Development of a first-principles code based on the screened KKR method for large super-cells

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Abstract. The procedures of performing first-principles electronic structure calculation using the Korringa–Kohn–Rostoker (KKR) and the screened KKR methods are reviewed with an emphasis put on their numerical efficiency. It is shown that an iterative matrix inversion combined with a suitable preconditioning greatly improves the computational time of screened KKR method. The method is well parallelized and also has an $O(N)$ scaling property.

1. Introduction
First-principles electronic structure calculations based on the density functional theory (DFT) have been widely used and are now indispensable for both basic condensed matter physics and applications. Microscopic information obtained by first-principles calculations not only helps us to understand the physics behind experimental results but also makes it possible for us to predict properties of materials that have not been synthesized yet. This even opens up a possibility of computational materials design (CMD). However, in order for CMD to be a reality, it is necessary to execute first-principles calculations for larger scale systems than ever. In a first-principles calculation based on the DFT, the ground state electronic structure $\rho(r)$ (or the corresponding one body effective potential $V_{\text{eff}}(r)$) which satisfies the variational principle of energy functional $\delta E[\rho(r)] = 0$ is calculated by solving the so-called Kohn-Sham equations self-consistently [1, 2]. In each Kohn-Sham iteration cycle, one-body Schrödinger-like equation must be solved. As long as the usual band structure calculation algorithms are used, the numerical costs are proportional to the third power of the system size. Due to this property, large scale electronic structure calculations using a traditional method need large amount of numerical resources and computational time. To overcome these difficulties, algorithms and codes which have a property of $O(N)$ have been developed for many years. $O(N)$ means that the total steps of the calculation is proportional to the system size (i.e., the number of atoms $N$ in the system).

Among many other methods solving the Kohn-Sham equations, the KKR Green’s function method is one of the most accurate and powerful methods. Moreover, screened KKR, an extension of the method to $O(N)$, was introduced in 1995 [3]. The most time consuming part in the KKR Green’s function method is the process of inverting a matrix having the dimension proportional to the number of atoms in the system. In the screened KKR, by choosing a suitable reference system such that the matrix pattern becomes sparse, and by combining the matrix sparsity with an efficient matrix solver, the total computational work is greatly reduced without sacrificing the accuracy. The screened KKR method has been mainly applied to multilayered...
systems [4]. For multilayered systems, assuming 2D periodicity, the unit cell is extended to the layer growth direction. It was shown that in this case the total step needed for the calculation is exactly proportional to the number of layers $N$. Then, the electronic structure calculations for a system containing layers larger than ten thousands become possible on even a single CPU of present-day desktop PCs. This is due to the fact that in such layered systems, the problem is reduced to a matrix inversion of a simple block tridiagonal sparse matrix. Beyond multilayered structures, if the unit cell is extended to $x$, $y$, and $z$ directions, the matrix pattern becomes general block sparse. As long as a direct method like $LU$ factorization, which is used in traditional KKR and screened KKR for multilayered systems, is used, it is very difficult to obtain sufficient efficiency for the matrix inversion of these general block sparse matrices. Recently, high efficiency in using an iterative method, which has been widely used to solve simultaneous linear equations with sparse coefficient matrix, was proven for these large super-cell systems [5]. The electronic structure calculation code, which uses the iterative matrix inversion technique, for super-cell systems with massively parallel computing ‘KKRnano’ was developed by the Jülich group [6, 7]. Referring to their work, we have developed a first-principles electronic structure calculation code for super-cell systems by using the screened KKR method, based on the full-potential KKR (FP-KKR) code that was developed in our group [8].

We briefly outline the KKR Green’s function method in section 2. Section 3 describes the concept of screened KKR and explains how $O(N)$ scaling properties are obtained for both multilayered systems and general large super-cell systems. Implementation of parallel computing using the iterative method as a sparse matrix solver is also discussed. In section 4, results of some test calculations are shown. The efficiency of the iterative method when combined with a suitable preconditioning is confirmed. Section 5 is devoted for summary.

