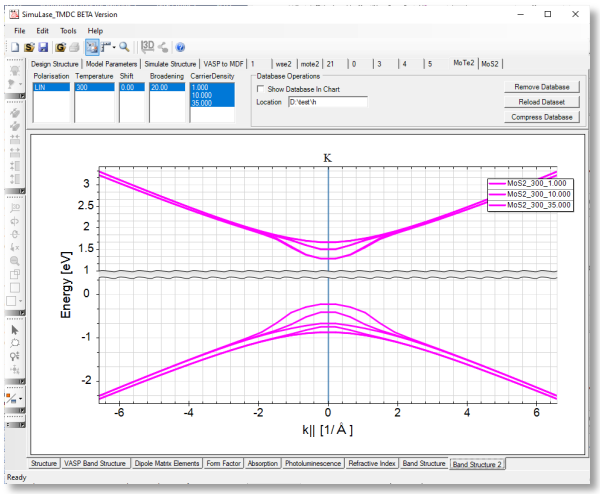


Figure 23 Bands Structure 2 chart

1.4 General Analysis tools

Analysis tools, accessible via the tab **'Tools'** of the main menu, generally operate on the loaded datasets and produce derived quantities usually independent of the device

geometry. Activating any tool will add a new tab to the **Control Group Tabs**, containing all the controls used to interact with the tool, and one or more new chart tabs to the **Data Chart Tabs** to plot the derived quantities.

DFT_to_MDF Tool

The DFT_to_MDF tool is used to determine essential material parameters required for a simulation within the MDF model from the output of a VASP DFT calculation. Once these parameters are combined with some additional parameters as described in main manual, the data can be used to describe a user defined TMDC material using the Material Type *DFT_MDF*.

Selecting the DFT_to_MDF tool from the Tools menu adds the control tab shown in Fig. 26 to the Control Groups Tab. The controls on this tab are used to:

1) Locate a pre-existing folder containing the output from a VASP simulation.

2) Identify a destination folder and name for the file into which to save the derived MDF model parameters.

3) Present a set of numeric controls that enable the user to modify the MDF fit parameters if the user is not fully satisfied with the auto generated fits.

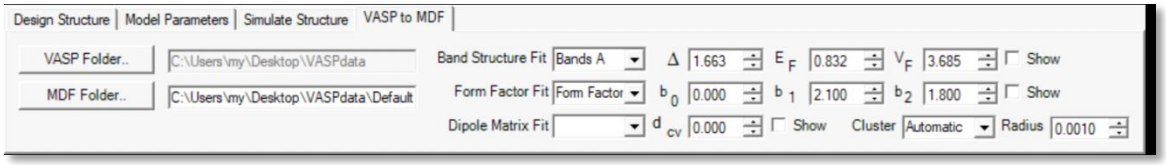


Figure 24 The VASP to MDF Controls

VASP Folder selects the location of the VASP data files on disk. At a minimum the folder must contain the WAVECAR file. By default, the automatically determined values for the MDF parameters are written to a file named *Default_fit.dat* in the VASP folder as specified here.

K Point selects the index of the VASP output momentum grid point that is the K-point of the Brillouin zone. The index '0' indicates the first k-point contained in the VASP output.

Generate Fit starts the determination of the MDF parameters.

Save MDF Data selects the location where the generated MDF parameters will be placed. The MDF name is initially assigned the name ***Default*** but should be changed by the user

when generating a new MDF material. The parameters are written to the file *name_fit.dat* where '*name*' is the name as specified here.

Once a VASP Folder and K-Point have been specified, Generate Fit will initialize the extraction of the electron and hole A- and B-bands from the WAVECAR file and calculate the dipole matrix elements and Coulomb form factors using the wavefunctions as saved in the WAVECAR file. Details of this analysis can be found in the full manual.

Once the evaluation of the VASP data has completed, three new tabs are added to the Data Chart Tabs along the bottom of the graphics window. These tabs display **VASP Band Structure**, **Dipole Matrix Elements** and **Form Factor** plots. The user may overlay plots showing fits to the data according to the MDF model.

In the example shown below the VASP calculation was done for a grid that contained three lines. One going from K halfway to Γ, one going from K halfway to K', and one going from K halfway to Λ.

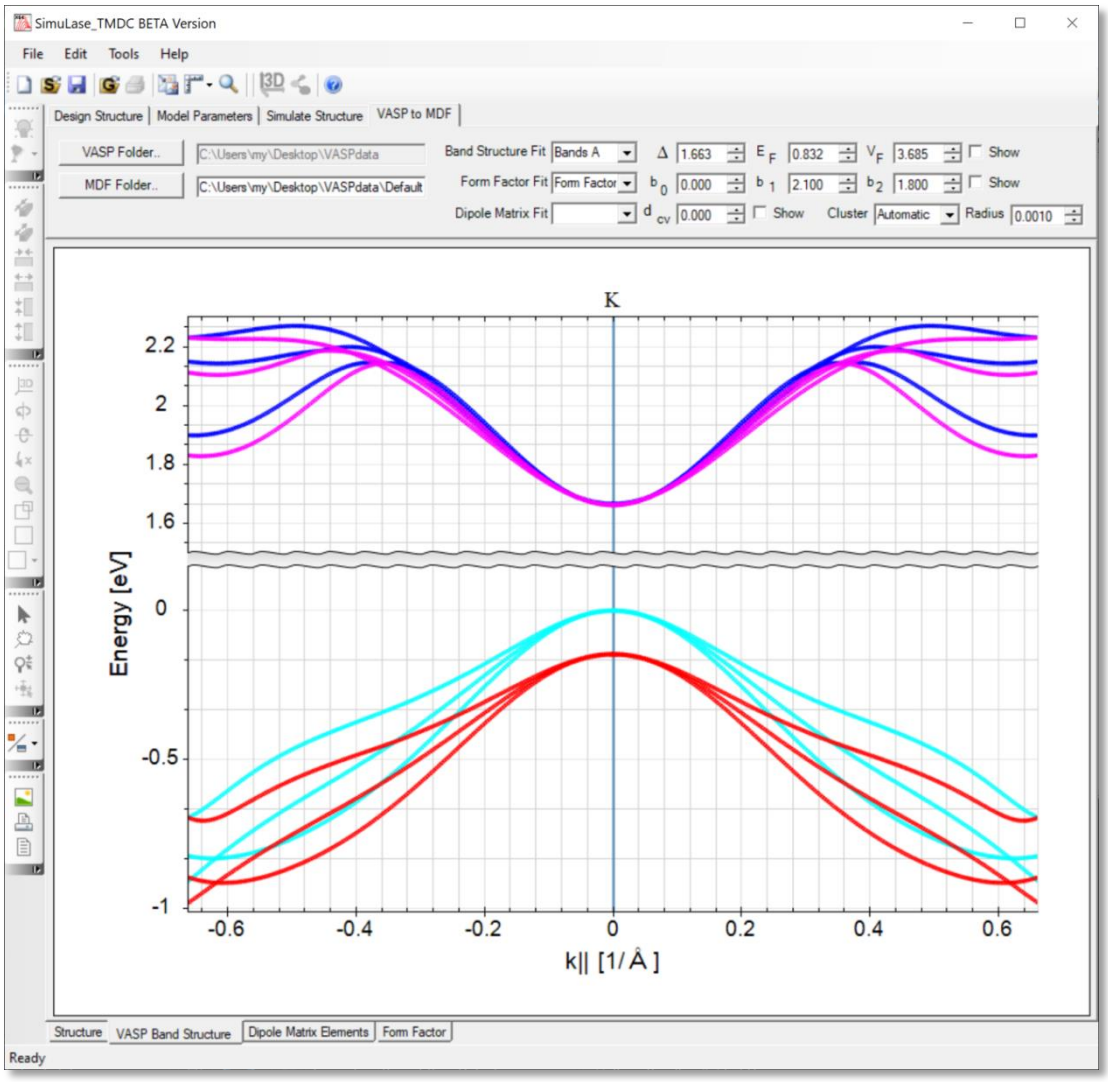


Figure 25 Band structure data imported directly from the VASP.

VASP Band Structure tab displays the band structure plots of the electron and hole A- and B-bands as determined from the VASP output. Additionally, plot showing the auto generated fits to the various conduction and valence bands may be overlaid on the chart. Changing any of the fit parameters will dynamically update the associated fit plot in the chart.

Dipole Matrix Elements tab displays plots of the dipole matrix elements between the A electron and hole band and between the B electron and hole band. The associated MDF fits to that data can be overlaid.

Form Factor tab displays plots of the Coulomb form factors and optionally the associated MDF fits to that data.

Form factors are calculated and displayed for interband transitions, *_mat_ns_cv.dat, as well as for conduction and valence band intra-band transitions, *_mat_ns_cc.dat, *_mat_ns_vv.dat. As described in the main manual, within the model implemented in SimuLase_TMDC the form factor is assumed to be directionally independent and given by an average over inter- and intraband form factors. The fit to the data should try to find an average with respect to the band association and directional dependence.

The rest of controls on the **VASP to MDF** tab enable the user to dynamically view and modify the MDF parameters fits to the VASP data and how the data is displayed on the chart.

Band Structure Fit This line of controls enables the user to select the A or B bands and to modify the fit parameters for the bandgaps Δ, for the Fermi energies, E_F, and for the Fermi velocities, v_F. The **Show** check box is used to show or hide the associated fit curve overlaid on the chart.

Form Factor Fit This line of controls enables the use to select either the Form factor and modify the fit parameters b_1 and b_2. The Show check box is used to Show or Hide the associated fit curve overlaid on the chart.

Dipole Matrix Fit This line of controls enables the use to select either Band A or Band B and modify the fit of the dipole matrix element d_{cv}. As outlined in the main manual, the dipole matrix element is the one at the K-point. The directional and momentum dependence of the dipole matrix elements is considered analytically within the MDF model. Thus, the fit shown here is just a line at the momentum independent value of the matrix element at K. The **Show** check box is used to show or hide the associated fit line on the chart.

In each of the VASP to MDF charts, visual representation of the Band Structure, Dipole Matrix Elements and Form Factor curves may be modified by using the **Cluster** and **Radius** controls. The controls dictate the density of points used to represent the raw data. Reducing the point density highlights the overlaid solid line fit curves.

