基于多目标差分演化算法进行逆向材料设计

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上半场

- 高通量计算及逆向材料设计的介绍

下半场

- 逆向材料设计软件包介绍

上半场内容提要

0 背景

1 算法

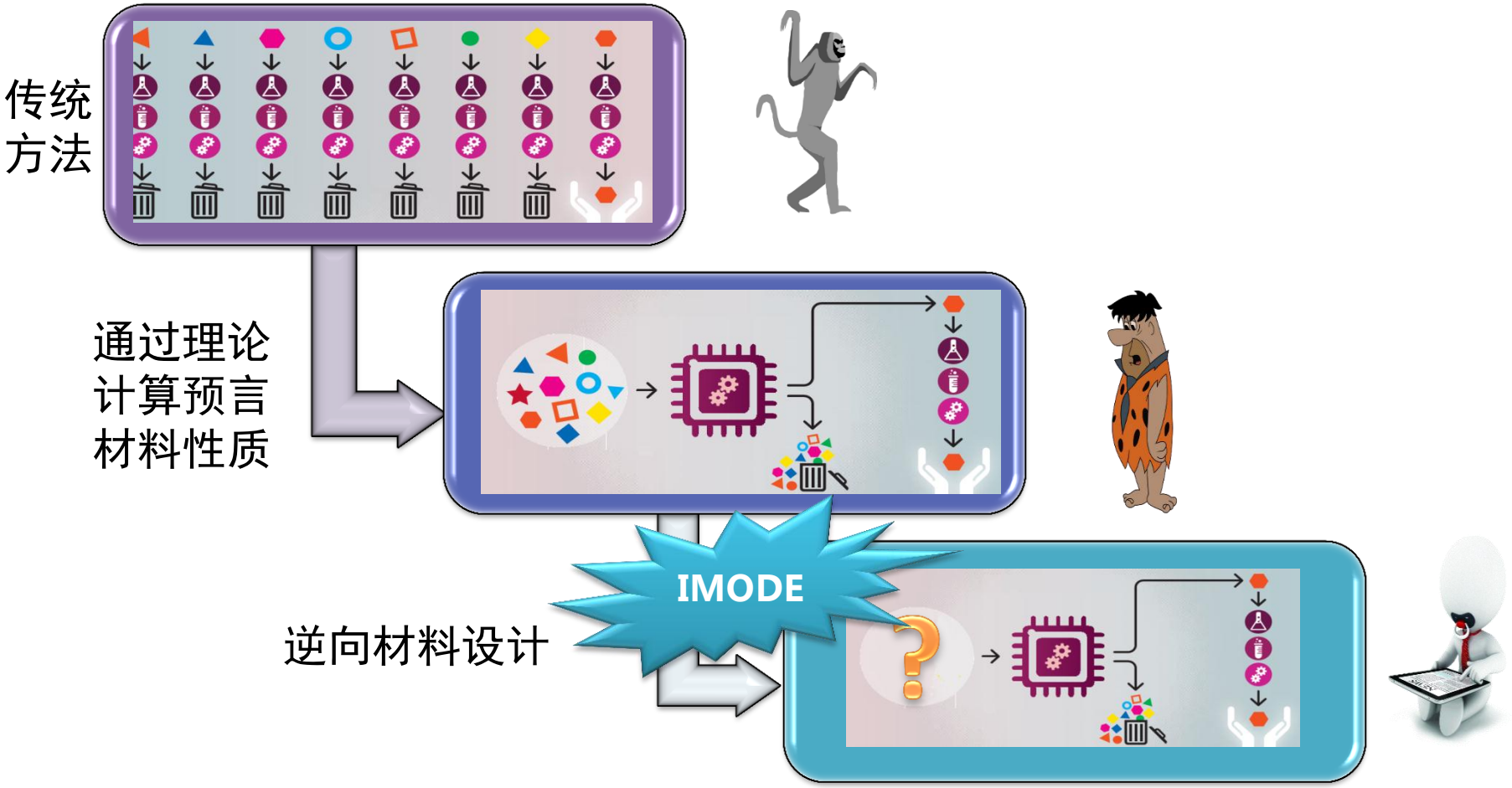
2 功能

3 技术特点

4 应用案例

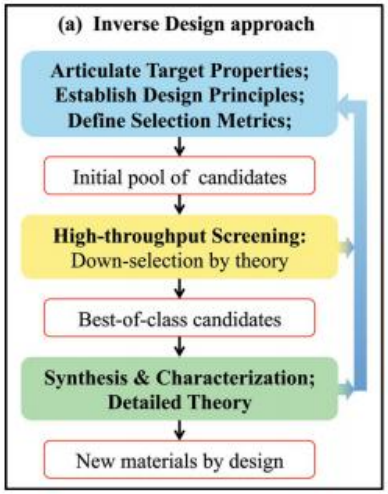
5 成果

背景：什么是逆向材料设计



随着超级计算机以及第一性原理计算方法的发展，如果一种物质的结构明确了，那么我们可以通过计算的手段预言它的一系列性质。但是，这一过程的逆向过程，根据性质预言一种物质的组成结构，仍然是一种巨大的挑战。

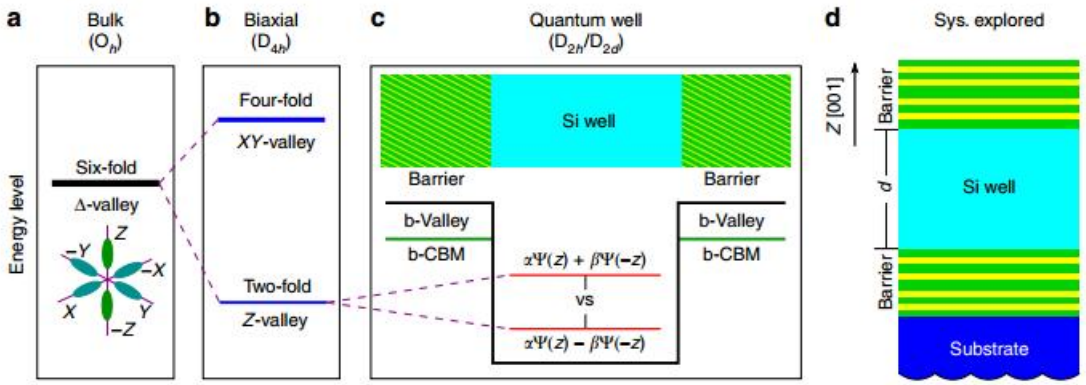
逆向材料设计思想在计算上的实现



(b) The search for *p*-type ternary Mn-oxides

Materials	Mn_2TiO_4	$Mn_2V_2O_7$	Ba_2MnO_3	Mn_2SiO_4	Mn_2GeO_4	Al_2MnO_4	Ga_2MnO_4	In_2MnO_4	$MnTiO_3$	$MnGeO_3$	$MnSnO_3$	Mn_2SnO_4	Cr_2MnO_4
Structure	ISp	*	*	Ol	Ol	Sp	Sp	Sp	Pyr	Ilm	Ilm	ISp	Sp
Stability	✗	✓	✓	✓	✓	○	✓	○	✓	✓	○	○	✓
Band gap		✗	✓	✓	✓	✓	✓	✓	✓	✓	✓	○	✓
m_h^*		✓	✗	✗	✗	✗	✗	✗	✗	✓	✓	✓	○
h.-killer										✗	✗	✓	✓
Polaron												✗	✓
h.-prod.													ext.

*For $Mn_2V_2O_7$ and Ba_2MnO_3, the ICSD collection codes are 24321 and 15508; ISp: Inverse Spinel; Ol: Olivine; Sp: Spinel; Pyr: Pyriophanite; Ilm: Ilmenite; h.-killer: hole killer; h.-prod.: hole-producer; ext.: extrinsic dopant

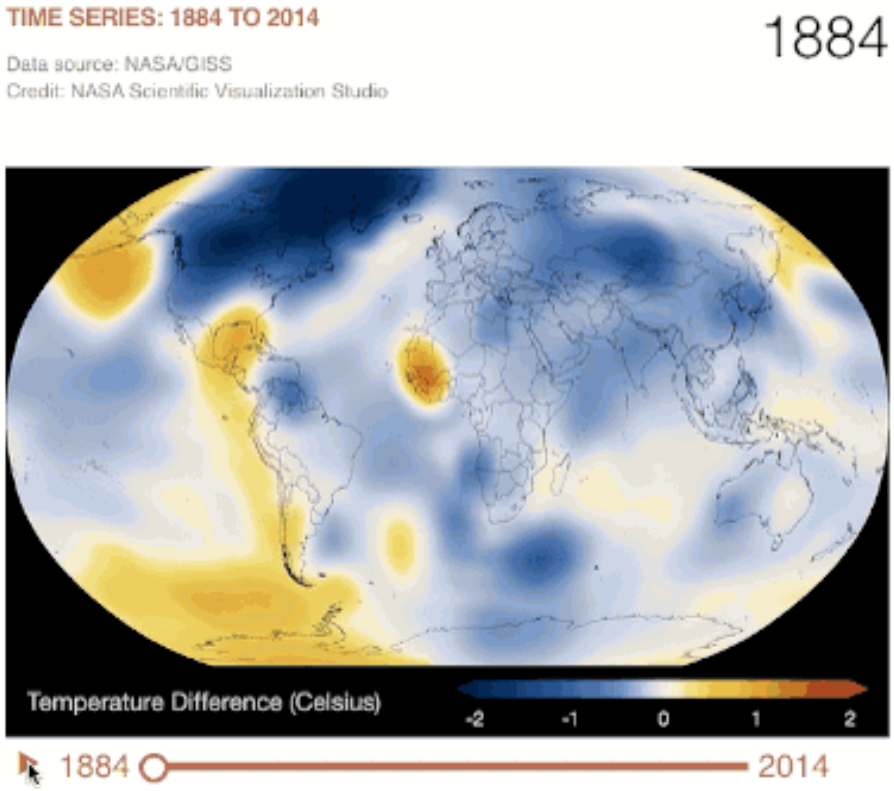


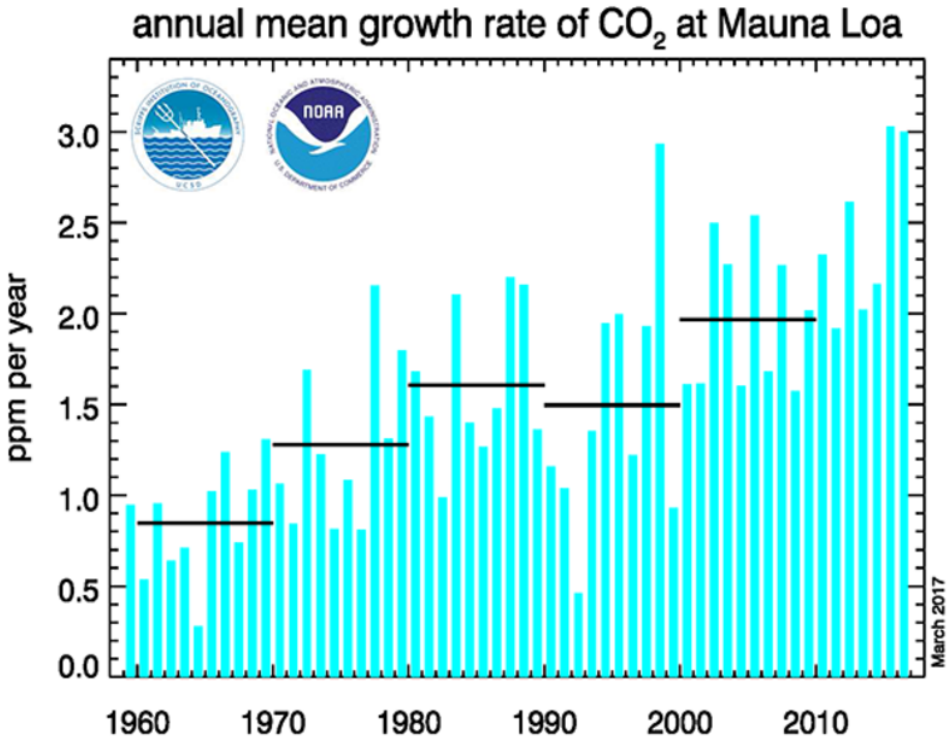
Zunger组首先提出了采用遗传算法对Si/Ge量子阱进行逆向材料设计，从而构建谷电子劈裂的体系。

L. Zhang, J.-W. Luo, A. Saraiva, B. Koiller, A. Zunger, Nat Commun, 4 (2013)2396.

背景：全球变暖

GLOBAL TEMPERATURE: 1884 to 2014
Data source: NASA/GISS
Credit: NASA Scientific Visualization Studio





