# How much time necessary to photo-generate Fermi surface from true electron vacuum?

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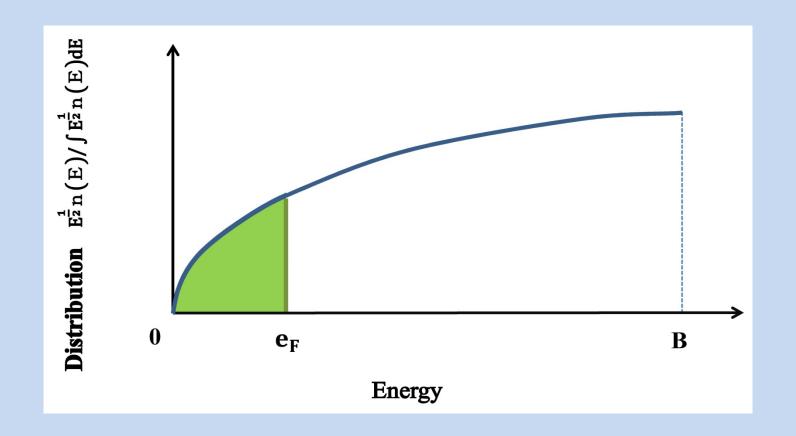
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# Presence of Fermi surface is the base of solid state science.

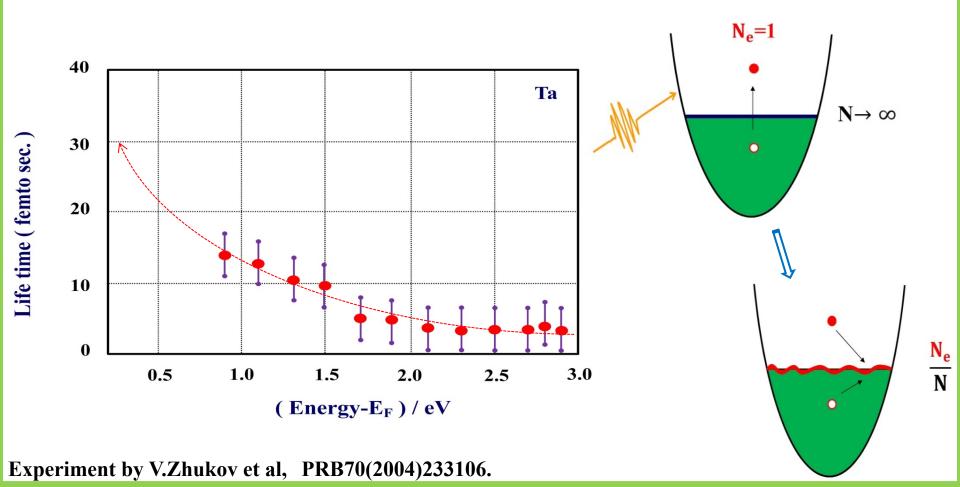


BCS, Kondo, C(S)DW, Plasmon theories are all assumed, it has been already well established.

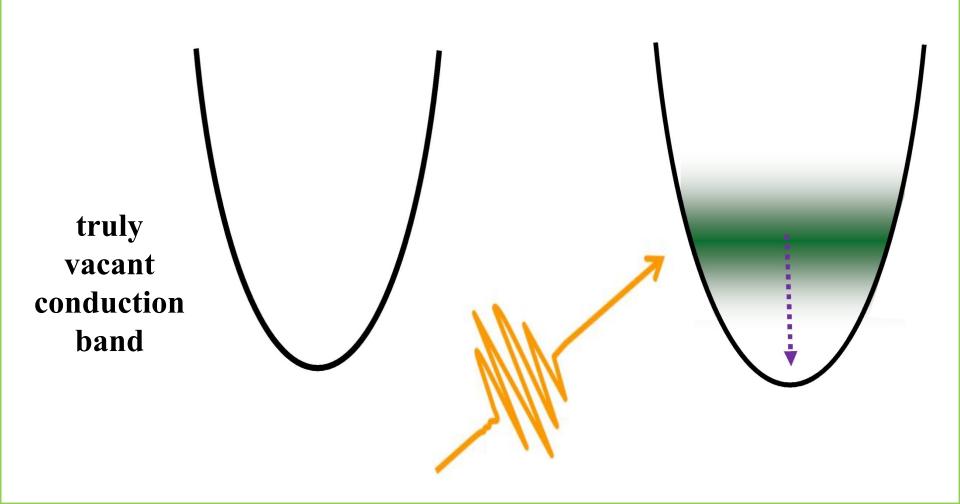
# How much time necessary to photo-generate Fermi surface from true electron vacuum?

#### **Motivation**

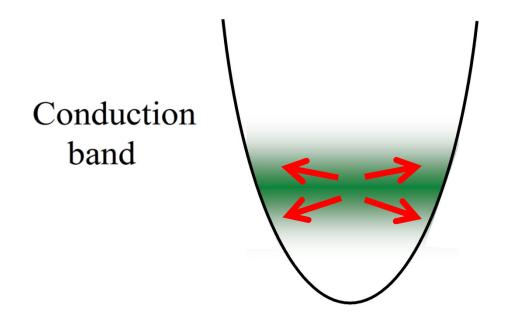
Rapid relaxation dynamics of optically excited electrons in metallic systems, has already been widely investigated. In most cases, however, only a few electrons are excited, while, the main part of electrons is still in the ground state, works as an infinite heart reservoir, resulting in quite rapid relaxation of newly given energy and momentum.



What occurs, if a macroscopic number of electrons are excited, at once, into a truly vacant conduction band, without electronic heat reservoir?



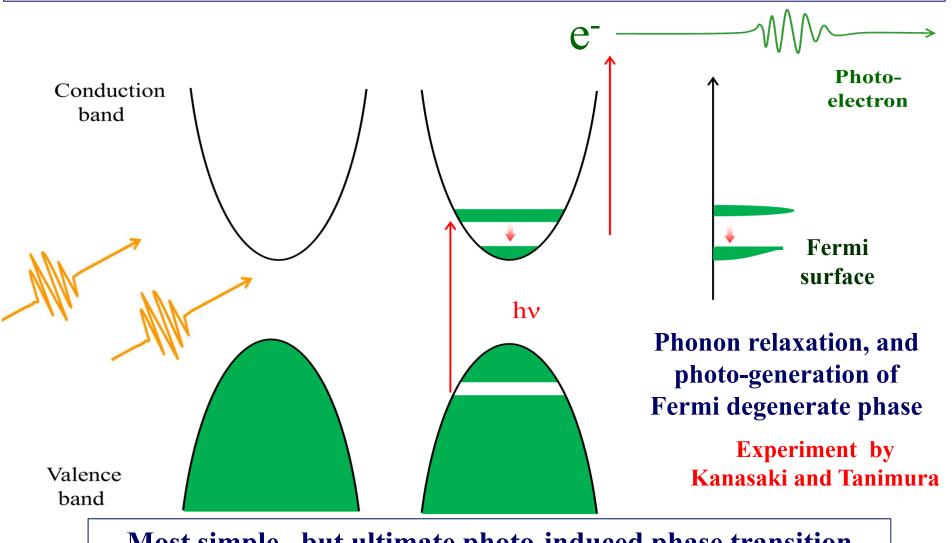
# Coulombic inter-electron scatterings within the conduction band, being completely elastic, can give no net energy relaxation.



