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Carlsson-Gelatt-Ehrenreich technique and the Möbius inversion theorem

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The Möbius inversion theorem is first applied to different lattices so as to improve the Carlsson-Gelatt-Ehrenreich technique and give very fast convergence.

It is well known that pairwise potentials are widely used in calculations of mechanical and defect properties of metals. The inverse cohesive energy problem is to determine the pairwise potential from the *ab initio* calculated or experimentally measured cohesive energy.[1] Carlsson, Gelatt, and Ehrenreich (CGE) have derived an exact procedure to convert the cohesive energy of an isostructural one-component system as a function of volume to find the radial pair potential, with evaluation of the applicability of this pairwise potential.[1-3] Here, we present an improved method based on the Möbius inversion formula[4-6] with very fast convergence.

Assuming that for a given crystal structure the cohesive energy E of one-component system can be written as a sum over atomic sites of a radial pairwise potential

$$E(r_1)=\tfrac{1}{2}\sum_{R\neq 0}\Phi(|\mathbf{R}|)=\tfrac{1}{2}\sum_{p=1}^{\infty}W_p\Phi(S_pr_1)\,, \tag{1}$$

where r_1 is the nearest-neighbor distance and Φ is the pairwise potential. In the final expression of Eq. (1) the sum is over spherical shell, p, containing W_p atoms at a distance $r_p=s_pr_1$ from the atom at the origin, and s_p is the dimensionless ratio of the pth-neighbor distance to the nearest one r_1. Equation (1) can be written in terms of a linear operator L:

$$\begin{aligned}E(r_1)&=L\Phi(r_1)=\sum_p T_p\Phi(r_1)\\&=\left[T_1\left(1+\sum_{p=2}^{\infty}T_1^{-1}T_p\right)\right]\Phi(r_1)\,,\end{aligned} \tag{2}$$

where

$$T_pf(x)=(1/2)W_pf(s_px)\,. \tag{3}$$

Therefore,

$$\begin{aligned}\Phi(r)&=L^{-1}E(r)=\left(1+\sum_{p=2}^{\infty}T_1^{-1}T_p\right)^{-1}T_1^{-1}E(r)\\&=\left(1-\sum_{p=2}^{\infty}T_1^{-1}T_p+\sum_{p,q=2}^{\infty}T_1^{-1}T_pT_1^{-1}T_q-\cdots\right)\\&\quad\times T_1^{-1}E(r)\,.\end{aligned} \tag{4}$$

Substituting Eq. (3) in Eq. (4), the final result can be obtained,

$$\begin{aligned}\Phi(r)&=\frac{2}{W_1}E\frac{r}{s_1}-\sum_{p=2}^{\infty}\frac{2}{W_1}\frac{W_p}{2}\frac{2}{W_1}E\frac{s_pr}{s_1^2}\\&\quad+\sum_{p,q=2}^{\infty}\frac{2}{W_1}\frac{W_p}{2}\frac{2}{W_1}\frac{W_q}{2}\frac{2}{W_1}E\frac{s_ps_qr}{s_1^3}-\cdots\end{aligned} \tag{5}$$

for the pairwise potential in terms of the cohesive energy function. Notice that all the parameters s_p and W_p are determined by the crystal structure under consideration.

For the linear atomic chain, $W_p=2$ and $s_p=p$, then (1) and (5) become

$$E(r)=\sum_{n=1}^{\infty}\Phi(nr) \tag{6}$$

and

$$\begin{aligned}\Phi(r)&=E(r)-\sum_{p=2}^{\infty}E(pr)+\sum_{p,q=2}^{\infty}E(pqr)\\&\quad-\sum_{p,q,t=2}^{\infty}E(pqtr)+\cdots\,,\end{aligned} \tag{7}$$

respectively. In practice, calculating Eq. (7) would be a time-consuming process with slow convergence. It would be convenient to combine the same $E(nr)$ terms together from different summations in Eq. (7). In other words, it is possible to express Eq. (7) as

$$\Phi(r)=E(r)+\sum_{n=2}^{\infty}\mu(n)E(nr) \tag{8}$$

if the inversion coefficient $\mu(n)$ in Eq. (8) can be determined. This is possible from the Möbius inversion theorem which states[4,5] that if

$$F(x)=\sum_{n=1}^{\infty}f(nx) \tag{9}$$

then

$$f(x)=\sum_{n=1}^{\infty}\mu(n)f(nx) \tag{10}$$

and vice versa, provided that the related sums converge. Thus, the coefficient $\mu(n)$ in (8) is just identical to the Möbius function in (10) defined[7] as

$$\mu(n)=\begin{cases}1, & \text{when } n=1,\\(-1)^s, & \text{when } n \text{ is a product of } s \text{ distinct primes},\\0, & \text{when } n \text{ includes repeated factors.}\end{cases}$$