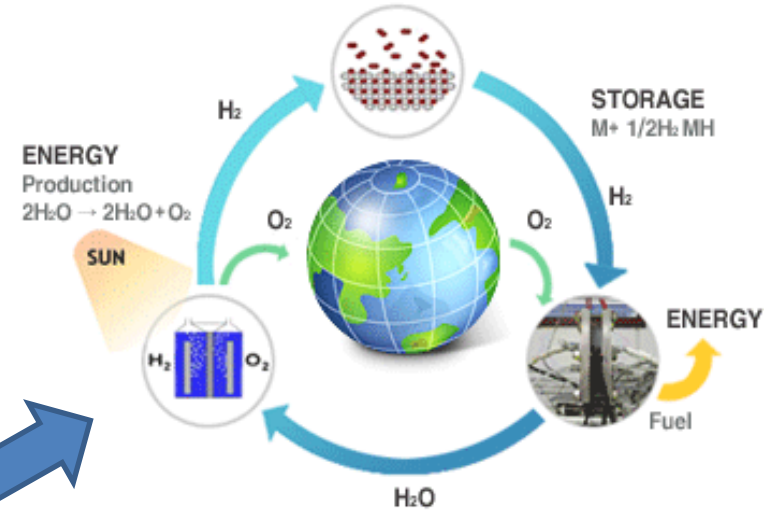
近一个世纪以来，全球温度急剧变暖，这与逐年上升的CO_2排放密不可分。

至2016年大气中CO2浓度已达405 ppm，远超350 ppm的安全上限。

背景：能源危机

化石能源引发的环境危机

寻找替代方案



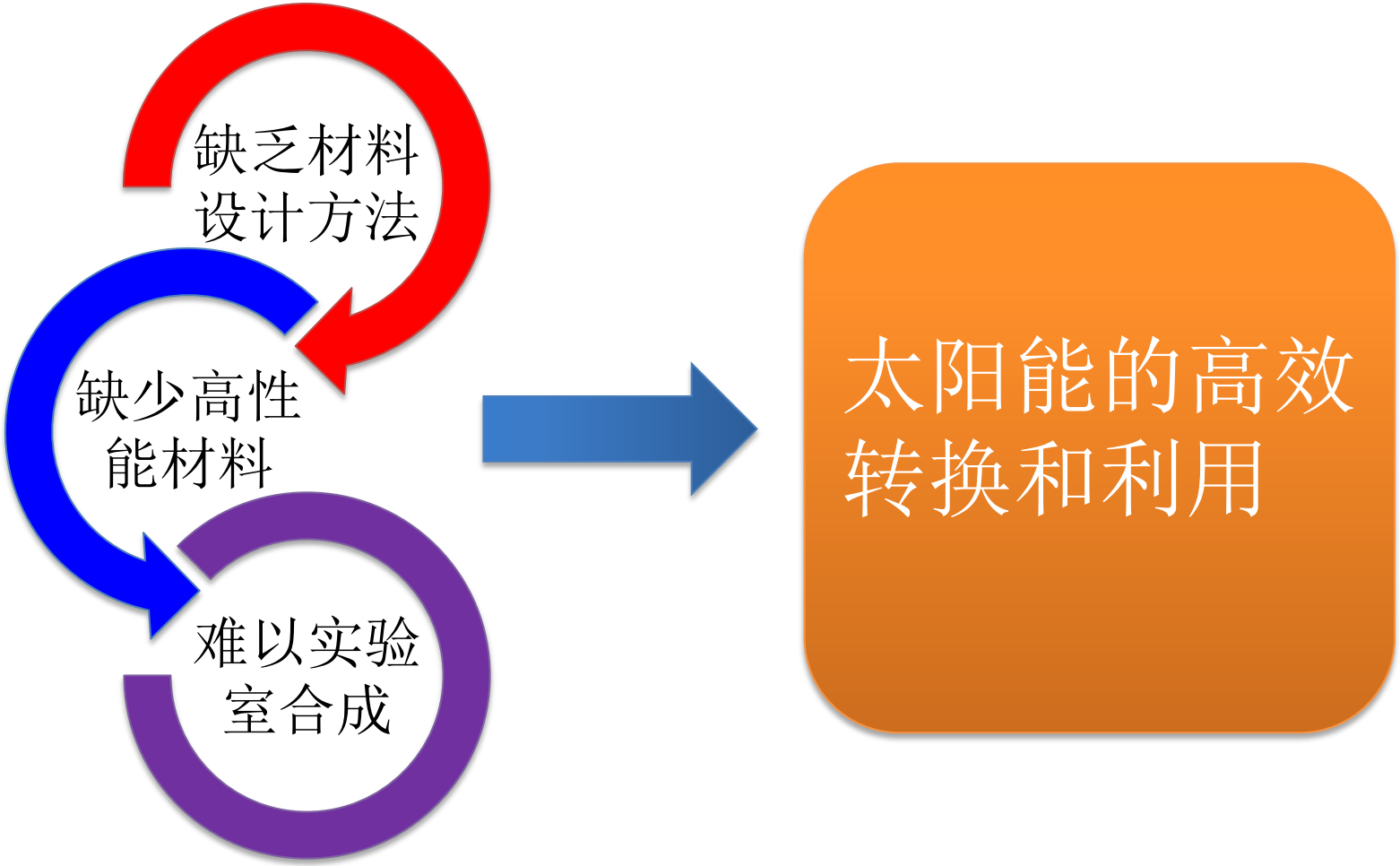
太阳能光解水

太阳能发电

化石能源的使用是CO_2过度排放的重要原因

理想替代：太阳能、氢能

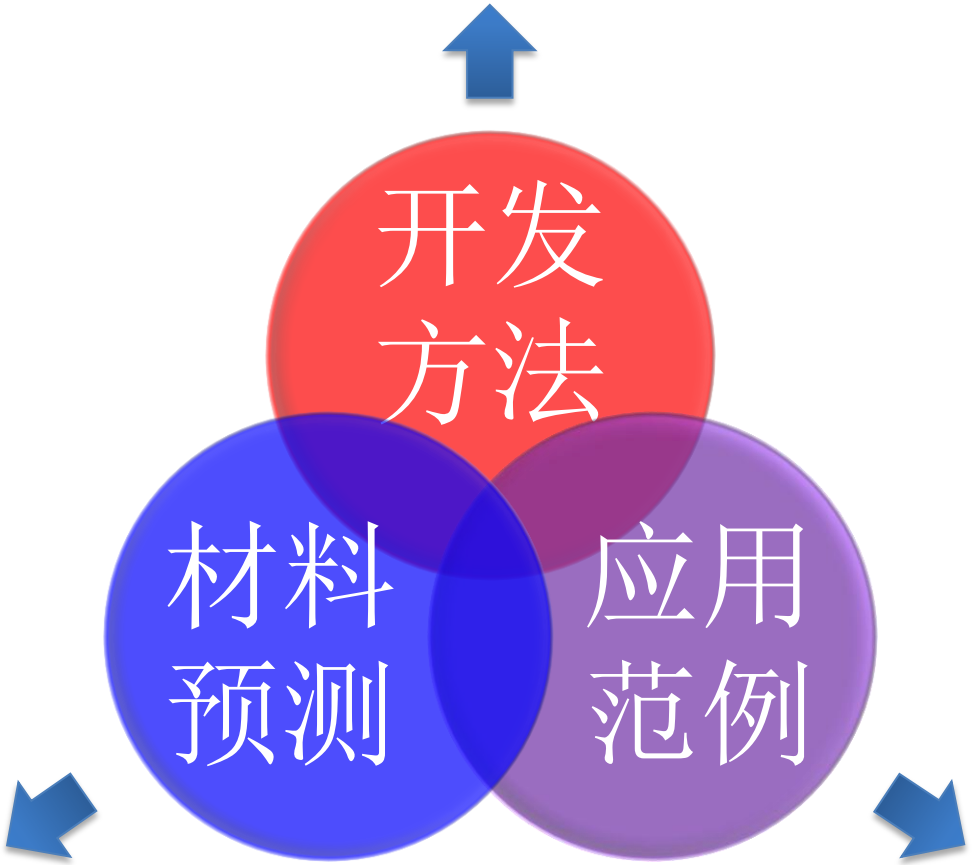
背景：太阳能材料的研发



长期以来新材料设计方法的缺乏使得我们难以系统地预言新型太阳能材料，预言能力的缺乏使得我们缺少对材料的合理设计，并最终影响到太阳能真正大规模的转换与利用。

一. 研究背景与思路

通过开发多目标全局优化算法的程序IM^{2}ODE，从而实现新型功能材料的设计



通过我们自己开发的方法，预言新型太阳能吸收材料

指导实验组，设计新型太阳能材料的组分和形貌

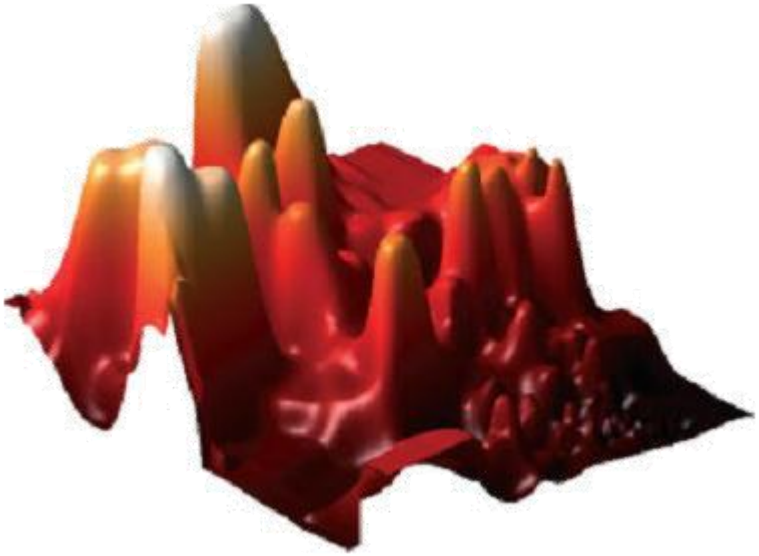
1算法

IM^{2}ODE

Inverse Design of Materials by Multi-Objective Differential Evolution

基于多目标差分演化算法进行逆向材料设计

逆向材料设计的三大主要步骤

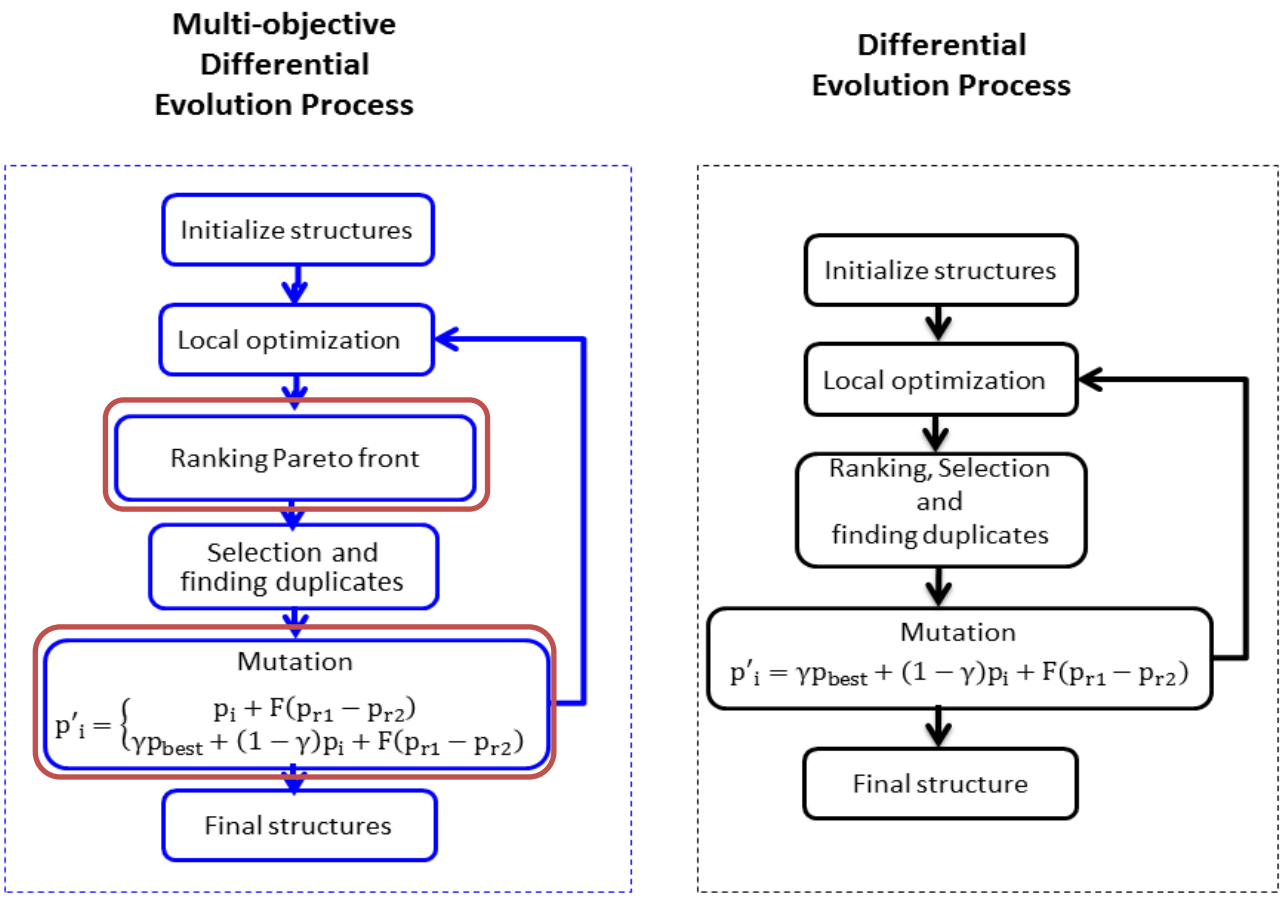


对材料的势能面进行重点采样，寻找具有特定性质的稳态或亚稳态材料

Zhang, Y.-Y.; Gao, W.; Chen, S.; Xiang, H.; Gong, X.-G. *Comput. Mater. Sci.* **2015**, 98, 51.

1算法

IM^{2}ODE程序包的算法流程图

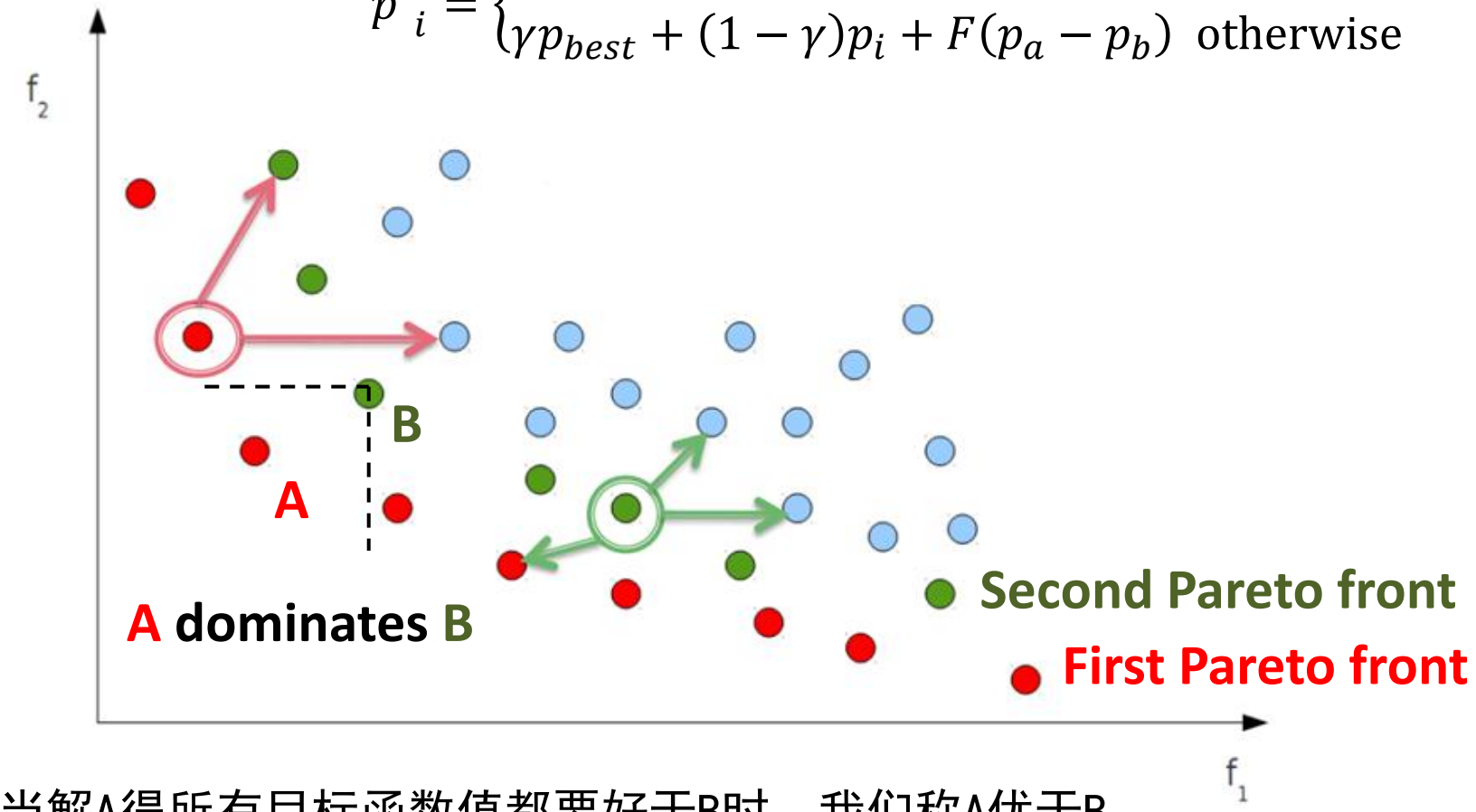


与传统的单目标差分演化算法相比，多目标的算法主要的改进在于排序和变异操作。

1算法

多目标差分算法的排序操作

$$p'_i = \begin{cases} p_i + F(p_a - p_b) & \text{if } p_i \text{ is non-dominated} \\ \gamma p_{best} + (1-\gamma)p_i + F(p_a - p_b) & \text{otherwise} \end{cases}$$

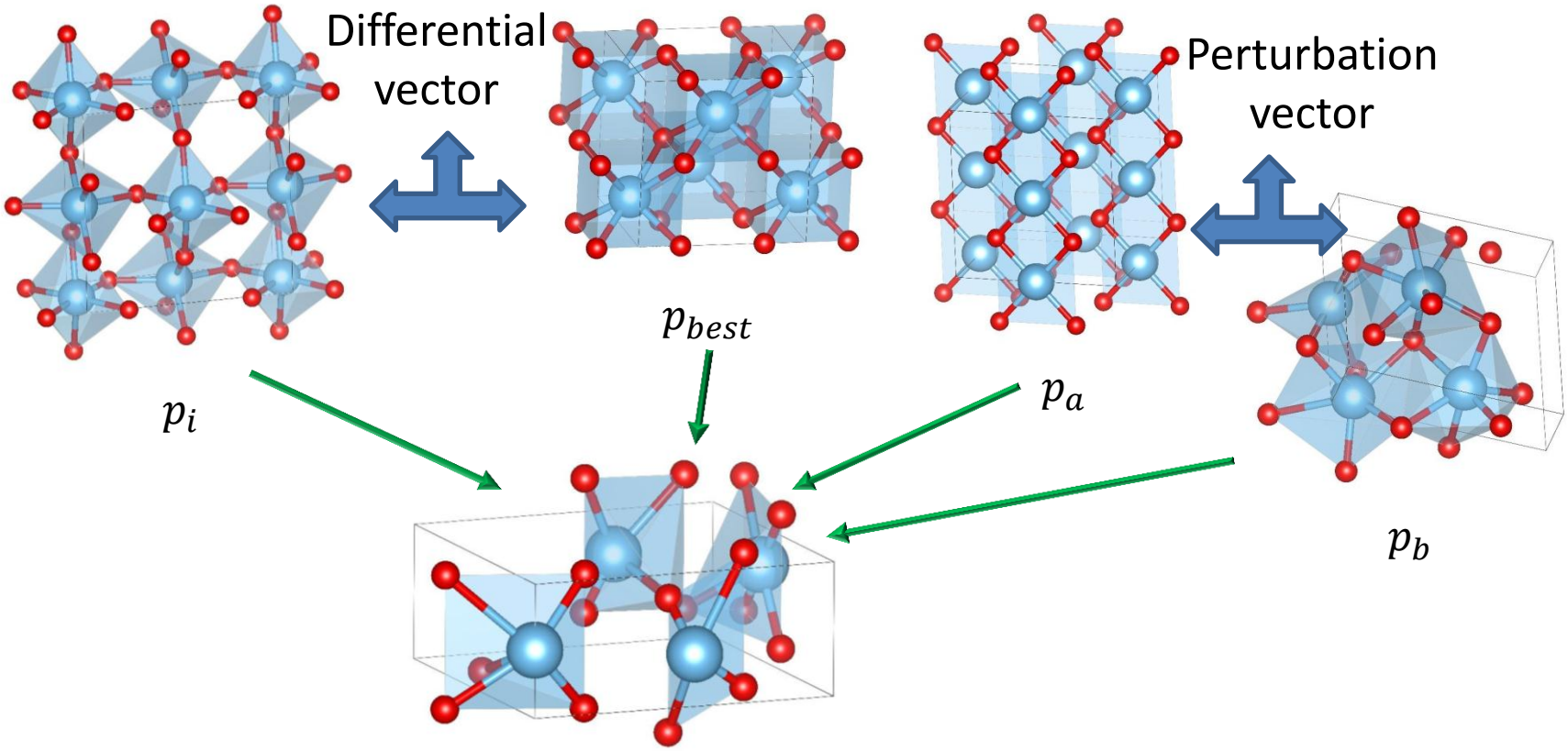


优于：当解A得所有目标函数值都要好于B时，我们称A优于B

Pareto最优解：当没有其他解“优于” A时，我们称A在Pareto最优解集中

1算法

多目标差分算法的变异操作

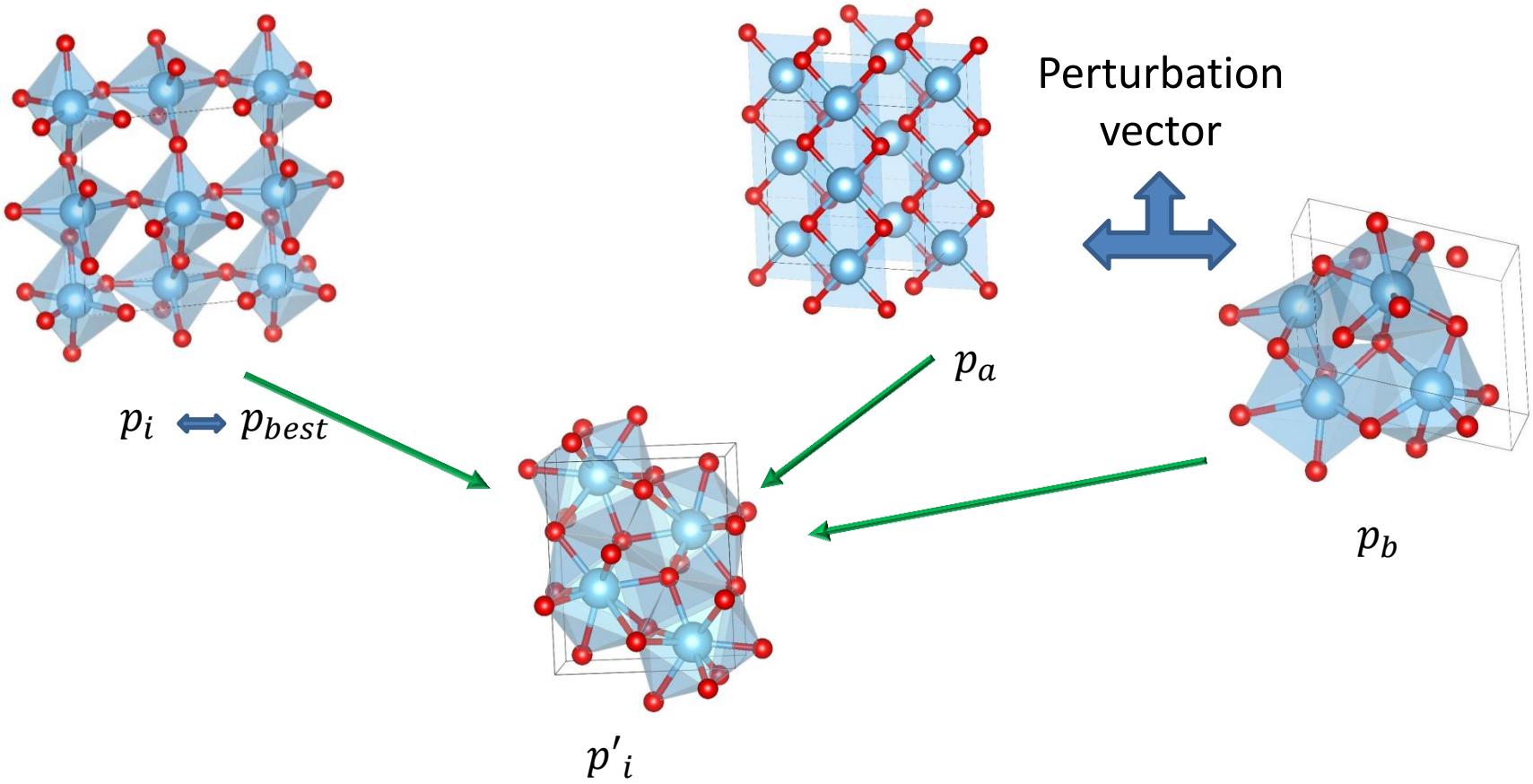


$$p'_i = \gamma p_{best} + (1-\gamma)p_i + F(p_a - p_b)$$

对于一个不在Pareto最优解集中的解，其差分矢量由学习项和微扰项构成

1算法

多目标差分算法的变异操作



$$p'_i = p_i + F(p_a - p_b)$$

对于一个已经在Pareto最优解集中的解，其差分矢量仅由微扰项构成

2功能

IM^{2}ODE软件包的功能

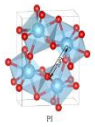
Property

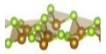
Structure

Electronic structure

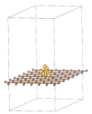
Optical absorption

Mechanical property

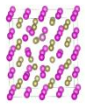

Crystal


2D


Cluster


Surface


Interface


Defect

3技术特点

- 程序采用模块化的结构，便于维护和作进一步的开发
- 在产生初始的块体结构时采用空间群优化，有效提高采样效率
- 采用种群并行的策略，可以进行上千核并行的高通量材料筛选，提高效率

4应用案例

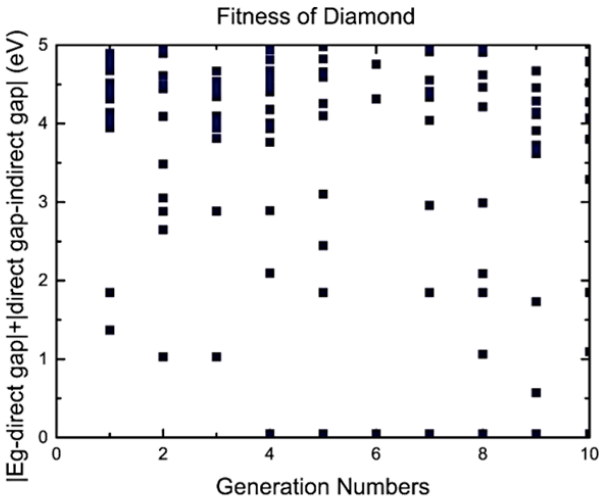
寻找具有特定带隙的材料

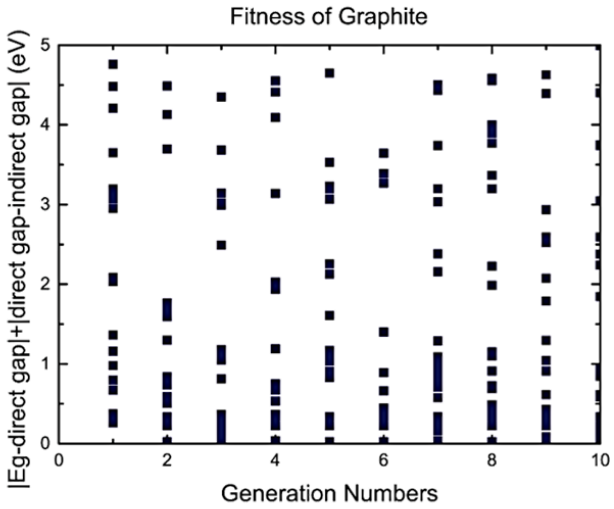
- design material with a target direct band gap E_g

$$\min z_1 = \text{total energy}$$

$$\min z_2 = \left|E_g - \text{direct gap}\right| + \left|\text{direct gap} - \text{indirect gap}\right|$$

System	E_g (eV)	Average generation
α-Al_2O_3	6.4	2
Diamond (Carbon)	4.1	3
Graphite (Carbon)	0.0	3