Intra-band Coulomb scattering

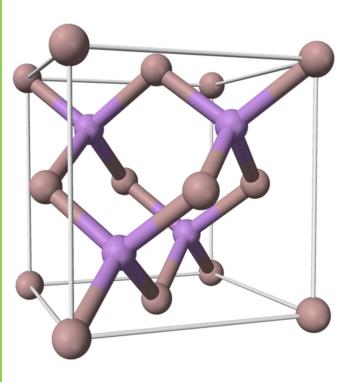
# Phonon relaxation, two time regions avalanching initially, but soon it slows down infinitely, as approaches Fermi degeneracy, since only low energy phonons are available. c.f. Luttinger theorem PR 118(1960)1417. Avalanching Fermi **Critical** degeneracy slowing down

# Two pulse excitations of GaAs, InP by visible laser Time resolved photo-emission spectrum of conduction band electron

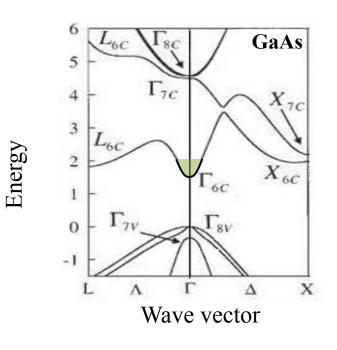


Most simple, but ultimate photo-induced phase transition from true electron vacuum.

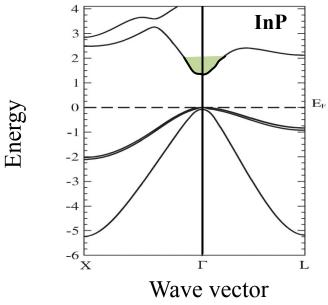
# Lattice structure of GaAs, InP



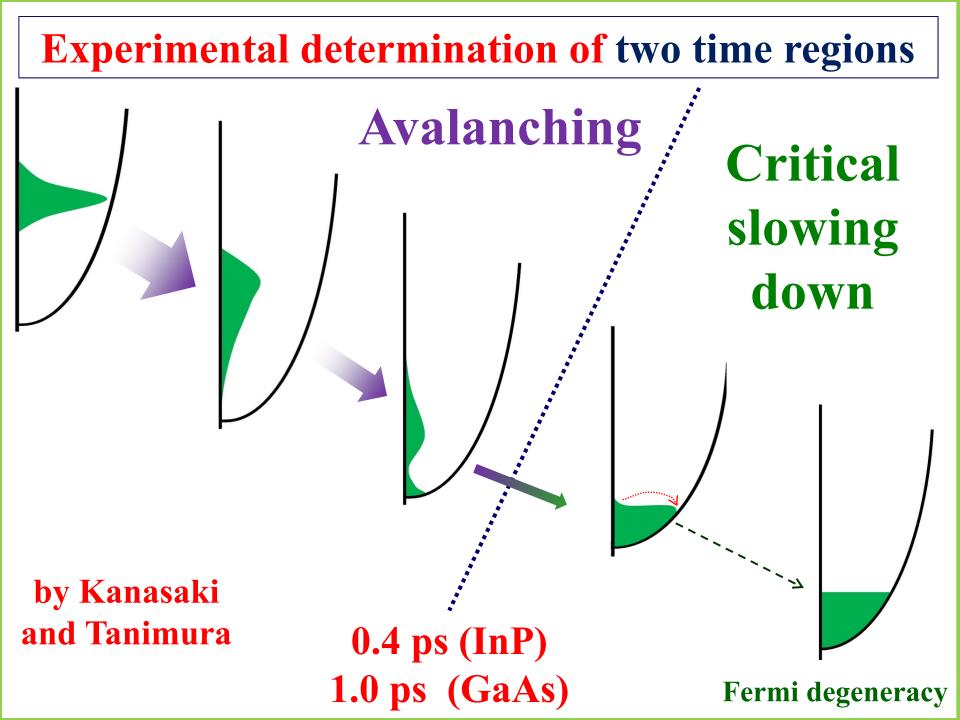
#### **Energy bands**



N.Cavassilas et al, PRB 64(2001)115207.



L.Dacal et al, SSC 151(2011)781.



#### Many-electron and acoustic phonon coupled system $(\equiv H)$

$$\mathbf{H} = \mathbf{H_0} + \mathbf{H_i}, \qquad \mathbf{H_0} \equiv \mathbf{H_e} + \mathbf{H_p}, \qquad \mathbf{H_i} \equiv \mathbf{H_{ep}} + \mathbf{H_{ee}},$$

$$H_e \equiv \sum_{k,\sigma(=\alpha,\beta)} (e(k)-\mu) a_{k,\sigma}^+ a_{k,\sigma}$$
,  $0 \le e(k) \le B$ ,  $B = 5 \text{ eV}$ ,

$$n_M \equiv (2N)^{-1} \sum_{k,\sigma(=\alpha,\beta)} a^+_{k,\sigma} a_{k,\sigma}, \quad n_M = 0.001 \sim 0.005, \quad \mu \equiv \ \ \text{Chemical potential}$$

Electron-phonon coupling and Hubbard type weak Coulomb repulsion

$$H_p \equiv \sum_q \omega_q b_q^+ b_q^-$$
 ,  $\omega_q \equiv c_s^- |q|$  ,  $~0 \leq \omega_q \leq \omega_\text{M} (\equiv 24 \text{meV})$  ,  $c_s^- = 30 \text{Å/(pico.sec.)}$  ,

$$H_{ep} \equiv S(2N)^{-1/2} \sum_{q,k,\sigma(=\alpha,\beta)} (b_q^+ + b_{-q}) a_{k-q,\sigma}^+ a_{k,\sigma}, \quad S \cong 1 \text{ eV}$$

$$H_{ee} \equiv U \sum_{\rho} (n_{\ell,\alpha} - n_M) (n_{\ell,\beta} - n_M), \quad U \cong 1eV,$$

$$n_{\ell,\sigma} \equiv a_{\ell,\sigma}^{+} a_{\ell,\sigma}, \quad a_{\ell,\sigma} \equiv (2N)^{-1/2} \sum_{k} e^{ik \cdot \ell} a_{k,\sigma},$$

The whole electronic system is always in a plan wave state only around the bottom of the conduction band minimum, with only a low carrier density, well described by one-boy H<sub>e</sub>, and effects of H<sub>i</sub> is weak.

Density matrix at a time t  $\rho(t)$ 

$$\rho(t) \rightarrow \rho_e(t) \rho_p \text{,} \qquad \qquad \rho_p \equiv e^{-H_p/k_B T_p} \text{,} \quad T_p = 0 \text{K} \label{eq:rho_p}$$

Phonon system is always heat reservoir.

$$< n_{\boldsymbol{\ell},\sigma}(t)> \equiv Tr(n_{\boldsymbol{\ell},\sigma}\rho(t))/\,Tr(\rho(t)), \qquad < \cdots > \equiv Tr(\cdots\rho(t))/\,Tr(\rho(t)),$$
 
$$< n_{\boldsymbol{\ell},\sigma}(t)> \to n_M, \qquad \text{independent of time} \quad t$$

The first order effect is always absent

$$< H_i > = < H_{ee} > = < H_{ep} > = 0$$

## I. Statistical relaxation theory, Electron temperature ( $\equiv T_e(t)$ ) is always well defined.

At each time t, electron temperature  $T_e(t)$  (,  $0 \le T_e(t) \le 200$ K) is always well established in electronic system, prescribed by one-body H<sub>e</sub>, due to intra-system multiple scattering by H<sub>ee</sub>, but gradually decreases, as it releases its energy to the phonon system through  $H_{ep}$ . We can forget about  $H_{ee}$ , except  $T_{e}(t)$ 

$$\mathbf{H}_{\mathrm{i}} \rightarrow \mathbf{H}_{\mathbf{ep}}$$

Density matrix is

$$\rho(t) \rightarrow \rho_e(t)\rho_p, \ \rho_e(t) \equiv e^{-\frac{H_e}{k_BT_e(t)}}$$

