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Method to construct transferable minimal basis sets for *ab initio* calculations

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We propose a scheme to construct transferable minimal basis of localized orbitals for *ab initio* calculations. We first extract a set of highly localized Wannier-type orbitals from the reference systems. For each orbital, we decompose it to a pseudoatomic orbital, augmented by small local functions centered at its nearest-neighbor atoms. When applied for a real system, the center of each local function moves with its associated atoms, without changing its shape and amplitude. We have done intensive tests of this scheme for III–V and group IV semiconductors and find the modified orbitals have very good transferability while still keep the basis size minimal. This work discusses why Wannier and Wannier-type functions are not transferable as one may expect.

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I. INTRODUCTION

The first-principles method based on the density-functional theory (DFT) (Refs. 1 and 2) is nowadays an indispensable tool to explore the physical properties of condensed-matter and materials systems. Plane-wave basis (PWB) set has been used widely because it has many attractive features.[3] For instances, the basis set is independent of atom positions, therefore it is easy to implement, and the forces can be easily calculated. Furthermore the quality of the basis can be systematically improved by a single parameter, energy cutoff. Plane wave basis is very efficient for a system of modest size.[3] However, it become very inefficient as the system size grow up since the computational effort needed increase as N^3 with respect to the number of inequivalent atoms N in the system, which limit the simulation in the systems up to hundreds of atoms.

Alternatively, highly localized basis set can be much more efficient, especially for the large systems. Because of the localization character, the localized basis are very suitable for the so-called order-N method.[4] Therefore, there has been continuous interests in constructing high quality localized basis set for DFT calculations.

In principle, there are two ways to construct local basis set. One is the "bottom up" approach, including various scheme to construct numerical orbitals.[5–16] For example, the numerical orbitals can be constructed by applying certain confinement potentials to the isolated atoms.[6,14] To ensure the transferability of the orbitals, one could use larger basis set, by using more than one radial function for each angular moment (multi ζ), or by including higher angular moment orbitals (polar orbitals). It is also possible to use smaller basis set, provided the radial functions are optimized as part of the self-consistency cycle.[16] The alternative way to construct the local basis is a "top down" method. One may extract local orbitals, e.g., the Wannier functions[17,18] from the Bloch states calculated for some reference systems, assuming that they are transferable to systems which are similar to the reference systems. These local orbitals form the minimal basis that are exact for the reference states. This scheme could potentially have many applications since many nanosized systems (e.g., the self-assembled quantum dots[19]) are extremely close to their bulk materials, except that they are usually strained or alloyed. One may expect that Wannier function should work very well in these situations. Unfortunately, it has been shown that the transferability of the Wannier functions is unsatisfactory even when the chemical environment is very similar to the reference systems.[4,20]

In this work, we propose a scheme to improve the transferability for Wannier (or Wannier-type) functions that could be used as transferable minimal basis for *ab initio* calculations. Our starting point is a set of highly localized Wannier-type orbitals constructed from some reference systems. For each orbital, we decompose it to a pseudoatomic orbital, augmented by small local functions centered at its nearest-neighbor (NN) atoms. When applied for a real system, the center of each local function moves with its associated atoms without changing its shape and amplitude. We have done intensive tests of this scheme for III–V and group IV semiconductors. We found the modified orbitals have very good transferability while still keep the basis size minimal. The work also discusses on why Wannier and Wannier-type functions are not transferable as one may expect.

The remainder of this paper is organized as follows. In Sec. II we describe the details on how to construct the minimal local basis. We present the test results of the basis sets for typical III–V and group IV semiconductors in Sec. III. We summarize in Sec. IV.

II. METHODS

The calculations are all based on DFT, within local-density approximation. We use norm-conserving,[21] fully separable pseudopotentials,[22] and Monkhorst-Pack k points sampling.[23]

The first step of our scheme is to construct a set of highly localized orbitals for a given reference system. There are many ways to do this, e.g., the maximally localized Wannier orbitals proposed by Marzari and Vanderbilt[24] and later on was generalized to include partially occupied bands[25] and to the nonothorgonal Wannier functions.[26] In this work, we use the highly localized quasiatomic minimal basis orbitals (QUAMBOs) proposed by Lu *et al.*[27] as our starting point to construct localized, transferable minimal basis.

