Chapter 28
Screened KKR

Krisztián Palotás and László Szunyogh

Abstract The concept of screening in the Korringa–Kohn–Rostoker (KKR) multiple scattering electronic structure method for solids is briefly presented. The main advantages of the screened KKR (SKKR) method and recent applications are highlighted.

28.1   Introduction

Since the publications of the seminal works of Korringa [1] and Kohn and Rostoker [2] with the aim at describing the electronic structure of periodic solids, the Korringa–Kohn–Rostoker (KKR) Green-function-based multiple scattering method has been considerably developed over the years [3–5]. This methodological progress has been fueled by the steadily growing computational (hardware) facilities and constantly developing efficiency in the software implementations of the KKR method. An important step has been the introduction of the so-called screening transformation [6, 7] that for systems with at best two-dimensional translational symmetry provides a fast and, in principle, exact numerical solution through the casting of big (infinite) matrices into a block-tridiagonal form. The Screened Korringa–Kohn–Rostoker (SKKR) method has been invented in the 1990s in the groups of Peter Weinberger at the Technical University of Vienna and László Szunyogh at the Technical University of Budapest, where it is still hosted and being continuously developed.

K. Palotás
Department of Theoretical Physics, Budapest University of Technology
and Economics, Budafoki út 8., Budapest 1111, Hungary
e-mail: palotas@phy.bme.hu

K. Palotás
Slovak Academy of Sciences, Institute of Physics, Dúbravská cesta 9.,
84511 Bratislava, Slovakia

L. Szunyogh (✉)
Department of Theoretical Physics and MTA-BME Condensed
Matter Research Group, Budapest University of Technology and Economics,
Budafoki út 8., Budapest 1111, Hungary
e-mail: szunyogh@phy.bme.hu

While a very detailed description of the SKKR method can be found in [8], in this chapter we briefly present the underlying concept and highlight the main advantages of the SKKR method together with selected recent applications.

28.2 The KKR Method

Unlike other popular electronic structure calculation methods using electron wave functions, the KKR formalism relies on the Green's function that has a conceptual advantage of straightforward evaluation of physical properties. The basic ingredients of the KKR method formulated within multiple scattering theory [4] are the single-site scattering t–matrices and the structure constants. The single-site t-matrix is an angular momentum representation of the single-site T–operator formally obtained in the following way: Assuming that a single-particle Hamiltonian, composed in the spirit of the Kohn–Sham formulation [9] of density functional theory, is given as a sum of an unperturbed (H_0) and a Hermitian perturbation (V) part, $H = H_0 + V$, the corresponding resolvents (with z a complex energy argument, and I the identity operator) are $G_0(z) = (zI - H_0)^{-1}$ and $G(z) = (zI - H)^{-1}$. They can be related to each other as $G(z) = G_0(z) + G_0(z)T(z)G_0(z)$ by defining the T–operator $T(z) = V(I - G_0(z)V)^{-1} = (I - VG_0(z))^{-1}V$, which implies $G_0(z)T(z) = G(z)V$ and $T(z)G_0(z) = VG(z)$. If only a single scattering potential centered at the lattice position $\boldsymbol{R}_n$ is present in the system then $\underline{t}^n(z)$ represents the single-site scattering t-matrix with respect to the product of the spherical Bessel functions and spherical harmonics, $j_L(z, \boldsymbol{r}) = j_\ell(\sqrt{z}r)Y_L(\hat{r})$ with the abbreviation $L = (\ell, m)$.

Generalizing the system by introducing non-overlapping potentials at various positions in space, e.g., at atomic sites, that individually act as single-site scatterers, their common effect is described by multiple scattering theory. The multi-site T–operator describes all possible scattering events: $T = \sum_n t^n + \sum_{nm} t^n G_0(1 - \delta_{nm})t^m + \sum_{nmo} t^n G_0(1 - \delta_{nm})t^m G_0(1 - \delta_{mo})t^o + \ldots$, which can be recast to $T = \sum_{nm} \tau^{nm}$ by introducing τ the so-called scattering path operator [3]. This implies that the multiple scattering can be taken into account as simple as $G = G_0 + \sum_{nm} G_0 \tau^{nm} G_0$. The real-space structure constants, $G_{0,LL'}^{nm}(z)$, result from the two-center expansion of $G_0(z)$, i.e. for $n \neq m$ and $\boldsymbol{r} = \boldsymbol{r}_n + \boldsymbol{R}_n$ and $\boldsymbol{r}' = \boldsymbol{r}'_m + \boldsymbol{R}_m$: $G_0(z; \boldsymbol{r}_n + \boldsymbol{R}_n, \boldsymbol{r}'_m + \boldsymbol{R}_m) = \sum_{LL'} j_L(z, \boldsymbol{r}_n)G_{0,LL'}^{nm}(z)j_{L'}(z, \boldsymbol{r}'_m)^\times$, where the superscript $^\times$ denotes that only the spherical harmonics are conjugated. Using a site-angular momentum supermatrix representation, the matrix of the structure constants is denoted by $\underline{\underline{G}}_0(z) = \underline{\underline{G}}_0^{nm}(z)(1 - \delta_{nm})$, while the t-matrices can also be structured in a site-angular momentum supermatrix, $\underline{\underline{t}}(z) = \underline{t}^n(z)\delta_{nm}$, i.e. block-diagonal in the site index.