This indicates a lot of cancellations in Eq. (7), which is important to speed the calculation based on the Möbius theorem. Now let us use the theorem to improve the CGE technique.

From the above example of the linear atomic chain, the effect of the T_1 in the original CGE scheme only applies to the nearest neighbors, and in our modified CGE scheme, the effective operator T_1^* is expanded significantly as

$$T_1^* f(x)=\sum_{n=1}^{\infty} f(nx)\,, \tag{11}$$

which applies to all the atoms in the chain, and we know the existence of T_1^{*-1} based on the Möbius theorem.

Since both the operators L and T_1 in Eqs. (2) and (3) are structure dependent, in this paper we only present some examples for illustrating the new scheme. The dominate point is to determine the operator T_1^*, and give the explicit expression of the corresponding inverse operator T_1^{*-1}.

For a square lattice, the cohesive energy can be written as

$$E(r)=2\sum_{p,q=1}^{\infty}\Phi\{[p^2+(q-1)^2]^{1/2}r\}\,. \tag{12}$$

According to the CGE scheme, when $(p,q-1)\le 8$, there exist 41 different s_p^2 terms with a maximum value of 128.

According to our modified CGE scheme, Eq. (13) can be rewritten as

$$E(r)=2\sum_{n=1}^{\infty}[\Phi(nr)+\Phi(\sqrt{2}nr)]$$
$$+4\sum_{p,q=1}^{\infty}\Phi\{[p^2+(p+q)^2]^{1/2}r\}\,. \tag{13}$$

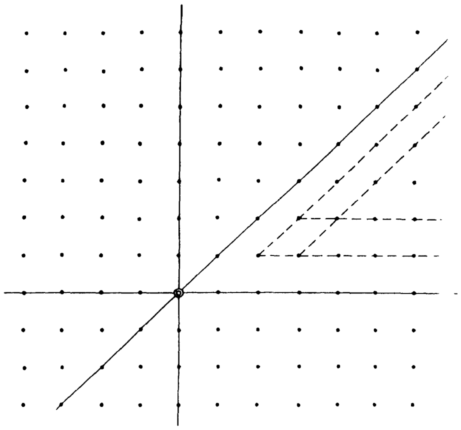

FIG. 1. A 2D square lattice that can be divided into eight equivalent sections.

where the terms $\Phi(nr)$ represents the coordinate axes and $\Phi(\sqrt{2}nr)$ corresponds to diagonals as shown in Fig. 1. Now let us define the operator T_1^* as

$$T_1^* f(r)=2\sum_{n=1}^{\infty}[f(nr)+f(\sqrt{2}nr)]\,, \tag{14}$$

then the corresponding inverse operator T^{*-1} can be expressed as

$$T_1^{*-1}f(r)=\tfrac{1}{2}\sum_{m,n=1}^{\infty}(-1)^{m-1}\mu(n)f(2^{(m-1)/2}nr)\,, \tag{15}$$

since

$$T_1^{*-1}T_1^* f(r)=\tfrac{1}{2}\sum_{m,n=1}^{\infty}(-1)^{m-1}\mu(n)T_1^* f(2^{(m-1)/2}nr)=\sum_{m,n,s=1}^{\infty}(-1)^{m-1}\mu(n)[f(2^{(m-1)/2}nsr)+f(2^{(m-1)/2}\sqrt{2}nsr)]$$
$$=\sum_{n=1}^{\infty}\mu(n)\left(\sum_{m=0}^{\infty}(-1)^m+\sum_{m=1}^{\infty}(-1)^{m-1}\right)f(2^{m/2}nsr)=\sum_{n,s=1}^{\infty}\mu(n)f(nsr)=\sum_{s=1}^{\infty}g(sr)=f(r) \tag{16}$$