Details on how to construct QUAMBOs can be found in Ref. 27. Here we only give a brief description to this method. The objective is to construct a set of highly localized atomic-like orbitals, $A_\alpha(\mathbf{r}-\mathbf{r}_i)$, centered at atomic position $\mathbf{r}_i$, by linear combinations of the Bloch orbitals. We first choose a set of bands $\phi_\mu(\mathbf{k},\mathbf{r})$ that we want to represent faithfully by the local orbitals. For self-consistent DFT calculations, all valence bands must be included to give correct charge density. We may also include certain low energy conduction bands, if we want to study, for example, the optical transitions of the systems. Additional higher Bloch bands are used to make the orbitals more localize. These bands were called *virtual* bands.[27] The virtual bands are not necessarily faithfully represented by the QUAMBOs. We can write the QUAMBOs as

$$A_\alpha(\mathbf{r}-\mathbf{r}_i) = \sum_{\mathbf{k},\mu} a_{\mu,\alpha}(\mathbf{k},\mathbf{r}_i)\phi_\mu(\mathbf{k},\mathbf{r}) + \sum_{\mathbf{k},p} b_{p,\alpha}(\mathbf{k},\mathbf{r}_i)\varphi_p(\mathbf{k},\mathbf{r}) \tag{1}$$

where,

$$\varphi_p(\mathbf{k},\mathbf{r}) = \sum_\nu T_{\nu p}(\mathbf{k})\phi_\nu(\mathbf{k},\mathbf{r}) \tag{2}$$

is the optimal subset of the virtual bands $\phi_\nu(\mathbf{k},\mathbf{r})$. $T_{\nu p}(\mathbf{k})$ is a rectangular matrix that satisfies the orthogonal condition $\Sigma_\nu T^*_{\nu p}(\mathbf{k})T_{\nu q}(\mathbf{k})=\delta_{pq}$. The central idea of this method is to choose the optimal subset of the Bloch states (i.e., the T matrix) so that $A_\alpha(\mathbf{r}-\mathbf{r}_i)$ have "maximal similarity" to the free-atom pseudoatomic orbitals (PAOs) $\Phi_\alpha(\mathbf{r}-\mathbf{r}_i)$ while still represent the selected Bloch bands $\phi_\mu(\mathbf{k})$ faithfully. More details on how to construct QUAMBOs can be found in Ref. 27. Generally, the QUAMBOs are more localized if more virtual bands are used. There is a special case, where the number of the bands we want to faithfully represent equals the number of the local orbitals. In this case, no virtual bands is used and the obtained local orbitals are just a set of generalized Wannier orbitals. For III–V semiconductors, we have eight local orbitals.

To construct QUAMBOs, we first carried out first-principles calculation using PW basis sets. We use the energy cutoff 36 Ry, and $6\times6\times6$ k mesh. In our calculations for the III–V compounds, we selected the lowest five bands (four valence bands and the lowest conduction band) to be represented faithfully. We use additional 59 bands as the virtual bands. We found no apparent difference if 123 bands are used as the virtual bands. Figure 1 depicts the QUAMBO s orbitals of Ga and As atoms alone the [111] direction, compared with the PAOs of free atoms. We can see that the QUAMBOs and corresponding PAOs are very similar to each other, except near the center of their adjacent atoms.

The obtained QUAMBOs can be expanded into Kubic functions[28] or into spherical harmonics. Here we choose spherical harmonic functions to do the expansion. The αth orbital of ith atom can be written as

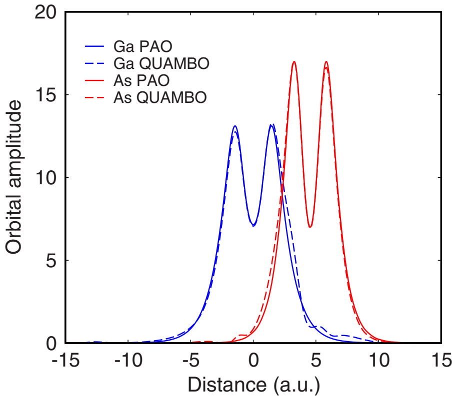

FIG. 1. (Color online) Profiles of the QUAMBOs s orbitals in GaAs alone the [111] direction, compared with the PAOs. The Ga and As atoms are located at 0 and about 5 a.u., respectively.

