Applications of the Quasiparticle self-consistent GW method to semiconductors:

towards first-principles-based device simulations

Takao Kotani, Tottori-u

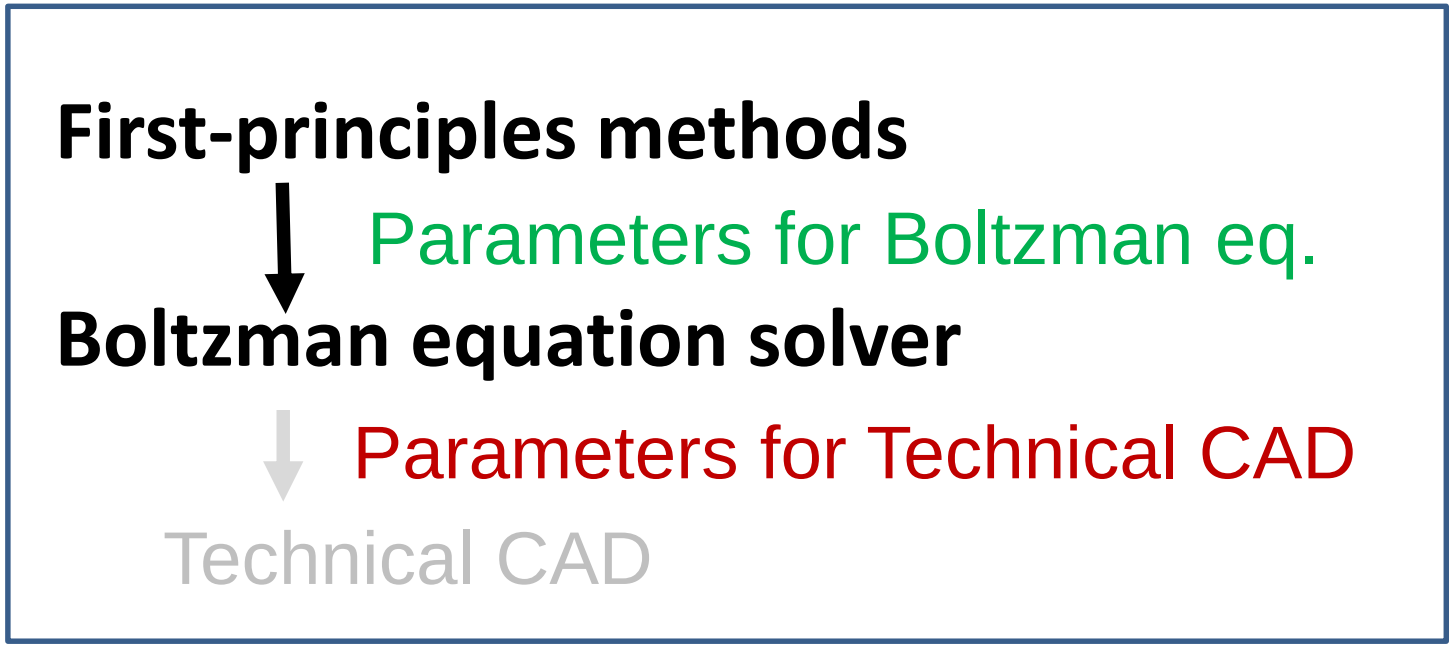
Collaboration:

- QSGW survey :
  K.Sato, D.Deguchi（Material, Osaka-u),H.Kino(NIMS)
- Power device :
  Y.Kamakura, R Fujita, K Konaga, Y Ueoka, N Mori（EE, Osaka-u)
- Superlattice for IR sensor :
  J.Otsuka,T.Kato, A.Sawamura（Sumitomo E.,LTD)

***ecalj* package(https://github.com/tkotani/ecalj/)**

Acknowledgement Dr.T.Miyake for a Wannier code in ecalj.

Our Purpose



TCAD requires many parameters.

It is necessary to go through Boltzman equation.

(band structures, band offsets, impact ionization rate, impurity level, cross, relaxation time…).

Problems

- **DFT(LDA) do not give good energy bands**

  → Use GW method (or QSGW method).

- **Varieties of physical quantities required.**

  → We still have to develop kinds of methods.

  Very recently, the mobility of GaAs is calculated
  J.J.Zhou et al, Phys. Rev. B 94, 201201(R) (2016)

- **Too much computational time**

  → parallel computing, Algorism.

  → Mapping to a tight-binding model

OUTLINE

1. QSGW gives good energy bands.

   (QSGW=Quasiparticle self-consistent GW)

2. Type-II Superlattice $(InAs)_n/(GaSb)_n$

   We can calculate energy bands and band offsets.

   Make a tight-binding model based on the QSGW results.

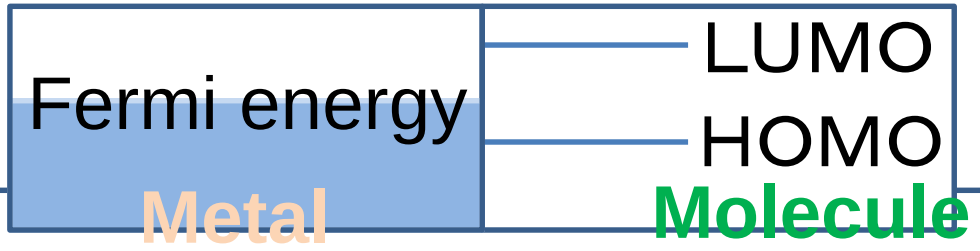
3. Power devices: C and 4h-SiC

   Energy bands and impact ionization.

Problems in DFT

Not gives a good independent-particle picture.

1. Excitation energies of semiconductors:
   Band gap, effective mass
2. Transition metal oxides:
   Magnetic order (moments), Relative positions of 3d and O(2p).
3. Molecules on Metal
   LUMO-HOMO/Fermi energy



How to solve these problems?

"GW approximation"

better one is QSGW (self-consistent version of GW)

1. QSGW gives good energy bands.

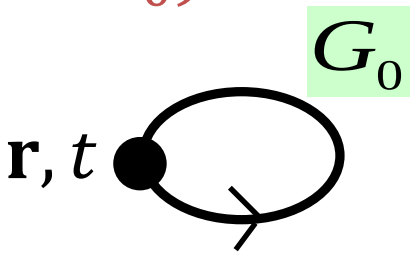
GW approximation: from H_0 to $H(\omega)$

We can perform GW from any H_0 , for example, H^{LDA}

$$H = H_0 + (H - H_0)$$

1. $$H_0 = -\frac{\nabla^2}{2} + V_{\mathrm{eff}}(\mathbf{r},\mathbf{r}') \Leftrightarrow G_0 = \frac{1}{\omega - H_0}$$

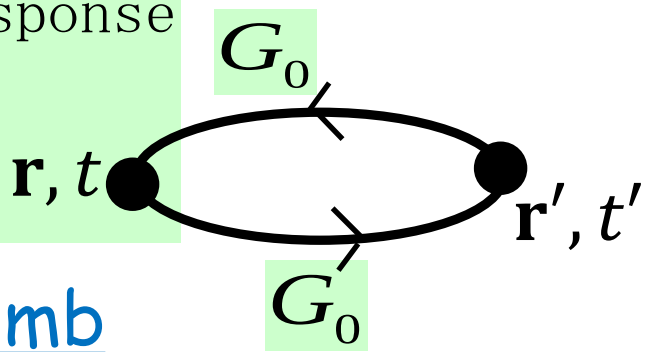
$$n(\mathbf{r}) = G_0(\mathbf{r},\mathbf{r},0-)$$



2. $\Pi = -iG_0 \times G_0$ Proper Polarization function

Π means non-interacting density-response

$$\Pi(\mathbf{r},\mathbf{r}',t-t') = \frac{\delta n(\mathbf{r},t)}{\delta V(\mathbf{r}',t')}$$



3. $$W = \varepsilon^{-1}v = (1 - \Pi v)^{-1} v$$

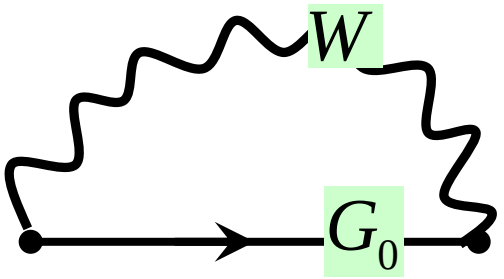
$$v(\mathbf{r},\mathbf{r}') = \frac{e^2}{|\mathbf{r} - \mathbf{r}'|}$$

Screened Coulomb $W(\mathbf{r},\mathbf{r}',\omega)$ in the RPA (Time-dependent Hartree approx. Sum of ring diagrams)

1. QSGW gives good energy bands.

4. $\Sigma = iG_0W$

Self-energy



(exchange-correlation effect)

Exchange effect + an electron moving in a polarizable medium.

Finally, GW gives

$$H(\mathbf{r},\mathbf{r}',\omega) = -\frac{\nabla^2}{2} + V^{\text{ext}}(\mathbf{r}) + V^{\text{H}}(\mathbf{r}) + \Sigma(\mathbf{r},\mathbf{r}',\omega)$$

GW gives $H_0 \rightarrow H(\omega)$

Many-body effect is down-folded into $H(\omega)$.

Quasiparticle equation,

$$H(\mathbf{r},\mathbf{r}',\varepsilon_i)\varphi_i(\mathbf{r}') = \varepsilon_i\varphi_i(\mathbf{r}),$$

determines QPs (not only eigenfunctions but also eigenvalues).

What is the QSGW?

Usual one-shot GW, only calculate

$$\varepsilon_i^{NEW} = \langle \varphi_i | H(\mathbf{r}, \mathbf{r}', \varepsilon_i) | \varphi_i \rangle$$

But… How to determine best H_0?

