All-electron relativistic four-component Dirac–Kohn–Sham theory for solids using Gaussian-type functions

Marius Kadek,[a] Michal Repisky,[a] and Kenneth Ruud[a]

[a] *Hylleraas Centre for Quantum Molecular Sciences, UiT The Arctic University of Norway, Tromsø, Norway*
marius.kadek@uit.no

I will present a four-component Dirac–Kohn–Sham theory for obtaining relativistic band structures of spin–orbit-coupled solids. The method was recently developed in our group [1], and uses Gaussian-type orbitals (GTOs) to compactly express all operators in real space. The four-component self-consistent field equation of a translationally invariant system

$$\begin{pmatrix} \mathcal{V}^{LL}(\boldsymbol{k}) & \mathcal{T}(\boldsymbol{k}) \\ \mathcal{T}(\boldsymbol{k}) & \frac{1}{4c^2}\mathcal{V}^{SS}(\boldsymbol{k}) - \mathcal{T}(\boldsymbol{k}) \end{pmatrix} \begin{pmatrix} c_L(\boldsymbol{k}) \\ c_S(\boldsymbol{k}) \end{pmatrix} = \epsilon(\boldsymbol{k}) \begin{pmatrix} \mathcal{S}(\boldsymbol{k}) & 0_2 \\ 0_2 & \frac{1}{2c^2}\mathcal{T}(\boldsymbol{k}) \end{pmatrix} \begin{pmatrix} c_L(\boldsymbol{k}) \\ c_S(\boldsymbol{k}) \end{pmatrix} \tag{1}$$

is then solved in its matrix form in reciprocal space using the restricted kinetically balanced basis. The atom-centered nature of GTOs allows for explicit handling of one-, two-, and three-dimensional periodic systems while avoiding the need to introduce vacuum layers. The spin–orbit coupling is treated variationally, which allows for studying topological insulators as well as spin–orbit-induced splittings of bands in materials containing heavy elements. The GTO-based all-electron methodology makes no assumptions about the electronic density in the vicinity of nuclei, and can be used to calculate core-related properties, such as nuclear magnetic resonance parameters. To enable large-scale relativistic calculations of solids containing thousands of heavy atoms in the simulation supercell, the presented approach exploits fast-multipole methods, and is implemented in the ReSpect program package [2] that uses the quaternion algebra for the time-reversal-adapted basis.

References

1. M. Kadek, M. Repisky, and K. Ruud, *Phys. Rev. B* **99** (2019), 205103.
2. http://www.respectprogram.org/