where $g(r)=\sum_{n=1}^{\infty}\mu(n)f(nr)$ is used. From the second summation in Eq. (14), one can construct the s_p^2 with 63 different s_p^2; the maximum is 320. This indicates the faster convergence of the modified CGE scheme than the original one. After choosing T_1^*, Eqs. (3) and (4) are still available except that the atoms in the most close-packed direction need not be counted again. Therefore, we have

$$\Phi(x)=\left(1-\sum_{p=2}^{\infty}T_1^{*-1}T_p+\sum_{p,q=2}^{\infty}T_1^{*-1}T_pT_1^{*-1}T_q-\cdots\right)T_1^{*-1}E(x)=\tfrac{1}{2}\sum_{m,n=1}^{\infty}(-1)^{m-1}\mu(n)E(2^{(m-1)/2}nx)$$
$$-\tfrac{1}{4}\sum_{m_1,m_2,n_1,n_2=1}^{\infty}(-1)^{m_1+m_2-2}\mu(n_1)\mu(n_2)\sum_{p=2}^{\infty}T_pE(2^{(m_1+m_2-2)/2}n_1n_2x)+\cdots\,, \tag{17}$$

where, as in Eq. (14), the T_p is equal to

$$T_pE(x)=4E(s_px),\ \text{for } s_p^2\le 64\,.$$

In practice, these terms alone are enough for calculating with high accuracy and stability.

The hexagon lattice essentially is a complex lattice. As shown in Fig. 2(a), one can introduce a line to separate the lattice into the upper and lower parts. Let us reflect all the lattice points marked by circles in the upper half-space to the lower one; all the lattice points marked with open circles will go to the centers of hexagons in the lower half-space [see Fig. 2(b)]. After this so-called partial symmetry operation, each of the half-spaces has a single structure. The upper one has an elementary parallelogram with a length of $\sqrt{3x}$, and the lower one with the length x. Since the partial symmetry operation does not change the distance of any lattice point from the origin, it does not affect our calculation. Therefore, the cohesive energy can be rewritten as

$$E(x)=\tfrac{3}{2}\sum_{n=1}^{\infty}[\Phi(nr)+2\Phi(\sqrt{3}nr)+\Phi(3nr)]$$
$$+3\sum_{p,q=1}^{\infty}\{\Phi[(3p^2+q^2+3pq)^{1/2}r]$$
$$+\Phi[3(3p^2+q^2+3pq)^{1/2}r]\}\,. \tag{18}$$

In Eq. (19), the terms of $\Phi(nr)$ represent the most close-packed directions, and $\Phi(\sqrt{3}nr)$ and $\Phi(3nr)$ correspond to secondmost and thirdmost close-packed directions. According to our modified CGE scheme, the operator T_1^* can be chosen as

$$T_1^* f(x)=\tfrac{3}{2}\sum_{n=1}^{\infty}[f(nr)+2f(\sqrt{3}nr)+f(3nr)] \tag{19}$$

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and the corresponding inverse operator is

$$T_1^{*-1}f(x)=\tfrac{2}{3}\sum_{m,n,u=1}^{\infty}(-1)^{m+u}\mu(n)f(3^{(m+u)/2-1}nr)\,, \tag{20}$$

since

$$\begin{aligned}T_1^{*-1}T_1^* f(x)&=\tfrac{2}{3}\sum_{m,n,u=1}^{\infty}(-1)^{m+u}\mu(n)T_1^* f(3^{(m+u)/2-1}nr)\\&=\sum_{m,n,u,v=1}^{\infty}(-1)^{m+u}\mu(n)[f(3^{(m+u)/2-1}nvr)+2f(3^{(m+u)/2-1}\sqrt{3}nvr)+f(3^{(m+u)/2-1}3nvr)]\\&=\sum_{n,n,u,v=1}^{\infty}(-1)^{m+u}\mu(n)[g(3^{(m+u)/2-1}nvr+g(3^{(m+u)/2-1}\sqrt{3}nvr)]\,,\end{aligned} \tag{21}$$

where

$$g(x)=f(x)+f(\sqrt{3}x)\,. \tag{22}$$

Similar to Eq. (17), it is given

$$\begin{aligned}T_1^{*-1}T_1^* f(x)&=\sum_{m,n,v=1}^{\infty}(-1)^{m-1}\mu(n)\left(\sum_{u=0}^{\infty}(-1)^{u}+\sum_{u=1}^{\infty}(-1)^{u-1}\right)g(3^{(m-1)/2+u/2}nvr)\\&=\sum_{m,n,v=1}^{\infty}(-1)^{m-1}\mu(n)[f(3^{(m-1)/2}nvr)+f(3^{(m-1)/2}\sqrt{3}nvr)]=\sum_{n,v=1}^{\infty}\mu(n)f(nvr)=f(r)\,.\end{aligned} \tag{23}$$

