

# Quantum Transport:

ATOM TO TRANSISTOR

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## Lecture 36: Radiative Transitions

Ref. Chapter 10.1 & 10.2



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## Summarizing previous lectures

- We want to calculate the rate at which an electron in an upper level relaxes to a lower level and emits light.

- There are two views which may be used to understand this phenomena:

**1. The one-particle picture (Easy to understand but conceptually problematic):**

- The reason electron relaxes from 2 to 1 is because it feels some potential due to the surroundings. This potential is the scattering potential in the Hamiltonian matrix. But to explain what we observe the potential that relaxes the electron should be bigger than the one exciting it.

- The 1-level picture:  
→ Results in a non-Hermitian Hamiltonian matrix

$$i\hbar \frac{d}{dt} \begin{Bmatrix} \psi_1 \\ \psi_2 \end{Bmatrix} = \begin{bmatrix} \varepsilon_1 & [U_S]_{12} \\ [U_S]_{21} & \varepsilon_2 \end{bmatrix} \begin{Bmatrix} \psi_1 \\ \psi_2 \end{Bmatrix}$$

where  $[U_S]_{12} \propto (N+1)$  and  $[U_S]_{21} \propto (N)$

Note:  $N$  is the number of photons present such that

$$N = \frac{1}{e^{\hbar\omega/k_B T} + 1}$$

## 2. The multi-particle picture (More difficult to understand):

- Radiative lifetime is understood in terms of  $N$  to  $N+1$  photon system transitions. Notice that in that big system  $N$  and  $N+1$  are degenerate levels and any small coupling will take electron to the other level. We will use this picture to write down the appropriate radiative lifetime.

For this we can use the ideas that we used when discussing contacts:

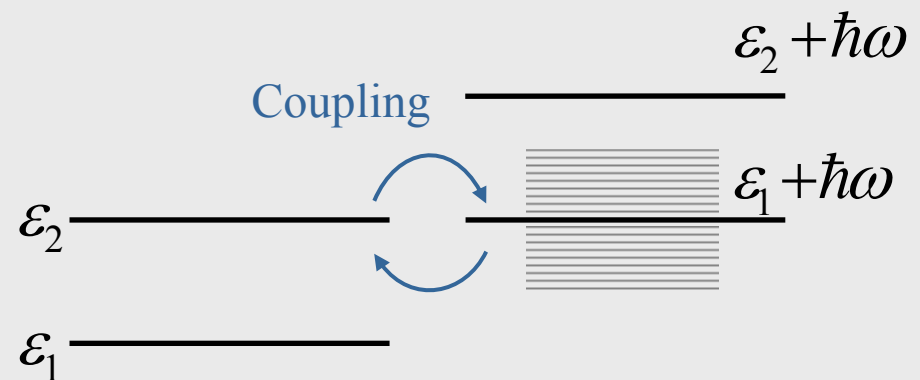
- We can apply the NEGF method to a multi-particle system formed as a product of the  $n$  electron and  $N$  photon, that is

$$|n, N\rangle = |n\rangle \otimes |N\rangle$$

- With NEGF one may calculate both broadening and the transition rate i.e. for the zero photon subspace

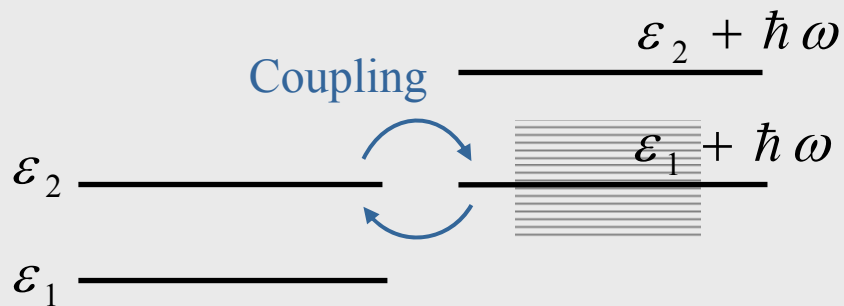
$$\Gamma = \frac{\hbar}{\tau} = \tau_c a \tau_c^+ = \sum |K|^2 \delta(E - \varepsilon_1 - \hbar\omega)$$

