Convergence rate for numerical computation of the lattice Green's function

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Flexible boundary-condition methods couple an isolated defect to bulk through the bulk lattice Green's function. Direct computation of the lattice Green's function requires projecting out the singular subspace of uniform displacements and forces for the infinite lattice. We calculate the convergence rates for elastically isotropic and anisotropic cases for three different techniques: relative displacement, elastic Green's function correction, and discontinuity correction. The discontinuity correction has the most rapid convergence for the general case.

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Atomic-scale simulation of isolated defects with a computationally tractable number of atoms requires careful choice of boundary conditions. Periodic or fixed boundary conditions introduce fictitious forces when relaxing the geometry of defects; reducing the error requires increasing the number of atoms. Flexible boundary-condition methods avoid these errors by instead using harmonic lattice response for atoms away from the defect. In particular, the bulk lattice Green's function (LGF) gives the short- and long-range displacements in response to a point or line force. Sinclair *et al.* [1] introduced flexible boundary conditions for studying defects such as cracks [2,3], dislocations [4–7], vacancies and free surfaces [8] with classical potentials, and isolated screw or edge dislocations with density-functional theory (DFT) [9–12]. Evaluation of the LGF in real space involves the inverse Fourier transform of a function with a singularity at the Γ point ($k=0$), which requires algorithmic approaches to evaluate numerically. Relative displacement method [2,13], elastic Green's function (EGF) correction [14], and discontinuity correction [15] are three techniques to numerically evaluating the bulk LGF; we compare these and find that the discontinuity correction has the fastest convergence rate. We verify our predicted convergence rates with a simple model and density-functional theory results for Al.

The lattice Green's function $\underset{\sim}{G}^L(\vec{R}-\vec{R}')$ and the force-constant matrix $\underset{\sim}{D}(\vec{R}-\vec{R}')$ relate the internal displacements $\vec{u}(\vec{R})$ and forces $\vec{f}(\vec{R}')$ of atoms $\vec{R}$ and $\vec{R}'$ of the lattice through $\vec{u}(\vec{R})=\Sigma_{\vec{R}'}\underset{\sim}{G}^L(\vec{R}-\vec{R}')\vec{f}(\vec{R}')$ and $\vec{f}(\vec{R})=-\Sigma_{\vec{R}'}\underset{\sim}{D}(\vec{R}-\vec{R}')\vec{u}(\vec{R}')$. Translational invariance of an infinite lattice makes $\underset{\sim}{G}^L$ a function of the relative positions of two atoms. Substituting one of the above relations into the other gives $\Sigma_{\vec{R}'}\underset{\sim}{G}^L(\vec{R}-\vec{R}')\underset{\sim}{D}(\vec{R}')=-\mathbf{1}\delta(\vec{R})$, where $\delta(\vec{R})$ is the Kronecker delta function. A constant shift in the atom positions does not produce internal forces, giving the sum rule $\Sigma_{\vec{R}}\underset{\sim}{D}(\vec{R})=0$ and making $\underset{\sim}{G}^L(\vec{R})$ the pseudoinverse of $\underset{\sim}{D}(\vec{R})$ in the subspace without uniform displacements or forces. Fourier transform of the lattice functions are defined as $\underset{\sim}{G}^L(\vec{k})=\Sigma_{\vec{R}}e^{i\vec{k}\cdot\vec{R}}\underset{\sim}{G}^L(\vec{R})$, $\underset{\sim}{G}^L(\vec{R})=\int_{BZ}\frac{d^3k}{(2\pi)^3}e^{-i\vec{k}\cdot\vec{R}}\underset{\sim}{G}^L(\vec{k})$ for $\vec{k}$ in the Brillouin zone (BZ). The integral can be approximated by a discrete sum of N_k points as $\underset{\sim}{G}^L(\vec{R})=\frac{1}{N_k}\Sigma_{\vec{k}}e^{-i\vec{k}\cdot\vec{R}}\underset{\sim}{G}^L(\vec{k})$. In reciprocal space, the matrix inverse relation and the sum rule are $\underset{\sim}{G}^L(\vec{k})\underset{\sim}{D}(\vec{k})=1$ and $\underset{\sim}{D}(\vec{0})=0$, respectively. For a single atom crystal basis, $\underset{\sim}{D}(\vec{k})$ expands for small $\vec{k}$ as $\underset{\sim}{D}(\vec{k})=\Sigma_{\vec{R}}\underset{\sim}{D}(\vec{R})[1-\frac{(\vec{k}\cdot\vec{R})^2}{2!}+\cdots]\simeq-\frac{1}{2}\Sigma_{\vec{R}}(\vec{k}\cdot\vec{R})^2\underset{\sim}{D}(\vec{R})$, due to the inversion symmetry of $\underset{\sim}{D}(\vec{R})$. At the Γ point, $\underset{\sim}{D}(\vec{k})$ is of the order k^2 so $\underset{\sim}{G}^L(\vec{k})$ has a second-order pole. Due to this singularity, the three-dimensional (3D) inverse Fourier transform of $\underset{\sim}{G}^L(\vec{k})$ converges very slowly while the two-dimensional (2D) version does not converge at all. Different behaviors of 3D and 2D integrals lie in the integration factors which are proportional to k^2 and k, respectively. The 3D integration factor cancels out the second-order pole leaving a discontinuity in the Γ point that causes a poor convergence. The 2D integrand is still singular which results in a nonconvergent integral.

