Introduction to Tight-Binding

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Outline

- Theoretical Part

- Common approximations in tight-binding

- Practical case : Band Structure (Graphene)

- Practical case : Optical Spectrum (Bulk Silicon)

- *Ab initio* tight-binding

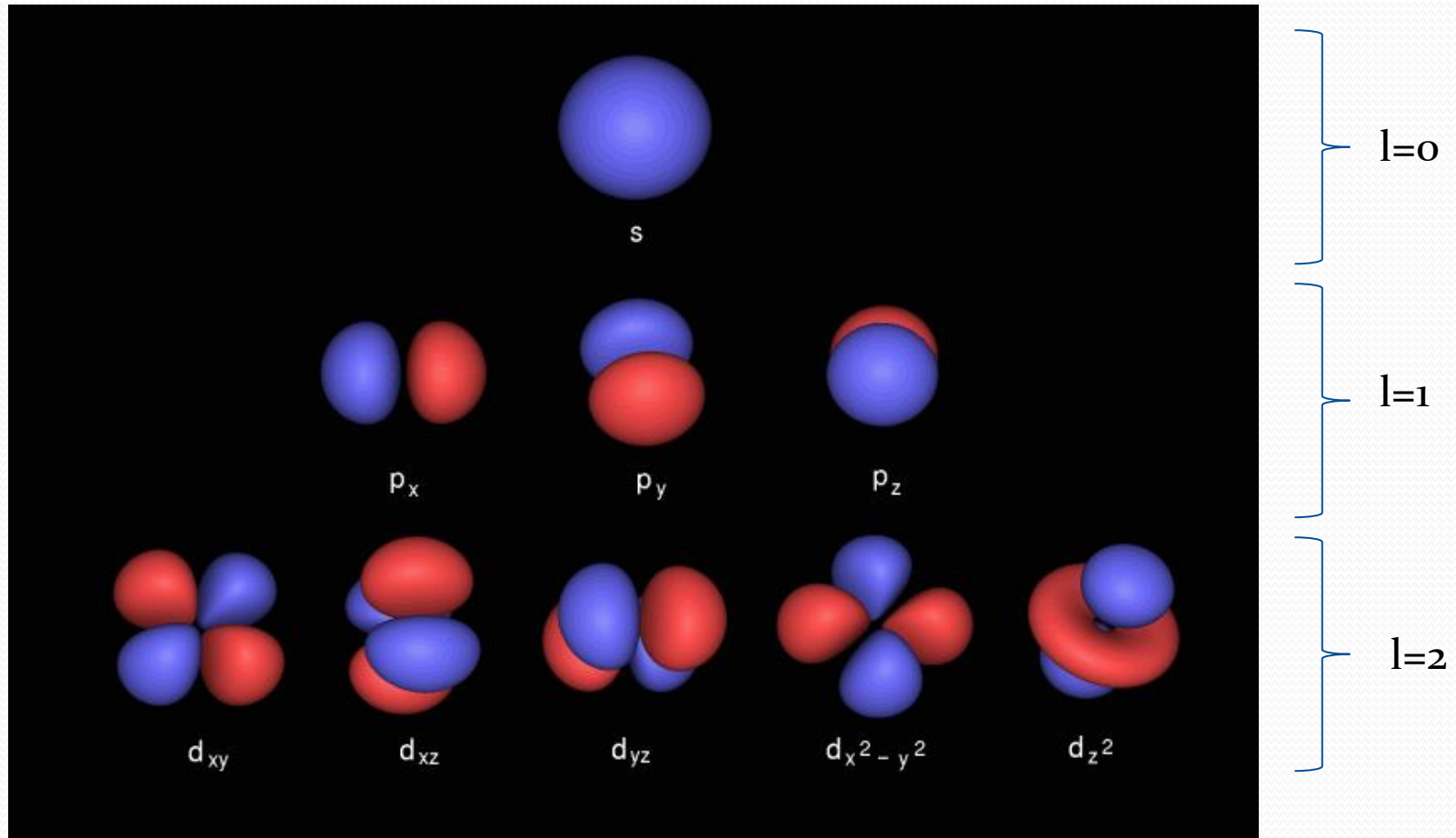- Going further

Theoretical Part

Linear Combination of Atomic Orbitals(LCAO)

Originally proposed as *ab initio* technique for computing electronic properties from atomic wave functions [1]

But very demanding in terms of calculations
=> relatively little success

[1] F.Bloch, Z. Phys. 52, 555 (1928)

Atomic orbitals (n=3)

l=0

l=1

l=2

From www.chemcomp.com/journal/molorbs.htm

Construction of LCAO Hamiltonian

We consider a set of atomic-like orbitals located on atomic positions $\vec{R_i}$

The LCAO idea is to express one-electron wave functions as a combination of atomic orbitals

$$\psi(\vec{r}) = \sum_{\alpha,i} c_{\alpha,i}\chi_\alpha(\vec{r} - \vec{R_i})$$

Orbital   Atom

Construction of LCAO Hamiltonian

Considering two orbitals α and β located on atomic sites at $\vec{R_i}$ and $\vec{R_j}$

- The Hamiltonian matrix elements are

$$H_{\alpha,\beta}(\vec{R_i}, \vec{R_j}) = \int d\vec{r}\chi_\alpha(\vec{r} - \vec{R_i})H\chi_\beta(\vec{r} - \vec{R_j})$$

- The overlap matrix elements are

$$S_{\alpha,\beta}(\vec{R_i}, \vec{R_j}) = \int d\vec{r}\chi_\alpha(\vec{r} - \vec{R_i})\chi_\beta(\vec{r} - \vec{R_j})$$

Construction of LCAO Hamiltonian

- We use the definition of Hamiltonian and overlapping matrices to Schrödinger equation

$$H\psi(\vec{r}) = E\psi(\vec{r})$$

- The equation we have to solve is

$$\sum_{\beta,j}[H_{\alpha,\beta}(\vec{R}_i, \vec{R}_j) - ES_{\alpha,\beta}(\vec{R}_i - \vec{R}_j)]c_{\beta,j} = 0$$

(generalized eigenvalue problem)

Using periodicity

- Considering a periodic crystal, on can define a new set of orbitals

Atom

$$\chi_{\alpha i \vec{k}}(\vec{r}) = \frac{1}{\sqrt{N}} \sum_{\vec{T}} e^{i \vec{k} \vec{T}} \chi_{\alpha}(\vec{r} - (\vec{T} + \vec{\tau}_i))$$

Atom position in unit cell

Orbital

Translation vector (to other unit-cells)

Using periodicity

- Considering a periodic crystal, on can define a new set of orbitals

$$\chi_{\alpha i \vec{k}}(\vec{r}) = \frac{1}{\sqrt{N}} \sum_{\vec{T}} e^{i\vec{k}\vec{T}} \chi_\alpha(\vec{r} - (\vec{T} + \vec{\tau}_i))$$

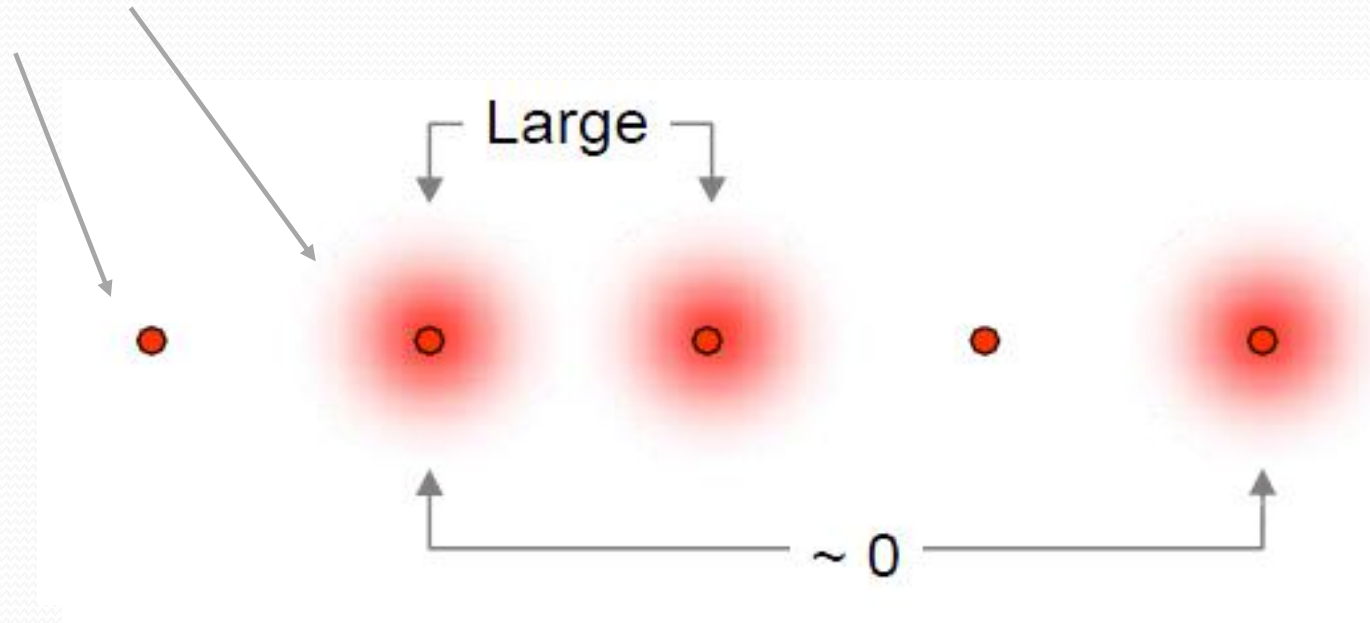$$\psi_{n\vec{k}}(\vec{r}) = \sum_{\alpha,i} c_{n,\alpha,i}(\vec{k}) \chi_{\alpha i \vec{k}}(\vec{r})$$

The Hamiltonian becomes

$$H_{\alpha,\beta,i,j}(\vec{k}) = \frac{1}{N} \sum_{\vec{T},\vec{T}'} e^{-i\vec{k}((\vec{T}+\vec{\tau}_i)-(\vec{T}'+\vec{\tau}_j))} H_{\alpha,\beta}(\vec{T} + \vec{\tau}_i, \vec{T}' + \vec{\tau}_j)$$

