A Self-Interaction Corrected Ultrasoft Pseudopotential Scheme for Highly Correlated and Magnetic Systems

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Among the innumerable successes of Local-Spin-Density approximation (LSDA), there are also well known, systematic failures that compromise the possibility of obtaining accurate predictions for a range of properties in highly correlated and magnetic compounds. These problems are particularly evident in the case of magnetic perovskites, in which LSDA fails to predict the correct ground state magnetic ordering, and generally favors metallic and ferromagnetic ground states in compounds that are experimentally observed to be antiferromagnetic insulators (e.g. $LaMnO_3$).

The misrepresentation of an insulator as a conductor is especially harmful if this system is ferroelectric or piezoelectric (e.g. $YMnO_3$), since the metallic character makes it impossible to calculate their dielectric properties. Furthermore, the local dependence of the one-electron potential on the charge density does not allow the incorporation of important mechanisms of energy stabilization such as orbital or charge orderings.

Many of these problems can be traced back to the presence of self-interaction in the LSDA potential, which causes an underestimation of the binding energy of the strongly localized d states of the cation. This effect, in turn, widens the interatomic d-p hybridizations, and overestimates the electron hopping with respect to the on-site Coulomb energy. As a results, materials with strongly localized (atomic-like) electrons are generally not correctly described in LSDA.

Since the direct application of a self-interaction corrected (SIC) functional is highly impracticable (due to the consequent breaking of translational symmetry), it is necessary to apply the SIC in some semplified form. Here we propose an approach that casts the SIC potential as a non-local projector acting upon the Bloch states. This potential is first derived from the free atom (i.e. is pseudopotential-like), and then it is updated during the self-consistent procedure. This SIC scheme, implemented within the ultrasoft pseudopotental formalism, promises to be a powerful and general approach, capable of a systematic improvement over the LSDA description, without sacrificing its conceptual semplicity, and, moreover, without considerably increasing the computational cost. Here we introduce our formalism and we show results of its application to important cases of magnetic perovskites.