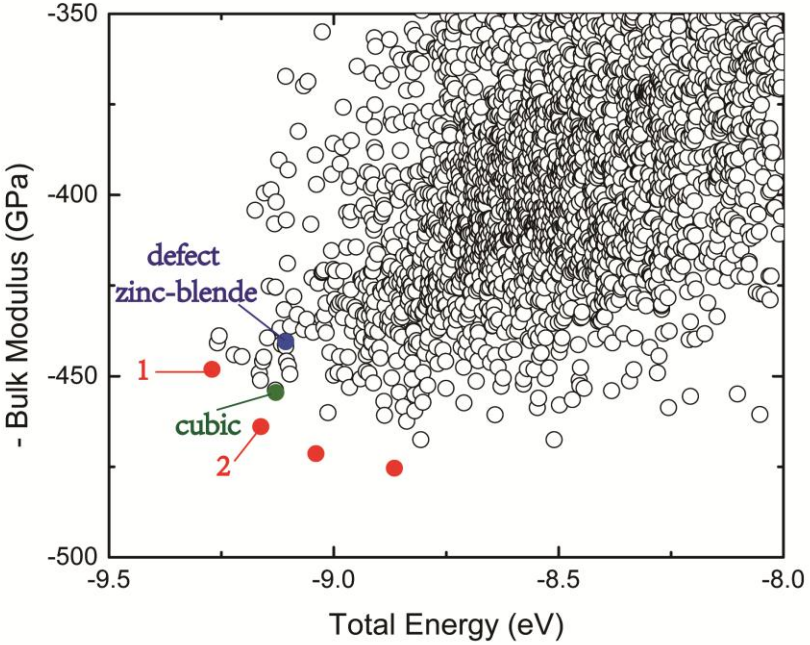




在种群数30的情况下，IM^{2}ODE能在2-3代内找到具有特定带隙的碳同素异形体和Al_2O_3的晶体结构

4应用案例

寻找超硬材料



- Tested system: C_3N_4
- objective functions:

$$\min z_1 = \text{total energy}$$

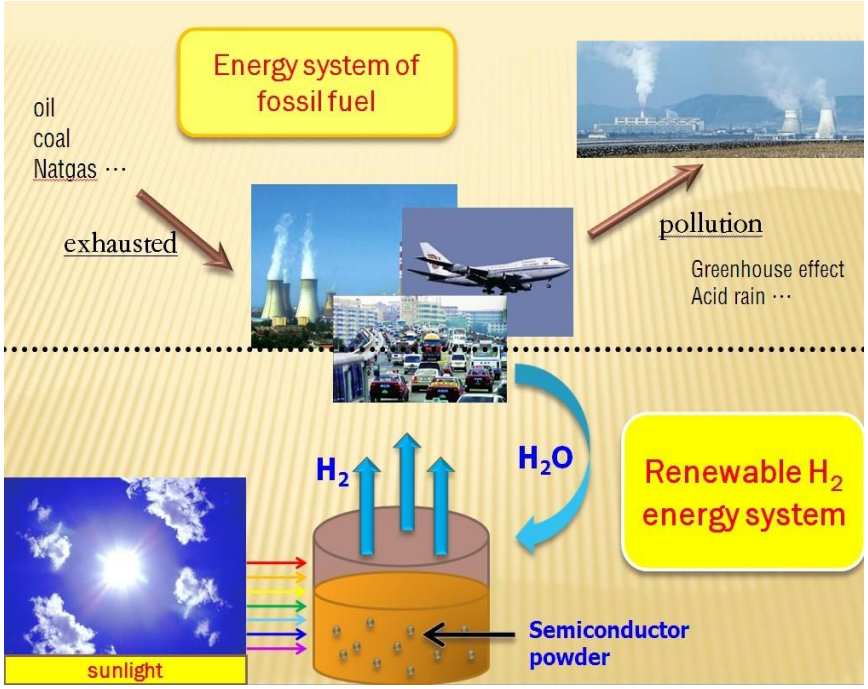
$$\min z_2 = -B = -\frac{N_c}{4}\frac{0.624 - 0.070I}{d^{3.5}}$$

	Total Energy / atom (eV / atom)	B(empirical equ)(GPa)	B(ab initio)(GPa)
Defect zinc-blende	0.0	440.557	414.718
cubic	-0.022	454.546	462.48
1	-0.164	448.134	30.859
2	-0.055	463.917	140.35

IM²ODE能够重复出经典的cubic和defect zinc-blende相的晶体结构

4应用案例

预言具有合适带隙的二氧化钛材料



Titanium Oxide (TiO_2):

✓ great potential in PEC water splitting

✓ low cost, nontoxic …

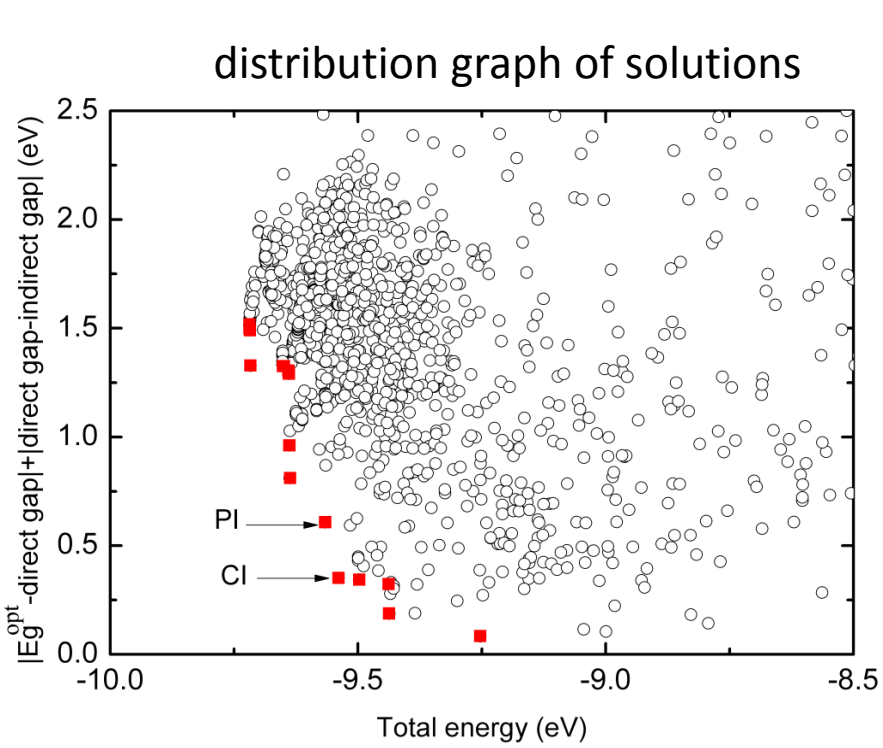
✓strong catalytic activity, high chemical stability

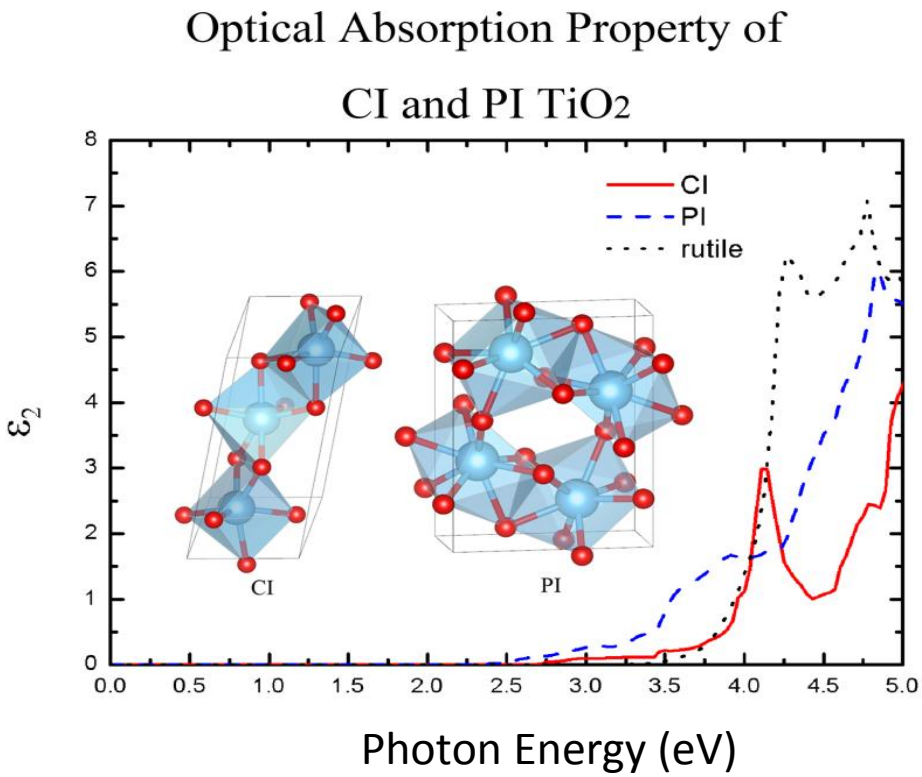
- Large intrinsic band, absorbs only UV

➢ A dopant-free TiO_2 phase with a suitable band gap is highly desirable.

4应用案例

预言具有合适带隙的二氧化钛材料





IM²ODE能够设计出有着更强可见光吸收的TiO_2相

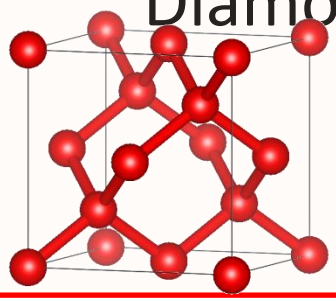
HZ Chen, YY Zhang, X Gong, H Xiang, *J. Phys. Chem. C*, **2014**, *118* (5)

4应用案例：纯碳材料的设计

Carbon: a Versatile Element

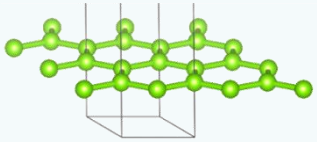
Carbon Allotropes

Diamond



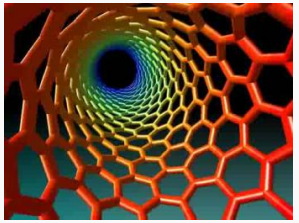
Wide band gap

Graphene



Zero band gap

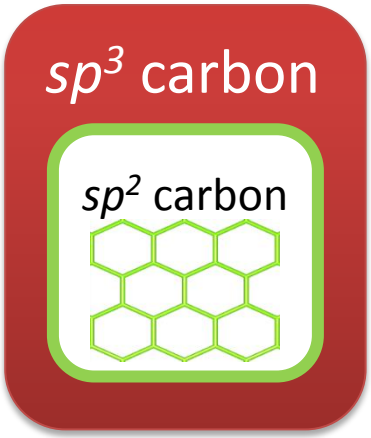
Carbon Nanotube



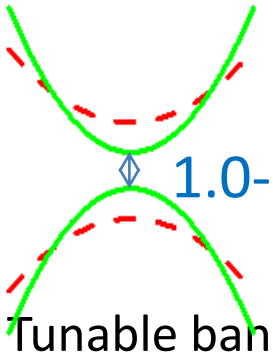
Zero band gap

None of them is suitable to be used as solar cell

4应用案例：纯碳材料的设计



Hybrid sp^2 & sp^3

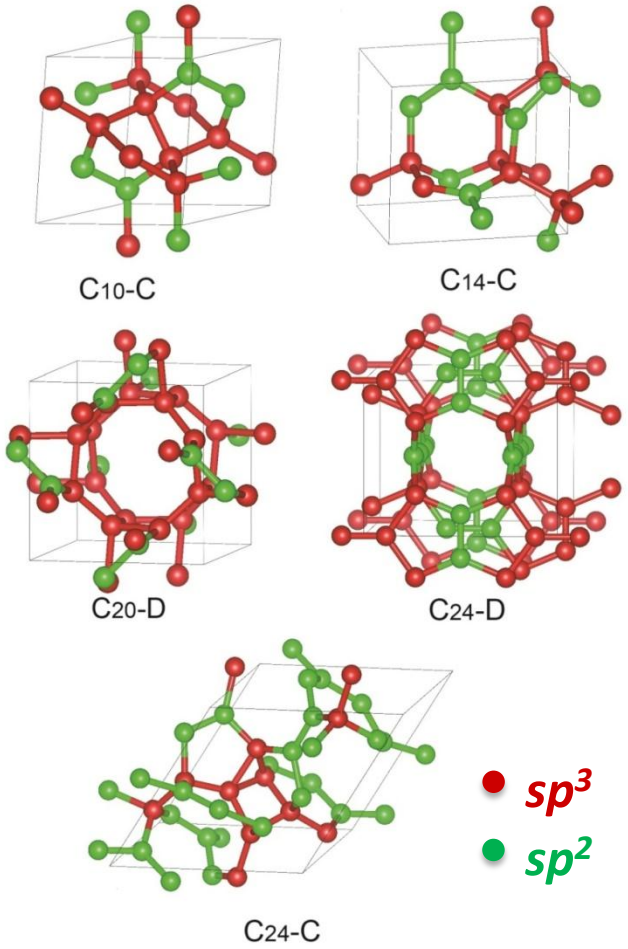


1.0-1.5 eV

Tunable band gap

IM²ODE

Inverse Design

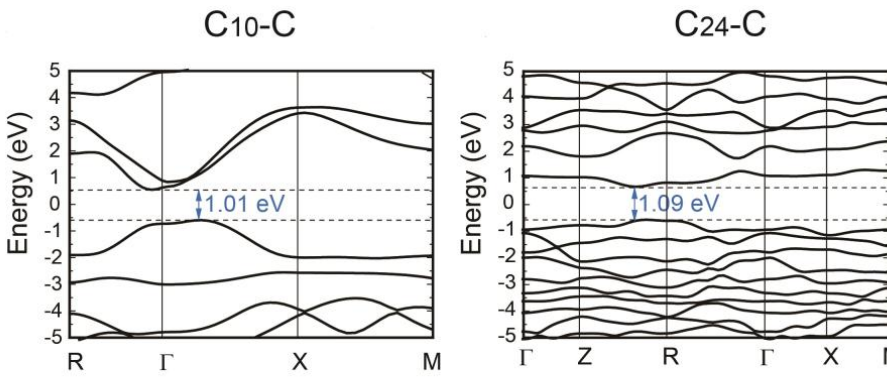


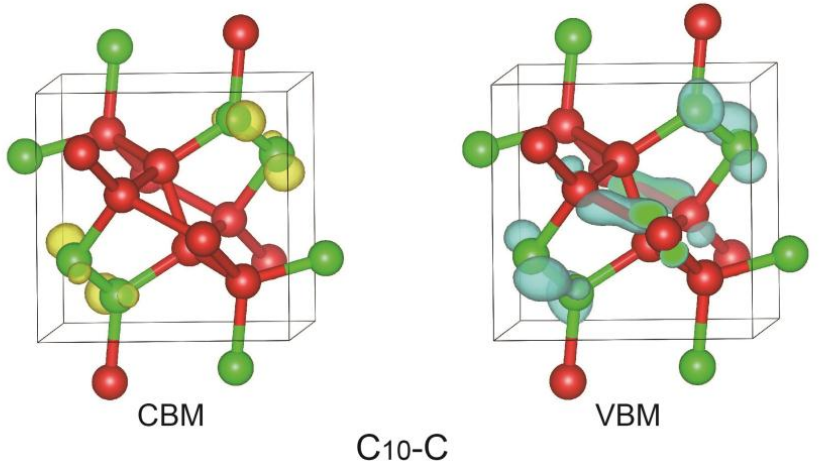
通过IM²ODE软件包的搜索，我们成功地预言了5种有混合sp^2-sp^3 杂化的碳同素异形体

Zhang, Y.-Y.; Chen, S.; Xiang, H.; Gong, X.-G. *Carbon*, 2016, 109, 246-252

4应用案例：纯碳材料的设计

电子结构性质



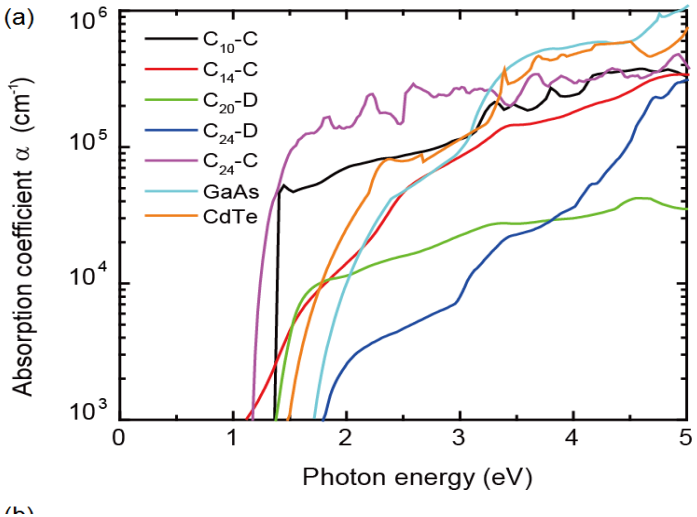


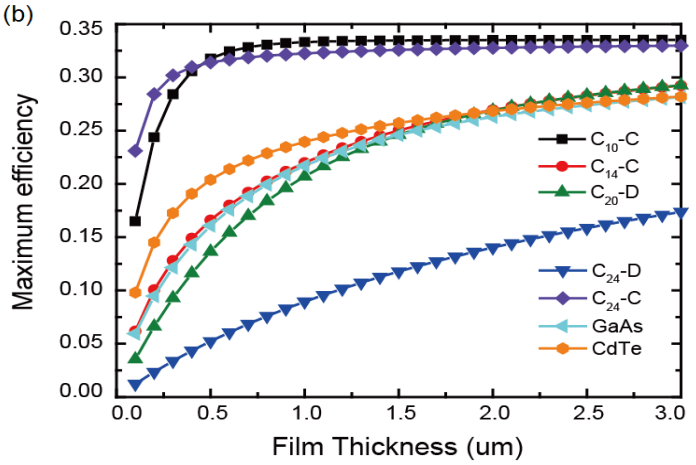
Band gap between 1.0 – 1.5 eV

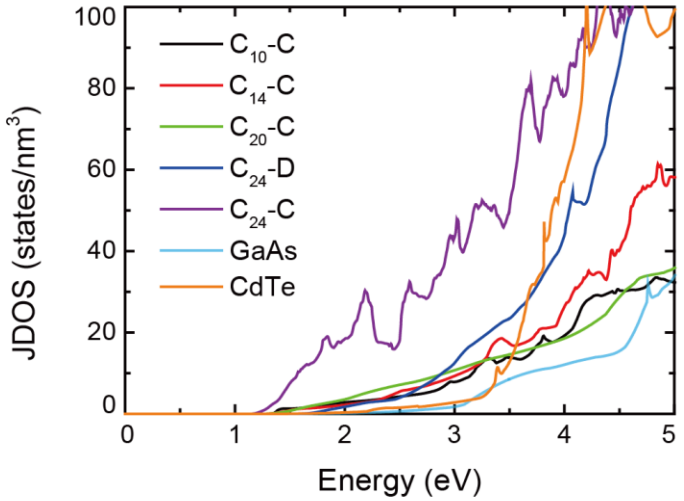
Optical dipole transition between the π and π* states is allowed

4应用案例：纯碳材料的设计

光学性质



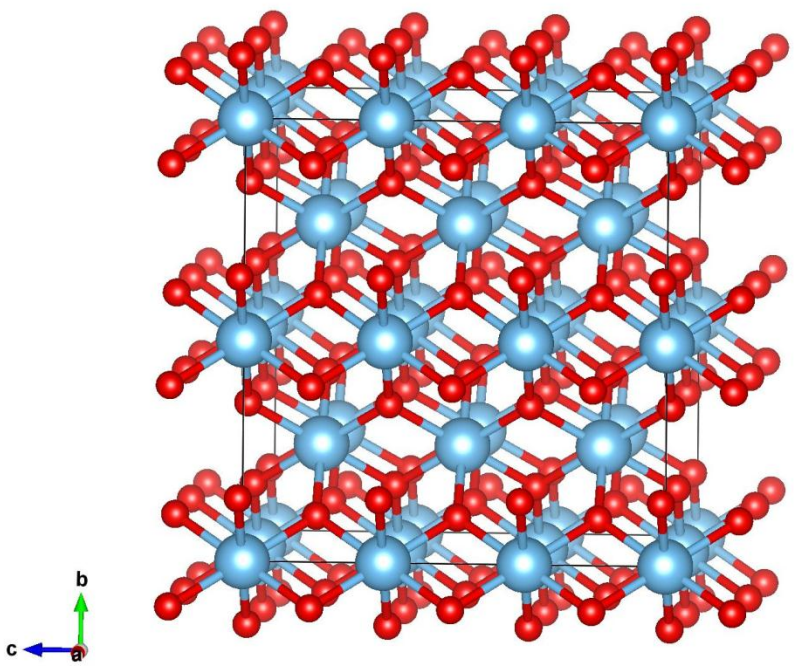




C10-C和C24-C在3 μm厚时，预测的转化效率可以高达33.5%和32.9%

4应用案例：多样的体系

体系1：块体材料测试案例



System: TiO2(rutile)
Ti: 16, O: 32
Optimized by GULP
Structures generated according to space group

