Implementation of non-equilibrium vertex corrections in KKR: transport through disordered layers

Christian Franz, Michael Czerner and Christian Heiliger

I. Physikalisches Institut, Justus Liebig University, Giessen, Germany

E-mail: Christian.Heiliger@physik.uni-giessen.de

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Abstract
The theoretical description of modern nanoelectronic devices requires a quantum mechanical treatment and often involves disorder, e.g. from alloys. Therefore, the ab initio theory of transport using non-equilibrium Green’s functions is extended to the case of disorder described by the coherent potential approximation. This requires the calculation of non-equilibrium vertex corrections. We implement the vertex corrections in a Korringa–Kohn–Rostoker multiple scattering scheme. In order to verify our implementation and to demonstrate the accuracy and applicability we investigate a system of an iron-cobalt alloy layer embedded in copper. The results obtained with the coherent potential approximation are compared to supercell calculations. It turns out that vertex corrections play an important role for this system.

(Some figures may appear in colour only in the online journal)

1. Introduction
Modern applications require an accurate description of transport processes on the nanometre scale, where quantum coherence effects cannot be neglected, making fully quantum mechanical calculations necessary. A popular tool for these calculations is the non-equilibrium Green’s function (NEGF) method (or Keldysh NEGF) [1]. This method can be implemented within the Korringa–Kohn–Rostoker (KKR) multiple scattering scheme [2, 3]. Advantages of the NEGF method include the capability to calculate the transport under applied bias self-consistently [4, 5], the simple inclusion of inelastic scattering events by additional self-energies [1], and a numerically more stable description of the transmission coefficient in comparison with methods using the current operator [2].

The presence of chemical disorder from alloying breaks the translational symmetry of the crystal and thus complicates the calculation. In order to restore periodicity one can use an effective medium scheme, i.e. the alloy is replaced by an effective medium which approximates the properties of the alloy but has the periodicity of the underlying lattice. The most popular scheme is probably the coherent potential approximation (CPA) [6]. In the CPA, the effective medium is defined by a self-consistent condition. The idea is that the Green’s function (GF) of the effective medium should resemble the full configurational average of the alloy GF. Therefore, the single-site CPA condition requires that the additional scattering from a real atom placed on one site in the effective medium crystal averages to zero when the alloy average is taken. While this approximation is very successful in many applications, it neglects the effect of correlations due to local clusters in the alloy. The single-site CPA can be extended to include several sites, e.g. in the nonlocal CPA [7], thus taking the effects of short range order into account.

An alternative approach is the use of large supercells averaging over a large number of configurations. The drawbacks of this method are high computational cost, possible supercell effects, and the limited number of realizable concentrations. It has the advantage that it can be applied within most band structure methods without additional implementation. Further, the accuracy of the description can be enhanced simply by using larger supercells and more configurations. In this sense, it is possible to find the results...
2. Non-equilibrium Green’s functions method

The NEGF method was successfully implemented in the KKR scheme [2, 3]. Here, we present a more detailed and more general derivation of the formulae. The usual way to apply the NEGF formalism to a system with steady state transport is to divide the system of interest into three regions along the transport direction [1]: a middle region connected to a left and a right lead (see figure 1). The middle is the region of interest and contains e.g. the alloy or a potential barrier. It is contacted by the leads, which are considered as ideal conductors. To describe the influence of the leads on the middle region one considers a fictitious decoupled system with a Hamiltonian

$$H_{dc} = \begin{pmatrix}
h_L & 0 & 0 \\
0 & h_M & 0 \\
0 & 0 & h_R \\
\end{pmatrix}$$

(1)

and the corresponding Green’s function (GF)

$$\tilde{G}_{dc} = [z - H_{dc}]^{-1} = \begin{pmatrix}
\tilde{G}_L & 0 & 0 \\
0 & \tilde{G}_M & 0 \\
0 & 0 & \tilde{G}_R \\
\end{pmatrix}.$$  

(2)

