Results and Discussion: Universal DFT + U Generation Scheme

Instructions pertaining to the use of the org-mode python code UGCA_in_DFTU.org to generate and run DFT+U linear response calculations pursuant to the calculation of self-consistent Hubbard U values are listed in the code itself. In order to process these results and find self-consistent U values, the following steps were completed:

- After all calculations have finished running, make a directory titled according to the perturbed atom and inputted value of the perturbed U (mkdir Ucalc_100_Co). Then, copy the output file (run.o …) into this directory and rename it OUTPUT.

- Copy aggregator_1st_QE_DFT+U into this directory and run it to separate the initial SCF calculation and all subsequent perturbation calculations into distinct files. After this separation, the initial and final responses of each atom, which are taken with respect to appropriately enumerated perturbed atoms, will be formed by this code. They are named according to the convention: " dn / dn0 (final / initial response)" _ "atom # or index of current atom" _ dalpha _ "atom index of perturbed atom".

- The occupations of the perturbed orbitals are listed in files where the two atomic indices within file names match. For U = 0, the initial and final responses must have equal occupation traces (sum of diagonals of occupation matrices) when their perturbations are both zero. Both responses must also be linear over perturbations sampled. This is verified for examples tested by this code – 2x2x2 AFM (FCC) NiO (tested by Cococcioni at U = 0, files stored at media.quantum espresso.org/ santa_barbara_2009_07/index.php) and 1x1x1 FM (R-3c) $LaCoO_3$ – (tested by Wentzcovitch *et al.* at Physical Review B 85, 140404(R) 2012) on the next page.