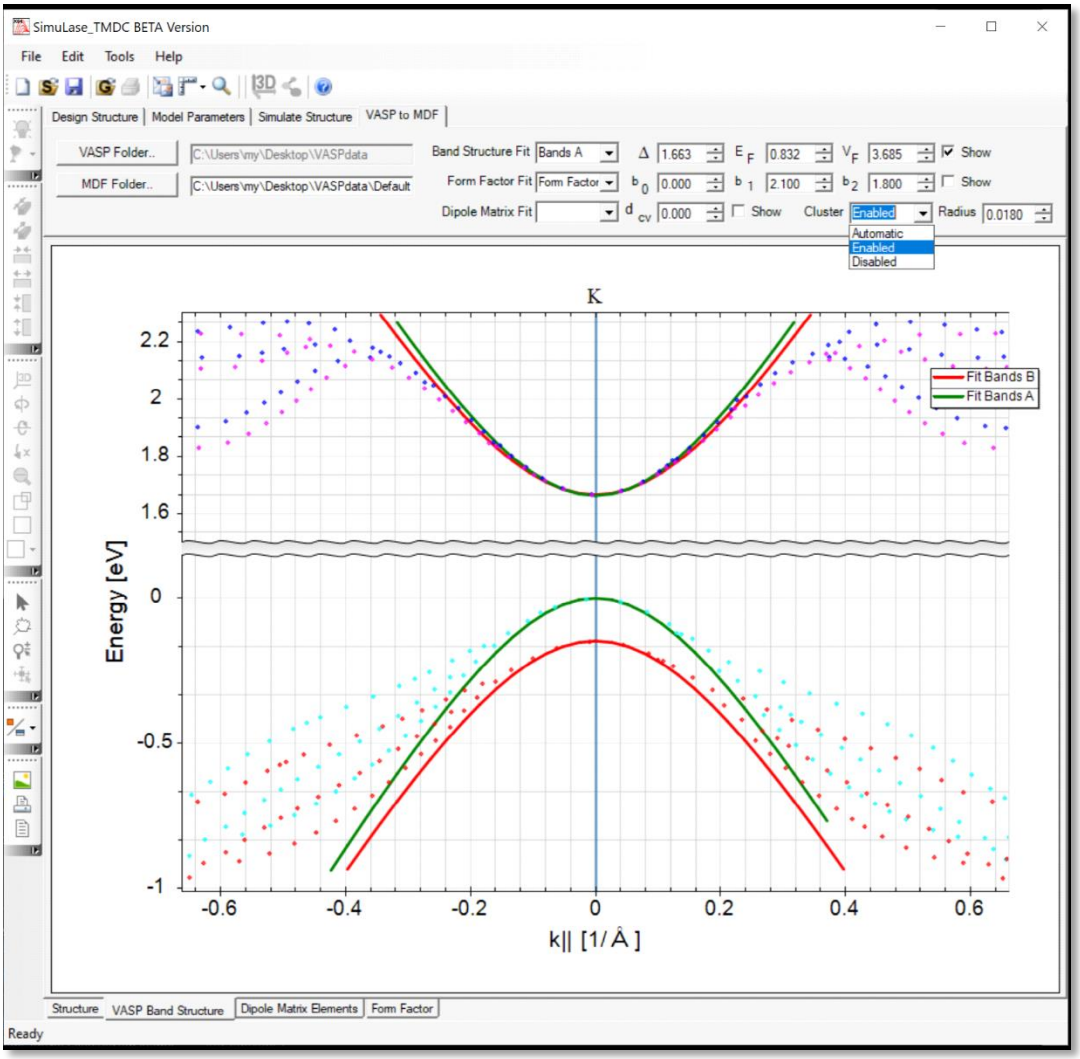


Figure 26 VASP Band Structure plots with overlaid MDF bands.

Cluster determines how many datapoints of the calculated data are displayed in the graphs. This control can be used to reduce the amount of displayed data points in order to be able to see more clearly the fit-lines amongst the symbols representing the VASP data. The clustering mode can be set to three possible modes:

Automatic attempt to render the dataset as a smooth continuous curve without over plotting unnecessary data point.
Enabled allows the user to manually set a density of points used to represent the data set.
Disabled displays a single dot symbol on the chart for every data point in the original.

Radius sets an exclusion radius around the displayed data points. Increasing the radius value reduces the level of detail with which the raw data is plotted. Lower radius values will result in more accurate representation of the data set, at the cost of more points being plotted.

1.5 3D Structure Visualization

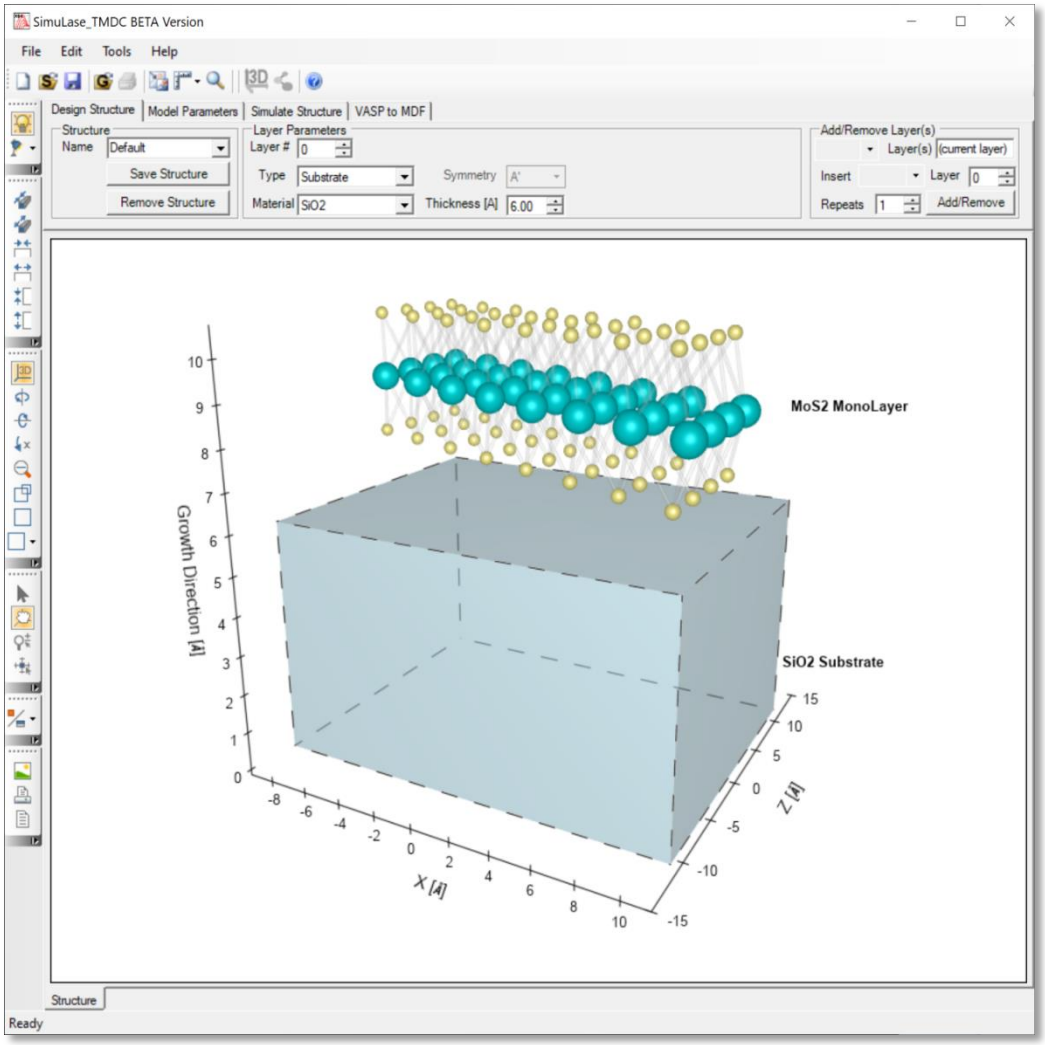


Figure 27 The layered device structure rendered as 3D object.

SimuLase_TMDC possesses the capability to render layered structures as 3D objects with full control over visual aspects such as camera orientation and lighting. The primary use of these 3D capabilities is to produce 3D renderings of TMDC based structures for presentation purposes. The 3D visualizations of the structure have no influence on the numerical simulation of the structures and should be considered a graphical rendering tool only.