Here, $z = E + i \eta$ is the complex energy. We will always assume $\eta \geq 0$, thus $G(E) = \lim_{\eta \rightarrow 0} G(z)$ is a retarded and $G(E)^\dagger = \lim_{\eta \rightarrow 0} G(z^*)$ an advanced GF. All physical quantities (density, transmission, etc) are obtained by taking the limit $z \rightarrow E$, i.e. $\eta \rightarrow 0^+$. In practice, this limit is taken by using a sufficiently small $\eta$, determined by convergence tests. The coupling between the leads and the middle region is given by the coupling matrix

$$\tau = \begin{pmatrix}
0 & \tau_{LM} & 0 \\
\tau_{ML} & 0 & \tau_{MR} \\
0 & \tau_{RM} & 0 \\
\end{pmatrix}. $$

(3)

This includes the assumption that there is no direct coupling between the left and the right lead but only indirect coupling via the middle region. Thus, the full system is described by the Hamiltonian $H_c = H_{dc} + \tau$ and the overall GF of the coupled system

$$G_c = [z - H_{dc} - \tau]^{-1} = \begin{pmatrix}
G_{LL} & G_{LM} & G_{LR} \\
G_{ML} & G_{MM} & G_{MR} \\
G_{RL} & G_{RM} & G_{RR} \\
\end{pmatrix}. $$

(4)

By solving this system of equations for $G_{MM}$, the GF of the middle region coupled to the leads, $G_{MM}$ can be described by

$$G_{MM} = [z - h_M - \Sigma]^{-1}, $$

(5)

where the so-called self-energy of the leads is introduced

$$\Sigma = \Sigma_L + \Sigma_R, $$

$$\Sigma_L = \tau_{ML} \tilde{G}_L \tau_{LM}, $$

$$\Sigma_R = \tau_{RM} \tilde{G}_R \tau_{RM}. $$

(6)

These self-energies can be interpreted as fluxes of incoming and outgoing electrons at the connection between leads and the middle region [1]. They describe the influence of the semi-infinite leads on the middle region and are determined by the surface GFs of the leads and the coupling to the middle region. The coupled and decoupled GFs in the middle region are connected by the self-energies via the Dyson equation

$$G_{MM} = \tilde{G}_M + \tilde{G}_M \Sigma_L G_{MM} + \tilde{G}_M \Sigma_R G_{MM}. $$

(7)
Using the self-energies and the coupled GF the density can be calculated [1, 13, 14]:

\[ n(E) = \frac{1}{2\pi} G_{\text{MM}}(E) \Gamma(E) G_{\text{MM}}^\dagger(E), \]  

(8)

where \( \Gamma(E) = i(\Sigma(E) - \Sigma(E)\dagger) \). The density can be decomposed into the fraction of electrons originating from the left or right lead by using \( \Gamma L = i(\Sigma L - \Sigma L\dagger) \) or \( \Gamma R = i(\Sigma R - \Sigma R\dagger) \) instead of \( \Gamma = \Gamma L + \Gamma R \). This way, it is possible to obtain the non-equilibrium density or, in other words, the density of the transport electrons. This is required, for example, to calculate the spin-transfer torque [3, 15]. Another application is the analysis of the transport states, e.g., during tunnelling.

On the other hand, the equilibrium density can also be calculated by [6]

\[ n(E) = -\frac{1}{\pi} \text{Im} [G_{\text{MM}}(E)]. \]  

(9)

Now, \( n(E) \) in (8) involves a product of GFs, whereas (9) does not. Therefore, for \( n(E) \) in (8) vertex corrections are required in the presence of CPA. But both have to give the same results in equilibrium. Consequently, this provides a simple and stringent way to test the vertex corrections, including their convergence.

Many transport properties of the system can be calculated from the transmission function \( T(E) \), which is given by [1, 16]

\[ T(E) = \text{Tr}[G_{\text{MM}}(E) \Gamma_L(E) G_{\text{MM}}(E)\dagger \Gamma_R(E)], \]  

(10)

where the trace is over the basis set.