Level coupling



- So we need to find the coupling  $|K|$  in

$$\Gamma = \frac{\hbar}{\tau} = \sum |K|^2 \delta(\varepsilon_2 - \varepsilon_1 - \hbar\omega)$$



- This view is not exact since the  $N$  photon subspace is coupled to the  $N+1$  subspace which is in turn coupled to the  $N+2$  subspace, etc. And this will result in not completely accurate spectral function  $a$ .

- To calculate coupling between the 0-photon and 1-photon states we use the vector potential due to 1 photon:

$$\vec{A} = \hat{v} \frac{A_0}{2} \left( e^{i\vec{\beta} \cdot \vec{r}} e^{-i\omega_\beta t} + e^{-i\vec{\beta} \cdot \vec{r}} e^{i\omega_\beta t} \right)$$

where  $A_0 = \sqrt{\frac{2\hbar}{\omega\varepsilon\Omega}}$

Note: Due to canceling terms the final expression for  $\Gamma$  is independent of the volume of the box  $\Omega$

# Vector Potential

- In the photon vector potential we denote

$$\vec{A}_{\text{abs}}(\vec{r})e^{-i\omega_{\beta}t} = \hat{v} \frac{A_0}{2} \left( e^{i\vec{\beta}\cdot\vec{r}} e^{-i\omega_{\beta}t} \right)$$

as the absorption term and

$$\vec{A}_{\text{em}}(\vec{r})e^{+i\omega_{\beta}t} = \hat{v} \frac{A_0}{2} \left( e^{-i\vec{\beta}\cdot\vec{r}} e^{+i\omega_{\beta}t} \right)$$

as the emission term

- Thus, the emission coupling term is

$$\begin{aligned} K_{\text{em}} &= \int d\vec{r} \phi_1^* (q/m) (\vec{A}_{\text{em}} \cdot \vec{p}) \phi_2(\vec{r}) \\ &= \frac{qA_0}{2m} \int d\vec{r} e^{i\vec{\beta}\cdot\vec{r}} \phi_1^*(\vec{r}) (\hat{v} \cdot \vec{p}) \phi_2(\vec{r}) \end{aligned}$$

Note: from the original operator

$$(\vec{p} + q\vec{A}) \cdot (\vec{p} + q\vec{A}) / 2m$$

we neglect  $\vec{A} \cdot \vec{A}$  as it is assumed to be very small and for

$$\vec{p} \cdot (\vec{A} \varphi) = \vec{A} \cdot (\vec{p} \varphi) + \varphi (\vec{p} \cdot \vec{A})$$

we have  $\vec{\nabla} \cdot \vec{A} = 0 \Rightarrow \vec{p} \cdot \vec{A} = \vec{A} \cdot \vec{p}$

# Emission Coupling

- To evaluate  $K_{\text{abs}}$  we simply replace  $e^{i\vec{\beta}\cdot\vec{r}}$  by  $e^{-i\vec{\beta}\cdot\vec{r}}$  in  $K_{\text{em}}$
- So now we know how to add a vector potential to the Schrödinger equation
- Phonons induce scalar rather than vector potentials and are much more easily incorporated into the Schrödinger equation

- However,  $e^{\pm i\vec{\beta}\cdot\vec{r}}$  is often ignored when analyzing effects on the atomic scale because  $\vec{\beta}$  varies on the scale of microns and  $\phi_{1,2}$  on the scale of angstroms. This leads to the approximation

$$K_{\text{em/abs}} = \frac{qA_0}{2m} \hat{v} \cdot \vec{p}$$

where  $\vec{p} = \int d\vec{r} \phi_1^*(\vec{r}) \vec{p} \phi_2(\vec{r})$

and  $\vec{p} = -i\hbar\vec{\nabla}$

# Hydrogen Atom

- For example consider emission from the degenerate hydrogen 2s, 2p<sub>x</sub>, 2p<sub>y</sub>, 2p<sub>z</sub> levels to the 1s level

