Supplementary information: New developments in the GDIS simulation package: integration of VASP and USPEX.

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March 17, 2021

The GDIS software[1] is a multipurpose visualization software which aims to provide computational chemists with an easy and fast way to render molecular and periodic systems. As of today, GDIS can read the input and/or output of 28 different sources for a total of about 50 file formats (see Table 1 of the manuscript). GDIS is written in C using GTK and OpenGL for its rendering. This supplementary materials details briefly the function and features of GDIS, not limited to the scope of the main manuscript: VASP and USPEX integration. The basic functionality of GDIS was detailed in the original reference (Ref. 1). The GDIS software must be compiled by users, its released sources are available in a free repository[2]. The latest developments are often available for testing in a separate repository[3].

S1 The Graphical User Interface

The GDIS software Graphical User Interface (GUI) can be divided into 8 different areas, as shown in Fig. S1. While the manuscript empathizes on the new features: the integration of VASP and USPEX to the GDIS software, a brief summary of the GDIS capabilities will be presented for each area. To avoid confusion with the text, all elements that belong to the GUI will be underlined. For example, the Tree view area refers to the tree view area of the GDIS software, as indicated in Fig. S1.

The menu area functions are presented in Table S1. A submenu of the Menu area will be described thereafter by using a path: for example, Tools/Computation/GULP refers to the GULP[5] software GUI that is access through the Computation submenu of the Tools menu. Some of the functions present in the Menu area are also available as a quick icon in the Tool area, for convenience. This area functions are described in the Table S2. Note that some functions, while not directly related to the VASP and USPEX integration, were

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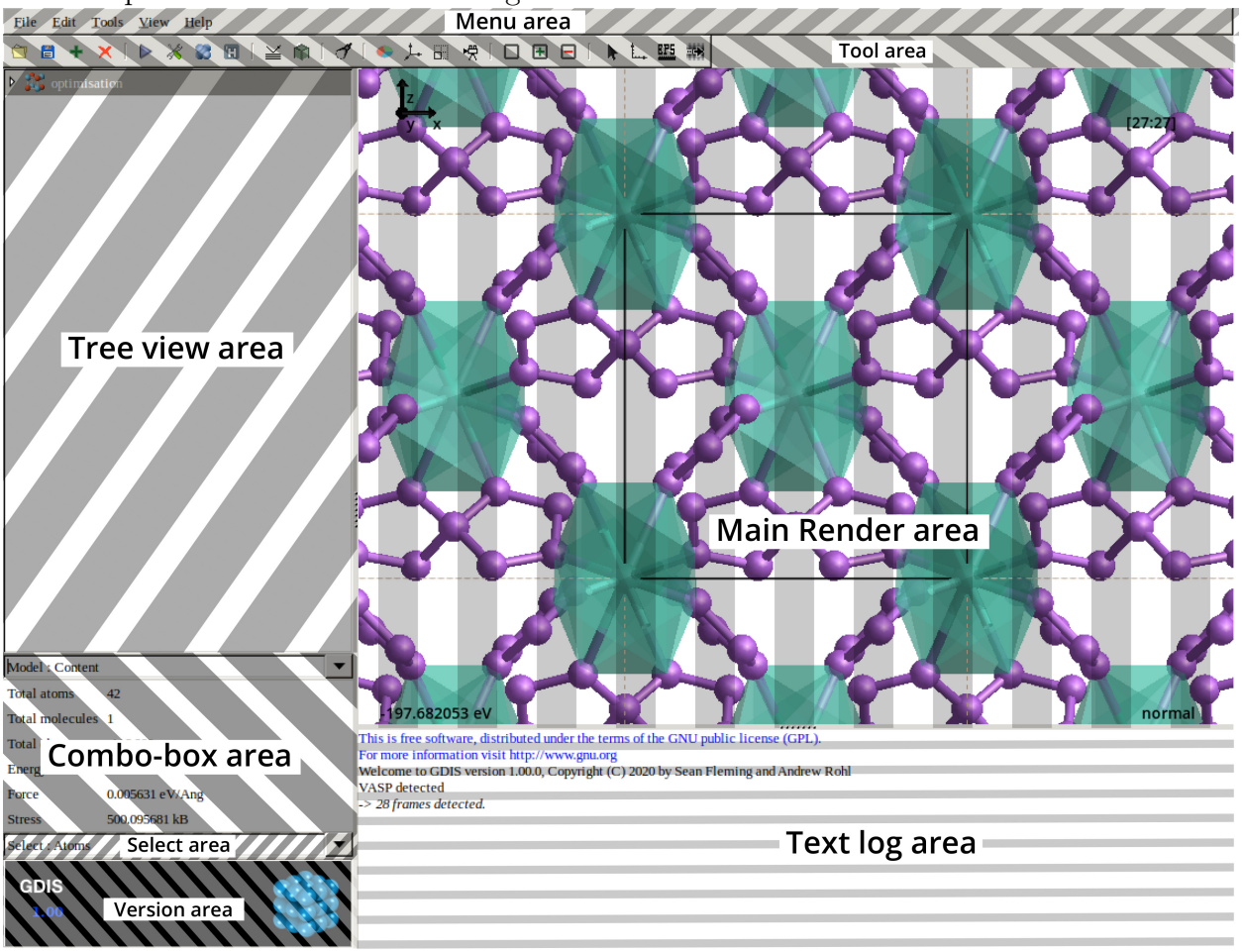
Figure S1: Principal areas of the graphical user interface (GUI) in the GDIS software. Each hatching correspond to a different area of the GUI. Currently displaying is an optimization of the $Sn(N_5)_4$ I-4 molecular crystal at 50 GPa[4], chosen identical to the Figure 1 of the manuscript for a better understanding.