In the KKR method, the GF of the coupled system is known and can be calculated via the decimation technique [6]. A difficulty occurs when describing the self-energy in terms of the decoupled system. The KKR method works with GFs and has no finite couplings \( \tau \) which could be set to zero. To solve this problem, one can introduce an artificial potential \( V_{\text{dc}} = V_{\text{dc}}^L + V_{\text{dc}}^R \) [2, 3], which decouples the middle region from the leads (see figure 1). In this case, the decoupled system is described by \( H_{\text{dc}} = H_c + V_{\text{dc}} \). This potential difference connects the two GFs via the Dyson equation

\[
\begin{align*}
G_c &= \tilde{G}_{\text{dc}}(V_{\text{dc}}^L + V_{\text{dc}}^R) G_{\text{dc}} \\
&= \tilde{G}_{\text{dc}}(V_{\text{dc}}^L + V_{\text{dc}}^R) G_{\text{dc}} + \tilde{G}_{\text{dc}}(V_{\text{dc}} + V_{\text{dc}}^L) G_{\text{dc}} + \tilde{G}_{\text{dc}}(V_{\text{dc}} + V_{\text{dc}}^R) G_{\text{dc}},
\end{align*}
\]  

(11)

where the second equation is derived by inserting the Dyson equation into itself. Some of the terms arising can be neglected if the following assumptions are made (written schematically)

\[
\tilde{G}_{\text{dc}}(V_{\text{dc}} + V_{\text{dc}}^L) G_{\text{dc}} \ll \tilde{G}_{\text{dc}}(V_{\text{dc}} + V_{\text{dc}}^R) G_{\text{dc}}
\]

(12)

and

\[
\begin{align*}
\tilde{G}_{\text{dc}}(V_{\text{dc}} + V_{\text{dc}}^L) G_{\text{dc}} &= \tilde{G}_{\text{dc}}(V_{\text{dc}}^L + V_{\text{dc}}^R) G_{\text{dc}} + \tilde{G}_{\text{dc}}(V_{\text{dc}}^L + V_{\text{dc}}^R) G_{\text{dc}} + \tilde{G}_{\text{dc}}(V_{\text{dc}}^L + V_{\text{dc}}^R) G_{\text{dc}}.
\end{align*}
\]  

(13)

The first assumption is obviously true if \( 1 \ll (V_{\text{dc}}^L + V_{\text{dc}}^R) G_c \), which can be ensured by choosing an appropriately high decoupling potential \( V_{\text{dc}} \). The second assumption makes sure that the self-energy of the leads can be written as a sum of right and left lead self-energy. This is true if the leads are well separated because the elements of \( \tilde{G}_{\text{dc}} \) relating the left and the right leads decay exponentially with respect to the thickness of the decoupling potential. By comparing the remaining terms with (7)

\[
G_c = \tilde{G}_{\text{dc}} G_{\text{dc}} + \tilde{G}_{\text{dc}}(V_{\text{dc}}^L + V_{\text{dc}}^R) G_{\text{dc}} + \tilde{G}_{\text{dc}}(V_{\text{dc}}^L + V_{\text{dc}}^R) G_{\text{dc}} + \tilde{G}_{\text{dc}}(V_{\text{dc}}^L + V_{\text{dc}}^R) G_{\text{dc}}
\]

(14)

one can identify the self-energies in the KKR scheme with

\[
\begin{align*}
\Sigma_L &= V_{\text{dc}}^L G_{\text{dc}} V_{\text{dc}}^L, \\
\Sigma_R &= V_{\text{dc}}^R G_{\text{dc}} V_{\text{dc}}^R.
\end{align*}
\]  

(15)

3. NEGF in the Korringa–Kohn–Rostoker basis

In the KKR scheme the Green’s function (GF) in cell centred coordinates can be written as [6]

\[
G(r + r', r' + R_n; z) = \delta_{n, z} g^{n}_{\text{dc}}(r, r'; z) + R_n(r; z) g^{n\prime}(r; z) \delta R_n(r; z),
\]

