Efficient calculation of magnetocrystalline anisotropy energy by Wannier interpolation

Junfeng Qiao\textsuperscript{1,2} and Weisheng Zhao\textsuperscript{1,2,*}

\textsuperscript{1}Fert Beijing Institute, BDBC, Beihang University, Beijing 100191, China
\textsuperscript{2}School of Electronic and Information Engineering, Beihang University, Beijing 100191, China

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\textit{Ab-initio} calculation of magnetocrystalline anisotropy energy (MCAE) often requires a strict convergence criterion and a dense $k$-point mesh to sample the Brillouin zone, making its convergence problematic and time-consuming. The force theorem for MCAE states that MCAE can be calculated by band energy difference between two magnetization directions at a fixed potential. The maximally localized Wannier function can be utilized to construct a compact Hilbert space of low-lying electron states and interpolate band eigenvalues with high precision. We combine the force theorem and the Wannier interpolation of eigenvalues together to improve the efficiency of MCAE calculations with no loss of accuracy. We use an iron chain as an example and demonstrate that the Wannier interpolation method for MCAE is able to reduce the computational cost significantly and remain accurate simultaneously, compared with a direct \textit{ab-initio} calculation on a very dense $k$-point mesh. This efficient Wannier interpolation approach makes it possible for large-scale and high-accuracy MCAE calculations, which could benefit the design of spintronics devices.

I. INTRODUCTION

In ferromagnetic (FM) materials, the magnetic anisotropy energy (MAE) is the difference of magnetizing energies along two directions. MAE can be mainly separated into extrinsic part and intrinsic part. The extrinsic shape anisotropy, that MAE correlates with the shape of the FM material, originates from the magnetic dipole-dipole interaction \cite{1}. The intrinsic magnetocrystalline anisotropy energy (MCAE), stems from the collective effect of the crystal structure and spin-orbit coupling (SOC). The orbital motions of electrons are restricted to specific orientations by the crystal field, and the SOC couples the spin degree of freedom to the orbital motion. Consequently the energy of the crystal depends on the magnetization orientation \cite{2}.

Apart from the interest in fundamental science, the practical applications have also stimulated lots of research into the understanding and optimization of MCAE \cite{3–10}. One of the applications is the magnetic tunnel junction (MTJ), which is the core structure of magnetic random access memory (MRAM) \cite{3, 11}. MTJ stores one bit of information by the relative magnetization orientation of the two FM layers, and the read process is based on tunneling magnetoresistance \cite{12–13}. Materials of large MCAE are beneficial to the stability of the information.

The MCAE is hard to be captured because of its small magnitude. The total energy of a crystal unit cell is at the order of $1 \times 10^3$ eV, while that of MCAE is at the order of $1 \times 10^{-3}$ eV, this significant contrast poses serious challenges not only to experimental instruments but also to numerical algorithms. As a result, calculation of MCAE still remains as an intriguing problem in \textit{ab-initio} community \cite{15–19}.

In \textit{ab-initio} calculations, plane wave (PW) algorithm is widely adopted because it is mathematically simple and in principle complete. The PW method provides the same accuracy at all points in space, this advantage on one side is appealing but on the other side is disappointing since it often requires large amounts of plane waves to converge and results in bad scaling behavior when system size increases. A more efficient approach is always desired. Maximally localized Wannier function (MLWF) is the Fourier transform of the reciprocal space PW wave function under the criterion of maximal localization of Wannier function (WF) in real space \cite{20}. MLWF not only gives a clearer physical insight of chemical bonds but have also manifested its necessities and accuracies in the calculations of many physical properties, such as electric polarization \cite{21}, anomalous hall conductance \cite{22} and orbital magnetization \cite{23}, etc. \cite{20}.