Tight-Binding approximation

The interaction between nearest neighbors is large and decrease quickly with the distance
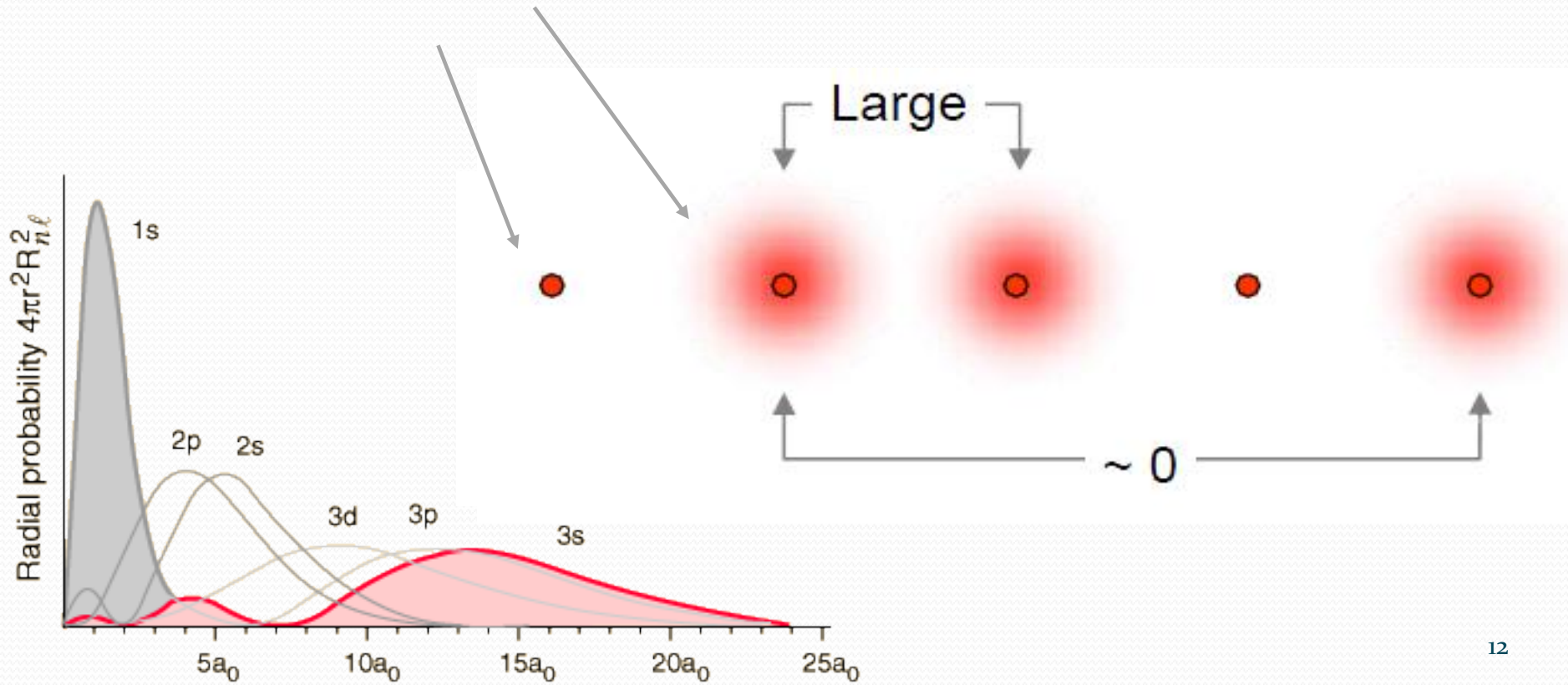
Atomic sites

Tight-Binding approximation

The interaction between nearest neighbors is large and decrease quickly with the distance

Atomic sites

Construction of the Tight-Binding Hamiltonian

The Tight-Binding Hamiltonian becomes a truncation of the sum in Hamiltonian expression

$$H_{\alpha,\beta,i,j}(\vec{k}) = \frac{1}{N} \sum_{\vec{T},\vec{T'}} e^{-i\vec{k}((\vec{T}+\vec{\tau_i})-(\vec{T'}+\vec{\tau_j}))} H_{\alpha,\beta}(\vec{T}+\vec{\tau_i}, \vec{T'}+\vec{\tau_j})$$

Limited by the tight-binding approximation

Common approximations in Tight-Binding

Common approximations in TB

- Nearest-neighbors

- Two centers approximation

- Orthogonal tight-binding

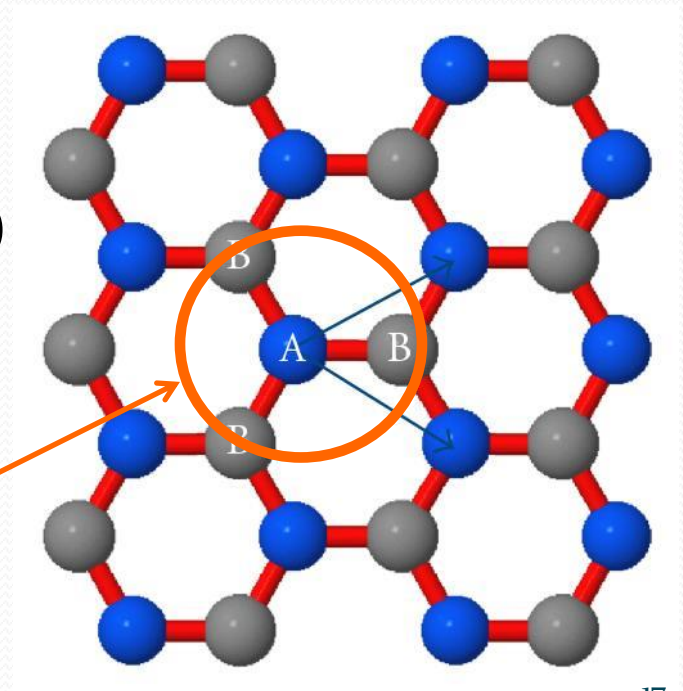- Semi-empirical Tight-Binding (SETB)

Nearest-Neighbors (N-N)

- One has to choose the extension of the interactions between neighbors

- In general

    first nearest-neighbors (1N-N)
    or third nearest-neighbors (3N-N)
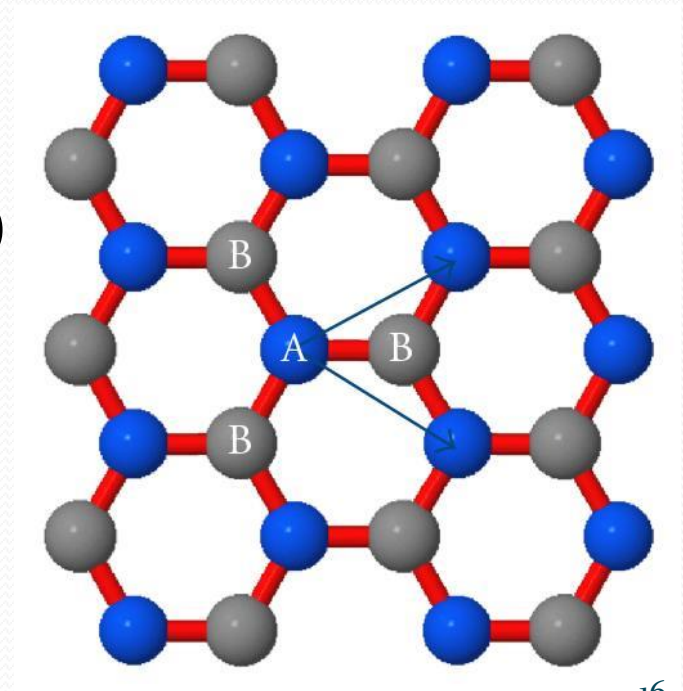
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1st neighbors

Nearest-Neighbors (N-N)

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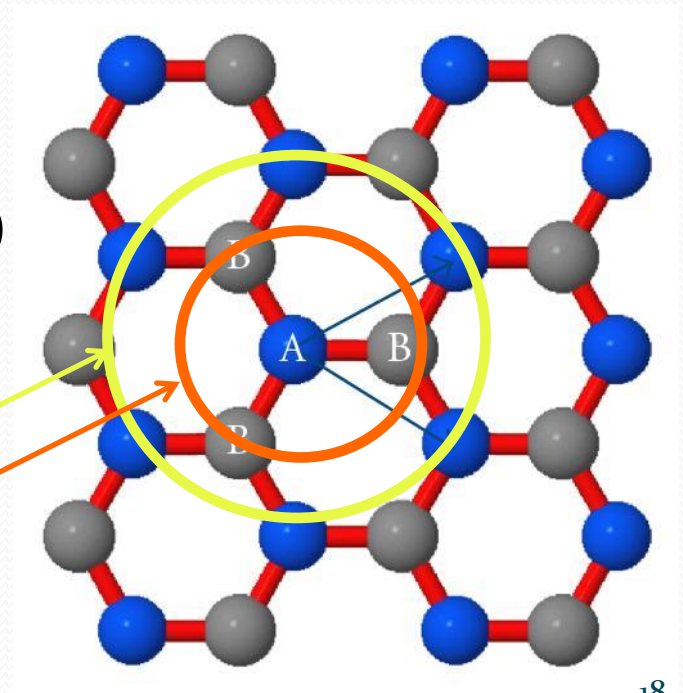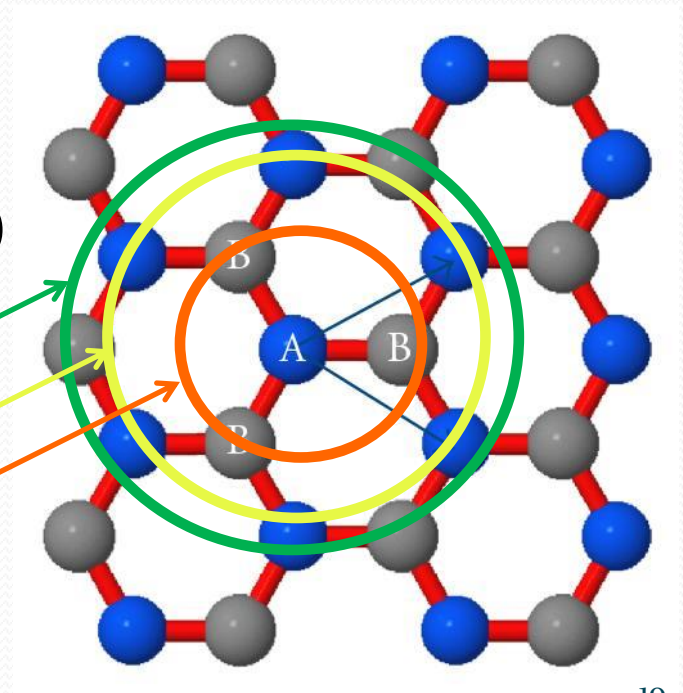

2nd neighbors
1st neighbors

Nearest-Neighbors (N-N)

- One has to choose the extension of the interactions between neighbors

- In general

  first nearest-neighbors (1N-N)
  or third nearest-neighbors (3N-N)

3rd neighbors

2nd neighbors

1st neighbors

Common approximations in TB

- Nearest-neighbors

- Two centers approximation

- Orthogonal tight-binding

- Semi-empirical Tight-Binding (SETB)

Two centers approximation

All overlap terms and Hamiltonian matrix elements involve only orbitals and potentials on two atomic sites