The fundamental KKR equation defines the site-angular momentum supermatrix representation of the scattering path operator, $\underline{\underline{\tau}}(z)$, in terms of the single-site t-matrix and the structure constant matrix as

$$\underline{\underline{\tau}}(z) = \left(\underline{\underline{t}}(z)^{-1} - \underline{\underline{G}}_0(z) \right)^{-1} . \tag{28.1}$$

Finally, the single-particle Green's function is obtained from the scattering path operator as

$$
\begin{aligned}
G(z; \boldsymbol{r}_n + \boldsymbol{R}_n, \boldsymbol{r}'_m + \boldsymbol{R}_m) = \sum_{LL'} Z_L^n(z, \boldsymbol{r}_n) \tau_{LL'}^{nm}(z) Z_{L'}^m(z, \boldsymbol{r}'_m)^\times - \\
\delta_{nm} \sum_L Z_L^n(z; \boldsymbol{r}_{n,<}) J_L^n(z; \boldsymbol{r}_{n,>})^\times ,
\end{aligned}
\tag{28.2}
$$

where Z_L^n and J_L^n are properly normalized regular and irregular scattering solutions, respectively, and $r_< = \min(r, r')$ and $r_> = \max(r, r')$. From the above Green's function the physical quantities can straightforwardly be calculated.

28.3   Screening Transformations in the KKR Method

Solving the KKR equation (28.1) for systems in reduced dimension requires the inversion of a big matrix, and its computational time scales with N^3 (N being the size of the system in real space). This is highly unfavorable at a large number of scatterers (atoms) and one possible way to overcome the problem is to use a *screening transformation*. Here, a reference potential, V_r, is added to the Hamiltonian of the unperturbed system, $H'_0 = H_0 + V_r$, such that $H = H_0 + V = (H_0 + V_r) + (V - V_r) = H'_0 + V'$ with $V' = V - V_r$. The resolvent of H can then be expressed as $G(z) = G_0(z)(I + VG(z)) = G'_0(z)(I + V'G(z))$, where $G'_0(z) = (zI - H'_0)^{-1} = (zI - H_0 - V_r)^{-1}$. Once the potential V_r is repulsive, $G'_0(z)$ gets localized in real space for $\Re(z) < V_r$, which makes the calculation of $G_0(z)$ feasible.

Turning to multiple scattering, such a reference potential is written as a superposition of non-overlapping potentials $V_r = \sum_n V_r^n(\boldsymbol{r}_n)$. If the corresponding single-site t-matrices are $\underline{t}_r^n(z)$ then the Green's function matrix of the reference system, termed as *screened structure constant*, is obtained as

$$\underline{\underline{G}}_r(z) = \underline{\underline{G}}_0(z) \left(\underline{\underline{I}} - \underline{\underline{t}}_r(z) \underline{\underline{G}}_0(z) \right)^{-1} . \tag{28.3}$$

By defining the *screened t-matrix* as the difference, $\underline{\underline{t}}_\Delta(z) = \underline{\underline{t}}(z) - \underline{\underline{t}}_r(z)$, a formally equivalent equation to (28.1) can be obtained,

$$\underline{\underline{\tau}}_\Delta(z) = \left(\underline{\underline{t}}_\Delta(z)^{-1} - \underline{\underline{G}}_r(z) \right)^{-1} , \tag{28.4}$$

related to τ as

$$\underline{\underline{\tau}}(z) = \underline{\underline{t}}(z) \left(\underline{\underline{t}}_\Delta(z)^{-1} \underline{\underline{\tau}}_\Delta(z) \underline{\underline{t}}_\Delta(z)^{-1} + \left(\underline{\underline{t}}(z)^{-1} - \underline{\underline{t}}_\Delta(z)^{-1} \right) \right) \underline{\underline{t}}(z) . \tag{28.5}$$

By using repulsive V_r^n *screening potentials*, (28.3) can be solved so that $\underline{G}_r^{nm}(z) \simeq \underline{0}$ for all $|\underline{R}_n - \underline{R}_m| > d$, where d is a distance of some atomic spacings, i.e., the structure constants in (28.4) are indeed *screened* and their solution reduces to the inversion of a sparse matrix [6, 7]. In case of layered systems, the corresponding matrix gets block-tridiagonal and the required computational time scales with N [10]. Using (28.5), the τ-matrix in *screened representation* can be transformed to the physical representation and the Green's function is obtained from (28.2).

