Fluorine clusters at CeO2(111) - A DFT+U and Monte Carlo study

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STM experiments on CeO2(111) reveal depressions in the surface oxygen sublattice which are observed to form clusters of various shapes and sizes [1]. While these depressions were assumed to be oxygen vacancies, subsequent DFT calculations have indicated that clusters of oxygen vacancies are energetically unstable [2-4]. Recently, we showed theoretically that fluorine impurities should appear almost identical to oxygen vacancies in STM experiments, but that their properties are more in line with those of the defects observed in experiments [5]. Here, I will present the results of a further investigation into the distribution of F impurity clusters at CeO2(111), using a combination of DFT+U calculations, and Monte Carlo sampling based on a simple but accurate pair potential which was fitted to the DFT results. The distribution is characterised in terms of the number of clusters of a certain size, and also on their topology, i.e. whether they are compact or open/linea r. Our results compare favourably with the experiments, and also exhibit some interesting physics in their own right.

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