Sometime called *Slater-Koster two-centers approximation*[1] or *Two centers tight-binding model*

[1]*Phys. Rev 94, 1498 (1956)*

Two centers approximation

$$H_{\alpha\beta}\left(\mathbf{R}_{i,}\mathbf{R}_j\right)=\left\langle\varphi_\alpha\left(\mathbf{r}-\mathbf{R}_i\right)\middle|h\middle|\varphi_\beta\left(\mathbf{r}-\mathbf{R}_j\right)\right\rangle$$

$$=\left\langle\varphi_\alpha\left(\mathbf{r}-\mathbf{R}_i\right)\middle|-\frac{\hbar^2}{2m_0}\Delta_\mathbf{r}+\sum_k v_k\left(\mathbf{r}-\mathbf{R}_k\right)\middle|\varphi_\beta\left(\mathbf{r}-\mathbf{R}_j\right)\right\rangle$$

$$=\left\langle\varphi_\alpha\left(\mathbf{r}-\mathbf{R}_i\right)\middle|-\frac{\hbar^2}{2m_0}\Delta_\mathbf{r}\middle|\varphi_\beta\left(\mathbf{r}-\mathbf{R}_j\right)\right\rangle+\sum_k\left\langle\varphi_\alpha\left(\mathbf{r}-\mathbf{R}_i\right)\middle|v_k\left(\mathbf{r}-\mathbf{R}_k\right)\middle|\varphi_\beta\left(\mathbf{r}-\mathbf{R}_j\right)\right\rangle$$

Keeping only k=i or k=j give the two-center approximation otherwise it's a three center tight-binding model

Common approximations in TB

- Nearest-neighbors

- Two centers approximation

- Orthogonal tight-binding

- Semi-empirical Tight-Binding (SETB)

Orthogonal Tight-Binding

The overlapping is given, on the same atomic site

$$S_{\alpha\beta}(\mathbf{R}_i, \mathbf{R}_i) = \left\langle \varphi_\alpha(\mathbf{r} - \mathbf{R}_i) \middle| \varphi_\beta(\mathbf{r} - \mathbf{R}_i) \right\rangle = \delta_{\alpha\beta}$$

But for two different sites, we only have

$$S_{\alpha\beta}(\mathbf{R}_i, \mathbf{R}_j) = \left\langle \varphi_\alpha(\mathbf{r} - \mathbf{R}_i) \middle| \varphi_\beta(\mathbf{r} - \mathbf{R}_j) \right\rangle \approx \delta_{ij}\delta_{\alpha\beta}$$

Orthogonal Tight-Binding

The *Orthogonal Tight-Binding Approximation* is an approximation where your basis is chosen to have the property

$$S_{\alpha\beta}\left(\mathbf{R}_i, \mathbf{R}_j\right) = \left\langle \varphi_\alpha\left(\mathbf{r} - \mathbf{R}_i\right) \middle| \varphi_\beta\left(\mathbf{r} - \mathbf{R}_j\right) \right\rangle = \delta_{\alpha\beta}$$

This is not an *Orthogonalized Tight-Binding* where you transform your orbitals to be orthogonal (Löwdin theorem[1])

[1] P.O Löwdin, J. Chem. Phys. 18, 365 (1950)

Orthogonal Tight-Binding

The equation we have to solve is now an eigenvalue problem

$$\sum_{\beta,j}[H_{\alpha,\beta,i,j}(\vec{k}) - E_n(\vec{k})\delta_{\alpha,\beta}\delta_{i,j}]c_{n,\beta,j}(\vec{k}) = 0$$

$$\mathbf{H}\Psi = ES\Psi \longrightarrow \mathbf{H}\Psi = E\Psi$$

Common approximations in TB

- Nearest-neighbors

- Two centers approximation

- Orthogonal tight-binding

- Semi-empirical Tight-Binding (SETB)

Semi-empirical Tight-Binding

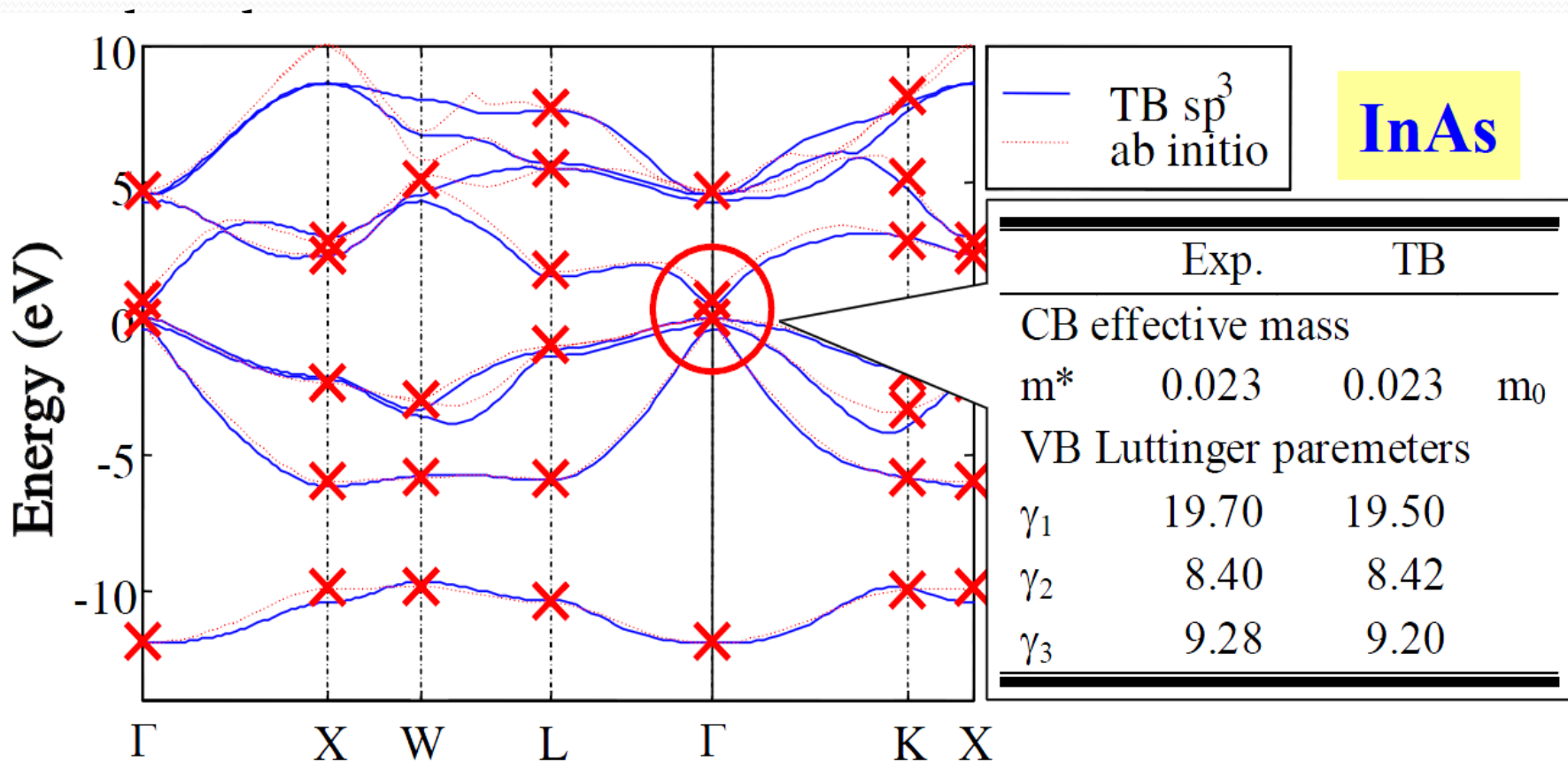Proposed by Slater and Koster   *Phys. Rev 94, 1498 (1956)*

The idea was to parameterize the matrix elements $H_{\alpha,\beta}(\vec{R}, \vec{R}')$ that become the tight-binding parameters

Advantage : orbitals are never defined !

Parameters are obtained by fitting or adjusting band structures

Fitting the parameters

Fitting from experiment or Hatree-Fock, DFT, GW, ... calculations

Ways for obtaining parameters

- Fitting (e.g. least square minimization) : Minimization of the discrepancy between the band structures

- Adjusting on band structure : The band structure have some points fixed

- Analytic expression of values of the band structure at some high symmetry point
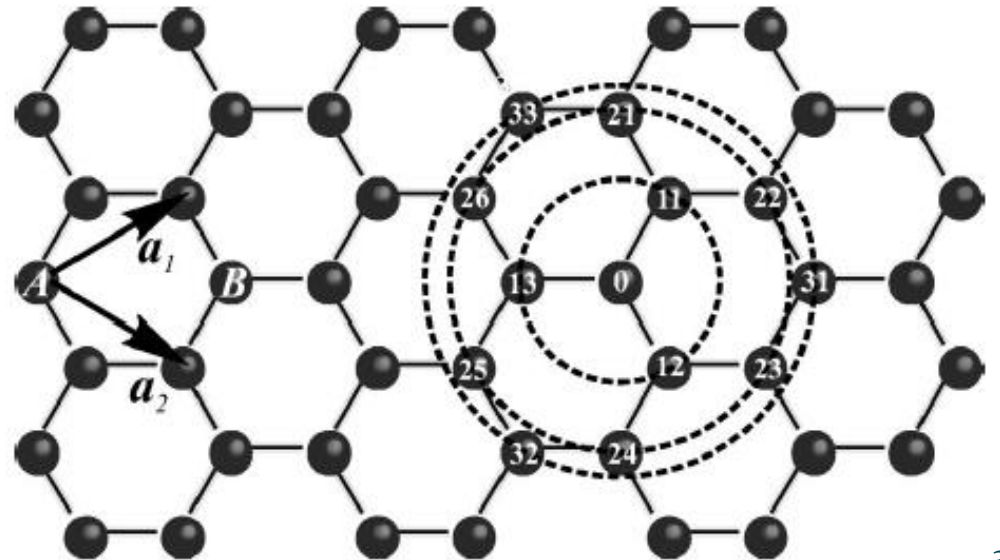
Practical case : Band Structure

Graphene

Application to Graphene

We start from the most general problem for p_z orbital only

$$\begin{vmatrix} H_{AA}(k) - E(k)S_{AA}(k) & H_{AB}(k) - E(k)S_{AB}(k) \\ H_{AB}^*(k) - E(k)S_{AB}^*(k) & H_{AA}(k) - E(k)S_{AA}(k) \end{vmatrix} = 0.$$