28.4 Suitability of the SKKR Method

The above described strategy of the SKKR method can uniformly be used for a non-relativistic or fully relativistic angular momentum expansion, for spin-polarized systems and for scattering potentials of spherically symmetrical or arbitrary (full-potential) shape. The SKKR method has traditionally been used for layered systems, i.e., materials with two-dimensional translational symmetry, like thin films, multilayers, surfaces and interfaces [8]. This combined with the Coherent Potential Approximation (CPA) [3] enables the investigation of substitutionally disordered random alloys described by an effective medium. The Embedded Cluster Method (ECM) [11] enables the investigation of real-space nanostructures, like impurities, surface islands, atomic contacts. The SKKR within a fully relativistic spin-polarized description is extremely suitable to study diverse magnetic properties, like local spin/orbital moments, magnetic anisotropy (MAE), domain walls [12, 13], interlayer exchange coupling (IEC), tensorial exchange interactions [14], spin wave (magnon) spectroscopy [14, 15]. Combined with the linear response Kubo-Greenwood theory, electrical (e.g., conductance, magnetoresistance) and magneto-optical transport (e.g., Kerr spectroscopy) properties can be studied at a fully relativistic first principles level. For a more detailed and structured overview the reader is referred to [8].

It is important to note that the massively parallelized KKRnano program package [16], developed mainly in the Research Center Jülich in Germany, takes advantage of the SKKR concept to provide an order-N electronic structure code suitable for studying solid-state systems consisting of tens of thousands of atoms. For more details, the reader is referred to Chap. 17 of this book.

28.5 Using SKKR to Explore Complex Magnetism
of Nanostructures

Recent advancements in experimental techniques of imaging complex magnetic structures in real space above surfaces using spin-polarized scanning tunneling microscopy [17] require theoretical efforts for the understanding of the formation of

complex magnetic patterns in thin films. Based on the SKKR method, the parameters of a classical spin Hamiltonian, including tensorial exchange interactions, can be extracted using either the relativistic torque method (RTM) [14] or the spin cluster expansion (SCE) technique [18]. A suitable spin Hamiltonian is

$$H = -\frac{1}{2}\sum_{i \neq j} \frac{1}{m_i m_j} \boldsymbol{m}_i \underline{\underline{J}}_{ij} \boldsymbol{m}_j + \sum_i \frac{1}{m_i^2} \boldsymbol{m}_i \underline{\underline{K}}_i \boldsymbol{m}_i - \sum_i \boldsymbol{m}_i \cdot \boldsymbol{b}_{\text{ext}} \,, \qquad (28.6)$$

with $\boldsymbol{m}_i$ the classical spin moment of atom i, $\underline{\underline{J}}_{ij}$ the exchange tensor, $\underline{\underline{K}}_i$ the on-site anisotropy matrix, and $\boldsymbol{b}_{\text{ext}}$ the external magnetic field. Based on this, the magnetic ground state can be estimated in the following way: The energy of a spin spiral with propagation vector $\boldsymbol{q}$ corresponds to the maximal eigenvalue of the Fourier transform of the exchange tensor. When calculating spin spiral energies by sweeping $\boldsymbol{q}$ in the Brillouin zone, the maximal obtained value corresponds to the estimated magnetic ground state. Another method to find the ground state magnetic structure is based on the zero temperature (deterministic) Landau–Lifshitz–Gilbert (LLG) equation of atomistic spin dynamics,

$$\frac{\partial \boldsymbol{m}_i}{\partial t} = -\frac{\gamma}{1+\alpha^2} \boldsymbol{m}_i \times \boldsymbol{b}_i - \frac{\alpha\gamma}{(1+\alpha^2)m_i} \boldsymbol{m}_i \times (\boldsymbol{m}_i \times \boldsymbol{b}_i) \,, \qquad (28.7)$$

where γ is the gyromagnetic ratio, α the Gilbert damping, and the effective field is $\boldsymbol{b}_i = -\partial H / \partial \boldsymbol{m}_i$.