Total energy decrease of electrons, due to temperature decrease from  $T_e$  to  $(T_e - \Delta T_e)$ ,  $\rightarrow$  Electronic heat capacity  $(, \equiv C(T_e))$ 

$$\begin{split} C(T_e) = & \frac{\partial < H_e >}{\partial T_e} \text{,} \qquad < H_e > = \sum\nolimits_{k,\sigma(=\alpha,\beta)} (e_k - \mu) < n_{k,\sigma} > \text{,} \quad n_{k,\sigma} \equiv a_{k,\sigma}^+ a_{k,\sigma} \\ \text{Fermi distribution:} & < n_{k,\sigma} > = \frac{e^{-(e(k)-\mu)/k_BT_e}}{1 + e^{-(e(k)-\mu)/k_BT_e}} \text{,} \end{split}$$

$$< n_{k,\sigma} > = \frac{e^{-(e(k)-\mu)/k_BT_e}}{1+e^{-(e(k)-\mu)/k_BT_e}}$$

where,  $\mu(T_e)$  should be determined at given  $T_e$  from the self-consistent condition

$$n_{M}\!=\!\left(2N
ight)^{-1}\sum_{k,\sigma(=lpha,eta)}< n_{k,\sigma}>$$

Thus we get

$$\Delta < H_e(T_e) > = C(T_e)\Delta T_e, \quad C(T_e) \propto T_e$$

which is well known to be linear at low temperature? Luttinger, PR 119(1960)1153.

# Total energy increase of phonon system through second order of $H_{ep}$ , within a time interval $\Delta t$ from t.

Time evolution of  $\rho(t + \Delta t)$  from  $\rho(t)$ 

$$\rho(t + \Delta t) = e^{-i\Delta tH} \rho_e(t) \rho_p e^{i\Delta tH}$$

$$< H_p(t + \Delta t) > \equiv Tr \left( H_p e^{-i\Delta t H} \rho_e(t) \rho_p e^{i\Delta t H} \right) / Tr \left( \rho_e(t) \rho_p \right),$$

$$e^{-i\Delta tH} = \ e^{-i\Delta t(H_0+H_i)} = e^{-i\Delta tH_0} \ exp_+ \left\{ -i \int_0^{\Delta t} d\tau \ \widetilde{H}_i(\tau) \right\} \!, \label{eq:epsilon}$$

exp<sub>+</sub> positive chronologically ordered exponential, from left to right

$$e^{i\Delta t H_0} e^{-i\Delta t H} = \exp_+ \left\{ -i \int_0^{\Delta t} d\tau \, \widetilde{H}_i(\tau) \right\}$$

Here, the interaction representation  $\tilde{O}$  of an operator O is

$$\widetilde{\mathbf{O}}(\Delta t) \equiv e^{i\Delta t H_0} \mathbf{O} e^{-i\Delta t H_0}$$

Its straight forward expansion is

$$\begin{split} exp_+ \left\{ -i \int_0^{\Delta t} d\tau \ \widetilde{H}_i(\tau) \right\} &= 1 + (-i) \int_0^{\Delta t} d\tau_1 \ \widetilde{H}_i(\tau_1) + (-i)^2 \int_0^{\Delta t} d\tau_1 \int_0^{\tau_1} d\tau_2 \widetilde{H}_i(\tau_1) \widetilde{H}_i(\tau_2) \\ &+ (-i)^3 \int_0^{\Delta t} d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 \widetilde{H}_i(\tau_1) \widetilde{H}_i(\tau_2) \widetilde{H}_i(\tau_3) + \dots \dots \quad , \end{split}$$

Its complex conjugate

$$e^{i\Delta tH}=\;e^{i\Delta t(H_0+H_i)}=exp_-\left\{i\int_0^{\Delta t}\!d\tau'\;\widetilde{H}_i(\tau')\right\}\,e^{i\Delta tH_0}\text{,}$$

exp\_ negative chronologically ordered exponential from right to left.

$$e^{i\Delta tH}e^{-i\Delta tH_0} = \exp_{-}\left\{i\int_{0}^{\Delta t} d\tau' \,\widetilde{H}_i(\tau')\right\}$$

Its straight forward expansion is

$$\begin{split} exp_{-} \left\{ i \int_{0}^{\Delta t} d\tau' \, \widetilde{H}_{i}(\tau') \right\} &= 1 + (i) \int_{0}^{\Delta t} d\tau'_{1} \, \widetilde{H}_{i}(\tau'_{1}) + (i)^{2} \int_{0}^{\Delta t} d\tau'_{1} \int_{0}^{\tau'_{1}} d\tau'_{2} \widetilde{H}_{i}(\tau'_{2}) \widetilde{H}_{i}(\tau'_{1}) \\ &+ (i)^{3} \int_{0}^{\Delta t} d\tau'_{1} \int_{0}^{\tau'_{1}} d\tau'_{2} \int_{0}^{\tau'_{2}} d\tau'_{3} \widetilde{H}_{i}(\tau'_{3}) \widetilde{H}_{i}(\tau'_{2}) \widetilde{H}_{i}(\tau'_{1}) + \dots \dots , \end{split}$$

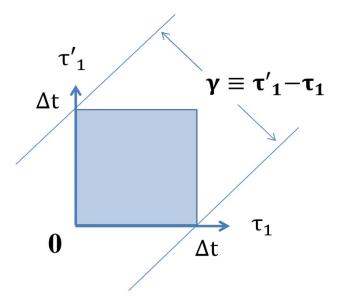
Thus

$$\rho(t+\Delta t) = e^{-i\Delta t H_0} \; exp_+ \left\{ -i \int_0^{\Delta t} \! d\tau \; \widetilde{H}_i(\tau) \right\} \rho_e(t) \rho_p exp_- \left\{ i \int_0^{\Delta t} \! d\tau' \; \widetilde{H}_i(\tau') \right\} \, e^{i\Delta t H_0}, \label{eq:rho_total_prob}$$

Non zero second order term at phonon vacuum,  $\rho_p$  at  $T_p$  (= 0 K)

$$<\text{H}_{\text{p}}(t+\Delta t)> = \frac{\text{Tr}\left(\text{H}_{\text{p}}\int_{0}^{\Delta t}d\tau_{1}\,\widetilde{\text{H}}_{\text{ep}}(\tau_{1})\rho_{\text{e}}(t)\rho_{\text{p}}\int_{0}^{\Delta t}d\tau'_{1}\,\,\widetilde{\text{H}}_{\text{ep}}(\tau'_{1})\right)}{\text{Tr}\big(\rho_{\text{e}}(t)\rho_{\text{p}}\big)}$$

$$=\int\limits_{0}^{\Delta t}d\tau'_{1}\int\limits_{0}^{\Delta t}d\tau_{1}\frac{\text{Tr}\big(\widetilde{H}_{ep}(\tau'_{1})H_{p}\widetilde{H}_{ep}(\tau_{1})\rho_{e}(t)\rho_{p}\big)}{\text{Tr}\big(\rho_{e}(t)\rho_{p}\big)}=\Delta t\int\limits_{-\Delta t}^{\Delta t}d\gamma\frac{\text{Tr}\big(\widetilde{H}_{ep}(\gamma)H_{p}\widetilde{H}_{ep}(0)\rho_{e}(t)\rho_{p}\big)}{\text{Tr}\big(\rho_{e}(t)\rho_{p}\big)}$$



= .......