$$A_{i\alpha}(\mathbf{r}) = \sum_{l=0}^{l_{\max}}\sum_{m=-l}^{l}\chi_{lm}(r)Y_{lm}(\hat{r}). \tag{3}$$

Here, l and m are the angular-momentum quantum number and the magnetic quantum number, respectively. In practice, we found use $l_{\max}=18$ can represent the QUAMBOs very well.

One may expect that the QUAMBOs could be a transferable minimal basis set because they are very localized and are extremely close to the PAOs. However, as will be shown in latter sections, the QUAMBOs are not transferable as one might think. The reason, which can be seen from Fig. 1, is that the orbitals are different from PAOs near the center of their adjacent atoms. The deviation from PAOs is unavoidable since we know that PAOs cannot faithfully represent the electronic structures of the solid system.[8] In this work, we propose a unique scheme to improve the transferability of the QUAMBOs. This scheme can be used to other type of local basis set, such as Wannier functions as well.

We rewrite Eq. (3) into a multicenter projection form

$$A_{i\alpha}(\mathbf{r}) = c_{i\alpha}\Phi_{i\alpha}(\mathbf{r}) + \sum_{j=1}^{\mathrm{NN}} a_{i\alpha j}\Theta_{i\alpha j}(\mathbf{r}-\mathbf{r}_j), \tag{4}$$

where, $\Phi_{i\alpha}$ is the αth PAO centered at the ith atom and $\Theta_{i\alpha j}(\mathbf{r}-\mathbf{r}_j)$ is a set of local wave function centered at $\mathbf{r}_j$ which is the position of its adjacent atoms. Usually $\Theta_{i\alpha j}(\mathbf{r})$ is extremely small except at the NNs of the ith atom because the QUAMBOs is constructed to have most overlaps with the PAOs. We therefore only sum j over the NNs of the ith atom. When use this basis set, we assume the coefficients $c_{i\alpha}$, $a_{i\alpha p}$ and the shape of $\Phi_{i\alpha}(\mathbf{r})$ and $\Theta_{i\alpha j}(\mathbf{r}-\mathbf{r}_j)$ remain unchanged (except an overall normalization factor) while only their position move with the center of the atoms. The scheme is motived by the fact that the Wannier functions are the solutions of the on-site (pseudo)potential plus *screened* potentials of nearby atoms.[29] The augmentations to the PAO are due to the screened potentials of nearby atoms and should move with the atoms. $\Theta_{i\alpha j}(\mathbf{r}-\mathbf{r}_j)$ can also be expanded into spherical harmonic functions. Here, we use $l_{\max}=2$. To ensure strict localization, we smoothly truncate the wave function,[12,14] us-

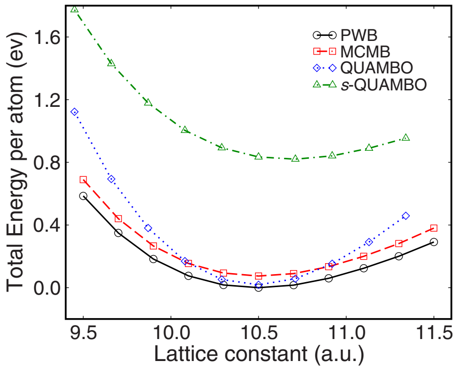

FIG. 2. (Color online) Comparison of the total energies of GaAs as functions of lattice constant using the PWB, QUAMBOs, *s*-QUAMBOs, and MCMB.

ing a Fermi-distribution-like function. The truncation radii are chosen to be 6 a.u. unless otherwise noticed in the paper. In all calculations, we use the periodic boundary conditions and the linear combination of atomic orbitals Hamiltonian is obtained in Bloch space. As a consequence, the Hamiltonian includes interactions up to infinite neighbors.[8] We name the basis set constructed based on the above scheme as the multicenter minimal basis (MCMB). When the MCMB is applied in *ab initio* calculations, every atom in the system has different orbitals. However, the orbitals can be easily constructed according to some simple rules, i.e., by simply moving the centers of the augmentation functions. The cost of constructing the orbitals is minima and proportional to the number of atoms in the system. Importantly, the size of resulting Hamiltonian matrix is the same as if an usual minimal basis set is used. Therefore, it is very efficient in the following iteration process, which is the most time consuming part of the calculation.

