

## 時間依存密度汎関数法によるグラフェンリボンからの レーザー刺激電界電子放射シミュレーション

Time-Dependent Density Functional Theory Calculations  
of Laser-Assisted Electron Field Emission  
from Graphene Nanoribbons

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# OUTLINE

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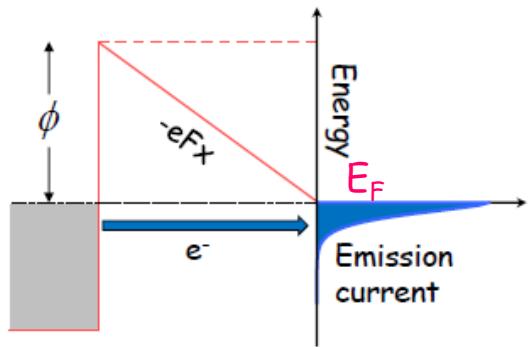
1. Background: FE & LAFE
2. Objectives : Mechanism
3. Method: TDDFT
4. Models: GNR
5. Results: Excitation & tunneling are correlated.
6. Summary: Controlled emission path.

# 1. BACKGROUND

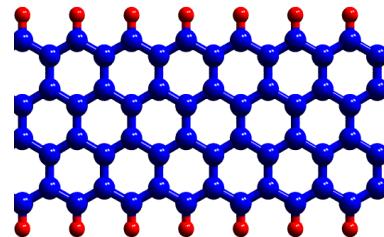
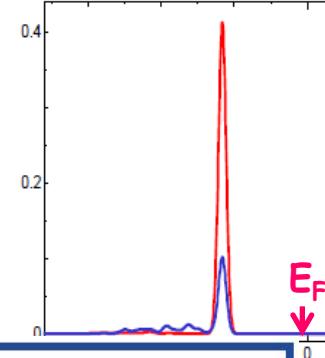
## Field Emission from Carbon Nanostructures

Review: *Carbon Nanotubes and Related Field Emitters*, ed. Y. Saito

- FE from metals

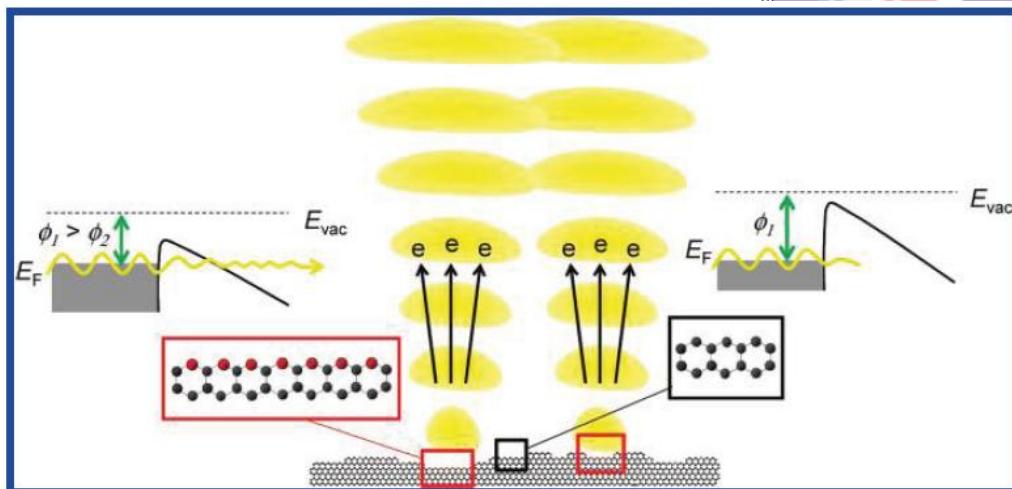


- FE from graphene nanoribbon (GNR)



Theory

K.Tada,KW:PRL88,127601(2000),  
M.Araida,Y.Nakamura,KW:  
PRB70,245410(2004).



