



UNIVERSITY OF STRASBOURG

Radiation Matter Interaction Exam

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I - Coherent transients and $\pi/2$ -pulses

We consider a two-level system which stationary eigenenergies and eigenstates are noted $\{E_2, E_1\}$ and $\{|1\rangle, |2\rangle\}$ respectively. We set $E_2 - E_1 = \hbar\omega_0$, E_1 being the ground-state energy. The system is in interaction with a quasi-resonant electromagnetic field $E_0 \cos(\omega t)$. The solution of the time-dependent problem is expanded on the basis of the stationary states:

$$|\psi(t)\rangle = \tilde{C}_1(t)e^{i\delta/2t} |1\rangle + \tilde{C}_2(t)e^{-i\delta/2t} e^{-i\omega_0 t} |2\rangle$$

with

$$\begin{cases} \tilde{C}_1(t) = \cos\left(\frac{\Omega}{2}t\right) - i\frac{\delta}{\Omega} \sin\left(\frac{\Omega}{2}t\right) \\ \tilde{C}_2(t) = -i\frac{\Omega_1}{\Omega} \sin\left(\frac{\Omega}{2}t\right) \end{cases}$$

where Ω_1 is the Rabi frequency, Ω the generalized Rabi frequency and δ the detuning.

1. Give the expressions of Ω and δ . For the remainder, we assume that the incident field is tuned exactly on resonance.

We can write

$$\Omega = \sqrt{\Omega_1^2 + \delta^2}$$

but, if we are exactly on resonance, we have $\delta = 0$.

2. Write the corresponding expressions of $\tilde{C}_1(t)$, $\tilde{C}_2(t)$ and $|\psi(t)\rangle$.

$$\tilde{C}_1(t) = \cos\left(\frac{\Omega_1}{2}t\right), \quad \tilde{C}_2(t) = -i \sin\left(\frac{\Omega_1}{2}t\right)$$

leading to,

$$|\psi\rangle(t) = \cos\left(\frac{\Omega_1}{2}t\right) |1\rangle - i \sin\left(\frac{\Omega_1}{2}t\right) e^{-i\omega_0 t} |2\rangle$$

We consider the situation described in 2 but we suppose now that the electromagnetic field is applied under the form of a $\pi/2$ -pulse switched on at $t = 0$, the system being initially in the ground-state.

3. Define what a $\pi/2$ -pulse is. Calculate the numerical values of $\tilde{C}_1(t)$ and $\tilde{C}_2(t)$ that characterize the new state of the system after the interaction with the $\pi/2$ -pulse. What are the

probabilities for the system to be in state $|1\rangle$, in state $|2\rangle$? Conclusion ?

A $\pi/2$ -pulse is a pulse that is switched on at $t = 0$ and switched off at $t = \pi/2\Omega_1$. Thus,

$$\tilde{C}_1(t) = \cos\left(\frac{\Omega_1}{2} \frac{\pi}{2\Omega_1} t\right) = \frac{1}{\sqrt{2}}, \quad \tilde{C}_2(t) = -i \sin\left(\frac{\Omega_1}{2} \frac{\pi}{2\Omega_1} t\right) = -\frac{i}{\sqrt{2}}$$

Consequently,

$$|\psi(t)\rangle = \frac{1}{\sqrt{2}} |1\rangle - \frac{i}{\sqrt{2}} e^{-i\omega_0 t} |2\rangle$$

and we notice that,

$$|\langle 1|\psi\rangle|^2 = |\langle 2|\psi\rangle|^2 = \frac{1}{2}$$

Thus, the probabilities for the atom to be in either state does not change in time.

The system is still in the state prepared in 3. The operator associated with the electric dipole moment is written $\hat{D} = \begin{pmatrix} 0 & d \\ d & 0 \end{pmatrix}$

4. Explain why \hat{D} does not present any diagonal terms. Calculate $\langle \hat{D} \rangle$, the expectation value of \hat{D} . How does it evolve in time ? Use this result to explain what a coherent transient is.

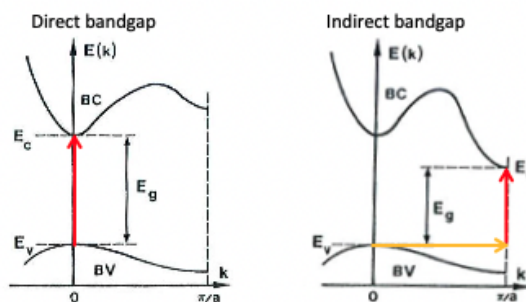
$$\begin{aligned} \langle \psi(t) | \hat{D} | \psi(t) \rangle &= \left(\frac{1}{\sqrt{2}} \langle 1 | + \frac{i}{\sqrt{2}} e^{+i\omega_0 t} \langle 2 | \right) \hat{D} \left(\frac{1}{\sqrt{2}} |1\rangle - \frac{i}{\sqrt{2}} e^{-i\omega_0 t} |2\rangle \right) \\ &= \frac{i}{2} e^{i\omega_0 t} d - \frac{i}{2} e^{-i\omega_0 t} d \\ &= \frac{id}{2} 2i \sin(\omega_0 t) \\ &= -d \sin(\omega_0 t) \end{aligned}$$