→**self-consistent perturbation.**

$H(\mathbf{r}, \mathbf{r}', \omega)$ → QP (eigenvalues and eigenfunctions).
We choose H_0 so that QP are reproduced well.

Self-consistency

$H_0 \rightarrow H(\mathbf{r}, \mathbf{r}', \omega) \rightarrow H_0 \rightarrow H(\mathbf{r}, \mathbf{r}', \omega) \rightarrow H_0 \rightarrow \ldots$

(Next page: $\Sigma(\mathbf{r}, \mathbf{r}', \omega) \rightarrow V_{\mathrm{xc}}(\mathbf{r}, \mathbf{r}')$. How?)

$$H_0 \to \{\varepsilon_i, \varphi_i\} \to \Sigma(\mathbf{r}, \mathbf{r}', \omega) \to V_{\mathrm{xc}}(\mathbf{r}, \mathbf{r}')$$

We use a simple average in usual QSGW as

$$V_{\mathrm{xc}}(\mathbf{r}, \mathbf{r}') = \sum_{i,j} \varphi_i(\mathbf{r}) \frac{\langle \varphi_i | \mathrm{Re}[\Sigma(\boldsymbol{r}, \boldsymbol{r}', \varepsilon_i) + \Sigma(\boldsymbol{r}, \boldsymbol{r}', \varepsilon_j)] | \varphi_j \rangle}{2} \varphi_j^*(\boldsymbol{r}')$$

(Re means Hermitian part)

This determines mapping

$$H_0 \to H(\mathbf{r}, \mathbf{r}', \omega) \to H_0 \to H(\mathbf{r}, \mathbf{r}', \omega)$$

Then $\{\varphi_i\}$ is a orthonormalized complete set.
Best one-particle picture. http://dx.doi.org/10.7566/JPSJ.83.094711

Advantage in QSGW

LDA（GGA）→ Homogeneous gas OK! （Physics）
Hartree-Fock → H-atom OK! （chemistry）

Truth is somewhere between LDA and HF.

➔ Hybrid methd（B3LYP, HSE…） $\frac{1}{4}\times \mathrm{HF} + \frac{3}{4}\times \mathrm{LDA}$

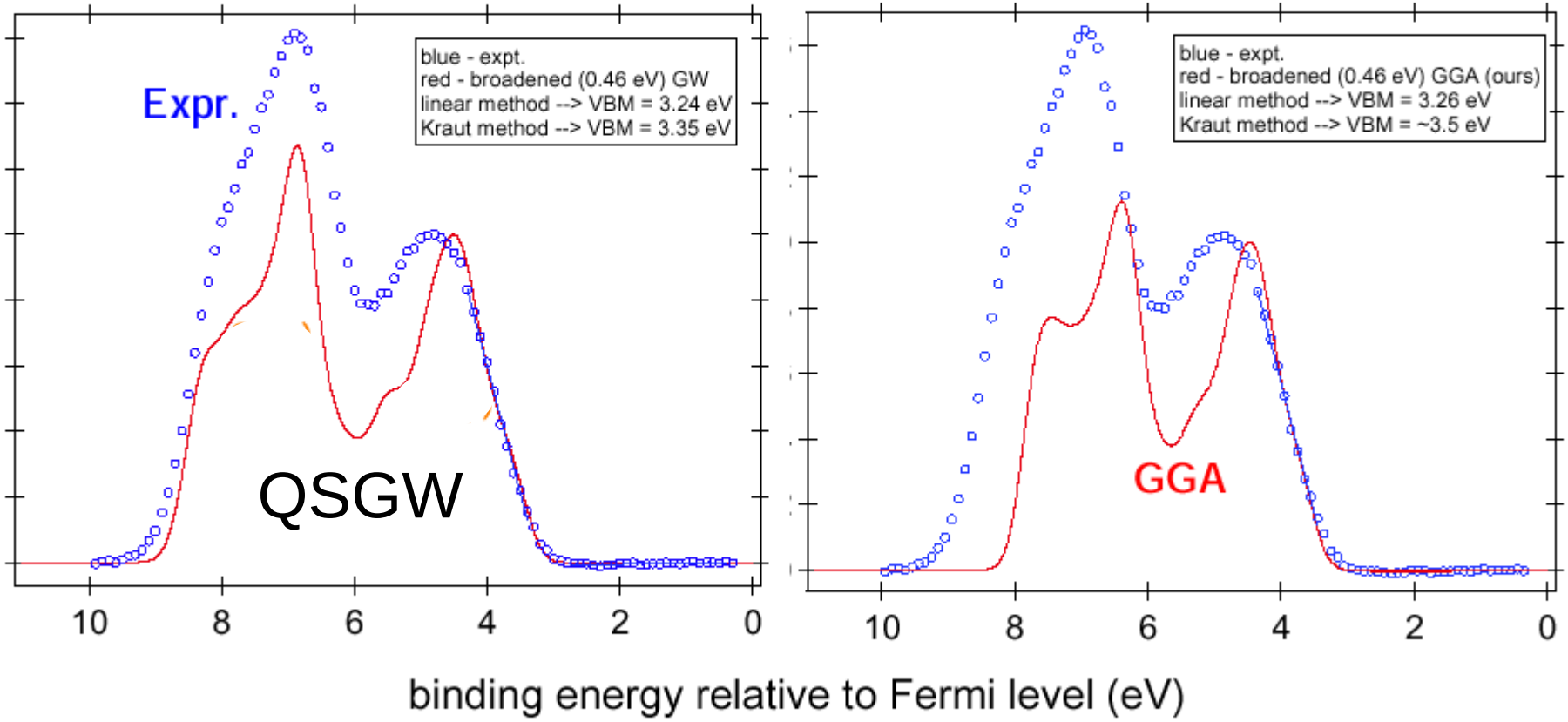
Problem：the mixing ratio can be dependent on materials.

QSGW：**”Instead of bare Coulomb v”**
we use ” Dynamically Screened Coulomb interaction W ”.
$W(\mathbf{r},\mathbf{r}',\omega)$ is determined self-consistently.

Independent-particle picture, where we take into account the charge fluctuation in the RPA self-consistently.

SrTiO3 Valence DOS

Band width. Relative position between 3d and O2p

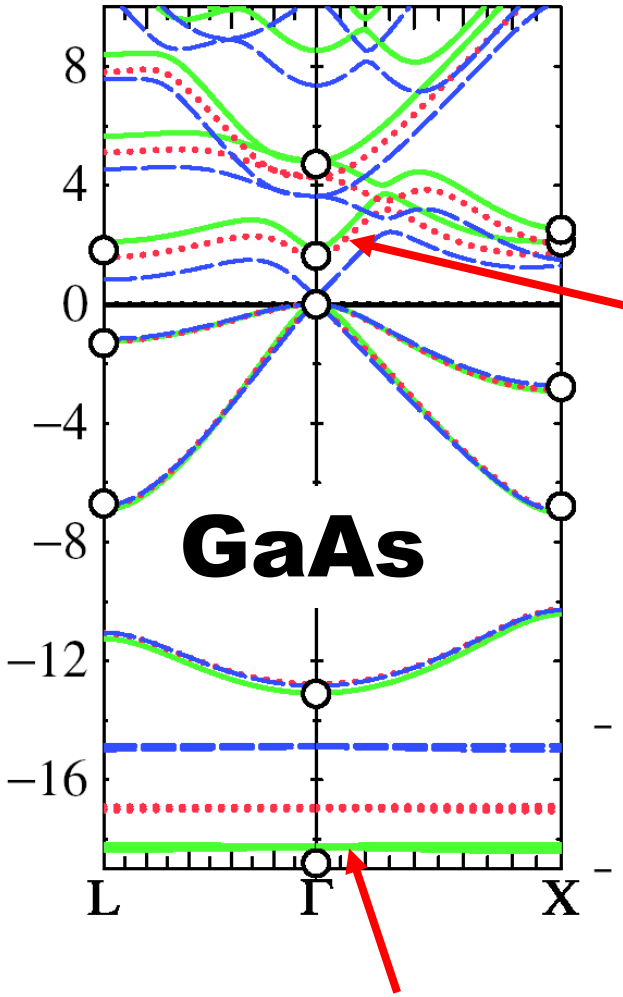


We have accurate description for O2 band width.