Here we extend the Wannier interpolation method to MCAE calculation for the purpose of improving efficiency. Our steps are summarized as follows: First, an \textit{ab-initio} calculation is performed on a relatively coarse $k$-point mesh (kmesh). Second, MLWFs are extracted from the acquired \textit{ab-initio} wave functions. Third, MCAE calculations are performed on a much denser kmesh in the Wannier representation. Compared to a direct \textit{ab-initio} calculation on the dense kmesh, this Wannier interpolation approach has equivalent accuracy but greatly reduced computational cost. The initial coarse kmesh is too sparse for MCAE to converge but the extracted WFs meet the criterion of "good" localization. Our Wannier interpolation approach could not only reduce the computational cost but also make it possible to calculate MCAE for large systems requiring extremal computational resources, and calculations requiring exceptional accuracy such as high order effect of MCAE with respect to the polar angle $\theta$ and azimuthal angle $\phi$ \cite{24–25}.
II. METHODS

The Wannier interpolation method for calculating MCAE is composed of two parts: The force theorem states that MCAE can be calculated by the difference of band energies of two magnetization directions; The MLWF provides an efficient way for interpolating eigenvalues to arbitrary dense kmesh based on coarse kmesh \textit{ab-initio} calculation. We first give a brief and self-contained introduction to the Wannier interpolation of band structure, then show our procedures for combining Wannier interpolation with the force theorem of MCAE. We followed the theory and mathematical notations used in [Yates, et al. (Ref. [20]) and Marzari, et al. (Ref. [21])]. The disentanglement of entangled bands is omitted for conciseness and detailed theory on MLWFs can be found in these papers.

A. Wannier interpolation

1. Construction of Wannier functions

In periodic crystal, the Bloch theorem allows us to write down the wave function as

\[ \psi_{nk}(r) = e^{ikr}u_{nk}(r), \]  

where \( k \) is the \( k \)-point vector, \( n \) is the band index, \( \psi_{nk}(r) \) is the Bloch wave function, \( u_{nk}(r) \) is periodic in real space.

Inserting Bloch function into the Kohn-Sham equation, we arrive at the equation for the periodic part of the Bloch function,

\[ \hat{H}(k)u(k) = \epsilon_k u(k), \]  

where \( k \) is the \( k \)-point vector, \( \hat{H}(k) \) is the transformed Hamiltonian \( \hat{H}(k) = e^{-ikr}\hat{H}e^{ikr}, \epsilon_k \) is the eigenvalue.

Usually one needs more than 10000 plane waves to expand the \( u(k) \) in PW method and the diagonalization of the 10000 \times 10000 Hamiltonian matrix is performed on each \( k \)-point. This is why a direct \textit{ab-initio} calculation on a dense kmesh is rather time-consuming.

Generally, Fourier transform enables us to have a different representation of functions in another domain, which may be useful for analysis of the problem, e.g. the frequency spectrum of audio signals and images. If treated properly, the inverse domain should be equivalent to the original domain, i.e. they are equivalent Hilbert space. We apply the Fourier transform to the periodic Bloch wave function \( \psi_{nk}(r) \) which lies in the Brillouin Zone (BZ),

\[ |R_n\rangle = \frac{V}{(2\pi)^3} \int_{BZ} dk e^{-ikr}R |\psi_{nk}\rangle, \]  

thus \( |R_n\rangle \) form an orthonormal set and span the same Hilbert space as \( \psi_{nk}(r) \).

In principle, a smooth function in real space results in a localized function in its reciprocal space, and vice versa. It is not naturally guaranteed that the simply summed Bloch function of Equ. (3) results in a smooth function \( |R_n\rangle \) in real space.

Fortunate enough, there is a gauge freedom left in the definition of Bloch function,

\[ |\tilde{\psi}_{nk}\rangle = e^{i\varphi_n(k)}|\psi_{nk}\rangle, \]  
or equivalently,

\[ |\tilde{u}_{nk}\rangle = e^{i\varphi_n(k)}|u_{nk}\rangle. \]

We can utilize this freedom to construct localized WF s in real space, the so-called maximally localized Wannier function.

We define the unitary transformation which takes the original Bloch function \( |u_{nk}\rangle \) to the smoothed function \( |\tilde{u}_{nk}\rangle \) (from now on written as \( |u_{Wnq}\rangle \), we use \( q \) here as to differentiate another kmesh in the following Wannier interpolation step) as

\[ |u_{Wnq}\rangle = \sum_{m=1}^{M} |u_{mnq}\rangle \mathcal{U}_{mn}(q), \]

where \( M \) is the number of states needed to be considered for our targeted physical properties, usually the low-lying states below Fermi energy or plus few empty states above Fermi energy. We call this unitary transformation as the transformation from Bloch gauge to Wannier gauge.