- First note,

$$\phi_{1s} = \frac{1}{\sqrt{\pi a_0^3}} e^{-r/a_0}$$

$$\phi_{2s} = \frac{1}{\sqrt{32\pi a_0^3}} \left( 2 - \frac{r}{a_0} \right) e^{-r/2a_0}$$

- Also,

$$\phi_{2p_x} = \frac{1}{\sqrt{16\pi a_0^3}} \left( \frac{x}{a_0} \right) e^{-x/2a_0}$$

$$\phi_{2p_y} = \frac{1}{\sqrt{16\pi a_0^3}} \left( \frac{y}{a_0} \right) e^{-y/2a_0}$$

$$\phi_{2p_z} = \frac{1}{\sqrt{16\pi a_0^3}} \left( \frac{z}{a_0} \right) e^{-z/2a_0}$$

- Solving for  $\vec{p}$  in  $K_{\text{em/abs}} = (qA_0/2m)\hat{v} \cdot \vec{p}$  we can see that 2s→1s emission is isotropic and the remainder (2p<sub>x</sub>→1s, 2p<sub>y</sub>→1s, and 2p<sub>z</sub>→1s) are polarized

# Hydrogen Emission

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- For  $2s \rightarrow 1s$   $\vec{p}$  is...

$$\vec{p} = \int d\vec{r} \frac{1}{\sqrt{\pi a_0^3}} e^{-r/a_0} \left( \frac{\partial}{\partial r} \hat{r} + \frac{1}{r} \frac{\partial}{\partial \theta} \hat{\theta} + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} \hat{\theta} \right) \frac{1}{\sqrt{32\pi a_0^3}} \left( 2 - \frac{r}{a_0} \right) e^{-r/2a_0}$$

$= \hat{r} \times \text{Const} \quad \therefore \text{Isotropic}$

- For  $2p_x \rightarrow 1s$   $\vec{p}$  is...

$$\vec{p} = \int d\vec{r} \frac{1}{\sqrt{\pi a_0^3}} e^{-r/a_0} \left( \hat{x} \frac{\partial}{\partial x} + \hat{y} \frac{\partial}{\partial y} + \hat{z} \frac{\partial}{\partial z} \right) \frac{1}{\sqrt{16\pi a_0^3}} \left( \frac{x}{a_0} \right) e^{-x/2a_0}$$

$= \hat{x} \times \text{Const} \quad \therefore \text{Polarized in } \hat{x} \text{ direction}$

- Likewise for  $2p_y \rightarrow 1s$  we get polarization in the  $\hat{y}$  direction and for  $2p_z \rightarrow 1s$  polarization in the  $\hat{z}$  direction



- Summary:

Transition	$\vec{p}$
$2s \rightarrow 1s$	$\hat{r}$ (isotropic)
$2p_x \rightarrow 1s$	$\hat{x}$
$2p_y \rightarrow 1s$	$\hat{y}$
$2p_z \rightarrow 1s$	$\hat{z}$

- Polarization limits the emission vector of a photon to the plane perpendicular to the direction of polarization

- Thus,  
 $2p_x \rightarrow 1s$  emits in the (y,z) plane.  
 $2p_y \rightarrow 1s$  emits in the (x,z) plane  
 $2p_z \rightarrow 1s$  emits in the (x,y) plane

- In a sense atomic transitions such as these behave just like a classical dipole such that

$$\vec{I}d = \frac{2q}{m} \vec{p}$$

Often we refer to this as an atomic dipole

## Emission Details

- In the actual hydrogen atom an electron in fact spreads out evenly amongst the 2px, 2py, and 2pz level resulting in isotropic emission.
- Note, a coupling exists between the 1s and (2px,2py,2pz) levels whose resulting self energy creates a small energy shift between the two levels. Hence, in actuality the s and p levels are in fact not degenerate