S. A. Chambers et al, Surface Sci 554,81-89 (2004)

1. QSGW gives good energy bands.

How QSGW works?: Band gap and effective mass



LDA: Broken blue

QSGW: green

O: Experiment

m^* (QSGW) = 0.073

m^* (expt) = 0.067

m^* (LDA) = 0.022

too large Gap by ~0.3 eV

PhysRevB.76.165106

Ga *d* level well described

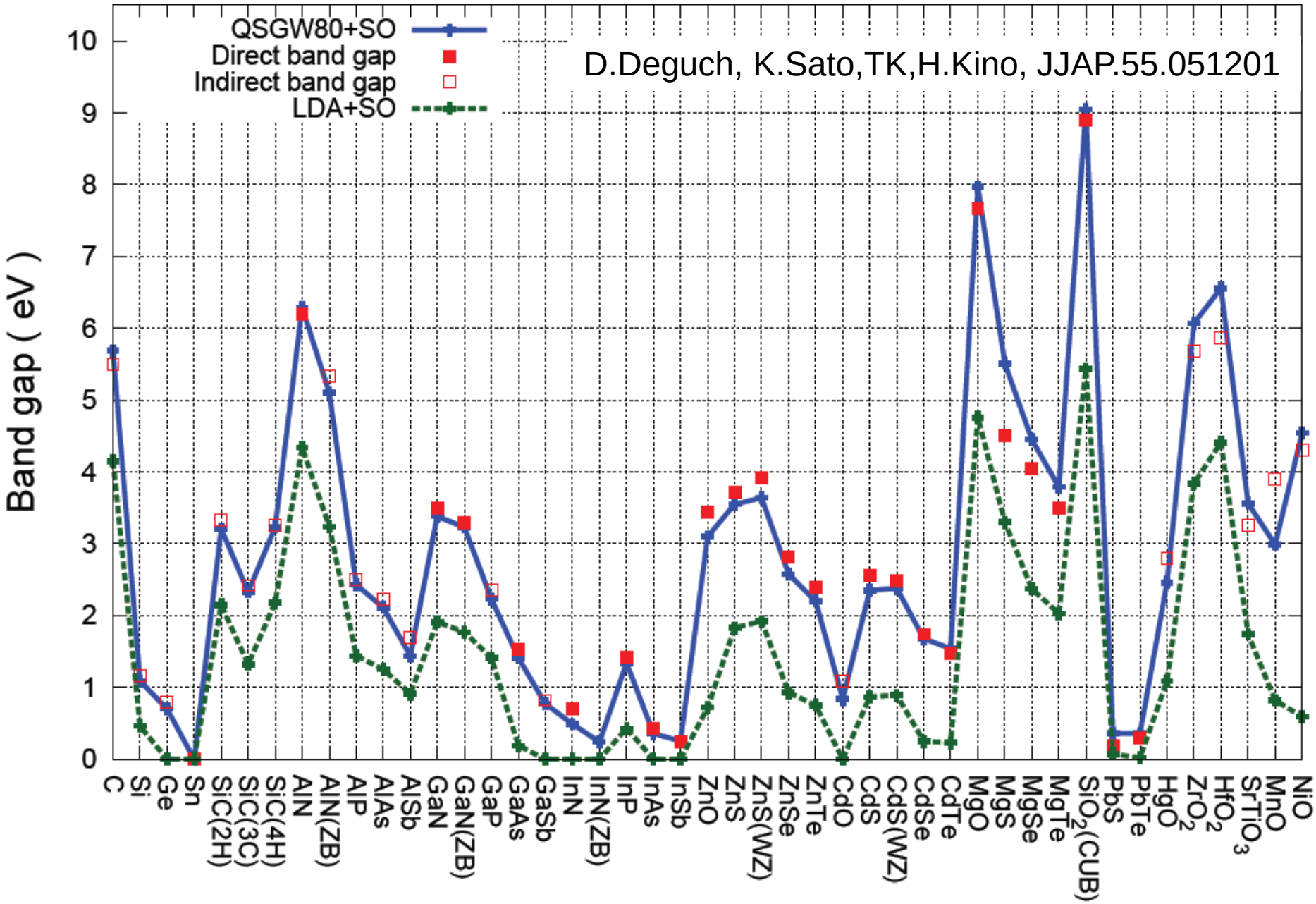
1. QSGW gives good energy bands.

QSGW gives systematic overestimation of band gaps (and effective mass)

→empirical correction

We use QSGW80(80%QSGW+20%LDA).

D.Deguch, K.Sato,TK,H.Kino,
https://doi.org/10.7567/JJAP.55.051201



1. QSGW gives good energy bands.

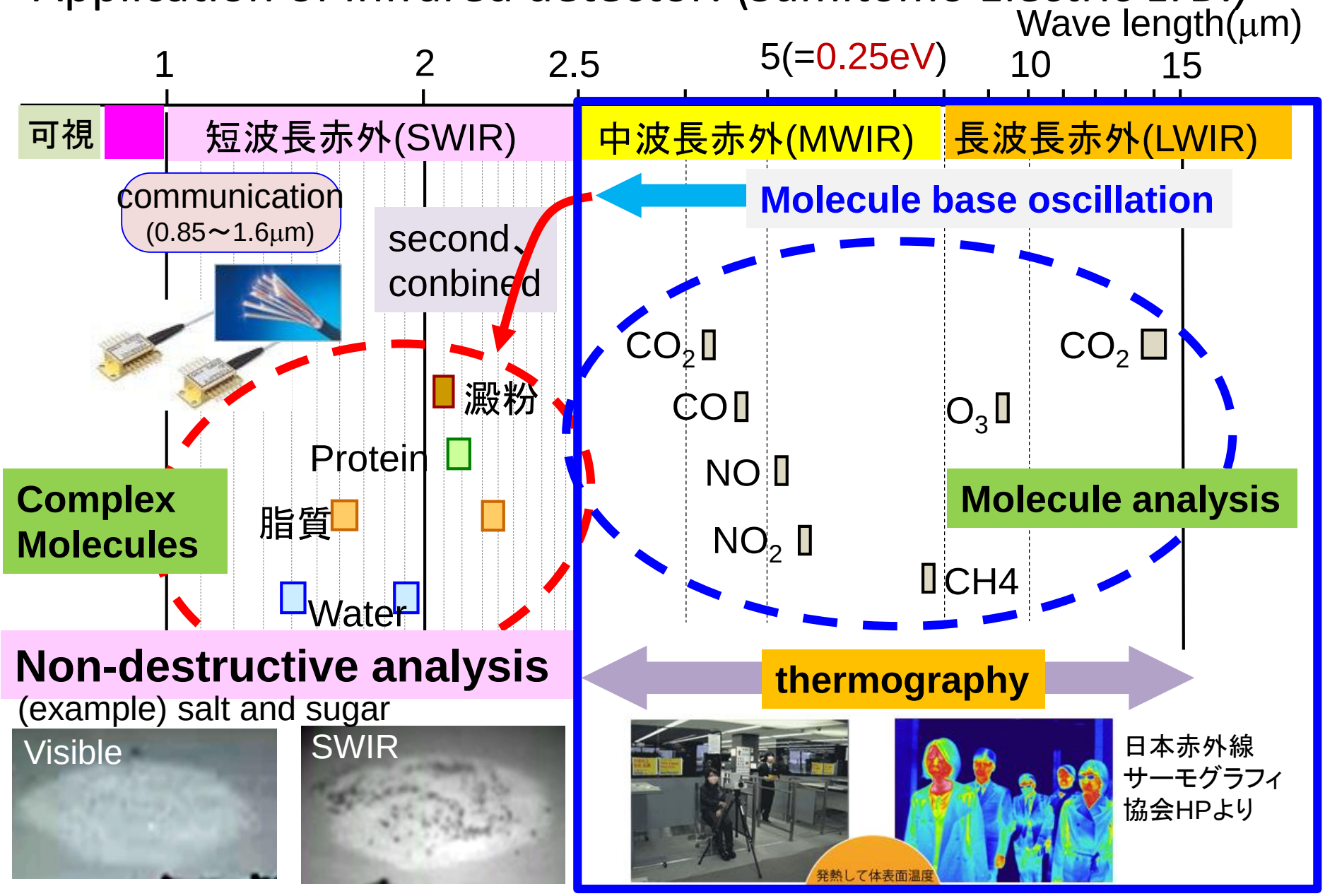
Effective mass(QSGW80+SO)

	m_e	m_{lh}	m_{hh}	m_{so}
GaAs	0.067	0.085	0.319	0.166
	(0.067)	(0.090)	(0.350)	(0.172)
GaSb	0.043	0.049	0.231	0.142
	(0.039)	(0.044)	(0.250)	(0.120)
InP	0.080	0.103	0.411	0.174
	(0.080)	(0.121)	(0.531)	(0.210)
InAs	0.027	0.033	0.346	0.106
	(0.026)	(0.027)	(0.333)	(0.140)
InSb	0.019	0.021	0.254	0.129
	(0.014)	(0.015)	(0.263)	(0.110)
InN(ZB)	0.035	—	1.027	—
ZnS	0.188	0.254	0.643	0.377
ZnSe	0.130	0.178	0.542	0.316
ZnTe	0.115	0.138	0.396	0.288
CdS	0.155	0.205	0.695	0.336
CdSe	0.108	0.148	0.578	0.292
CdTe	0.097	0.118	0.420	0.288
MgS	0.251	0.415	1.252	0.639
MgSe	0.203	0.332	1.038	0.558
MgTe	0.177	0.263	0.734	0.500

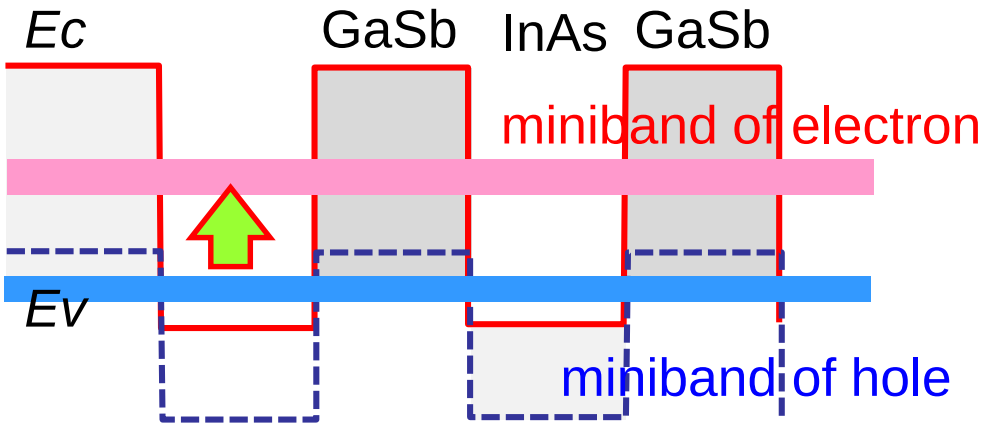
→importan for superlattice of (InGa)(AsSb)

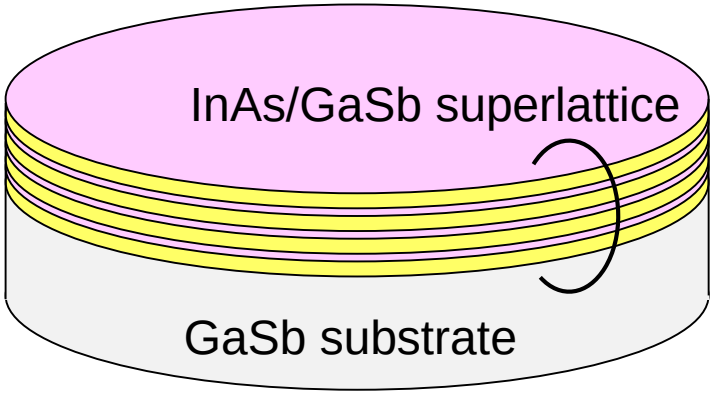
2. Type-II Superlattice $(InAs)_n/(GaSb)_n$

Application of Infrared detector: (*Sumitomo Electric LTD.*)



InAs/GaSb TypeII superlattice for infrared detector





● Why?