Thus, the Fourier transform pair between the smoothed Bloch functions and the MLWFs are

\[ |R_n\rangle = \frac{1}{N} \sum_{q} e^{-iq \cdot R} |u_{Wnq}\rangle, \]

\[ \mathcal{U}_{mn}(q) = \sum_{R} e^{iq \cdot R} |R_n\rangle, \]

where, \( N \) is the number of points in BZ.

2. Interpolation on arbitrary kmesh

For the reciprocal space Hamiltonian operator \( \hat{H}(q) \), we define the \( M \times M \) Hamiltonian matrix in the Wannier gauge as

\[ H_{nm}^{(W)}(q) = \langle u_{Wnq} \mid \hat{H}(q) \mid u_{Wm}\rangle = \langle u_{Wnq} \mid H(q) \mathcal{U}(q) \mid u_{Wm}\rangle_{nm}, \]

where \( H_{nm}(q) = \mathcal{E}_{nq}\delta_{nm} \), \( \delta_{nm} \) is the Kronecker delta function.

If diagonalizing \( H_{nm}^{(W)}(q) \) by

\[ \mathcal{U}(q) H^{(W)}(q) \mathcal{U}(q)^\dagger = H^{(H)}(q), \]

where \( H_{nm}^{(H)}(q) = \mathcal{E}_{nq}\delta_{nm} \), then \( \mathcal{E}_{nq} \) will be identical to the original \textit{ab-initio} \( \mathcal{E}_{nq} \).
Transforming the Hamiltonian operator from reciprocal space to real space,

$$H_{nm}^{(W)}(R) = \frac{1}{N} \sum_{q} e^{-i \mathbf{q} \cdot \mathbf{R}} H_{nm}^{(W)}(q),$$

(10)

and then performing inverse Fourier transform

$$H_{nm}^{(W)}(k) = \sum_{\mathbf{R}} e^{i \mathbf{k} \cdot \mathbf{R}} H_{nm}^{(W)}(\mathbf{R}),$$

(11)

we succeed in interpolating the Hamiltonian operator on arbitrary k-point \( \mathbf{k} \).

Since the WFs we chose are maximally localized, the \( H_{nm}^{(W)}(R) \) is expected to be exponentially localized in real space, a few \( R \) are sufficient in the summation of Equ. (11).

The final step is to diagonalize \( H_{nm}^{(W)}(k) \),

$$U(k)^{\dagger} H^{(W)}(k) U(k) = H^{(H)}(k),$$

(12)

then the acquired eigenvalues on arbitrary k-point \( \mathbf{k} \) can be used for latter extractions of the targeted physical properties.

We comment here that since \( H^{(W)}(k) \) are of dimensions \( M \times M \), their diagonalizations are very "cheap", compared with the diagonalizations of the Hamiltonian matrices in PW method, which are on the order of \( 10000 \times 10000 \) dimensions.

Figure 1 shows the interpolated band structure of our latterly used iron chain compared with the original coarse kmesh \( \text{ab-initio} \) calculation and a dense kmesh \( \text{ab-initio} \) calculation. The calculation details will be described in section IIIA. The interpolated band structure is in excellent agreement with the dense kmesh \( \text{ab-initio} \) band, whether it locates at the coarse kmesh \( \text{ab-initio} \) \( q \)-points or the interpolated \( k \)-points between the \( q \)-points. Apparently, the coarse kmesh \( \text{ab-initio} \) calculation, from which the WFs are constructed, is far from convergence with respect to the dense kmesh \( \text{ab-initio} \) calculation.

**B. Magnetocrystalline anisotropy energy**

As mentioned in the introduction, one of the difficulties of calculating MCAE is its small magnitude compared to the total energy. Another difficulty is its rapid variation in \( k \)-space. As shown in the Fig. 2(b), the \( k \)-space resolved MCAE by Equ. (14), displays sharp peaks and steps along \( k_z \) direction. To capture these tiny but critical features, a sufficiently dense kmesh must be adopted, and slow convergences relative to kmesh are often the case in the calculations of MCAE.

