Ab-initio pseudopotential theory of the orbital magnetization

D. Ceresoli[1†], U. Gerstmann[2], A. P. Seitsonen[2] and F. Mauri[2]

1. Scuola Internazionale Superiore di Studi Avanzati (SISSA) and DEMOCRITOS, via Beirut 2–4, I-34014 Trieste, Italy.

2. Institute de Minéralogie et de Physisque des Milieux Condensés, Université Pierre et Marie Curie, 140 rue de Lourmel, F-75015 Paris, France

† Present address: Dept. of Materials Science and Engineering, Massachusestts Institute of Technology (MIT), 77 Massachusetts Avenue, Cambridge, MA 02139-4307.

The position operator $\mathbf{r}$ is ill-defined within periodic boundary conditions: owing to this, both the macroscopic (electric) polarization and the macroscopic orbital magnetization are nontrivial quantities. While the former has been successfully tamed since the early 1990s, the latter remained a long-standing unsolved problem. A successful formula within DFT for crystalline systems has been recently found [1]. The formula allows to evaluate $\langle \mathbf{r} \times \mathbf{v} \rangle$ within periodic boundary condition, and is valid for all-electron (AE) hamiltonians.

On the other hand, pseudopotential (PS) methods are highly desirable in order to reduce the computational cost of AE calculations. However, PS methods carry some additional drawbacks: (i) the non local potential couples to the electromagnetic field in a non trivial way; (ii) the nodal structure of the wavefunction close to the nuclei is lost. This last drawback prevents the accurate evaluation of magnetic response properties such as the NMR chemical shifts and the EPR g-tensor.

In order to circumvent these drawbacks, we employed the GIPAW formalism [2], which takes care of the coupling to electromagnetic fields and allows to "reconstruct" the AE wavefunctions from the PS wavefunctions.

We applied our pseudopotential formula to compute the EPR g-tensor of paramagnetic defects in a non-perturbative way and to the calculation of the orbital magnetization of bulk Fe, Ni and Co.

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