

# SCDM-k Wannierization from AO basis sets

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**Abstract**—We present a workflow to construct localized Wannier bases from large-scale electronic-structure calculations performed in non-orthogonal atomic-orbital (AO) representations. The approach leverages the SCDM-k formalism, avoiding iterative localization. We apply it to two MoS<sub>2</sub>/oxide interfaces with up to 880 atoms. The Wannier basis enables band interpolation and material-projected densities of states on dense k-meshes, and compact real-space Hamiltonians suitable for efficient NEGF transport calculations.

**Index Terms**—DFT, interfaces, wannierization, NEGF.

## I. INTRODUCTION

First-principles electronic-structure calculations of interface or disordered materials often require large supercells, e.g., 2D semiconductors on amorphous oxides. In such settings, direct Brillouin-zone sampling and post-processing of Kohn–Sham eigenstates become prohibitively expensive. Localized Wannier functions (WF) provide a compact representation that enables interpolation of observables on dense k-meshes, as well as downstream modeling, such as quantum transport. The maximally localized WF (MLWF) framework is powerful but involves iterative spread minimization and careful initialization, which can become fragile for large, entangled manifolds [1]. In contrast, Selected Columns of the Density Matrix (SCDM) and its k-point generalization SCDM-k construct localized orbitals without iterative optimization, providing gauge-independent and robust automation well suited to large and disordered systems [1]–[3].

Here we combine SCDM for non-orthogonal AO bases [3] with the k-point generalization of SCDM-k [2], to construct Wannier bases directly from AO Hamiltonian and overlap matrices, scaling to large unit cells. We demonstrate it on two MoS<sub>2</sub>/oxide stacks from [4]: monolayer (ML) MoS<sub>2</sub> with sapphire ( $\alpha$ -Al<sub>2</sub>O<sub>3</sub>) (Fig. 1a) and ML MoS<sub>2</sub> with amorphous (am) Al<sub>2</sub>O<sub>3</sub> (Fig. 1b) with 768 and 880 atoms, respectively.

## II. ALGORITHM

Starting from AO Hamiltonian  $H(R)$  and overlap matrices  $S(R)$ , the workflow constructs a smooth Bloch subspace using energy-weighted SCDM-k projections and transforms it into a localized real-space Wannier Hamiltonian  $h(R)$ . The workflow is summarized in Table I. We follow the SCDM-k algorithm from [2] operating on the quantities of SCDM for AO bases from [3] with minor modifications. For brevity, we omit the k-index in the following paragraph. The procedure consists of seven steps: (1) solve the generalized eigenvalue problem

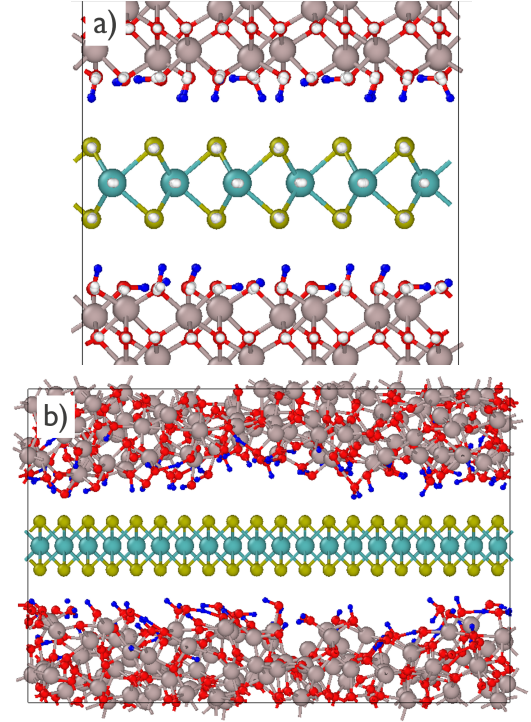


Fig. 1. Atomic configurations of the MoS<sub>2</sub>/ $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (a) and MoS<sub>2</sub>/am-Al<sub>2</sub>O<sub>3</sub> (b) interfaces. Yellow, cyan, red, gray, and blue spheres represent S, Mo, O, Al, and H. The white spheres visualize the Wannier centers of the MoS<sub>2</sub>/ $\alpha$ -Al<sub>2</sub>O<sub>3</sub> interface. The unit cells contain 768 and 880 atoms and measure  $1.88 \times 1.63 \times 2.22$  nm and  $3.14 \times 1.63 \times 2.04$  nm, respectively.