Figure 1 shows the relative displacement method, elastic Green's function correction, and discontinuity correction which are used to avoid the singularity in LGF. With regards to relative displacement [2,13,16,17], rigid body translations leave the potential energy of the lattice unchanged. Choosing an arbitrary atom as an undisplaced origin for the relative displacements of atoms requires calculation of $\underset{\sim}{G}^L(\vec{R})-\underset{\sim}{G}^L(\vec{0})=\int\frac{d^3\vec{k}}{(2\pi)^3}\underset{\sim}{G}^L(\vec{k})e^{i\vec{k}\cdot\vec{R}}-\int\frac{d^3\vec{k}}{(2\pi)^3}\underset{\sim}{G}^L(\vec{k})e^{i\vec{k}\cdot\vec{0}}$, which reduces to

$$\underset{\sim}{G}^L(\vec{R})-\underset{\sim}{G}^L(\vec{0})=\int\underset{\sim}{G}^L(\vec{k})[\cos(\vec{k}\cdot\vec{R})-1]\frac{d^3\vec{k}}{(2\pi)^3},\qquad(1)$$

due to the sum rule and inversion symmetry. For small k, $\cos(\vec{k}\cdot\vec{R})-1$ is of the order k^2 which cancels out the second-order pole in $\underset{\sim}{G}^L(\vec{k})$ leaving a $\vec{k}$-direction dependent discontinuity at the Γ point. The discretized version of Eq. (1) is $\frac{1}{N_k}\Sigma_{\vec{k}}[\cos(\vec{k}\cdot\vec{R})-1]\underset{\sim}{G}^L(\vec{k})$. With regards to elastic Green's function correction, following the procedure and notations of [15], $\underset{\sim}{G}^L(\vec{k})$ for small k expands as $\underset{\sim}{G}^L(\vec{k})=[\underset{\sim}{D}(\vec{k})]^{-1}=k^{-2}[\tilde{\Lambda}^{(2)}(\hat{k})]^{-1}+[\tilde{\Lambda}^{(2)}(\hat{k})]^{-1}\tilde{\Lambda}^{(4)}(\hat{k})[\tilde{\Lambda}^{(2)}(\hat{k})]^{-1}+O(k^2)=\underset{\sim}{G}^E(\vec{k})+\underset{\sim}{G}^{\mathrm{dc}}(\vec{k})+O(k^2)$, where $k^2\tilde{\Lambda}^{(2)}(\hat{k})$ and $k^4\tilde{\Lambda}^{(4)}(\hat{k})$ are the second and fourth order terms in a small k expansion of $\underset{\sim}{D}(\vec{k})$. The Fourier transform of the elastic Green's function $\underset{\sim}{G}^E(\vec{k})$ is the second-order pole and $\underset{\sim}{G}^{\mathrm{dc}}(\vec{k})$ is a $\vec{k}$-direction dependent discontinuity [15]. The elastic part $\underset{\sim}{G}^E(\vec{k})$ should be inverse Fourier transformed analytically and the remaining part which no longer has a pole (it still has a discontinuity) can be