developed to exploit their results. These are the Plot Control, to EPS (or to PNG), and Track mode functions, which are described in the manuscript. The Tree view area contains a list of the loaded models that can be selected for display. This list consists in the name of the loaded model or, if the model name is not available, the default Model_i name (i being the model index) is used. In the left of the model name is an icon that gives an indication about the type of model. The detail of each possible icon is given in the Table S3.

The Combo-box area provides shortcuts to useful functions related to model viewing and editing. The first element, Model: Content display some information gathered by GDIS on the loaded model. For all models, the total number of atoms, molecules, and the total charge of the system are given. This numbers are provided as an indication only. The total energy, force, and stress values are given whenever GDIS can detect them in an input file. For VASP and USPEX, the exact figures displayed on the combo-box are detailed in the manuscript. For other computational software, such as GULP[5], CASTEP[6], Quantum-Espresso[7], and SIESTA[8], other fields are available depending on the calculation. For example, the calcula-

tion type (optimization, PWSCF, CPMD, etc) can be given in the Calculation field. Other important parameters like the exchange-correlation function name, in the Ex-Corr field, and the pseudopotential type in use, in the Pseudopotentials field, are available depending on the computational software. The next element in the combo-box, Model: Editing, displays information about a selected atom or, when several atoms are selected, the position of their centroid. The displayed information can also be modified there. Below these information a group of 4 buttons allows the mouse interaction with the Main render area to be changed into either adding atoms, bonds, or removing bonds. The last button switches back to the default mode of GDIS, which is the selection mode. The bottom part of the Model: Editing element allows to mark atoms as ghosts, GDIS then render them as transparent, or switch them back to the default, non-ghost mode. Note that this feature will only be recorded if the corresponding output file format supports ghost atoms definition. The Model: Display element of the Combo-box area gathers some commonly used render modes from the View/Display properties submenu (or equivalently the Display tool). Note that the Zone based display mode in this element is an experimental feature, mostly intended for development purposes. The Model: Images tool of the combo-box allows to create a supercell of the structure currently selected in the Tree view area. It is done by replicating a cell for a user-defined number of times in either the positive or negative direction of each of the cell axes. The Model: Symmetry tool is a brute-force symmetry analysis that can ascribe a space group to a molecule species, given a GDIS internal tolerance. Finally, the last item of the combo-box area is Model: Viewing which allows to rapidly switch the viewing axis in the main render area to the x, y, z, a, b, or c axis. Note that for a molecular model, the x and a, y and b, and c and z axes are equivalent.