The three-dimensional (3D) close-packed hexagonal structure lattice can be treated in the similar way.

The cohesive energy for fcc structure can be written as

$$\begin{aligned}E(x)=&3\sum_{n=1}^{\infty}\Phi(nx)+6\sum_{p,q=1}^{\infty}(\Phi[(p^2+q^2)^{1/2}x]+\Phi\{[(p-\tfrac{1}{2})^2+(q-\tfrac{1}{2})^2]^{1/2}x\})\\&+4\sum_{p,q,u=1}^{\infty}(\Phi[(p^2+q^2+u^2)^{1/2}x+3\Phi\{[(p-\tfrac{1}{2})^2+(q-\tfrac{1}{2})^2+u^2]^{1/2}x\})\\=&6\sum_{n=1}^{\infty}[\Phi(nx/\sqrt{2})+\tfrac{1}{2}\Phi(nx)]+6\sum_{p,q=1,p\neq q}^{\infty}(\Phi[(p^2+q^2)^{1/2}x]+\Phi\{[(p-\tfrac{1}{2})^2+(q-\tfrac{1}{2})^2]^{1/2}x\})\\&+4\sum_{p,q,u=1}^{\infty}(\Phi[(p^2+q^2+u^2)^{1/2}x]+3\Phi\{[(p-\tfrac{1}{2})^2+(q-\tfrac{1}{2})^2+u^2]^{1/2}x\})\,.\end{aligned} \tag{24}$$

Introducing an operator T_1^* so that

$$T_1^* f(x)=6\sum_{n=1}^{\infty}[f(nx/\sqrt{2})+\tfrac{1}{2}f(nx)]\,, \tag{25}$$

it can be shown, as before, that

$$T_1^{*-1}f(x)=\tfrac{1}{6}\sum_{m,n=1}^{\infty}(-\tfrac{1}{2})^{m-1}\mu(n)f(2^{m/2}nx)\,, \tag{26}$$

where $f(x)$ is any function which makes the above summations convergent. Based on this formula, Mookerjee *et al.* have completed an *ab initio* pairwise potential calculation for copper successfully[8] and Li *et al.* have completed the calculation for Cu, Al, and Ni.[9]

The cohesive energy for a bcc structure can be given as

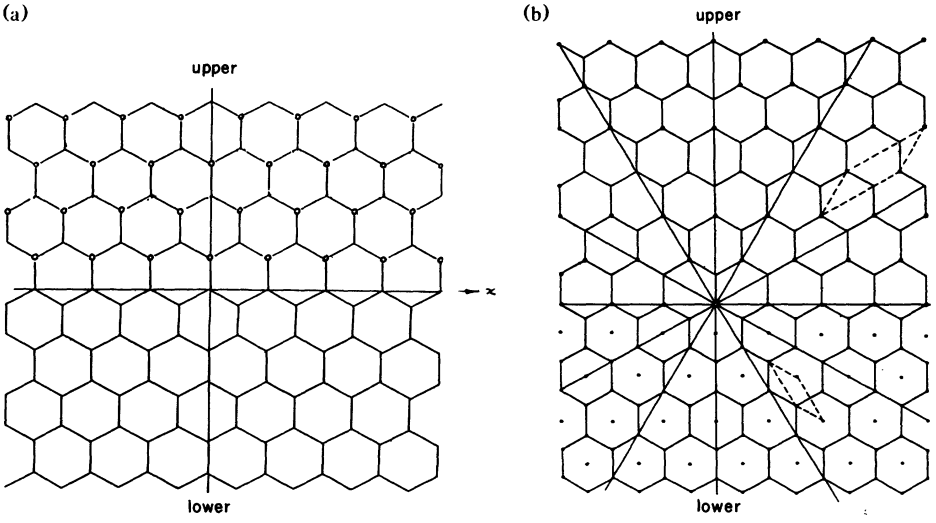

FIG. 2. (a) A 2D hexagonal lattice is separated into two parts by the line shown in this figure. All the points marked by ○ in the upper part are reflected to the lower part as the centers of all the hexagons. This is the so-called partial symmetry operation. (b) After the partial symmetry operation, both the upper and lower semiplanes become single structures.