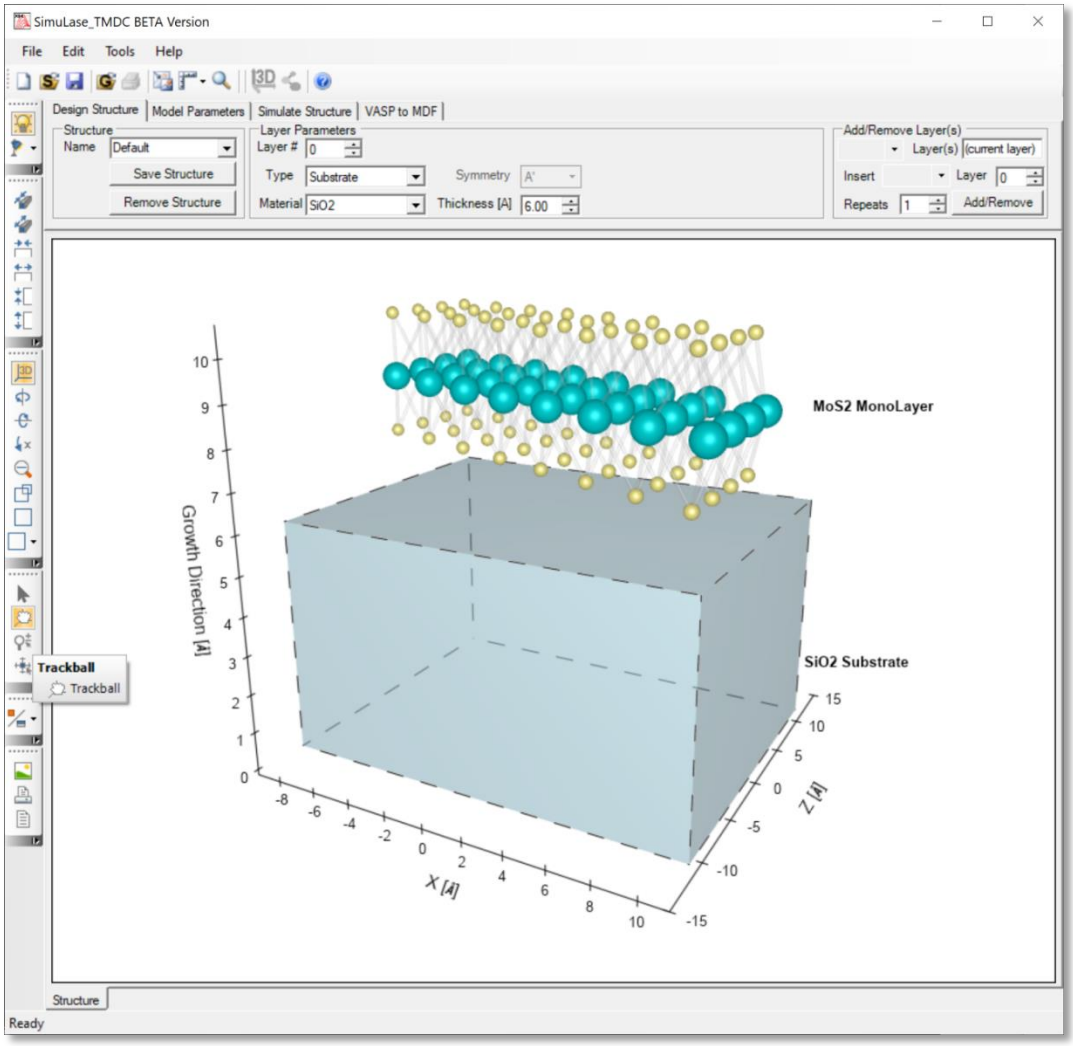


Figure 28 Rotating the structure into the third dimension using the Trackball tool.

Most of the controls related to the 3D aspects of a structure rendering appear as icons in a vertical toolbar on the left-hand side of the main window. Hovering the mouse pointer over an icon will pop-up a tool tip describing the action the icon will perform.

By default, SimuLase_TMDC renders structures in 2D with false shading applied to the ball and stick renderings of TMDC molecules giving them a spherical appearance. A flat unshaded 2D view of the structure is also available and is always applied when outputting graphics to PDF file format.

Activating Full 3D Visualization Mode

To activate full 3D Mode, click on the on the "Trackball" icon, on the vertical tool bar at the left-hand side of the screen, and then drag the mouse anywhere on the 2D structure, causing the 2D rendering to rotate into the third dimension. This simultaneously enables all the 3D manipulation control icons on the vertical toolbar.

To produce a visually more convincing 3D rendering of the structure the user should enable the "Perspective Projection" icon in the vertical 3D toolbar.

3D Structure rendering controls

A brief description of each of the 3D control icons is given below. The most frequently used controls controls are expanded on in the following sections. Controls are grouped according to the operations they perform on the 3D rendering of the structure.

Lighting Controls

Toggle Lighting: Turns and off the virtual light sources illuminating the model
Toggle Lighting: Turns and off the virtual light sources illuminating the model

Chart Export and Printing Controls

Export Chart Image: Export the current chart to file as a raster, (BMP, PNG, JPEG etc.) or vector (SVG) image.
Print: Opens the built-in print manager dialog
Page Setup: Configure size and orientation of the page layout before printing

Structure Aspect Ratio Controls

Decrease Chart Depth: Decreases the 3D depth of the structure rendering
Increase Chart Depth : Increases the 3D depth of the structure rendering
Decrease Chart Width: Decreases the 3D width of the structure rendering
Increase Chart Width: Increases the 3D width of the structure rendering
Decrease Chart Height: Decreases the 3D height of the structure rendering
Increase Chart Height: Increases the 3D height of the structure rendering

Hardware Rendering Controls

Render Surface: Selects if rendering is done on CPU (bitmap) or employs GPU acceleration (window)

Render Surface determines the hardware (CPU or GPU) used to render the 3D image on the screen. The default render surface in SimuLase_TMDC is set to ***Window***, in this mode the chart will leverage installed GPU hardware, which results in much higher rendering speed. An alternative render mode, ***Bitmap*** is also available. When this mode is selected the entire graphics rendering is done using the CPU. This mode is more compatible with a broader range of hardware since it does not depend on a installed GPU and the associated drivers. The most noticeable effect of using bitmap rendering is a reduction in graphic rendering performance and responsiveness when rotating complex structure containing many lattice atoms.

Mouse Interaction Controls

Mouse Select tool: This tool is used to interactively select structure layers via the mouse
Trackball tool : This tool helps you interactively change the camera position to rotate it around the structure rendering
Mouse Zoom tool: This tool helps you interactively zoom in and out the structure rendering
Mouse Offset tool: This tool helps you interactively position the structure rendering within the main window

3D View and Rendering Controls

Toggle 3D: Toggles between a full 3D rendering or 2D flat rendering of the structure
Rotate Right: Rotate structure rendering around the Y-Axis
Elevation Up: Rotate structure rendering around the X-Axis
Viewer Rotation Right: Rotate structure rendering around the X-Axis
Zoom Out: Zoom out of the viewer, decreases the apparent size of the structure rendering
Perspective Projection: Enables the perspective projection view of the structure rendering
View 2D: Enables a predefined 2D orthogonal projection of the structure onto the screen maintaining the 3D shading.
Predefined Projection: List of predefined viewing directions of the structure at various angles and rotations

1.6 Exporting Chart Images and Printing

From the main menu bar the user may select **File | Save Chart As Image...** to export the currently active chart or 3D visualizations of the device structure into many standard raster and vector graphic formats such as JPG, SVG, PNG and PDF, at user selectable resolutions.

From the main menu bar the user may access the Windows printer subsystem, enabling printing capabilities, such as Page Preview, Page Setup and Printer selection etc., for any SimuLase_TMDC chart or 3D rendering of a device structure.

2. Implemented Models

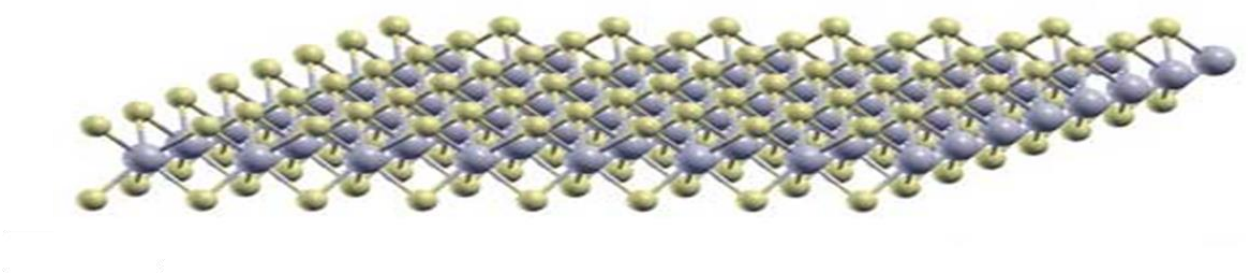


Figure 29 Schematic of a TMDC monolayer with the metal atoms (gray) in the plane and the dichalcogenide atoms (green) out of the plane.

A truly predictive software design tool requires the minimization of adjustable model parameters. The new SimuLase_TMDC software is built on a first-principles comprehensive microscopic physics foundation, namely solution of the Semiconductor Dirac Bloch many-body equations. To extract key optical properties, we only need to compute these in the vicinity of the so-called K and K' symmetry points of the 2D Brillouin zone where optical transitions occur.

The relevant scientific paper on which the software was built is: Ref. [1]: L. Meckbach, J. Hader, et al., PRB **101**, 075401 (2020). It is important to understand the complexity of this model to fully appreciate the achievement involved with porting it to a laptop-based interactive compute environment.

Figure 29 shows a computed 2D single-electron bandstructure output from DFT and a 1D cut through the high symmetry points of the 2D Brillouin zone.

These 2D bandstructure energies and associated dipole moments linking the bands are key inputs to the Semiconductor Dirac-Bloch many-body equations. The above is a simple two-band representation, but, in general, many more valence and conduction bands need to be included. Any external environmental influences, such as substrates or other species, are accounted for by solving a Poisson equation.

2.1 Dirac-Bloch Equations

The computational back-end implemented in the new SimuLase_TMDC software are the Semiconductor Dirac-Bloch equations:

$$i\hbar\frac{d}{dt}p_{\mathbf{k}i} = \left(\varepsilon^e_{\mathbf{k}i} - \varepsilon^h_{\mathbf{k}i}\right)p_{\mathbf{k}i} - \left(1 - f^e_{\mathbf{k}i} - f^h_{\mathbf{k}i}\right)\Omega_{\mathbf{k}i} - i\hbar\left.\frac{d}{dt}p_{\mathbf{k}i}\right|_{\text{corr}}$$

$$\frac{d}{dt}f^{e/h}_{\mathbf{k}i} = -\frac{2}{\hbar}\,\text{Im}\left(\Omega_{\mathbf{k}i}p^*_{\mathbf{k}i}\right) - \left.\frac{d}{dt}f^{e/h}_{\mathbf{k}i}\right|_{\text{corr}}$$