The Select area, just below the previous area, is a combo-box in which elements allow the selection mode of GDIS to be modified. Here, the user can switch between the default selection of atoms in Select: Atoms to more complex selection modes. Selecting all atoms that share the same label, force-field name, or chemical element symbol within the same molecule, molecule-fragment, or predefined region is also possible.

Table S1: Content and description of the functions in the menu area of the GDIS software.

Menu title	Sub-menu	Brief description
File	Open	Open a file from one of the 28 supported sources (see Table 1 of the manuscript, with the addition of VASP and USPEX).
	Save	Save a file in one of the 19 supported sources (see Table 1 of the manuscript).
	Close	The model selected in the Tree view area (current model) will be closed.
	Import	Import a previously exported geometry (Geomview) or graph document. Projects created with the docking tool (see below) can also be imported here.
	Export	Export a geometry file (Geomview) or graph data.
	Quit	Leave GDIS. Note that when a calculation or task is running in the Task manager (see below), an additional confirmation will be required.
Edit	Copy	Copy the selection from the current model. Atom, molecule, or region selections are supported.
	Paste	Paste the previously copied selection into the current model. Note that for the paste operation to succeed, the copy and destination models must be different.
	Delete	Delete a selection from the current model.
	Undo	Undo one of the above copy, paste or delete operation.
	Colour	Allow to change the color of the selected elements, bypassing the internal defaults colors of GDIS.
	Hide	Hide the selected elements. Hiding elements only apply to the rendering process, and such elements are still part of the model and analysis.
	Unhide all	Display all previously hidden elements in the current model.
	Select all	Select all selectable elements the current model.
	Invert	Reverse the previous selection. Unselected elements become selected and the selected ones are unselected.
Tools	Visualization	This submenu gathers some tools related to the rendering of model additional features. The Animation tool is a small GUI for the rendering or recording of the several frames that can be included in a model. The Iso-surfaces submenu allows several isosurfaces types to be rendered on the current model. Finally, the Periodic Table submenu displays a periodic table of the elements in which the user can change each element-related value in use by GDIS.

Table S1 Continued: Content and description of the functions in the menu area of the GDIS software.

Menu title	Sub-menu	Brief description
Tools	Building	These submenus tools are used to modify the current model or create new structures from it. The first submenu, Editing, allows to modify the representation of a model: its lattice can be modified and transformed, some spatial elements (planes, vectors) can be added, and all model editing functions are also available here. The Dislocations tool is a simple GUI for setting a crystal dislocation model using orientation and Burger vectors. The Docking tool allows to setup the docking of a subset selected on the main display of a surface model. The Dynamics tool is used to setup a simple molecular simulation of a solvent (the first loaded molecular model) and a selectable number of solutes molecules. The Surfaces tool allows a surface or set of surfaces to be created from a loaded crystal model. Equilibrium crystal shape can then be calculated. Finally, the Zmatrix submenu is a tool used to generate or modify a Z-matrix.
	Computation	This submenu is used to setup calculations using external simulation codes: GULP[5], GAMESS[9], Monty[10], and SIESTA[8]. Additionally, VASP[11,12] and USPEX[13–15] are now available, which is the topic of the current publication. The Diffraction tool uses internal GDIS calculations to render a simulated pattern for X-ray, neutron, or electron diffraction.
	Analysis	This submenu is dedicated to the postprocessing of calculated results. The first part, Dynamics, is used to process results from models consisting of several frames. The pair count, radial distribution function (RDF), and several geometric measurements can be displayed as a function of the frame index. The second submenu, Measurements, provides an interface for the calculation of interatomic distances, bond distances, 3-atoms angles, and 4-atoms torsion angles, selected on the model or systematically (by atom type). The results are indicated on display and in an included table. The last tool, Plots, was introduced for the analysis of VASP results. It gives the possibility to plot the energy, force, volume, and pressure for each model frame, and the total electronic density of states (DOS) and band structures, when available.