 $\Delta t \rightarrow \infty$ , long time limit

So called golden rule

$$\begin{split} \int\limits_{-\Delta t}^{\Delta t} \mathrm{d}\gamma \, \mathrm{e}^{-\mathrm{i}\bar{\mathrm{e}}(k-q)\gamma t} (1-&< n_{k-q,\sigma}>) \mathrm{e}^{\mathrm{i}\bar{\mathrm{e}}(k)\gamma t} < n_{k,\sigma}> \omega_{\mathrm{q}} \mathrm{e}^{-\mathrm{i}\omega_{\mathrm{q}}\gamma} \\ &= 2\pi \Delta t \omega_{\mathrm{q}} (1-&< n_{k-q,\sigma}>) < n_{k,\sigma}> \delta(\omega_{\mathrm{q}}+\bar{\mathrm{e}}(k-\mathrm{q})-\bar{\mathrm{e}}(k)) \\ &= \Delta t \Gamma(T_{e}), \end{split}$$

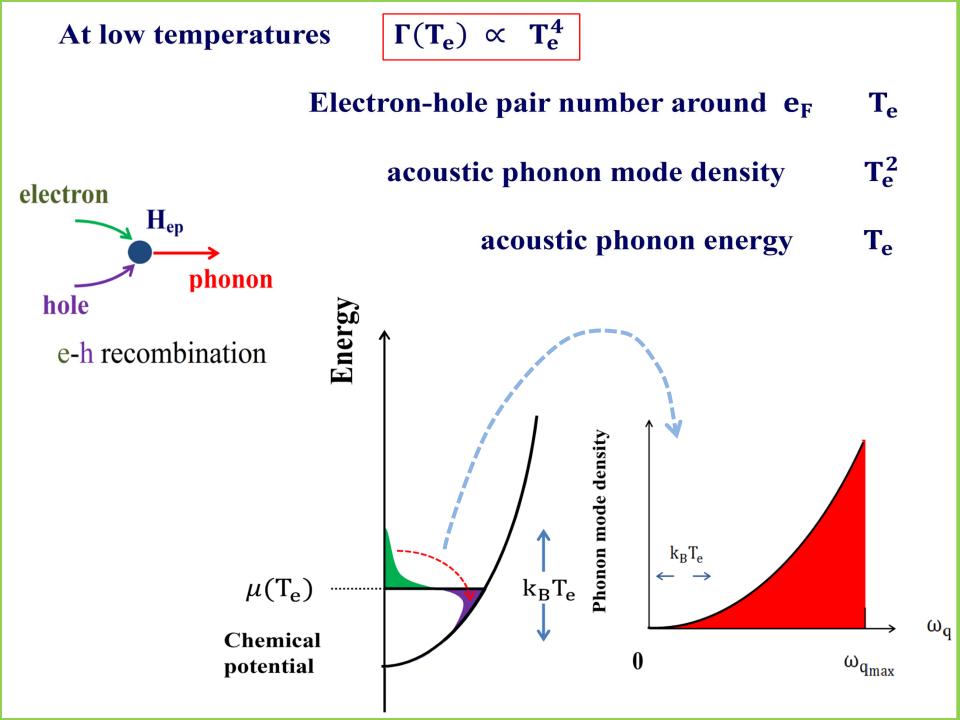
electron

### e-h recombination by phonon emission

$$\Gamma(T_e) = 2\pi \frac{S^2}{2N} \sum\nolimits_{q,k,\ \sigma(=\alpha,\beta)} \omega_q(1 - < n_{k-q,\sigma} >) < n_{k,\sigma} > \delta \left( \omega_q + e(k-q) - e(k) \right)$$

$$C(T_e)|\Delta T_e| = \Gamma(T_e)\Delta t$$
,  $\frac{\partial T_e}{\partial t} = -\frac{\Gamma(T_e)}{C(T_e)}$ 

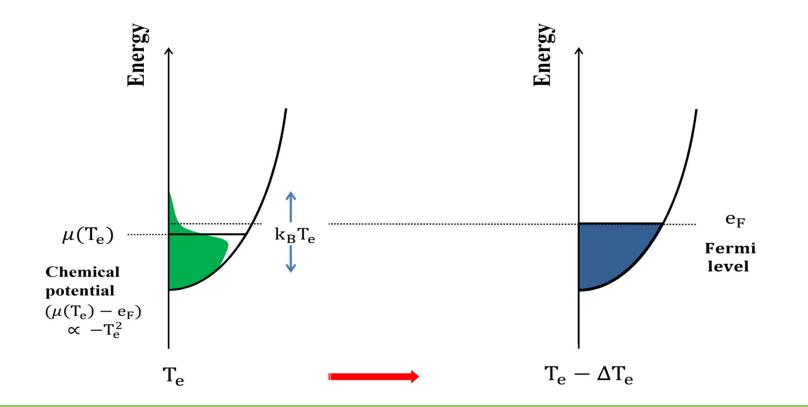
Thus, we finally get the equation for electronic system cooling.



While, total energy decrease of electrons, due to temperature decrease from  $T_e$  to  $(T_e - \Delta T_e)$ , at low temperatures

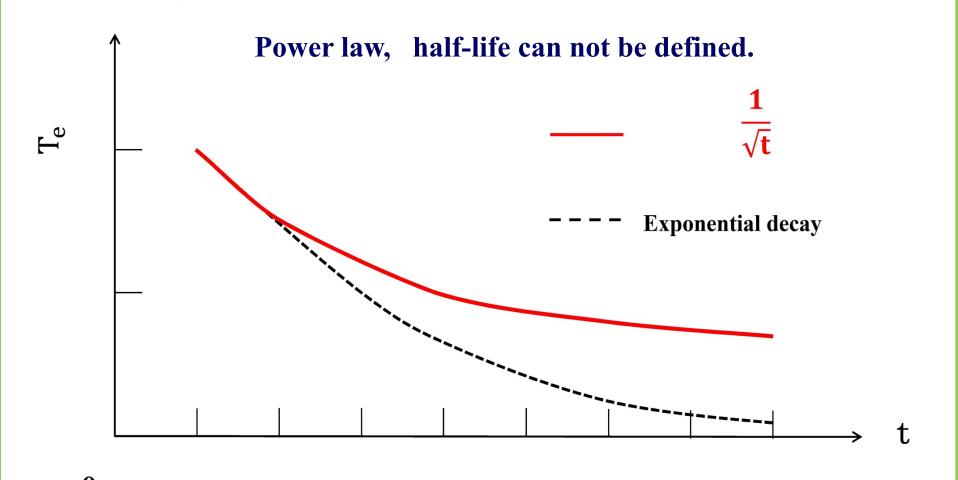
$$[<\text{H}_{\text{e}}(\text{T}_{\text{e}})>-<\text{H}_{\text{e}}(\text{T}_{\text{e}}-\Delta\text{T}_{\text{e}})>]\propto~\text{T}_{\text{e}}^2\rightarrow\text{T}_{\text{e}}|\Delta\text{T}_{\text{e}}|$$

 $|energy - e_F| \times (number of electron, or hole around e_F) \\ k_B T_e \qquad \qquad k_B T_e/e_F$ 