IM^{2}ODE
1000 structure generated
50 rutile
Hit rate: 5%

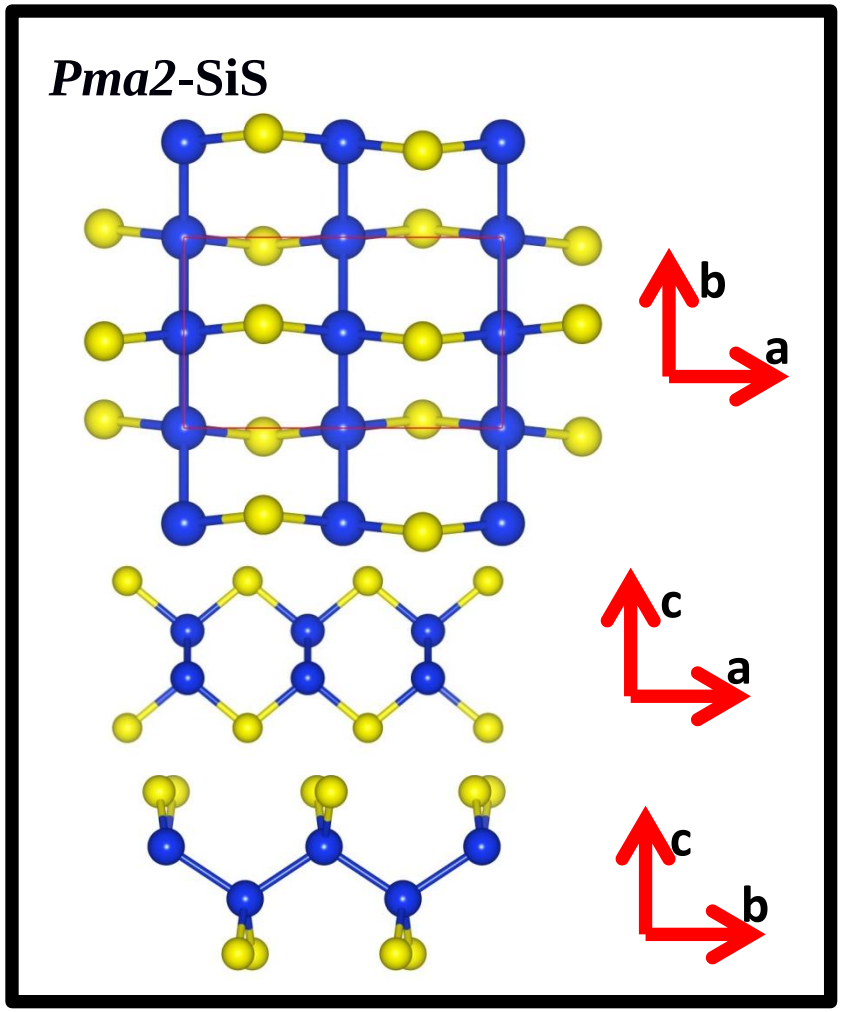
CALYPSO
3250 structure generated
203 rutile
Hit rate: 6.2%

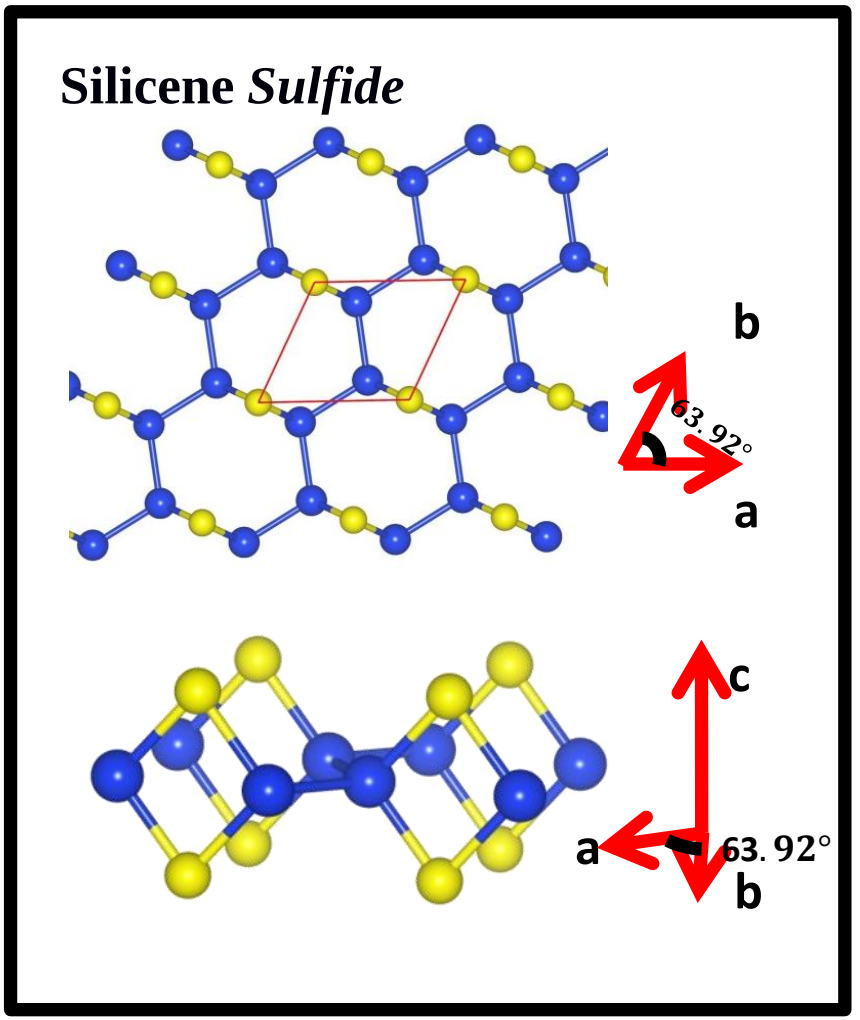
在初始结构采用空间群优化的情况下，IM^{2}ODE的百次命中率与CALYPSO相仿

Wang et al, Computer Physics Communications, Volume 183, Issue 10, October 2012

4应用案例

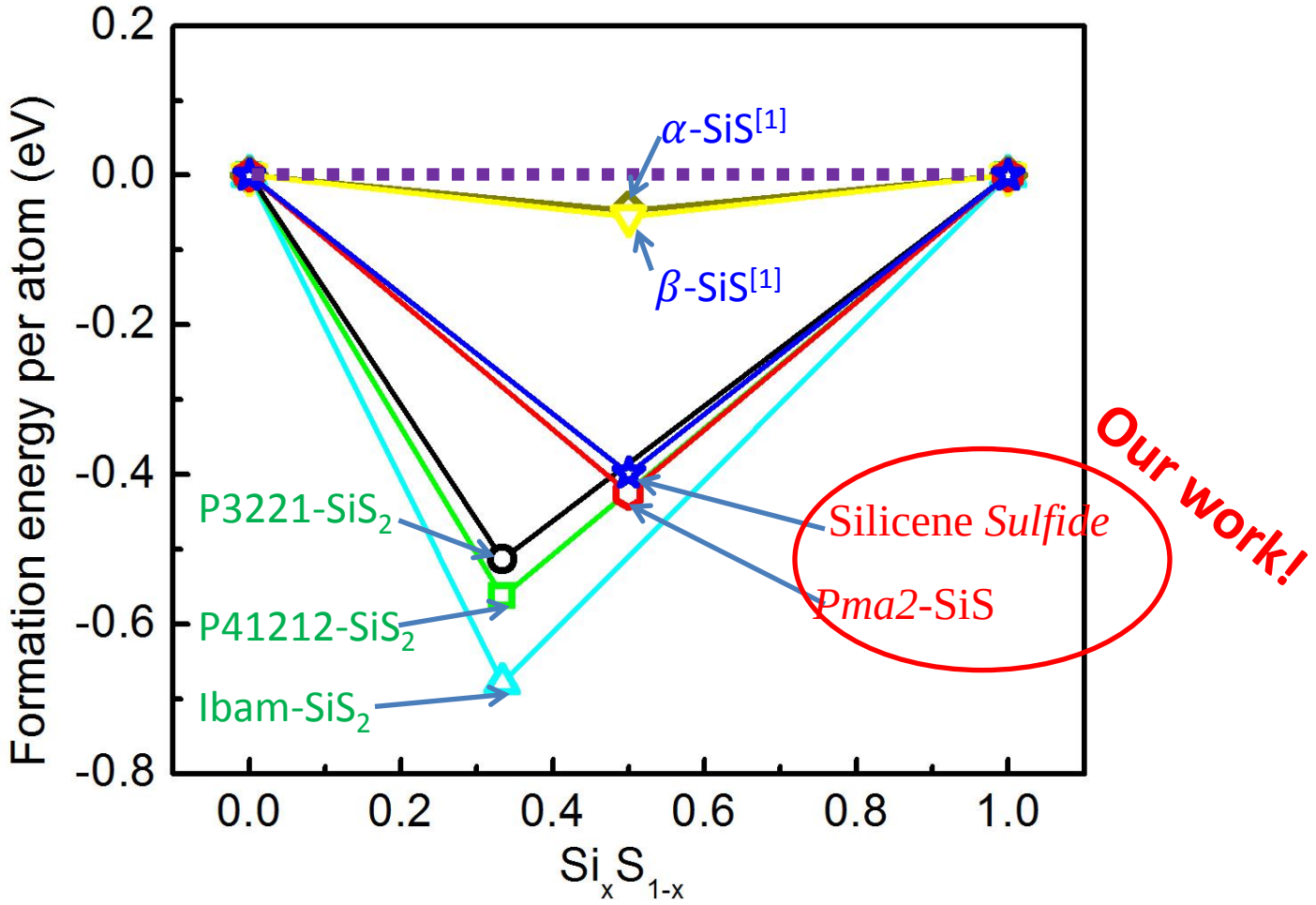
体系2：二维材料测试案例





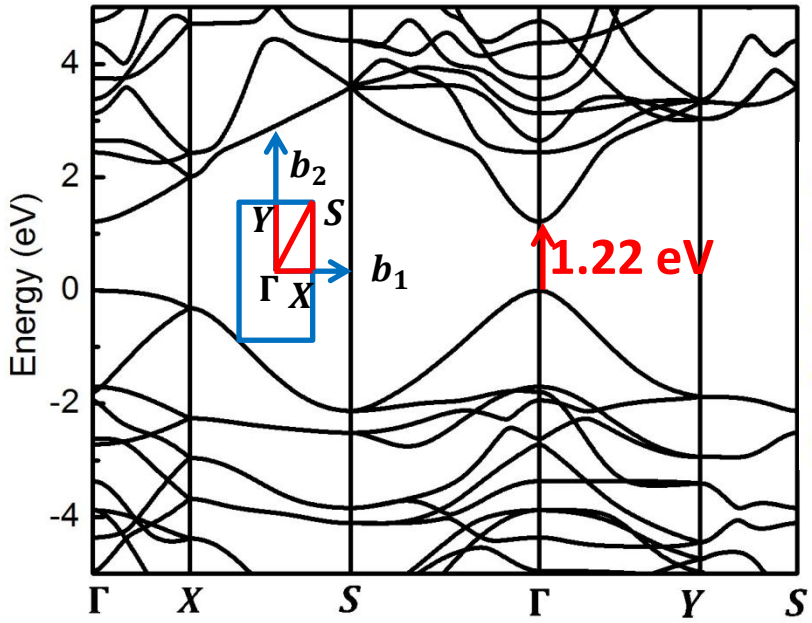
Yang, J.-H.; Zhang, Y.-Y; Yin, W.-J.; Gong, X.-G. Yakobson, B. I.; Wei. S.-H. *Nano Lett, 2016, 16 (2)*

Formation energy diagram of Si_xS_{1-x}

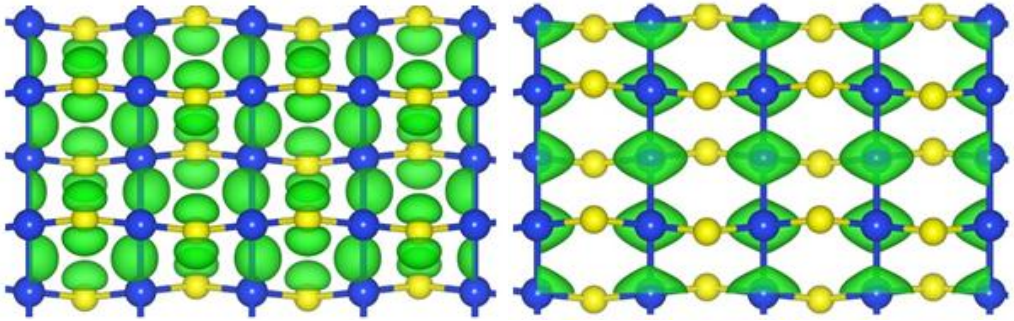


- [1] Zhu, Z.; Guan, J.; Liu D.; Tománek, D. *ACS Nano* **2015**, 9, 8284–8290
- SiS2 structures are all 3D, obtained from open data base.
- Formation energy is referenced to 3D Si bulk and S_8 molecule.