III. RESULTS AND DISCUSSION

We have done intensive tests of the MCMB for typical III–V and group IV semiconductors under isotropic strain, biaxial strain, and alloys. The overall results for the MCMB are very satisfactory. We show the most results for GaAs as an example but similar results are obtained for other materials too.

A. Isotropic strain

Figure 2 depicts the total energies of GaAs as functions of lattice constant calculated by using different basis sets. The total energy calculated by using QUAMBOs is almost exact to PW result (within 38 meV) at equilibrium lattice constant (the reference state). However, away from the equilibrium lattice constant, the total energy increase much faster than that calculated from PW basis. This is because that the QUAMBOs have many details that worsen the transferability. To improve the transferability of the basis, we may smooth the QUAMBOs by using less spherical Harmonics functions in Eq. (3). The results for the smoothed QUAMBOs (*s* QUAMBOs) with $l_{\max}=2$ are shown in a blue curve in Fig. 2. We see that the *s* QUAMBOs are much more transferable than the original QUAMBOs. However, the total energies calculated using the *s* QUAMBOs are much higher than those of the PWB. The total energy is 0.83 eV per atom higher than that from PWB, even at the reference state. Therefore, the direct use QUAMBOs or *s* QUAMBOs are not satisfactory. We now turn to the MCMB. We see that the transferability in the sense of total energy is very good. The total-energy curve as the function of lattice constant is almost identical to that of the PW results with energy cutoff 18 Ry, which is about 74 meV higher per atom than the PW results with energy cutoff 36 Ry. This performance is comparable to the optimized DZP, (i.e, two radial functions for each angular moment plus one polar orbital), even TZP (i.e., three radial functions for each angular moment plus one polar orbital) bases in Ref. 14, which have much larger basis sizes.

In Table I, we compare the calculated lattice constants and bulk modulus using the MCMB to those calculated from PW basis and to the experimental values, for a wide range of semiconductor materials. We see that the error of lattice constants compared with PW results are generally within 0.4%. The errors for In compounds are slightly lager. For bulk modulus, the errors are somehow larger compared to the PWB results, especially for InAs and InSb, in which the bulk modulus are about 14% overestimate PWB calculation. We find, however, by including the In *d* orbitals in the basis can reduce the errors to about one third of those if no *d* orbitals are used for both lattice constants and bulk modulus of In compounds. On the other hand, although the equilibrium lattice constants calculated from the original QUAMBOs are very good, the calculated bulk modulus is way overestimated. For example, the QUAMBOs give 170 GPa for GaAs, which is more than twice of the PWB result.

We then test the performance of MCMB in the band-structure calculations. Figure 3 shows the lowest eight bands (four valence bands plus four conduction bands) calculated at lattice constants $a=9.45$, 10.50, and 11.55 a.u., respectively. The results of PWB are plotted in solid lines as the references, whereas the MCMB bands are plotted in the dotted lines. We see that for the valence states, the bands of the MCMB are almost identical to the PWB ones at all three lattice constants. The lowest conduction bands are slightly worse but still quite good. However, the higher conduction bands are shifted to higher energy and very different from the PW results even at the reference state. This is because when constructing the QUAMBOs, we only require that the lowest five bands to be faithfully represented. The higher conduction bands are not guarantee to be correct in the first place. We also calculate the band structures using MCMB constructed using only eight bands (i.e., the Wannier orbitals). At the reference lattice constant, these orbitals gives much better band structure for the conduction bands than the above results. But they also fail for the higher conduction band when away from the reference state. We attribute this failure to the antibonding nature of the conduction bands, which are very sensitive to the local basis.