In the \( \text{ab-initio} \) calculation of MCAE, the most natural and rigorous method is performing two self-consistent calculation of different magnetization directions, and the MCAE should be the difference of the total energies.

$$\text{MCAE} = E^{\theta=0^\circ} - E^{\theta=90^\circ},$$

(13)

where \( \theta \) is the spherical polar angle with respect to crystallographic \( c \) axis, \( E^{\theta=\alpha} \) is the total energy of magnetization pointing towards the \( \theta = \alpha \) direction, which in our case we choose \( \alpha = 0^\circ \) and \( 90^\circ \). The azimuthal angle \( \phi \) is kept fixed to 0 in all the calculations.

However, the cumbersome diagonalizations are rather time-consuming, especially when SOC is included, and reaching the energy minimal becomes much harder in the self-consistent iterations. According to force theorem [16, 17, 27, 29], the main contribution to the MCAE originates from the difference of band energies between two magnetization directions at fixed potential,

$$\text{MCAE}(k) = \sum_{i} n_{i,k} \epsilon_{i,k}^{\theta=0^\circ} - \sum_{i'} n_{i',k} \epsilon_{i',k}^{\theta=90^\circ},$$

\( MCAE = \frac{1}{N} \sum_{k} \text{MCAE}(k), \)

(14)

where \( \mathbf{k} \) is the \( k \)-point vector, \( i, i' \) are the band indexes of magnetization directions along \( \theta = 0^\circ \) and \( \theta = 90^\circ \), respectively. \( n_{i,k} \) is occupation number and \( \epsilon_{i,k} \) is the energy of band \( i \) at \( k \)-point \( \mathbf{k} \), \( N \) is the number of \( k \)-points.

Thus by non-self-consistent calculation of only a one-step diagonalization, the eigenvalues are obtained and the
MCAE can be calculated by Eqn. (14). The force theorem is widely adopted in literatures and its validity has been discussed in [Wang, et al. (Ref. [30])] and [Błoński, et al. (Ref. [19])]. Good matches between experimental and ab-initio results can be found in literatures [31, 32].

Another merit of force theorem is that by Eqn. (14) some k-space resolved analyses can be performed, such as the contribution of the quantum well states to the oscillation of MCAE [3].

The practical calculation of MCAE involves two steps. First, charge density is acquired self-consistently without taking into account SOC. Second, reading the self-consistent charge density, two non-self-consistent calculations are performed including SOC, with magnetization pointing towards the $\theta = 0^\circ$ direction and the $\theta = 90^\circ$ direction. Finally, MCAE is calculated by Eqn. (14).

Before we concluding the force theorem for MCAE, we would like to emphasize the importance of Fermi energy $E_F$. Since MCAE is such a small quantity that is on the order of $1 \times 10^{-3}$ eV, a small displacement of $E_F$ will propagate into the occupation numbers $n_{i\k}$, and then the MCAE by Eqn. (14). The MCAE will be significantly modified, even the sign could be changed. To accurately calculate MCAE, the small difference of Fermi energy between magnetizations along $\theta = 0^\circ$ and $\theta = 90^\circ$ must be considered. Since the number of electrons $N_{elec}$ are the same between two magnetization directions, the Fermi energies are calculated separately for $\theta = 0^\circ$ and $\theta = 90^\circ$ by

$$N_{elec} = \int_{E_F^{\theta=0^\circ}}^{E_F^{\theta=90^\circ}} n_{\theta^=0^\circ}(E)dE = \int_{E_F^{\theta=90^\circ}}^{E_F^{\theta=0^\circ}} n_{\theta^=90^\circ}(E)dE,$$

where $E_F^{\theta=\alpha}$ and $n_{\theta^=\alpha}(E)$ are the Fermi energy and density of states for magnetization along $\theta = \alpha$, $\alpha = 0^\circ$ or $90^\circ$. The occupation numbers $n_{i\k}$ are calculated separately for the two magnetization directions and finally MCAE is calculated by Eqn. (14).