2. KKR Green’s function method

2.1. KKR Green’s function method

In contrast to usual schemes of first-principles methods, in the KKR Green’s function method, instead of solving eigenvalue problems, the corresponding one-body Green’s function $G(r, r', E)$ which satisfies

$$[-\Delta + V_{\text{eff}}(r) - E]G(r, r', E) = -\delta(r - r')$$

is calculated. The electron density $\rho(r)$ is constructed directly from the energy integration of the Green’s function up to the Fermi level $E_F$ [9]:

$$\rho(r) = \frac{1}{\pi} \Im \int_{-\infty}^{E_F} dE G(r, r, E)$$

$G(r, r', E)$ has a very complex structure near the real energy axis, and evaluation of the integral (2) along real energy axis requires many energy sampling points. However, off the real energy axis, $G(r, r, E)$ becomes structureless without any singularity on the upper half of complex energy plane. Therefore by an analytic continuation, the integral (2) can be evaluated by numerical integration with a few tens of energy sampling points accurately. Practically, core electrons can be considered as well localized and they can be determined by atomic calculation for given effective potentials. In this case the complex energy contour can start from $E_B$ (between the bottom of valence band and the top of core level) and approaches to $E_F$ through the upper half of complex energy plane. This procedure is important not only for the numerical integration but also for the matrix inversion in the iterative method to obtain the rapid convergences.

2.2. Calculation of crystal Green’s function

The one-body Green’s function in a crystal is obtained by multiple scattering theory. The system is divided into cells around each lattice point and local effective potential $V_n(r)$ for the $n$-th cell
is defined within the cell. Green’s function can be expressed in a cell-centered form:

\[ G(\mathbf{r} + \mathbf{R}^n, \mathbf{r}' + \mathbf{R}^{n'}, E) = \delta_{nn'} G_{LL'}^n(\mathbf{r}, \mathbf{r}', E) + \sum_{LL'} R_{LL'}^n(\mathbf{r}, E) G_{LL'}^{nn'}(E) R_{LL'}^{n'}(\mathbf{r}', E), \]  

(3)

where \( \mathbf{R}^n \) denotes the center of the \( n \)-th cell, \( L \equiv (l, m) \) the combined indices of the orbital angular momentum \( l \) and the magnetic quantum number \( m \), \( R_{LL'}^n(\mathbf{r}, E) \) the regular solution of the single-site Schrödinger equation for the local potential. The single-site Green’s function \( G_{LL'}^n \) is defined as

\[ G_{LL'}^n(\mathbf{r}, \mathbf{r}', E) = \sqrt{E} \sum_L R_{LL'}^n(\mathbf{r}_<, E) H_{ll}^L(\mathbf{r}_>, E), \]  

(4)

where \( \mathbf{r}_< (\mathbf{r}_>) \) means the smaller (larger) of \( \mathbf{r} \) and \( \mathbf{r}' \), \( H_{ll}^L(\mathbf{r}, E) \) irregular solution of the single-site Schrödinger equation for the local potential. In the original KKR formulation, the potential shape is assumed to be muffin-tin potentials. However, an FP extension is possible if all regions are Voronoi cells and the potentials are expanded into real spherical harmonics of the expansion coefficients \( \Omega_n \).

In the standard KKR, the free space, which has no potential scatterers, is chosen as a reference system. For the free space reference, an analytical expression of the expansion coefficient \( G_{LL'}^n(\mathbf{r}, \mathbf{r}', E) \) in the second term in (3). \( G_{LL'}^{nn'}(E) \) expresses the effect of the multiple scattering due to the existence of the t-matrices on the lattice points in the system and is connected to the expansion coefficient of an arbitrary reference system \( G_{LL'}^{nn'}(E) \) through the Dyson equation:

\[ G_{LL'}^{nn'}(E) = G_{LL'}^n(\mathbf{r}, \mathbf{r}', E) + \sum_{n'', L''} G_{LL'''}^n(\mathbf{r}) \sum_{L'''} [t_{LL'''}^{n'''}(E) - t_{LL'''}^{n'''}(E)] G_{LL'''}^{n'''}(E). \]  