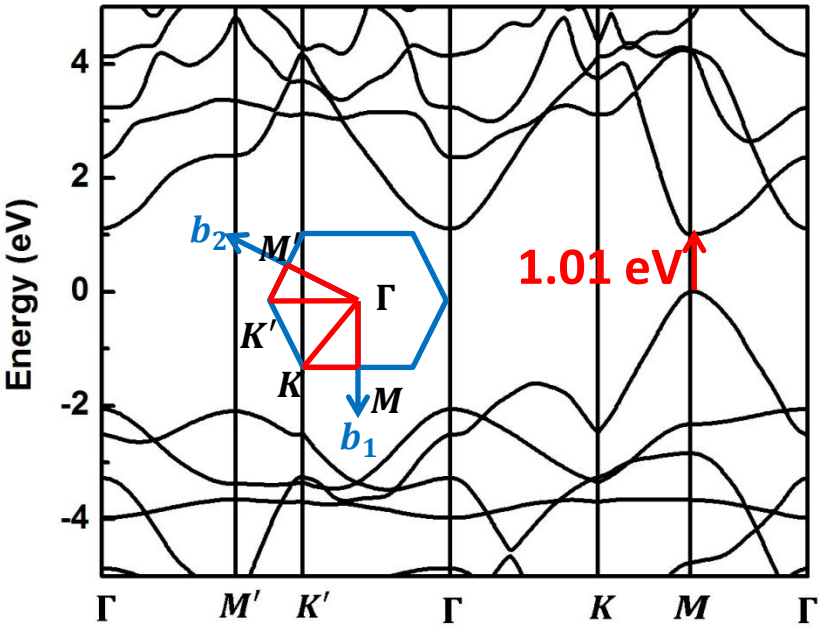
Band structures and DOS



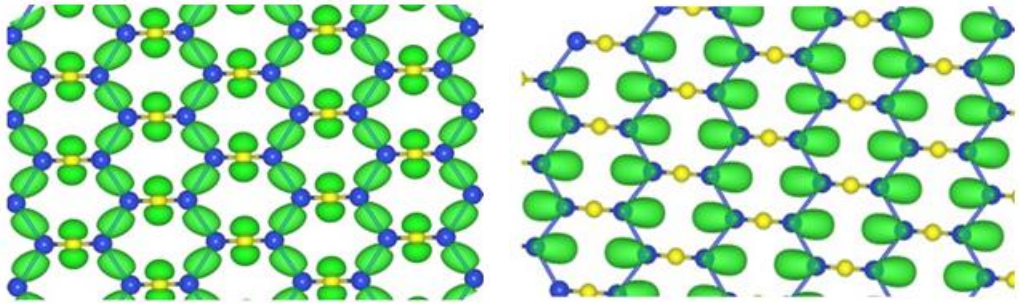
***Pma2*-SiS**



VBM CBM

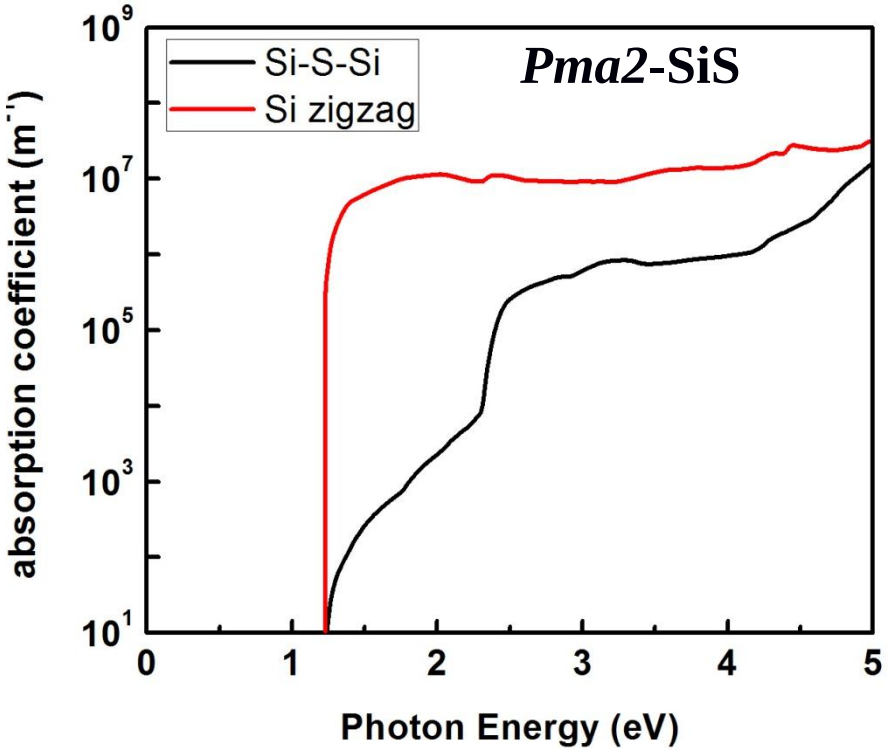


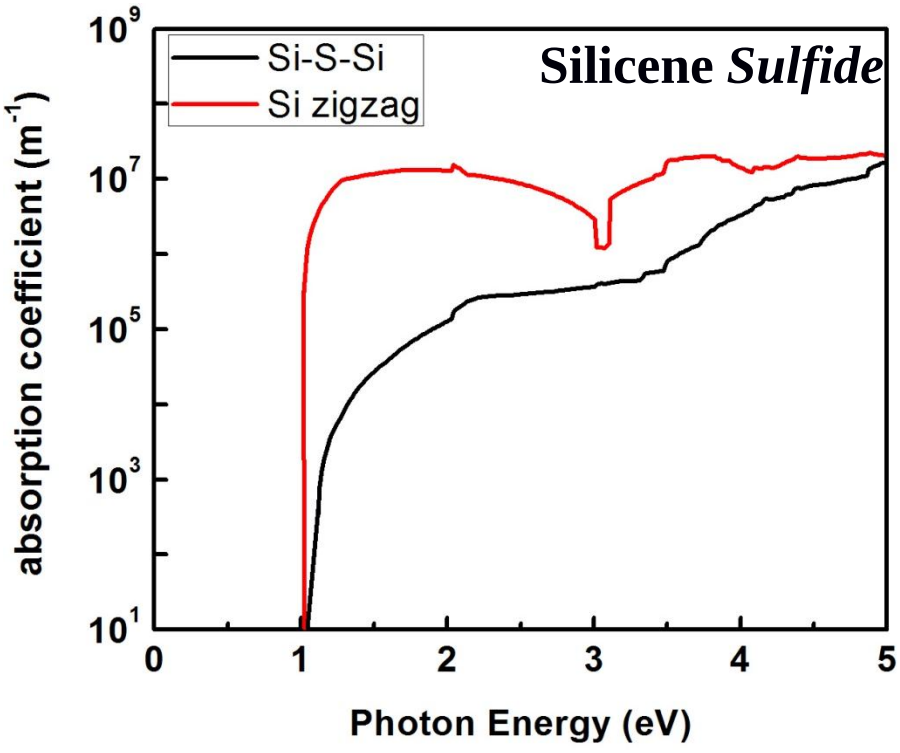
Silicene *Sulfide*



VBM CBM

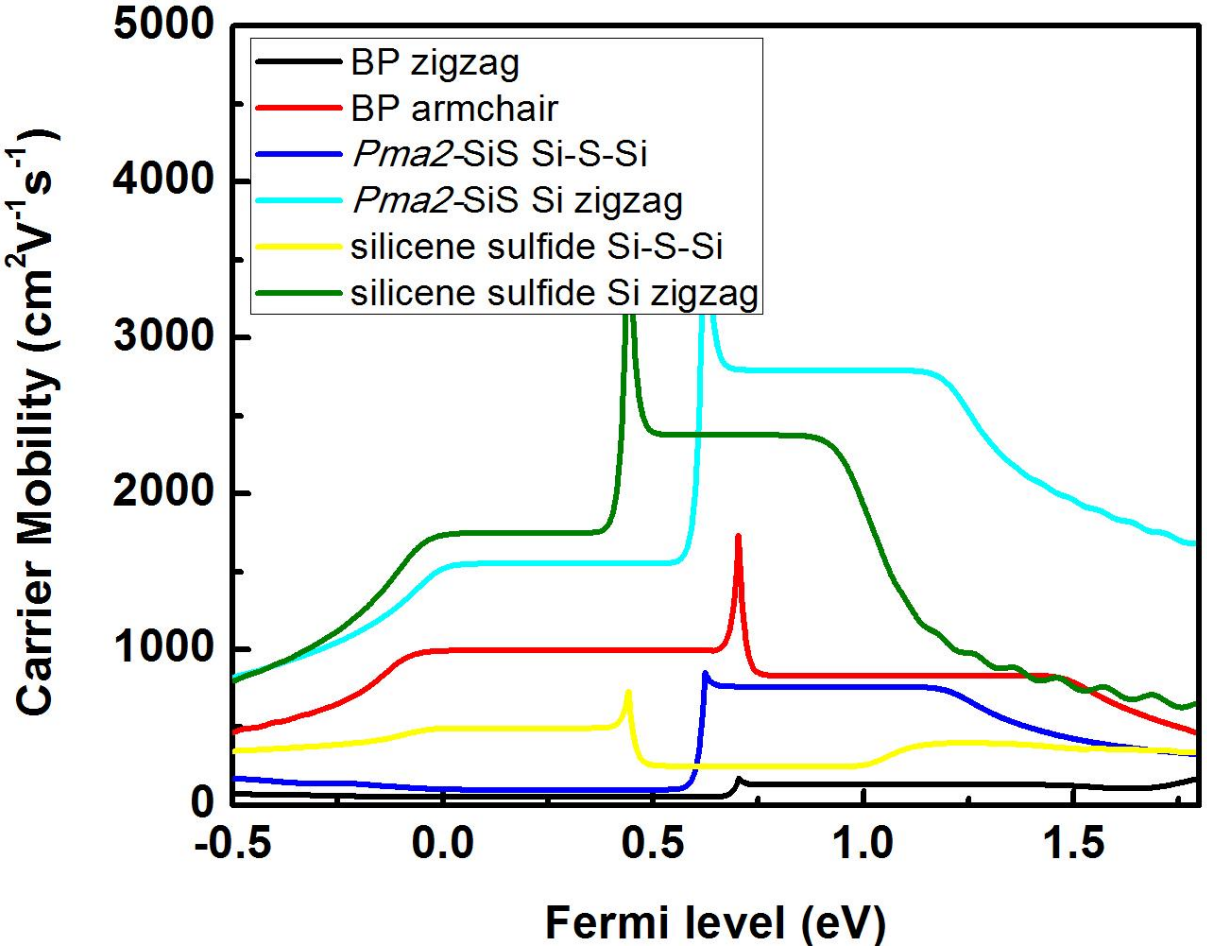
Optical properties





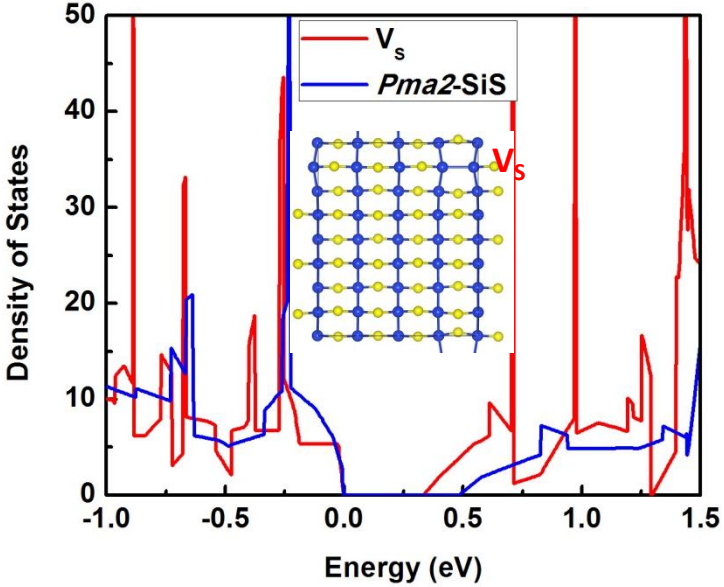
Both Pma2-SiS and Silicene Sulfide have direct bandgaps which can allow optical transitions at band edges and have values close to the optimal requirement for solar cell applications.

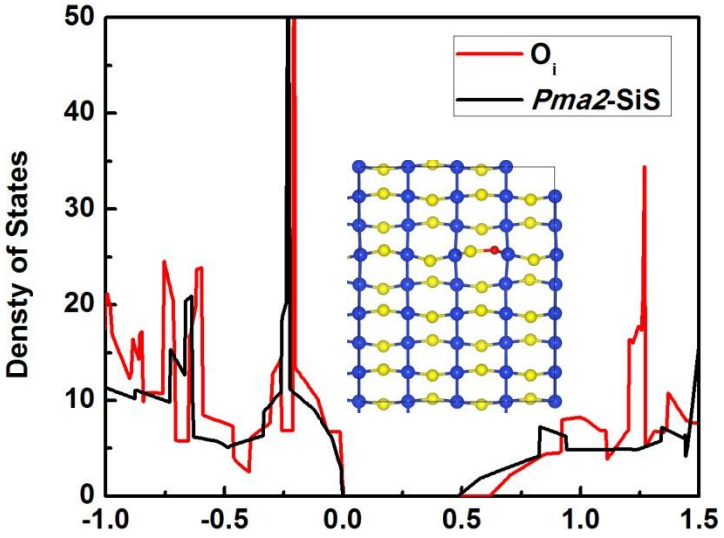
High carrier mobility in SiS in comparison with phosphorene

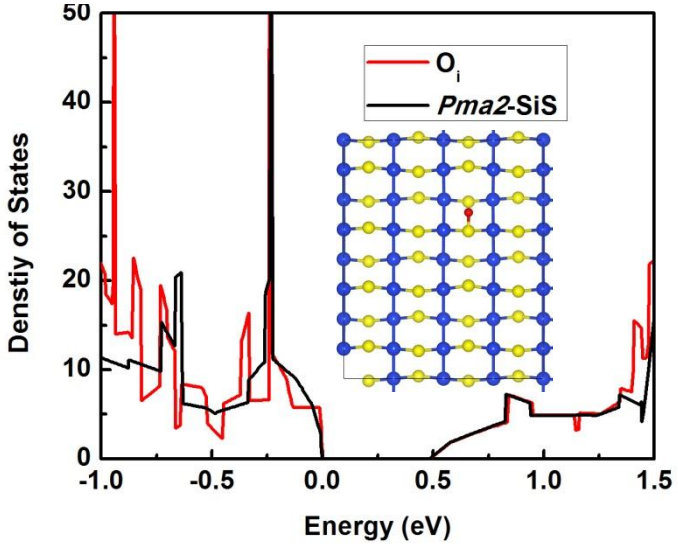


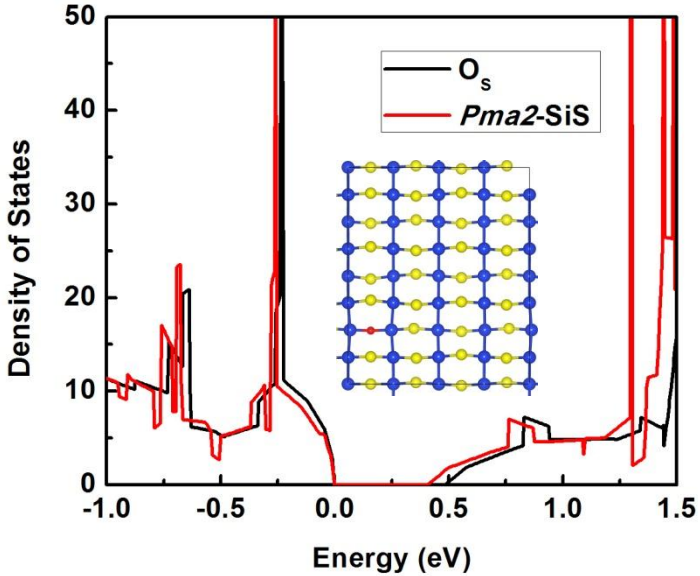
- Mobility $\mu = \frac{\sigma}{e*n}$, σ is conductivity, e is elemental charge, and n is carrier density.
- Both σ and n are functions of electron potentials or Fermi levels, so is μ.
- The VBMs of each materials are set as zero Fermi energy.
- Our SiS systems have high carrier mobility and are suitable for FETs.

Good defect properties of SiS: no gap states, air stablility









- S vacancy has no deep gap states;
- O-related defects have no deep gap states.

4应用案例

体系3：搜索表面测试案例

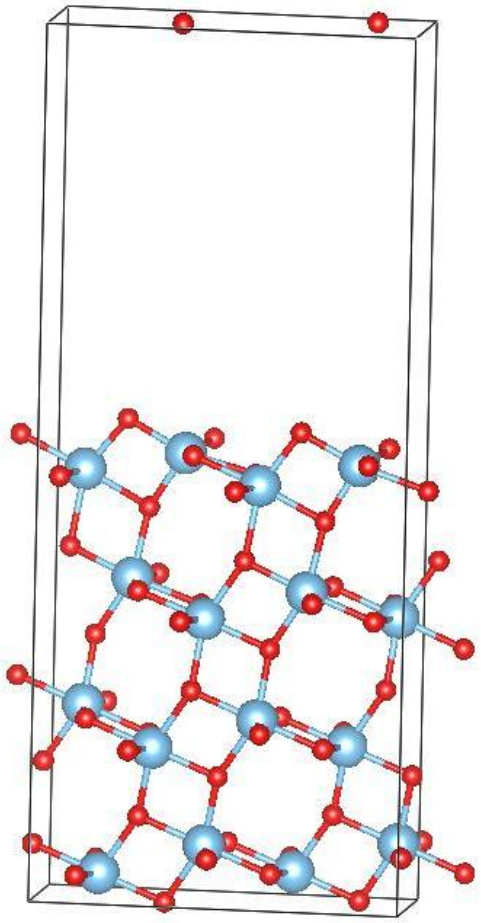
System: TiO_2 (anatase[101])

Substrate:
Ti: 12; O: 24

Surface:
Ti: 4, O: 8

4 Ti atom and 8 O atoms for global optimization

Perfect surface could be found in an average of 3 generation



IM²ODE能够在种群数是30的情况下，平均3代把完整的表面重复出来

4应用案例

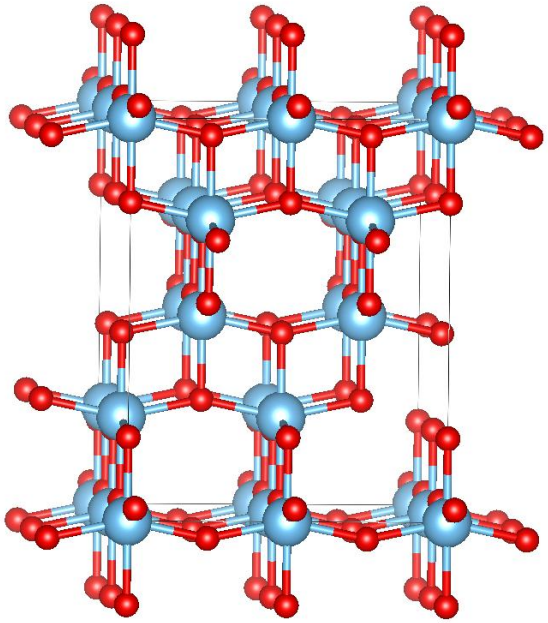
体系4：搜索复杂缺陷测试案例

System: Defect of TiO_2 (anatase)

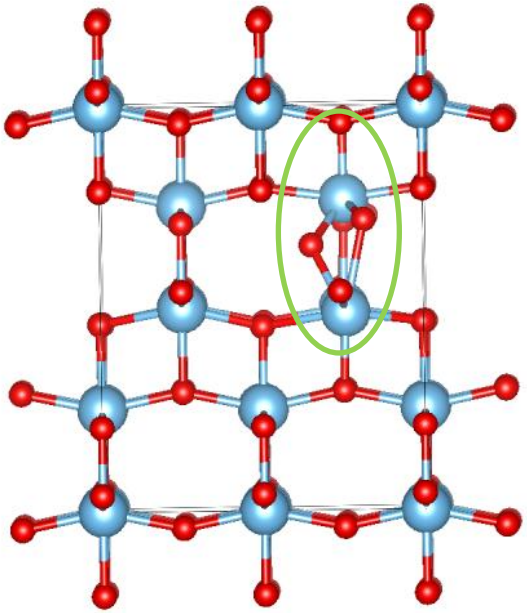
Ti: 16; O: 32

1 Ti atom and 6 atoms for global optimization

NELECT = 256, perfect structure



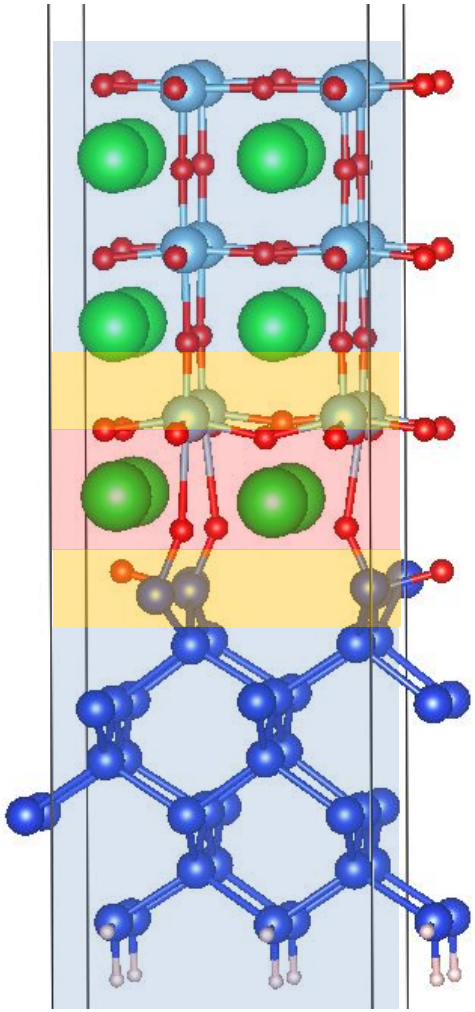
NELECT = 254, O atoms tends to be closer



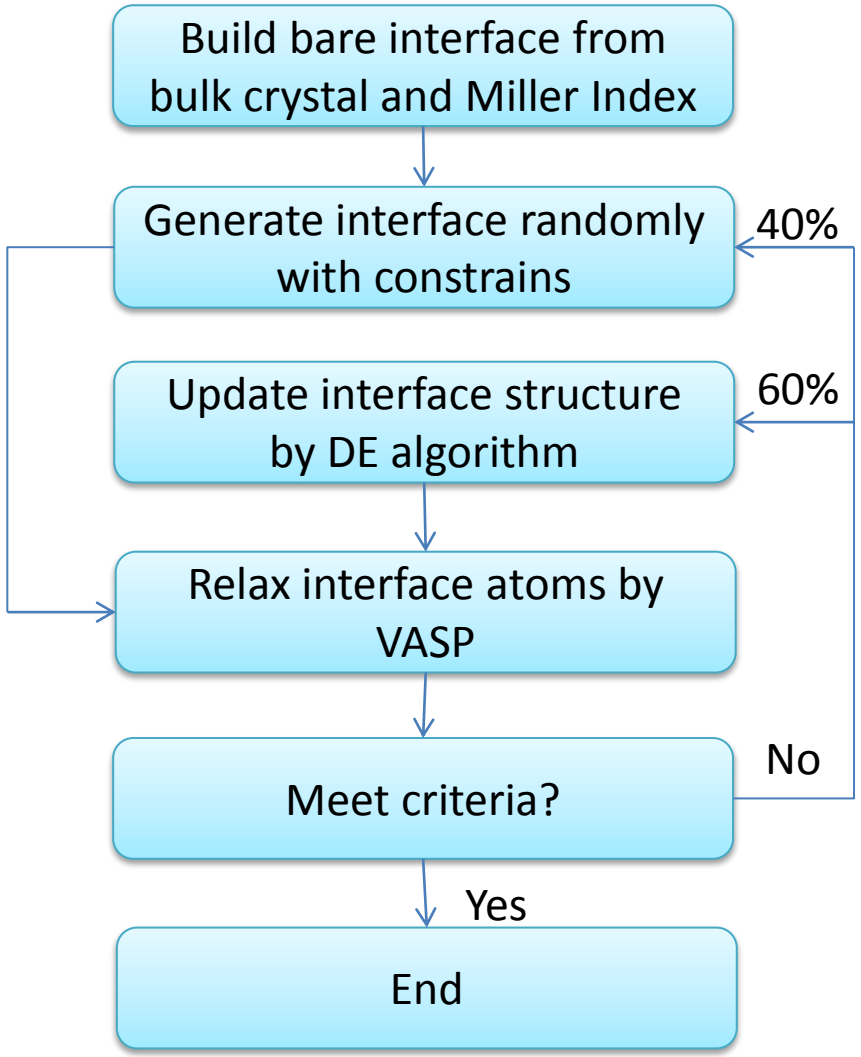
IM^{2}ODE能够只对块体中指定的部分原子做全局优化

4应用案例

体系5：搜索界面的测试案例



program chart



4应用案例

体系6：搜索界面的测试案例

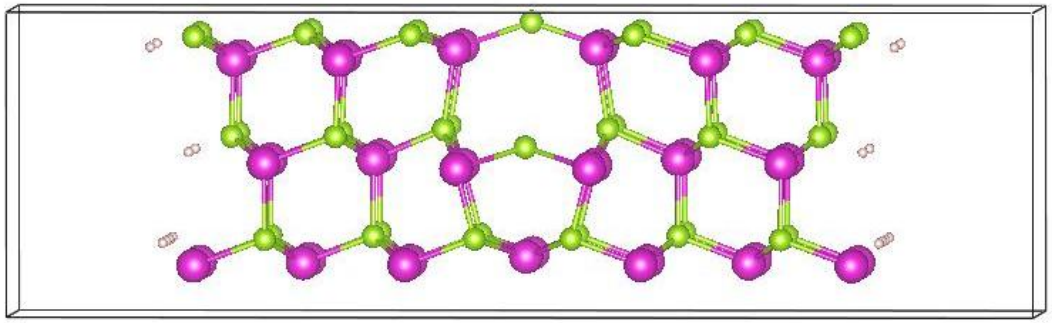
System: CdSe

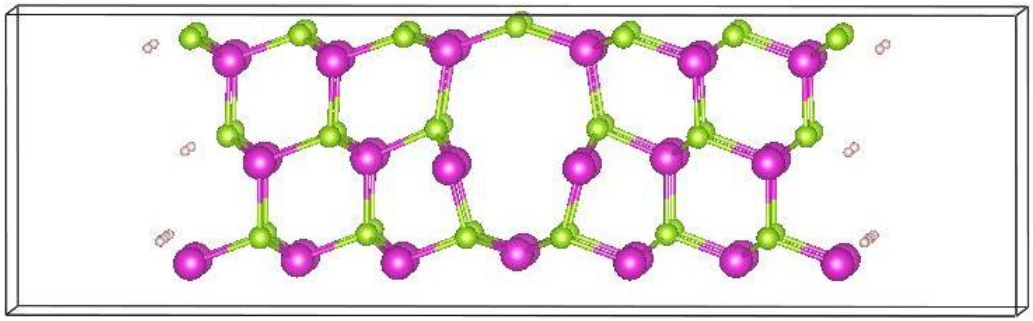
Substrate:
Cd: 36; Se: 36

Grain boundary:
Cd: 2, Se: 2

2 Cd atom and 2 Se atoms for global optimization

Stable and meta-stable structures could be found in 3 generations





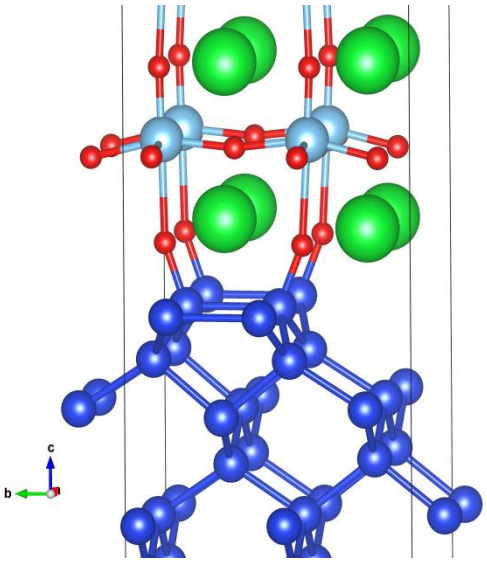
IM^{2}ODE能够在种群数是30的情况下，平均3代把能量最低的结构搜索出来

4应用案例

体系4：搜索界面的测试案例

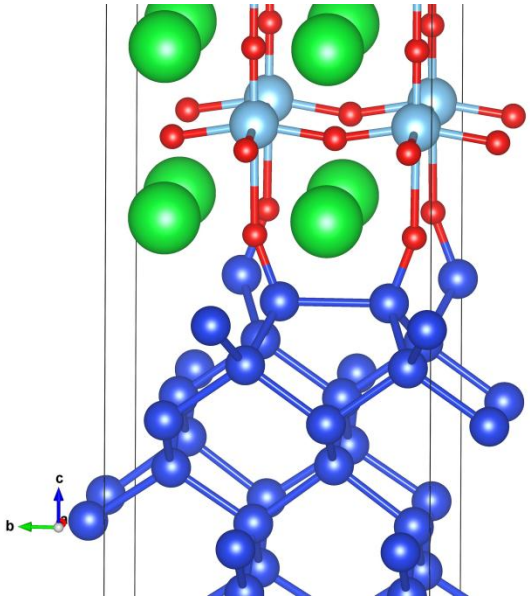
Interface of STO-Si

PRL105,217601 (2010)