Spillage[7,8] is a useful parameter that measures the ability of the local basis to represent PW eigenstates. The spillage for a given local basis set is

TABLE I. Comparison of the calculated lattice constants and bulk modulus B using PWB and MCMB for typical III–V and group IV materials. An energy cutoff of 36 Ry and $4\times4\times4$ k mesh are used for the PWB calculation unless otherwise noticed. The MCMB calculation is constructed using 64 bands and $6\times6\times6$ k mesh. Calculations using MCMB are carried out using $4\times4\times4$ k points and energy cutoff 36 Ry unless otherwise indicated.

	Lattice constant (a.u.)			B (GPa)		
Materials	PWB	MCMB	Experiment	PWB	MCMB	Experiment
GaAs	10.50	10.51	10.68[a]	74	72	75.57[c]
GaP[e]	10.09	10.11	10.30[a]	90	88	89[d]
GaSb	11.36	10.40	11.52[a]	59	61	57[d]
InAs[e]	11.28	11.22	11.45[a]	62	71	60[d]
InP[e]	10.93	10.87	11.09[a]	72	76	71[d]
InSb	12.09	11.97	12.24[a]	53	60	47[d]
AlAs[e]	10.59	10.60	10.70[a]	74	77	77[d]
AlP[f]	10.21	10.23	10.33[a]	84	86	86[d]
AlSb	11.54	11.52	11.59[a]	58	62	58[d]
Ge	10.61	10.58	10.70[c]	73	78	77.20[c]
Si[g]	10.21	10.25	10.26[b]	92	101	99[b]
C[h]	6.76	6.76	6.75[b]	466	465	442[b]

[a] Reference 31.
[b] Reference 32.
[c] Reference 33.
[d] Reference 34.
[e] A $6\times6\times6$ k mesh is used.
[f] A $6\times6\times6$ k mesh and energy cutoff 50 Ry are used.
[g] Local orbitals truncated at 5.5 a.u.
[h] Local orbitals truncated at 4.0 a.u.

$$\mathcal{S}=\frac{1}{N_k}\frac{1}{N_\alpha}\sum_{\mathbf{k}=1}^{N_k}\sum_{\alpha=1}^{N_\alpha}\langle\psi_\alpha(\mathbf{k})|1-\hat{P}(\mathbf{k})|\psi_\alpha(\mathbf{k})\rangle, \tag{5}$$

where $|\psi_\alpha(\mathbf{k})\rangle$ are the Bloch states calculated from PW basis for a certain k points. N_k and N_α are the number of k points and bands, respectively. $\hat{P}(\mathbf{k})$ is the projector operator defined as

$$\hat{P}(\mathbf{k})=\sum_{\mu\nu}|A_\mu(\mathbf{k})\rangle S_{\mu\nu}^{-1}(\mathbf{k})\langle A_\nu(\mathbf{k})|, \tag{6}$$

where

$$A_\mu(\mathbf{k})=\sum_{\mathbf{R}}A_\mu(\mathbf{r}-\mathbf{r}_\mu-\mathbf{R})e^{i\mathbf{k}\cdot(\mathbf{r}_\mu+\mathbf{R})} \tag{7}$$

in which $\mathbf{r}_\mu$ is the atomic coordinates in the unit cell and $\mathbf{R}$ the lattice vector.

$$S_{\mu\nu}(\mathbf{k})=\langle A_\mu(\mathbf{k})|A_\nu(\mathbf{k})\rangle \tag{8}$$

is the overlap matrix of the local basis. We calculate the spillage of MCMB to the occupied bands for GaAs under isotropic strain. We found that the spillage is between $6-7\times10^{-4}$ as the lattice constant vary from $0.9a_0$ to $1.1a_0$. These values are almost an order of magnitudes smaller than 4×10^{-3} if simple truncate PAOs are used as the basis.