Table S1 Continued: Content and description of the functions in the menu area of the GDIS software.

Menu title	Sub-menu	Brief description
View	Display properties	This submenu gathers all editable rendering parameters of GDIS. The chemical model representation can be configured here together with the global camera, light, and colors settings. The 3D, stereo, and external rendering using the POVRay software[16] are also available here.
	Reset model images	This submenu resets the number of displayed cell images to a 1x1x1 supercell.
	Normal mode	This submenu is used to switch back to normal mode from the recorded mode (see below). All generated frames are then saved and the recording process can be continued at a later point.
	Recording mode	This submenu switches the recording mode on. Each transformation applied to a model (rotation, zoom, etc.) is recorded as a new frame of this model. This results in a recording of the user action on the model that can be rendered or saved using the Animation tool (see above). This recording mode cannot be initiated on a model already containing frames.
	Task manager	This submenu opens the task manager GUI which gathers all tasks and processes that have been initiated by GDIS since the software was launched. With the introduction of VASP and USPEX interfaces, GDIS can now launch concurrent and asynchronous tasks. Depending on whether a task is internal to GDIS or a spawned process, it can be interrupted by the user.
	Executable paths	This submenu displays a table containing the execution path of 3rd party software used by GDIS (Babel, GAMESS, GULP, Monty, POVRay, and the display image viewer). In this work, the path of VASP, USPEX, and mpirun commands have been added to perform the corresponding calculation (mpirun allows VASP and USPEX to perform the calculations in a parallel environment).
Help	Manual	This submenu shows a short manual of some GDIS functions. Acknowledgement for major GDIS contributors is also displayed here.

Table S2: Content and description of the functions in the tool area of the GDIS software.

Tool icon	Tool name	Brief description
	Open	identical to the File/Open function of the menu area.
	Save	identical to the File/Save function of the menu area.
	Add Model	Add an empty model to the list of opened model. The empty model does not contain any chemical structure information and is initially considered as a molecular model by GDIS.
	Remove Model	This closes the current model, which is then removed from the Tree view area. The model is completely unloaded from memory and this operation cannot be undone.
	Animate	Identical to the Tools/Visualization/Animation submenu.
	Edition	Identical to the Tools/Visualization/Editing submenu.
	Iso-surfaces	Identical to the Tools/Visualization/Iso-surfaces submenu.
	Periodic	Identical to the Tools/Visualization/Periodic Table submenu.
	Diffraction	Identical to the Tools/Computation/Diffraction submenu.
	Surfaces	Identical to the Tools/Building/Surfaces submenu.
	Measurements	Identical to the Tools/Analysis/Measurements submenu.
	Display	Identical to the View/Display properties submenu.
	Reset model	This tool will reset the rendering field of view of a chemical structure to its original position. Note that transformations and editing that were applied to atoms will remain, only the viewing angle and scaling (changed by rotation and zoom) will be reset to their initial values.
	Reset images	Identical to the View/Reset model images submenu.
	Record mode	Identical to the View/Recording mode submenu.

Table S2 Continued: Content and description of the functions in the tool area of the GDIS software.