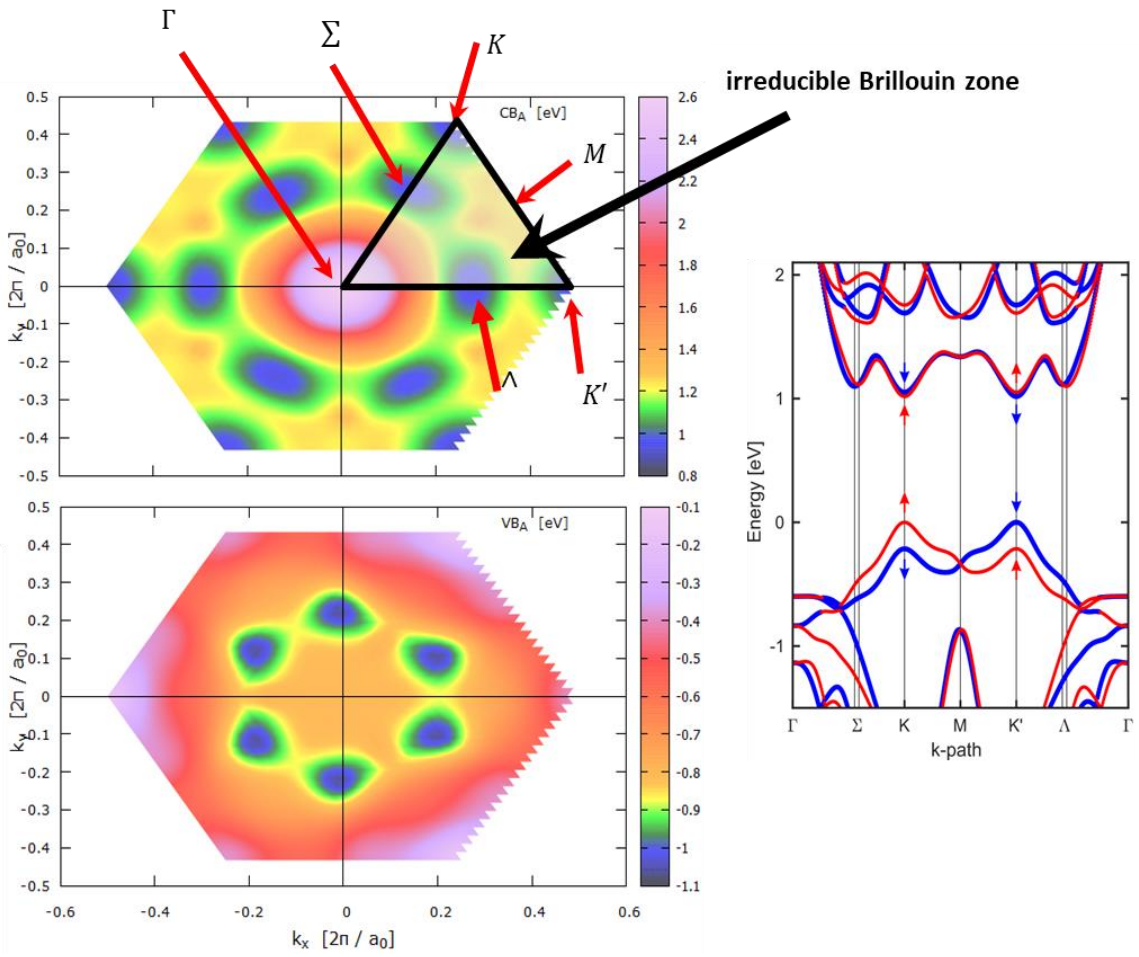


Figure 30. Left: Contour plots of the lowest conduction (top) and highest valence band (bottom) of a monolayer of MoS_2. Right: Bandstructure along Γ, K, M, K', Γ.

where p_k and f_k represent the microscopic polarization and carrier (electron and hole) populations, respectively. These many-body equations look deceptively simply when written in the above compact form. However, their true complexity becomes evident when identifying sources of energy and field renormalization below. These equations are a relativistic generalization of the usual Semiconductor Bloch equations and are approximated at the level of MDF in the vicinity of the K and K' valleys where the relevant direct optical transitions occur in TMDC monolayers.

The energy and field (Rabi frequency) renormalizations in the Dirac-Bloch equations are much more complicated than those in the conventional Semiconductor Bloch equations for 3D semiconductors underpinning our current SimuLase epitaxy design software. The energy renormalization:

$$\varepsilon_{\mathbf{k}i}^{e/h} = \epsilon_{\mathbf{k}i}^{e/h} - \sum_{\mathbf{k}'} \left[\left[V_{\mathbf{k}-\mathbf{k}';\mathbf{k}';\mathbf{k}}^{eeee/hhhh} - V_{\mathbf{k}-\mathbf{k}';\mathbf{k}';\mathbf{k}}^{eheh/hehe} \right] f_{\mathbf{k}'i}^{e/h} + \left[V_{\mathbf{k}-\mathbf{k}';\mathbf{k}';\mathbf{k}}^{ehee/hhhe} p_{\mathbf{k}'i} + c.c. \right] \right]$$

and field renormalization:

$$\Omega_{\mathbf{k}i} = \frac{e}{m_0 c} \mathbf{A} \cdot \boldsymbol{\mu}_{\mathbf{ki}} - \sum_{\mathbf{k}'} \left[V_{\mathbf{k}-\mathbf{k}';\mathbf{k}';\mathbf{k}}^{ehhh} \left(1 - f_{\mathbf{k}i}^{e} - f_{\mathbf{k}i}^{h}\right) - V_{\mathbf{k}-\mathbf{k}';\mathbf{k}';\mathbf{k}}^{ehhe} p_{\mathbf{k}'i} + V_{\mathbf{k}-\mathbf{k}';\mathbf{k}';\mathbf{k}}^{eehh} p_{\mathbf{k}'i}^{*} \right]$$

now contain additional terms involving Coulomb matrix elements of the type $V^{eheh/hehe}$ and $V^{ehee/hhhe}$. These mediate pair creation/annihilation and Auger processes, respectively, that, unlike the regular Hartree terms, do not maintain the number of carriers. Due to the much weaker Coulomb interaction in typical III-V semiconductors, these processes only influence the carrier dynamics at very high densities and, even there, only on a timescale of picoseconds or longer. There, they do not impact the polarization dynamics on a femtosecond timescale that is responsible for optical spectra. Due to the orders of magnitude stronger Coulomb interaction in the 2D materials, these processes must be considered, even for the optical response involving absorption/gain and photoluminescence spectra.

Correlation Terms, Scatterings

The correlation terms, $\frac{d}{dt} p_k \,|_{corr}$ and $\frac{d}{dt} f_k \,|_{corr}$, describe the dephasing of the polarizations and the carrier relaxation due to electron-electron and electron-phonon scattering. Traditionally, these terms were replaced by simple dephasing time constants. The work of the Marburg and Arizona groups including members of NLCSTR, showed that this simple approach leads to fundamentally flawed results, leading to wrong line-shapes, spectral positions, dispersions and amplitudes. Also, it introduces fit parameters required for the dephasing times, which reduces the predictability of the calculations. Instead, it was shown that an explicit calculation of the underlying scattering processes not only removes the need for fit parameters, but also leads to extremely good agreement with experiment, resulting in a truly quantitatively predictive theory. NLCSTR's established SimuLase software for III-V materials was and is the only commercially available software that implements a fully microscopic solution of the scattering processes, which is part of the reason for its commercial success.

A direct numerical implementation of the scattering processes is essential for the results of the Dirac-Bloch equations, as well. Implementing these processes has already been found to be very challenging in the traditional III-V materials. This is the reason why it is typically avoided in commercial software. Due to the exceptional strength of Coulomb interactions which mediate the scatterings in 2D TMDCs, the description and required numerical effort are also significantly

increased here. In III-V materials memory effects can be neglected in the description of the screening of the Coulomb interaction. Here, the so-called static Lindhard formula can be used to describe the screening. Ref. [1] found that the same treatment leads to significant errors in the description of the screening in 2D TMDCs. E.g., such a treatment leads to an unphysical decrease of the linewidth broadening with increasing carrier density despite the availability of more scattering partners. The static screening increases too quickly with density and diminishes the Coulomb interaction faster than the increase of scattering partners.

Dynamic Plasma Screening

For the correct description of the scattering processes in 2D TMDCs memory effects have to be included in the screening as described in Ref [1]. Instead of using the static screening in III-V semiconductors, here a dynamic screening model is required. This strongly increases the numerical effort beyond the already very involved treatment in III-V materials. With this, the screened Coulomb interaction, $W(\omega)$ becomes frequency-dependent in contrast to the frequency independent unscreened interaction V. The screening is described by Dyson's equation [1]:

$$W^{\alpha\alpha'}_{\mathbf{q};\mathbf{k}}(\omega) = \tilde{V}_{\mathbf{q}} + \tilde{V}_{\mathbf{q}}\Pi^{\alpha\alpha'}_{\mathbf{q};\mathbf{k}}(\omega)W^{\alpha\alpha'}_{\mathbf{q};\mathbf{k}}(\omega)$$

$$\Pi^{\alpha\alpha'}_{\mathbf{q};\mathbf{k}}(\omega) = \Pi_{\mathbf{q}}(\omega + (\Sigma_{\alpha\mathbf{k}} - \Sigma_{\alpha'\mathbf{k}-\mathbf{q}})/\hbar),$$

$$\Pi_{\mathbf{q}}(\omega) = \sum_{\beta\mathbf{k}'} \frac{f^{\beta}_{\mathbf{k}'-\mathbf{q}} - f^{\beta}_{\mathbf{k}'}}{\hbar\omega + \Sigma_{\beta\mathbf{k}'-\mathbf{q}} - \Sigma_{\beta\mathbf{k}'}}$$

where $\Pi(\omega)$ is the Lindhard polarization function. For the full electron-electron scattering equations, please see Ref. [1].

Similar to the Coulomb interaction in the electron-electron scattering, the electron-phonon coupling entering the respective scattering becomes frequency dependent:

$$\begin{aligned} \mathrm{i}\hbar\frac{d}{dt}P_{s\mathbf{k}}\Big|^{\mathrm{c,ph.}}_{\mathrm{corr.}} = \mathcal{F}\Bigg[& \sum_{\mathbf{q}} g^{0}_{\mathbf{q}} g^{\mathrm{vc}}_{\mathbf{q};\mathbf{k}} \left\{ \frac{\bar{f}^{\mathrm{c}}_{s\mathbf{k}-\mathbf{q}} n_{\mathbf{q}} + f^{\mathrm{c}}_{s\mathbf{k}-\mathbf{q}}(1+n_{\mathbf{q}})}{\hbar(\omega+\omega_{\mathbf{q}}) + \tilde{\Sigma}^{\mathrm{v}}_{s\mathbf{k}} - \tilde{\Sigma}^{\mathrm{c}}_{s\mathbf{k}-\mathbf{q}} + \mathrm{i}\eta} + \frac{\bar{f}^{\mathrm{c}}_{s\mathbf{k}-\mathbf{q}}(1+n_{\mathbf{q}}) + f^{\mathrm{c}}_{s\mathbf{k}-\mathbf{q}} n_{\mathbf{q}}}{\hbar(\omega-\omega_{\mathbf{q}}) + \tilde{\Sigma}^{\mathrm{v}}_{s\mathbf{k}} - \tilde{\Sigma}^{\mathrm{c}}_{s\mathbf{k}-\mathbf{q}} + \mathrm{i}\eta} \right\} P_{s\mathbf{k}} \\ & - \sum_{\mathbf{q}} g^{0}_{\mathbf{q}} g^{\mathrm{vc}}_{\mathbf{q};\mathbf{k}+\mathbf{q}} \left\{ \frac{\bar{f}^{\mathrm{c}}_{s\mathbf{k}} n_{\mathbf{q}} + f^{\mathrm{c}}_{s\mathbf{k}}(1+n_{\mathbf{q}})}{\hbar(\omega+\omega_{\mathbf{q}}) + \tilde{\Sigma}^{\mathrm{v}}_{s\mathbf{k}+\mathbf{q}} - \tilde{\Sigma}^{\mathrm{c}}_{s\mathbf{k}} + \mathrm{i}\eta} + \frac{\bar{f}^{\mathrm{c}}_{s\mathbf{k}}(1+n_{\mathbf{q}}) + f^{\mathrm{c}}_{s\mathbf{k}} n_{\mathbf{q}}}{\hbar(\omega-\omega_{\mathbf{q}}) + \tilde{\Sigma}^{\mathrm{v}}_{s\mathbf{k}+\mathbf{q}} - \tilde{\Sigma}^{\mathrm{c}}_{s\mathbf{k}} + \mathrm{i}\eta} \right\} P_{s\mathbf{k}+\mathbf{q}} \Bigg], \end{aligned}$$