(6)

Here, the reference system is defined as the system which has the same lattice structure as the crystal, but has a different kind of potential scatterers, described by a set of t-matrices \( t_{LL'}(E) \) on the lattice positions. Therefore, if \( G_{LL'}^{nn'}(E) \) is known, we can calculate \( G_{LL'}^{nn'}(E) \) by solving the Dyson equation (6).

2.3. Standard KKR Green’s function method

In the standard KKR, the free space, which has no potential scatterers, is chosen as a reference system. For the free space reference, an analytical expression of the expansion coefficient \( g_{LL'}^{nn'}(E) \) (called structure constant) is known:

\[ g_{LL'}^{nn'}(E) = -4\pi i \sqrt{E}(1 - \delta_{nn'}) \sum_{L''} \xi^{l+l''-l'''} C_{LL'L'''} h_{ll'}(\sqrt{E}|\mathbf{R}^n - \mathbf{R}^{n'}|) X_{L'''}(\mathbf{R}^n - \mathbf{R}^{n'}), \]  

(7)

where \( C_{LL'L'''} = \int d\mathbf{r} X_L(\mathbf{r}) X_{L'}(\mathbf{r}) X_{L'''}(\mathbf{r}) \) is the Gaunt number and \( h_l \) the spherical Hunkel function. The Dyson equation for the free space reference is now given by

\[ G_{LL'}^{nn'}(E) = g_{LL'}^{nn'}(E) + \sum_{n'', L''} g_{LL'''}^{nn'}(E) \sum_{L'''} t_{LL'''}^{n'''}(E) G_{LL'''}^{n'''}(E). \]  

(8)
For periodic crystals, by applying the 3D Fourier transform
\[
G_{\mu\mu'}^{LL'}(k, E) = \sum_{n} \exp(i \mathbf{k} \cdot (\mathbf{R}^n - \mathbf{R}^{n'})) G_{\mu\mu'}^{nn'}(E),
\]
the Dyson equation for each \(k\) point in the irreducible Brillouin zone (BZ) is obtained:
\[
G_{\mu\mu'}^{LL'}(k, E) = g_{\mu\mu'}^{LL'}(k, E) + \sum_{L'',\mu''} g_{\mu\mu'}^{LL''}(k, E) t_{L''\mu''}^{LL'}(E) G_{L''L'}^{\mu\mu'}(k, E).
\]

Here, \(\mu(= 1, \cdots, N)\) indicates an atom in the unit cell of periodic crystal \((N\) is the number of atoms in the unit cell). In the matrix notation of combined indices of \(\mu\) and \(L\),
\[
g(k, E) = \left\{ g_{\mu\mu'}^{LL'}(E) \right\},
\]
\[
G(k, E) = \left\{ g_{\mu\mu'}^{LL}(E) \right\},
\]
and \(t(E) = \{ \delta_{\mu\mu'} t_{LL'}^{LL'}(E) \}\), with matrix dimension \((l_{\text{max}} + 1)^2 \times N\), (10) can be rewritten as
\[
G(k, E) = t(E)^{-1} \left[ g(k, E) - t(E)^{-1} \right]^{-1} t(E)^{-1} - t(E)^{-1}.
\]

Then, the problem to solve Dyson equation (10) reduced to the problem of calculating the inverse of the matrix \(M(k, E) \equiv g(k, E) - t(E)^{-1}\). Matrix inversion is usually done by the \(LU\) factorization; then the number of steps needed increases as \(O(N^3)\). Therefore, if the system size becomes large, the computational time explodes rapidly. To overcome this difficulty, the concept of screened KKR described in the next section was proposed.