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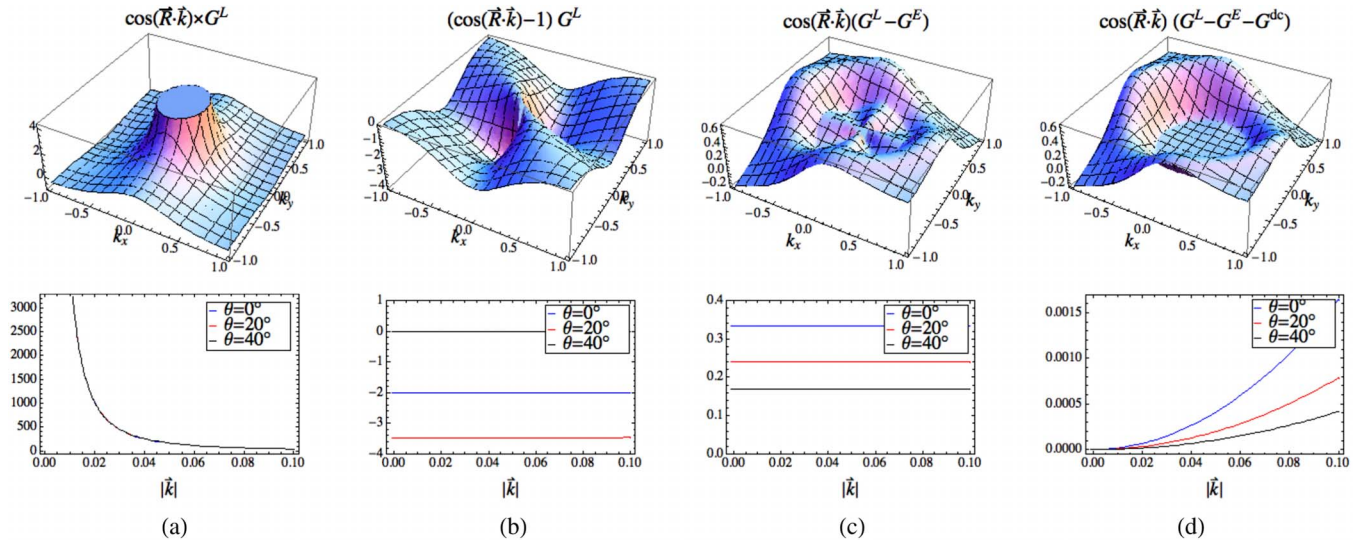

FIG. 1. (Color online) Integrand of the inverse Fourier transform for (a) LGF, (b) relative displacement method, (c) EGF correction, and (d) discontinuity correction at $\vec{R}=(1,1)$. $\underset{\sim}{G}^L(\vec{k})$ has a second-order pole at the Γ point. The relative displacement method avoids the pole by considering only the displacements relative to a fixed point. The EGF correction removes the second-order pole by subtracting a cutoff elastic Green's function. Removal of the pole creates a discontinuity independent of $|\vec{k}|$ at the Γ point. The discontinuity correction removes the discontinuity created by EGF correction. The remaining part of the integrand is smooth in the entire Brillouin zone. The bottom row shows the variation in the integrand as a function of $|\vec{k}|$ when the origin is approached from different angles $\theta=\tan^{-1}(k_y/k_x)$. The discontinuity created at the Γ point by the relative displacement method and EGF correction is independent of $|\vec{k}|$ but depends on the direction of approaching the origin. N.B.: the vertical scale changes from LGF to relative displacement and EGF correction to discontinuity correction.