- This process is completed for each perturbed atomic species, and all responses resulting from these perturbations are copied over to parent directories of the form Ucalc_#_combine (# = value of inputted U), along with lists generated that specify the names of the files containing occupation data (e.g.: files_NiO). These individual file lists are concatenated to match the ordering of elements written into a position file (e.g.: pos_NiO), which gives the cell geometry and coordinates of all atoms, as well as data detailing the magnitudes and signs of atomic magnetic moments.

- The executable (r.x) that runs the linear response calculation code (resp_mat.f90) is generated by the command "sh comp_resp_mat.j", while the command "./r.x < resp_mat.in" calculates effective outputted U values from a system with particular inputted values of U. The input file read into the executable (resp_mat.in), in addition to specifying the numbers of different types of atoms and perturbations present, also indicates whether magnetism is present in the system. Lastly, the input file can specify the addition of a "background" to the response matrix; the addition of a background, which is generally a weighted (evenly weighted in the 'neutral' case) sum of all response matrix elements added to newly appended column and row entries, can lead to faster U convergence with respect to supercell size.
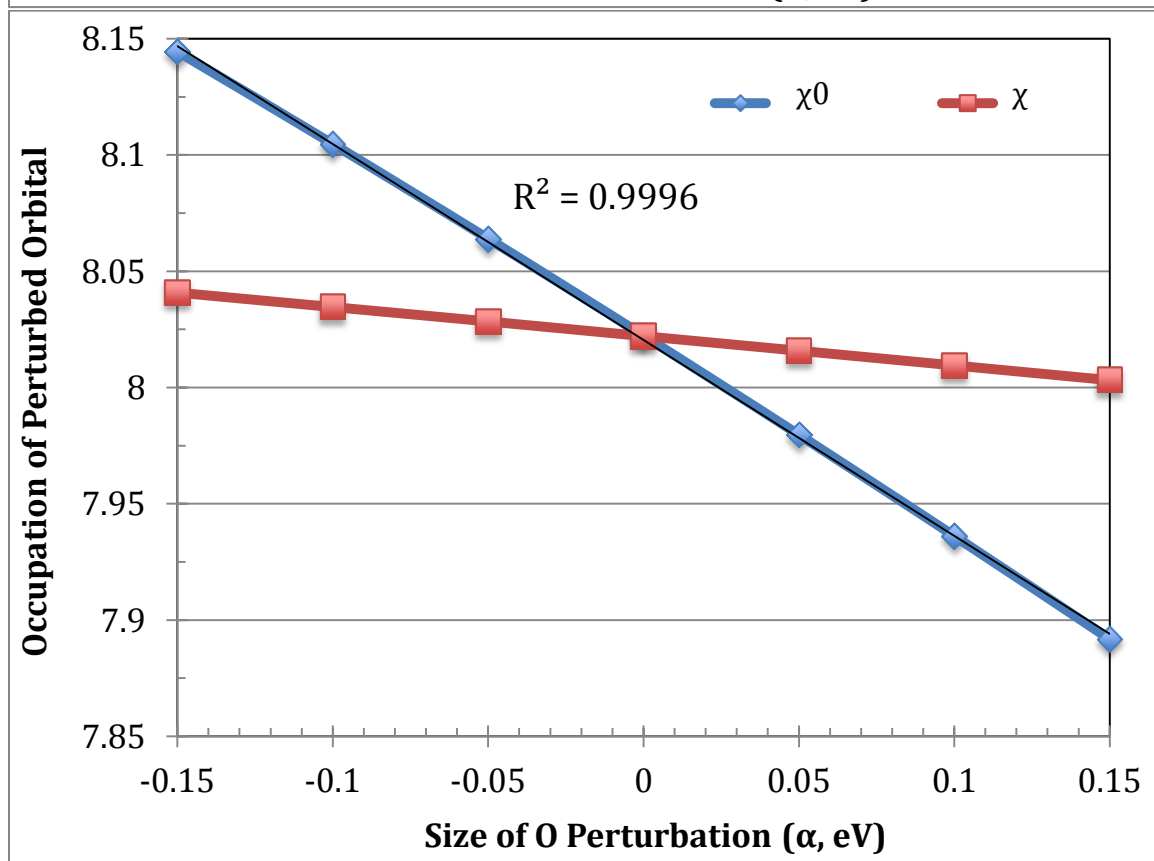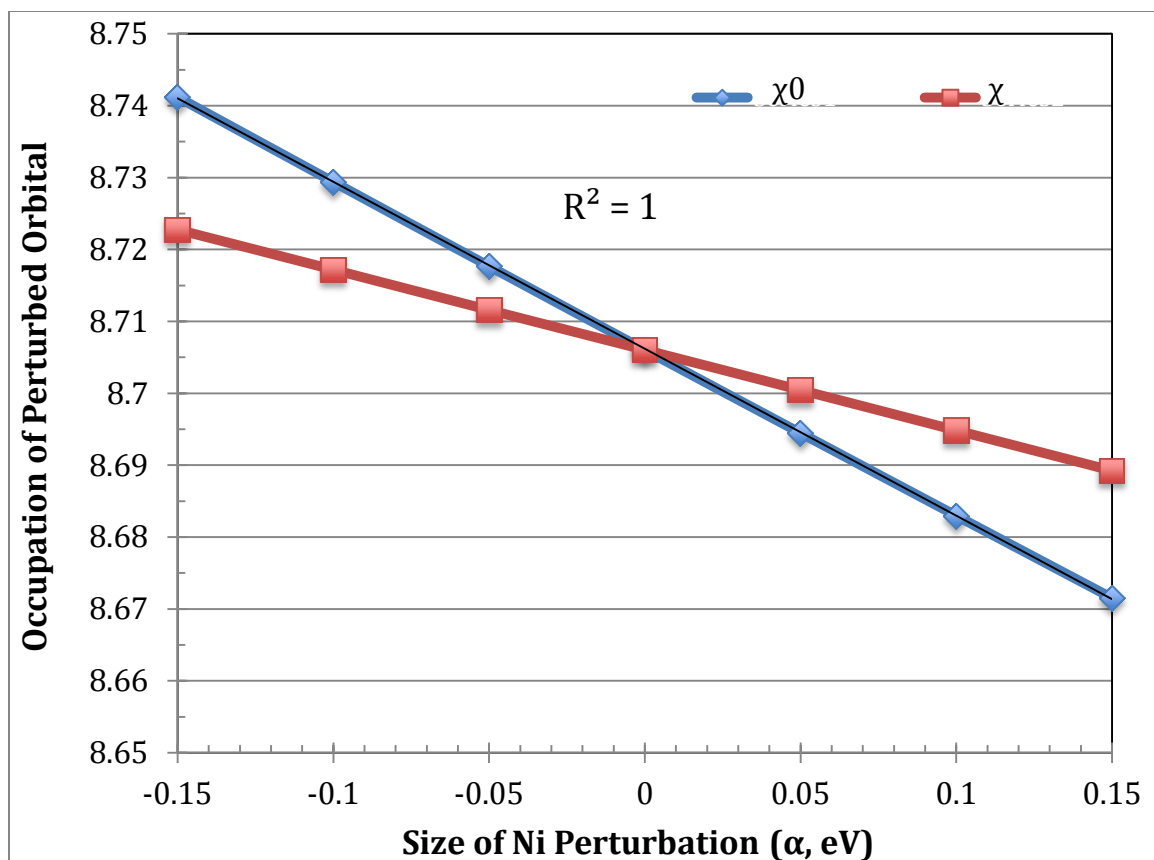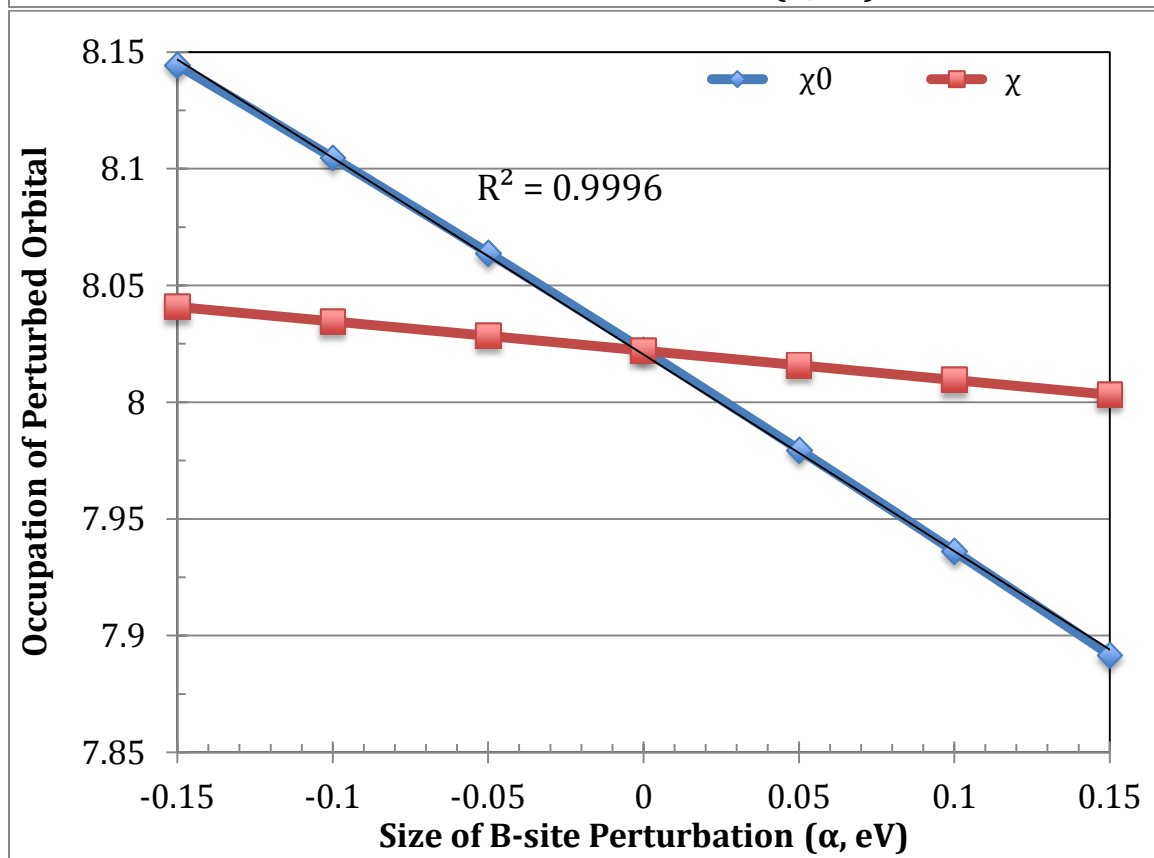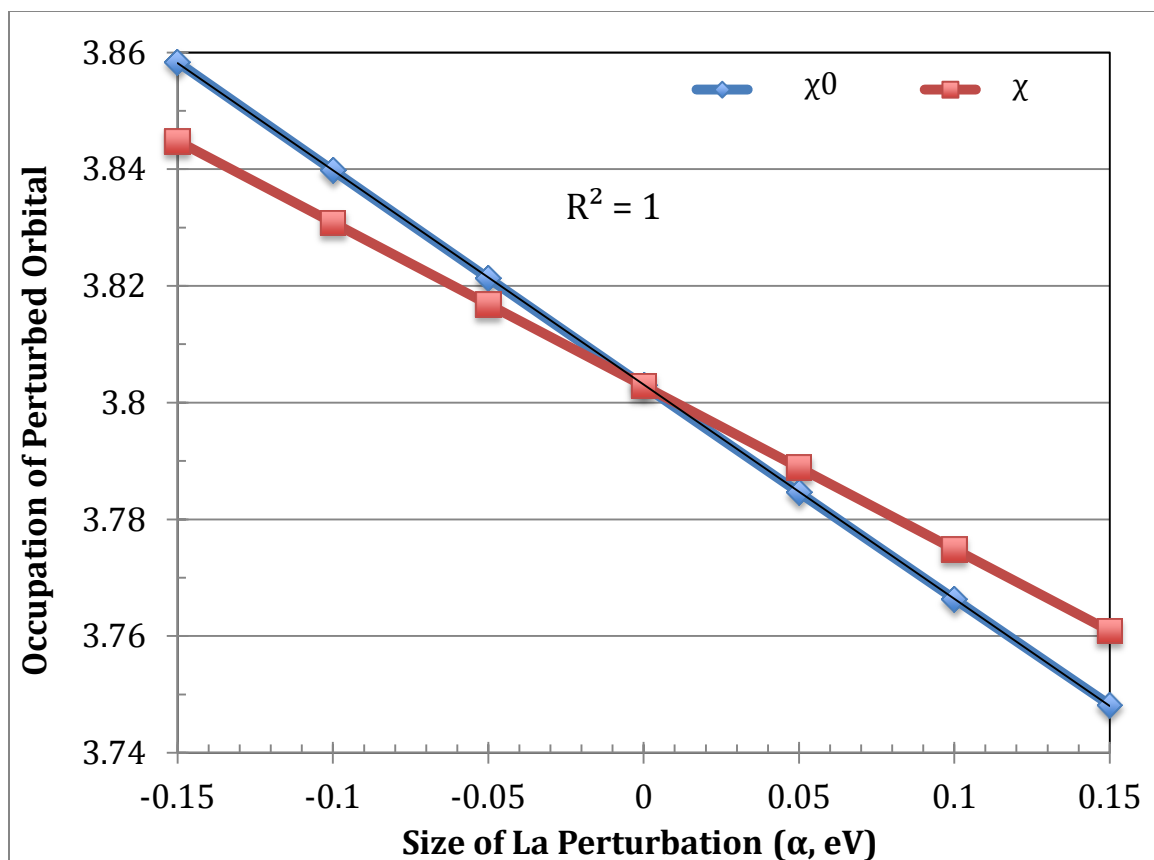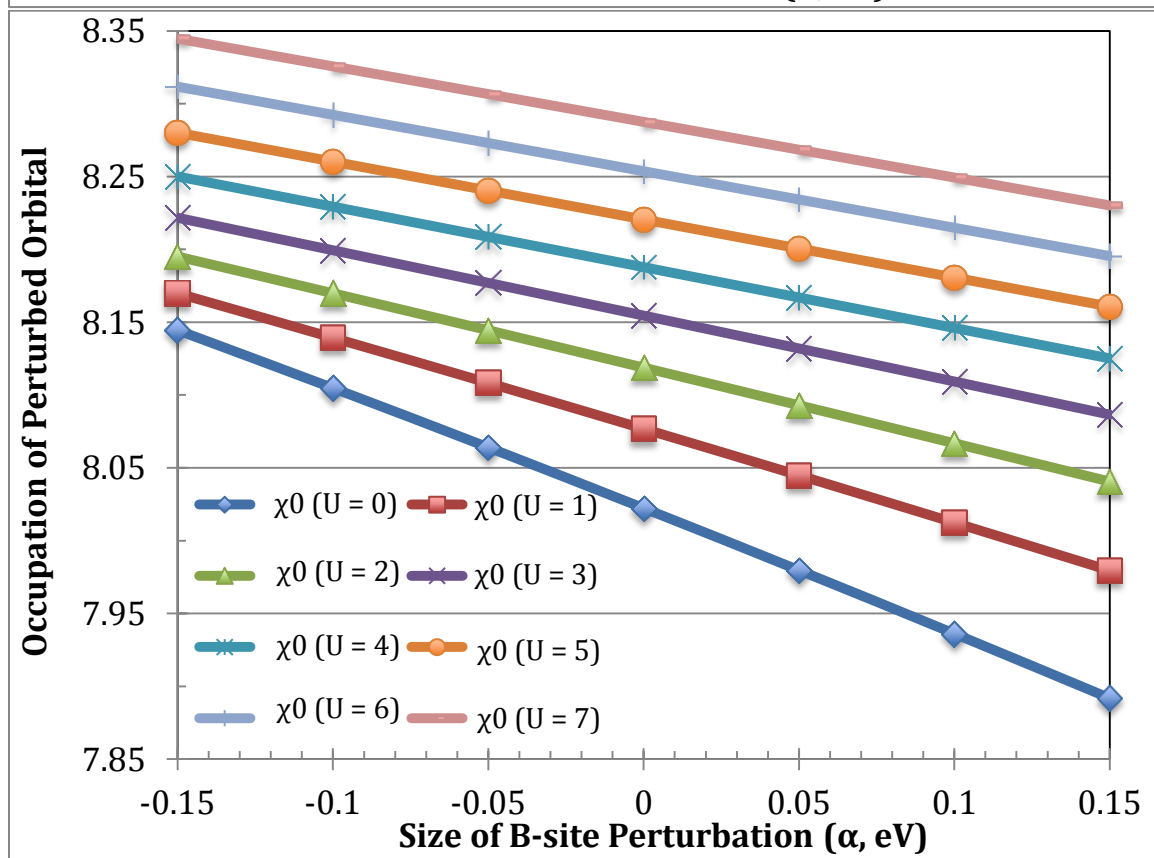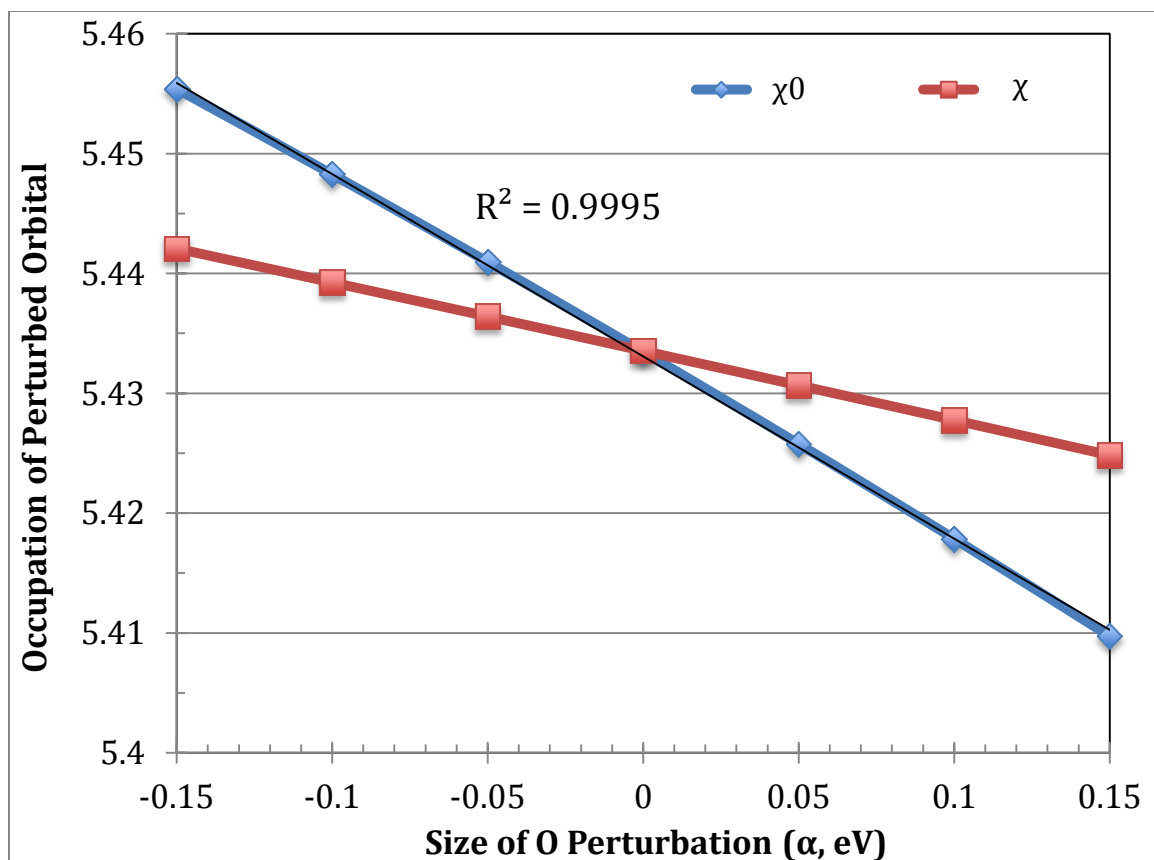
In addition to showcasing the linearity and intersection of the initial and final responses for all cases above, the variation in the initial response with increasing inputted U (U perturbing $3d$ orbitals of Co) is illustrated as well for the $LaCoO_3$ case. Note that $3d$ orbital perturbation occurs for Ni, $2p$ orbital perturbation occurs whenever O is considered, and La – due to the design of its pseudopotential – must be modeled with respect to its ground state (0 K) occupancy (i.e.: $5d$ orbital perturbation occurs). In order to obtain the self-consistent U of FM $LaCoO_3$, different values of inputted U are first placed on all Co cations in the unit cell, while the values of outputted U on those Co cations are resolved in every case via linear response theory. Outputted U values are regressed against inputted U values linearly – in cases where either a neutral background is added or none is considered – up to the point at which outputted and inputted U values are no longer linearly related (valid points for both $LaCoO_3$ cases are marked accordingly).