✓ Cutoff wave length from 3μm thru 30μm by the layer width control

✓ High quality Wafer. Suitable for 2D array detector.

✓ Good S/N due to small auger current (Heavy mass hole)

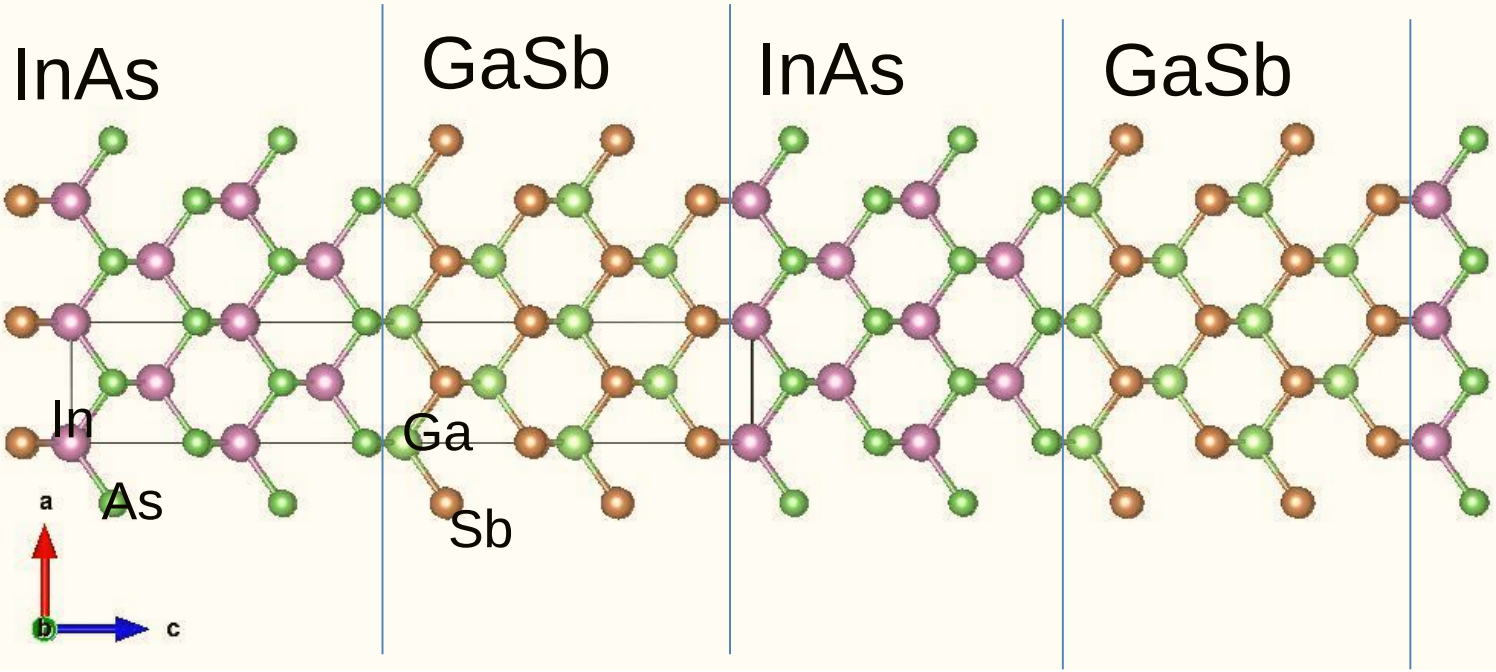
✓ Cd free

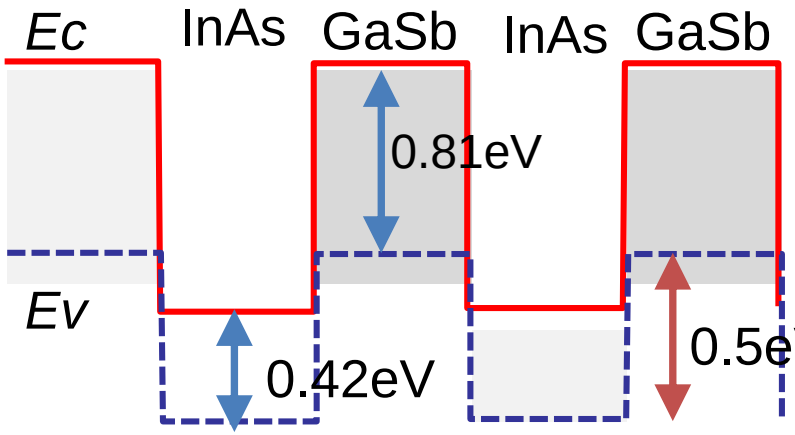
We apply QSGW80 to InAs/GaSb superlattice

- Energy band
- Band gap
- VBM,CBM eigenfunctions
- Type II band diagram(band offset)

J.Otsuka,T.Kato,T.K JJAP.56.021201

$(InAs)_n (GaSb)_m$ n=m=4 model (16atoms/cell)

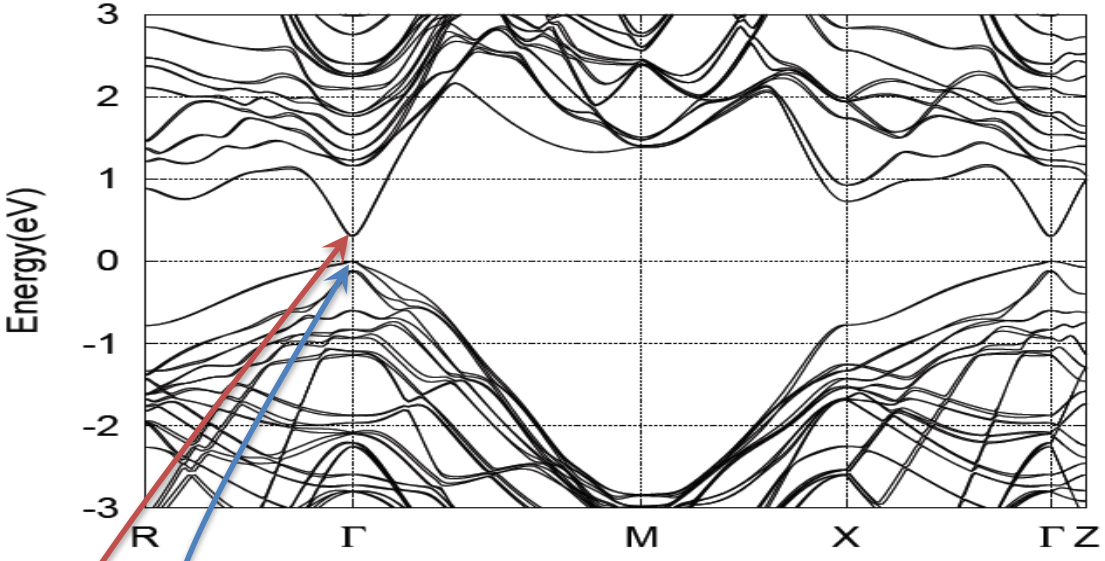




ZB(100) stacking

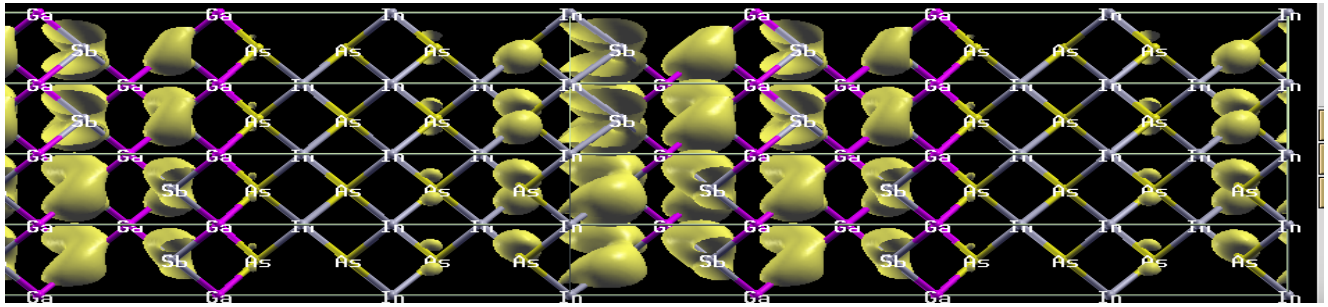
Type II

$(InAs)_n\ (GaSb)_m$ n=m=4 model (16atoms/cell)