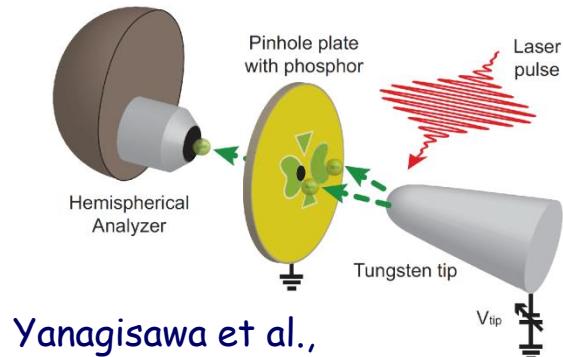
- FE from reduced graphene oxide edges

Experiment

H.Yamaguchi et al.,  
ACSNano 5,4945(2011)

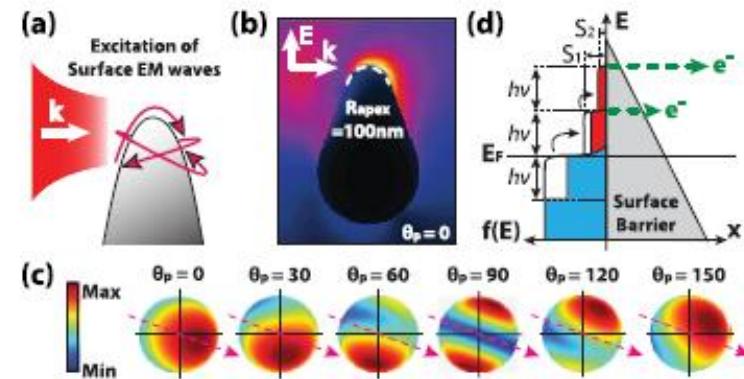
# Laser-Assisted FE from Tungsten Tip

- Experimental setup

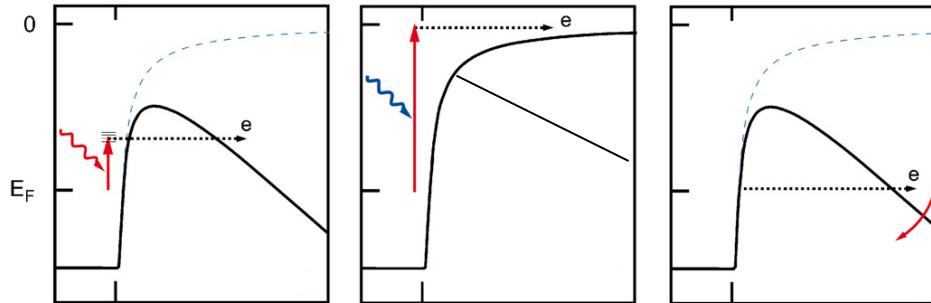


Yanagisawa et al.,  
PRL103, 257603(2009),  
ibid. 107, 087601(2011)

- Laser-assisted photo-field emission



- Mechanisms



P. Hommelhoff et al.,  
PRL96, 077401(2006),  
ibid. 97, 247402(2011)

1. Photo-field emission

2. Optical field emission

3. Photoemission  
(Over-barrier emission)

# Former Theories for LAFE

## Modified Fowler-Nordheim equation

$$J(t) = AF(t)^2 \exp\left(-\frac{B}{F(t)}\right), \quad F(t) = F_{dc} + F_{laser}(t)$$

P. Hommelhoff et al.,  
PRL96, 077401(2006),  
ibid. 97, 247402(2011)

→ 2. optical field emission

## Boltzmann equation

L. Wu and L.K. Ang, PRB78, 224112(2008)  
B. Rethfeld, et al., PRB65, 214303(2002)  
H. Yanagisawa et al., PRL107, 087601(2011)

$$\frac{\partial f(k)}{\partial t} = \frac{\partial f(k)}{\partial t}\Big|_{e-e} + \frac{\partial f(k)}{\partial t}\Big|_{e-ph} + \frac{\partial f(k)}{\partial t}\Big|_{e-laser},$$

$$J(t)_{emission} = \frac{em}{2\pi^2\hbar^3} \iint_W D(W) f(E, t) dE dW, \quad D(W) = \text{Tunneling probability with WKB.}$$

→ 1.photo-field emission

## Needs for Ab-initio Dynamics Simulation

Because, semi-classical theories assume

- Free electrons in a jellium, instead of atomic structures.
- One-dimensional system (2D perfectly flat surface ).
- Electron tunneling is not fully quantum (WKB).
- Electronic excitation is “adiabatic”.

So, 

Time-dependent density functional theory (TDDFT)

Fully quantum electron dynamics excited  
in real materials.

## 2. OBJECTIVES

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- ◆ Explore excited electron dynamics under fs laser and dc field by ab-initio calculations.
- Reveal emission mechanism depending on laser and materials parameters, and predict & control the emission path.