(GEVP), (2) select parameters  $\mu$  and  $\sigma$  for the complementary error function for the energy weight function  $w(E)$  (Fig. 2) [2] and the number of WF  $N_W$ , (3) solve QR with column pivoting (QRCP) at a reference k-point  $k_{\text{ref}}$  on the weighted eigenvectors  $w\Psi'S$  and select the first  $N_W$  pivot indices  $J$ , (4) compute S-orthonormal projector  $\phi$ , (5) orthonormalize the projector:  $\Phi$ , (6) project the Hamiltonian onto the subspace, and (7) Fourier transform to real space. The resulting Wannier Hamiltonian  $h(R)$  can be used for, e.g., quantum transport or interpolation.

Unlike [3] we perform QRCP on the fat matrix  $\Psi_k'W_kS_k$  (see Table I) instead of the density matrix for performance reasons, same as in SCDM-k [1], [2]. Furthermore, we use a modified expression for the transformation matrix (see Table I)

using the Moore–Penrose inverse. Empirically, we find that it improves the Wannier interpolation accuracy for the entangled manifolds.

The Wannier centers and spreads are estimated directly in the AO representation using a Mulliken-style population analysis, where the position operator  $\mathbf{r}$  is approximated by the center position of each AO basis function. The center of WF  $n$  is obtained from its first moment,  $\mathbf{r}_n = \langle \mathbf{r} \rangle_n$ , and the spread from the second central moment,  $\Omega_n = \langle r^2 \rangle_n - |\mathbf{r}_n|^2$ . The moments are evaluated from the full real-space Wannier representation, including contributions from all lattice vectors  $\mathbf{R}$  retained in the Wannier Hamiltonian. The resulting spread therefore yields a lower bound to the true Wannier spread because the spatial extent of the AO basis functions is neglected.

SCDM-k depends on a few parameters, namely the energy window characterized by the weight function  $w(E)$ ,  $N_W$ , and  $k_{\text{ref}}$ . We follow [1], [2] and use the complementary error function, characterized by  $\mu$  and  $\sigma$ , for entangled bands and step functions elsewhere as illustrated in Fig. 2. We select  $N_W$  from the weights  $w_k$  (see Table I) as the number of weights larger than 0.01:  $N_W = \max_k(\text{count}(w_k > 0.01))$ . We find this criterion to provide a robust and fully automatic choice of  $N_W$  to obtain low interpolation errors. Lastly, similar to [2] we find that  $k_{\text{ref}}$  has little impact, particularly for the large unit cells we aim to transform. Hence we select the k-point closest to the center of the Brillouin zone:  $k_{\text{ref}} = \min_k(|k|)$ .

TABLE I

SUMMARY OF THE SCDM-K ALGORITHM.  $W_k$  IS A DIAGONAL MATRIX WITH THE WEIGHTS  $w_k$  ON THE DIAGONAL. SQUARE BRACKETS INDICATE PYTHON-STYLE MATRIX SUBSCRIPTS, THE  $'$  SYMBOL THE HERMITIAN TRANSPOSED, AND  $^+$  THE MOORE–PENROSE INVERSE, AND  $^{-1/2}$  THE INVERSE MATRIX SQUARE ROOT. STEPS 1-2, 4-6 ARE PERFORMED AT EACH K-POINT AND ARE EMBARRASSINGLY PARALLEL. FOR MEMORY SAVINGS, 1-6 CAN BE PERFORMED SEQUENTIALLY FOR EACH K-POINT, STARTING WITH  $K_{\text{REF}}$ .