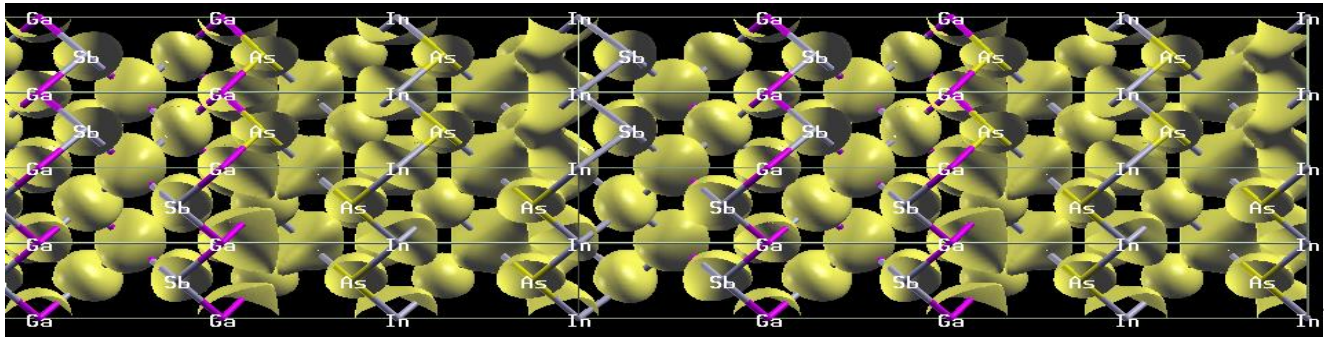


Square of eigenfuncitons

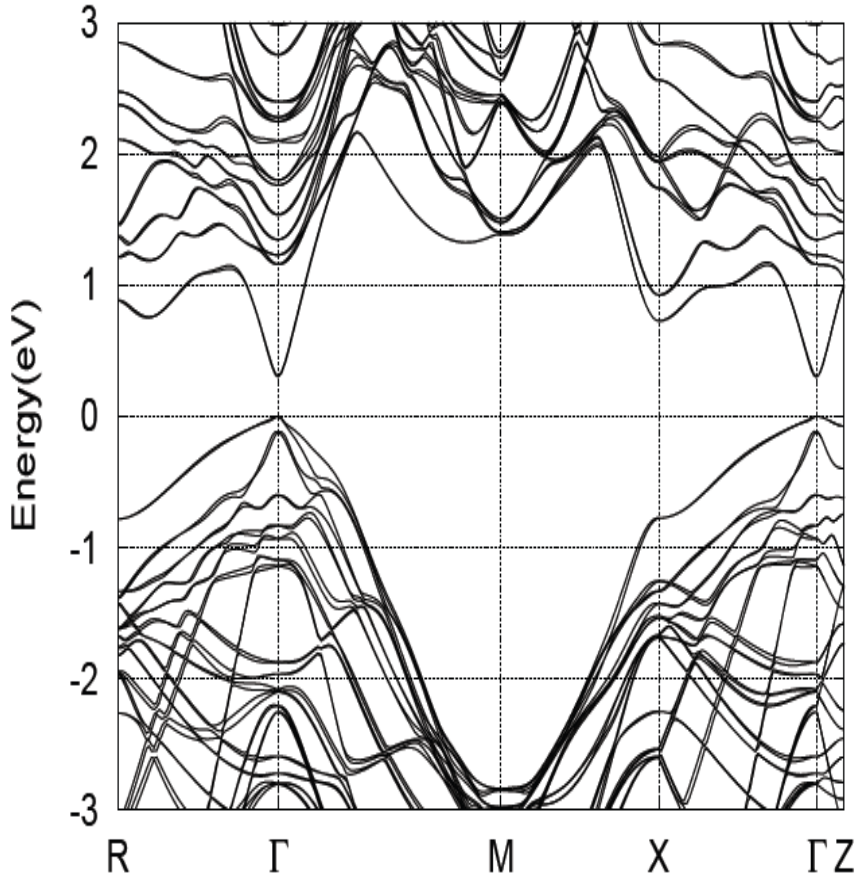
VBM

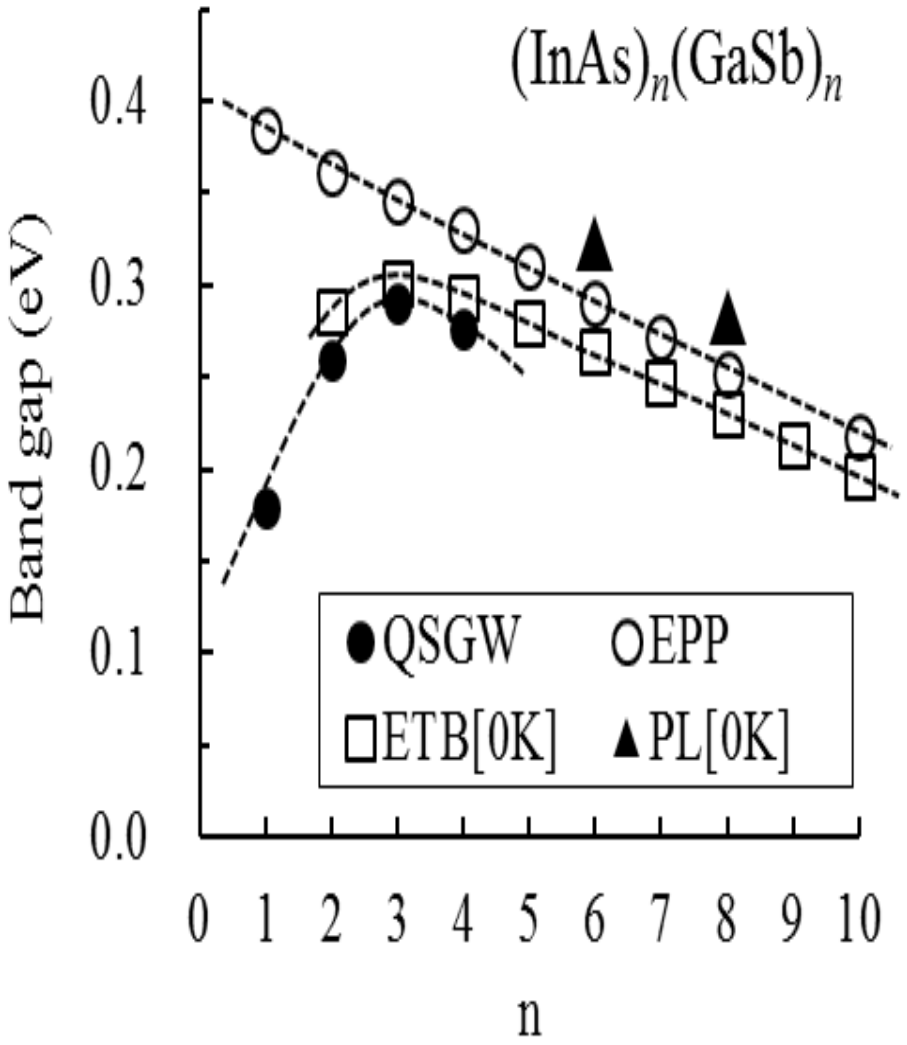


CBM



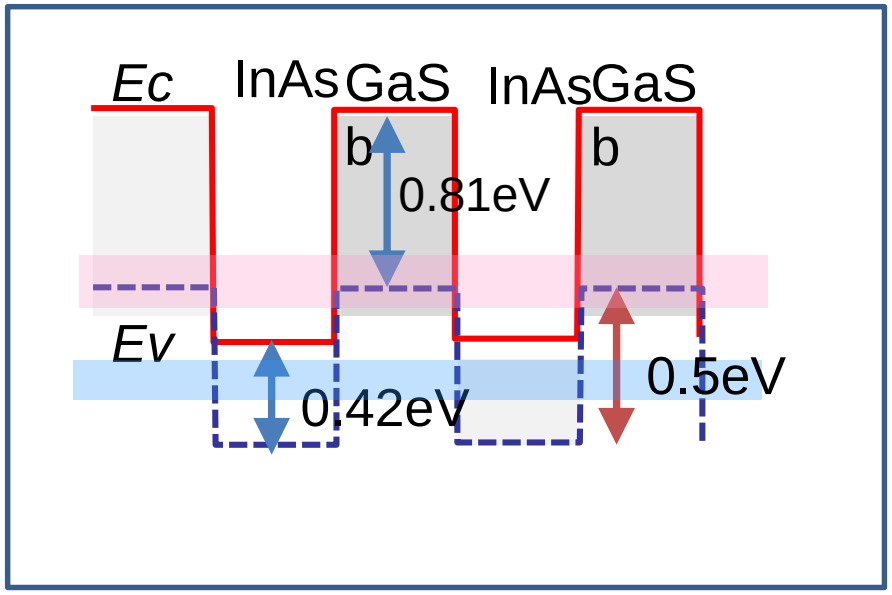
[EPM] P. Piquini, A. Zunger, R. Magri,PRB77, 115314(2008)
[ETBM] Wei, Yajun, Ph.D Thesis, Northwestern Univ (2005)
[PL] A. P. Ongstad, et al, JAP89 2185(2001)