inverse transformed numerically by $\frac{1}{N_k}\Sigma_k \cos(\vec{k}\cdot\vec{R})[\underset{\sim}{G}^L(\vec{k}) - \underset{\sim}{G}^E(\vec{k})f_{\text{cut}}(\vec{k})]$, where f_{cut} is a cutoff function that smoothly vanishes on the Brillouin-zone edges. Removal of the second-order pole by subtraction of a cutoff version of elastic Green's function is used in the semicontinuum method of Tewary and Bullough [14]. With regards to discontinuity correction [15], to further improve convergence, the discontinuity correction treats the $\underset{\sim}{G}^{\text{dc}}(\vec{k})$ part analytically. The remaining portion of $\underset{\sim}{G}^L(\vec{k})$ given by $\frac{1}{N_k}\Sigma_k \cos(\vec{k}\cdot\vec{R})\{\underset{\sim}{G}^L(\vec{k}) - [\underset{\sim}{G}^E(\vec{k}) + \underset{\sim}{G}^{\text{dc}}(\vec{k})]f_{\text{cut}}(\vec{k})\}$ is smooth and can be integrated numerically more efficiently.

We expect the convergence rate of the discontinuity correction method to be consistent with the results for integration of smooth periodic functions while the convergence of relative displacement and elastic Green's function correction methods should be dominated by the discontinuity. For N_{div} partitions in each direction, midpoint rule gives a N_{div}^{-4} scale for convergence rate of such integrals in all dimensions [18,19]. The number of k points N_k is N_{div}^d for dimensionality $d=1,2,3$; therefore, the convergence rate of the midpoint rule scales as $N_k^{-4/d}$. In the EGF correction and relative displacement method, the integrand is smooth everywhere except the Γ point so we expect the error to be dominated by the area/volume around Γ point and therefore be of the order of N_k^{-1} or N_{div}^{-d}.

We check the predictions of convergence for the three methods using (1) a simple-cubic nearest-neighbor model and (2) fcc Al. First, as a simplified case we consider a square (cubic in 3D) elastically isotropic lattice with nearest-neighbor interactions and lattice constant $a_0=\pi$. The nonzero component of the LGF matrix is $G^L(k_x,k_y)=[\sin^2(\pi k_x/2) + \sin^2(\pi k_y/2)]^{-1}$. The second-order pole is given by the elastic Green's function $G^E(k_x,k_y)=4/(\pi|\vec{k}|^2)$ which is multiplied by a cutoff function to vanish smoothly at the BZ edges. The discontinuity correction is given by $G^{\text{dc}}(k_x,k_y)=(\hat{k}_x^4 + \hat{k}_y^4)/3|\vec{k}|^2$ which is also multiplied by the cutoff function. The three-dimensional G^L, G^E, and G^{dc} are obtained by replacing the 2D vector $\vec{k}=(k_x,k_y)$ with $\vec{k}=(k_x,k_y,k_z)$. For the elastically anisotropic Al, we obtain the force-constant matrix $\underset{\sim}{D}(\vec{R})$ from density functional theory (DFT) [ultrasoft pseudopotentials with generalized gradient approximation (GGA) [12]]. Numerical integration over the BZ is done with a uniform mesh evaluating the integrand at the midpoints. Even and odd values of N_{div} give meshes that include or avoid the Γ point—what we call Γ and non-Γ centered meshes, respectively. When applying the relative displacement method and EGF correction, the value of the integrand—which is discontinuous at Γ—is assigned zero at the Γ point. We calculate the numerical error as a function of N_k and N_{div} to compare the efficiency of the three methods.