**Figure 1.** Repulsive potential reference system. \(G_{LL'}^{r,nr'}(E)\) decays exponentially in real space.

**Figure 2.** Real space cluster to obtain \(G_{LL'}^{r,nr'}(E)\) in solving Dyson equation (12) with the number of repulsive potential arrays \(N_{cl}\)

### 3. Screened KKR method

#### 3.1. The concept of screened KKR method

For the free space reference system, Green’s function has the analytical form \(G^{F}(\mathbf{r}, \mathbf{r}', E) \equiv g(\mathbf{r}, \mathbf{r}', E) = -\exp(i \sqrt{E} |\mathbf{r} - \mathbf{r}'|)/4\pi |\mathbf{r} - \mathbf{r}'|\). From this expression, we can see that \(g(\mathbf{r}, \mathbf{r}', E)\) decays exponentially in real space when \(E\) is negative. In other words, \(g(\mathbf{r}, \mathbf{r}', E)\) decays exponentially in real space if energy region is lower than the continuous eigenstates region of free space.

As suggested by the above fact, in screened KKR, a reference system with constant repulsive potential arrays of muffin-tin radius \(R_{\text{MT}}\) for each lattice position (figure 1) is adopted [3]. For these repulsive potential systems, continuous eigenstates are pushed up to a energy region much higher than \(E_F\) if the height of repulsive potential is sufficiently high. Then the Green’s function of the reference system decays exponentially in real space over the energy region used in evaluating (2). In this case, it is justified to neglect electron propagation longer than
some threshold distance without sacrificing the accuracy. Correspondingly, the matrix pattern appearing in the Dyson equation becomes sparse. By combining this feature with an efficient sparse matrix solver, it is possible to reduce the computational time drastically while keeping the accuracy.

Structure constants $G_{LL'}^{r,n'\prime}(E)$ of the reference system with repulsive potentials are easily calculated by solving a Dyson equation

$$G_{LL'}^{r,n'\prime}(E) = g_{LL'}^{n,n'}(E) + \sum_{n',L'} g_{LL'}^{n,n'}(E) \sum_{L''} t_{L'L''}(E) G_{L''LL'}^{r,n'\prime}(E)$$

in real space within small cluster (with the number of repulsive potential $N_{cl}$ within the cluster) as shown in figure 2, due to exponential decay of $G_{LL'}^{r,n'\prime}(E)$. After obtaining $G_{LL'}^{r,n'\prime}(E)$, we can calculate the crystal Green’s function through (6). In short, the reference system with repulsive potentials is inserted as an intermediate step, between initial free space and final crystal system in the screened KKR.

3.2. Screened KKR method for multilayered systems

If the screened KKR is applied to a multilayered system which is extended to the layer growth direction (figure 3), assuming 2D periodicity for the rest two directions, the Dyson equation can be solved for each $k_{||}$ in the 2D BZ, and the matrix pattern becomes just block tridiagonal [4, 10]. For a block tridiagonal matrix, the site-diagonal elements of its inverse, which are necessary for the construction of charge density in Kohn-Sham iteration, can be calculated in $O(N)$ ($N$ is the number of layer) using the LU factorization. This technique have been widely and successfully applied to layered systems since the screened KKR was first proposed.

3.3. Screened KKR method for super-cell systems

If we consider systems which are extended to x, y, and z directions (figure 4), the sparse matrix pattern becomes general block sparse form, differently from the case of multilayered systems. For a general block sparse matrix, there is no known $O(N)$ direct algorithm to calculate its inverse such as the LU factorization for block tridiagonal matrices. If LU factorization is applied, so called fill-in elements appear which require much larger memory storage than that of the original sparse matrix. Now, the problem is, for given matrix $A$, a calculation of matrix $A^{-1}$ which satisfies $AA^{-1} = I$ ($I$ is the unit matrix). And for the $i$-th column vector $x_i$ of $A^{-1}$, it satisfies $Ax_i = I_i$ ($I_i$ : the $i$-th column of $I$). It means that the problem can be considered as a simultaneous linear equation for each $x_i$ sharing a sparse coefficient-matrix $A$.