In all, Wannier interpolation gives us the ability to efficiently interpolate band energies on arbitrary kmesh, the force theorem tells us the MCAE can be calculated by the difference of band energies. Combining these two theories together, MCAE calculations can be carried out on a much denser kmesh with no loss of accuracy but greatly reduced computational cost.

We implemented the code for MCAE calculations on the basis of WANNIER90 code [32, 35]. In the next section, we choose an iron chain as an example to demonstrate our method of MCAE calculation. On one hand, due to the lowered crystal symmetry, the iron chain system is expected to have medium magnitude MCAE; on the other hand, this system is small enough that an extremely accurate dense kmesh ab-initio calculation can be performed so that our Wannier interpolation method can be directly compared with high accuracy ab-initio results.

III. IRON CHAIN

A. ab-initio calculation and Wannierization

The ab-initio calculation were performed using QUANTUM ESPRESSO package based on projector-augmented wave (PAW) method and a plane wave basis set [30, 37]. The exchange and correlation terms were described using generalized gradient approximation (GGA) in the scheme of Perdew-Burke-Ernzerhof (PBE) parameterization, as implemented in the PS LIBRARY version 0.3.1 [38]. We used a wave function cutoff of 90 Ry and electron density cutoff of 1080 Ry. A Marzari-Vanderbilt cold smearing [39] of width 0.002 eV was adopted. Convergence relative to smearing width and k mesh will be detailedly discussed in the subsequent paragraph. Since the MCAE of the iron chain is on the order of $1 \times 10^{-3}$ eV, the energy convergence criteria of all the calculations were set as $1.0 \times 10^{-3}$ Ry. We kept a 15 Å vacuum space in the xy plane to eliminate interactions between periodic images. Since enough vacuum space was left in the xy plane, we set the Monkhorst-Pack k-point mesh to $1 \times 1 \times 24$ and confirmed that increasing it to $2 \times 2 \times 24$ or more had a negligible impact on MCAE. The unit cell contains one Fe atom [shown in Fig. 2a)] and the lattice constant along c axis (i.e. the atomic spacing along the iron chain) was set as 2.2546 Å, which was acquired by relaxation until the force acting on the Fe atom was less than $1 \times 10^{-2}$ Ry/Bohr.

To transform Bloch functions into MLWFs, first the overlap matrices $M_{mn}^{\alpha\beta}$ and the projection matrices $A_{mn}^{k}$ are extracted from the Bloch functions [20]. Then after disentanglement and Wannierization the MLWFs are constructed. We used 20 spinor WFs having the form of $sp3d2$, $dxy$, $dxz$, $dyz$ and s-like Gaussians. The disentanglement outer energy window was set as $[-100.0,10.0]$eV, and the inner window was set as $[-100.0,-3.41]$eV, which is slightly higher than the Fermi energy $-4.38$ eV. The spreads of WFs signify the quality of localization, as de-
where $|0n⟩$ are the WFs in the home unit cell. The convergence threshold of spread for both the disentanglement and Wannierization were set as $1 \times 10^{-10}$ Å$^2$. Apart from 2 WFs having the spread $Ω$ around 2.0 Å$^2$, the spread of the remaining 18 WFs were smaller than 1.0 Å$^2$, mostly around 0.2 Å$^2$. The total spread of WFs were around 10.0 Å$^2$ and the convergence issues will be discussed in the next section.

### B. Convergence issues of Wannier interpolation

Three main parameters should be tested to reach a reliable result of MCAE: the number of coarse $ab$-initio kmesh $n_k^{ab}$ for the construction of MLWFs, the number of Wannier interpolation kmesh $n_k^{wan}$ for the interpolation of MCAE from MLWFs, and the smearing width $σ$ that determines the fictitious smearing contributions to the total energies.

Note since the kmesh in xy plane is always set as $1 \times 1$, we will use the notation that for a function $f(x; y)$, the x before the semicolon is the variable while the y after the semicolon is the parameter which is held fixed. For example, MCAE($n_k^{ab}; n_k^{wan}$) represents the variation of MCAE relative to the coarse $ab$-initio kmesh $n_k^{ab}$ when the Wannier interpolation kmesh $n_k^{wan}$ is held fixed.