We further compare the self-consistent charge densities of the MCMB to those calculated from PWB. The error of charge density is calculated as

$$\Delta\rho/N_e=\frac{1}{N_e}\int|\rho_{\mathrm{MCMB}}(\mathbf{r})-\rho_{\mathrm{PWB}}(\mathbf{r})|d^3\mathbf{r}, \tag{9}$$

where N_e is the total electrons in a unit cell. The error calculated at the equilibrium lattice constant is 1.08% and is smaller than 1.5% in the range of 9.5–11.5 a.u. We recalculated the band structures at the three lattice constants using the PWB but fixed the charge densities to those calculated from the MCMB. The results are shown in Fig. 4, compared to the full PW calculations. Remarkably, the bands calculated from the MCMB charge densities have virtually no difference with the full PW calculations. We may then have a very useful scheme if the high accuracy band structures (especially the conduction bands) are needed: the charge density which is the most time consuming part can be calculated by the MCMB, whereas the band structure can be calculated by the PWB. This scheme can be best combined with the so-called folded spectrum method to give a very efficient method,[30] if only a small number of bands around certain reference energy is needed.

B. Biaxial strain

In this section, we test the transferability of the MCMB under biaxial strains. In many systems, the material is biaxially strained, for examples, the epitaxially grown superlattice, self-assembled quantum dots, etc. The biaxial strain is defined as

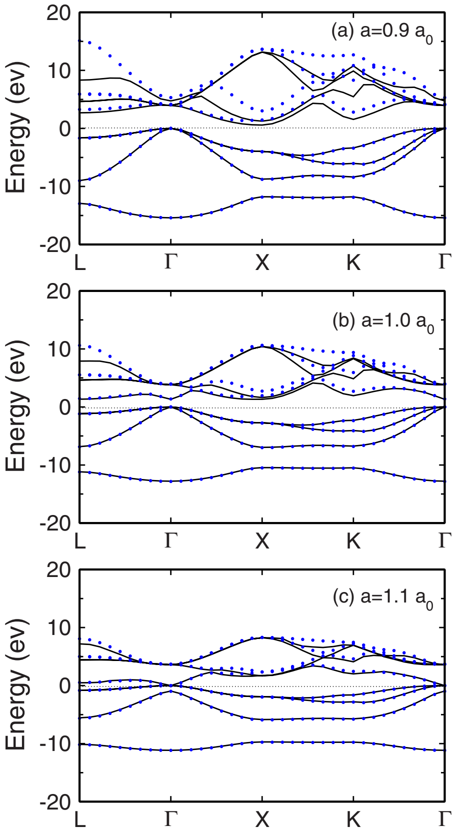

FIG. 3. (Color online) Band structures of GaAs of different lattice constants, (a) $0.9a_0$, (b) a_0, and (c) $1.1a_0$. The PWB results are shown in the solid black lines, whereas the results from MCMB are shown in dotted blue lines.

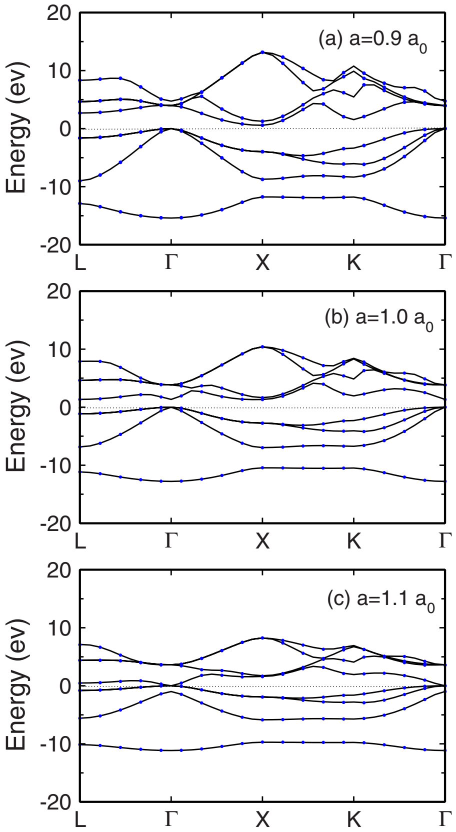

FIG. 4. (Color online) Band structure of GaAs under different lattice constants, (a) $0.9a_0$, (b) a_0, and (c) $1.1a_0$. The solid black lines are the band structures from full PWB calculations, whereas the dotted blue lines are the band structures calculated by PWB using the MCMB charge densities.