From PRB 66 035412 (2002)
Tight-binding description of graphene

Application to Graphene

The general solution is

$$E(k)^{\pm} = \frac{-(-2E_0+E_1) \pm \sqrt{(-2E_0+E_1)^2 - 4E_2E_3}}{2E_3}$$

with

$$E_0 = H_{AA}S_{AA}, \quad E_1 = S_{AB}H_{AB}^* + H_{AB}S_{AB}^*$$

$$E_2 = H_{AA}^2 - H_{AB}H_{AB}^* \quad E_3 = S_{AA}^2 - S_{AB}S_{AB}^*.$$

Graphene : Simple TB

We limit the sums to the first nearest-neighbors

$$H_{AB} = \frac{1}{N} \sum_{R_A} \sum_{R_B} e^{ik(R_B - R_A)} \langle \varphi_A(r - R_A) | H | \varphi_B(r - R_B) \rangle$$

$$= \gamma_0 (e^{ikR_{11}} + e^{ikR_{12}} + e^{ikR_{13}})$$

with

$$\gamma_0 = \langle \varphi_A(r - R_A) | H | \varphi_B(r - R_A - R_{1i}) \rangle \quad (i = 1,2,3),$$

Graphene : Simple TB

We limit the sums to the first nearest-neighbors

The Tight-Binding approximation

$$H_{AB} = \frac{1}{N} \sum_{R_A} \sum_{R_B} e^{ik(R_B - R_A)} \langle \varphi_A(r - R_A) | H | \varphi_B(r - R_B) \rangle$$

$$= \gamma_0 (e^{ikR_{11}} + e^{ikR_{12}} + e^{ikR_{13}})$$

with

The parameter

$$\gamma_0 = \langle \varphi_A(r - R_A) | H | \varphi_B(r - R_A - R_{1i}) \rangle \quad (i = 1, 2, 3),$$

Graphene : Simple TB

We limit the sums to the first nearest-neighbors

We apply the same to overlap matrix

$$S_{AB} = s_0 (e^{ikR_{11}} + e^{ikR_{12}} + e^{ikR_{13}})$$

Second parameter

with

$$s_0 = \langle \varphi_A(r - R_A) | \varphi_B(r - R_A - R_{1i}) \rangle \quad (i = 1, 2, 3)$$

Graphene : Simple TB

On-site energy

The solution becomes

$$E^{\pm}(k) = \frac{\varepsilon_{2p} \mp \gamma_0 \sqrt{f(k)}}{1 \mp s_0 \sqrt{f(k)}}$$

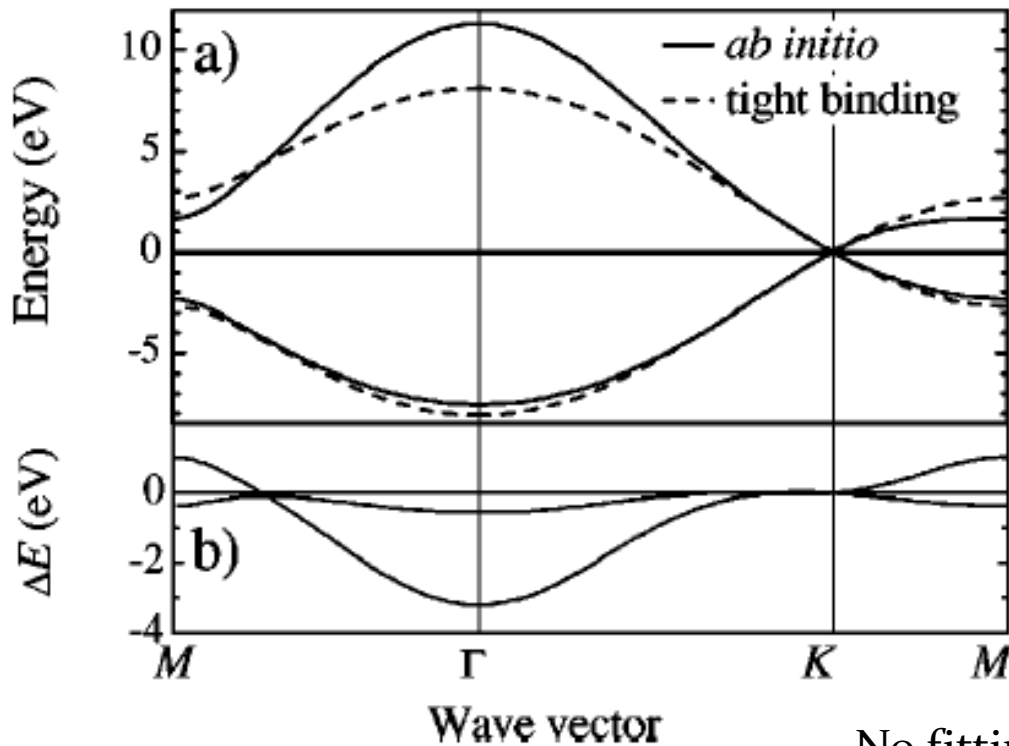# Graphene : Simple TB

On-site energy

$$E^{\pm}(k) = \frac{\varepsilon_{2p} \mp \gamma_0 \sqrt{f(k)}}{1 \mp s_0 \sqrt{f(k)}}$$

The solution becomes

a) — ab initio · · · tight binding

b)

$\gamma_0 = -2.7 eV$
$s_0 = 0 eV$
$\varepsilon_{2p} = 0 eV$

No overlapping
= Orthogonal TB

No fitting, only adjustment at K point

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Non orthogonal 3N-N TB

With a better choice of parameters (fitting) and considering overlapping and third-neighbors interaction
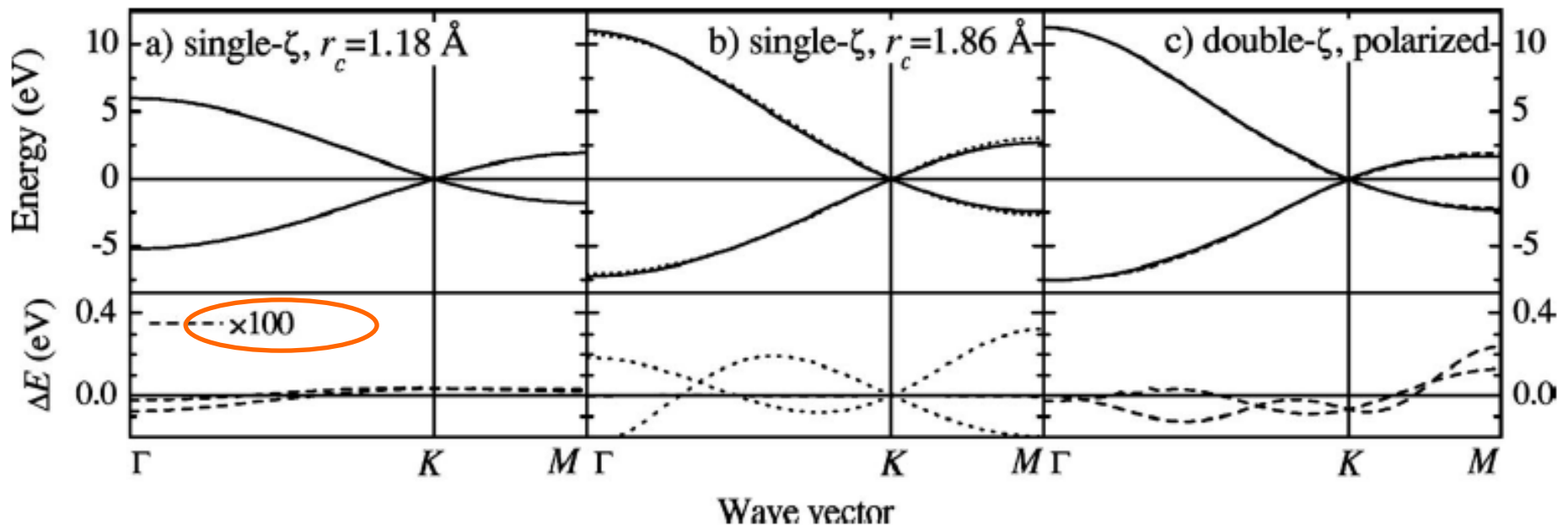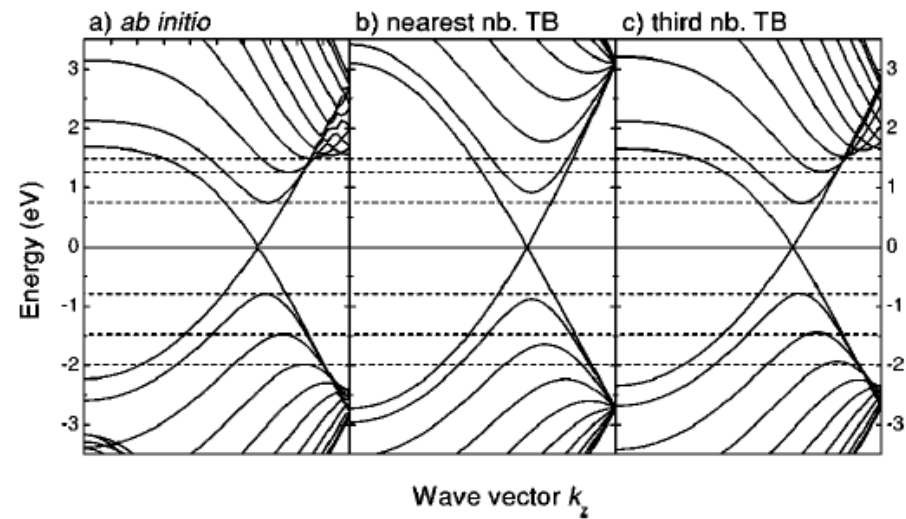
Application to Nanotubes