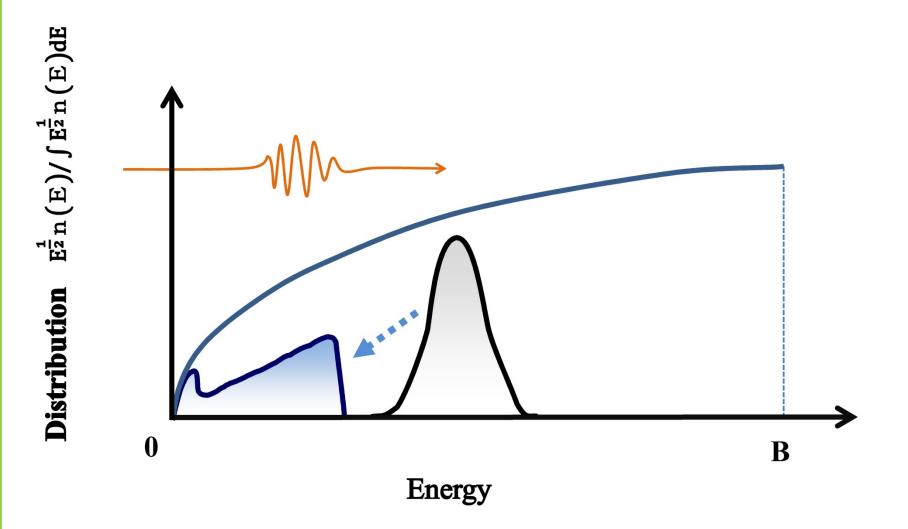


$$T_{\rm e} |\Delta T_{\rm e}| \propto \Delta t \, T_{\rm e}^4, \quad \frac{\partial T_{\rm e}}{\partial t} \propto - T_{\rm e}^3, \quad T_{\rm e} \propto t^{-\frac{1}{2}}$$

#### Slowing down of relaxation speed than exponential decay.



# Iterative theory for real time relaxation dynamic without electron temperature approximation



# II.Iterative theory for relaxation dynamics without electron temperature approximation

We now recover the full interactions for multiple scattering.

$$\mathbf{H}_{i} = (\mathbf{H}_{ep} + \mathbf{H}_{ee}),$$

$$ho(t) 
ightarrow 
ho_e(t)
ho_p, \quad 
ho_p \equiv e^{-rac{H_p}{k_B T_p}}, \ T_p = 0K$$

 $\rho_e(t)$  is now non-equilibrium state starting from the photo-excitation.

$$\tilde{\rho}(t+\Delta t) = exp_{+} \left\{ -i \int_{0}^{\Delta t} d\tau \; \widetilde{H}_{i}(\tau) \right\} \rho_{e}(t) \rho_{p} exp_{-} \left\{ i \int_{0}^{\Delta t} d\tau' \; \widetilde{H}_{i}(\tau') \right\}$$

Second order time evolution  $\Delta t$  from the transient state at t

$$\begin{split} \widetilde{\rho}(t+\Delta t) &= \rho_e(t)\rho_p \\ &+ \int_0^{\Delta t} d\tau_1 \, \widetilde{H}_{ep}(\tau_1) \rho_e(t) \rho_p \int_0^{\Delta t} d\tau'_1 \, \widetilde{H}_{ep}(\tau'_1) \\ &- \int_0^{\Delta t} d\tau_1 \int_0^{\tau_1} d\tau_2 \, \widetilde{H}_{ep}(\tau_1) \widetilde{H}_{ep}(\tau_2) \, \, \rho_e(t) \rho_p \, - \rho_e(t) \rho_p \int_0^{\Delta t} d\tau'_1 \int_0^{\tau'_1} d\tau'_2 \, \, \widetilde{H}_{ep}(\tau'_2) \, \widetilde{H}_{ep}(\tau'_1) \\ &+ \int_0^{\Delta t} d\tau_1 \, \widetilde{H}_{ee}(\tau_1) \, \rho_e(t) \rho_p \int_0^{\Delta t} d\tau'_1 \widetilde{H}_{ee}(\tau'_1) \end{split}$$

$$- \int_{0}^{\Delta t} d\tau_{1} \int_{0}^{\tau_{1}} d\tau_{2} \, \widetilde{H}_{ee}(\tau_{1}) \widetilde{H}_{ee}(\tau_{2}) \, \, \rho_{e}(t) \rho_{p} \, - \rho_{e}(t) \rho_{p} \int_{0}^{\Delta t} d\tau'_{1} \int_{0}^{\tau'_{1}} d\tau'_{2} \, \, \widetilde{H}_{ee}(\tau'_{2}) \, \widetilde{H}_{ee}(\tau'_{1})$$

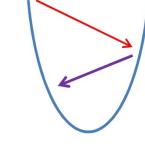
What we want know finally is the time evolution of electron number  $n_{k,\sigma}(t + \Delta t)$ 

$$rac{\partial n_{k,\sigma}(t+\Delta t)}{\partial \Delta t}$$
 ,  $n_{k,\sigma}(t+\Delta t) \equiv \langle n_{k,\sigma} \, 
angle$ 

# Rate equation for $n_{k,\sigma}(t)$

Gain of  $n_{k,\sigma}(t)$  proportional to  $(1-\langle n_{k,\sigma} \rangle)$ , loss proportional to  $\langle n_{k,\sigma} \rangle$ .

$$\frac{\partial n_{k,\sigma}(t)}{\partial t} = (1 - \langle n_{k,\sigma}(t) \rangle)(\Gamma_{ep,k}^+(t) + \Gamma_{ee,k}^+(t)) - \langle n_{k,\sigma}(t) \rangle (\Gamma_{ep,k}^-(t) + \Gamma_{ee,k}^-(t))$$



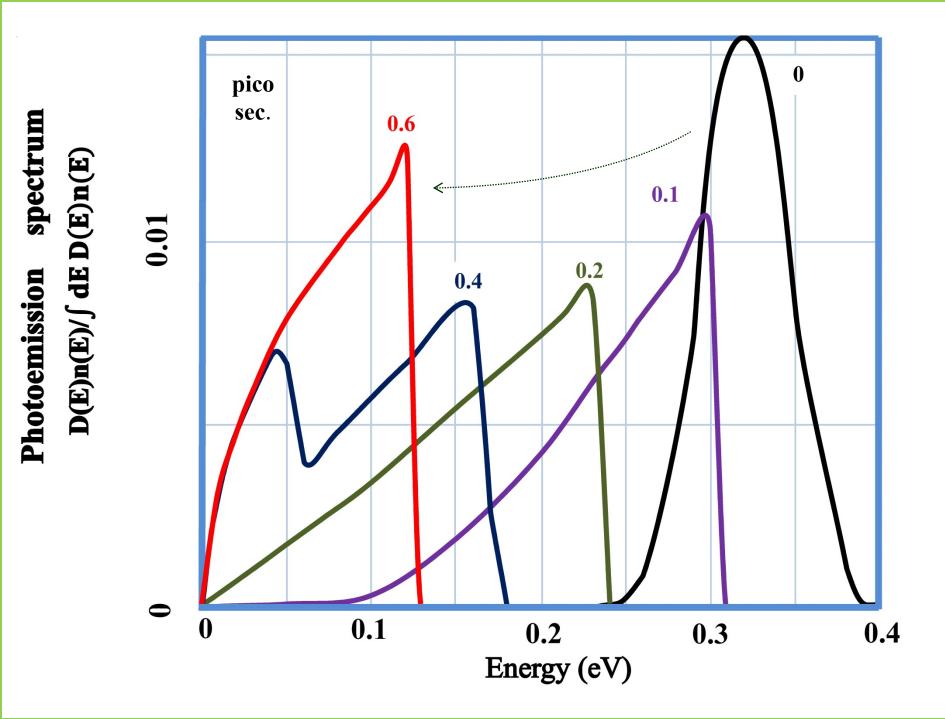
 $\Gamma_{ep,k}^{+} \equiv \pi S^2 N^{-1} \sum_{q} < n_{k+q,\sigma} > \delta(e(k) + \omega_q - e(k+q))$   $\Gamma_{ep,k}^{-} \equiv \pi S^2 N^{-1} \sum_{q} (1 - < n_{k+q,\sigma} >) \, \delta(e(k+q) + \omega_q - e(k))$ 