- 1) solve  $H_k \Psi_k = S_k \Psi_k E_k$ , where  $H_k = \sum e^{-ikR} H_R$  and  $S_k = \sum e^{-ikR} S_R$
- 2) compute the energy weights  $w_k = 0.5 \times \text{erfc}((E_k - \mu)/\sigma)$
- 3) solve  $((\Psi_k W_k)' S_k) P = QR$  at a reference point  $k = k_{\text{ref}}$ ,  $J = P[:N_W]$
- 4) compute Wannier transformation  $\phi_k = \Psi_k W_k \chi_k^+$ ,  $\chi_k = (S_k \Psi_k W_k)[:J]$
- 5) orthonormalize Wannier transformation:  $\Phi_k = \phi_k (\phi_k' S_k \phi_k)^{-1/2}$
- 6) transform the Hamiltonian  $h_k = \Phi_k' H_k \Phi_k$
- 7) Fourier transform of the Wannier Hamiltonian:  $h_R = \sum e^{ikR} h_k$

### III. RESULTS

The electronic structure of the two interfaces is computed with CP2K [5]. We used the PBEsol functional together with the DFT-D3(BJ) dispersion correction with Becke–Johnson damping [6], [7]. Furthermore, we employed DZVP MOLOPT basis sets and GTH pseudopotentials [8]. Due to the large unit cell only  $\Gamma$ -point sampling was used. For SCDM-k we used a  $4 \times 4 \times 1$  Monkhorst-Pack k-point mesh. Timings for SCF and SCDM-k are given in Table II. The dominant cost of SCDM-k is solving the GEVP of the AO matrices, making it comparable to a single SCF iteration on the same k-mesh (see Table II). The maximum (average) interpolation errors on 10 points between  $\Gamma$ -X for  $\text{MoS}_2/\alpha\text{-Al}_2\text{O}_3$  and  $\text{MoS}_2/\text{am-Al}_2\text{O}_3$

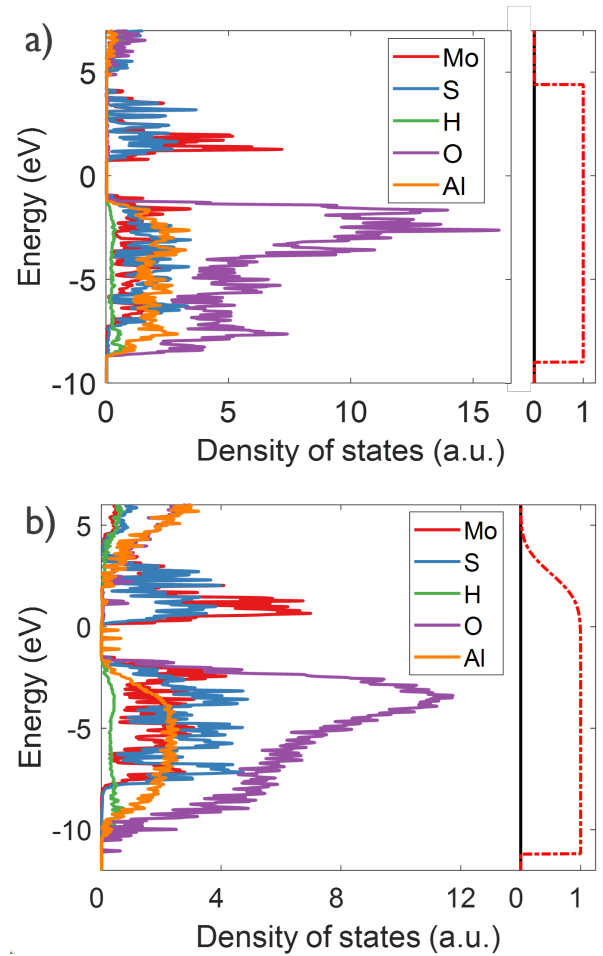


Fig. 2. Left panel: PDOS of the (a)  $\text{MoS}_2/\alpha\text{-Al}_2\text{O}_3$  and (b)  $\text{MoS}_2/\text{am-Al}_2\text{O}_3$  interface. The right panels show the energy weight function  $w(E)$  used in the “disentanglement via entanglement” paradigm of SCDM-k [2]. With  $\text{am-Al}_2\text{O}_3$  the upper edge of the window of interest cuts through the oxide tail states. For disentanglement with  $w(E)$  a complementary error function with  $\mu = E_F + 2.63$  eV, and  $\sigma = 1.5$  eV is used. The other window edges are step functions.

are 0.88 (0.059) meV and 2.49 (0.064) meV, respectively, within 2 eV of the band edges. These low errors confirm that the Wannier interpolation faithfully reproduces the coarse mesh electronic structure at a fraction of the cost. In addition, the standard Wannier interpolation paradigm can be leveraged to efficiently obtain denser Brillouin-zone quantities: High-resolution material-projected DOS can be obtained at low computational cost using Wannier interpolation, as shown in Fig. 3.