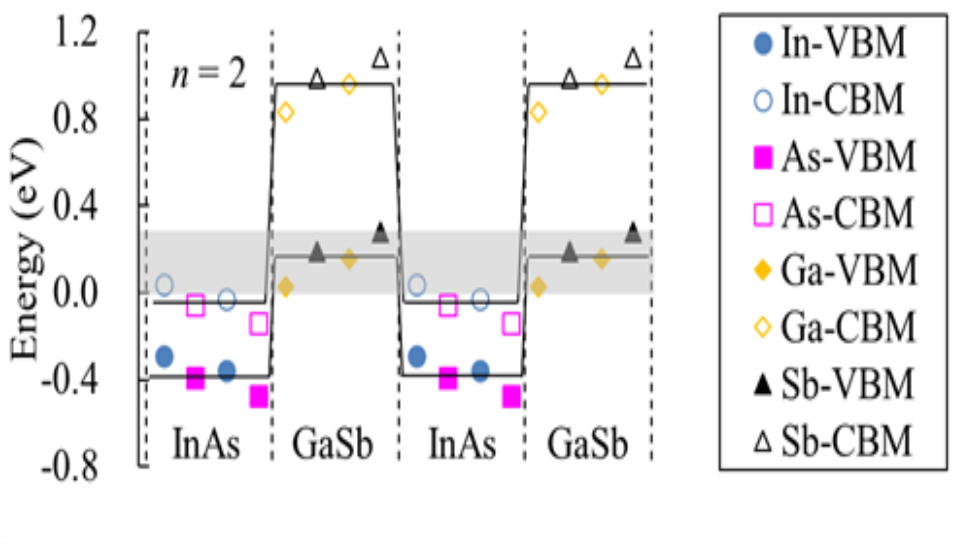




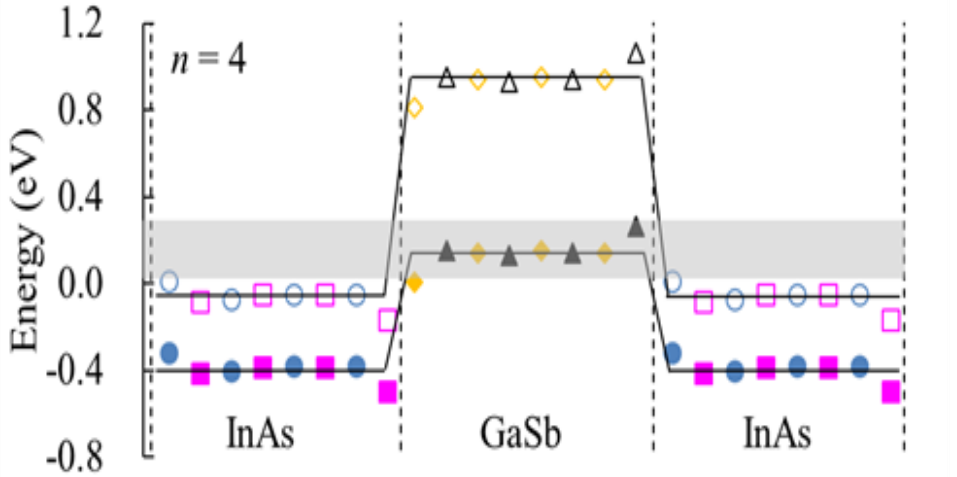
2. Type-II Superlattice $(InAs)_n/(GaSb)_n$

Type II valence band offset calculated by QSGW80





We plot real-axis band diagrams by core levels

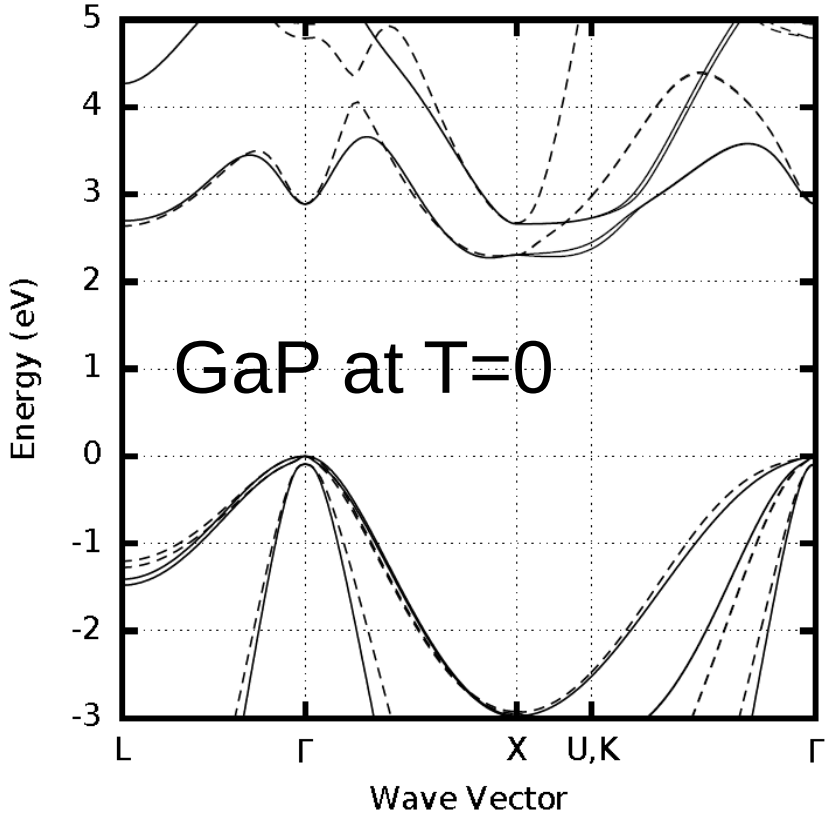


We reproduce experimental band offset very well

How we treat large systems?→Mapping to the model

Nearest-Neighbor sp3s* model based on QSGW

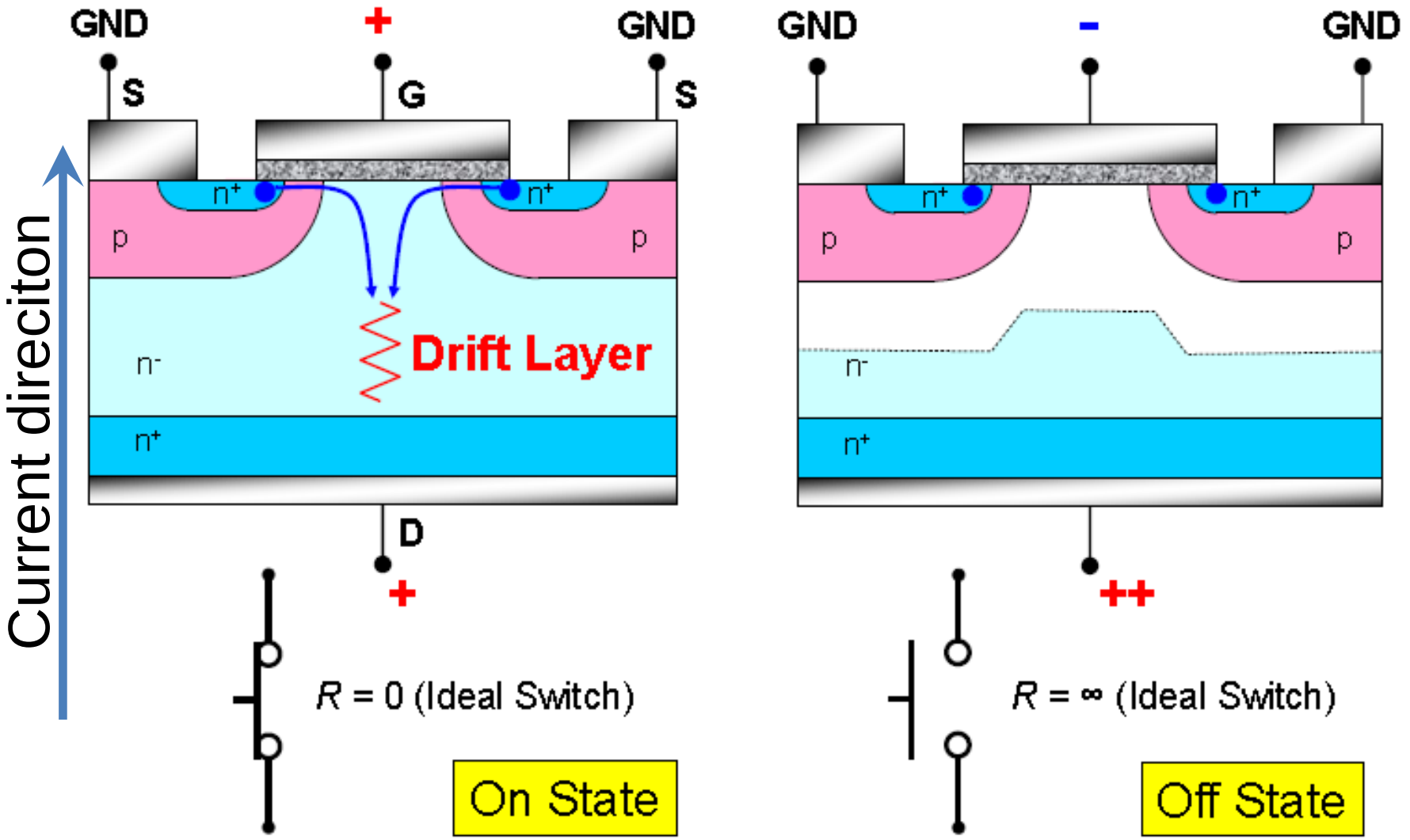
(http://dx.doi.org/10.1063/1.4986658
and supplementary material in it.)