Next, we test the convergence of the spread relative to the number of coarse $ab$-initio kmesh $Ω(n_k^{ab})$, the convergence of MCAE relative to the number of coarse $ab$-initio kmesh MCAE($n_k^{ab}; n_k^{wan}, σ$), the convergence of MCAE relative to the number of Wannier interpolation kmesh MCAE($n_k^{wan}; n_k^{ab}, σ$), the convergence of MCAE relative to the smearing width MCAE($σ; n_k^{ab}, n_k^{wan}$) and the minimum number of Wannier interpolation kmesh needed relative to smearing width $n_k^{wan}(σ)$.

We use the subtracted function $∆f(x) = f(x) − f_{conv}$ to clearly displays the deviation of a function $f(x)$ when approaching the converged value of $f_{conv}$. Meanwhile, we use $δf(x) = |\max_{x≥x} f(y) − \min_{y≥x} f(y)|$ to display the convergence trend of a function $f(x)$, where $| . . . |$ means taking the absolute value.

#### 1. Spread of Wannier functions

The construction of Wannier functions relies on a uniform $ab$-initio kmesh, we call it as coarse $ab$-initio kmesh, since this kmesh does not need to be dense enough for the MCAE to be converged but is sufficient only when "good" localization of Wannier functions is reached.

Here we show the relation of WFs’ spread and the density of $ab$-initio kmesh in Fig. 3(a). In the MCAE calculations, the "good" criterion for the localization of WFs is ultimately determined by the convergence of the MCAE. We mention it in advance that a $1 \times 1 \times 2600$ interpolation kmesh and a smearing width of 0.0012 eV are sufficient for MCAE to be converged at the order of $1 \times 10^{-6}$ eV. The subsequent paragraphs will detailedly discuss the convergence issue related to interpolation kmesh and smearing width. We use this conclusion here to exclude the influence of the convergence of Wannier interpolation kmesh and the smearing width when discussing the relation of the convergence of the WFs’ spread and the MCAE relative to the density of the $ab$-initio kmesh.

We use $δΩ(n_k^{ab})$ to represent the convergence trend of the spread $Ω$ calculated on $ab$-initio kmesh $n_k^{ab}$

$$δΩ(n_k^{ab}) = |\max_{i≥n_k^{ab}} Ω(i) − \min_{i≥n_k^{ab}} Ω(i)|$$ (17)

As shown in the inset of Fig. 3(a), after $n_k^{ab} \geq 24$, the $δΩ$ is less than 0.2 Å$^2$.

We use $ΔMCAE(n_k^{ab})$ to represent the deviation of MCAE($n_k^{wan}$) relative to the converged MCAE$_{conv}$, which is defined as

$$ΔMCAE(n_k^{ab}) = MCAE(n_k^{ab}) − MCAE_{conv}$$ (18)

Then $n_k^{ab}$ is the $ab$-initio kmesh used for the construction of WFs. MCAE$_{conv}$ is the mean value of MCAE calculated with $ab$-initio kmesh from 22 to 30 and this should be the converged value since the variation between these 9 values are on the order of $1 \times 10^{-6}$ eV, as shown in the inset of Fig. 3(b). To validate the fixed Wannier interpolation kmesh of $n_k^{wan} = 2600$ is high enough, we define the convergence trend of the MCAE($n_k^{wan}; n_k^{ab}$) relative to $n_k^{wan}$ as

$$δMCAE(n_k^{wan}; n_k^{ab}) = |\max_{i≥n_k^{wan}} MCAE(i; n_k^{ab}) − \min_{i≥n_k^{wan}} MCAE(i; n_k^{ab})|$$ (19)

where $n_k^{wan}$ is in the range of 2500 to 2700. As shown by the red error bar in the inset of Fig. 3(b), at each specific $n_k^{ab}, δMCAE(n_k^{wan}; n_k^{ab})$ only varies less than $3 \times 10^{-6}$ eV when $n_k^{wan}$ changes from 2500 to 2700.