(16)

where \( R_n \) is the regular solution of the single scatterer (isolated atom) at site \( n \), \( g_{\text{dc}} \) is the single scatterer GF, and \( g^{\prime}\) is the so-called structural GF. All these quantities carry angular momentum indices, which are either \( L = (l, m, s) \) for non-relativistic treatment or the \( Q = (\kappa, \mu) \) representation in the full-relativistic case. The \( R_n \) are row vectors in angular momentum space and can be written as \( R_n(r; z) = [R_n^L(r; z) Y_L(r/r)] \) for the case of a non-relativistic treatment with spherical potentials. In accordance with the vector notation of [6] we define \( R_n(r; z)^\prime = [R_n^L(r; z)^\prime Y_L(r/r)]^\dagger \), where \( ^\dagger \) stands for the transpose. For the other quantities we use the usual matrix notation, e.g., \( g^{\prime\prime} = [g^{\prime\prime}_{LL}] \) and \( g = [g^{\prime\prime}] \). For the full-potential and full-relativistic case it will be sufficient to note that they can be written in the form of (16).

By inserting the KKR GF into the transport formula (10) and into the density formula (8) one obtains

\[
T(E) = \lim_{\varepsilon \to E} \text{Tr}[g(z) \gamma_L(z, z^\prime) g(z^\prime) \gamma_R(z^\prime, z)],
\]

(17)

\[
n_n^{L/R}(r, E) = \frac{1}{2\pi} \lim_{\varepsilon \to E} R_n(r; z) g^{<, n}_{L/R}(r) R_n(r; z)^\prime \delta R_n(r; z),
\]

(18)

with

\[
g^{<, n}_{L/R}(z) = \sum_k g^{<, n}_{kL/R}(z) Y_k^L(r/r) Y_k^L(z^\prime),
\]

(19)

where the trace runs over atoms and basis representation and the quantity \( \gamma_L \) is defined as
Please note that the single scatterer GF $g_{dc}^\alpha$ in (16) does not contribute in (17) and (19) because only non-site-diagonal elements of the GF enter into these formulae. On the other hand, the single scatterer GF enters in (20) as we see in the following.

Using the definition of $\Gamma(z) = i(\Sigma(z) - \Sigma(z^*))$ one gets

$$-i\gamma_{L/R}^{mn}(z, z^*) = \int \mathrm{d}r \int \mathrm{d}r' R^\nu(r, z) \times \Gamma_{L/R}^{mn}(r, r'; z) R^\nu(r'; z^*)$$

With the use of the self-energy (15) and the KKR GF (16), $\gamma_{L/R}$ is given by

$$-i\gamma_{L/R}^{mn}(z, z^*) = \int \mathrm{d}r \int \mathrm{d}r' R^\nu(r, z) \times \left[ V^{\nu}(r) \left( \tilde{g}_{dc}^\nu(r, r'; z) \delta_{nn'} \right. \right. + R^{\nu}_{dc}(r) \tilde{g}_{dc}^\nu(r, r'; z) \delta_{nn'} + R^{\nu}_{dc}(r') \tilde{g}_{dc}^\nu(r', z) V^{\nu}(r') \left. \left. - V^{\nu}_{dc}(r) \left( \tilde{g}_{dc}^\nu(r', r'; z^*) \delta_{nn'} \right. \right. \right. + R^{\nu}_{dc}(r') \tilde{g}_{dc}^\nu(r', z^*) V^{\nu}(r') \left. \left. \right. \right] R^\nu(r'; z^*)$$

where for $\gamma_{L/R}$ both site indices $n, n'$ are restricted to sites where $V_{dc,L}(V_{dc,R})$ is nonzero. The Lippmann–Schwinger equation for the single scatterer solution

$$\int \mathrm{d}r \ R^\nu(r, z) \times V_{dc}^\nu(r) \tilde{g}_{dc}^\nu(r, r'; z) \delta_{nn'}$$

reduces the expression to the simple form

$$-i\gamma_{L/R}^{mn}(z, z^*) = \delta_{nn'} \left( -\Delta^\nu(z, z^*) + \Delta^\nu(z^*, z) \right) + \Delta^\nu(z) \tilde{g}_{dc}^\nu(z) \delta_{nn'} \Delta^\nu(z^*, z) - \Delta^\nu(z^*, z) \tilde{g}_{dc}^\nu(z^*) \Delta^\nu(z^*, z),$$

where the definition of the $t$-matrix, $\Delta^\nu(z)$ and an expression $\Delta^\nu(z, z^*)$ similar to the $t$-matrix were used:

$$\Delta^\nu(z) = \int \mathrm{d}r \ R^{\nu}_{dc}(r; z) \times V^{\nu}_{dc}(r) R^{\nu}(r; z)$$

$$\Delta^\nu(z, z^*) = \int \mathrm{d}r \ R^{\nu}_{dc}(r; z) \times V^{\nu}_{dc}(r) R^{\nu}(r; z^*).$$