FIG. 4. Band structure of a (10,10) armchair nanotube. (a) *Ab initio* calculation. (b) Nearest-neighbor tight-binding calculation with $\gamma_0 = -2.7$ eV. (c) Third-nearest-neighbor tight-binding calculation with parameters obtained from a fit to the optical energy range; see Table I. The dashed lines denote *ab initio* calculated energies of the singularities in the density of states.
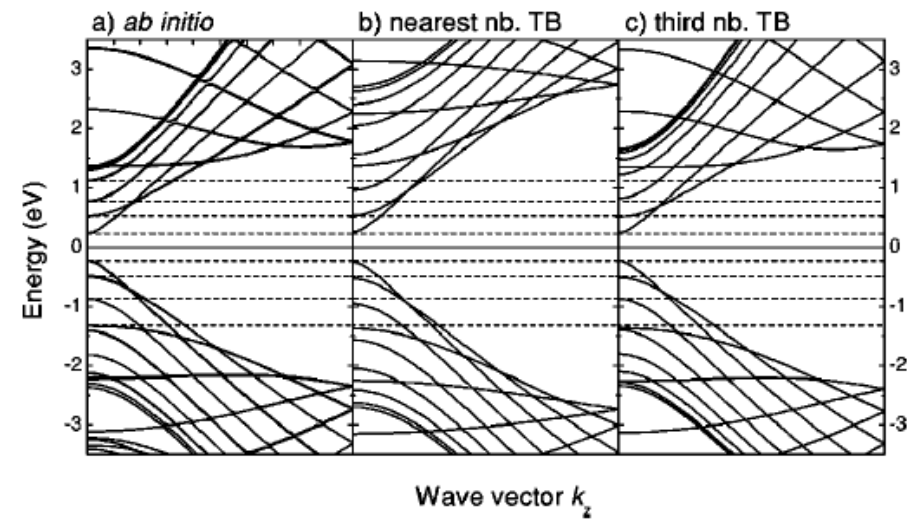
FIG. 5. Band structure of a (19,0) zigzag nanotube. (a) *Ab initio* calculation. (b) Nearest-neighbor tight-binding calculation with $\gamma_0 = -2.7$ eV. (c) Third-nearest-neighbor tight-binding calculation with parameters obtained from a fit to the optical energy range; see Table I. The dashed lines denote *ab initio* calculated energies of the singularities in the density of states.
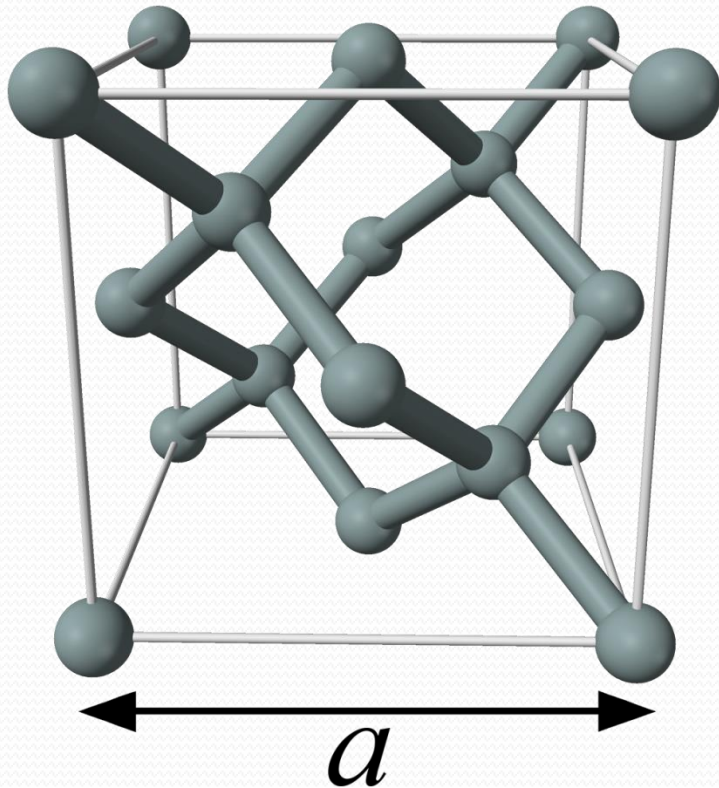
Application to Silicon

Approximation :

-Two center approximation

-Orthogonal TB

Which basis to choose ?
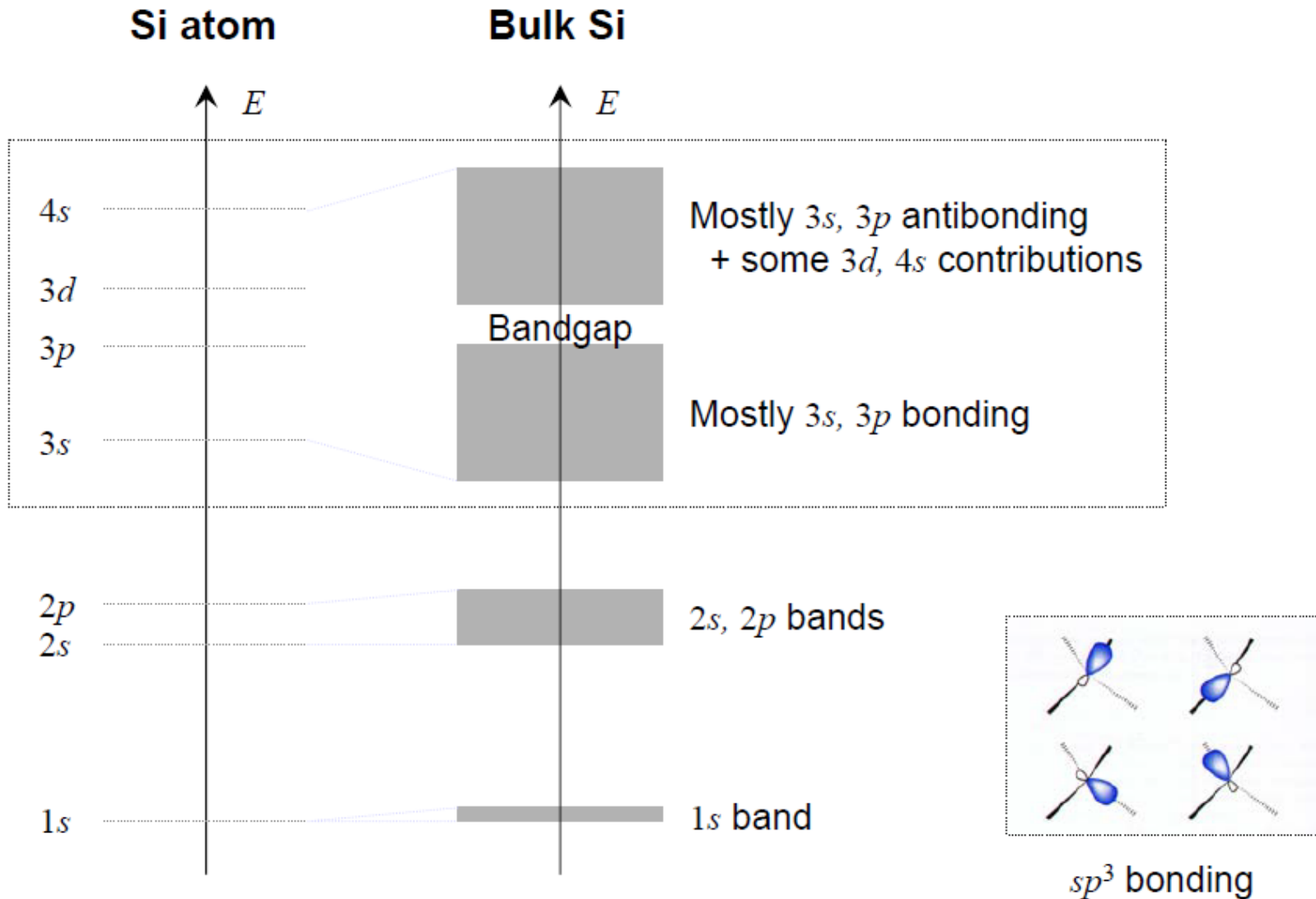
Si atom   **Bulk Si**

- $4s$
- $3d$
- $3p$
- $3s$

Mostly $3s$, $3p$ antibonding + some $3d$, $4s$ contributions

Bandgap

Mostly $3s$, $3p$ bonding

- $2p$
- $2s$

$2s$, $2p$ bands

- $1s$

$1s$ band

sp^3 bonding

From Y. M. Niquet presentation : Introduction of tight-binding description of semiconductor nanostructures

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Basis choice for Silicon

Depending on the number of parameters, one can describe correctly more or less bands

- sp3 basis :

Nearest-Neighbours	First	Third
Number of parameters	6	20

- sp3d5s* basis First Nearest-Neighbors :

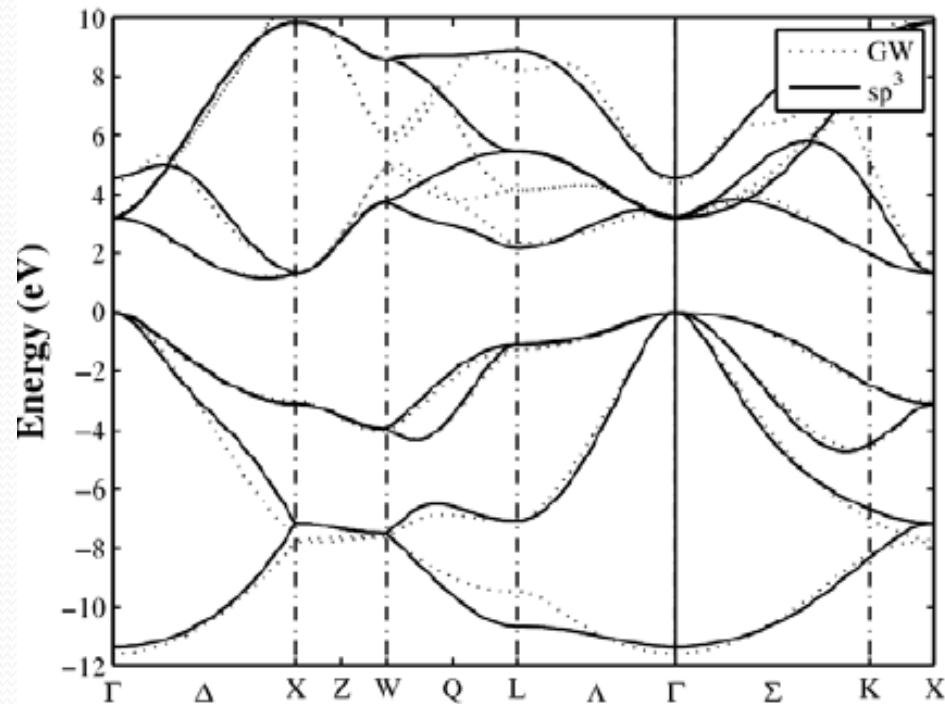| Nearest-Neighbours	First
Number of parameters	18

Basis choice for Silicon

- sp3 : quite accurate for the valence bands, somewhat less for the conduction band, especially at high energy