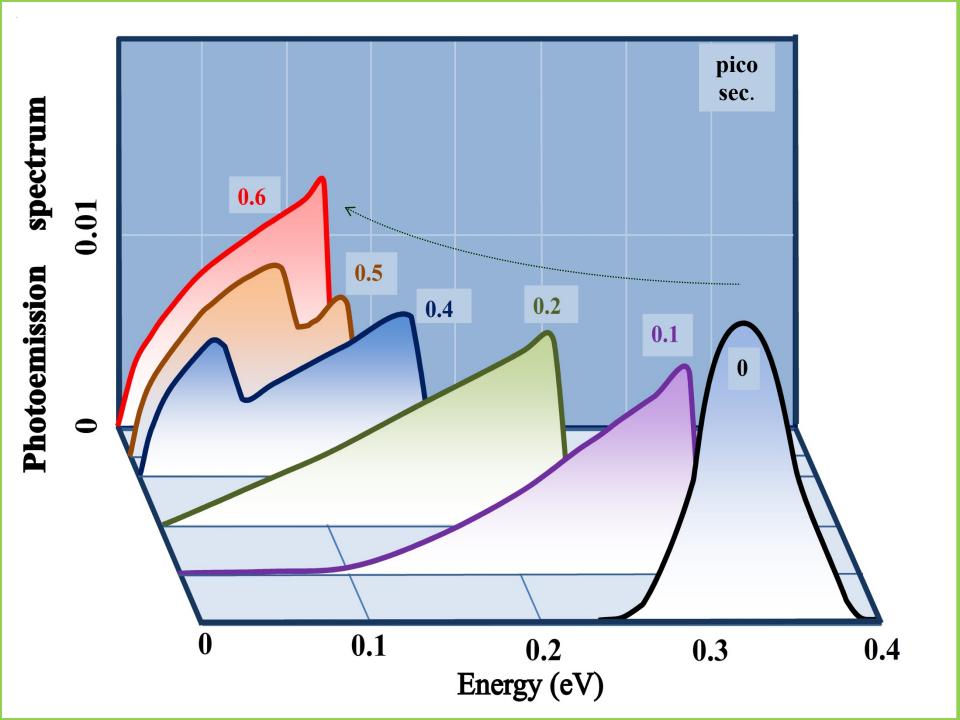
 $\Gamma_{ee,k}^{+} \equiv 2\pi U^{2}N^{-2} \sum\nolimits_{l,l'} \left(1 - < n_{k',-\sigma} > \right) \sum\nolimits_{l,l''} < n_{k-k'',\sigma} > < n_{k'+k'',-\sigma} > \delta(e(k) + e(k') - e(k-k'') - e(k'+k''))$ 

 $\Gamma_{ee,k}^{-} \equiv 2\pi U^2 N^{-2} \sum\nolimits_{k',-\sigma} > \sum\nolimits_{k',-\sigma} > \sum\nolimits_{k',-\sigma} > (1-< n_{k+k'',\sigma}>) (1-< n_{k'-k'',-\sigma}>) \delta(e(k+k'') + e(k'-k'') - e(k) - e(k'))$ 

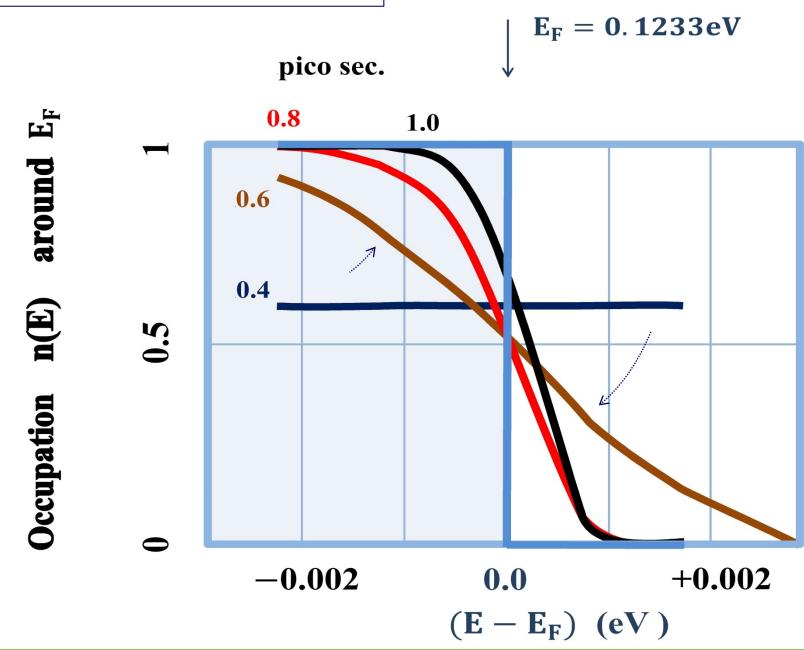
**Iterative theory for dynamics**  $n_{k,\sigma}(t+\Delta t) = n_{k,\sigma}(t) + \Delta t \big\{ (1 - < n_{k,\sigma}(t) >) (\Gamma_{ep,k}^+(t) + \Gamma_{ee,k}^+(t)) - < n_{k,\sigma}(t) > (\Gamma_{ep,k}^-(t) + \Gamma_{ee,k}^-(t)) \big\}$ 

 $n_{k,\sigma}(t + 2\Delta t) = n_{k,\sigma}(t + \Delta t) + \Delta t \left\{ (1 - \langle n_{k,\sigma}(t + \Delta t) \rangle)(\Gamma_{ep,k}^+(t + \Delta t) + \Gamma_{ee,k}^+(t + \Delta t)) - \langle n_{k,\sigma}(t + \Delta t) \rangle (\Gamma_{ep,k}^-(t + \Delta t) + \Gamma_{ee,k}^-(t + \Delta t)) \right\}$ 