For $n_k^{ab} > 22$, the total spread of WFs converge to less than 0.2 Å$^2$, at the same time the MCAE converge at the order of $1 \times 10^{-6}$ eV as shown in the inset of Fig. 3(b). As in many other cases, the interpolated band structure agrees very well with high-density kmesh $ab$-initio calculation [20]. While the band structure of the coarse $ab$-initio kmesh, which is used for the construction of MLWFs, deviates largely to the "true" dense kmesh band.
structure needed for MCAE to converge. The comparison of band structures is shown in Fig. 4.

2. Interpolation kmesh

The former paragraphs considered the convergence of MCAE with respect to ab-initio kmesh, which should be dense enough for constructing MLWFs. From now on, we fix the \( n_k^{\text{ab}} \) to 24. Upon acquiring of high-quality MLWFs, the next step for achieving converged MCAE is the Wannier interpolation. This is where the computational cost is significantly reduced.

In such circumstance, the \( \Delta \text{MCAE} \) is defined as

\[
\Delta \text{MCAE}(n_k^{\text{wan}}) = \text{MCAE}(n_k^{\text{wan}}) - \text{MCAE}_{\text{conv}},
\]

\[
\text{MCAE}_{\text{conv}} = \frac{1}{201} \sum_{n_k^{\text{wan}}=2500}^{2700} \text{MCAE}(n_k^{\text{wan}}), \tag{20}
\]

and this is shown in Fig. 4.

The inset of Fig. 4 shows the overview of the \( \Delta \text{MCAE}(n_k^{\text{wan}}) \) for \( n_k^{\text{wan}} \) from 100 to 800. Apparently, \( n_k^{\text{wan}} \) on the order of 100 is far from convergence. For \( n_k^{\text{wan}} \) around 2600, the MCAE converged under \( 4 \times 10^{-6} \text{eV} \).

3. Smearing contribution

Smearing is very important for the convergence of ab-initio calculations. Large smearing is beneficial for fast convergence, but this may induce a fictitious and negligible contribution to the total energy. Too small smearing may slow down the convergence significantly, and often one needs a much denser kmesh to converge. This problem can be largely alleviated because the Wannier interpolation is very "cheap"—it only involves diagonalizations of \( M \times M \) matrices. One could easily increase kmesh density and willfully reduce smearing width, or even do not resort to smearing.

The relation between the minimum number of interpolation kmesh \( n_k^{\text{wan}} \) and the smearing width \( \sigma \) is shown in the inset of Fig. 5 and the specific values are tabulated in Table I. When \( \sigma \) is in the range of 0.20 eV to 0.05 eV, only less than 300 \( n_k^{\text{wan}} \) is needed to converge the MCAE on the order of \( 2 \times 10^{-6} \text{eV} \). However, as indicated by the blue line in the inset of Fig. 5, the smearing contribution to the MCAE, defined as

\[
\Delta \text{MCAE}(\sigma; n_k^{\text{wan}}) = \text{MCAE}(\sigma; n_k^{\text{wan}}) - \text{MCAE}_{\text{conv}},
\]

\[
\text{MCAE}_{\text{conv}} = \frac{1}{10} \sum_{\sigma \in \Sigma} \text{MCAE}(\sigma; n_k^{\text{wan}}),
\]

where \( \Sigma = \{0.0001 \times i | 0 \leq i \leq 9\} \),

\[
\tag{21}
\]

is on the order of \( 2 \times 10^{-3} \text{eV} \), leading to unreliable MCAE. The \( n_k^{\text{wan}} \) is Eqn. (21) are fixed to 2600, and the 0.0000 in \( \Sigma \) means calculation without smearing.

We find a smearing width of less than 0.003 eV is able to reduce the smearing contribution to under
the minimum number of Wannier interpolation kmesh \( n_{kmesh}^{\text{Wann}} \) needed for calculations with each smearing width \( \sigma \).