with the screened phonon coupling matrix elements:

$$g^{\alpha\alpha'}_{\mathbf{q},\mathbf{k}}(\omega) = g^{0}_{\mathbf{q}} + g^{0}_{\mathbf{q}}\Pi^{\alpha\alpha'}_{\mathbf{q},\mathbf{k}}(\omega)g^{\alpha\alpha'}_{\mathbf{q},\mathbf{k}}(\omega)$$

While the semiconductor Bloch equations can effectively be solved by time integration—which is how they are solved in the traditional SimuLase software—the frequency dependence of the coupling matrix elements favors a solution of the Dirac-Bloch equations in frequency space via matrix inversion. Such a solution has been implemented in SimuLase_TMDC.

Dielectric Screening

The matrix elements $\tilde{V}_q$ appearing in Dyson's equation for the plasma-screened Coulomb interaction are the ones for the Coulomb interaction in the absence of excited carriers. They differ from the ideal 2D Coulomb matrix elements, V_q, through the fact that they include the dielectric screening due to the finite out-of-plane layer extension and potential neighboring TMDC layers, substrates, and coatings. This dielectric screening is calculated by solving Poisson's equation, as described in Ref. [2].

Spontaneous Emission (Photoluminescence, PL) Spectra

The spontaneous emission (photoluminescence) is a critical means of optical characterization of these quasi-2D materials. It yields insights about crystal structure quality, composition, and potential performance in a rather simple and non-destructive manner. It also yields the radiative carrier lifetime, which is obtained from a spectral integral of the emission. The latter is an important loss mechanism that determines the quantum efficiency of optoelectronic devices.

To solve this problem, we need to augment the above Semiconductor Dirac-Bloch system with the following quantum Semiconductor Dirac-Luminescence equations [4]:

$$\frac{d}{dt}P_{\mathbf{k}}^{ji} = \frac{1}{i\hbar}\left\{\sum_{i',j'}\left[\mathcal{E}_{jj',\mathbf{k}}^{h}\delta_{ii'} + \mathcal{E}_{ii',\mathbf{k}}^{e}\delta_{jj'}\right]P_{\mathbf{k}}^{j'i'} + \left[f_{\mathbf{k}}^{e,i}f_{\mathbf{k}}^{h,j}\right]\mathcal{U}_{i,j,\mathbf{k}} - E(t)\,X_{i,j,\mathbf{k}}\right\}$$

which are equations of motion for photon assisted polarizations. Formally, these equations are very similar to the Semiconductor-Dirac Bloch equations—except for replacing the inversion $(1 - f^e - f^h)$ by the product $(f^e f^h)$. However, they contain additional source terms, $X_{i,j,k}$, that are absent in the equations of motion for the optical polarizations. These terms represent higher excitonic correlations beyond the Hartree-Fock level. Numerically, they are on the same level as the electron-electron scattering terms. Except for SimuLase, these terms have been neglected in all commercial software so far. While they modify the spontaneous emission and resulting carrier lifetimes only moderately in III-V materials, they are essential for the correct

description of these properties in the 2D TMDC materials due to the much stronger Coulomb interaction there.

Figure 3 shows an example for spontaneous emission spectra for a monolayer $MoTe_2$, calculated with and without the excitonic source terms. Without these terms, the emission is underestimated by about one order of magnitude. In addition, the line-shape is wrong and unphysical negative emission occurs. The resulting radiative carrier lifetime would be too long by an order of magnitude.

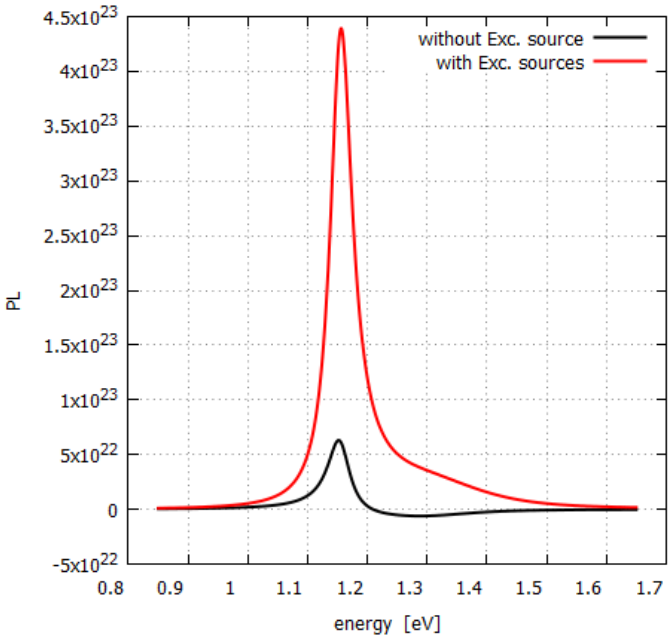


Figure 31 Spontaneous emission from a monolayer $MoTe_2$. Black/red: calculated with/without higher excitonic correlation source terms.

Besides the excitonic source terms, the solution of the Dirac-Luminescence Equations involves similar electron-electron and electron-phonon scattering terms and dynamic treatment of the screening as the Dirac-Bloch equations outlined above.

2.2 Massive Fermi Dirac (MDF) Approach

Instead of integrating the full Brillouin bandstructure as derived through DFT, SimuLase_TMDC adopts the so-called MDF model. The Dirac-Fermion model had been used to describe Graphene. As with TMDCs, the Graphene crystal structure is hexagonal with the bandgap at the three degenerate K and K' points. However, since the Graphene crystal is built exclusively of

Carbon atoms, there is no bandgap. In the vicinity of the K/K' points, the bandstructure is linear, resulting in quasi-particles that are massless Dirac-Fermions. Since the TMDC crystal consists of two different materials, the bandstructure has a finite gap which leads to Massive Dirac-Fermions that can be described by the MDF model.

In this model, the bandstructure is given by:

$$\varepsilon_N^{e/h}(k) = E_N^F \pm \frac{1}{2}\sqrt{\Delta_N^2 + 4\, v_N^{F\,2}\, \hbar^2 k^2},$$

where N is A or B and denotes the first and second lowest electron and highest and second highest hole bands. Here $\Delta_{A/B}$ are the bandgaps between A and B electron and hole bands at the K-point, $E_{A/B}^F$ are the Fermi energies, and $\hbar v_{A/B}^F$ the Fermi velocities for the A and B bands. These parameters are obtained by fitting the bandstructure, as derived from DFT.

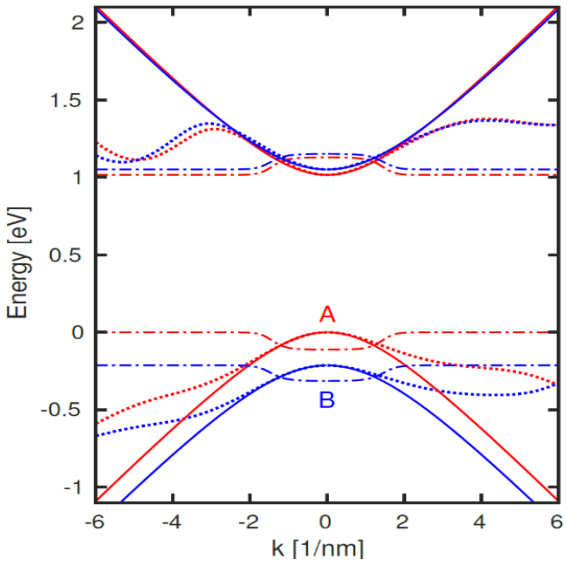


Figure 32 Spin up A (red) and spin down B (blue) bands near K. Solid lines: MDF Dotted: DFT data. Dash-dotted: carrier occupations in each band for a carrier density in the high gain regime.

Figure 31 shows DFT bandstructure together with the MDF fit for the example of a monolayer of $MoTe_2$. As can be seen there, the MDF model describes the DFT bandstructure well within momentum and energy range relevant for optical transitions. Even under high carrier excitation conditions as in the limit of very high optical gain, all carriers occupy states that are well represented by the MDF model. As shown in Figure 2, bands other than the A and B band are energetically very remote in typical TMDC materials and can be omitted.

A marked advantage of the MDF model is that relevant matrix elements can be determined analytically, and the directional dependence can be considered through simple pre-factors, as

described in Ref. [2]. The momentum dependence of the MDF dipole matrix elements is given by:

$$d_{A/B}^{cv} = e\sqrt{2}\hbar v_{A/B}^{F} \,/\, \Delta_{A/B}.$$

Coulomb Form Factor

Generally, the Coulomb matrix elements take the form:

$$V_{q_{||},k_{||},k'_{||}}^{\alpha\beta\beta'\alpha'} = F_{k_{||},k'_{||}}^{\alpha\beta\beta'\alpha'}(q_{||})\, V_{q_{||}}^{2D},$$

with

$$F_{k_{||},k'_{||}}^{\alpha\beta\beta'\alpha'}(q_{||}) = \int_{uc} d^3r \int_{uc} d^3r'\, u_{k_{||}-q_{||}}^{\alpha *}(r) u_{k'_{||}+q_{||}}^{\beta *}(r')\, e^{-q_{||}\,|z-z'|}\, u_{k'_{||}}^{\beta'}(r') u_{k_{||}}^{\alpha'}(r')$$

is the so-called form factor. It describes the deviation of the Coulomb potential, from the ideal 2D case, $V_{q_{||}}^{2D} = 1/q_{||}$, due to the finite out-of-plane extension of the TMDC layers [3]. Within the MDF model as implemented in SimuLase_TMDC, the Form factor is approximated by:

$$\tilde{F}(q_{||}) \approx e^{-b_1 q_{||} - b_2 q_{||}^2}$$

with fit-parameters, $b_{1,2}$, determined from DFT calculations.