In a similar procedure one can calculate the quantity $\gamma_{L/R}(z^*, z)$ with interchanged energy variables

$$-i\gamma_{L/R}^{mn}(z^*, z) = \delta_{nn'} \left( -\Delta^\nu(z, z^*)^* + \Delta^\nu(z^*, z)^T \right) + \Delta^\nu(z, z^*)^* \tilde{g}_{dc}^\nu(z) \Delta^\nu(z^*, z)^T - \Delta^\nu(z^*, z) \tilde{g}_{dc}^\nu(z^*) \Delta^\nu(z^*, z^*)^T.$$  

This is the representation of the operator $\Gamma$ in the KKR basis similar to the structural GF $g_{dc}^\alpha(z)$. The task of calculating the self-energies is transformed into the task of calculating the structural GF of the system with the decoupling potential. However, in the screened KKR [6] one solves a system with a repulsive potential, the reference system, which already fulfills the requirements of the decoupling potential. With the help of the screened KKR and the decimation technique the surface GF of a system can be calculated, which turns out to be the GF of the decoupled system needed in (22) and (25).

Another point is the use of $z$ and $z^*$ in (22) and (25), which requires solving the involved terms for two energies. One can make use of the following properties

$$\tilde{g}_{dc}(z) = \tilde{g}_{dc}(z^*)^T$$

$$\Delta^\nu(z) = \Delta^\nu(z^*)^T$$

$$\Delta^\nu(z, z^*) = \Delta^\nu(z^*, z)^T,$$

to decrease the numerical effort by solving the $t$-matrix, $\rho^\nu$, and the structural GF $\tilde{g}_{dc}$ only for the energy $z$. However, the modified $t$-matrix $\Delta^\nu$ given by (24) still requires the single scattering wavefunctions for $z$ and $z^*$. In the non-relativistic case with spherical potentials the scattering solutions $R^\nu(r; z)$ obtain a simple phase factor from changing the energy from $z$ to $z^*$ [17]. The screened reference system in the KKR is usually calculated for this case and is then transformed into the required representation (full-relativistic or full-potential or both). This is exact and not an approximation because the reference system has no physical meaning. Therefore, one can simply calculate $R^\nu_{dc}(r, z^*)$ from $R^\nu_{dc}(r, z)$ even for the full-potential and full-relativistic case. With this, one can obtain $\gamma$ by solving all quantities only at energy $z$.

4. Coherent potential approximation

In the single-site KKR-CPA the effective medium corresponds to placing (site-diagonal) effective medium $t$-matrices $T^{nn'} = \delta_{nn'} \tilde{T}$ on all alloy sites. The $\tilde{T}$ are chosen to restore the periodicity of the underlying lattice and hence the Green’s function of the effective medium (GF), defined by

$$\tilde{g}(z) = [g(z)^{-1} - \tilde{T}(z)]^{-1},$$

can be calculated using standard methods (lattice Fourier transform, decimation [6]), where $\tilde{g}$ is the free GF. Following Butler [8], we can show that the alloy GF for a fixed configuration and the effective medium GF are related by

$$g^{nn'}(z) = \tilde{g}^{nn'}(z) + \sum_{n'' n'''} g^{nn'''}(z) T^{n'n'''}(z) \tilde{g}^{n''n'}(z).$$

The alloy $T$-matrix can be calculated from

$$T^{nn'}(z) = x^{nn}(z) \left( \delta_{nn'} + \sum_{n'' \neq n} g^{nn''}(z) T^{n'n''}(z) \right) x^{nn'}(z).$$
where $x^a$ is given by
\begin{equation}
  x^a(z) = \left[ 1 - (t^a(z) - P^a(z)) \tilde{g}^m(z) \right]^{-1} (t^a(z) - P^a(z)).
\end{equation}

Here, the lattice sites $t^a$ are occupied according to the fixed configuration. The $x^a$ describe the additional scattering of the alloy relative to the effective medium.