Figure 2 shows the convergence rates of relative displacement method, EGF correction, and discontinuity correction in the square lattice case. The discontinuity correction and EGF correction scale as N_k^{-2} and N_k^{-1}, respectively, as expected. The value of $\vec{R}$ does not affect the power-law scalings of the convergence but the prefactors are changed in the relative displacement method, and are of the same order in EGF and discontinuity corrections. While the N_k^{-1} convergence for relative displacement method obtained by a Γ centered mesh is in accordance with the analytical predictions, use of a non-Γ centered mesh produces a convergence faster than expected for this method. This is an artifact of the isotropy of the EGF.

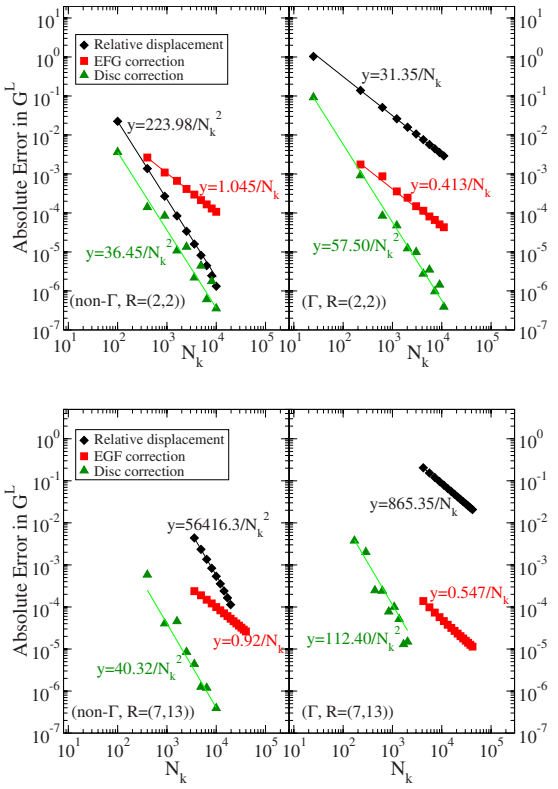

FIG. 2. (Color online) Convergence rate with number of k points of the relative displacement method, EGF correction, and discontinuity correction in a 2D square lattice. We expect N_k^{-2} convergence for discontinuity correction, and poorer N_k^{-1} convergence for EGF correction and relative displacement method. Using a non-Γ centered mesh (left) causes an unusually fast convergence for the relative displacement method in elastically isotropic materials. The exponents in the power-law scalings are not affected by the value of $\vec{R}$ while prefactors are changed in relative displacement method and are of the same order in EGF and discontinuity corrections.

The integrand in the relative displacement method is $I(\vec{k})=[\cos(\vec{k}\cdot\vec{R})-1]\underset{\sim}{G}^L(\vec{k})$. Near the Γ point, $\underset{\sim}{G}^L(\vec{k})$ matches $\underset{\sim}{G}^E(\vec{k})$ and the leading term in the integrand is $I^{(\Gamma)}(\vec{k})=-k^2R^2(\hat{k}\cdot\hat{R})^2\underset{\sim}{G}^E(\hat{k})/2k^2$. For an isotropic EGF, $\underset{\sim}{G}^E(\hat{k})$ is constant so $I_{\text{iso}}^{(\Gamma)}(\vec{k})=-\frac{1}{2}\underset{\sim}{G}^E R^2\cos^2(\theta_{\hat{k},\hat{R}})$, where $\theta_{\hat{k},\hat{R}}$ is the angle between vectors $\hat{k}$ and $\hat{R}$. The value of the integral over a square $k_0\times k_0$ region around $k=0$, for small k is

$$\int_{k_0^2} I_{\text{iso}}^{(\Gamma)}(\vec{k})d^2k=-\int\int_{k_0^2}\frac{\underset{\sim}{G}^E R^2\cos^2\theta}{2}dk_xdk_y=-\frac{k_0^2}{4}\underset{\sim}{G}^E R^2. \tag{2}$$