Interface to DFT Data for User-Defined TMDC Materials

NLCTR developed and implemented routines to post-process DFT output data in order to calculate the form factors and the dipole matrix elements directly from the Kohn-Sham wavefunctions. These routines allow the user to post-process their own material DFT data and find fits within the GUI for the relevant MDF parameters. This allows the user to implement new materials beyond these in the current version of SimuLase_TMDC. A description of the details of this DFT-to-MDF procedure and the underlying tools can be found in the SimuLase_ DFT_to_MDF guide.

2.3 Example Data

Comparison between MDF and Green's function GW Results

Density functional theory alone, being an effective single-particle method, cannot account for excitons in any semiconducting material. As excitons are dominant spectral features at room temperature, they can be accounted for via the so-called GW Green's function approach, which accounts for two-body correlations. The GW approach has been used extensively in the literature to compute TMDC excitons.

NLCSTR compared excitonic bandgaps as calculated with the many-body correlated MDF model with the single particle (Kohn Sham DFT) gaps and published GW-correlated gaps for five different monolayer TMDCs. The results are summarized in Table 1.

	$E_{g,s.p.}$	$E_{g,cor.,MDF}$	$E_{g,\ cor.,GW}$
MoS_2	1.682	2.467	2.82[1], 2.54[2], 2.66[3], 2.90[4], 2.41[5]
$MoSe_2$	1.410	2.112	2.41[1], 2.12[2], 2.31[3], 2.60[4]
$MoTe_2$	1.017	1.585	1.77[1], 1.77[3]
WS_2	1.626	2.410	2.88[1], 2.53[2], 2.91[3], 2.81[4]
WSe_2	1.385	2.072	2.42[1], 2.51[3], 2.40[4]

Table 1 Comparison of bandgaps in monolayer TMDC materials suspended in air. $E_{g,s.p}$ are single particle gaps. $E_{g,\ cor.,MDF}$ are the correlated gaps within MDF. $E_{g,\ cor.,\ GW}$ are the correlated gaps from the GW method in various literature sources.

Table 2 shows a comparison between the exciton binding energies for the same monolayers from the MDF many-body approach and the GW approach.

	$E_{b,MDF}$	$E_{b,GW}$
MoS_2	0.456	1.04[5], 0.74[8], 0.90[10]
$MoSe_2$	0.431	0.91[5], 0.81[8]
$MoTe_2$	0.365	0.71[5]
WS_2	0.428	1.04[5], 0.61[8]
WSe_2	0.392	0.91[5], 0.62[8], 0.59[11]

Table 2 Exciton binding energies in monolayer TMDCs suspended in air. $E_{b,MDF}$ are binding energies calculated within the MDF model. $E_{b,GW}$ are calculated in the literature using the GW method.

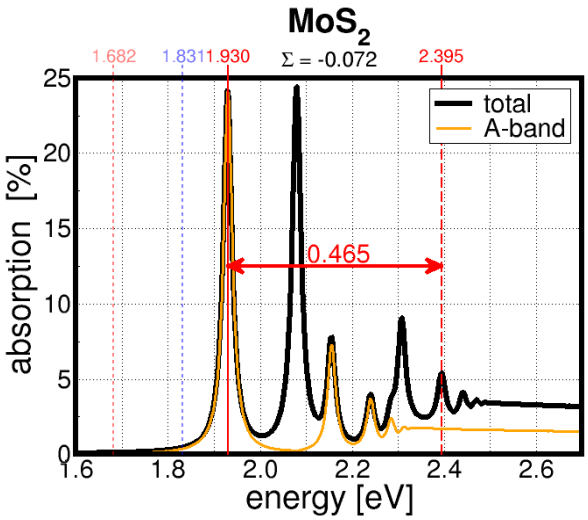


Figure 33 Absorption of a monolayer of MoS2 suspended in air. Black: total absorption. Orange: A-bands only. The A-band exciton is found 0.465 eV below the correlated A band gap.

Within the MDF approach, the binding energies were determined by calculating absorption spectra and measuring the energetic distance between the 1s A-exciton and the correlated bandgap. Figure 32 shows an example of the absorption for a monolayer MoS_2.

Here, the total absorption is shown together with just the absorption from transitions between the A electron and hole band. The latter allows for an easier analysis of the A-band excitons. The red dashed line marks the correlated bandgap. The dotted red and blue lines are the un-correlated single particle A- and B-bandgap from DFT. These are corrected by a scissor shift, Σ, for better agreement between experimental and calculated spectral positions.

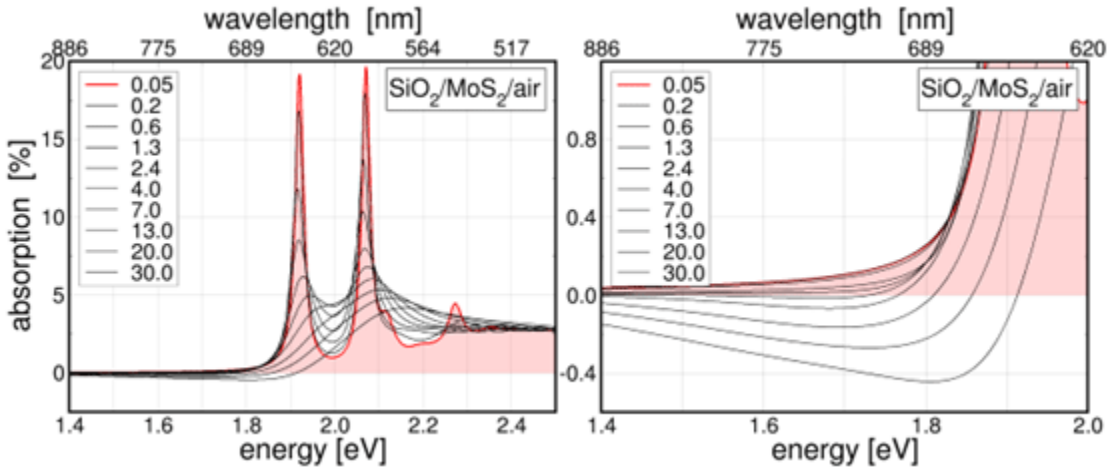


Figure 34 Absorption of a monolayer of MoS_2 for various carrier densities [1012/cm2] calculated using the MDF model.

While the published results of earlier GW calculations show strong variations between different sources, more recent calculations [private communications] agree well with the numbers of the MDF model. The latter have also been tested against various experimental sources.

Fig. 33 shows a representative example of absorption and gain spectra computed from the Semiconductor Dirac-Bloch equations for a monolayer of MoS_2 on a SiO_2 substrate for different carrier densities.

Prominent exciton absorption features at low densities of incoherent carriers are gradually suppressed with increasing density and switch over to gain at the highest values. In Figure 33, the right graph is a zoom into a region of the top the graph to display the gain region of the spectra.

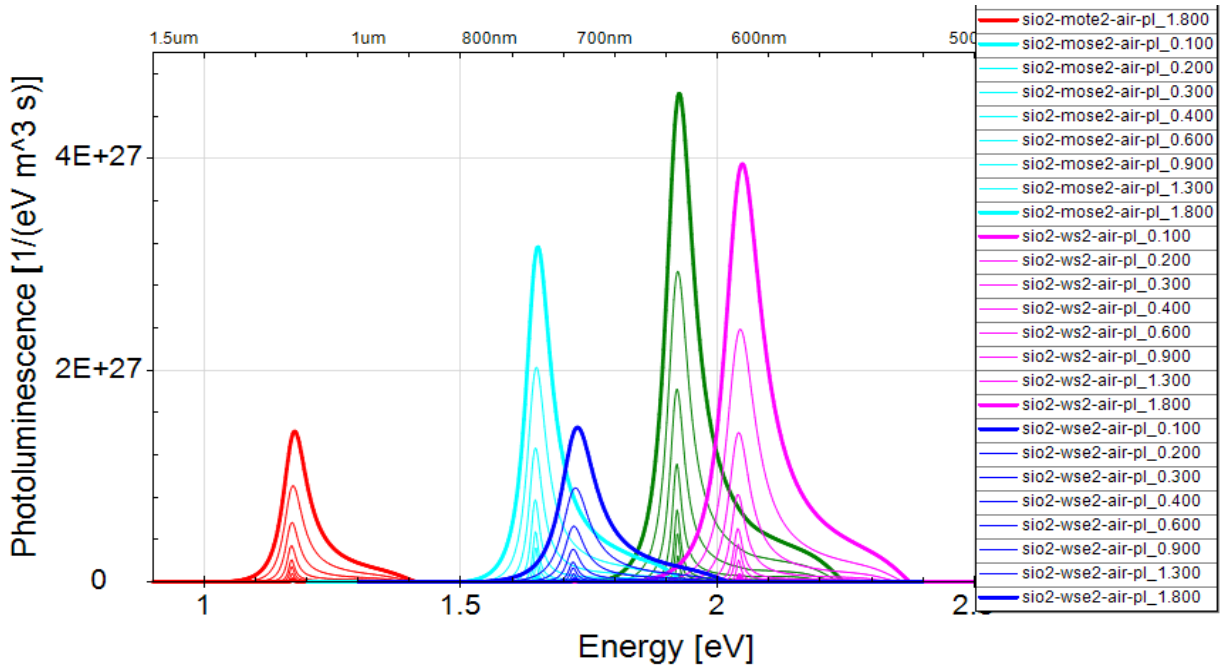


Figure 35 PL of TMDC monolayers on SiO_2 substrate. Red: $MoTe_2$. Turquois: $MoSe_2$. Blue: WSe_2. Green: MoS_2. Magenta: WS_2.