The resulting WF are spatially localized and chemically interpretable. For both interfaces, the procedure yields 3, 3, and 5 WF localized on each O, S, and Mo atom, respectively, consistent with O/S p-like and Mo d-like valence manifolds. This amounts to a total of 1404 and 1758 WF, respectively. In the latter case, 113 additional WF are introduced, resulting in a final basis size of 1871 WF. No WF are obtained for Al and H atoms with  $\alpha\text{-Al}_2\text{O}_3$  as all oxide conduction states were

TABLE II  
TIME PER K-POINT (S) OF 0) SCF IN CP2K AND THE DOMINANT SCDM-K STEPS (TABLE I), IMPLEMENTED IN MATLAB, FOR  $\text{MoS}_2/\alpha\text{-Al}_2\text{O}_3$  ON 4 CPUs. STEPS 1, 4-7 SUM UP TO 116 S PER K-POINT. STEP 3 ONLY NEEDS TO BE PERFORMED ONCE.

|                               |         |
|-------------------------------|---------|
| 0) SCF iteration:             | 123.7 s |
| 1) GEVP:                      | 93.0 s  |
| 3) QRCP:                      | 9.6 s   |
| 4) Computing $\phi_k$ :       | 5.2 s   |
| 5) Compute $\Phi_k$ :         | 8.0 s   |
| 6) Project $H(R)$ :           | 9.3 s   |
| 7) Fourier transform $h(R)$ : | 0.5 s   |

excluded from the energy window. The Wannier centers for  $\text{MoS}_2/\alpha\text{-Al}_2\text{O}_3$  are visualized in Fig. 1. For  $\text{am-Al}_2\text{O}_3$ , the 113 additional WF appear from the oxide conduction tail states as well as high energy  $\text{MoS}_2$  states that are included due to  $w(E)$ . These reflect the increased electronic complexity arising from the overlap between  $\text{MoS}_2$  and  $\text{am-Al}_2\text{O}_3$  conduction states (see Fig. 2).

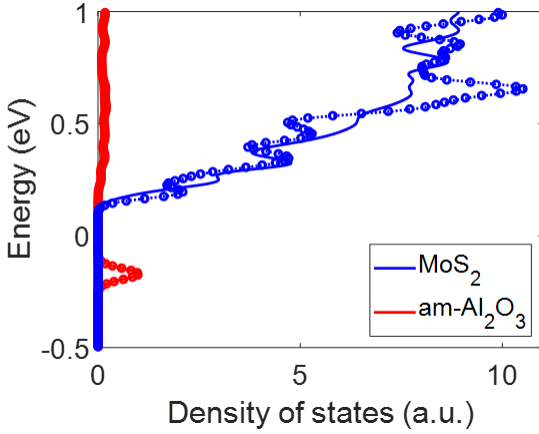


Fig. 3. Density of states projected onto  $\text{MoS}_2$  and  $\text{am-Al}_2\text{O}_3$  around the conduction band minimum of the  $\text{MoS}_2$ . The dashed line indicates the density from WF at  $\Gamma$ , the blue circles the density from DFT in the AO basis. The solid line shows the density from Wannier interpolation on a  $10 \times 10 \times 1$  Monkhorst-Pack grid. Interpolation smooths the density in the  $\text{MoS}_2$ , while the oxide state has no dispersion and does not change with interpolation.