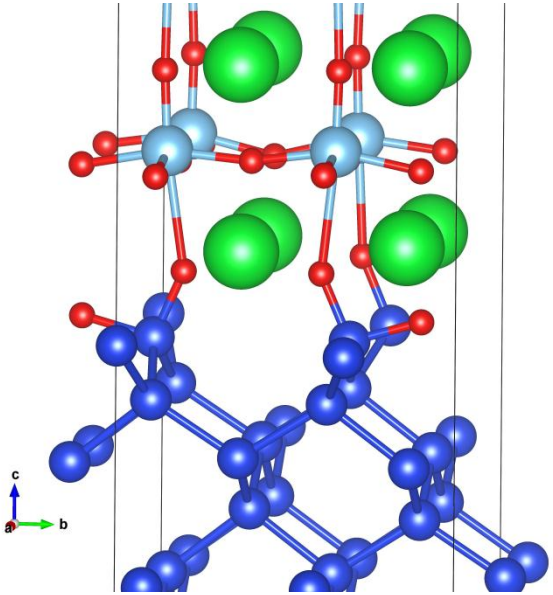
0 eV

IM^{2}ODE



-0.272 eV

IM^{2}ODE



-0.478 eV

IM^{2}ODE能够搜索到比传统的Si-SrO面能量更低的结构

5已取得成果

RAPID COMMUNICATIO

PHYSICAL REVIEW B **92**, 201413(R) (2015)

Structural evolution and optoelectronic applications of multilayer silicene

Zhi-Xin Guo,[1,*] Yue-Yu Zhang,[2] Hongjun Xiang,[2] Xin-Gao Gong,[2] and Atsushi Oshiyama[3]

Hybrid crystalline sp^2–sp^3 carbon as a high-efficiency solar cell absorber

Yue-Yu Zhang [a, b], Shiyou Chen [c], Hongjun Xiang [a, b], Xin-Gao Gong [a, b, *]

NANO LETTERS

Letter

pubs.acs.org/NanoLett

Two-Dimensional SiS Layers with Promising Electronic and Optoelectronic Properties: Theore

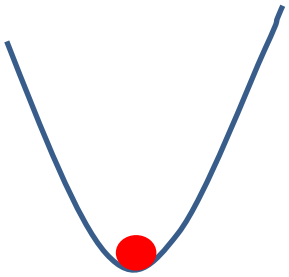
Ji-Hui Yang,[*,†,∥] Yueyu Zhang,[‡,§] Wan-Jian Yin,[†] X.

multi-objective inverse b
sorption, and the simula
materials such as GaAs, w
application as light-abso

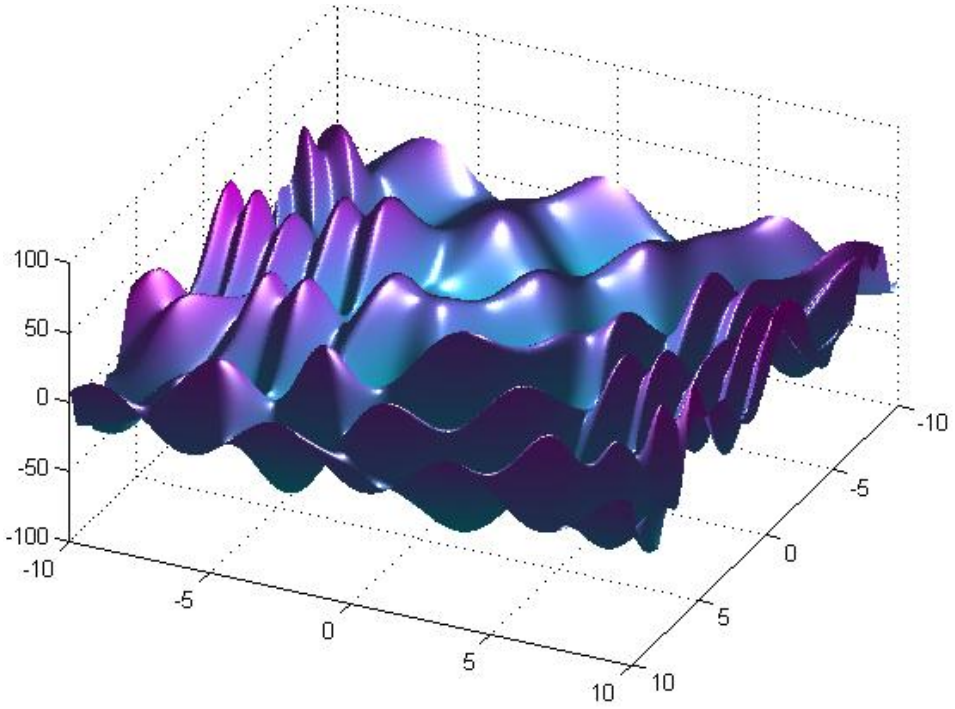
ding.
r too
rbon

自2015年面世以来，IM^{2}ODE成功预言的材料已发表SCI论文7篇，包括功能性块体材料、二维材料、团簇等。

未来的展望



Global Optimization

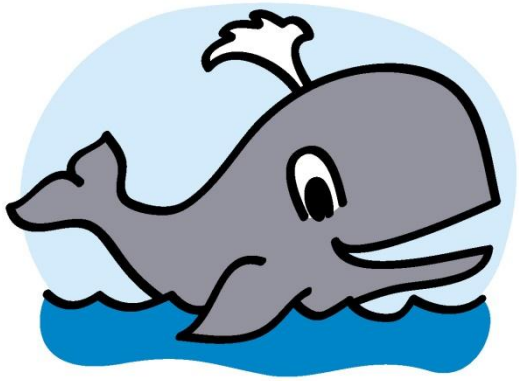


We believe that the number of functional meta-stable compounds which human beings can synthesis is infinity. And IM^{2}ODE offers some way to search for useful meta-stable states for further studying.

∞

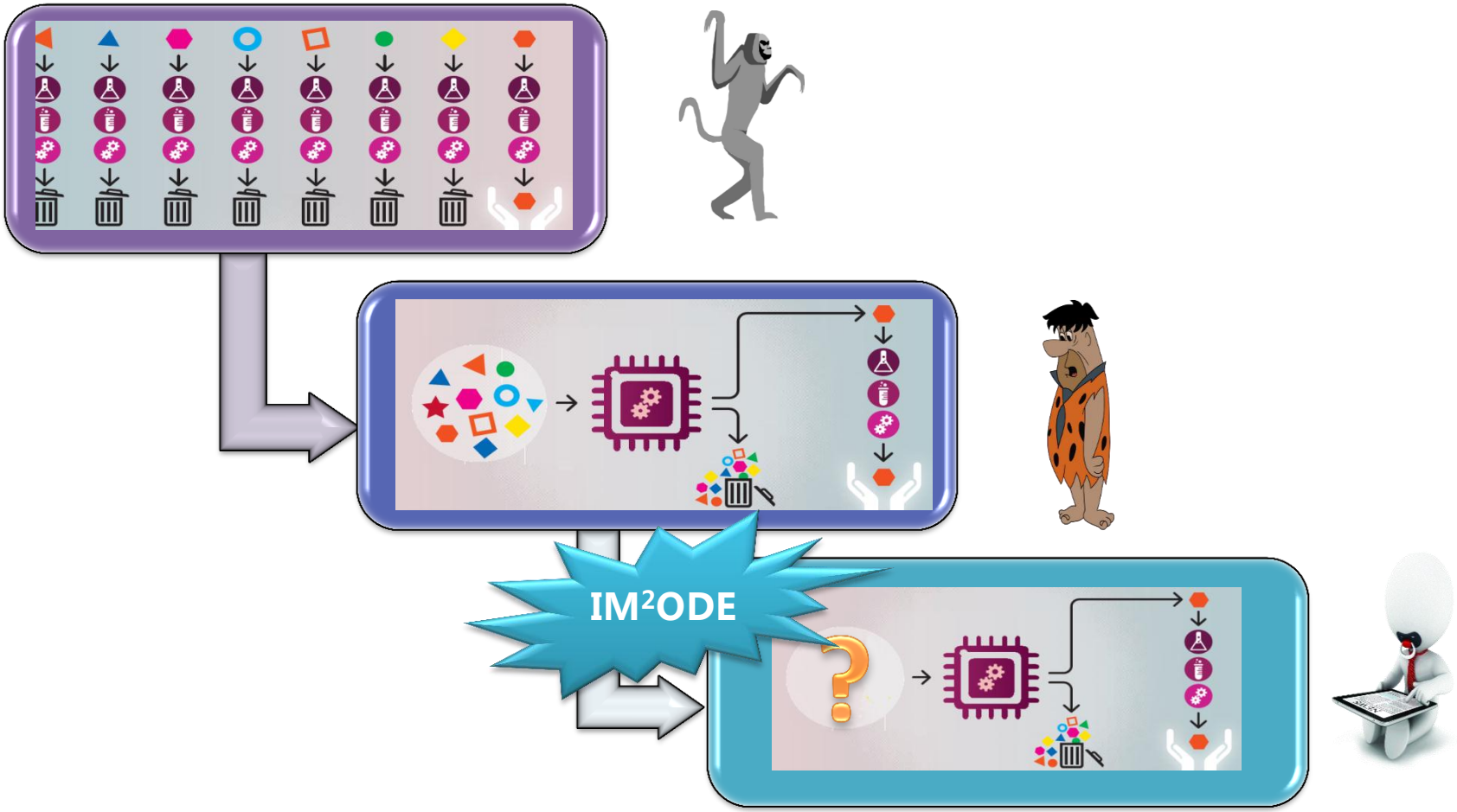
6结论

- 我们开发了一款**逆向材料设计的软件包IM^{2}ODE**，可以根据所需的性质来设计材料.
- IMODE采用**多目标全局优化**算法，具有很好的普适性.
- 我们给出了在不同体系中的应用案例，来说明IMODE的**普适**和**高效**.



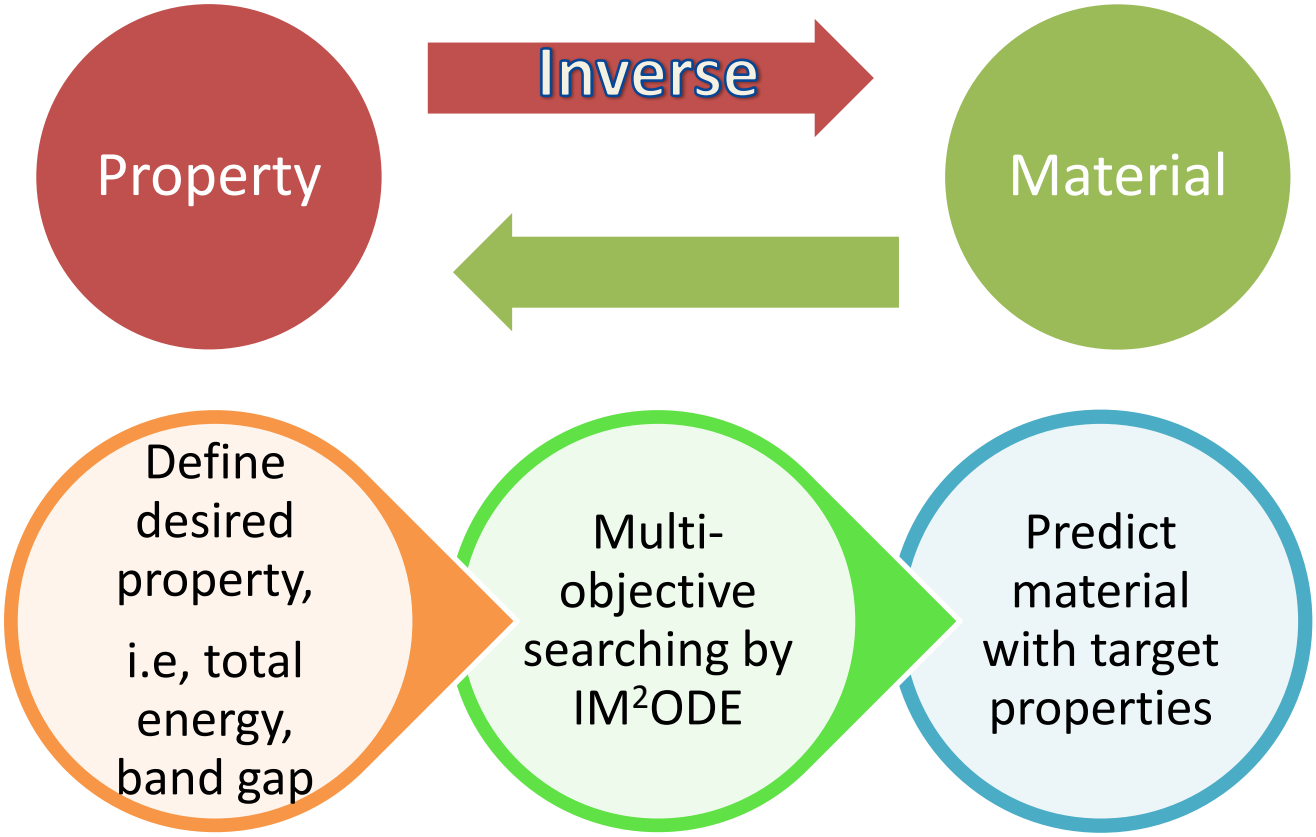
Thank you!

Inverse Design of Materials



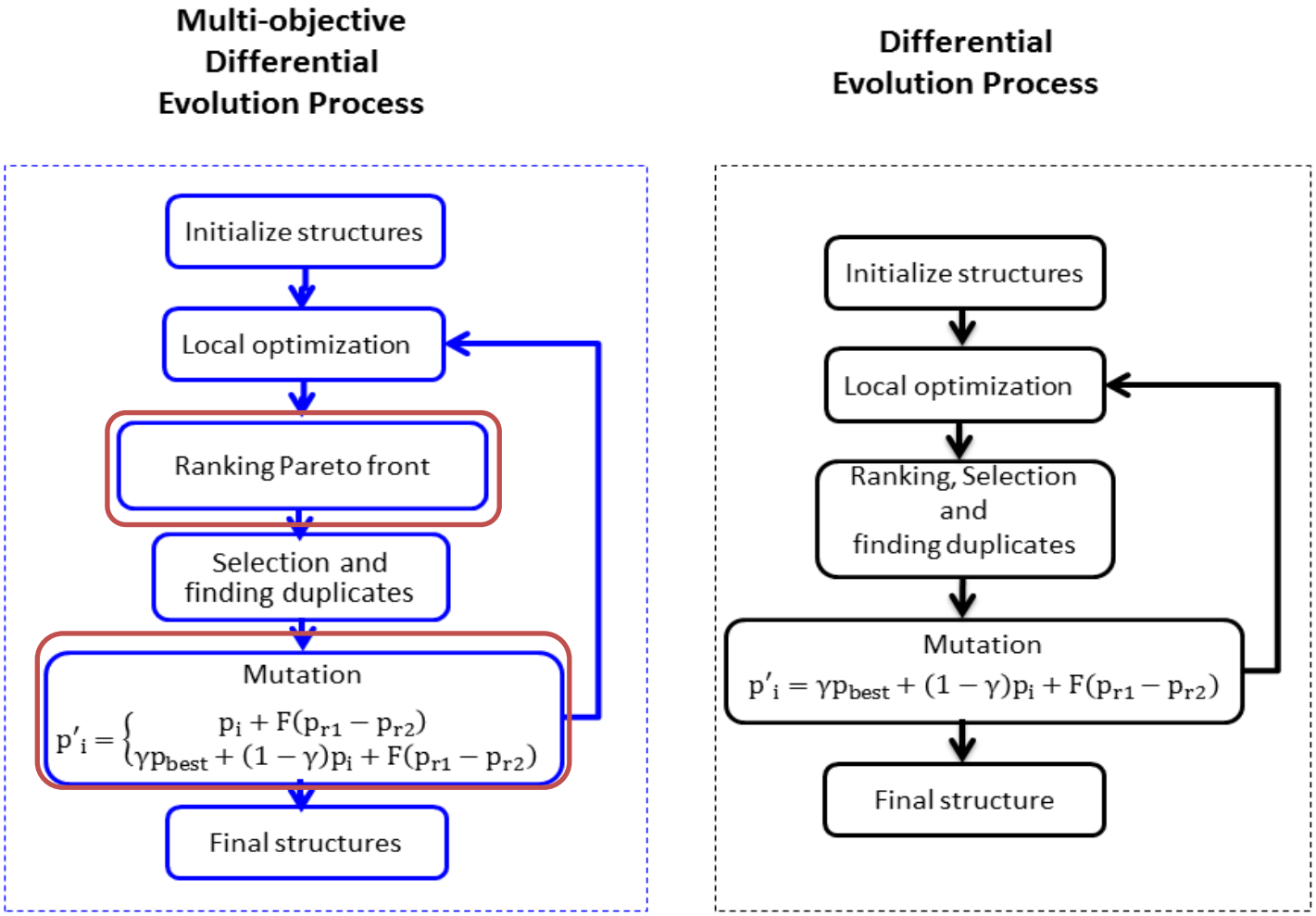
IM^{2}ODE

Inverse Design of Materials by Multi-Objective Differential Evolution



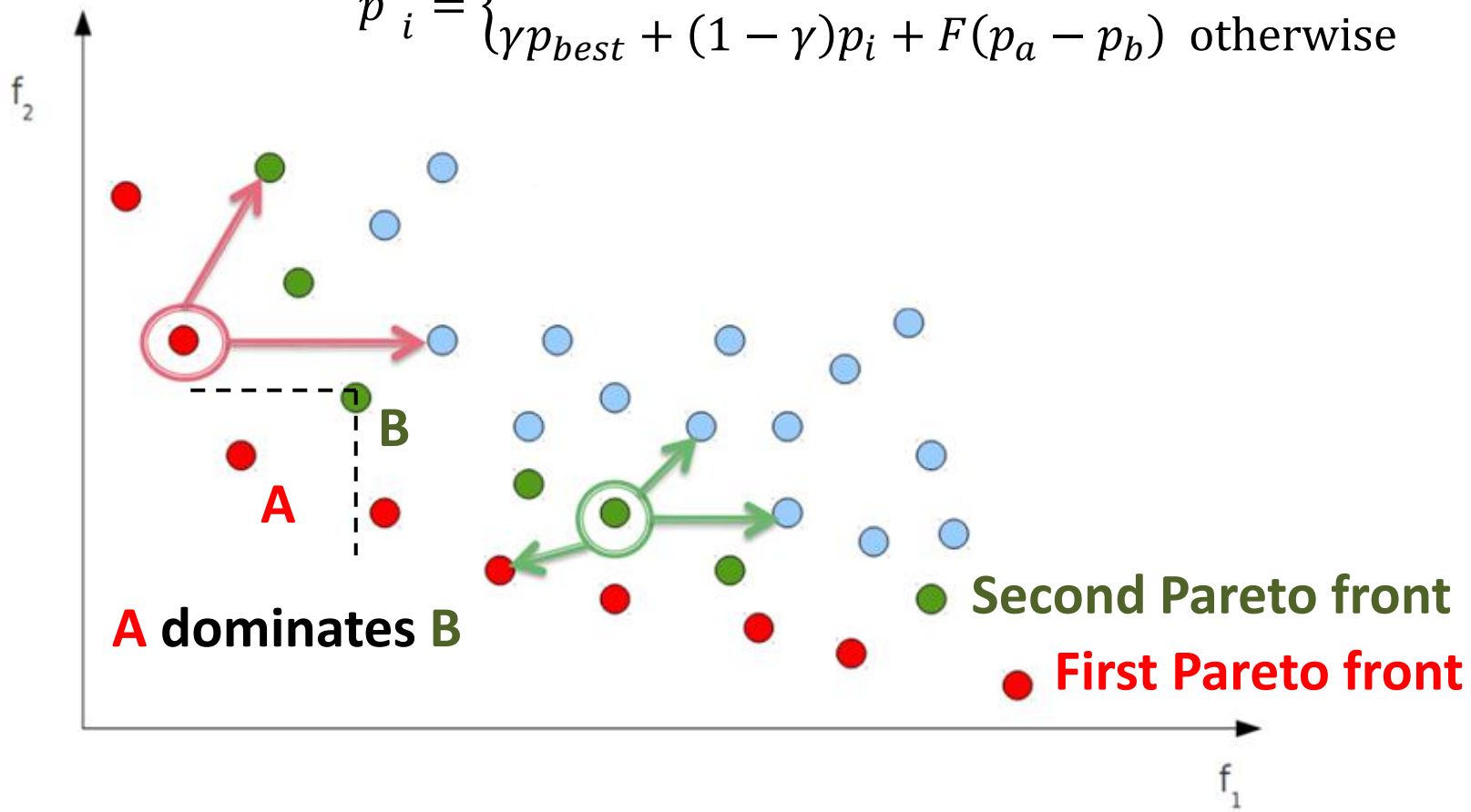
Zhang, Y.-Y.; Gao, W.; Chen, S.; Xiang, H.; Gong, X.-G. *Comput. Mater. Sci.* **2015**, 98, 51.