The dipole-moment oscillates at the Bohr-frequency ω_0 . This oscillation goes along with emission of light at the same frequency. Although emitted at the same frequency ω_0 as spontaneous emission between the same two energy levels, this light has different properties related to the coherence of the emission. The phase of the oscillations of the atomic dipole is uniquely determined with respect to that of the incident wave. An assembly of atoms all prepared by the same $\pi/2$ -pulse will therefore all emit light with the same phase. This is in contrast to what occurs with spontaneous emission, when individual atoms emit light with a random phase.

II - Optical transitions in a bulk semiconductor

We plan to establish the optical selection rules for an optical transition, under the influence of an electric field $E_0 \cos(\omega t - i\vec{k}_0 \cdot \vec{r})$, between the valence band ($E_v, |\psi_{v,k_v}(\vec{r})\rangle$) and the conduction band ($E_c, |\psi_{c,k_c}(\vec{r})\rangle$) of a bulk semiconductor that presents a direct bandgap located at $\vec{k} = 0$. For simplicity, we assume that the potential seen by electrons in the crystal lattice is invariant under a translation of vector \vec{a} along the three directions of space.

1. What is a direct bandgap semiconductor? What is an indirect bandgap semiconductor?



2. The periodic parts of the wave-functions are noted $u_{v,\vec{k}_v}(\vec{r})$ and $u_{c,\vec{k}_c}(\vec{r})$ for the valence band and the conduction band respectively. Write the total wave-functions $|\psi_{c,k_c}(\vec{r})\rangle$ and $|\psi_{v,k_v}(\vec{r})\rangle$ associated with the electronic bands considered here. What are the properties of $u_{v,k_v}(\vec{r})$ and $u_{c,k_c}(\vec{r})$? How are expanded the corresponding energies E_c and E_v in the reciprocal space?

$$|\psi_{v,k_v}(\vec{r})\rangle \propto N \sum_n e^{-ik_v n a} u_{v,k_v}(\vec{r}), \quad |\psi_{c,k_c}(\vec{r})\rangle \propto N \sum_n e^{-ik_c n a} u_{c,k_c}(\vec{r})$$

The properties of the Bloch-part of the wavefunction is to be periodic on the lattice-spacing. At $k \neq 0$, the dispersion lift the degeneracy between bands of different j . Near the band extrema, the Taylor expansions of the different energies are quadratic:

$$E_{c;v} = E_{c;v}(k_0) + (k - k_0) \frac{\partial E_{c;v}}{\partial k} + \frac{1}{2} (k - k_0)^2 \frac{\partial^2 E_{c;v}}{\partial k^2} + \dots$$

3. We denote $|W_{cv}|$ the matrix element of the dipole moment operator $-qE_0 \hat{r} \exp(-i\vec{k}_0 \cdot \vec{r})$ evaluated between $|\psi_{v,k_v}(\vec{r})\rangle$ and $|\psi_{c,k_c}(\vec{r})\rangle$. What is the general definition of optical selection rules associated with the transition between two electronic states?

$$|W_{cv}| = -qE_0 \langle c, k_c | \hat{r} e^{-i\vec{k}_0 \cdot \vec{r}} | v, k_v \rangle \propto \int u_{c,k_c}^*(\vec{r}) u_{v,k_v}(\vec{r}) e^{i\vec{r} \cdot (\vec{k}_c - \vec{k}_v - \vec{k}_0)} \hat{r} d^3\vec{r}$$

The optical transition is **allowed** if $\vec{k}_c - \vec{k}_v = \vec{k}_0$.