Then, an iterative method for simultaneous linear equations is a good candidate as an
efficient sparse matrix solver. For sparse coefficient-matrices of large dimension, nonstationary iterative methods categorized as the Krylov subspace methods have been widely used. In the Krylov subspace method, to solve an equation $Ax = b$, the true solution which minimizes the norm of the residual vector $||r|| ≡ ||x - b||$ is searched in the Krylov subspace defined as $K_n(c, A) = \text{span}(c, Ac, A^2c, \ldots, A^{n-1}c)$ by iterations. Here $c$ is an arbitrary vector. Starting from the initial guess $x_0$ and, normally, using $r_0 ≡ x_0 - b$ as $c$, an approximate solution $x_n \in x_0 + K_n(r_0, A)$ is updated in the $n$-th iteration. For each iteration, only (sparse matrix)-by-(vector) manipulations are required and no additional memory larger than $A$ is needed, in contrast to the case of a direct method. If the iteration converges rapidly, the method has an advantage over direct method from the view point of both the memory usage and the computational time. Since non-Hermitian general sparse matrices appear in the present cases, the transpose-free quasi-minimal residual (TFQMR) algorithm [11] is suitable as an iterative method [5, 12]. Further, as long as iterative methods are used, the calculation for each column vector is independent on the other columns. Therefore, parallelization with respect to the number of atoms is easily realized.

Usually, iterative methods are used in combination with a preconditioning of the coefficient matrix. For a linear equation $Ax = b$, the preconditioning matrix is defined as

$$A'(M_2x) = M_1^{-1}b,$$

where

$$A' = M_1^{-1}AM_2^{-1}.$$  \hspace{1cm} (14)

$M_1$ is a left preconditioning matrix and $M_2$ a right preconditioning matrix. As a general characteristics of the Krylov subspace method, the iteration convergence is fast when a distribution of eigenvalues of $A$ is dense near a non-zero point. Therefore, the acceleration in the convergence is expected if $A'$ approaches to $I$ through preconditioning. The efficiency of the preconditioning is shown by test calculations in the next section.

4. Test calculations and preconditioning

Following previous studies [12, 5, 6, 7], we have developed a code for first-principles electronic structure calculation of large super-cell systems using the screened KKR with the TFQMR algorithm. The modification of TFQMR routine available from QMRPACK in Netlib [13] is implemented, where the sparse matrix vector manipulation for each iteration is executed for $(l_{\text{max}} + 1)^2$ column vectors at the same time, in contrast to the original routine where each column is treated separately. This elaboration is implemented so as to take an advantage of cache access as mentioned in [6, 7]. MPI parallelization with respect to atoms in the super-cell is implemented. To check the performance and validity of the code, intensive test calculations were made. As a system for the test calculations, we took a single Ni impurity in a large Cu super-cell of the face-centered-cubic (fcc) structure. The primitive cubic unit cell consists of four atoms forming a fcc lattice with the lattice constant $a = 6.83$ a.u. is considered. Repeating this unit cell along three directions eight times, we construct super-cell containing $2048(= 4 \times 8 \times 8 \times 8)$ Cu atoms of a g structure. Then, one of 2048 Cu atom is replaced by a single Ni atom (figure 5).

The local electronic structure of a Ni impurity site in the super-cell is calculated using a single core of Intel Core2 Quad Z6700 2.66GHz. As a minimal setting, $l_{\text{max}} = 2$ and only nearest neighbor propagations of screened structure constants are considered ($N_{\text{cl}} = 13$), the Dyson equation is solved only at $\Gamma$-point in the BZ of the super-cell to construct the charge density. The convergence of iterative method is strongly dependent on the electronic structure at the sampling points used for the integration on the complex energy plane. Especially at a
Figure 5. Ni1Cu2047 super-cell system for test calculation. One atom in Cu super-cell is substituted for Ni impurity.

