VESTA & DFT Workshop: GaAs Models

Materials Theory and Modeling Group

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Contents

0 Software

0.1 VESTA

Visit the following link to download the VESTA program:
http://www.jp-minerals.org/vesta/en/download.html

Scroll the page to **"Latest stable versions"** and select the correct version for your computer. After completing the download, run the installer and follow the prompts to complete installation. Then, launch the application.

The following figure shows the main VESTA window with color coded sections that will be referred to later.

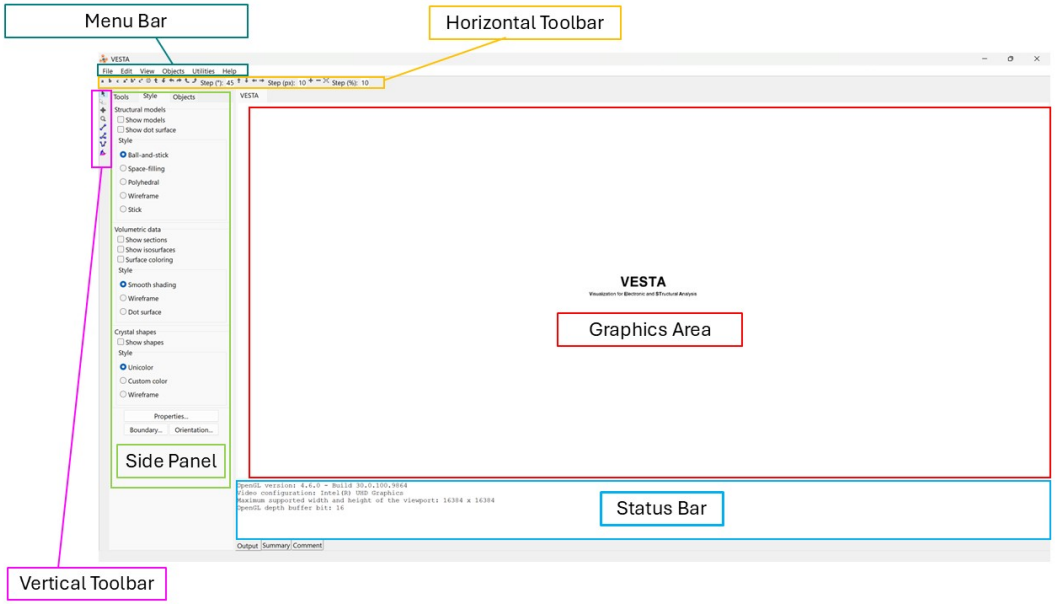

Figure 1: Major regions of the main VESTA window, color coded as menu bar, horizontal bar, vertical bar, side panel, graphics area, and status bar.

0.2 nanoHUB and SIESTA

Visit the following link to sign up for or login to nanoHUB:
https://nanohub.org/register/

(See the top right corner of the page.)

After logging in, hover your cursor over "Explore" near the top of the page, then click "Tools". In the search bar, enter: "MIT Atomic-Scale Modeling Toolkit" and then click on the top result. Alternatively, visit the link:
https://nanohub.org/resources/ucb_compnano

This is the MIT Atomic-Scale Modeling Toolkit. Launch the tool to make sure it works for you. It may take a moment to load.

To get to SIESTA, the DFT software we'll be demonstrating today, use the "Application" drop-down menu that appears next to the MIT logo. Select the entry "DFT for Solids, Nanostructures, and Molecules (SIESTA)".

0.3 Docker and Quantum ESPRESSO

Visit the following link to download and install Docker for free:
https://www.docker.com/

Click "Download Docker Desktop" and choose the correct version for your computer.

Afterwards, follow the prompts to install Docker and make sure it's running after the installation is complete.

Visit this GitHub link to download the files necessary to run our example calculation:
https://github.com/kerbyic/QuantumESPRESSO-Docker-GaAs

These files have instructions for Docker to compile Quantum ESPRESSO and run it all in one command.

Click on the green button that says "Code". In the drop-down menu, click "Download ZIP" and save it to your computer. Extract the ZIP to a location on your computer.

When you are ready to run, you will need to use a terminal window ("Terminal" on Mac, "Command Prompt" or "PowerShell " on Windows) and navigate to the folder where you extracted the ZIP. If you list the files (enter "`ls`" in Terminal and PowerShell or "`dir`" in Command Prompt), you should see:

- `data/`
- `qe/`
- `compose.yml`

If this is the first time running the command, enter:
"`docker compose up --build`"

This will take some time to build and then run the calculation.