- sp3s* : s* orbital to mimic d orbitals

- sp3d5s* : accurate valence and first conduction bands

Band Structure

Third nearest neighbors orthogonal sp^3 model

Nearest neighbors orthogonal $sp^3d^5s^*$ model

Y. M. Niquet et al., Phys. Rev. B **62**, 5109 (2000)
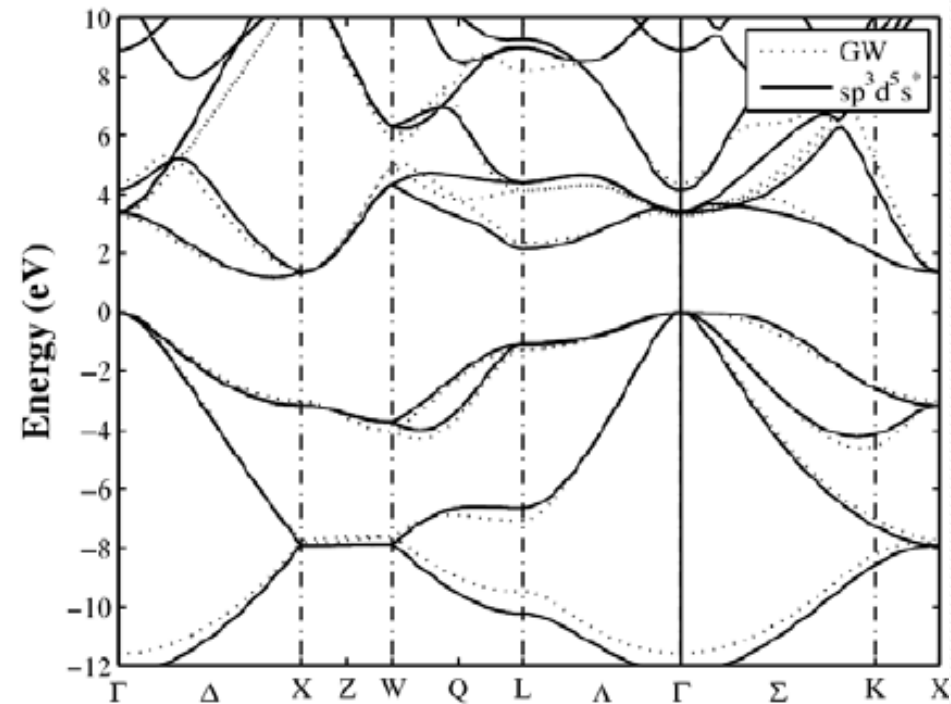
J. M. Jancu et al., Phys. Rev. B **57**, 6493 (1998)

Density of States

Comparison between TB and ab initio (ABINIT) density of states
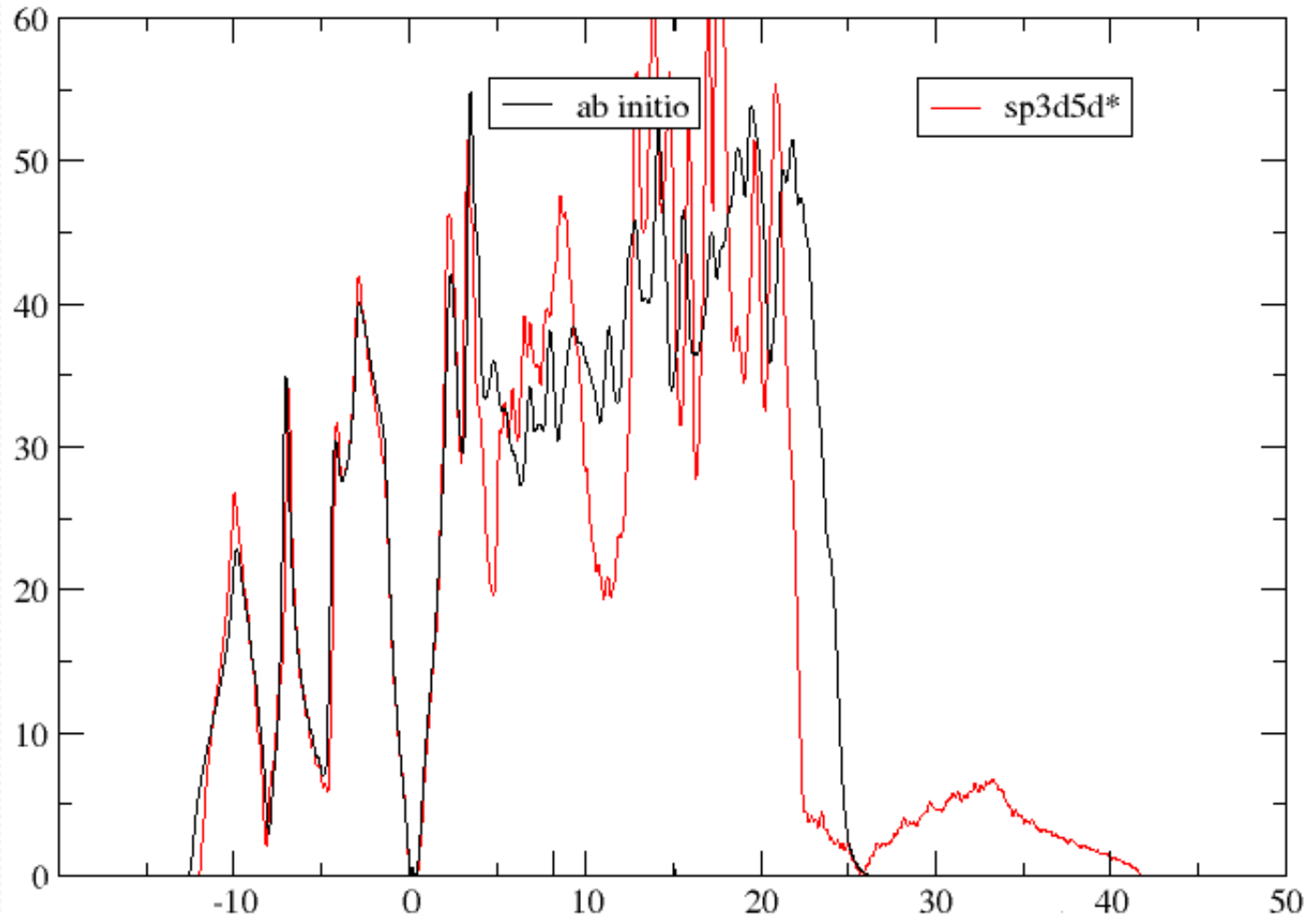
Computing momentum matrix elements

From k.p theory

$$H_{\vec{k}+\vec{q}} = H_{\vec{k}} + \vec{q}\vec{v} - \frac{i}{2}[\vec{q}\vec{r}, \vec{q}\vec{v}] + O[(\vec{q})^3]$$

$$H_{\vec{k}+\vec{q}} = H_{\vec{k}} + \nabla_{\vec{k}} H_{\vec{k}} \vec{q} + \frac{1}{2}\vec{q}\nabla_{\vec{k}}\nabla_{\vec{k}} H_{\vec{k}} \vec{q} + O[(\vec{q})^3]$$

Momentum matrix elements are obtained from Hamiltonien matrix elements directly

$$\vec{q}\vec{v} = \nabla_{\vec{k}} H_{\vec{k}} \vec{q}$$

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Computing momentum matrix elements

Hamiltonian matrix elements

$$< \lambda, \vec{k} | H(\vec{k}) | \mu, \vec{k} > = H_{\lambda,\mu}(\vec{k}) = \sum_{\vec{T}} e^{i\vec{k}.\vec{T}} t_{\lambda,\mu}(\vec{T})$$

Momentum matrix elements

$$< \lambda, \vec{k} | \nabla_{\vec{k}} H(\vec{k}) | \mu, \vec{k} > = \sum_{\vec{T}} (i\vec{T}) e^{i\vec{k}.\vec{T}} t_{\lambda,\mu}(\vec{T})$$

and $\quad \vec{q}\vec{v} = \nabla_{\vec{k}} H_{\vec{k}} \vec{q}$

Computing momentum matrix elements

Hamiltonian matrix elements

$$< \lambda, \vec{k}|H(\vec{k})|\mu, \vec{k} >= H_{\lambda,\mu}(\vec{k}) = \sum_{\vec{T}} e^{i\vec{k}.\vec{T}} t_{\lambda,\mu}(\vec{T})$$

Tight-Binding Parameters

Momentum matrix elements

$$< \lambda, \vec{k}|\nabla_{\vec{k}}H(\vec{k})|\mu, \vec{k} >= \sum_{\vec{T}} (i\vec{T}) e^{i\vec{k}.\vec{T}} t_{\lambda,\mu}(\vec{T})$$

and $\vec{q}\vec{v} = \nabla_{\vec{k}}H_{\vec{k}}\vec{q}$

Linear Spectrum

TB Parameters : PRB 79 245201 (2012)

GW – RPA : PRB 69 155112 (2004)

Legend:
- GW
- sp3d5s*

Y-axis: $\text{Im}\{\varepsilon_M\}$

X-axis: ω (eV)

Linear Spectrum

TB Parameters : PRB 79 245201 (2012)

GW - RPA : PRB 69 155112 (2004)