### 3. THEORY

#### TDDFT studies on FE & LAFE

- FE from CNT: S.Han & J.Ihm, PRB66,241402(R)(2002).
- FE from GNR: K.Tada & KW, PRL88, 127601(2000).
- FE from Diamond: M.Araida, KW, JJAP42,L666(2003).
- LAFE from CNT: J.A.Driscoll et al., PRB83,233405(2011).

#### What is TDDFT ?

E. Runge, E.K.U Gross, PRL52,997 (1984)

Electrons:

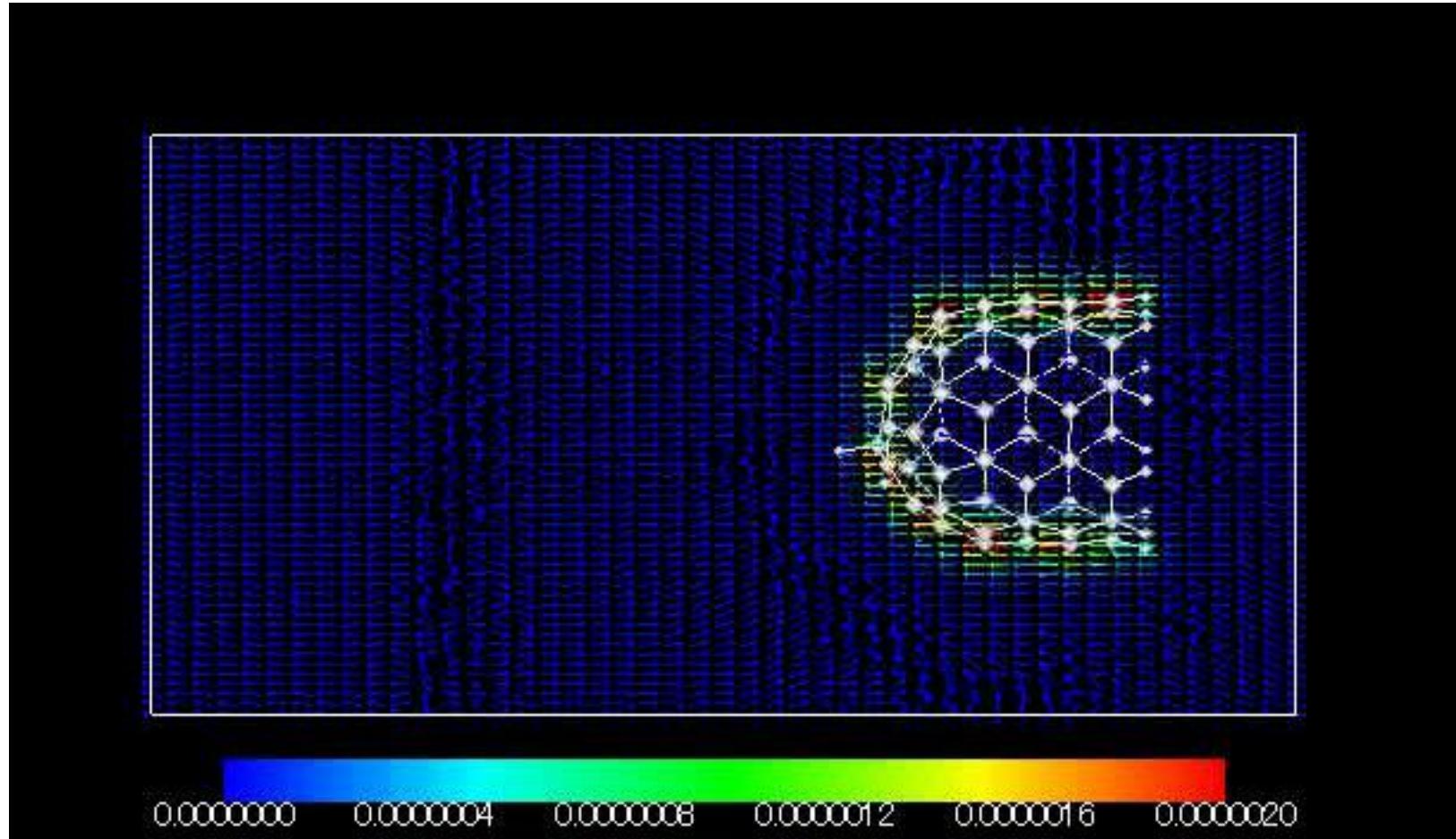
$$i\hbar \frac{\partial}{\partial t} \psi_i(\vec{r}, t) = H_{KS}(\vec{r}, t) \psi_i(\vec{r}, t)$$

$$H_{KS}(\vec{r}, t) = H_0(\vec{r}, t) + v_{\text{laser}}(\vec{r}, t), \quad v_{\text{laser}}(\vec{r}, t) = e\vec{E}(t) \cdot \vec{r}$$