If you have successfully run this in the past, you can enter:
"`docker compose up --no-build`"

This will skip building the image again and run the calculation.

When the calculation is finished, the output files will be in the "`data/`" folder. Take a look at "`GaAs.png`" for the band structure plot.

(If you would like to adjust the number of cores used in compilation, you can change the variable "`BUILD_THREADS`" in the "`.env`" file. If you would like to change the number of cores/threads used in running the calculation, you can change the "`NCORE`" variable in "`data/run.sh`".)

1 Bulk Crystal

We'll begin with creating a pure, crystalline, GaAs structure to learn some of the basics of structure generation using the utilities within VESTA.

1.1 New Structure

To start creating anything within VESTA, we need to create a new structure by clicking on "File > New Structure...". in the menu bar. This action will open a new window titled "New Data" and prompt you for information.

1.2 Phase

There are five information tabs. You are started on "Phase" by default. In VESTA, a "phase" is best thought of as a "layer" in a graphics editing program (like Photoshop), each with its own separately defined structure.

We will only be using one structure. **To rename the default structure, triple-click on its title to edit it. Rename it to "GaAs".**

1.3 Unit Cell

Change to the "Unit Cell" tab by clicking on it. The window will change to prompt you for symmetry information and lattice parameters. All structures can be represented with the default P1 space group, however, by using symmetry we can create our structure far more efficiently.

Using the symmetry data in section 5.1, choose the corresponding space group. We can filter our space group options by selecting "Cubic" under "System". (Some space groups will have multiple options available in "Setting", however, here there is only one choice.)

Next, we will input our lattice parameters. Notice that only the lattice parameter a can be edited. This is because our symmetry applies constraints to possible values. **Change a to the correct length using the lattice info in section 5.1.** Lattice parameters b and c will be updated automatically. Leave the second field "s.u." as 0.0.

At this point, our graphics area in VESTA is still blank. **Let's see our unit cell by clicking "OK" at the bottom of the new window.** The window will close, returning us to the main VESTA window. You'll see a compass and a box appear in the graphics area. You can rotate your view by left-clicking and dragging within the graphics area.

Save your progress at this point. Use "File > Save As..." in the menu bar and choose a location. Name this file "GaAs.vesta".

1.4 Structure Parameters

We need to continue editing the data. **In the menu bar, click "Edit > Edit Data > Structure parameters..." to re-open the last window to the "Structure parameters" tab.** You will see an empty table where we can add atoms to our structure. **Add an atom by clicking "New" to the right of the table.**

A row will appear, highlighted grey, and the fields above the table become enabled. We only need to edit two of these fields for now. Let's enter the data for the first atom in the atomic positions table in section 5.1. **Select the element by clicking on "Symbol..." or typing its abbreviation in place of "XX". Change "Label" to be the same. Then, enter the position coordinates in *x*, *y*, and *z*.** This is the minimum required to create an atom in our structure. *(You can enter the coordinates with fractions. They will automatically be converted to decimals.)*

Let's see the changes you made here in the graphics area; click "Apply" at the bottom of the window. *(Do not click "OK" yet, which will close the window.)* Multiple atoms will appear, rather than only one. This is because VESTA automatically adds atoms to the equivalent positions according to the symmetry we defined. **Now, add all the remaining atoms in the atomic positions table in section 5.1.** After creating all the atoms, you can click "OK" to apply your changes and close the window.

Save your progress at this point.

1.5 Bonds

You will see the complete structure in the graphics area, but there will not be any bonds shown. Let's fix this by adding the bond data. **In the menu bar, click "Edit > Bonds...".** A new window will open, allowing us to add our bond data. VESTA uses a pair searching algorithm to determine where to draw bonds. We'll need to tell it what to look for. **Add a new row to the empty table by clicking "New" to the right of the table.**

Bonds will be drawn between atoms matching "Atom 1" and "Atom 2". **By default, the entry for "Atom 1" and "Atom 2" will be gallium. Let's change "Atom 2" to arsenic by using the drop-down menu next to "A2", above the table.** Then, we need to change the maximum length for our bond. To determine a good value for this, let's move our attention to the main VESTA window. *(You can interact with both windows at the same time.)* **In the vertical toolbar, at the left of the main window, select the "distance" tool (5th icon from the top). Measure the distance between an arsenic and gallium atom pair by left-clicking on them sequentially (good atoms to use will be** [0 0 0] **and** [1/4 1/4 1/4]**).**

(When you select the distance tool, you will not be able to rotate by clicking and dragging in the graphics area. To rotate again, you will need to select the "Rotate" tool (1st icon from the top) in the vertical toolbar.)