- Interestingly, one may also examine transitions between two extended states (not atomic wave functions). Such events are often referred to as intraband transitions. The intraband coupling term expressed in its simplest form is:

$$K_{\text{em/abs}} = \int d\vec{r} \left[ e^{\pm i\vec{\beta}\cdot\vec{r}} e^{-i\hat{k}'\cdot\vec{r}} \times (\vec{p} \cdot \hat{v}) e^{+i\vec{k}\cdot\vec{r}} \right]$$

- Note that for

$$K_{\text{em/abs}} = \int d\vec{r} e^{\pm i\vec{\beta}\cdot\vec{r}} e^{-i\hat{k}'\cdot\vec{r}} (\vec{p}\cdot\hat{v}) e^{+i\vec{k}\cdot\vec{r}}$$

coupling will be non-zero iff  $k - \hat{k}' \pm \vec{\beta} = 0$ . This follows from the fact that

$$e^{i(\vec{k} - \vec{k}' \pm \vec{\beta})\cdot\vec{r}} \text{ integrated over } d\vec{r} \text{ is non-zero iff } \vec{k} - \vec{k}' \pm \vec{\beta} = 0.$$

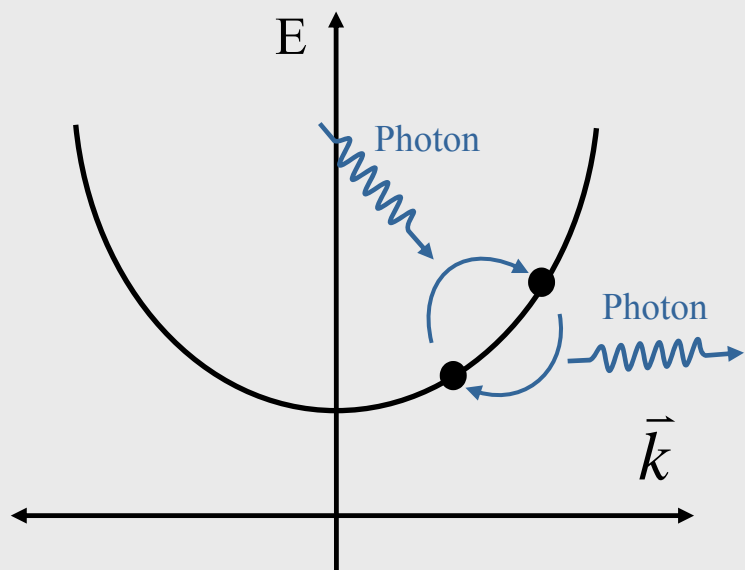
Furthermore, the relation affects the broadening term,  $\Gamma$ . For delta function to have any contribution, the argument must be 0. Thus we have:

$$\begin{aligned} \varepsilon_{\vec{k}'} &= \varepsilon_{\vec{k}} - \hbar\omega_{\vec{\beta}} & \} & \text{emission} \\ \varepsilon_{\vec{k}'} &= \varepsilon_{\vec{k}} + \hbar\omega_{\vec{\beta}} & \} & \text{absorption} \end{aligned}$$

## Final Intraband Comments

- So, to make an intraband transition, the final energy,  $\mathcal{E}_{\vec{k}'}$ , equals the initial energy,  $\mathcal{E}_{\vec{k}}$ , plus or minus  $\hbar\omega_{\vec{\beta}}$

### Intraband Transitions



- Mathematically we can show that intraband transitions occur only if the electron travels faster than the speed of light in the medium of interest. Usually this is called Cerenkov radiation. Cerenkov radiation is permitted in many solids because the speed of light is a fraction of that in a vacuum (i.e.  $1/3$ ). This effect also applies to the speed of sound and results in the frequent emission of phonons