While the excitonic resonances are diminished with increasing density due to screening of the Coulomb interaction and excitation induced dephasing, they remain spectrally close to the original position of the A-exciton. This demonstrates a complicated balance between the spectral blue-shift induced by the reduction of the binding energy and the red-shift due to the density dependent bandgap renormalization. It would not be possible to correctly obtain this complicated balance without the correct treatment of all Coulomb correlations, including carrier scatterings and the dynamic screening.

Figure 34 shows the families of spontaneous emission (photoluminescence, PL) spectra computed from the Semiconductor Dirac-Luminescence Equations for different material systems and carrier densities of 0.1, 0.2, 0.3, 0.4, 0.6, 0.9, 1.3 and 1.8 x $10^{12}/cm^2$. All data is for 300 K.

3. Material Parameters

3.1 TMDC Materials

The models implemented in SimuLase_TMDC require the following 18 material parameters to describe TMDC monolayers:

- $\Delta_{A/B}$ [eV] The bandgaps between the lowest/second lowest (A/B) electron and hole bands at the K-point.
- $E^F_{A/B}$ [eV] The Fermi energies for the A and B bands.
- $\hbar v^F_{A/B}$ [eÅ] The Fermi velocities for the A and B bands.
- $d^{cv}_{A/B}$ [eÅ] The dipole matrix elements for the A and B bands at K.
- b_1 [Å] Parameter entering the form factor part of the Coulomb interaction.
- b_2 [Å^2] Parameter entering the form factor part of the Coulomb interaction.
- E^S [eV] The so-called scissor operator that often is required to adjust the bandgap as calculated in DFT to agree with an experimentally known bandgap. This energy is added to the single particle energies of all conduction band states.
- d^L [Å] The bulk inter-layer distance, i.e., the out-of-plane lattice constant.
- $\epsilon^B_{||}$ [1] The (off-resonant) bulk in-plane dielectric constant.
- $\epsilon^B_{\perp}$ [1] The (off-resonant) bulk out-of-plane dielectric constant.
- A^{uc} [Å^2] The area of the unit cell.
- E_{LO} [meV] LO-phonon energy near K.
- g_{LO} [meV] LO-phonon coupling constant near K.
- r_{eff} [Å] Effective screening length.

The values implemented in SimuLase_TMDC for these parameters for monolayer TMDC materials are summarized in Table 3. Values for d^{cv}_A are derived from the parameters for the Fermi velocities and bandgaps as described in [18]. Values for the scissor operator were obtained by comparing results based on the DFT data as derived within the work of Ref. [12] to the experimental results published in the references as indicated in the table.

	MoS_2	$MoSe_2$	$MoTe_2$	WS_2	WSe_2
Δ_A [eV], [3]	1.682	1.410	1.017	1.626	1.385
Δ_B [eV], [3]	1.831	1.614	1.266	2.022	1.801
E^F_A [eV], [3]	0.000	0.000	0.000	0.000	0.000
E^F_B [eV], [3]	-0.0715	-0.0810	-0.0895	-0.2300	-0.2520
$\hbar v^F_A$ [eVÅ], [3]	3.532	3.027	2.526	4.433	3.941
$\hbar v^F_B$ [eVÅ], [3]	3.467	3.001	2.574	4.208	3.757
d^{cv}_A [eÅ]	2.9697	3.0361	3.5126	3.8556	4.0242

$\boldsymbol{d_B^{cv}}$ [eÅ]	2.6778	2.6300	2.8754	2.9432	2.9502
$\boldsymbol{b_1}$ [Å], [12]	2.10	2.06	1.41	2.46	2.42
$\boldsymbol{b_2}$ [Å^2], [12]	1.82	2.21	5.21	1.01	1.45
$\boldsymbol{E^S}$ [eV], [12]	-0.0715[14]	-0.029[15]	-0.044[14]	0.080[16]	0.050[17]
$\boldsymbol{d^L}$ [Å], [3]	6.18	6.52	6.99	6.22	6.51
$\boldsymbol{\epsilon_{||}^B}$ [1], [3]	11.65	12.81	15.43	10.56	11.81
$\boldsymbol{\epsilon_{\perp}^B}$ [1], [3]	6.38	7.81	10.90	5.93	7.65
$\boldsymbol{A^{uc}}$ [Å^2]	8.758	9.546	10.976	8.758	9.488
$\mathbf{E_{LO}}$ [meV], [13]	46.333	34.406	27.720	42.886	29.682
$\mathbf{g_{LO}}$ [meV], [13]	355.0	521.0	819.0	165.0	323.0
$\mathbf{r_{eff}}$ [Å], [13]	46.5	53.2	69.5	42.0	48.7

Table 3 Material parameters used in SimuLase_TMDC for monolayer TMDC materials.

In the case of hetero bi-layers one has to solve the Dirac-Bloch equations for the intra-layer cases where the electron and hole are in the same layer as well as for the inter-layer case where the electron is in one layer and the hole in the other [18]. In addition to the band-energies within each layer one also needs the hetero-offset between the top of the valence band in one layer versus in the other layer. For this case one has to run a DFT calculation for a system including both layers. Then one has to check which bands are located in which layer by checking the relative ion contribution to the corresponding states as described in Ref. [18]. The ion contribution information is written by VASP into the file PROCAR which is created when the switch LORBIT is enabled. For intra-layer part of the calculations the A and B bands used in the MDF calculations are the lowest electron and highest hole bands located within the layer. For the inter-layer calculation, the A and B electron-/ hole-bands are the overall lowest/highest conduction/valence bands. Thus, a total of four electron and hole bands and corresponding MDF parameters are required here instead of the two each for the monolayer case. Also, inter-layer dipole matrix elements are required in addition to the intra-layer ones for monolayers.

NLCSTR will implement an adaptation of the ***DFT_to_MDF*** tool and '*dft_mdf_fit.dat*' file for custom hetero-bi-layer materials in Version 1.1 of *SimuLase_TMDC* or as soon as customer demand is voiced.

Tables 4 and 5 list the MDF parameters as required for the hetero-bi-layer materials currently implemented in *SimuLase_TMDC*. For parameters not listed there the same values are used as for the monolayers of the respective material.

Similar to the hetero-bi-layer case, homo-mono-layer simulations also require additional material parameters. While the band energies are the same as in the monolayer case, additional inter-layer dipole matrix elements are required. As for the hetero-bi-layer case, NLCSTR will implement an adaptation of the ***DFT_to_MDF*** tool and '*dft_mdf_fit.dat*' file for

custom homo-multi-layer materials in Version 1.1 of *SimuLase_TMDC* or as soon as customer demand is voiced.

	MoS_2 /MoS_2	WSe_2/WSe_2	MoS_2/WSe_2	WSe_2/MoS_2
Δ_A [eV], [18]	1.456	2.013	0.835	2.634
Δ_B [eV], [18]	1.609	1.688	0.409	2.888
$\boldsymbol{E_A^F}$ [eV], [18]	-0.322	0.578		
$\boldsymbol{E_B^F}$ [eV], [18]	-0.396	0.844		
$\hbar\boldsymbol{v_A^F}$ [eVÅ], [18]	3.645	3.658		
$\hbar\boldsymbol{v_B^F}$ [eVÅ], [18]	3.607	4.142		
$\boldsymbol{d_A^{cv}}$ [eÅ]	3.54	2.57	0.42	0.14
$\boldsymbol{d_B^{cv}}$ [eÅ]	3.17	3.47	0.46	0.08

Table 4 Material parameters used in SimuLase_TMDC for the hetero-bi-layer system MoS_2/WSe_2. A and B refer here to spin-up and spin-down instead of the usual energetically lowest and second lowest bands since the lowest electron states in MoS_2 and WSe_2 have opposite spins.

	$MoSe_2$ /$MoSe_2$	WSe_2/WSe_2	$MoSe_2$/WSe_2	WSe_2/$MoSe_2$
Δ_A [eV], [18]	1.407	1.824	1.577	1.654
Δ_B [eV], [18]	1.633	1.393	1.117	1.909
$\boldsymbol{E_A^F}$ [eV], [18]	0.394	0.432		
$\boldsymbol{E_B^F}$ [eV], [18]	0.301	0.697		
$\hbar\boldsymbol{v_A^F}$ [eVÅ], [18]	3.084	3.676		
$\hbar\boldsymbol{v_B^F}$ [eVÅ], [18]	3.233	4.048		
$\boldsymbol{d_A^{cv}}$ [eÅ]	3.10	2.85	0.94	1.06
$\boldsymbol{d_B^{cv}}$ [eÅ]	2.80	4.11	0.48	0.33

Table 5 Material parameters used in SimuLase_TMDC for the hetero-bi-layer system $MoSe_2$/WSe_2.

3.2 TMDC Materials

Metals are only included in the calculations of the dielectric screening of the Coulomb interaction through the substrate. Thus, the only relevant material parameters are the refractive indices, n_s, and dielectric constants, $\varepsilon_{S.}$, of the material.
Currently implemented metals are: Silver, Gold, Titanium and Chromium. The material parameters are taken from Ref. [19]. NLCSTR will implement additional metals on demand for free with user-provided information for the parameter values.

Refractive indices and dielectric constants for dielectric coatings are derived from various literature sources and private communications. For the refractive indices the wavelength- and temperature-dependence is taken into account. Due to a lack of information, for the dielectric constants only a constant value is implemented.
Currently SiO2, SiC, Y_2O_3, TiO_2, Ta_2O_5, Al_2O_3, Si_3N_4, and hBN are implemented dielectric coatings. NLCSTR will implement additional coatings on demand for free with user-provided information for the parameter values.

4. Coupling SimuLase_TMDC with User-Generated DFT Data

The ***DFT_to_MDF*** tool allows the user to determine the essential MDF input parameters from first principles DFT output. The tool is set up to expect the DFT data as written by VASP to the WAVECAR output file. The parameters that will be extracted are:

- $\Delta_{A/B}$ [eV] The bandgaps between the lowest/second lowest (A/B) electron and hole bands at the K-point.
- $E^F_{A/B}$ [eV] The Fermi energies for the A and B bands.
- $\hbar v^F_{A/B}$ [eÅ] The Fermi velocities for the A and B bands.
- $d^{cv}_{A/B}$ [eÅ] The dipole matrix elements for the A and B bands at K.
- b_1 [Å] Parameter entering the form factor part of the Coulomb interaction.
- b_2 [Å^2] Parameter entering the form factor part of the Coulomb interaction.