Tool icon	Tool name	Brief description
	Single Canvas	The canvas define an artificial separation of the Main render area to display simultaneously several systems. Each separation is taken by GDIS as an independent render, named under the generic term of canvas. This tool reset that separation to the initial, un-separated behaviour.[a]
	Increase Canvas	This will increase the number of display canvas in the Main render area. At present, only a vertical separation or both a vertical and horizontal separations modes are supported. This means that the number of canvas must be lower or equal to 4.[a]
	Decrease Canvas	This will decrease the number of canvas. The minimum number is 1, effectively resetting to the initial mode of GDIS Main render area.[a]
	Reset mode	Identical to the View/Reset model images submenu.
	Plot Control	This tool was developed together with the new VASP and USEX integration. It enables a better control of the plot parameters, such as legends, axes title, scale, etc.
	to EPS	Save the main render area into an encapsulated postscript (EPS) file. This tool will only appear if the computer running GDIS can handle the EPS file format.
	to PNG	Save the main render area into a portable network graphic (PNG) file. This menu will only appear in the rare case where a computer cannot handle the previous to EPS mode. Due to the library used by GDIS, this mode is always possible.
	Track mode	In the case VASP or USPEX files are opened in GDIS, this tool allows to follow the progress of an ongoing calculation. If the calculation was started by GDIS, the tracking is then automatically switched on. Another click on this tool icon will stop the tracking mode.

[a] Note that canvas feature is, in this version of GDIS, an experimental feature.

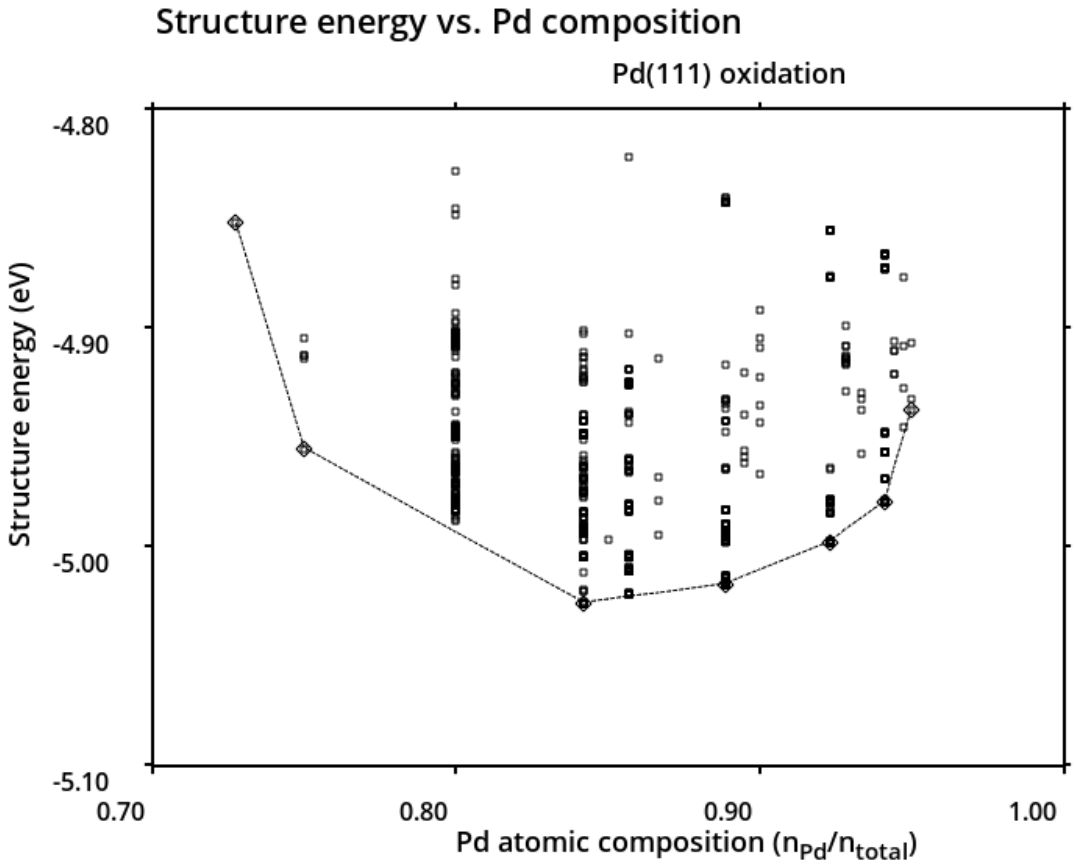
Table S3: Content and description of the icons that appear in the combo-box area of the GDIS software.