GW

sp3d5s*

Elapsed Time : 0.205s
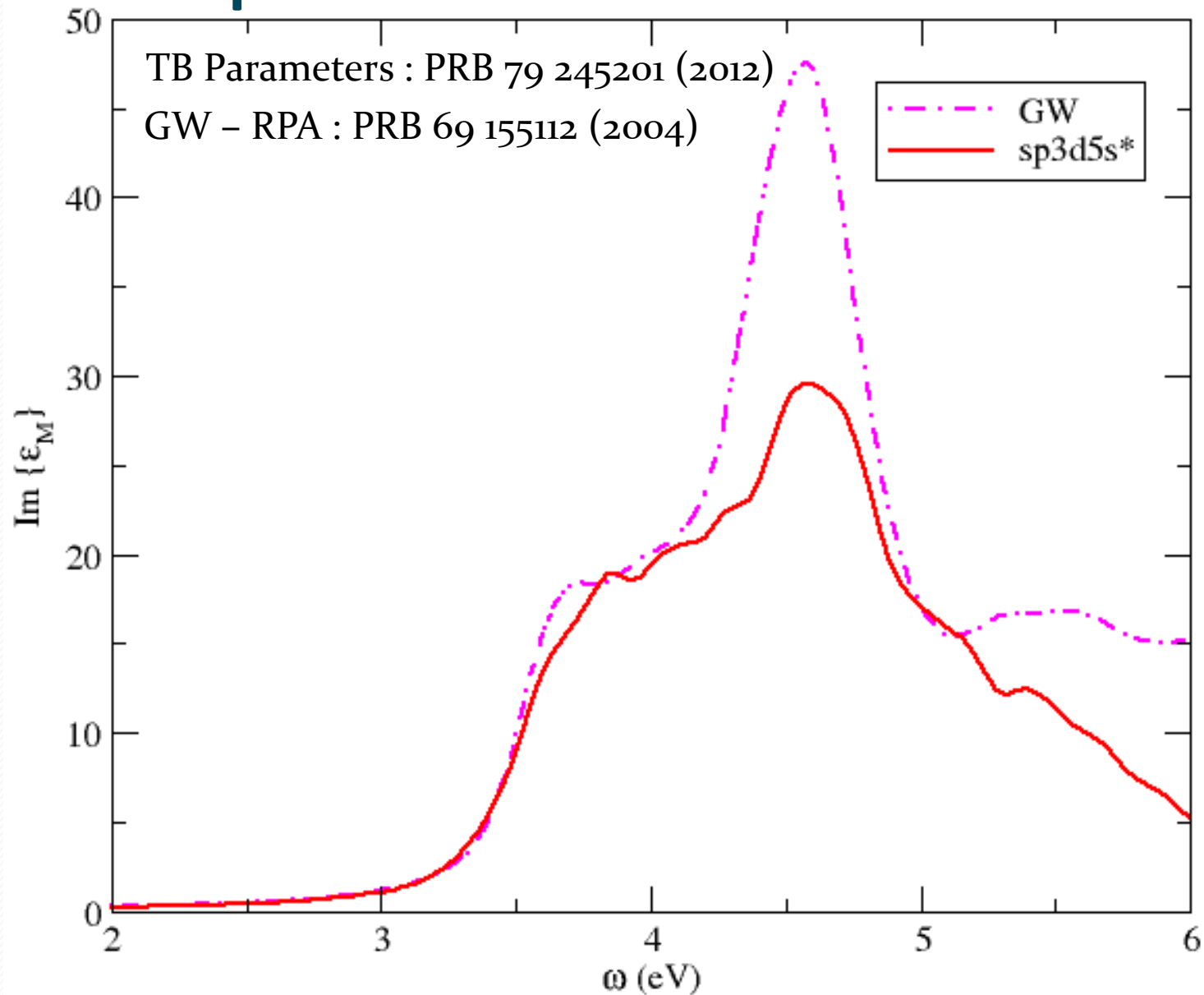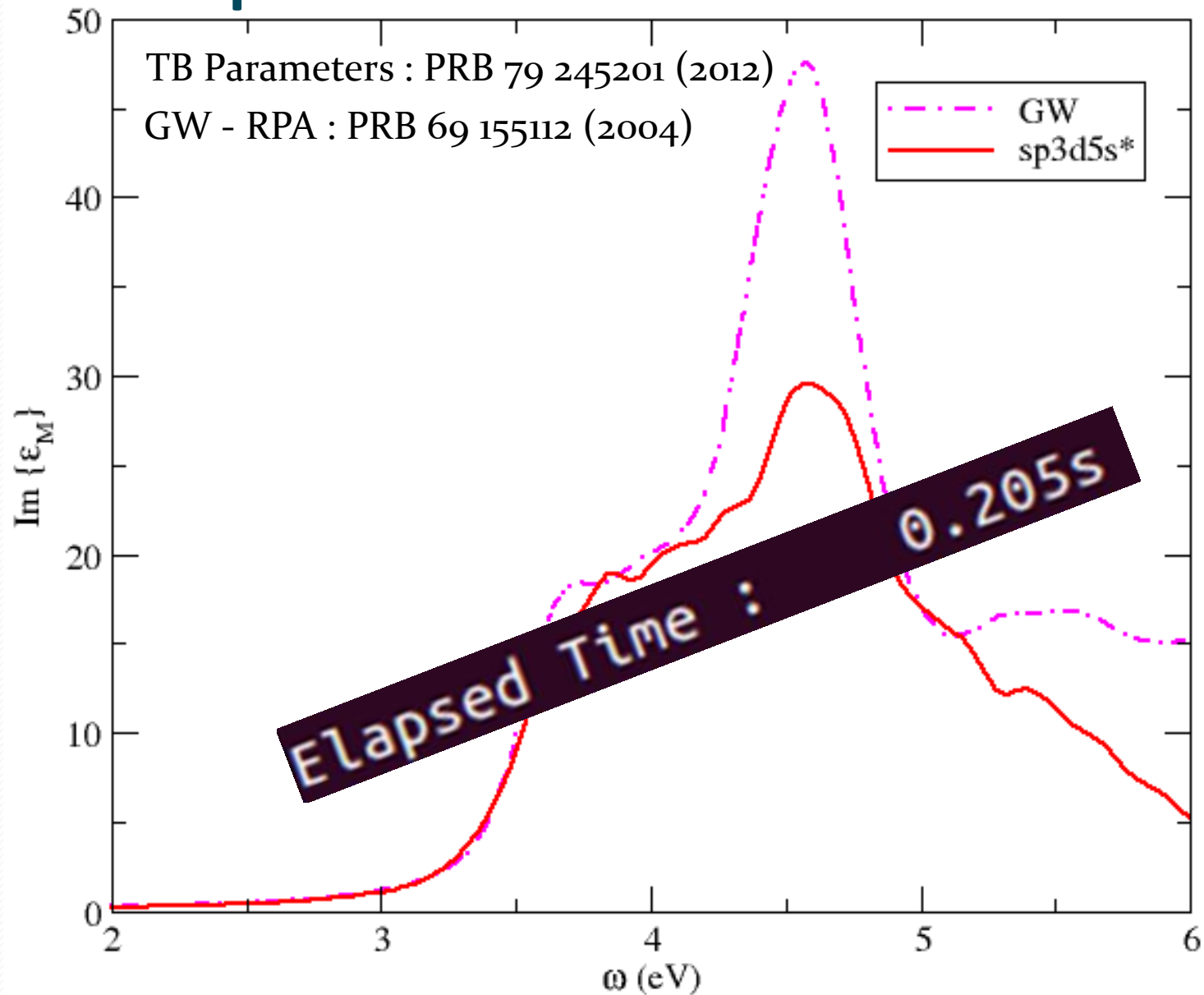
Conclusion from linear spectrum

- Tight-Binding is good for ground state properties :
Good for band structures (e.g. Silicon or Graphene)

- Not really accurate for computing spectra

Inclusion of Local-Field and excitonic effects

Possible in (semi-empirical) tight-binding

- Local Field effects with a Semi-Empirical Tight-Binding

C. Delerue, M. Lannoo, and G. Allan, Phys. Rev. B 56, 15306 (1997)

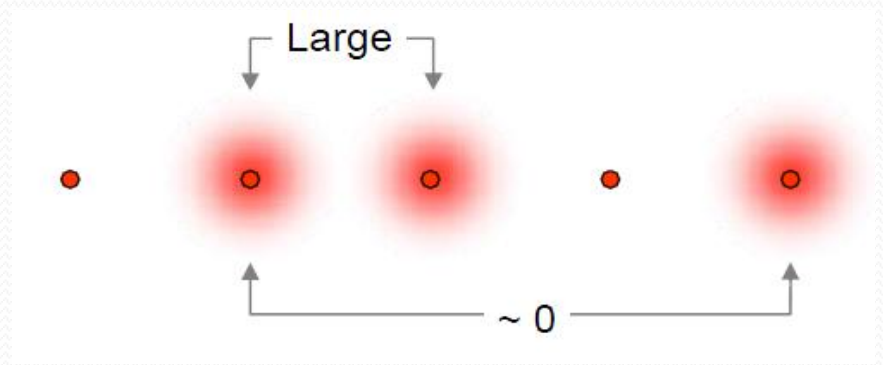- Excitonic effects via BSE

J.Jiang et al., Phys. Rev. B 75, 035407 (2007)

Ab initio tight-binding

Ab initio tight-binding

One can define the orbitals rather than using parameterization

Orbitals can be

- analytic functions, *e.g.* gaussian-type orbitals

- from DFT calculation, *e.g.* from atomic Kohn-Sham calculations with PBE exchange-correlation functional JCP 33, 1165 (2012)

Improving the tight-binding model

New Tight-Binding schemes

- Linear-muffin-tin orbitals tight-binding (LMTO-TB) [1]
- Hartree-Fock-based TB [2]
- *ab initio* multicenter TB [3]
- DF-based TB (DFTB) [4]
- Self-consistent charge DFTB [5]

1 - Phys. Rev. Lett. **53**, 2571 (1984) 2 - Phys. Rev. B **44**, 6169 (1991)
3 - Phys. Rev. B **40**, 3979 (1989) 4 - Phys. Rev. B **51**, 12 947 (1995)
5 – Phys. Rev. B 58, 7260 (1998)

Density Functional based Tight-Binding (DFTB)

DFT equations + tight-binding approximation

Interest : quite accurate and really quicker than DFT

Formamide	DFTB	SCC-DFTB	DFT-LSD (Ref. 39)	Expt. (Ref. 39)
C=O	1.296	1.224	1.223	1.193
C−N	1.296	1.382	1.358	1.376
N−H	1.003	0.996	1.022	1.002
C−H	1.130	1.131	1.122	1.102
OCN	127.0	125.5	124.5	123.8

From Phys. Rev. B 58, 7260 (1998)

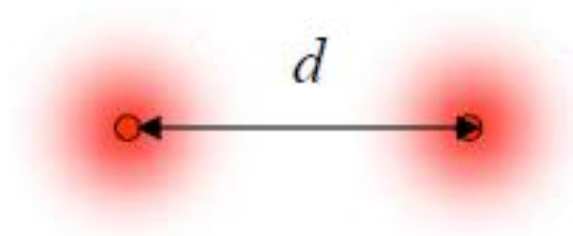# Going further

Deformation potentials in SETB

Spin-orbit coupling in SETB

Examples of applications

Deformation potentials in SETB

Generalized Harrison's law :

$$V_{ss\sigma}(d) = V_{ss\sigma}(d_0)\left(\frac{d_0}{d}\right)^{\alpha_{ss\sigma}}$$

Harrison's law[1] : $\alpha_{ss\sigma} = 2$

This is the value for a free electron

[1] Phys. Rev. B 10, 1516 (1974)

Deformation potentials in SETB

Examples of applications :

- Strained crystal

- Atomic displacements

- Surface reconstruction

Spin-orbit coupling in SETB

- Spin orbit coupling can be introduce (on p orbitals for instance, under approximations)