The computed Wannier centers are found close to the associated atomic sites, as shown in Fig. 4a. The maximum, median, and mean distances to the nearest atom are 0.16 Å, 0.07 Å, and 0.07 Å for  $\text{MoS}_2/\alpha\text{-Al}_2\text{O}_3$  and 1.56 Å, 0.08 Å, and 0.1 Å for  $\text{MoS}_2/\text{am-Al}_2\text{O}_3$ . With the amorphous oxide the distribution is heavily skewed due to the artificial states introduced by the "disentanglement via entanglement" [2]. Analogously, the WF are well localized as shown in Fig. 4b. The maximum, median, and mean spreads are 0.91 Å<sup>2</sup>, 0.33 Å<sup>2</sup>, and 0.37 Å<sup>2</sup> for the  $\text{MoS}_2/\alpha\text{-Al}_2\text{O}_3$  and 21.56 Å<sup>2</sup>, 0.46 Å<sup>2</sup>, and 1.13 Å<sup>2</sup> for the  $\text{MoS}_2/\text{am-Al}_2\text{O}_3$ . The large-spread WF occur predominantly among the additional functions introduced by the disentanglement procedure. These WF are associated with the gradual weighting ( $w(E)$ ) of high-energy oxide and  $\text{MoS}_2$  states at the upper boundary of the target energy window as illustrated in

Fig. 2. Their larger extent is therefore expected and does not affect the accuracy of the low-energy manifold targeted by the Wannierization. Together, the estimates of centers and spreads support the success of the SCDM-k procedure and provide a simple measure of WF localization and chemical character.

Finally, we use  $h(R)$  for self-consistent dissipative mod-  
espace NEGF-Poisson simulations, performed with ATOMOS [9], [10] (see Fig. 5) to obtain current-voltage (I-V) characteristics for both stacks and validate against AO. The good agreement between the AO and WF I-V characteristics (Fig. 6) demonstrates that the Wannier basis preserves the transport physics. We further observe an approximately threefold reduced current at  $V_{\text{GS}} = 0.7$  V and an increase in subthreshold slope (SS) from 60 to 70 mV/dec for  $\text{am-Al}_2\text{O}_3$  (Fig. 6). For these interfaces, the Wannier bases reduce the NEGF simulation cost by factors of 3.7-4.3, and peak memory usage by up to an order of magnitude (see Table III).

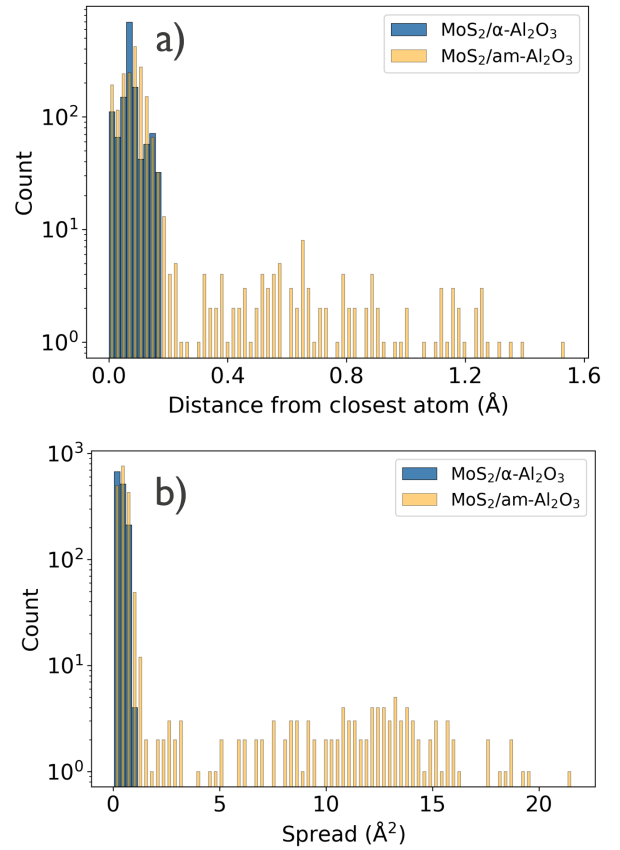


Fig. 4. (a) Distances of the Wannier centers to the nearest atom and (b) spreads of the Wannier functions computed for the two interface structures. Note the log scale to show the tails resulting from the disentanglement in the  $\text{MoS}_2/\text{am-Al}_2\text{O}_3$  interface.