Icon	Name	Brief description
	molecule	This icon indicates that the loaded model is of molecular type. This is the default when a new model is created, as no lattice information is provided.
	polymer	Indicates that the currently loaded model is a 1-dimensional material.
	surface	Indicates that the currently loaded model is a surface, ie. a 2-dimensional material. When saving a surface in the Tools/Building/Surfaces submenu, a model of that type is created.
	crystal	Indicates a 3-dimensional system. Note that it was also chosen to represent USPEX jobs, regardless of the dimension of the structure prediction, since structures in USPEX are represent in VASP format, which always include all cell parameters.
	morphology	Indicates that the currently loaded model represents a crystal shape. When using the tool from the Tools/Building/Surfaces submenu, it is possible to export the crystal shape relative to the calculated surface energies in a GDIS gmf format. The representation is then equivalent to a Wulff construction of the equilibrium crystal shape.
	graphics	This icon appears as a child of a model parent to indicate that some analysis is available in a separate plot graphic. When using the Remove Model tool while a graphics is selected in the Tree view area, only the graphics will be removed: the model and other graphics, if any, will remain available in GDIS.
	tracking	Tracking mode is a new feature introduced in the manuscript. It allows to follow a calculation, updating information as it becomes available by calculation. To indicate that a specific model is being track, its icon will be replaced by a tracking icon. Such icon displays a short (3-frame) animation only when GDIS is actually receiving new data from the calculation. Since tracking is only implemented in VASP and USPEX, only the crystal icon can be replaced by a tracking one.

S2 The Convex-Hull

The convex-hull is a precious indication in USPEX that allows a user to have an idea of the relative stability – or metastability – of a structure in a variable composition calculation of a binary structure A_xB_y. It is generally calculated by a simple algorithm that will first add as the origin the lowest energy structure for the lowest x composition. Then, the structure that, on a line with that origin, presents the most negative slope will be added to the convex hull, and set as the new origin. The last sentence is then repeated with that new origin until it reaches the highest x composition.

Figure S2: Example of a convex-hull graph obtain by GDIS on a Pd(111) surface oxidation variable composition simulation, from the USPEX example calculations.



Since GDIS is following ongoing calculation, such a simple algorithm had proven quite inefficient. With an ongoing calculation, any of the origins can be changed, which will cause drastic changes to the convex-hull. Furthermore, since the convex-hull is calculated after each structure optimization in a single USPEX generation, we cannot rely on the previously calculated best structures. GDIS, however, have a list of the minimum energy structures for each composition, due to the continuous display of the results. This list is then used for the

determination of the convex-hull which components are also minimum energy structures. Starting from the minimal energy structure, regardless of the composition, which is also known, the algorithm used in GDIS will calculate the left and right part of the convex-hull separately, considering the most negative and lowest positive slopes, respectively. Should the starting point be one of the minimum or maximum x compositions, only the relevant part of the graph will be calculated. The convex-hull is then rendered as a line on each graph that plots the structure energies as a function of the atomic of one of its constituents. An example graph is presented in Fig. S2. An important factor, as shown in the figure, is that if the convex-hull is obtained only from the USPEX generated structures, it will not necessarily include the references of pure materials. Another problem is that the references might not be of the expected thermodynamics standard. For example, one might expect the reference energy for oxygen to be that of the oxygen molecule, while USPEX calculation might not allow a structure to reach. This is why the reference for pure materials with the minimum energy can be provided externally for GDIS visualisation. For this, a separate file, chem.in, can be simply written in the directory of the USPEX results. That file can contain a line for each species which lists the species number (in the same order as provided in USPEX), the number of atoms of the reference, and the energy in eV. Note that only the references that need correction shall be entered in this way. Entering an energy for all references is not mandatory. The feature will be extended in the future to non-homoatomic references.

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