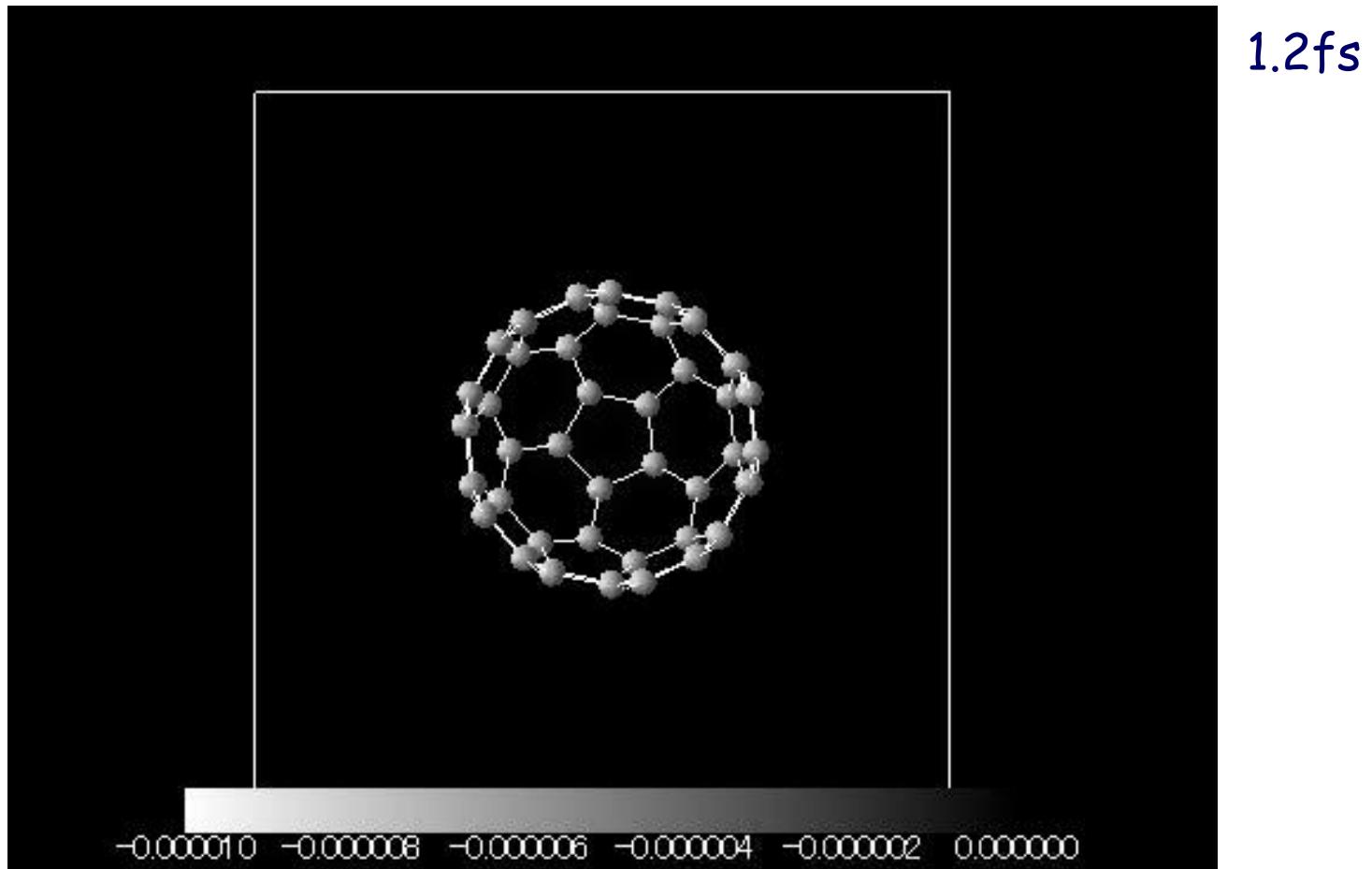
$$\psi_i(\vec{r}, t + \Delta t) \cong \exp[-i\Delta t / \hbar \cdot H_{KS}(\vec{r}, t)] \psi_i(\vec{r}, t)$$