Using the single site approximation (SSA) [18] leads to the decoupling of the averaged $T$-matrix \( (T) = (x^\alpha) (\delta_{\alpha\beta} + \sum_{n',n''} g_{\alpha n'} (T^n_{n''})) \). Therefore, if the single-site CPA condition \( (x^\alpha) = 0 \) is met, we find \( (T) = 0 \) and the GF of the configurationally averaged system is identical to the GF of the effective medium \( \tilde{g}^m(z) = \tilde{g}^m(z) \).

For the non-equilibrium applications we need to calculate alloy averages like \( \langle G A G \rangle \) for some operator $A$. In order to express this in terms of the effective medium GF one demands the relation
\begin{equation}
  \langle g(z) A(z) g(z') \rangle = \tilde{g}(z) A(z) \tilde{g}(z') + \tilde{g}(z) \Omega_A(z) \tilde{g}(z'),
\end{equation}
where the first term on the right-hand side represents the coherent contribution while the second term defines the non-equilibrium vertex corrections (NVC) $\Omega_A$. As we will show, the latter are the result of multiple scattering relative to coherent but damped motion in the effective medium. Thus, they can be interpreted as accounting for diffusive contributions. Note that the NVC are specific to the operator $A$.

As explained below, in our case the operator $A$ does not depend on the alloy configuration and using (30) and the SSA we find that
\begin{equation}
  \Omega_A(z) = (T(z) \tilde{g}(z) A(z) \tilde{g}(z') T(z')).
\end{equation}
Using (31) and the SSA we find that the vertex corrections are site-diagonal $\tilde{\Omega}_A = \delta_{\alpha\beta} \Omega_A$ and a closed set of equations can be derived [10-12]:
\begin{equation}
  \Omega_A(z) = \left( x^\alpha(z) \right) \frac{\delta_{\alpha\beta}}{\tilde{g}^m(z)} \left[ \tilde{g}(z) A(z) \tilde{g}(z') \right]_{\alpha\beta} x^\beta(z')
  + \sum_{n' \neq n} \left( x^n(z) \tilde{g}^m(z) \Omega^\alpha_{n'} A(z) \tilde{g}^n(z') x^\alpha(z') \right)
  = \sum_{\alpha} \delta_{\alpha} \delta^\alpha(z) \frac{\tilde{g}(z) A(z) \tilde{g}(z')}{\tilde{g}(z)} \left[ \tilde{g}(z) A(z) \tilde{g}(z') \right]_{\alpha\alpha} x^\alpha(z')
  + \sum_{\alpha \neq \alpha} \tilde{g}(z) A(z) \tilde{g}(z') \Omega^\alpha_{\alpha'} \tilde{g}^n(z') x^\alpha(z'),
\end{equation}
where $\alpha$ enumerates the species on site $n$, $n^\alpha(z)$ is the particular concentration, and $x^\alpha(z)$ is $x^\alpha$ when the site is occupied by $\alpha$. The NVC are zero for non-alloy sites, thus one has to calculate the NVC only for CPA sites.