The measurement will be displayed in the status bar, in the "Output" tab. The bottom three lines will show details about the distance measurement. **The first of the three will show the measurement; take note of this number. Now, let's return to the bond window and enter a number slightly larger than the measurement in the "Max. length" field,** (2.5 Å **is a good choice).**

Click "Apply" to see the bonds appear. You will also see more atoms beyond the boundary appear, due to the default "Boundary mode" setting. **Change the boundary mode from "Search additional atoms if A1 is included in the boundary" to "Do not search atoms beyond the boundary" to clean up the view a little bit. Click "OK" to apply and close the window again.**

Save your progress at this point.

(If you can't rotate your view in the graphics area, you likely have the distance measurement tool selected. Change back to the rotate tool by clicking the top icon in the vertical bar. Alternatively, you can press 'r' on your keyboard after clicking at least once in the graphics area.)

2 Export Data

At this point, we can export our data for use in a simulation. VESTA supports exporting its data to a variety of formats accepted by other software. Each format may export different information and in different styles. VESTA may ask for additional information when you export, so it's formatted as you expect.

To export, go to the menu bar and select "File > Export Data...". This will open a file browser window, allowing you to choose where to export it, what to name it, and what file extension.

The file extension you select will determine how VESTA writes the data and change any following prompts accordingly.

3 Defects

Before starting this section, let's create a new file to work on. **In the menu bar, go to "File > Save As..." and save this as a new file. Name it "GaAs Defects.vesta".**

3.1 Supercell

Sometimes we need a larger cell than the unit cell to build our models. VESTA provides matrices to make this adjustment. **In the menu bar, click "Edit > Edit Data > Unit cell..." to open the "Edit Data" window. Then click "Transform..." to open the "Unit Cell Transformation" window.** You'll be presented with a 3x3 rotation matrix, P, and a 3x1 origin shift matrix, p. Let's prepare to add a point defect with a concentration of $n_d \approx 2 \times 10^{20}\,\mathrm{cm}^{-3}$ by changing our cell to a 3x3x3 supercell. **Change the rotation matrix to** [3 3 3] **in the diagonal. Then click "OK".** This operation will change the volume of the unit cell, prompting a warning from VESTA. **Click "Yes" to continue.** VESTA will then ask what to do about additional lattice points (atomic positions). **Leave the first option selected, "Add new equivalent positions to a list of symmetry operations", and click "OK". Now, click "Apply" in the "Edit Data" window.**

The graphics area will change to show a larger cell.

(For future projects, you should calculate the minimum size of the supercell from the defect density you are interested in.)

3.2 Breaking Symmetry

Defects change or interrupt the crystalline structure of a material. We will need to remove the symmetry operations we've applied to our model to allow for this, otherwise, defects we introduce may propagate into other, undesirable, locations in the cell.

To break the symmetry, click the "Remove symmetry" button near the bottom of the "Edit Data" window in the "Unit cell" tab. This will remove the space group we have selected and convert equivalent positions to independent sites in the cell.

However, since we increased the size of our cell and used symmetry operations to populate the sites, we need to take another step. The "Remove symmetry" button does not remove these additional symmetry operations, so we have to remove them manually. **Click the "Customize..." button to open the "Equivalent Positions" window.** *(If a warning appears, disregard it by clicking "OK".)* Here, all the symmetry operations are listed under the table "General equivalent positions". **Click "Clear" to remove all but the first (mandatory) row. Then click "OK". Back in the "Edit Data" window, click "Apply", and then change the tab from "Unit Cell" to "Structure parameters".**

You will see many more entries in the table, since all equivalent positions were converted. In the case of GaAs, which has 8 atoms in a unit cell, we should see $8 \times 3 \times 3 \times 3 = 216$ atoms. **Verify you have 216 atoms by scrolling to the bottom row of the table.**

(In the main window, you may notice the status bar update and write ~ 280 *atoms. This number does not reflect atomic sites, but the number of atoms drawn in the graphics area.)*

Save your progress here.