Flow Chart of IM^{2}ODE

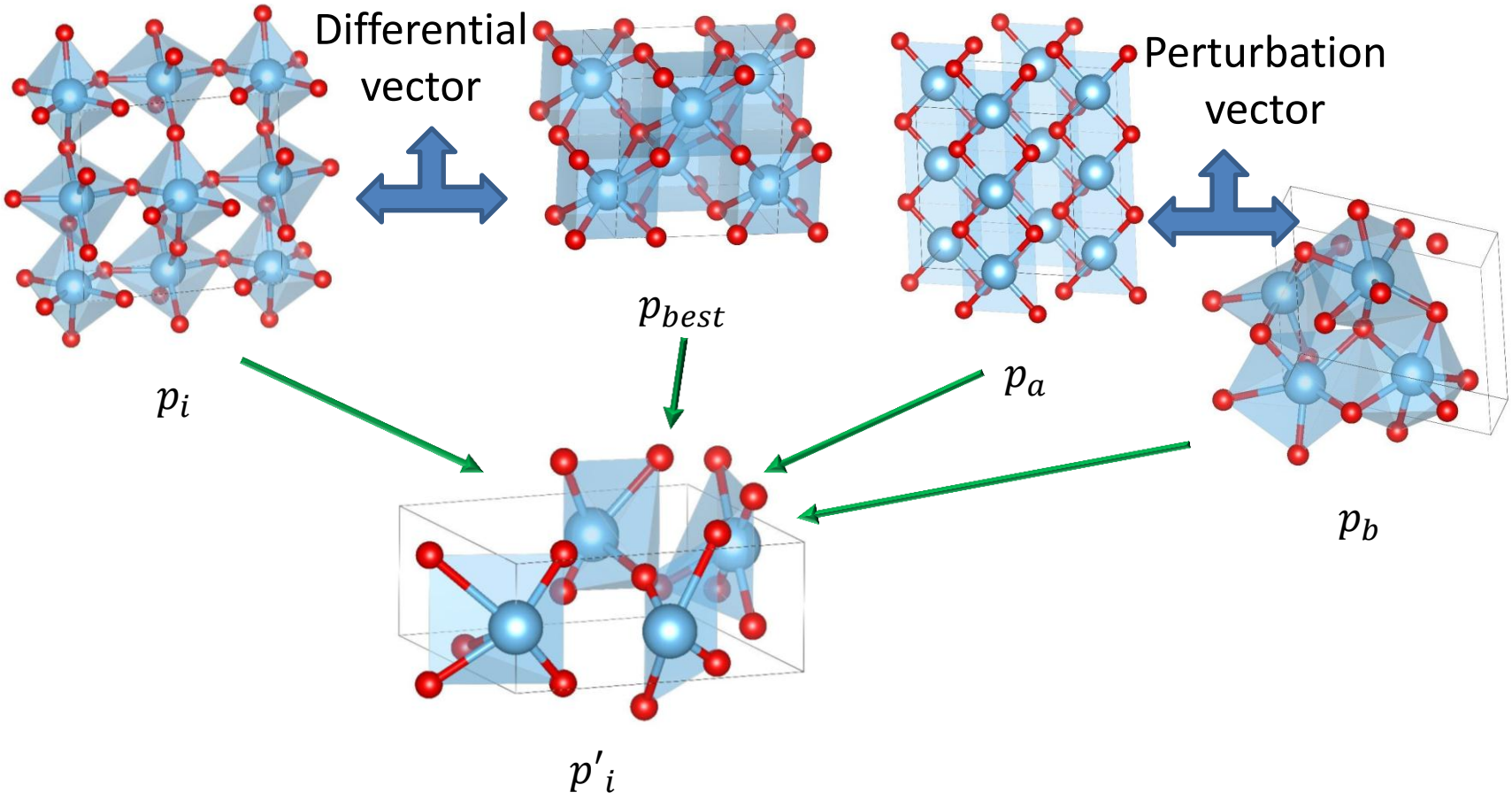


Multi-Objective Differential Evolution

$$p'_i = \begin{cases} p_i + F(p_a - p_b) & \text{if } p_i \text{ is non} - \text{dominated} \\ \gamma p_{best} + (1-\gamma)p_i + F(p_a - p_b) & \text{otherwise} \end{cases}$$

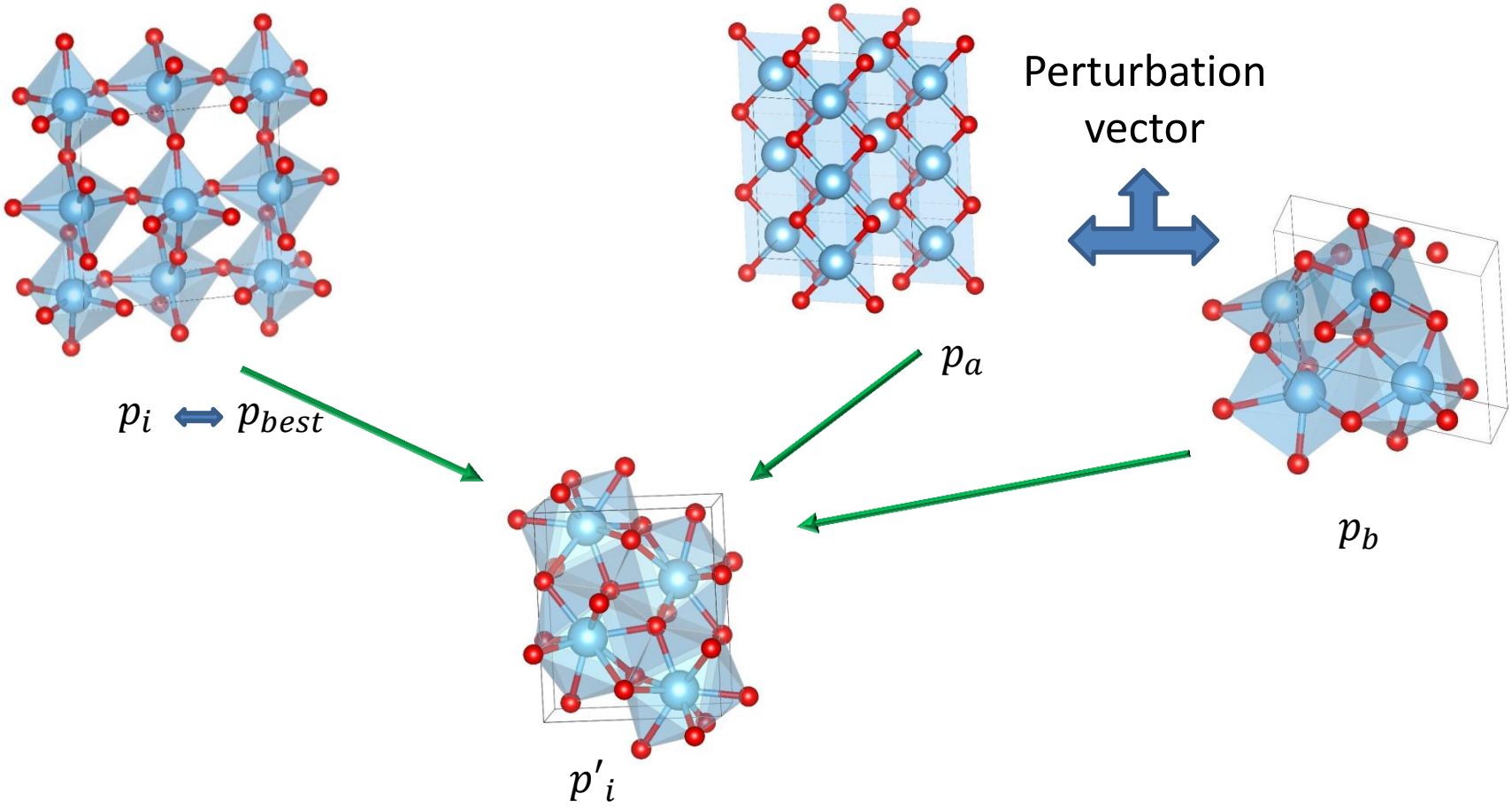


Mutation Operation (dominated)



$$p'_i = \gamma p_{best} + (1-\gamma)p_i + F(p_a - p_b)$$

Mutation Operation (non-dominated)



$$p'_i = p_i + F(p_a - p_b)$$

MODE

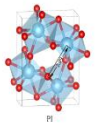
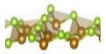
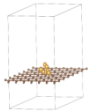
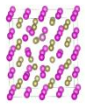
Flow Chart

Capability of IM^{2}ODE

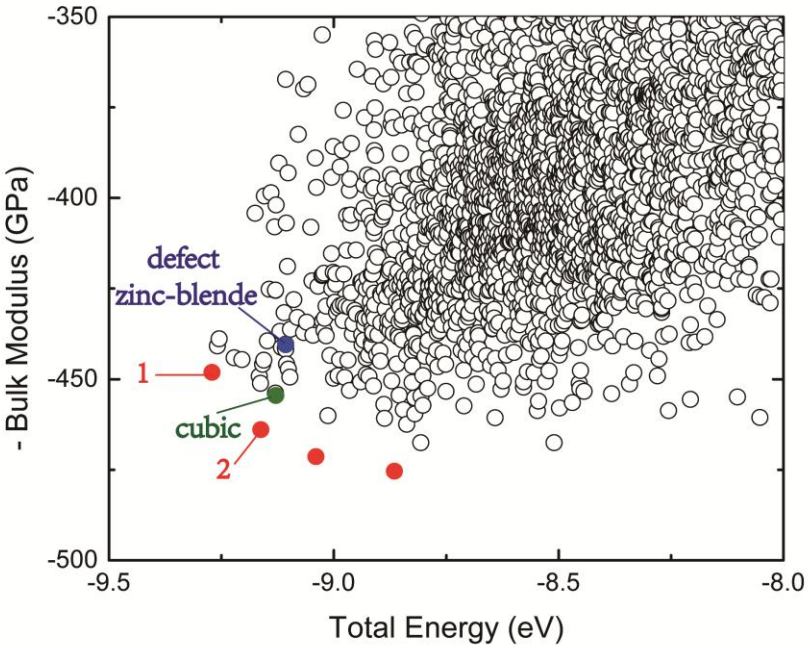
Property

- Electronic structure
- Optical absorption
- Mechanical property

Structure

-  Crystal
-  2D
-  Cluster
-  Surface
-  Interface
-  Defect

Find Materials with Large Bulk Modulus



- Tested system: C_3N_4
- objective functions:

$$\min z_1 = \text{total energy}$$

$$\min z_2 = -B = -\frac{N_c}{4}\frac{0.624 - 0.070I}{d^{3.5}}$$

	Total Energy / atom (eV / atom)	B(empirical equ)(GPa)	B(ab initio)(GPa)
Defect zinc-blende	0.0	440.557	414.718
cubic	-0.022	454.546	462.48
1	-0.164	448.134	30.859
2	-0.055	463.917	140.35

➢**YY Zhang**, W Gao, S Chen, H Xiang, XG Gong, *Comp. Mat. Sci*, 2015, 98 (51)

Find Materials with Desired Band Gap

- design material with a target direct band gap E_g

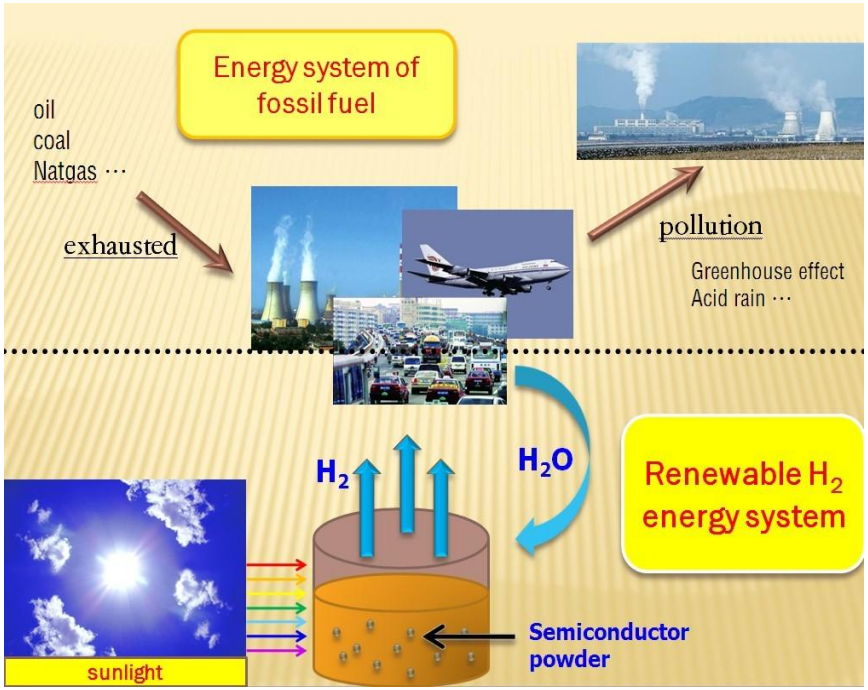
$$\min z_1 = \text{total energy}$$

$$\min z_2 = |E_g - \text{direct gap}| + |\text{direct gap} - \text{indirect gap}|$$

System	E_g (eV)	Average generation
α-Al_2O_3	6.4	2
Diamond (Carbon)	4.1	3
Graphite (Carbon)	0.0	3

- design material with a target direct band gap E_g

Motivation to Design TiO_2 with better Optical property



Titanium Oxide (TiO_2):

✓ great potential in PEC water splitting

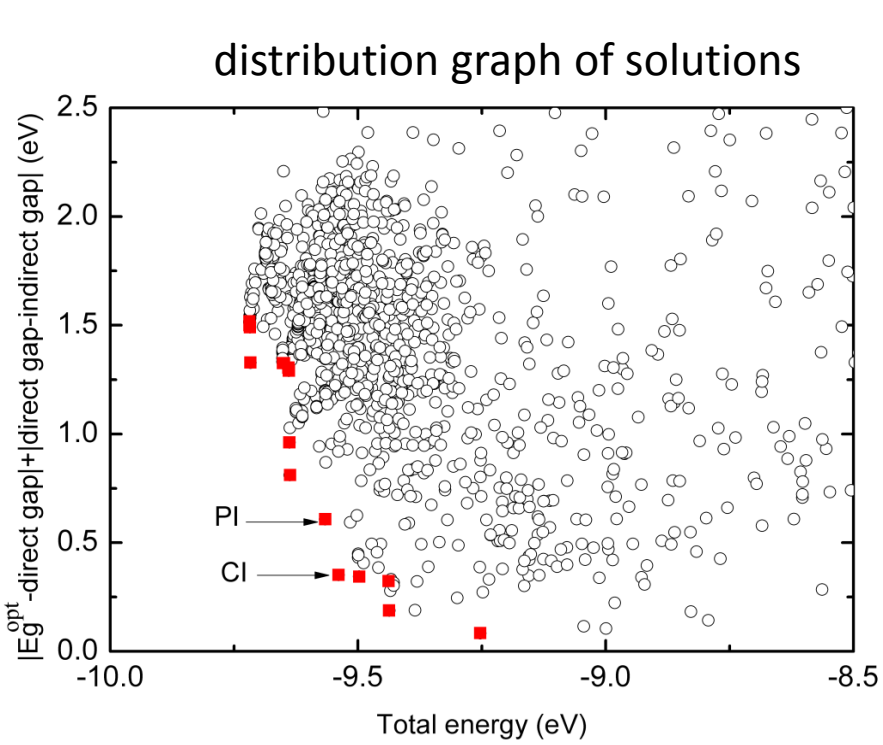
✓ low cost, nontoxic …

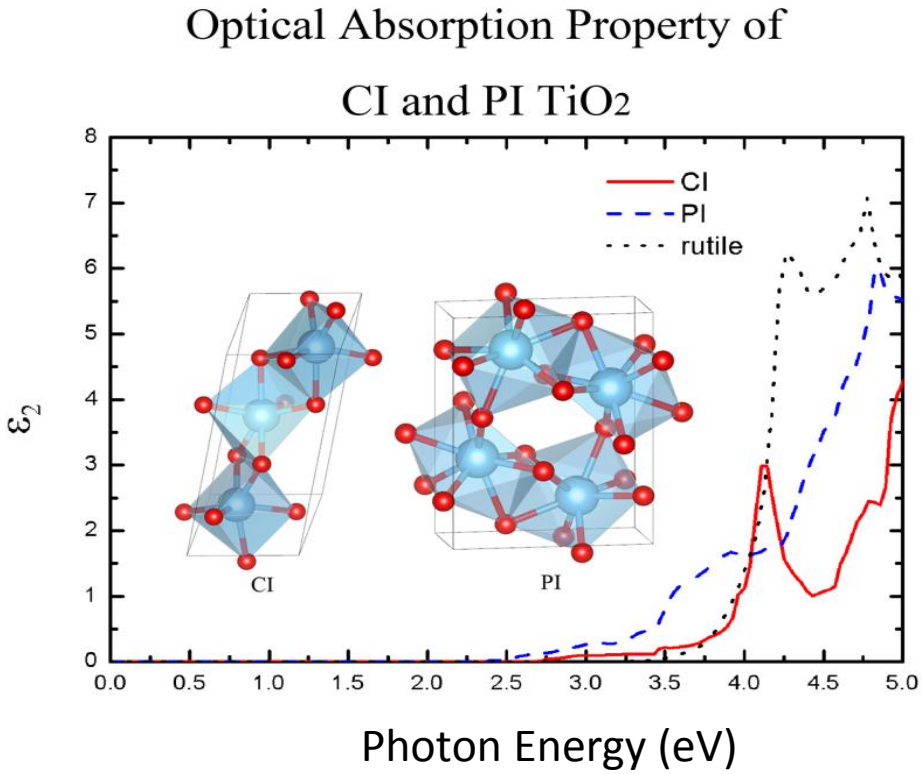
✓strong catalytic activity, high chemical stability

- Large intrinsic band, absorbs only UV

➢ A dopant-free TiO_2 phase with a suitable band gap is highly desirable.