The midpoint rule integration of the same region with a non-Γ centered mesh uses the k points $\vec{k}_1=(k_0/2,k_0/2)$, $\vec{k}_2=(-k_0/2,k_0/2)$, $\vec{k}_3=(-k_0/2,-k_0/2)$, and $\vec{k}_4=(k_0/2,-k_0/2)$ each contributing area $k_0^2/4$. The angle between each $\vec{k}_i$ and $\hat{R}$ are θ_1, $\theta_2=\theta_1+\pi/2$, $\theta_3=\theta_1+\pi$, and $\theta_4=\theta_1+3\pi/2$. Therefore, the numerical approximation for the integral around the Γ point is

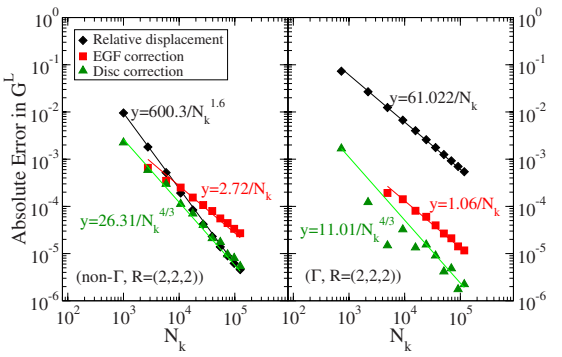

FIG. 3. (Color online) Convergence rate with number of k points of the relative displacement method, elastic GF correction, and discontinuity correction in a 3D cubic lattice. The error for discontinuity correction method scales as $N_k^{-4/d}$ with dimension d equal to three. Note that using a non-Γ centered mesh creates a faster convergence for the relative displacement method as observed in the 2D case.

$$\bar{I}_{\text{iso}}=\frac{k_0^2}{4}\sum_{i=1}^{4}I_{\text{iso}}^{(\Gamma)}(\vec{k}_i)=-\frac{k_0^2}{4}\underset{\sim}{G}^E R^2, \tag{3}$$

which is equal to the exact value of the integral around Γ point given by Eq. (2). To avoid the effect of the discontinuity at the origin using a Γ centered mesh, the Γ-point contribution to the integral is considered zero while its actual value is given by Eq. (2). This is the source of the dominant error in relative displacement method on a Γ centered mesh, and accounts for the R^2 dependence of the error. This dependence is verified by comparing the ratio of prefactors of the relative displacement convergence laws for different R values and the corresponding R^2 in Fig. 2 which are both approximately 27. On the other hand, the non-Γ centered mesh automatically gives the exact value of the integral around the origin based on Eq. (3) and thus produces a faster convergence limited only by the convergence of smooth periodic functions. For elastically anisotropic materials, $\underset{\sim}{G}^E(\hat{k})$ depends on $\hat{k}$, and the numerical integration around Γ will not equal the analytic value—eliminating the special convergence for non-Γ meshes.

Figure 3 shows that the 3D results follow the same trend as the 2D ones in accordance with the expected values. Both Γ centered and non-Γ centered meshes give $N_k^{-4/d}(d=3)$, and N_k^{-1} scale for the convergence rate of discontinuity and EGF corrections, respectively. Similar to the trend observed in 2D case the Γ centered mesh produces the expected N_k^{-1} scale for the convergence of relative displacement method and the non-Γ centered mesh produces faster convergence due to the isotropy of the elastic Green's function.

Figure 4 shows that convergence trends are unchanged for anisotropic long-range interactions in fcc Al except for relative displacement method. The lattice is periodic in the threading direction [110], appropriate for screw dislocations in fcc. With a non-Γ centered mesh, the anisotropy of the elastic Green's function eliminates the fast convergence of the relative displacement method. The convergence trends of the three methods show that these trends are not specific to the simplifying assumptions of isotropy or short-range interactions and therefore can be trusted in realistic calculations.