3.3 Point Defect

Now that symmetry will no longer be enforced, we can easily add defects. **Make sure your "Edit Data" window is open and on the "Structure parameters" tab.**

Vacancy

To introduce a vacancy, we need only to remove an atom. To demonstrate, let's remove the atom at [0 0 1/3]. You can read through all the entries in the table until you find the matching row, but an easier way is to use the graphics area. **Rotate your view in the graphics area to see the correct site, then double-click on the atom there (it should highlight yellow). The status bar will update with information about the selected atom, including its number in the table (shown as "`Atom:  #`"). Now, in the "Edit Data" window, scroll the table until the atom with the same "No." is in view. Click on its row, then click on "Delete" to the right of the table. Then, click "Apply" to see the change reflected in the graphics area.** You should notice the site at [0 0 1/3] is now vacant.

Substitution

In the menu bar, go to "File > Open..." to re-open the supercell file, before we introduced the vacancy.

Next, let's substitute an arsenic atom with phosphorus by modifying the arsenic atom at [1/12 1/12 1/12]. **Find the correct atom number using the method described in the vacancy section. Select the correct row, then edit the fields "Symbol..." and "Label" each to 'P'. Then, click "Apply".** You'll see the atom at site [1/12 1/12 1/12] change from arsenic to phosphorus.

(You'll also see bonds around it disappear since we do not have any bond in our bonds table regarding phosphorus. Later, you can change this by using the method described in 1.5).

Interstitial

Finally, let's add an entirely new atom. We'll add a nitrogen interstitial at site [1/6 0 0]. To do this, we'll again follow the procedure described in section 1.4. **Click "New" to the right of the table. Then, for "Symbol" and "Label", enter 'N'. For the positions x, y, and z, enter 1/6, 0, and 0 respectively. Then, click "OK" to close the window and see the change in the graphics area.**

At this point, you can continue any combination of these techniques to create your desired point defects. When your model is complete, you can save it and export it according to section 2.

4 Surface Model

Previously, our model has remained a "bulk" model. However, if we introduce a large enough region of vacuum, we can effectively create a "slab" or "surface" model.

To start off, let's open the unit cell of GaAs we saved at the end of section 1.5. **Open that file by clicking "File > Open..." in the menu bar and selecting it in the file browser window that opens.** Before continue this section, let's create a new file to work on. **In the menu bar, go to "File > Save As..." and save this as a new file. Name it "GaAs Surface.vesta".**

Next, let's create a 3x3x2 supercell and break the symmetry by following the steps in sections 3.1 and 3.2. When creating the supercell, make the rotation matrix diagonal [3 3 2]**.**

(Breaking symmetry is always required when making a surface model.)

4.1 Vacuum

Let's return to the "Edit Data" window through "Edit > Edit Data... > Unit Cell" in the menu bar. Click the "Transform..." button to bring up the "Unit Cell Transformation" window. The previous transformation we applied may still be present in the matrices. **Reset the matrices by clicking "Initialize current matrix". Then, change the diagonal to** [1 1 4] **and click "OK".** At the volume change prompt, **click "Yes" to continue.** The next window will prompt you for how to convert the structure. Since we are creating a slab from our current model, we do not want to convert anything in our structure. **Change the selection to "Do nothing", then click "OK".**

Click "OK" in the "Edit Data" window. In the graphics area, you will see the model change, drawing a rectangular prism long in the $\boldsymbol{c}$ direction. You'll also see a large region of empty space (vacuum) along the same direction. The length of this vacuum region will be $(4-1) \times |\boldsymbol{c}|$, since we resized the cell to $4\boldsymbol{c}$ but still have $1\boldsymbol{c}$ of matter.

Now, save and export your data according to section 2.

(Also, note how you can see matter at the furthest face in the $\boldsymbol{c}$ direction. This is because of the periodic boundary conditions in space groups.)

For other models, use the above principle to create a vacuum layer of any desired depth in any direction. The rotation matrices can accept fractional values, too.

5 Data

5.1 GaAs

Lattice

a	5.75 Å
b	5.75 Å
c	5.75 Å
α	90°
β	90°
γ	90°

Atomic Positions

Atom	x	y	z
Ga	0	0	0
As	$1/4$	$3/4$	$3/4$

Symmetry

Crystal System	Cubic
Lattice System	Cubic
Point Group	$\bar{4}$3m
Space Group	F$\bar{4}$3m
Intl. Symbol	216

Data sourced from The Materials Project:

https://next-gen.materialsproject.org/materials/mp-2534