### TABLE I. The MCAE values and the Wannier interpolation kmesh needed for convergence with respect to smearing width \( \sigma \). The data in this table are plotted in the inset of Fig. 5.

| \( \sigma \) (eV) | MCAE \( (\times 10^{-5} \text{ eV}) \) | \( n_{kmesh}^{\text{Wann}} \) |
|----------------|-----------------|-----------------|
| 0.2000         | -367.862        | 72              |
| 0.1700         | -429.854        | 81              |
| 0.1500         | -469.843        | 93              |
| 0.1300         | -503.960        | 113             |
| 0.1000         | -514.434        | 158             |
| 0.0700         | -385.764        | 225             |
| 0.0500         | -212.176        | 319             |
| 0.0200         | -211.546        | 725             |
| 0.0030         | -235.876        | 1575            |
| 0.0012         | -235.821        | 2670            |
| 0.0000         | -235.739        | 2598            |

* Calculated without smearing

2 \( \times 10^{-6} \) eV, and at such a minimal smearing width a kmesh of nearly 1600 should be adopted. To totally exclude the smearing contribution (i.e., calculation without smearing), a dense kmesh of 2600 should be used [data listed in Table II].

### IV. CONCLUSIONS

We combine the Wannier interpolation of eigenvalues and the force theorem of magnetocrystalline anisotropy energy together to develop an efficient and accurate method for calculating MCAE other than brute-force \( ab\text{-}initio \) calculation. First, coarse kmesh \( ab\text{-}initio \) calculations are performed for different magnetization directions. Second, maximally localized Wannier functions are constructed based on the overlap matrices and projection matrices obtained from the first step. Third, MCAE is

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**FIG. 5.** MCAE convergence relative to smearing width \( \sigma \), defined by Equ. (21). The inset is the overview for \( \sigma \) from 0.20 eV to 0.00 eV. The pink line in the inset is the minimum number of Wannier interpolation kmesh \( n_{kmesh}^{\text{Wann}} \) needed for calculations with each smearing width \( \sigma \).

**TABLE II. Computational resources spent by direct dense kmesh \( ab\text{-}initio \) calculation and by Wannier interpolation method.** The cost is defined by Equ. (22).

| Calculations           | CPU cores | Time\(^a\) | Cost\(^b\) |
|------------------------|-----------|------------|------------|
| direct \( ab\text{-}initio \), kmesh 2600 | 96        | 9.0 h      | 864        |
| Wannier interpolation  |            |            |            |
| a. coarse \( ab\text{-}initio \), kmesh 24 | 96        | 5.8 min    |            |
| b. projections \& overlaps | 96        | 3.8 min    | 18         |
| c. disentanglement \& Wannierization | 1          | 2.5 h      |            |
| d. interpolation, kmesh 2600 | 1          | 0\(^c\)     |            |

\(^a\) Wall time. The time is the sum of calculations of two magnetization directions.

\(^b\) Unit: hour-core. The cost is the sum of all the steps.

\(^c\) Less than a second, negligible.
calculated based on force theorem and Wannier interpolation of eigenvalues on a dense kmesh. This Wannier interpolation method serves as a post-processing step for economically calculating MCAE. The ultimate accuracy of the calculated MCAE is determined by the underlying \textit{ab-initio} code (the choice of exchange-correlation functionals, type of basis sets, etc.), since it is the \textit{ab-initio} code that produces the Bloch functions from which the MLWFs are constructed. Nevertheless, the Wannier interpolation is independent from the choice of the underlying code, since the only quantities needed from the code are the overlap and projection matrices which can be obtained merely from Bloch functions.

We take an iron chain as an example and demonstrate that our Wannier interpolation approach for MCAE achieves a 48 times speedup compared to the direct dense kmesh \textit{ab-initio} calculation. In summary, to get the most accurate result, no smearing should be adopted and the coarse kmesh \textit{ab-initio} calculation should be fully tested. The convergence relative to the Wannier interpolation kmesh can be easily achieved.

This Wannier interpolation method reduces the computational cost significantly and maintains high accuracy simultaneously. Besides, it makes it possible for MCAE calculations which are hard to converge or feasible due to the computational cost, as well as calculations requiring exceptional accuracy such as high order effect of MCAE.

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