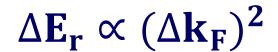


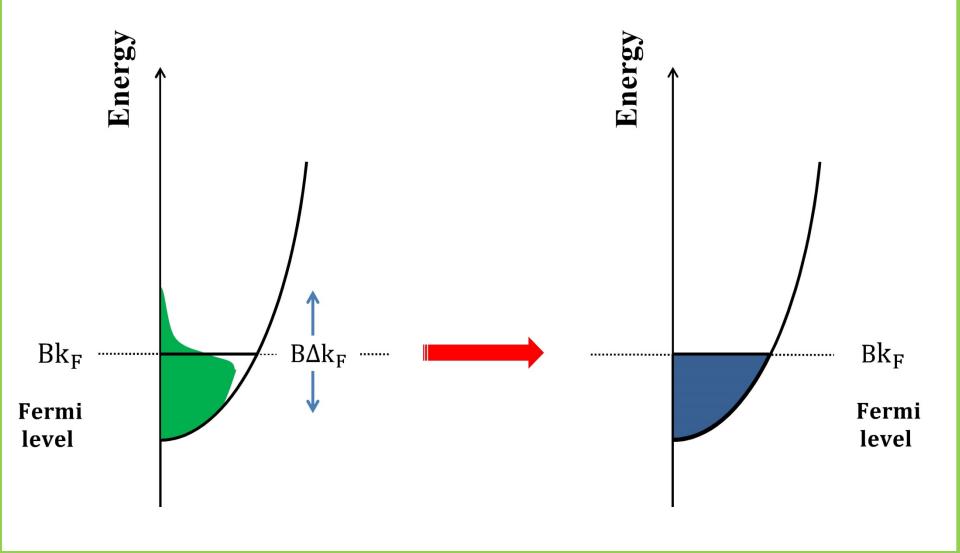


## Occupation around E<sub>F</sub>



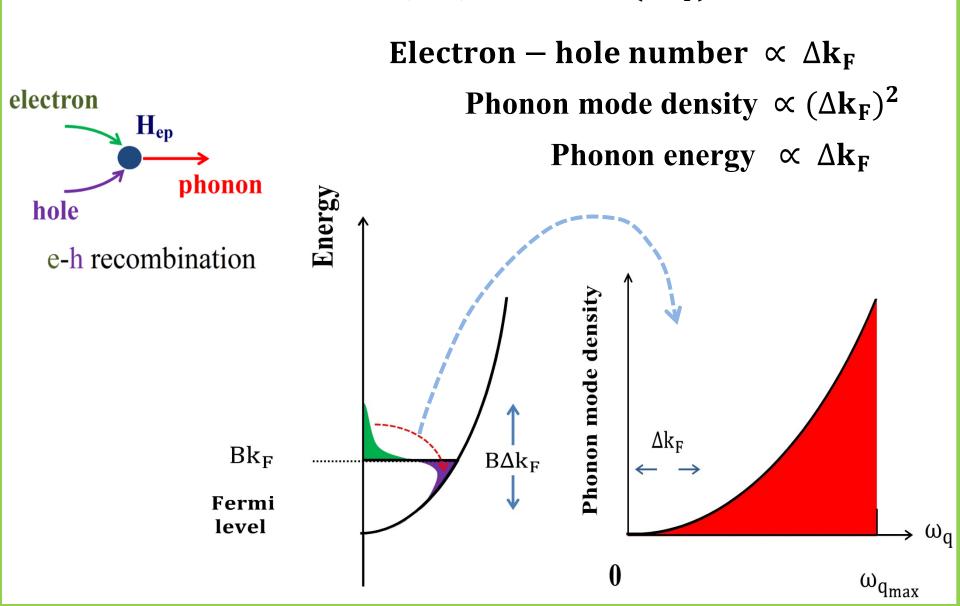
#### Residual photo-excitation energy ( $\equiv \Delta E_r$ ) at final stage





#### Final stage of phonon relaxation

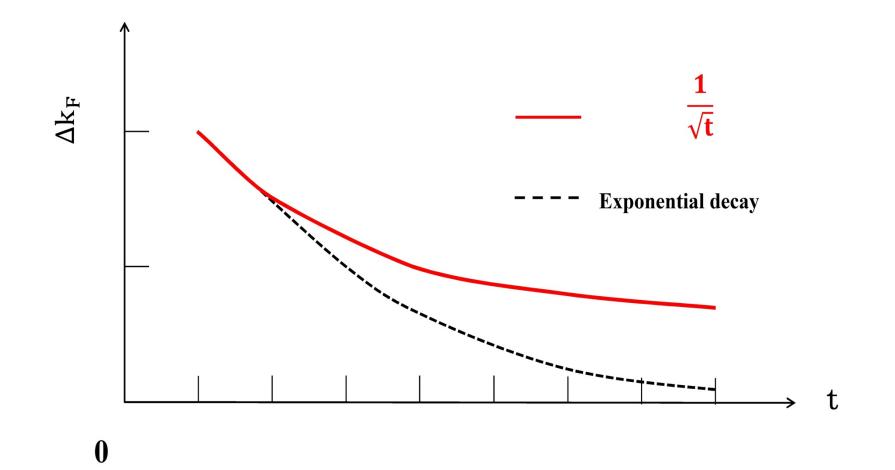
Transition rate  $(\equiv \Gamma)$ ,  $\Gamma \propto (\Delta k_F)^4$ 



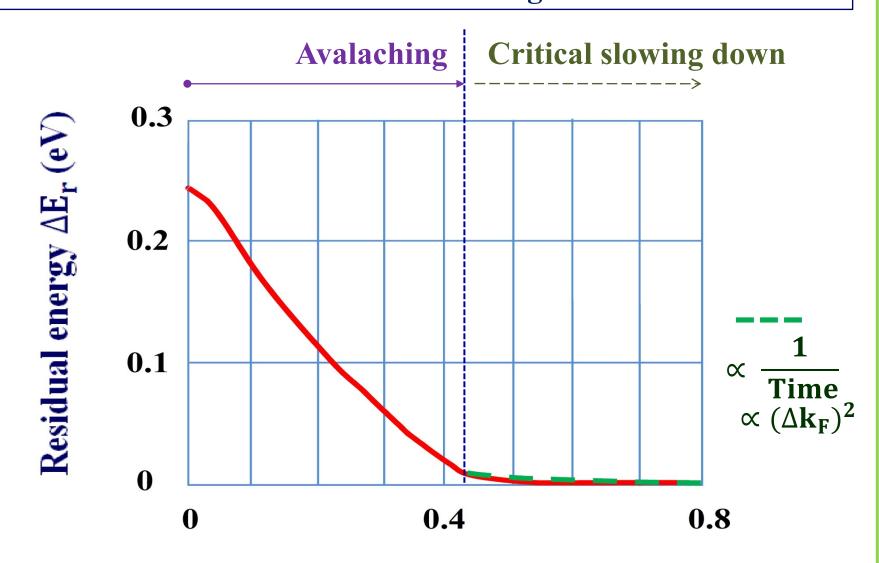
#### **Finally**

$$(\Delta k_F)^2 \propto \Delta t (\Delta k_F)^4$$
,  $\frac{\partial (\Delta k_F)}{\partial t} \propto (\Delta k_F)^3$ ,  $\Delta k_F \propto t^{-\frac{1}{2}}$ 

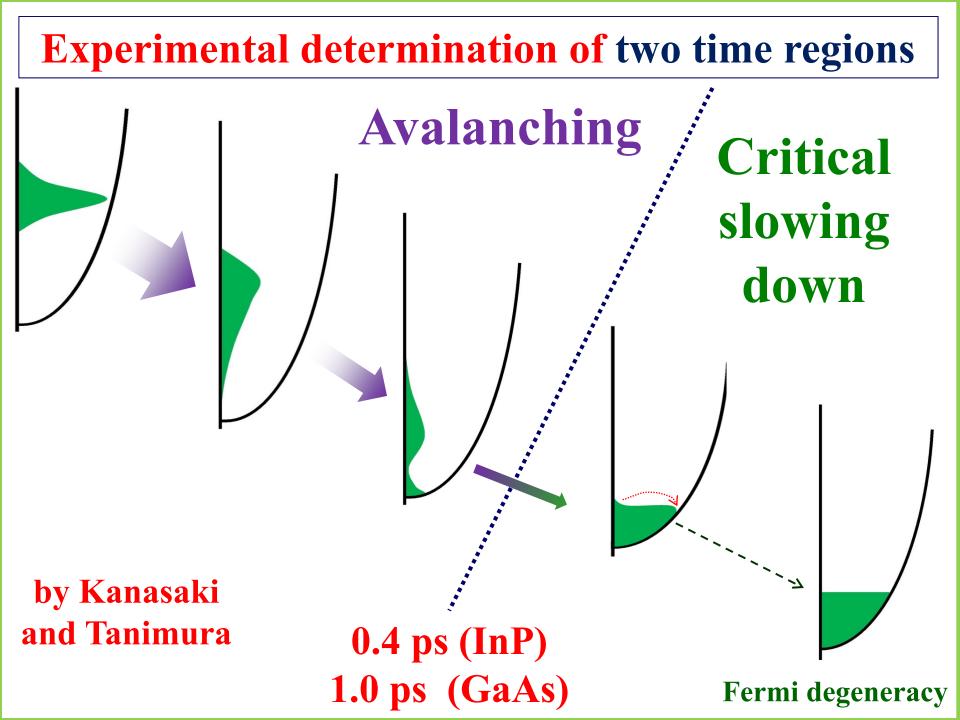
#### Slowing down of relaxation speed than exponential decay.