Predicting New TiO_2 Phases with Low Band Gaps



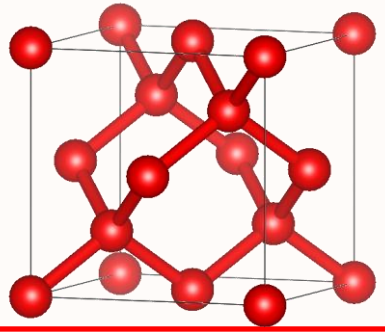


HZ Chen, YY Zhang, X Gong, H Xiang, *J. Phys. Chem. C*, **2014**, *118* (5)

Carbon: a Versatile Element

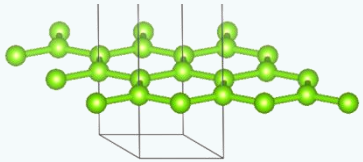
Carbon Allotropes

Diamond



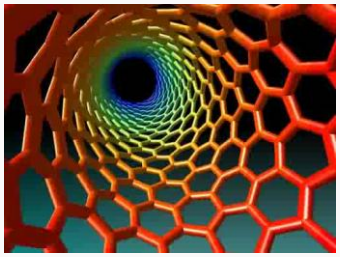
Wide band gap

Graphene



Zero band gap

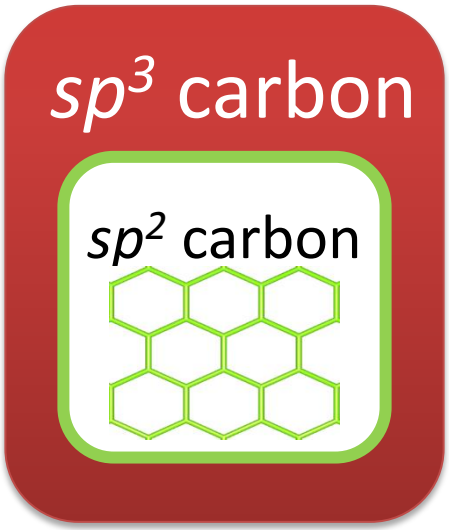
Carbon Nanotube



Zero band gap

None of them is suitable to be used as solar cell

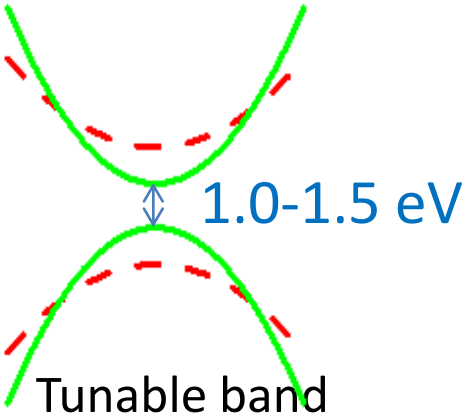
Hybrid Crystalline sp^2-sp^3 Carbon Allotropes



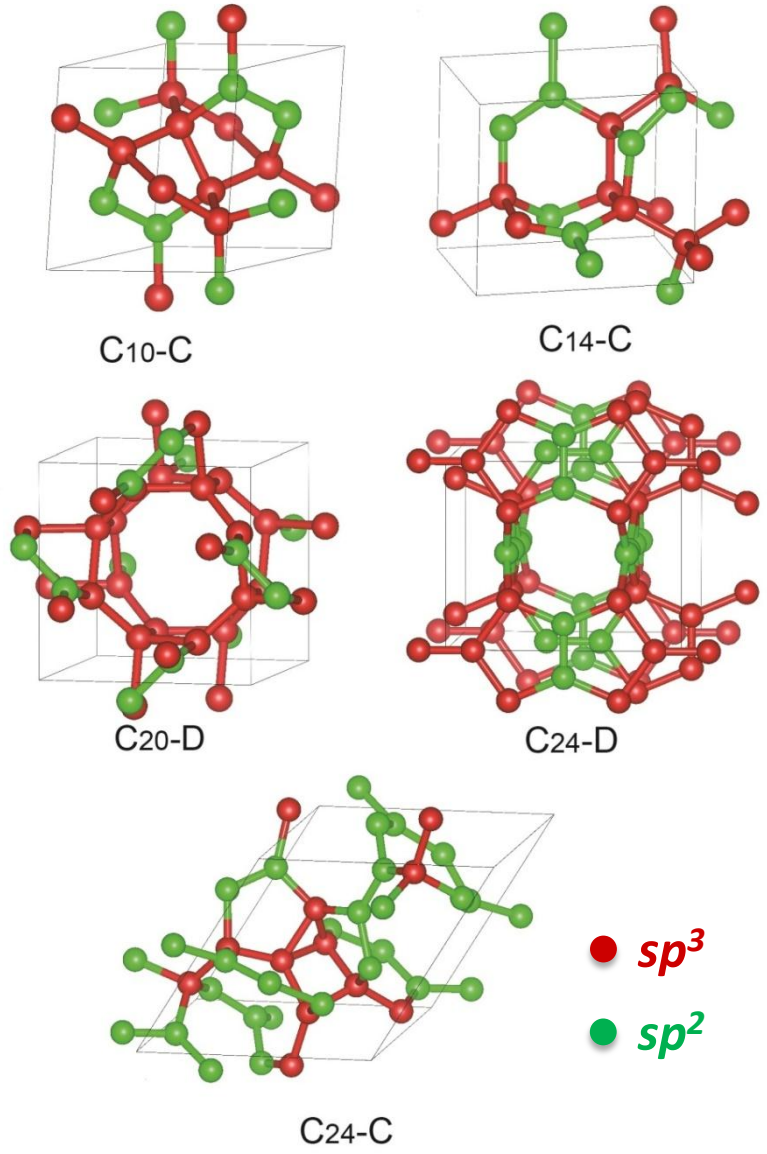
Hybrid sp^2 & sp^3

IM^{2}ODE

Inverse Design



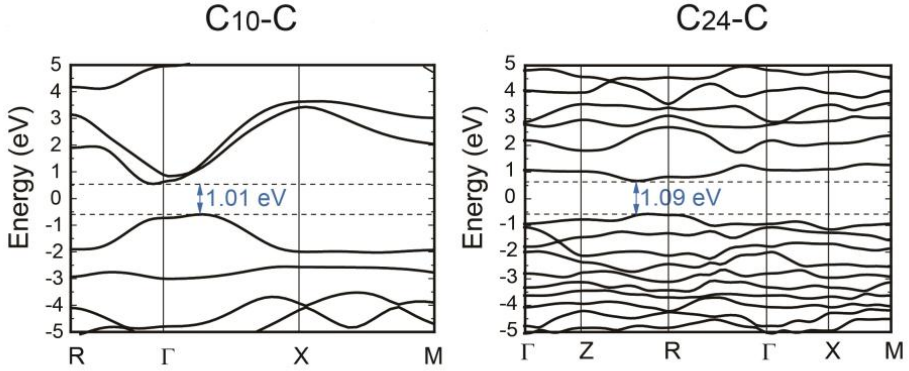
Tunable band gap



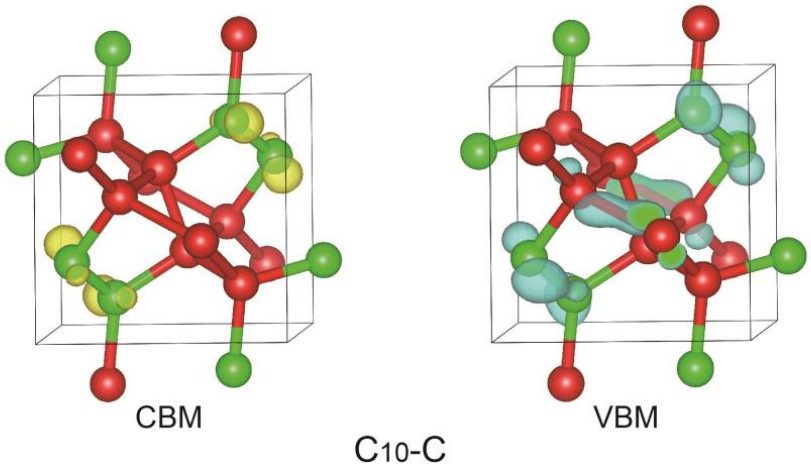
Zhang, Y.-Y.; Chen, S.; Xiang, H.; Gong, X.-G. *under revision*

High efficiency solar cell absorber

Electronic structure

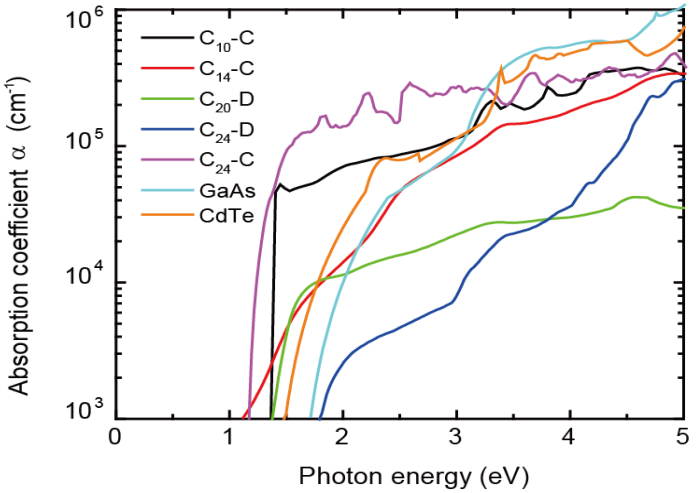


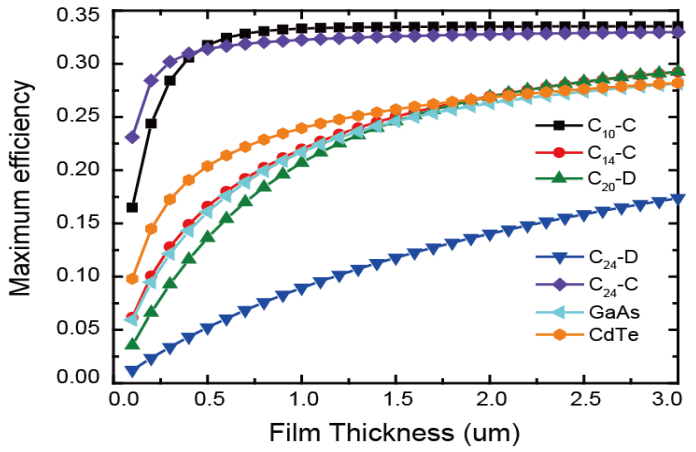
Band gap between 1.0 – 1.5 eV



Optical dipole transition between the π and π* states is allowed

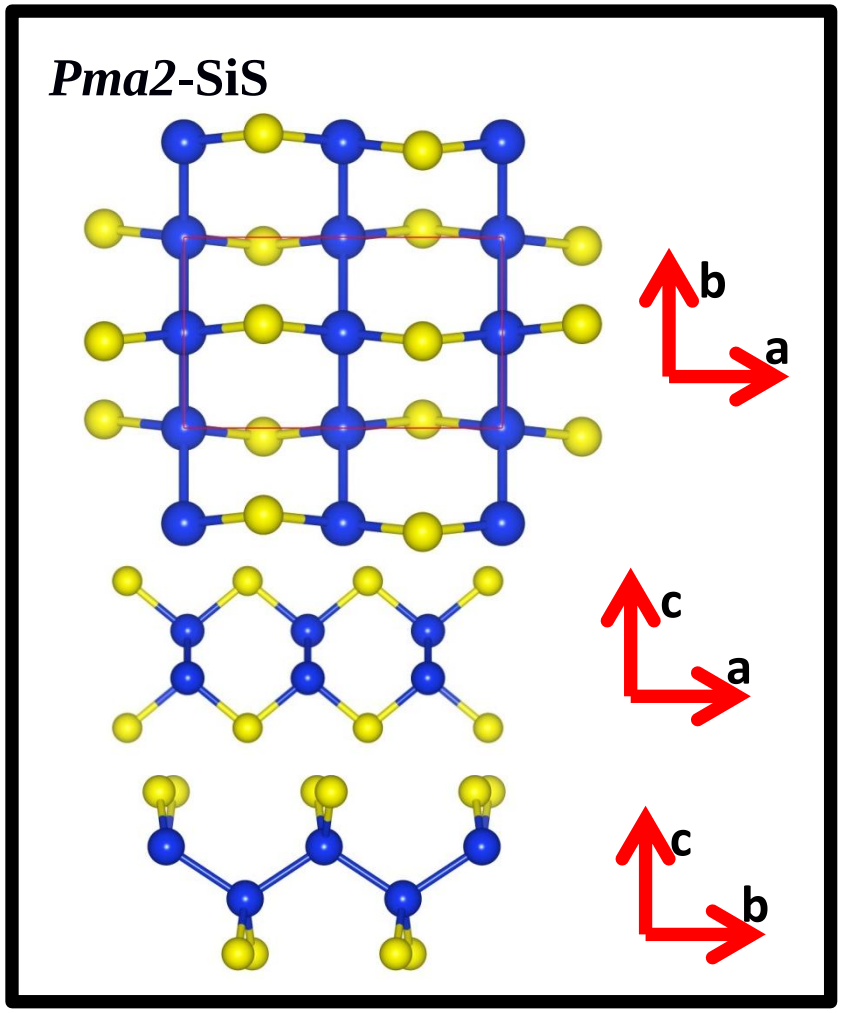
Optical property

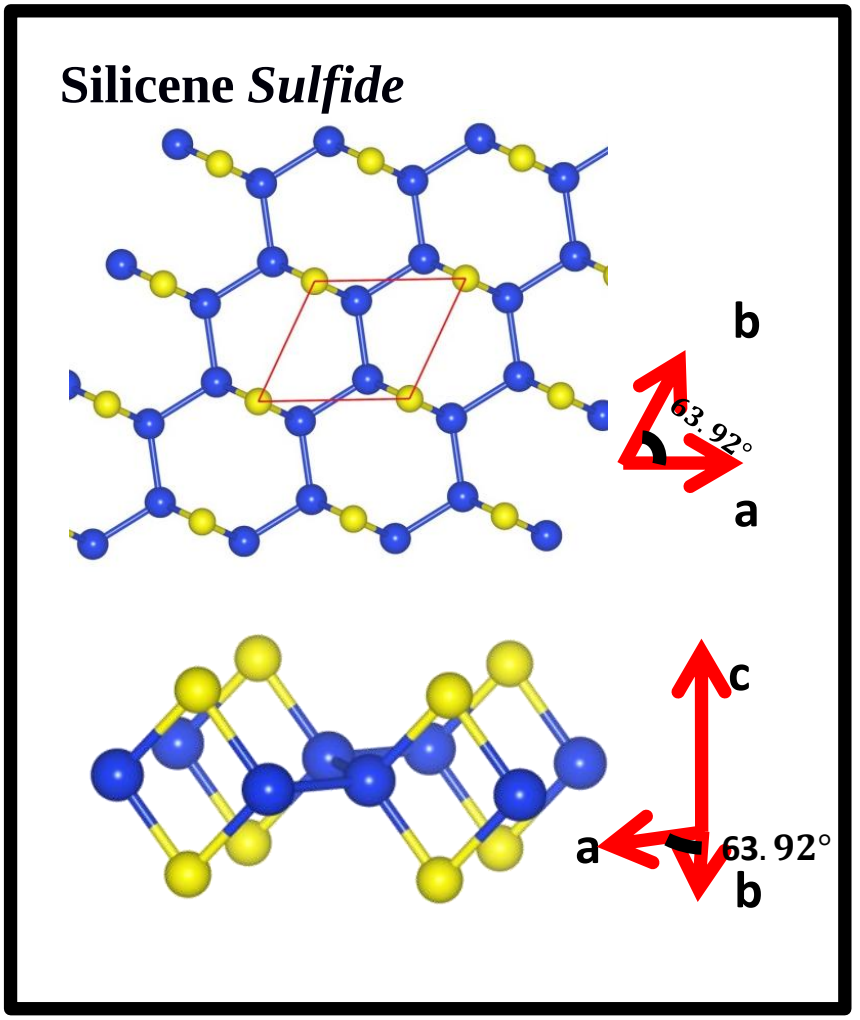




Large light absorption coefficients and high efficiencies

The lowest energy structures of SiS systems





Yang, J.-H.; Zhang, Y.-Y; Yin, W.-J.; Gong, X.-G. Yakobson, B. I.; Wei. S.-H. *Nano Lett, 2016, 16 (2)*

Publications

RAPID COMMUNICATIO

PHYSICAL REVIEW B **92**, 201413(R) (2015)

Structural evolution and optoelectronic applications of multilayer silicene

Zhi-Xin Guo,[1,*] Yue-Yu Zhang,[2] Hongjun Xiang,[2] Xin-Gao Gong,[2] and Atsushi Oshiyama[3]

Hybrid crystalline sp^2–sp^3 carbon as a high-efficiency solar cell absorber

Yue-Yu Zhang [a, b], Shiyou Chen [c], Hongjun Xiang [a, b], Xin-Gao Gong [a, b, *]

NANO LETTERS

Letter

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Two-Dimensional SiS Layers with Promising Electronic and Optoelectronic Properties: Theore

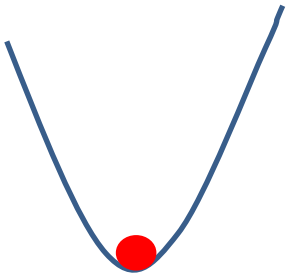
Ji-Hui Yang,[*,†,∥] Yueyu Zhang,[‡,§] Wan-Jian Yin,[†] X.

multi-objective inverse b
sorption, and the simula
materials such as GaAs, w
application as light-abso

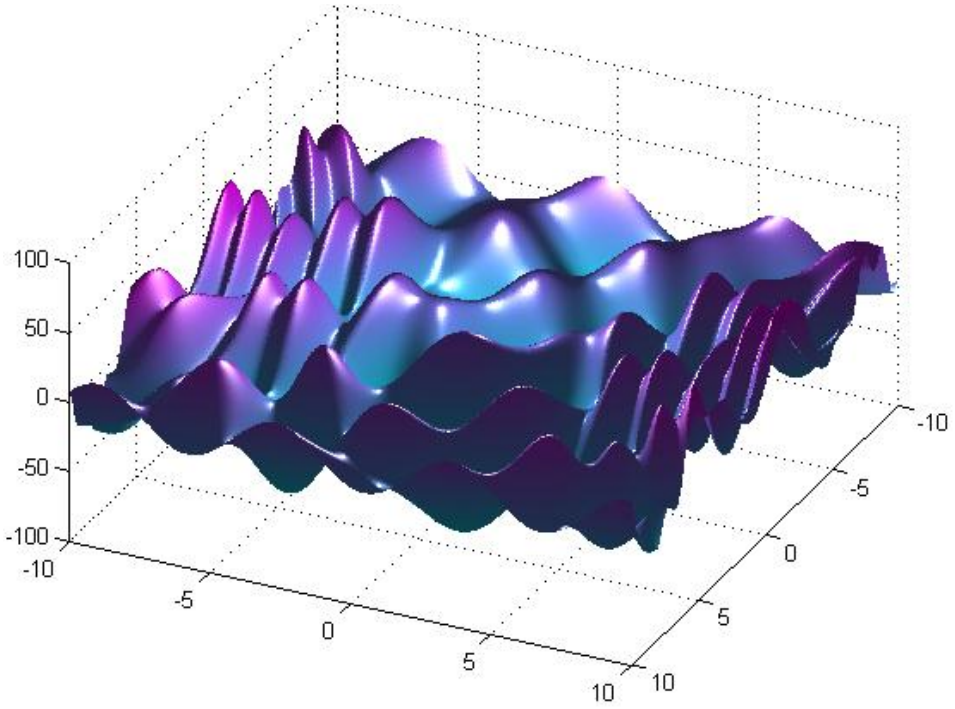
ding.
r too
rbon

自2015年面世以来，IM^{2}ODE成功预言的材料已发表SCI论文7篇，包括功能性块体材料、二维材料、团簇等。

Future Works



Global Optimization

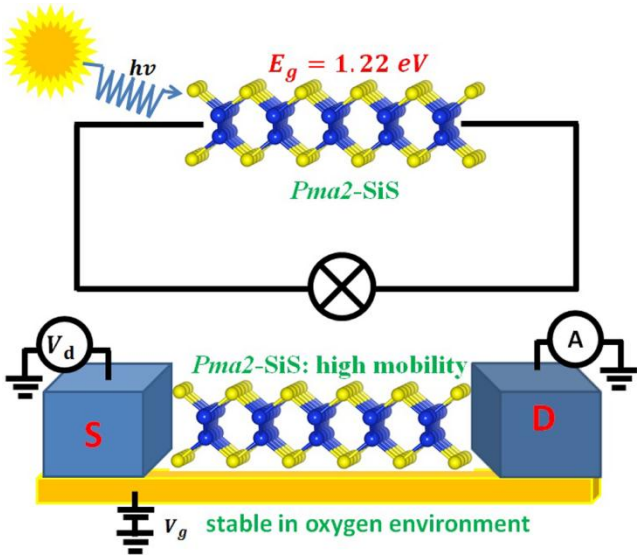


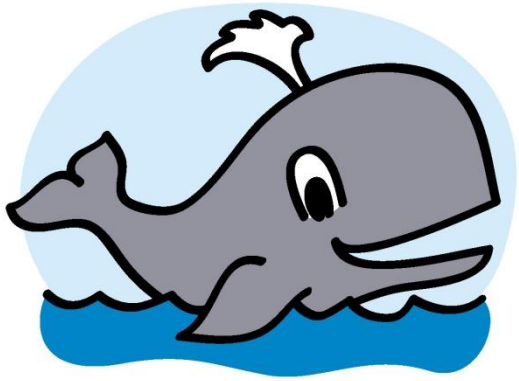
We believe that the number of functional meta-stable compounds which human beings can synthesis is infinity. And IM^{2}ODE offers some way to search for useful meta-stable states for further studying.

∞

Conclusion

- We developed a powerful tool, **IM^{2}ODE**, to search for materials with desired properties.
- **Five carbon allotropes** are found to be **good solar cell absorbers**.
- The predicted **SiS systems** have both **good optical property** and **high carrier mobility**.





Thank you!