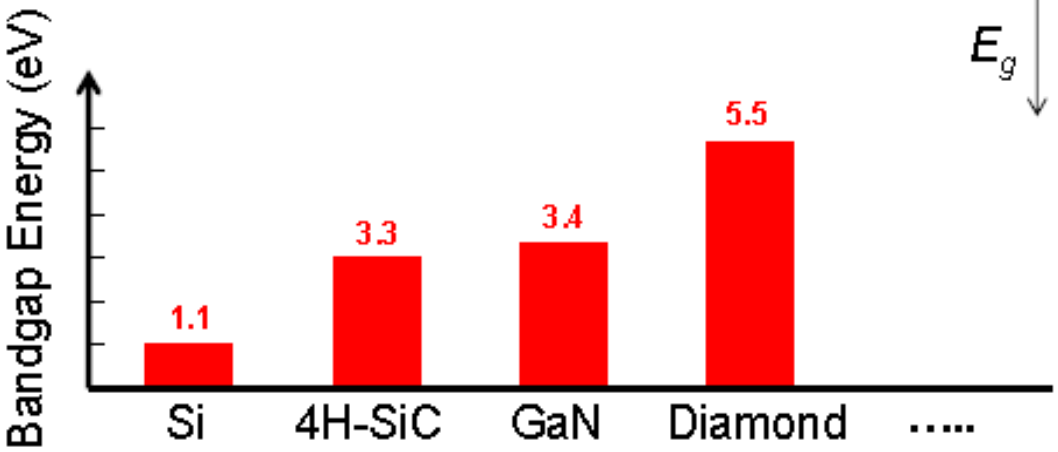
3a. The band structure of GaP obtained by the hybrid QSGW (dashed curves) and the present TB (solid curves) s at $T = 0$K.

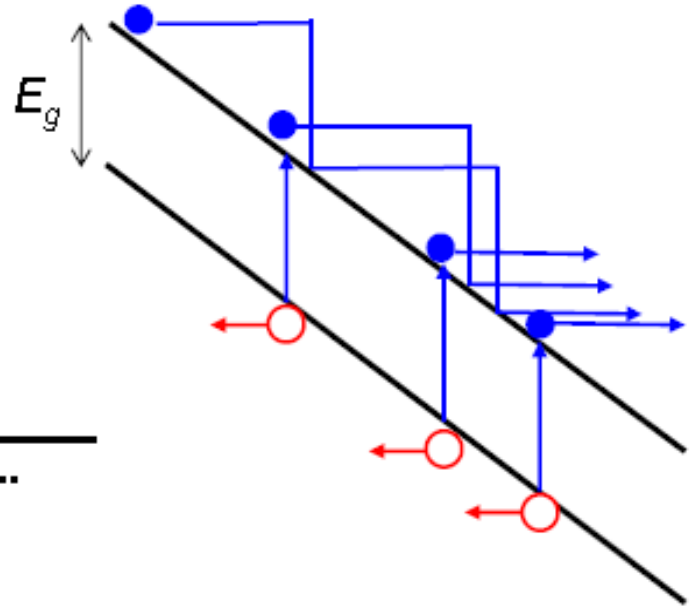
3.Power device

Power MOSFET Structure



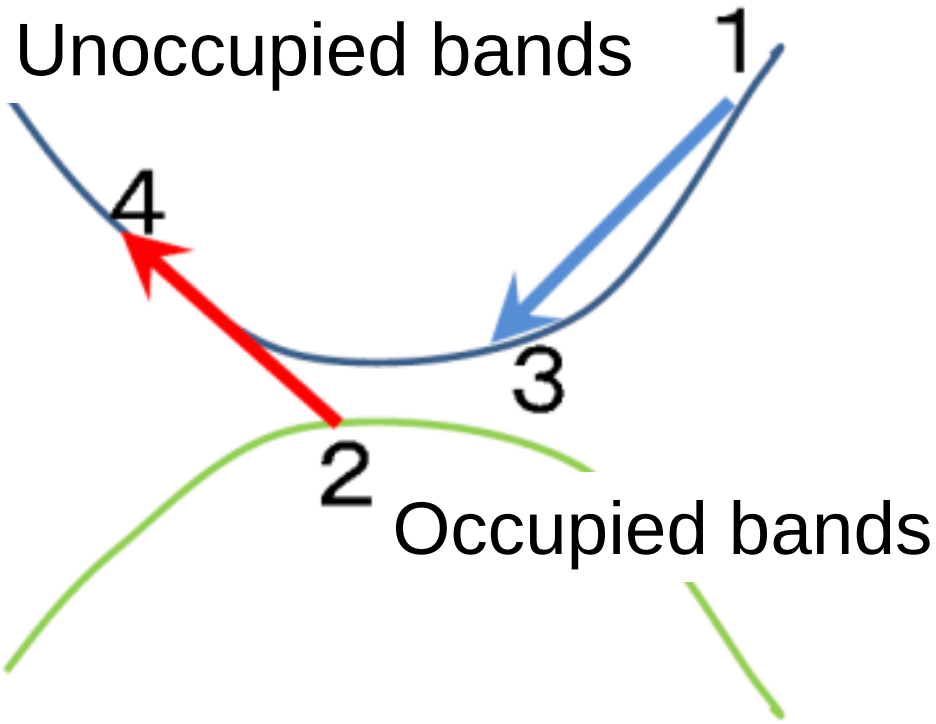
Wide bandgap semiconductors are promising for power devices





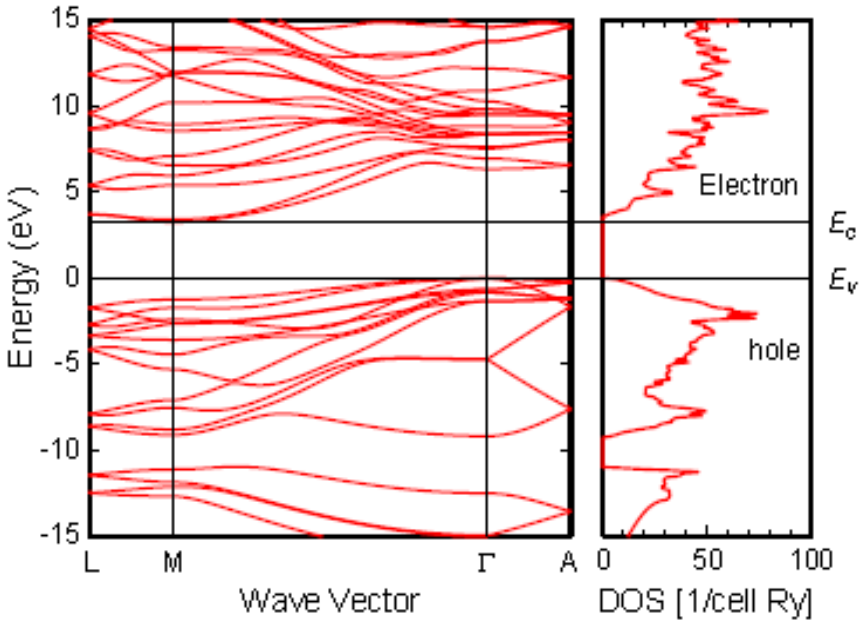
Bandgap E_g ↗ → Breakdown Field F_{BD} ↗ ⇢ Power Loss ↘

Impact ionization rate



= Imaginary part of self-energy

Band Structure (4H-SiC)

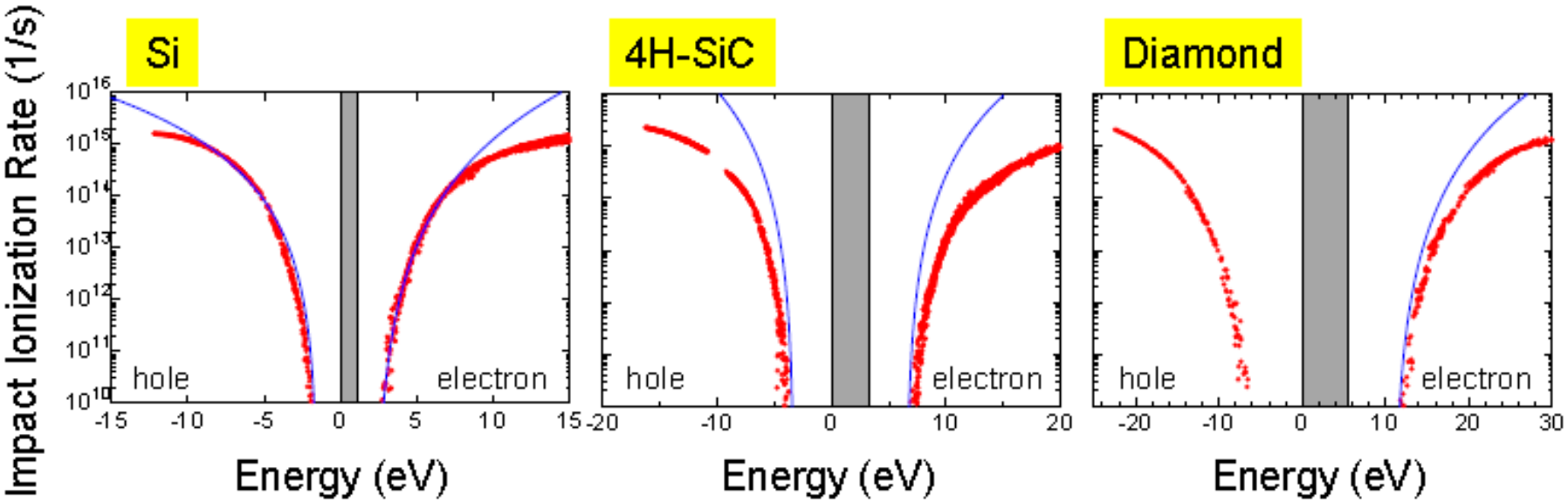


79.5% QSGW + 20.5% LDA to obtain E_g = 3.26 eV

	Direct band gap energy at symmetry points (eV)		Effective mass at conduction band bottom (m_0)	
	Γ	M	$m^*_{M-\Gamma}$	m^*_{M-K}
Experiment	6.18 [a]	4.56 [a]	0.58 [b]	0.31 [b]
This work	6.36	4.50	0.53	0.28

[a] Demir et al., Appl. Phys. Lett. **84**, 3540 (2004)
[b] Chen et al., Phys. Status Solidi A, **162**, 79 (2001)