In order to make use of the two-dimensional translational symmetry, we complete the sum over $n'$ and transform the infinite lattice sums to $k_1$-space integrations over the first Brillouin zone. Writing $n \rightarrow S + T_1$, where $T_1$ is a two-dimensional lattice vector and $S$ belongs to the primitive cell, we find
\begin{equation}
  \Omega_A(z) = \left( x^\alpha(z) \right) \frac{\delta_{\alpha\beta}}{\tilde{g}^m(z)} \left[ \tilde{g}(k_1; z) A(k_1; z) \tilde{g}(k_1; z') \right]_{\alpha\beta}
  \times \frac{\tilde{g}(z)}{\tilde{g}(z)} \left[ \tilde{g}(k_1; z) A(k_1; z) \tilde{g}(k_1; z') \right]_{\alpha\alpha}
  + \sum_{\alpha} \tilde{g}(z) \Omega^\alpha_{\alpha'} \tilde{g}^n(z') x^\alpha(z').
\end{equation}
Note that $\Omega_A$ is site-diagonal and hence does not depend on $k_1$. Therefore, we could pull $\Omega_A$ out of the $k_1$-sum and solve the linear equation by matrix inversion. Alternatively, one can obtain $\Omega_A$ directly by iterating (36). Matrix inversion is in general faster than the iterative procedure, but needs the full coupling matrix, which can become very large. The iterative procedure has the advantage that it allows for a splitting of the problem into smaller tasks (the vertex correction for one atom), which can be utilized in parallel computing. Additionally, only a part of the complete matrix is needed for every single task, which results in smaller memory consumption per task. It should be noted that the inversion has to be done only once after the Brillouin zone integration.

The theory of NVC can be directly applied to the transport equation. In this case, the operator $A$ is the self-energy of the left or right lead. Since the alloy is only inside the middle region $\gamma_{L/R}$ does not depend on the alloy configuration. Using (33) we find
\begin{equation}
  \langle T(E) \rangle = \lim_{E \rightarrow E} \text{Tr} [\tilde{g}(z) \gamma_{L,R} \tilde{g}(z') E_{\alpha}(z)|E_{\alpha}(z')].
\end{equation}
The averaged density at site $n$ is expressed as the weighted sum over the densities of the components
\begin{equation}
  \langle n^\alpha(r; E) \rangle = \lim_{E \rightarrow E} \sum_{\alpha} n^\alpha_n(r; z).
\end{equation}
This is necessary since the effective medium does not provide a scattering solution $R^\alpha$ or single scatterer GF $\tilde{g}^m_{\alpha\alpha}$. The component densities can be calculated from restricted averages, where only the atom on site $n$ is fixed. In equilibrium, they are obtained from projections of the effective medium GF [6] via the impurity matrix $D_n^\alpha$
\begin{equation}
  n^\alpha_n(r; z) = \langle \tilde{n}^\alpha(r; z) \rangle_{m=a}
  = \frac{-1}{\pi} \text{Im} \left[ D_n^\alpha(r; z) \tilde{g}^m_{\alpha\alpha}(z) R^\alpha_n(r; z) \right].
\end{equation}
For the non-equilibrium density in the presence of CPA alloys, $g^{<\alpha\alpha}$ in (18) is replaced by
\begin{equation}
  g^{<\alpha\alpha}(z) = \sum_{kl} \tilde{g}^{k\alpha}(z) \left( \gamma^{k\alpha}(z; z^*) + \delta_{k\alpha} \Omega^{k\alpha}_{\alpha}(z) \tilde{g}^{k\lambda}(z^*) \right)
  \text{for all sites. This means that the NVC also influence the non-equilibrium density for non-alloy sites.}
\end{equation}
Again, on alloy sites component densities must be used. Following [12], the component non-equilibrium densities for the special case of a binary alloy with the components $A$ and $B$ can be calculated via
\begin{equation}
  n^\alpha_n(r; z) = \frac{1}{2\pi} R^\alpha_n(r; z) \tilde{g}^{<\alpha\alpha}(z) R^\alpha_n(r; z)^*,
\end{equation}
where the quantity $g_A^{<,\alpha}(z)$ is given by

$$
\tilde{g}_A^{<,\alpha}(z) = (\tilde{\gamma}_A(z) - \tilde{\gamma}_0(z))^{-1} \left[ (\tilde{\gamma}_A(z) - \tilde{\gamma}_0(z)) \tilde{g}_A^{<,\alpha}(z) - \Omega_A^\alpha(z) \tilde{g}_A^{\alpha,\alpha}(z) \right],
$$

$$
\tilde{g}_B^{<,\alpha}(z) = (\tilde{\gamma}_B(z) - \tilde{\gamma}_0(z))^{-1} \left[ (\tilde{\gamma}_B(z) - \tilde{\gamma}_0(z)) \tilde{g}_B^{<,\alpha}(z) - \Omega_B^\alpha(z) \tilde{g}_B^{\alpha,\alpha}(z) \right],
$$

Unfortunately, the derivation in [12] relies on a system of equations, which is only closed and solvable for binary alloys. It does not allow for an obvious generalization to multicomponent alloys. Such a generalization is an important task for future developments.