## Field Emission from (5,5)CNT@H<sub>2</sub>

1000 steps = 1.2fs



# Field Emission "Microscopy" of (5,5)CNT



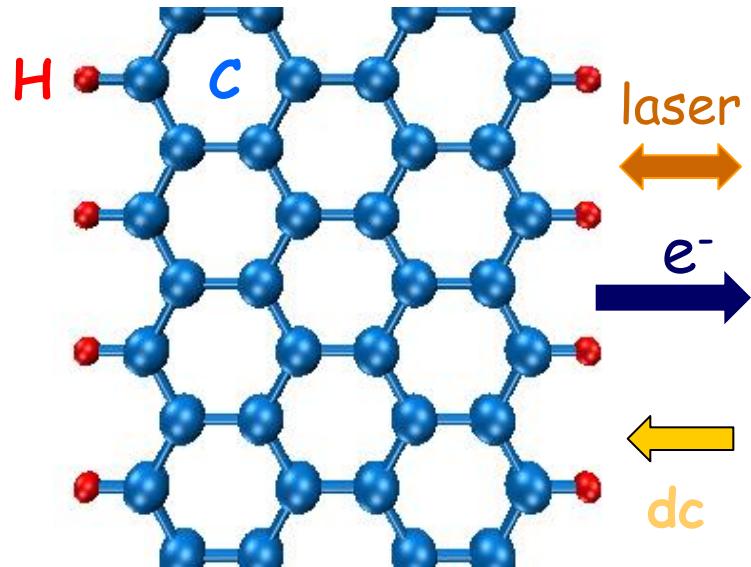
K. Nishida et al., APEX 4, 115102(2011)

(e)



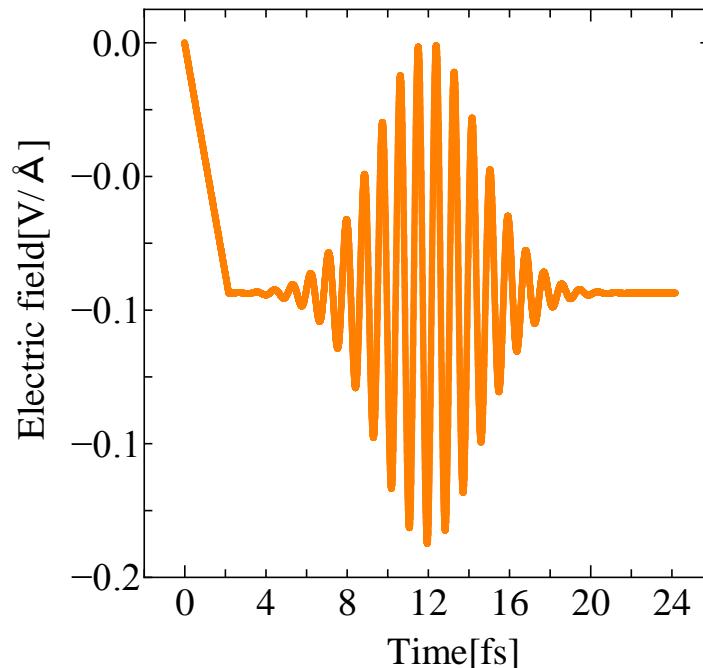
## 4. EMITTER & LASER

Graphene nanoribbon (GNR)



FS laser pulse

$$E_{dc} = 0 \sim 0.2 \text{ V/\AA}, \quad E_{laser} = 0 \sim 0.1 \text{ V/\AA},$$
$$I_{laser} \approx 1 \times 10^{11} \text{ W/cm}^2$$
$$\hbar\omega = 1 \sim 7 \text{ eV}$$