Impact Ionization Rates (Si, SiC, Diamond)



Red Dots: QSGW Calculation

Blue Lines: Empirical Pseudopotential Method

Si (electron); Kamakura et al., J. Appl. Phys. **75**, 3500 (1994)
Si (hole); Kunikiyo et al., J. Appl. Phys. **79**, 7718 (1996)
4H-SiC; Hjelm et al., J. Appl. Phys. **93**, 1099 (2003)
Diamond; Watanabe et al., J. Appl. Phys. **95**, 4866 (2004)

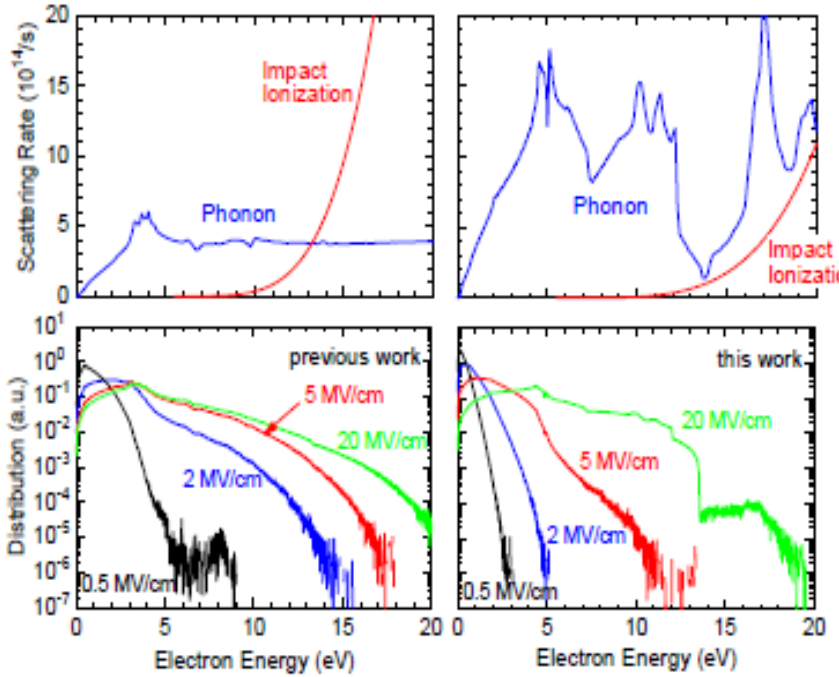


Fig. 7: Energy distribution of ele
energy is measured from the botto
initio calculation (right) are compa

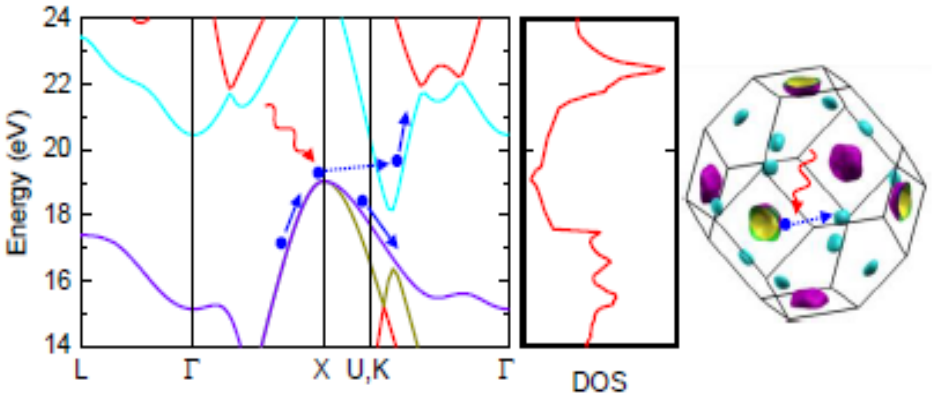
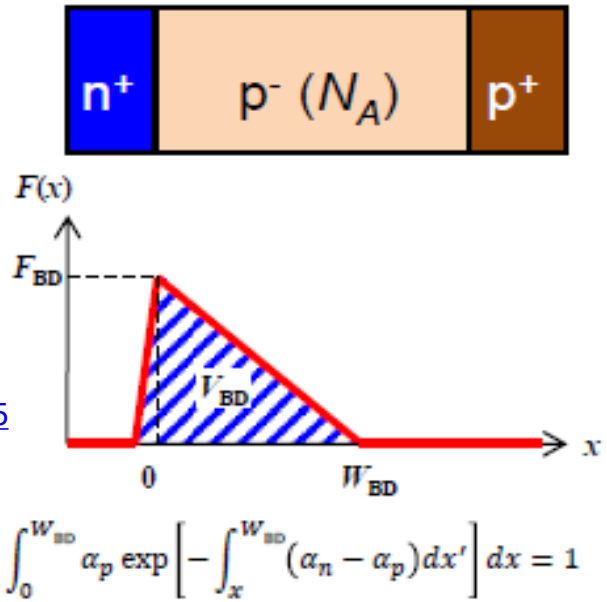
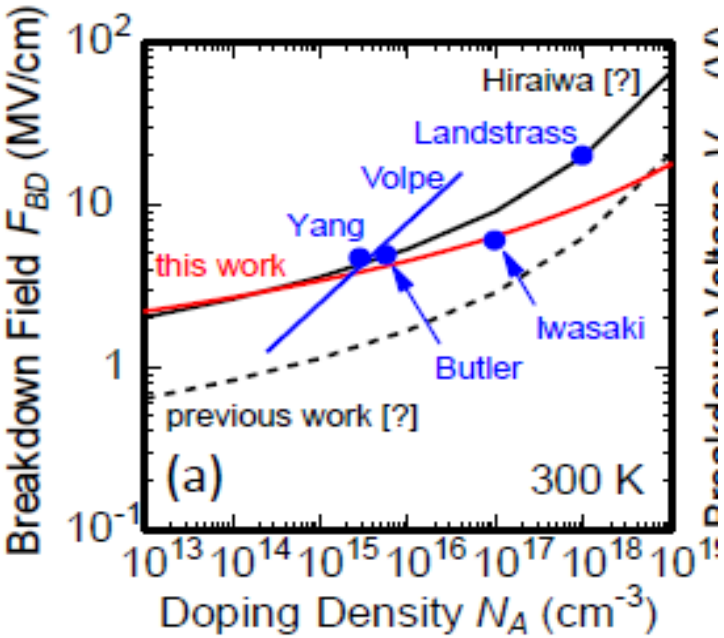


Fig. 8: Zoom-up view of the conduction band structure of diamond around the energy region showing valley-like DOS (see, Fig. 1). The band-to-band scattering process (whose rate is small) is needed for electrons to move to the upper band during the field acceleration. This significantly suppresses the hot electron distribution tail at $F = 20$ MV/cm as observed in Fig. 7.

Diamond

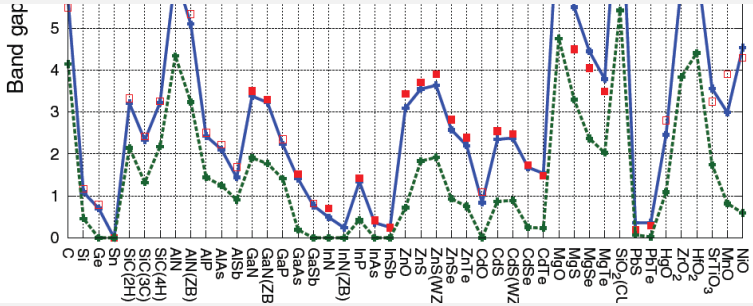




10.1109/IEDM.2015.7409632
10.1109/SISPAD.2016.7605145

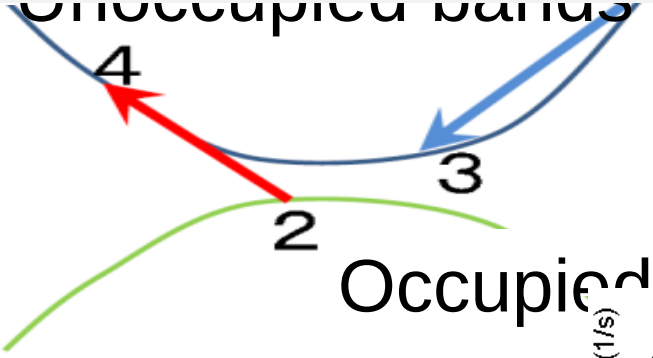
Summary

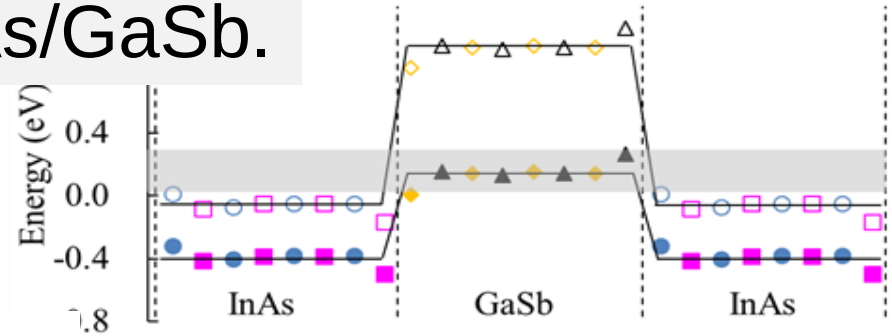
1. QSGW gives good band gap and effective mass.



GaAs	0.066	0.083	0.317	0.164
	(0.067)	(0.090)	(0.350)	(0.172)
GaSb	0.043	0.048	0.232	0.143
	(0.039)	(0.044)	(0.250)	(0.120)

2. Apply the QSGW to InAs/GaSb.





3. Application to power devices.