5. Results

In order to have a convincing test of the NVC we calculate the transport through an alloyed layer, comparing results from CPA calculations with and without NVC to supercell calculations. We choose a system of 10 atomic layers of disordered Fe$_x$Co$_{1-x}$ alloy in the bcc-structure of Fe connected to leads, which consist of Cu in the Fe bcc-structure. The applicability of the CPA for this material is well established [19]. For the supercells we use 16 atoms in-plane and average the results over 10 random configurations for each concentration. The self-consistent potentials were obtained using our all-electron screened KKR code within the local spin-density approximation (LSDA) and with a non-relativistic treatment. We use the parametrization of Vosko–Wilk–Nusair [20] for the local exchange–correlation potentials. Since we are interested in a test of the transport properties, we use CPA potentials for the supercell. The alloy and the Cu leads are considered in bcc-structure with $a = 0.287$ nm. The calculations were performed for concentrations in the range from 0 to 100%. The integrations in the 2D BZ were performed on a uniform mesh of $24^2$ k-points for the self-consistent calculation and $160^2$ ($40^2$) k-points at $\eta = 2 \times 10^{-5}$ Ryd for the CPA (supercell) transport calculations.

The results in figure 2 show that the CPA results including NVC are in excellent agreement with the supercell calculations and that the vertex corrections are necessary to obtain correct results. It is salient that the NVC are dominant in the spin-down channel while they are small in the spin-up channel. This can be easily understood by looking at the Bloch spectral density [21] $A^\alpha(k, E)$, which is shown in figure 3 for bulk Fe$_{0.5}$Co$_{0.5}$. We can see that the bands for spin-down show strong broadening at the Fermi-energy corresponding to strong alloy scattering. This leads to a strong effect of the vertex corrections and makes them non-negligible. However, the Bloch spectral density at the Fermi-energy of the spin-up bands is rather sharp and indicates weak alloy scattering at this energy, which results in small vertex corrections for the spin-up conductance. This demonstrates a severe difficulty for judging the importance of the NVC, since the impact of alloy scattering can be strongly energy dependent. Generally, neglecting the NVC (i.e. neglecting the diffusive current) breaks current conservation [10] and can lead to unphysical results.

We can also use the non-equilibrium density (8) to get an even more stringent test using the fact that

$$
n^L + n^R = n^{equ},
$$

where $n^{equ}$ is the equilibrium density. The NVC for the non-equilibrium density are calculated using (35). It is also necessary to perform projections onto the species resolved densities (see (38)–(42)). We perform this test for the FeCo system. The results are summarized in figure 4. One can see that the test is satisfied. The iterative solution shows sufficient convergence after about 100 iterations. On the other hand, the calculation of the NVC by inversion shows a perfect match with the equilibrium density. We find the same agreement for the component densities (not shown).

6. Conclusion

We have implemented the coherent potential approximation (CPA) and the necessary non-equilibrium vertex corrections.
(NVC) in a KKR method. This makes accurate *ab initio* description of alloys in equilibrium and non-equilibrium systems possible. The CPA includes the incoherent scattering of Bloch waves in the description. This leads to a broadening of the energy levels of the states, which is visible in the Bloch spectral density. It also leads to diffusive transport described by the NVC. We validate our implementation by calculating transport through FeCo alloys and comparing the CPA and supercell results. Additionally, we check an identity for the non-equilibrium density, which demonstrates the correctness of the non-equilibrium density and the projections. Our results emphasize the importance of the NVC.

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**Figure 4.** Layer and spin resolved density of states at the Fermi-energy $n(E_F)$ in a thin layer of Fe$_{0.3}$Co$_{0.7}$ between Cu leads, comparing the equilibrium density with several stages of the NVC calculation. Note that the non-equilibrium density is not defined in the outermost layers of the leads.

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