Following this multiscale approach based on the spin Hamiltonian parameters obtained within the SKKR method, the complex ground states of a variety of magnetic thin films have been obtained ranging from spin spirals to skyrmions, for example: 1–4 monolayers (ML) of Fe on Ir(001) surface [19]; 1 ML Fe on different substrates composed of $5d$ elements [20]; PdFe double layer on Ir(111) [21]; 1 ML Fe on Rh(001) surface [22]; 1 ML Fe on W(110) and Ta(110) substrates [23]. Theoretical analysis provides information on the relative importance and competition of isotropic exchange, Dzyaloshinskii–Moriya, and in certain cases of biquadratic and higher order spin interactions, partly considering the effect of layer relaxations as well. Extending the LLG equation (28.7) to include thermal effects on the effective field [24, 25], temperature dependence of the magnetic states can also be studied [23, 26, 27].

The above examples illustrate the importance of the SKKR method to contribute to the theoretical understanding of complex magnetism at the atomic scale.

References

1. J. Korringa, Physica **XIII**, 392 (1947)
2. W. Kohn, N. Rostoker, Phys. Rev. **94**, 1111 (1954)
3. B.L. Gyorffy, Phys. Rev. B **5**, 2382 (1972)
4. J.S. Faulkner, G.M. Stocks, Phys. Rev. B **21**, 3222 (1980)
5. H. Ebert, D. Ködderitzsch, J. Minár, Rep. Prog. Phys. **74**, 096501 (2011)
6. L. Szunyogh, B. Újfalussy, P. Weinberger, J. Kollár, Phys. Rev. B **49**, 2721 (1994)
7. R. Zeller, P.H. Dederichs, B. Újfalussy, L. Szunyogh, P. Weinberger, Phys. Rev. B **52**, 8807 (1995)
8. J. Zabloudil, R. Hammerling, L. Szunyogh, P. Weinberger, *Electron Scattering in Solid Matter*, 1st edn. (Springer, Berlin, 2005)
9. W. Kohn, L.J. Sham, Phys. Rev. **140**, A1133 (1965)
10. E.M. Godfrin, J. Phys. Condens. Matter **3**, 7843 (1991)
11. B. Lazarovits, L. Szunyogh, P. Weinberger, Phys. Rev. B **65**, 104441 (2002)
12. S. Gallego, P. Weinberger, L. Szunyogh, P.M. Levy, C. Sommers, Phys. Rev. B **68**, 054406 (2003)
13. L. Balogh, K. Palotás, L. Udvardi, L. Szunyogh, U. Nowak, Phys. Rev. B **86**, 024406 (2012)
14. L. Udvardi, L. Szunyogh, K. Palotás, P. Weinberger, Phys. Rev. B **68**, 104436 (2003)
15. L. Udvardi, L. Szunyogh, Phys. Rev. Lett. **102**, 207204 (2009)
16. KKRnano project (2016), http://www.fz-juelich.de/ias/jsc/EN/Expertise/High-Q-Club/ KKRnano/_node.html. Cited 30 Sep 2016
17. R. Wiesendanger, Rev. Mod. Phys. **81**, 1495 (2009)
18. L. Szunyogh, L. Udvardi, J. Jackson, U. Nowak, R. Chantrell, Phys. Rev. B **83**, 024401 (2011)
19. A. Deák, L. Szunyogh, B. Ujfalussy, Phys. Rev. B **84**, 224413 (2011)
20. E. Simon, K. Palotás, B. Ujfalussy, A. Deák, G.M. Stocks, L. Szunyogh, J. Phys. Condens. Matter **26**, 186001 (2014)
21. E. Simon, K. Palotás, L. Rózsa, L. Udvardi, L. Szunyogh, Phys. Rev. B **90**, 094410 (2014)
22. A. Deák, K. Palotás, L. Szunyogh, I.A. Szabó, J. Phys. Condens. Matter **27**, 146003 (2015)
23. L. Rózsa, L. Udvardi, L. Szunyogh, I.A. Szabó, Phys. Rev. B **91**, 144424 (2015)
24. L. Rózsa, L. Udvardi, L. Szunyogh, J. Phys. Condens. Matter **25**, 506002 (2013)
25. L. Rózsa, L. Udvardi, L. Szunyogh, J. Phys. Condens. Matter **26**, 216003 (2014)
26. G. Hasselberg, R. Yanes, D. Hinzke, P. Sessi, M. Bode, L. Szunyogh, U. Nowak, Phys. Rev. B **91**, 064402 (2015)
27. L. Rózsa, E. Simon, K. Palotás, L. Udvardi, L. Szunyogh, Phys. Rev. B **93**, 024417 (2016)