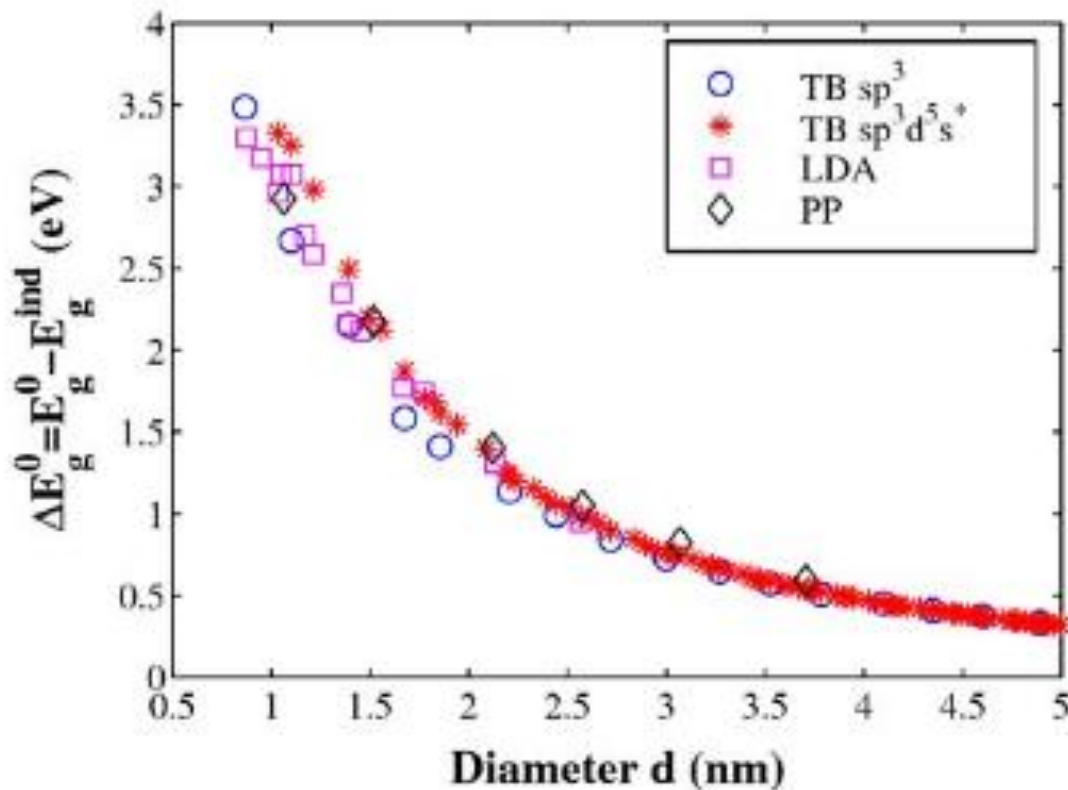$$H_i^{so} \approx \lambda_i \mathbf{L}_i \cdot \mathbf{S}$$

$$H_i^{so} = -\lambda_i \begin{bmatrix} 0 & -i & 0 & 0 & 0 & 1 \\ i & 0 & 0 & 0 & 0 & -i \\ 0 & 0 & 0 & -1 & i & 0 \\ 0 & 0 & -1 & 0 & i & 0 \\ 0 & 0 & -i & -i & 0 & 0 \\ 1 & i & 0 & 0 & 0 & 0 \end{bmatrix} \begin{array}{l} \left| p_x \uparrow \right\rangle \\ \left| p_y \uparrow \right\rangle \\ \left| p_z \uparrow \right\rangle \\ \left| p_x \downarrow \right\rangle \\ \left| p_y \downarrow \right\rangle \\ \left| p_z \downarrow \right\rangle \end{array}$$

Spin-orbit coupling parameter

Sol. State Comm. 62, 399 (1987)

Examples of applications

- Comparison between TB and LDA

$$\Delta E_g^0 = E_g(d) - E_g(\text{bulk Si})$$

where $E_g(\text{bulk Si}) = 1.17$ eV

From Y. M. Niquet presentation : Introduction of tight-binding description of semiconductor nanostructures
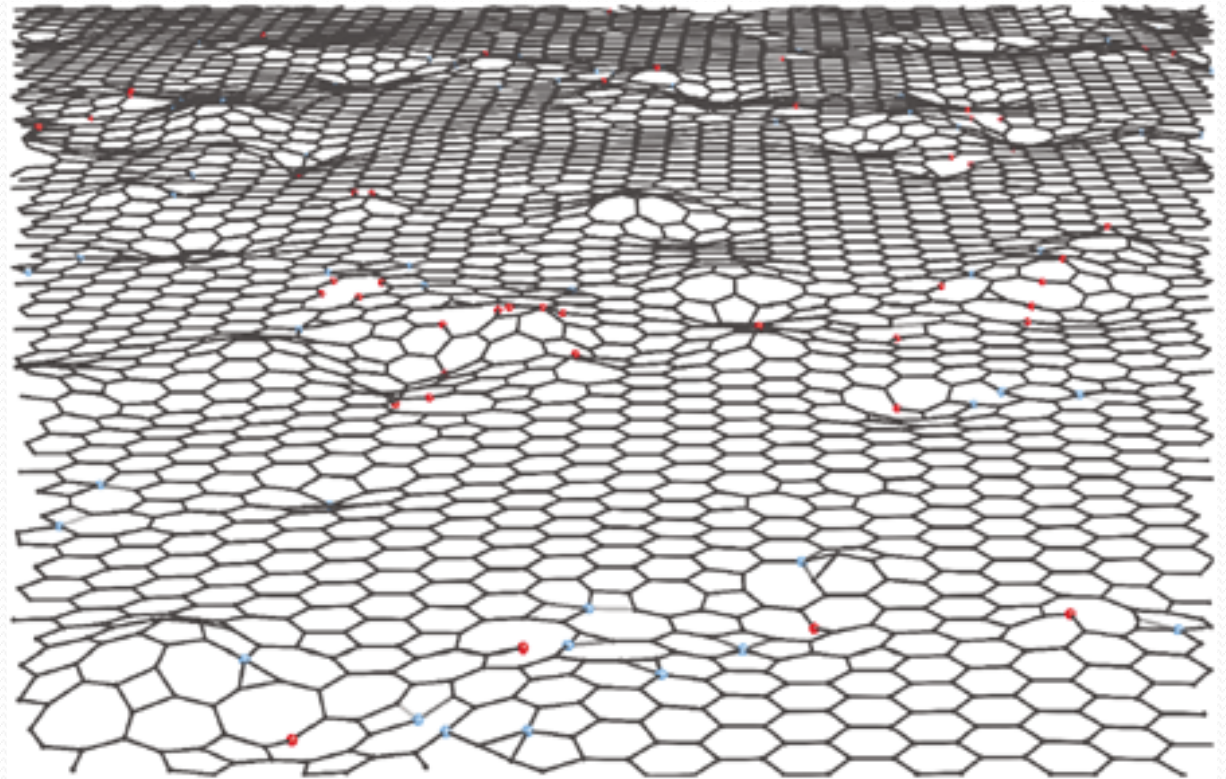
Examples of applications

Coallescence of nanotubes
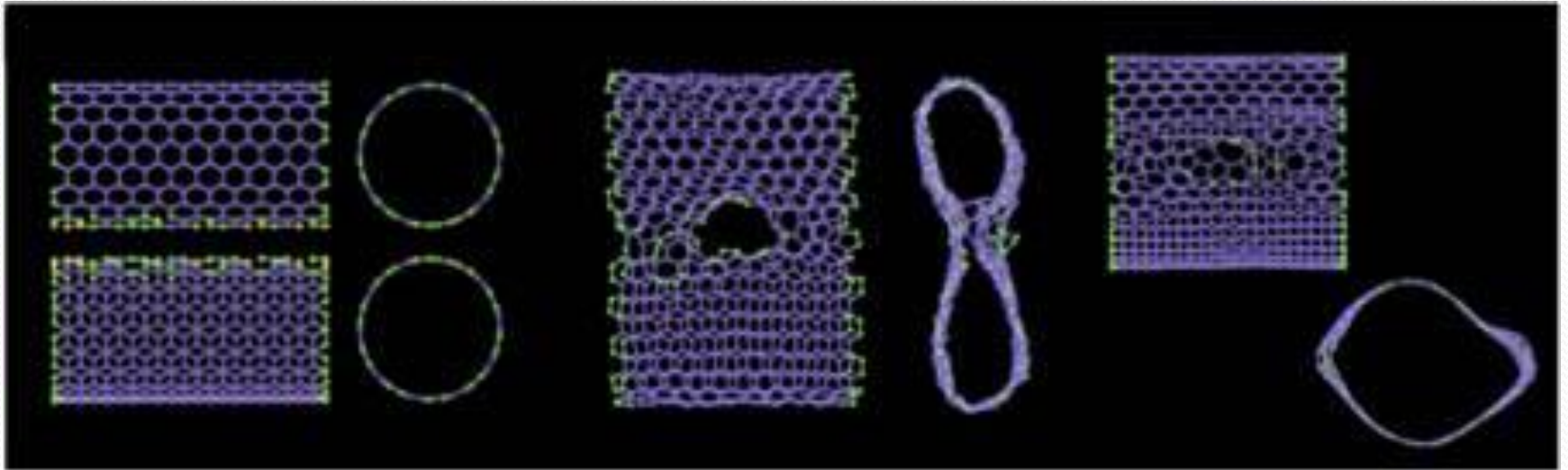
M. Terrones, H. Terrones, F. Banhart, J.-C. Charlier, and P. M. Ajayan
Science May 19 2000 : 1226-1229.

Examples of applications

- Highly defective graphene (2 millions of atoms)

Nano Research

Thank you for your attention !

Sp3 3N-N

$E_{ss}(000)$	-6.17334	eV	$E_{ss}(111)$	-1.78516	eV
$E_{xx}(000)$	2.39585	eV	$E_{sx}(111)$	0.78088	eV
Δ	0.04500	eV	$E_{xx}(111)$	0.35657	eV
			$E_{xy}(111)$	1.47649	eV
$E_{ss}(220)$	0.23010	eV	$E_{ss}(311)$	-0.06857	eV
$E_{sx}(220)$	-0.21608	eV	$E_{sx}(311)$	0.25209	eV
$E_{sx}(022)$	-0.02496	eV	$E_{sx}(113)$	-0.17098	eV
$E_{xx}(220)$	0.02286	eV	$E_{xx}(311)$	0.13968	eV
$E_{xx}(022)$	-0.24379	eV	$E_{xx}(113)$	-0.04580	eV
$E_{xy}(220)$	-0.05462	eV	$E_{xy}(311)$	-0.03625	eV
$E_{xy}(022)$	-0.12754	eV	$E_{xy}(113)$	0.06921	eV

Y. M. Niquet et al., Phys. Rev. B **62**, 5109 (2000)

sp3d5s* basis

PRB 69, 115201 (2004)

Parameter	Si	Ge
E_s	-2.15168	-1.95617
E_p	4.22925	5.30970
E_{s^*}	19.11650	19.29600
E_d	13.78950	13.58060
λ	0.01989	0.10132
$ss\sigma$	-1.95933	-1.39456
$s^*s^*\sigma$	-4.24135	-3.56680
$ss^*\sigma$	-1.52230	-2.01830
$sp\sigma$	3.02562	2.73135
$s^*p\sigma$	3.15565	2.68638
$sd\sigma$	-2.28485	-2.64779
$s^*d\sigma$	-0.80993	-1.12312
$pp\sigma$	4.10364	4.28921
$pp\pi$	-1.51801	-1.73707
$pd\sigma$	-1.35554	-2.00115
$pd\pi$	2.38479	2.10953
$dd\sigma$	-1.68136	-1.32941
$dd\pi$	2.58880	2.56261
$dd\delta$	-1.81400	-1.95120

$dd\sigma$

$dd\pi$

$dd\delta$

$pd\sigma$

$pd\pi$

$pp\sigma$

$pp\pi$

$sd\sigma$

$sp\sigma$

$ss\sigma$

CMS 47, 237 (2009)

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