#### IV. CONCLUSION

We demonstrate a SCDM-k Wannierization workflow for AO calculations that scales to thousand-atom heterostructures, yielding meV interpolation errors, 3.7–4.3× faster NEGF

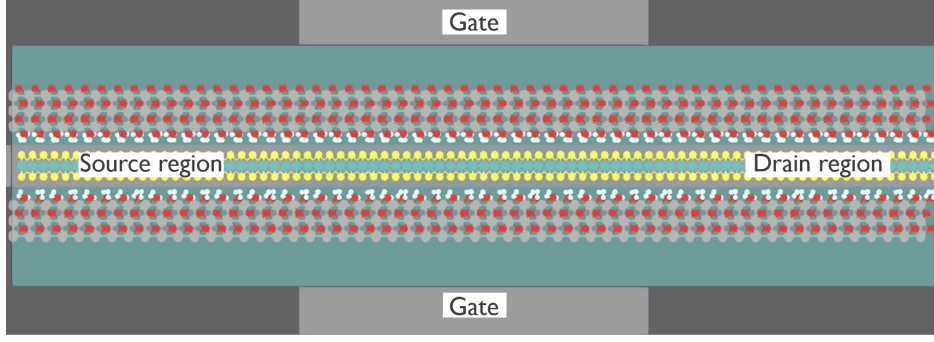


Fig. 5. (a) Schematic device layout (not to scale) for the NEGF simulations. The devices are built from periodically repeated unit cells of the two MoS<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> interfaces in both AO and Wannier bases. Source and drain lengths and doping are 25.7 nm and  $1.85 \times 10^{13} \text{ cm}^{-2}$ , respectively. The channel length is 14.4 nm, and the equivalent oxide thickness (EOT) is 0.5 nm. Electron-phonon scattering is included within the self-consistent Born approximation as described in [9], [10].

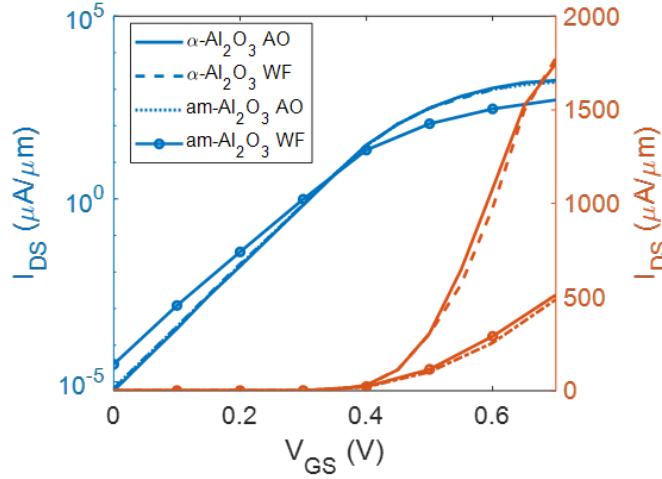


Fig. 6. NEGF I-V curves of the interfaces. The solid (dashed) lines are for  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> in the AO reference (Wannier) basis. The dotted (dash-dotted) lines are for am-Al<sub>2</sub>O<sub>3</sub> in the AO (Wannier) basis. The amorphous oxide causes a  $\sim 3\times$  reduction of the current at 0.7 V  $V_{GS}$  and an increase of the subthreshold slope (SS) from 60 mV/dec to 70 mV/dec.

TABLE III  
COMPARISON OF BASIS SIZES, TIMINGS, AND MEMORY REQUIREMENT OF NEGF SIMULATIONS FOR THE TWO INTERFACES IN THE AO AND WANNIER BASES.

| Basis type             | AO       |       | WF       |      |
|------------------------|----------|-------|----------|------|
| Oxide                  | $\alpha$ | am    | $\alpha$ | am   |
| Basis size per cell    | 9216     | 11148 | 1404     | 1871 |
| Mode space size        | 137      | 187   | 117      | 173  |
| Time per E-k point (s) | 2.96     | 6.22  | 0.81     | 1.44 |
| Peak memory (GB)       | 5.60     | 6.51  | 0.51     | 1.00 |

simulations, and up to  $10\times$  lower memory consumption. It enables (1) the standard Wannier interpolation paradigm for dense-k properties and (2) compact real-space Hamiltonians of realistic device stacks for NEGF transport. By avoiding iterative localization, the method is suited for large, complex, entangled manifolds typical of disordered or interface con-

figurations. The method therefore provides a practical route from large-scale AO DFT calculations to compact transport-ready Hamiltonians. The resulting basis also enables further applications such as downfolding models, linear-response calculations, as well as multiscale transport workflows.

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