Figure 6. Sampling points on complex energy plane. Numbers with blue character in this figure distinguish energy sampling points.

sampling point near the real energy axis with continuous eigenstates, the convergence is worse. It means that calculations at the energy sampling points near $E_F$ could be a bottleneck in the case of metallic systems. Therefore, it is desirable to use energy sampling points as apart from the real axis as possible. For this, the finite-temperature formalism obtained by introducing the Fermi–Dirac distribution function $f(E, E_F, T) = (\exp(1 + (E - E_F)/k_B T))^{-1}$ in (2), is useful [14]. In the finite-temperature formalism, the nearest sampling point from $E_F$ is the first ”Matsubara frequency” with a vertical distance $\pi k_B T$ from the real axis. In the test calculations, 27 sampling points, including five Matsubara frequencies with $T = 800$ K, are used in complex energy integration (figure 6). All the test calculations were started with the initial guess of $x_0 = (0,\ldots,0)^T$ for Ni impurity. The TFQMR iterations were repeated until relative residual norm approaches to $||r||/||r_0|| = 10^{-6}$.

4.1. TFQMR without preconditioning
The number of the iteration and CPU time needed in one self-consistent field (SCF) calculation for each energy sampling point is shown in figure 7 with red line and points. As can been seen from this figure, calculations at a few Matsubara energies near $E_F$ are the bottlenecks.

4.2. TFQMR with the block Jacobi (diagonal scaling) preconditioning
The block Jacobi (BJCB) (or called as diagonal scaling) preconditioning, which uses diagonal blocks of sparse matrix as left preconditioning matrix $M_1$ ($M_2 = I$) such that the diagonal blocks of $A'$ become unit matrices, is one of the simplest preconditionings. Results using BJCB left preconditioning are shown in figure 7 with green line and points. Even a simple preconditioning such as BJCB improves the convergence of TFQMR considerably. However, the bottlenecks near $E_F$ still exist.

4.3. TFQMR with the block-circulant matrix preconditioning
An efficient algorithm, named block-circulant matrix (BCM) preconditioning, suitable for super-cell systems, was proposed in [15]. Generally, a circulant matrix $C \in \mathbb{C}^{N \times N}$

$$C = \begin{pmatrix} c_0 & c_1 & \cdots & c_{N-1} \\ c_{N-1} & c_0 & \cdots & \vdots \\ \vdots & \vdots & \ddots & \vdots \\ c_1 & c_2 & \cdots & c_0 \end{pmatrix}$$

(15)
Figure 7. The numbers and CPU time of iteration for each sampling point in one SCF with/without using preconditioning. Numbers on the horizontal axis distinguish energy sampling points (see figure 6).

can be diagonalized by a Fourier matrix $F_N$ defined as $(F_N)_{j,k} = \exp(2\pi i \cdot jk/N)$. Once the matrix is diagonalized as $C = F_N \text{diag}(\lambda_1, \ldots, \lambda_N) F_N^{-1}$, where $\lambda_i$ $(i = 1, \ldots, N)$ are the eigenvalues of $C$, the inverse operation is easily done as $C^{-1} = F_N \text{diag}(\lambda_1^{-1}, \ldots, \lambda_N^{-1}) F_N^{-1}$. Also, a $d$-level circulant matrix ($d$-level means that the circulant matrix pattern has $d$-fold multiperiod, and hence $C$ appearing in (15) is a 1-level circulant matrix) can be diagonalized by a $d$-dimensional Fourier-transform. For the super-cell system consisting of a repeated unit cell structure without substitutional atoms, the sparse matrix pattern becomes just a 3-level block-circulant. Then, the Dyson equation for the super-cell of a pure system keeping the ideal structure of the unit cell without any substitutional impurities can be solved by using the fast-Fourier-transform (FFT) if the repetition of the primitive unit cell along x, y, and z directions in the unit super-cell are powers of two. If substitutional impurities or lattice distortions appear, the periodicity will be lost and the matrix pattern is no longer circulant. However, even in these cases, BCM can be constructed through the averaging over the corresponding blocks (figure 8). Once BCM is constructed, diagonalization is easy, and multiplying the inverse of BCM to an arbitrary column vector can be done within $\sim 0.1$ sec in the present calculation of setting. Therefore, BCM can be used as right preconditioning matrix $M_2$ ($M_1 = I$). Using $M_2 \neq I$ requires a $M_2^{-1} \times$ (vector) operation for each iteration, but from the view point of the parallelization and memory usage this procedure is favorable with respect to the case of $M_1 \neq I$ if the preconditioning matrix is not simple like BJCB. In contrast to the left preconditioning which requires memory resource to store $A'$ itself, as long as the right preconditioning is used, it is sufficient to store only the result of $M_2^{-1} \times$ (vector) operations.