The corresponding DFT data for the materials that are implemented in SimuLase_TMDC by NLCSTR have been derived as described in Refs. [3,18]. Here, a k-grid that starts from the K-point and goes into several directions of high symmetry was chosen to set up the data in order to see possible directional dependence of the parameters. About 100 points were used to resolve each direction in a line spanning from K to about half-way to K', Γ, and Λ, respectively.

The only strict requirement of the tool on the k-grid is that it has to contain the K-point and at least one line of equidistantly spaced grid-points. The user has to specify the index of the K-point within the grid through the field 'K Point' starting with 0 for the first grid-point. If the data contains several directions, only the index of the first occurrence is required.

MDF Fit Parameters Derived from DFT

The MDF fit of the DFT bandstructure for electrons/holes is given by:

$$\varepsilon^{e/h}_N(k) = E^F_N \pm \frac{1}{2}\sqrt{\Delta^2_N + 4\, v^{F\,2}_N\, \hbar^2 k^2}, \qquad (V1)$$

where N is A or B.

The interband dipole matrix elements, d^{cv}, are the matrix elements at $q_{||} = K$. Within the MDF model these matrix elements are connected to the Fermi velocities and bandgaps via [2,18,20]:

$$d^{cv}_{A/B} = e\sqrt{2}\hbar v^F_{A/B} \,/\, \Delta_{A/B}, \qquad (V2)$$

And the $q_{||}$-dependence is included through angular dependent prefactors as described in [3]. ***DFT_to_MDF*** uses formula (V2) to derive the dipole matrix element at K and displays those values as a k-independent line in the dipole fit plot window. This window also shows the momentum dependent dipole matrix elements as calculated directly from the DFT wavefunctions. The latter have been tested to agree with the values calculated internally by VASP and written to the file WAVEDERF if the option LOPTICS=.TRUE. has been used to set up the data. By default, SimuLase_TMDC uses the values as derived by (V2). However, the user can opt to adjust the values in order to agree with the independently calculated values.

The parameters $b_1$and b_1 are used in the description of the Coulomb matrix elements as described in Ref. [3]:

$$V^{\alpha\beta\beta'\alpha'}_{q_{||},k_{||},k'_{||}} = F^{\alpha\beta\beta'\alpha'}_{k_{||},k'_{||}}(q_{||})\, V^{2D}_{q_{||}}, \qquad (V2)$$

$$F^{\alpha\beta\beta'\alpha'}_{k_{||},k'_{||}}(q_{||}) = \int_{uc} d^3r \int_{uc} d^3r'\; u^{\alpha*}_{k_{||}-q_{||}}(r)\, u^{\beta*}_{k'_{||}+q_{||}}(r')$$

$$\times\; e^{-q_{||}\,|z-z'|}\, u^{\beta'}_{k'_{||}}(r') u^{\alpha'}_{k_{||}}(r') \qquad \text{(V3)}$$

$$\tilde{F}(q_{||}) \approx e^{-b_1 q_{||} - b_2 q_{||}^2}. \qquad (V4)$$

Here, the form factor, $F(q_{||})$, describes the deviation of the Coulomb potential, V, from the one in the ideal 2D case, V^{2D}, due to the finite out-of-plane extension of the TMDC layers [3]. The ideal 2D Coulomb matrix elements, $V^{2D}_{q_{||}}$, are simply given by $1/q_{||}$. The directional dependence is contained within the wavefunctions, u^i, and, thus, the form factor. Within the approach introduced and successfully tested in Ref. [3], the form factor is approximated by Eq. (V4). Here the directional dependence is considered outside the approximate form factor, $\tilde{F}$, through angular dependent MDF prefactors as described in Refs. [2,20]. An angle-averaged value is assumed for the form factor. Ref. [3] found that $\tilde{F}$ can be approximated by expression (V4) and that the same form factor can be used for all Coulomb matrix elements, those describing interband interactions between electron and hole bands as well as those describing intraband interactions within the hole- and electron-bands, respectively.

4.2 Defining a New Material from VASP Output Using DFT_to_MDF

The ***DFT_to_MDF*** tool determines the essential bandstructure, dipole and Coulomb parameters as listed in Sec. 4.1. By default, the data is written to a file named '*Default_fit.dat*' in the same directory that contained the VASP data and was specified through the field '*VASP Folder*'. If the

user modifies the default fit values he can save the modified values to a separate folder and filename as specified through the field '*MDF Folder*'.

The file contains the data mentioned in Sec. 4.1. For simulations within the MDF model several additional material parameters are required:

- E^S [eV] The so-called scissor operator that often is required to adjust the bandgap as calculated in DFT to agree with an experimentally known bandgap. This energy is added to the single particle energies of all conduction band states.
- d^L [Å] The bulk inter-layer distance, i.e., the out-of-plane lattice constant.
- $\epsilon_{||}^B$ [1] The (off-resonant) bulk in-plane dielectric constant.
- $\epsilon_{\perp}^B$ [1] The (off-resonant) bulk out-of-plane dielectric constant.
- A^{uc} [Å^2] The area of the unit cell.
- E_{LO} [meV] LO-phonon energy near K.
- g_{LO} [meV] LO-phonon coupling constant near K.
- r_{eff} [Å] Effective screening length.

The last four parameters are required to describe the electron-phonon coupling within the two-dimensional Fröhlich interaction model as described in Ref. [13]. Values for all parameters in this list have to be derived by *ab initio* calculations.

These eight parameters have to be written into a file named '*dft_mdf_fit.dat*' below the ten parameters that the ***DFT_to_MDF*** tool writes into the '*Default_fit.dat*' file or, if the user modified the default values, the file that the user specified for these parameters through the '*Save MDF Data*' option. The '*dft_mdf_fit.dat*' file then has to be placed in the folder into which the simulation data shall be written. The latter is specified once the '*Simulate*' button is clicked on the '*Simulate Structure*' panel.

We recommend copying the file that is generated through the ***DFT_to_MDF*** tool to the simulation output folder. After that, one should rename the file to '*dft_mdf_fit.dat*' and then concatenate the eight additional parameters in the order as shown above to the file. An example for a complete '*dft_mdf_fit.dat*' file can be found in the folder in which SimuLase_TMDC has been installed.

Once the file '*dft_mdf_fit.dat*' has been put in the correct place, one has to select the type '*VASP_MDF*' for the layer. SimuLase_TMDC will then automatically look for the parameter file and use the parameters within to describe the TMDC material.

Parameter values for the materials that are readily implemented in SimuLase_TMDC can be found in Sec. 3.

5. File and Data Formats

SimuLase_TMDC creates a variety of files containing data that comprises a GainDatabase like absorption/gain, PL and refractive index spectra, radiative losses, and bandstructures. All files are in ASCII-format in order to allow to investigate the data also outside the SimuLase_TMDC environment. The following describes the labeling of the files and the formatting of the data.

The names of all files belonging to a GainDatabase start with a common string, which is set through the file manager that opens once the button '*Simulate*' is clicked to start the calculation of a database. The files are located in the directory as specified through the same file manager dialog.

In the following description of the files the abbreviation:
• '**name'** stands for the GainDatabase name.
• '**PP'** stands for the polarization, with 'LIN' for linear polarized excitation and 'LC' and 'RC' for left circular and right circular, respectively.
• '**TTT'** stands for the temperature in Kelvin. An integer is used and all digits after the decimal point are dropped.
• '**s.ss**' stands for a spectral shift of the spectra in meV with percent accuracy.
• '**b.bb**' stands for the inhomogeneous broadening in meV (FWHM) with percent accuracy.
• '**N.NNN**' stands for the sheet carrier density in 10^{12}cm^2 with per mille accuracy.

The following files are created:

• **name_PP_TTT_im_s.ss_b.bb_N.NNN** : (Modal) absorption/gain spectra. The first column gives the energy in [eV]. The second column gives the total absorption, α. The third and fourth columns give the absorption of A-, and B-band transitions only. α is the fraction of light that is absorbed in one pass through the structure. The field intensity is scaled by $(1-\alpha)$ upon one pass through the structure with intensity increase for negative absorption, i.e., optical gain. 'im' in the file name stands for 'IMaginary part of the optical susceptibility'.

• **name_PP_TTT_re_s.ss_b.bb_N.NNN**: Carrier induced refractive index spectra. The first column gives the energy in [eV]. The second column gives the total carrier induced refractive index. The third and fourth columns give the index contribution from A- and B-band transitions only. The units are chosen to be the same as for the absorption in the 'im' files. These units have been chosen such that the linewidth enhancement factor (α -factor) can be derived by simply dividing the difference between the refractive index for two carrier densities by the corresponding difference between the absorption/gain without need for rescaling. In order to get from the refractive index spectra in these units, $b(\omega)$, to the dimensionless electronic contribution to the refractive index, $\delta_n(\omega)$, one has to use the conversion: $\delta_n(\omega) = \frac{c}{2L\omega}\, b(\omega)$. Here, L is the layer thickness in [m], c is the speed of light in vacuum in [m/s] and ω is the angular frequency of the transition energy in [1/s] ($\varepsilon = \hbar\omega\ /e$, where ε is the energy in [eV] as used in the file and e is the elementary charge).

In order to get to the total refractive index for a density N, one has to add $(\delta_n(N) - \delta_n(N_0))$ to the background refractive index. Here, N_0 should be zero or the lowest density for which δ_n has been calculated.
're' in the file name stands for 'REal part of the optical susceptibility'.

• **name_PP_TTT_pl_s.ss_b.bb_N.NNN**: Photo luminescence spectra (spontaneous emission). The first column gives the energy in [eV]. The second column gives the total spontaneous emission in [1/(eV s m^3)]. The third and fourth columns give the PL from A- and B-band transitions only. 'pl' in the _le names stands for 'Photo-Luminescence'.

• **name_PP_TTT_tsp**: Radiative carrier lifetimes. The first column gives the sheet carrier density in [10^{12}/cm^2]. The second gives the corresponding lifetime in [sec].

• **name_TTT_c_N.NNN**: Conduction bands. The first column gives the in-plane momentum in [1/nm]. The (n+1)'st column gives the energy of the n'th electron band in [eV].

• **name_TTT_v_N.NNN**: Valance bands. The first column gives the in-plane momentum in [1/nm]. The (n+1)'st column gives the energy of the n'th hole band in [eV].

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