$$
\begin{aligned}
E(x) &= 3\sum_{n=1}^{\infty}\Phi(nx)+6\sum_{p,q=1}^{\infty}\{\Phi[(p^2+q^2)^{1/2}x]\} \\
&\quad +4\sum_{p,q,u=1}^{\infty}(\Phi[(p^2+q^2+u^2)^{1/2}x]+\Phi[(p-\tfrac{1}{2})^2+(q-\tfrac{1}{2})^2+(u-\tfrac{1}{2})^2]^{1/2}x\}) \\
&= 4\sum_{n=1}^{\infty}[\Phi(\sqrt{3}nx/2)+\tfrac{3}{4}\Phi(nx)]+6\sum_{p,q=1}^{\infty}\Phi[(p^2+q^2)^{1/2}x] \\
&\quad +4{\sum}'(\Phi[(p^2+q^2+u^2)^{1/2}x]+\Phi\{[(p-\tfrac{1}{2})^2+(q-\tfrac{1}{2})^2+(u-\tfrac{1}{2})^2]^{1/2}x\})\,,
\end{aligned}
\tag{27}
$$

where the terms $\Phi(\sqrt{3}nx/2)$ are from diagonals (the most closed-packed direction in bcc structure), the terms $\Phi(nx)$ represent the axis which is the secondmost close-packed direction, and the $\sum'$ means that the sum is not over the case of $p=q=u$. For our modified CGE technique, we define the operator B by

$$
Bf(x)=4\sum_{n=1}^{\infty}[f(\sqrt{3}nx/2)+\tfrac{3}{4}f(nx)]\,, \tag{28}
$$

and the inverse operator can be obtained as

$$
B^{-1}f(x)=\tfrac{1}{4}\sum_{m,n=1}^{\infty}(-1)^{m-1}(\tfrac{3}{4})^{m-1}\mu(n)f[(2/\sqrt{3})^m nx] \tag{29}
$$

since

$$
\begin{aligned}
B^{-1}Bf(x) &= \tfrac{1}{4}\sum_{m,n=1}^{\infty}(-1)^{m-1}(\tfrac{3}{4})^{m-1}\mu(n)Bf[(2/\sqrt{3})^m nx] \\
&= \sum_{m,n,s=1}^{\infty}(-1)^{m-1}(\tfrac{3}{4})^{m-1}\mu(n)\{f[(2/\sqrt{3})^m(\sqrt{3}/2)nsx]+(3/4)f[(2/\sqrt{3})^m nsx]\} \\
&= \sum_{n,s=1}^{\infty}\mu(n)f(nsx)=f(x)\,.
\end{aligned}
\tag{30}
$$

Li *et al.* have completed a calculation for Mo and Cr successfully [9].

The Möbius function and the Möbius theorem are introduced to solve the important inverse cohesive energy problem proposed by Carlsson, Gilatt, and Ehrenreich, and a modified CGE scheme is obtained with very fast convergence. There are several reasons for the fast convergence. First, T_1^* has covered the most important series of atoms instead of only the nearest neighbors. Second, the Möbius function allows cancellations of many terms that have to be calculated in the original CGE scheme. Third, the higher-perturbation terms play a much smaller role since the operator $T_1^{*\,-1}$ is much more powerful then T_1^{-1}. By this method, many calculations for pairwise potentials become realistic since *ab initio* calculations for cohesive energy with variable lattice spacing are already available today. Of more importance is that the method indicates the potential application of number theory to physics.[5,6,10–13]

As Carlsson, Gelatt, and Ehrenreich have indicated,[1] there exist here the advantages and shortcomings of the radial pairwise potential approximation represented by Eq. (1). It might be desirable to include volume-dependent or density-dependent effects in a more explicit fashion, accompanied by the presented method. As well as the ion-ion interaction,[11] the anisotropic properties are also needed for further study.

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