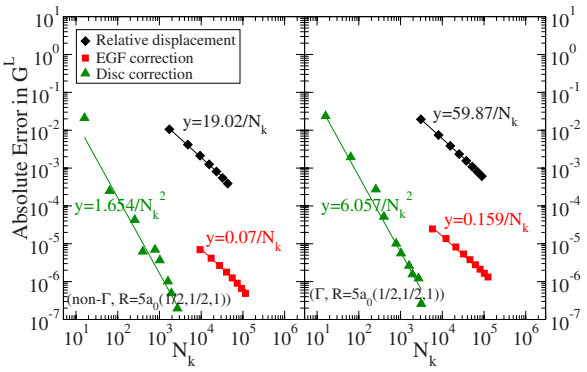

FIG. 4. (Color online) Convergence rate with number of k points of the relative displacement method, elastic Green's function correction, and discontinuity correction in computation of the G_{11} component of a 2D LGF in Al. The convergence of LGF calculations in a fcc lattice is the same as the one observed in the simplified problem in agreement with the expected values. Note that use of the non-Γ centered mesh does not cause a fast convergence for relative displacement method due to the anisotropy of the elastic Green's function.

Table I summarizes the convergence results for the three methods. The expected convergence rate for a numerical integral of a smooth periodic function evaluated by midpoint rule is N_{div}^{-4}. When expressed in terms of the number of k points N_k, the convergence rate would be proportional to $N_k^{-4/d}$. Since the discontinuity correction leaves a smooth periodic part of the integrand, it follows the above convergence rate. The EGF correction and relative displacement method also converge with the scale of N_k^{-1} or N_{div}^{-d}. The convergence rates imply that a certain amount of error is achieved with less N_k by discontinuity correction method compared to EGF correction or relative displacement method which means that the discontinuity correction requires the least computational effort. Although the EGF correction and relative displacement method require comparable computational effort, the R^2 dependence of the error suggests that the relative displacement method takes even more k points than the EGF correction. Also note that there is a tradeoff between less computational effort and more complex algorithms. EGF and discontinuity corrections calculate the elastic Green's function and discontinuity correction parts of the LGF analytically while relative displacement method does not require additional analytic evaluations.

TABLE I. Effect of dimension on the convergence rate with number of k points and number of divisions for the relative displacement method, EGF correction, and discontinuity correction. N_{div} is proportional to $1/h$, the inverse grid spacing and $N_k = N_{\mathrm{div}}^d$.

	2D		3D	
Power law scaling of error	N_k	N_{div}	N_k	N_{div}
Discontinuity correction	−2	−4	−4/3	−4
EGF correction	−1	−2	−1	−3
Relative displacement	−1	−2	−1	−3

The relative displacement method, elastic Green's function correction, and discontinuity correction have all been used in different calculations. These computational methods improve the slow rate of convergence for 3D and eliminate numerically divergent terms for 2D calculations. We find the discontinuity correction to be the most efficient method—improving the convergence rate to quadratic convergence for 2D over linear convergence for the relative displacement and elastic Green's function correction. The discontinuity correction method is general, applicable to any monatomic lattice, and can be generalized to any crystal. Beyond the monatomic case, the leading terms of $\underset{\sim}{G}^L(\vec{k})$ expansion for small k include an extra term of the order i/k without inversion symmetry. This term should be subtracted from $\underset{\sim}{G}^L(\vec{k})$ in addition to the k^{-2} elastic contribution before applying the discontinuity correction. Finally, while the convergence trends match the analytical values, there is an unusual exception for lattices with isotropic elastic Green's function. This connects the elastic anisotropy of a material to the efficiency of the computational methods used in Green's function calculations.

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