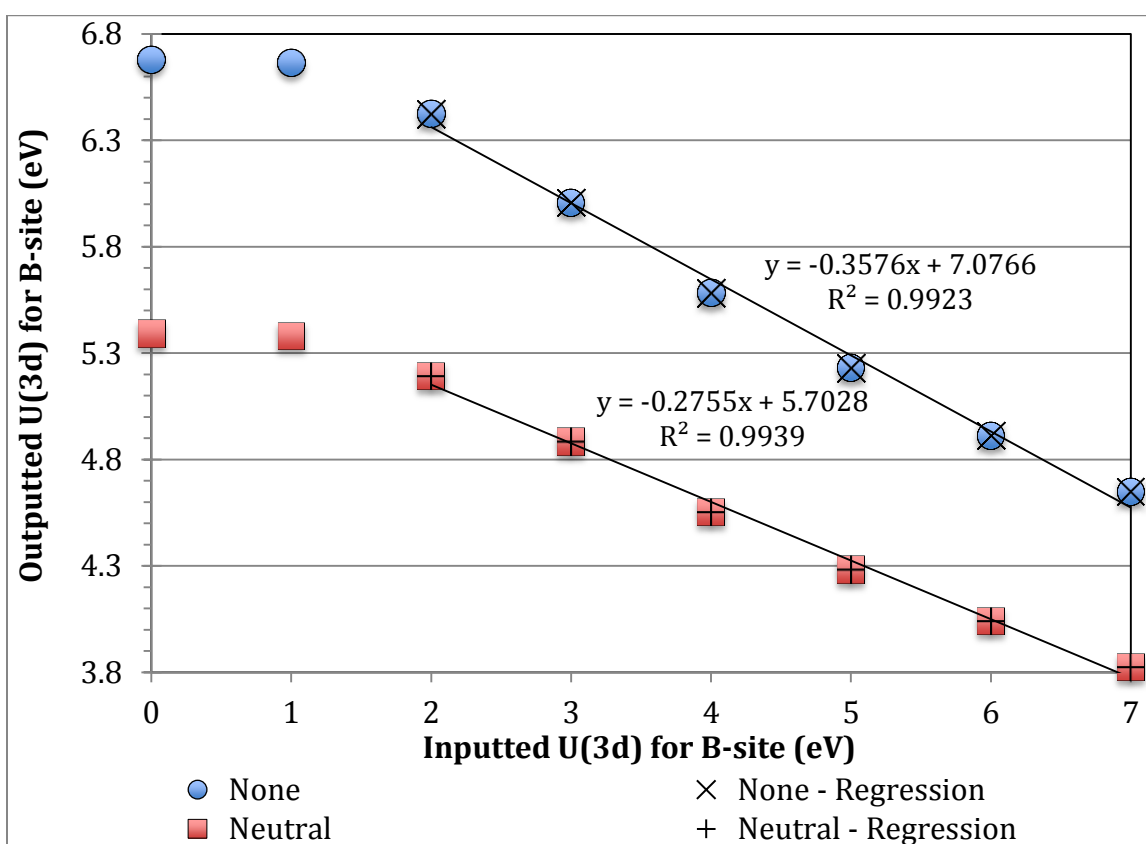
The y-intercepts formed by these regressions equal the calculated self-consistent U values for Co in the FM $LaCoO_3$ system. Despite the fact that only a unit cell representation of the R-3c cell was implemented in this calculation, the resolved U of 7.08 eV is in very strong agreement with corresponding results deduced by Wentzkovitch *et al* (7.0 eV). Implementation of a neutral background, which can – but does not necessarily – speed up convergence of calculated U values with respect to cell size, has a negligible effect in larger supercells.