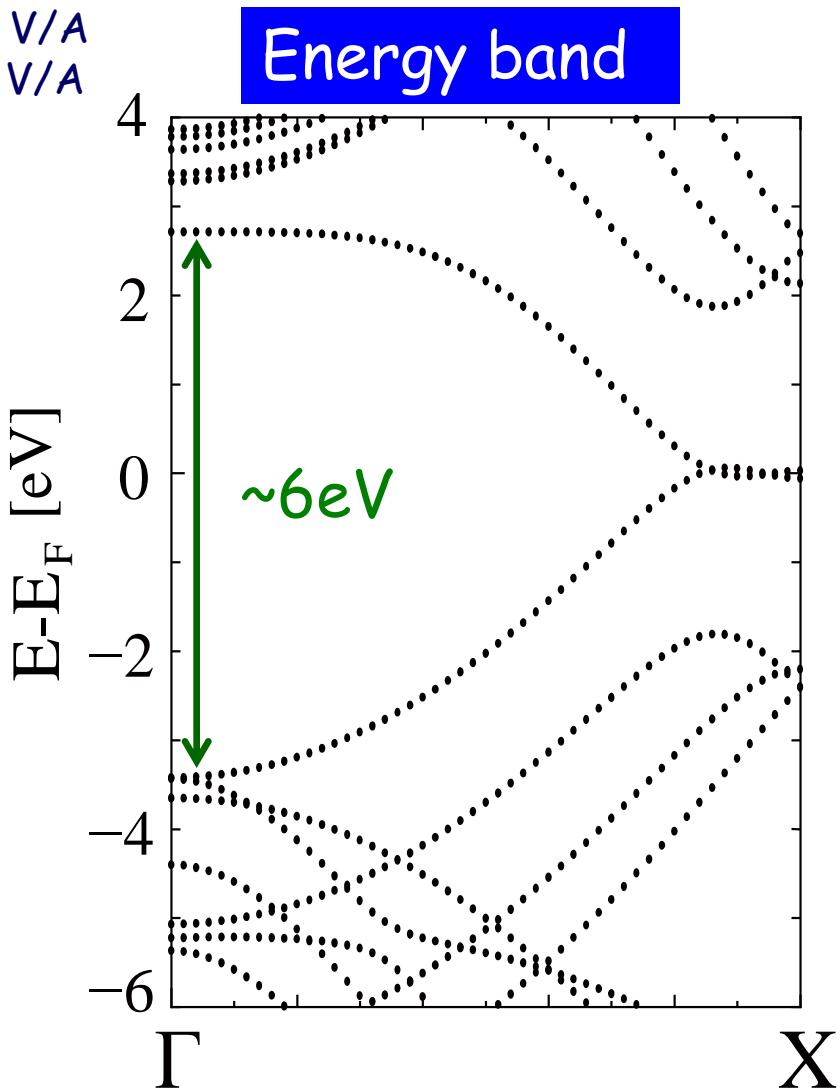


## 5. RESULTS: H-terminated GNR

### Emission current

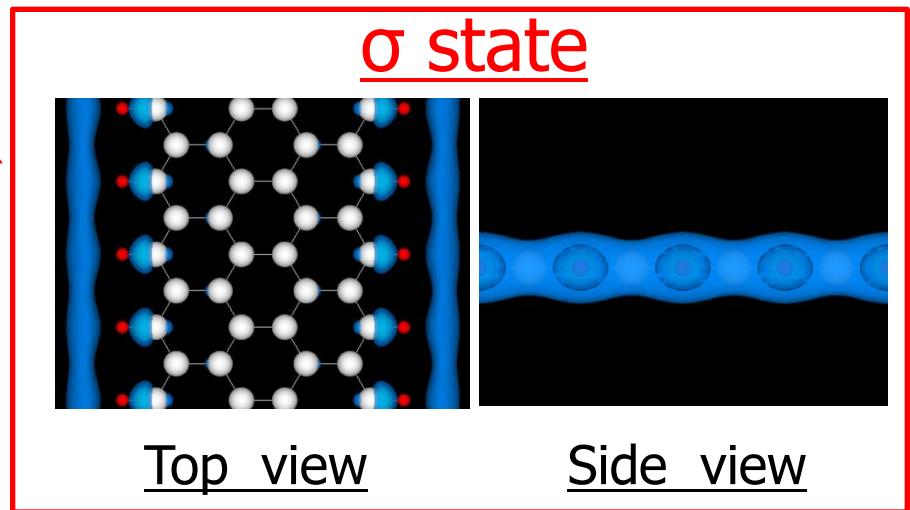
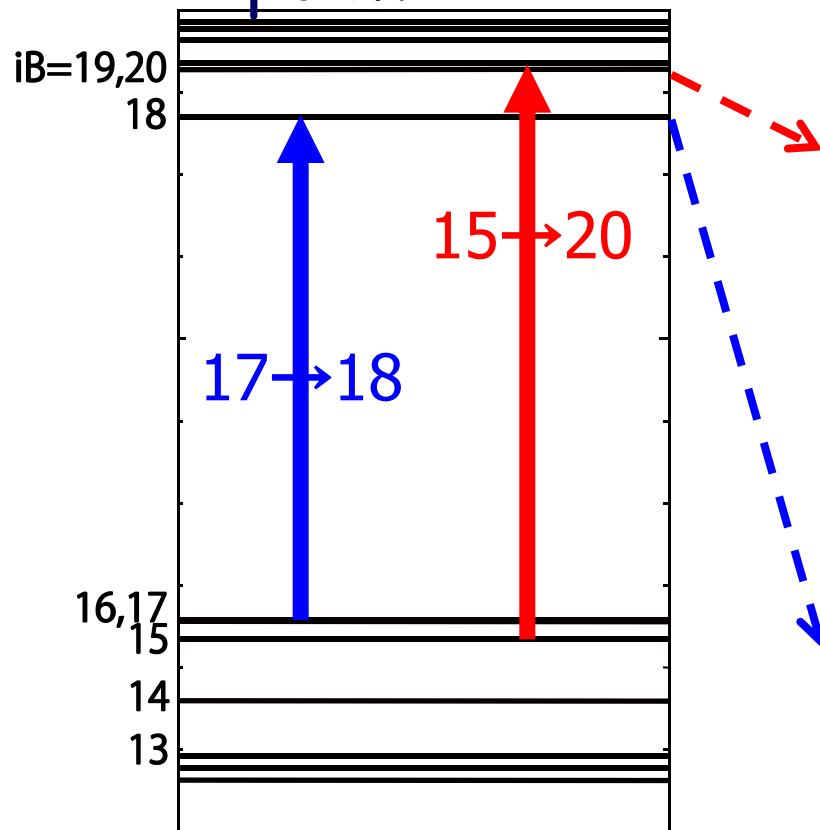
$E_{dc} = -0.1 \text{ V/A}$   
 $E_{laser} = 0.1 \text{ V/A}$

