Defect chemistry as a route to engineer oxide materials with new functionalities

C. Ricca[1], U. Aschauer[1]*

[1]University of Bern

Defect chemistry is a emerging parameter to take into account in the design and/or discovery of new functionalities in transition metal oxides. Defects, like oxygen vacancies or substitutional cations, can indeed tune the properties of these materials and even more importantly, new functionalities may arise because of defects. For example, in transition metal perovskite oxides, paraelectric to ferroelectric and antiferromagnetic to ferromagnetic transitions can be observed as a function of defect type and concentration.

It would thus be interesting to be able to screen, at the theoretical level, the effect of different types of defects, in different charge states, on the properties of these materials. In this way, general trends can be deduced and new material/defect combinations, together with the required experimental synthesis conditions, can be suggested to be used as guidelines by experimentalists. To reach this goal, we designed a computational strategy that will allow a systematic and automated exploration of the defect chemistry of oxide materials and which should be able to provide accurate results for experimental synthesis conditions.

The computation of reliable defect formation energies is complicated by the need for an accurate description of the electronic properties of the bulk and defective system, which can be obtained by the application of advanced density functional methods or application of appropriate corrections schemes. To adequately describe the electronic structure of strongly correlated materials containing transition metals within a reasonable computational time, we suggest to apply the DFT+U approximation, by determining the value of the U parameter from first principles and as a function of the distance from the defect sites. Accurate formation energies are then computed by applying adequate *a posteriori* corrections, like elastic,
band filling, and more importantly electrostatic correction in the case of charged defects.

In order to manage this sequence of calculations for a variety of material/defect combinations we use the AiiDA (Automated Interactive Infrastructure and Database for Computational Science)[1] informatics platform to chain the various calculation steps, allowing to obtain the formation energy for several defect types in automated way based solely on the bulk structure of a given material.

All the aspects relative to the theoretical and computational foundation for this systematic exploration of functional defects in materials are here described in detail. Furthermore, results on selected perovskite materials are also reported to demonstrate the validity and efficiency of the suggested computational strategy.

[1] G. Pizzi, A. Cepellotti, R. Sabatini, N. Marzari, and B. Kozinsky, *Comp. Mat. Sci.*, **2016**, 111, 218-230.