$$\epsilon_b = \epsilon_{zz} - (\epsilon_{xx} + \epsilon_{yy})/2. \tag{10}$$

In the pure InAs/GaAs quantum dots, the biaxial strain is usually about 0.15. The test is performed on the eight-atom cubic cell of GaAs. We apply the biaxial strain by varying the in-plane lattice constants $a=b$ while keep the volume of the unit-cell constant. The atomic positions are relaxed with the fixed lattice constants using PW codes.

Figure 5 depicts the total energy per atom with respect to the biaxial strain. At the reference state ($\epsilon_b=0$), the total energy calculated from QUAMBOs are extremely close to that calculated from PWB. However, again, the transferability of QUAMBOs is very bad. In contrast, MCMB show remarkable transferability in the wide range of biaxial stain from $\epsilon_b=-0.30$ to 0.30. The total energies are also within 74 meV per atom higher than the PW results with energy cutoff 36 Ry.

Figures 6(a)–6(c) compare the density of states (DOS) calculated using MCMB to those using PWB, at $\epsilon_b=0.15$, 0, and -0.15, respectively. The DOS is plotted with a Gaussian smearing of 0.07 eV. As we see, the valence states are almost identical for the two bases. The lowest states above the Fermi level are also very close. However, the higher conduction bands can not be described very well by MCMB and show large discrepancy to the PW results, as expected.

C. Alloy

Alloys provide a challenge test for the transferability of local orbitals because in addition to the atomic position distortions, the chemical environment are very different from their bulk materials. We carry out the tests in the $\mathrm{In}_{1-x}\mathrm{Ga}_x\mathrm{As}$ supercells containing 64 atoms. Since in the present case, we care only about the electronic structures, we relax the atomic position using a PW code. The lattice constants of the supercell are approximated by $a=x\cdot a_0(\mathrm{GaAs})+(1-x)\cdot a_0(\mathrm{InAs})$,

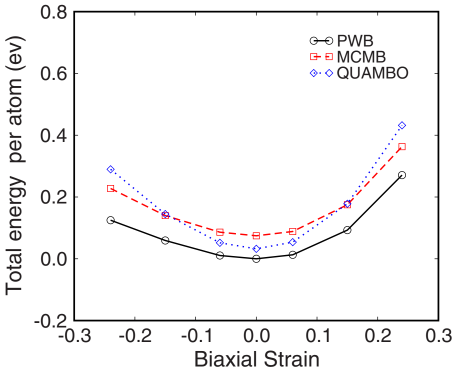

FIG. 5. (Color online) Comparison of the total energy verse biaxial strain using the PWB, QUAMBOs, and MCMB.

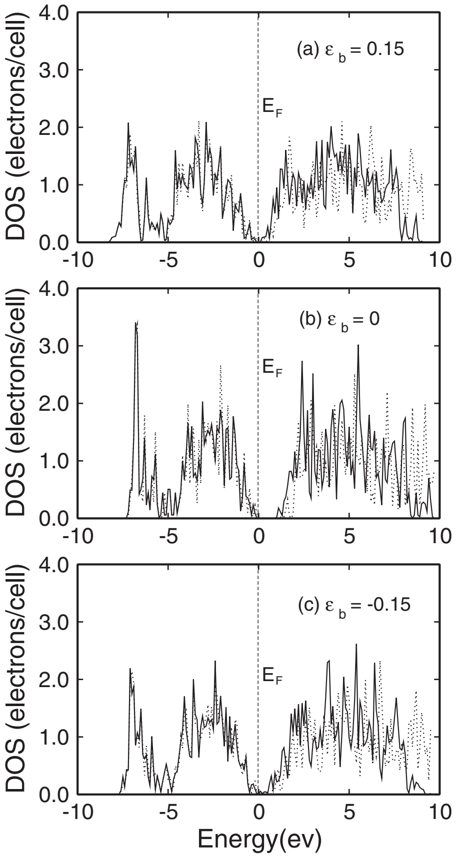

FIG. 6. Density of states of GaAs supercell containing eight atoms under biaxial strain for (a) $\epsilon_b=0.15$, (b) $\epsilon_b=0$, and (c) $\epsilon_b=-0.15$.