Emission starts under  
a laser of energy  
corresponding to  
dipole transition.



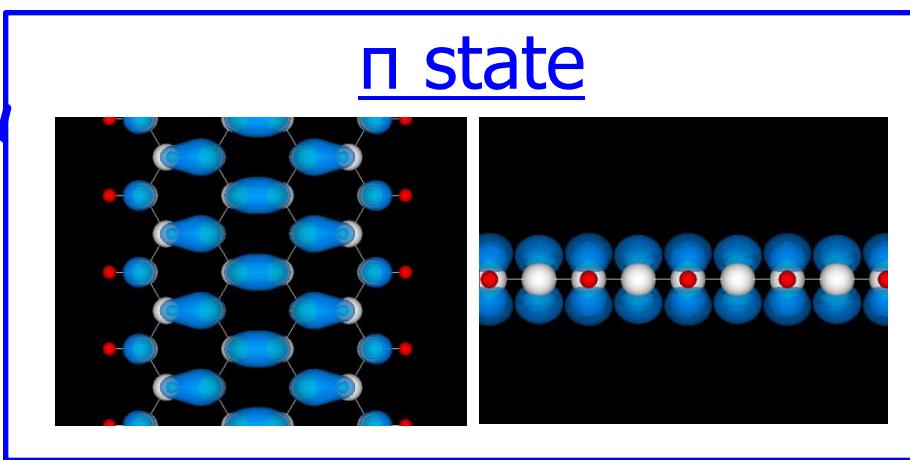
# Which electronic levels cause emission ?