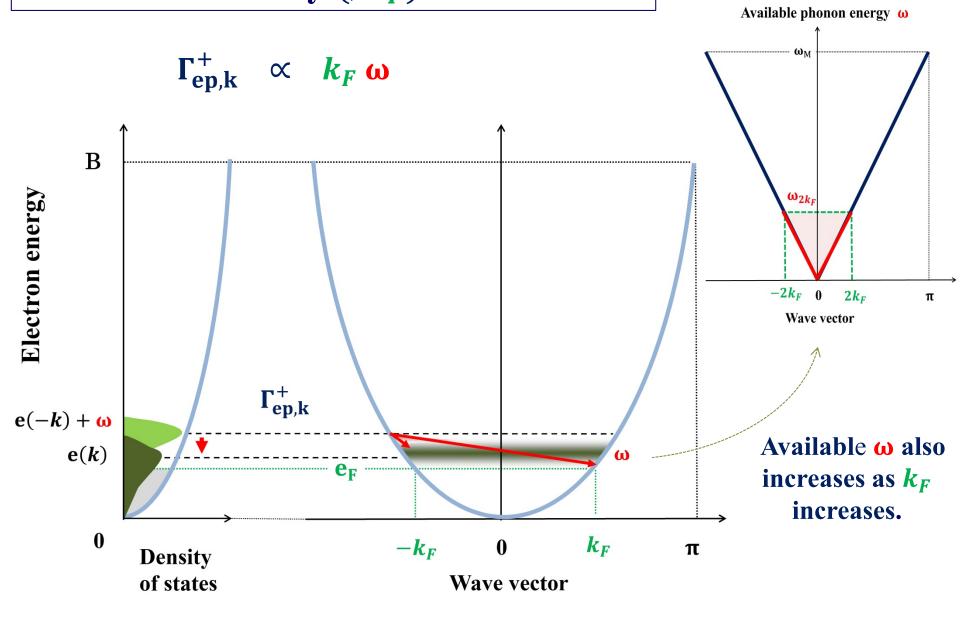
# Relaxation dynamics of residual excitation energy $\Delta E_r$ , Theory Two time regions



Time after photo-excitation (ps)



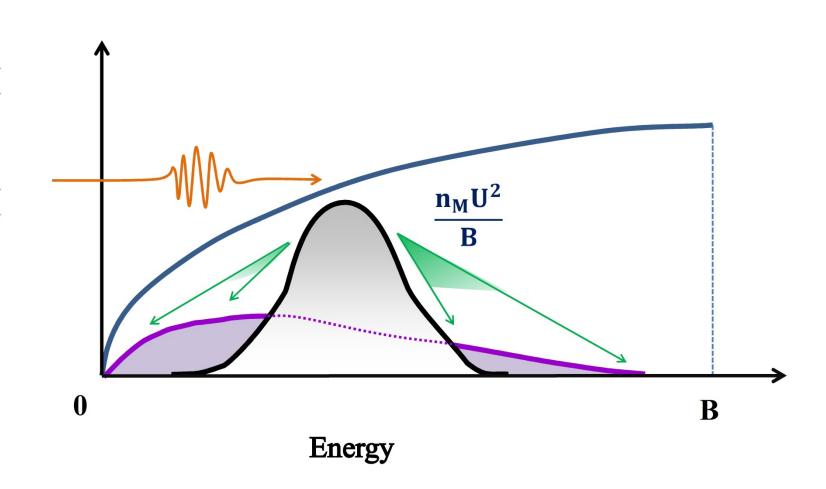
# Avalanching speed rapidly increases as electron density $(, k_F)$ increases.



# How much time necessary to photo-generate Fermi surface from true electron vacuum?

Never terminates.



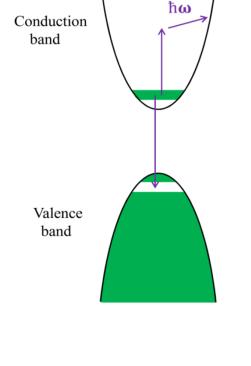


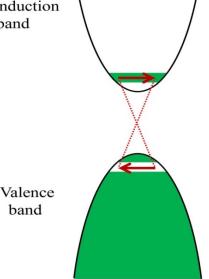
 $n_M \lesssim 10^{-3}$ , rare than e-ph

#### **Decay channels**

- 1. Radiative recombination of e-h pair,  $10^{-9} \text{ sec}$
- 2. Momentum, charge and spin fluctuations give no energy dissipation
- 3. Auger recombination of e-h pair with no energy dissipation,  $10^{-12} \sec$ Conduction

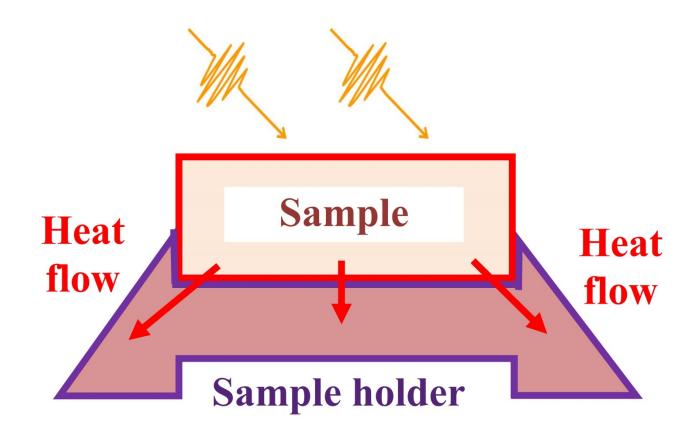
Inter-band Coulomb scattering, similarly to the intra-band one, gives no dissipation.





band

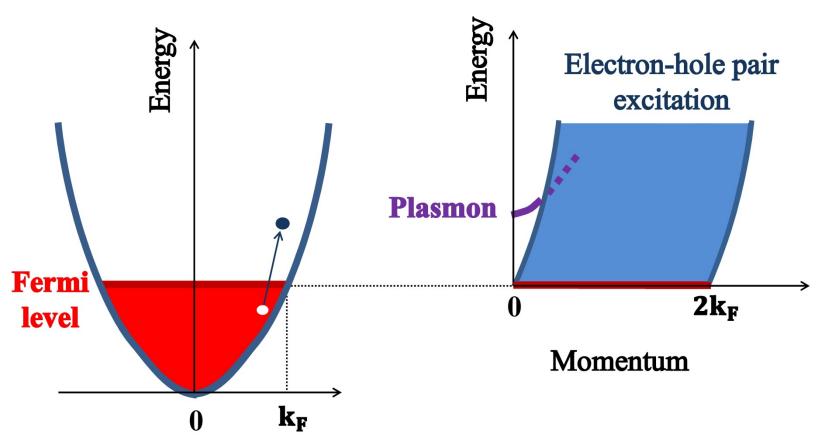
#### Importance of thermodynamic boundary condition



Uncontrolled boundary condition gives uncontrolled experimental results.

#### **Plasmon**

is the coulombic anti-bound state between electron-hole, above the well-established Fermi distribution.



Momentum

P. Anderson, Phys. Rev. 112(1958)1900.