4. Describe qualitatively the main steps of the procedure that is used to write $|W_{cv}|$ in a factorized form:

$$|W_{cv}| \propto f(\vec{k}_v, \vec{k}_c) \times g(u_{v,\vec{k}_v}, u_{c,\vec{k}_c})$$

where $f(\vec{k}_v, \vec{k}_c)$ and $g(u_{v,\vec{k}_v}, u_{c,\vec{k}_c})$ will be made explicit. What properties of the wavefunction enable one to obtain this result? What approximations are made?

$$|W_{cv}| = -qE_0 \langle c, k_c | \hat{r} e^{-i\vec{k}_0 \cdot \vec{r}}$$

$$\langle c, k_c | \hat{r} e^{-i\vec{k}_0 \cdot \vec{r}} | v, k_v \rangle = \frac{1}{V} \int_{\text{crystal}} u_{c,k_c}^*(\vec{r}) u_{v,k_v}(\vec{r}) e^{i\vec{r} \cdot (\vec{k}_c - \vec{k}_v - \vec{k}_0)} \hat{r} d^3\vec{r}$$

The crystal being periodic, we can express \vec{r} as:

$$\vec{r} = \vec{r}_j + \vec{R}$$

where \vec{r}_j denotes the location of the j^{th} unit cell and \vec{R} the position within this unit cell. The exponential functions are slowly varying in space as compared to u_c and u_v . The integral over the whole crystal is broken up into a sum of integrals over all the primitive cells

$$|W_{cv}| = -q \frac{E_0}{V} \sum_j \int_{\text{unit cell}} u_{c,k_c}^*(\vec{r}_j + \vec{R}) u_{v,k_v}(\vec{r}_j + \vec{R}) e^{i(\vec{r}_j + \vec{R}) \cdot (\vec{k}_c - \vec{k}_v - \vec{k}_0)} (\vec{r}_j + \vec{R}) d^3\vec{R}$$

The exponential functions are replaced by their average values in each primitive cell and pulled out of the integral.

$$|W_{cv}| = -q \frac{E_0}{V} \sum_j e^{i(\vec{k}_c - \vec{k}_v - \vec{k}_0) \cdot \vec{r}_j} \int_{\text{unit cell}} u_{c,k_c}^*(\vec{r}_j + \vec{R}) u_{v,k_v}(\vec{r}_j + \vec{R}) d^3\vec{R}$$

We use the periodicity of u_v and u_c ,

$$|W_{cv}| = -q \frac{E_0}{V} \sum_j e^{i(\vec{k}_c - \vec{k}_v - \vec{k}_0) \cdot \vec{r}_j} \int_{\text{unit cell}} u_{c,k_c}^*(\vec{r}_j) u_{v,k_v}(\vec{r}_j) (\vec{r}_j + \vec{R}) d^3\vec{R}$$

because of the orthogonality of the Bloch waves for different \vec{k} , we arrive at

$$|W_{cv}| = -q \frac{E_0}{V} \delta_{\vec{k}_c - \vec{k}_v, \vec{k}_0} \int_{\text{unit cell}} u_{c,k_c}^*(\vec{R}) u_{v,k_v}(\vec{R}) \vec{R} d^3\vec{R}$$

5. We assume that the wavelength of the incident light is in the visible spectral range. Discuss the possibility of a transition between the valence band and the conduction band as a function of the initial and final electronic wave-vectors. What are the practical consequences when comparing optical properties of indirect and direct bandgap semiconductors?

If our incident light is in the visible spectral range, at the scale of the first Brillouin zone, the transitions are vertical in the reciprocal space: $\vec{k}_c \approx \vec{k}_v$ and thus,

$$|W_{cv}| \approx -q \frac{E_0}{V} \int_{\text{unit cell}} u_{c,0}^*(\vec{R}) u_{v,0}(\vec{R}) \vec{R} d^3 \vec{R}$$

6. We are now interested in a transition occurring at $\vec{k} \approx 0$. In this case, $u_{v,\vec{k}_v}(\vec{r})$ and $u_{c,\vec{k}_c}(\vec{r})$ are eigenstates of the total angular momentum operator: $u_{v,\vec{k}_v=0}(\vec{r}) = |j; j_z\rangle$ and $u_{c,\vec{k}_c=0}(\vec{r}) = |s; s_z\rangle$. The incident light is circularly polarized in the plane perpendicular to the direction of quantization. The initial state is a valence state characterized by $|j; j_z\rangle$, the final state is a conduction state characterized by $|s; s_z\rangle$. What are the possible values of s_z as a function of j_z and of the light helicity ?

At the Γ point, $u_{c,k}$ and $u_{v,k}$ are eigenstate of the angular momentum operator

$$u_{c,0} = |s, s_z\rangle, \quad u_{v,0} = |j, j_z\rangle$$

When a circularly polarized photon is absorbed, this angular momentum is distributed between the photo-excited electron and hole according to the selection rules determined by the band structure of the semiconductor.

7. The valence band presents a p -like ($l = 1$) symmetry while the conduction band is s -like ($l = 0$). This results in the existence of 6 valence states and 2 conduction states. Draw a sketch of the different states at $\vec{k} = 0$ with the corresponding values of $|j; j_z\rangle$ and $|s; s_z\rangle$. Indicate the allowed transitions in the case of a circularly polarized light.