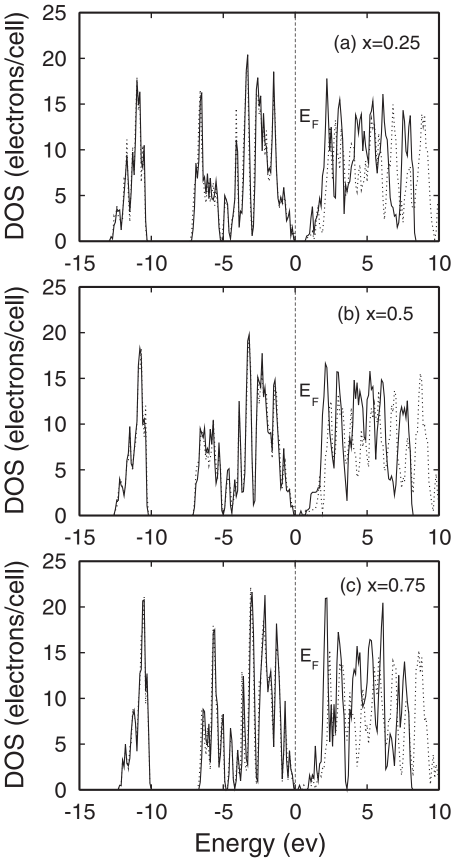

FIG. 7. Density of states of $\mathrm{Ga}_x\mathrm{In}_{1-x}\mathrm{As}$ alloys with (a) $x=0.25$, (b) $x=0.5$, and (c) $x=0.75$.

where a_0(GaAs) and a_0(InAs) are the lattice constants of GaAs and InAs, respectively. In the alloy, the As atoms may have Ga or In as their NN atoms at the same time. We then have two sets of $\Theta_{i\alpha j}$ functions for As atoms, one set from GaAs and the other from InAs, whereas the center of the As PAO $\Phi_{i\alpha}(\mathbf{r})$ are equal for the two sets. We choose the $\Theta_{i\alpha j}(\mathbf{r}-\mathbf{r}_j)$ functions according to the atomic type on $\mathbf{r}_j$ site. The DOS of the alloys are compared in Figs. 7(a)–7(c) for three compositions $x=0.25$, 0.5, and 0.75, respectively. We see that the DOS calculated from MCMB (dotted lines) are in remarkable agreement with those from PWB (solid lines) for valence electrons and the low energy conduction states. The DOS high energy states are very off from PWB results as expected.

Occasionally one has to deal with the defects that may change the coordination number of the bulk materials, such as vacancies, interstitials, etc. In these cases, the direct use of MCMB for the vacancies, interstitials, etc. may not be satisfactory. However, one can use (standard) larger local basis sets for the *few* atoms around vacancies, interstitials, etc. while use MCMB for the rest atoms to achieve high accuracy and the same time efficiency.

IV. SUMMARY

Wannier (or Wannier-type) functions are themselves minimal basis sets that are exact for the reference states. However, the Wannier and Wannier-type function are unavoidably have structures around their adjacent atoms that hamper their transferability for the *ab initio* calculations. We have proposed a unique scheme to construct transferable localized minimal basis for efficient, large scale DFT calculations. The local orbitals are composed of a (truncated) PAO Plus augmentation localized in its nearest atoms. We have tested the transferability of the basis sets under isotropic strain, biaxial strain, and alloys for typical III–V and group IV semiconductors, and obtained very satisfactory results. The scheme could be useful for studying extremely large systems that are very close to their bulk compounds, such as alloys, defects, super lattices, quantum wires, quantum dots, etc. We believe that the MCMB is a very useful supplement to the standard extended atomic basis sets (example, multi-ζ basis, etc.). The work also clarified why the generalized Wannier functions or Wannier-type functions are not transferable.

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