Results using BCM preconditioning are shown in figure 7 with blue line and points. As can be seen from this figure, linear equations are already virtually solved in the first step of the iteration for simple systems where a few impurities exist in a large super-cell such as the case of this test calculation. The needed number of iterations and CPU time are drastically reduced over all the sampling points. Even at the first Matsubara frequency, the TFQMR iteration converges after a few iterations, and the dependence of numerical effort on the position of sampling points is entirely smoothened. Now, the CPU time per single SCF cycle is reduced to $\sim 13$ sec, which is surprisingly fast for an all-electron level calculation.
Figure 8. Construction of BCM by averaging over corresponding blocks. If atoms in pure super-cell are substituted for impurities, diagonal blocks are substituted for that of impurities (shown as blue circle in the figure) due to the difference of t-matrix. Averaging over diagonal blocks (shown as purple circle), BCM can be constructed again.

Figure 9. Calculation time per one SCF cycle as a function of the number of MPI processes.

4.4. Parallel efficiency
The parallel efficiency of our code is explained in this subsection. Figure 9 shows the calculation time per one SCF cycle as a function of the number of MPI processes. The calculated super-cell is the same as in former test calculations. In contrast to the former test calculations, the electronic structure of an impurity atom as well as other atoms is calculated using many MPI processes. The calculation time is estimated on the calculation server which consists of Intel Xeon E5-2690 2.90GHz nodes. Note that the maximum number of MPI processes is limited to 256 because of a limitation of numerical resources which we can use (2048 MPI processes are required to determine the full electronic structure of super-cell with 2048 atoms). From this figure, one can confirm that the calculation time increases with the number of MPI processes. Ideally, the dependence of the calculation time with respect to the number of MPI process should be constant. This deviation from the ideal situation is due to the load of MPI communications which are required to construct sparse coefficient matrices from each piece of information of each MPI process. But the calculation time is still tens of seconds with 256 MPI processes,
which makes us expect that the calculation time per one SCF cycle for the full determination of electronic structure of the whole super-cell is several tens of seconds with thousands of MPI processes.

5. Summary
We have developed a code for electronic structure calculations of large super-cell systems. It uses the screened KKR and TFQMR algorithms combined with FP-KKR and is MPI parallelized with respect to the number of atoms. The efficiency of TFQMR combined with the BJCB and BCM preconditioning for super-cell systems is confirmed by several test calculations. The calculations shown here is rather limited for a single atom in a unit super-cell with a single MPI process. Due to the perfect MPI parallelization with respect to the number of atoms, the full determination of the electronic structure of the whole super-cell containing thousands of atoms is expected to be possible in an all-electron level within a reasonable CPU time comparable with that of the calculation that uses a single MPI process as is shown in the test calculations. The BCM preconditioning is efficient for super-cells with substitutional impurities, if ideal geometry of the unit structure or only with a slight distortion from ideal position is assumed. The TFQMR algorithm with a suitable preconditioning seems to surprisingly broaden the range of the application of the KKR Green’s function method.

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