$\Gamma$  point



Top view

Side view



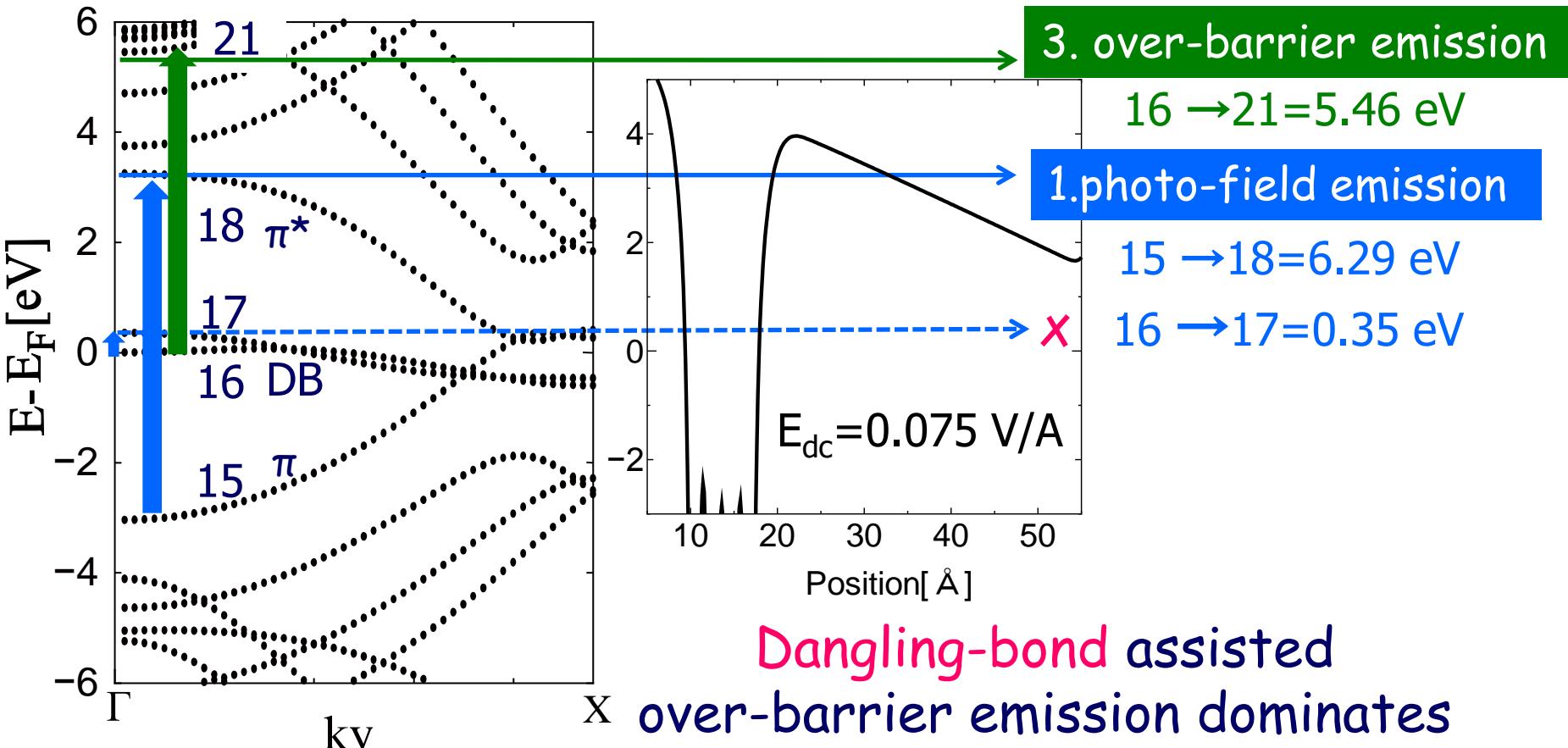
Transition is large for  $17 \rightarrow 18$ ,  
but, emission is large from 20,  
because of the **orbital symmetry**.

$$J \propto \int D(\varepsilon) E(\varepsilon) T(\varepsilon) d\varepsilon$$

Excitation  
in the band



Emission  
through the potential



Dangling-bond assisted

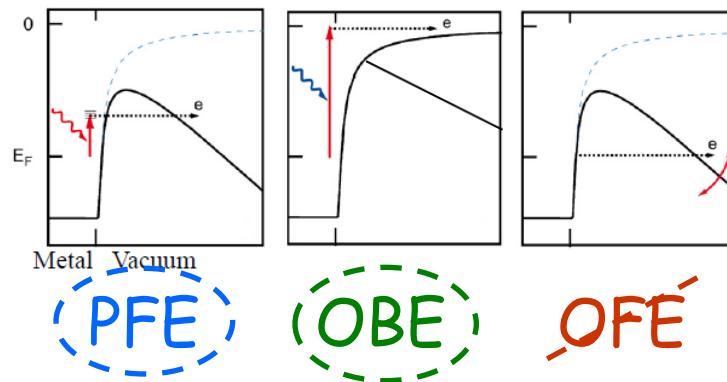
over-barrier emission dominates  
photo-field emission.

## 6. SUMMARY

### Laser-Assisted FE

- ◆ is highly non-stationary;  $J \neq \int D(\varepsilon)E(\varepsilon)T(\varepsilon)d\varepsilon$ ,
- ◆ reflects the electronic structures of the tip,
- ◆ changes critically depending on laser parameters, static field & work function.

### Emission Path



- ◆ can be selected by tuning laser parameters.