Chapter III

Light-matter interaction processes

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Outline

1) Light-matter interaction Hamiltonian
2) Emission and absorption
3) Light scattering

1) Light-matter interaction Hamiltonian

For one particle: $$\hat{H} = \frac{1}{2m}\left[\hat{\vec{P}} - q\vec{A}(\hat{\vec{R}})\right]^2 + qU(\hat{\vec{R}})$$

$$\hat{H} = \frac{\hat{\vec{P}}^2}{2m} - \frac{q}{2m}\underbrace{\left[\hat{\vec{P}}.\vec{A}(\hat{\vec{R}}) + \vec{A}(\hat{\vec{R}}).\hat{\vec{P}}\right]}_{[\hat{\vec{R}},\hat{\vec{P}}]\neq 0} + qU(\hat{\vec{R}}) + \frac{q^2\vec{A}^2(\hat{\vec{R}})}{2m}$$

Many-particle:

$$\begin{aligned}\hat{H} &= \sum_i \left\{\frac{\hat{\vec{P}}_i^2}{2m} - \frac{q}{2m}\left[\hat{\vec{P}}_i.\vec{A}(\hat{\vec{R}}_i) + \vec{A}(\hat{\vec{R}}_i).\hat{\vec{P}}_i\right] + qU(\hat{\vec{R}}_i) + \frac{q^2\vec{A}^2(\hat{\vec{R}}_i)}{2m}\right\} \\ &= \hat{H}_0 + \hat{H}_{int} \,.\end{aligned}$$

(same masse *m* and same charge *q*)

1) Light-matter interaction Hamiltonian

Paramagnetic courant operator:

$$\boxed{\hat{\vec{J}}(\vec{r}) \equiv \frac{1}{2m}\sum_i \left\{\hat{\vec{P}}_i \delta(\vec{r}-\hat{\vec{R}}_i) + \delta(\vec{r}-\hat{\vec{R}}_i)\hat{\vec{P}}_i\right\}}$$

Density operator: $\hat{n}(\vec{r}) \equiv \sum_i \delta(\vec{r}-\hat{\vec{R}}_i)$

$$\hat{H}_{int} = \int d\vec{r}\, \left\{-q\hat{\vec{J}}(\vec{r}).\vec{A}(\vec{r}) + \frac{q^2}{2m}\hat{n}(\vec{r})\vec{A}^2(\vec{r}) + q\hat{n}(\vec{r})U(\vec{r})\right\}$$

$$\hat{H}_{int} = \hat{H}_1 + \hat{H}_2 + \hat{H}_3$$

$$\boxed{\begin{aligned} \hat{H}_1 &= -q\int d\vec{r}\, \hat{\vec{J}}(\vec{r}).\vec{A}(\vec{r}) \ , \\ \hat{H}_2 &= \frac{q^2}{2m}\int d\vec{r}\, \hat{n}(\vec{r})\vec{A}^2(\vec{r}) \ , \\ \hat{H}_3 &= q\int d\vec{r}\, \hat{n}(\vec{r})U(\vec{r}) \ . \end{aligned}}$$

two choices

$\vec{A}(\vec{r})$: Classical description of the field

$\hat{\vec{A}}(\vec{r})$: Quantum description of the field

1) Light-matter interaction Hamiltonian

Using $\hat{\vec{A}}(\vec{r}) = \frac{1}{\sqrt{V}} \sum_{i,\vec{k}} \sqrt{\frac{\hbar}{2\omega_k \epsilon_0}} \left\{ \hat{a}_{i,\vec{k}}\, \hat{\epsilon}_{i,\vec{k}}\, e^{i\vec{k}.\vec{r}} + \hat{a}^{+}_{i,\vec{k}}\, \hat{\epsilon}^{*}_{i,\vec{k}} e^{-i\vec{k}.\vec{r}} \right\}$

$$\Rightarrow \quad \boxed{\hat{H}_1 = \frac{e}{\sqrt{V}} \sum_{i,\vec{k}} \sqrt{\frac{\hbar}{2\omega_k \epsilon_0}} \left\{ \hat{a}_{i,\vec{k}}\, \hat{\epsilon}_{i,\vec{k}} . \hat{\vec{J}}_{-\vec{k}} + \hat{a}^{+}_{i,\vec{k}}\, \hat{\epsilon}^{*}_{i,\vec{k}} . \hat{\vec{J}}_{\vec{k}} \right\}}$$

with $\hat{\vec{J}}_{\vec{k}} = \int d\vec{r}\; \hat{\vec{J}}(\vec{r})\; e^{-i\vec{k}.\vec{r}}$

2) Emission and absorption

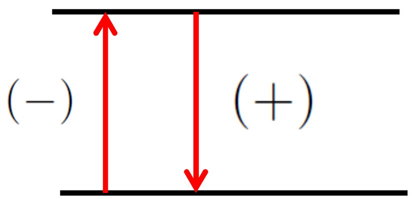
2.1) Emission and absorption rates

Fermi Golden Rule

(FGR)

First-order perturbation theory

$$\Gamma = \frac{2\pi}{\hbar} \left| \langle m; n_{i,\vec{k}} \pm 1 | \hat{H}_1 | n; n_{i,\vec{k}} \rangle \right|^2 \rho_f^{\pm}$$



$$E_m \quad |m; n_{i,\vec{k}} \pm 1\rangle$$

$$E_n \quad |n; n_{i,\vec{k}}\rangle$$

E_n Energies of the atomic, molecular, band…states

ρ_f Density of final states $\qquad \omega = c|\vec{k}| \equiv \omega_k$

$(-)$ absorption $\qquad \rho_f^- = \delta(E_m - E_n - \hbar\omega)$

$(+)$ emission $\qquad \rho_f^+ = \int \frac{d\vec{r}d\vec{p}}{(2\pi\hbar)^3}\delta(E_m - E_n + \hbar\omega)$

2) Emission and absorption

Note that the states of radiation can also contain **any number of photons** in other modes, but here we focus on a process in which **a single photon** is emitted or absorbed, so that these other photons are **passive spectators** who have no influence on the amplitude of the process.

The hamiltonian $\hat{H}_1$ is a sum over all the *em* modes but only one term of this sum contributes to the amplitude: the one associated to the mode $n_{i,\vec{k}}$

$\hat{H}_1$ is a product of an operator acting on the Hilbert space of the field and an operator acting on the Hilbert space of the material system

$$\hat{H}_1 = \frac{e}{\sqrt{V}} \sum_{i,\vec{k}} \sqrt{\frac{\hbar}{2\omega_k \epsilon_0}} \left\{ \hat{a}_{i,\vec{k}}\, \hat{\epsilon}_{i,\vec{k}} . \hat{\vec{J}}_{-\vec{k}} + \hat{a}^{+}_{i,\vec{k}}\, \hat{\epsilon}^{*}_{i,\vec{k}} . \hat{\vec{J}}_{\vec{k}} \right\}$$

field matter

2) Emission and absorption

$$\Gamma = \frac{2\pi}{\hbar} \left| \langle m; n_{i,\vec{k}} \pm 1 | \widehat{H}_1 | n; n_{i,\vec{k}} \rangle \right|^2 \rho_f^{\pm}$$

$$\begin{aligned} \langle n_{i,\vec{k}} + 1 | \hat{a}^+_{i,\vec{k}} | n_{i,\vec{k}} \rangle &= \sqrt{n_{i,\vec{k}} + 1} \\ \langle n_{i,\vec{k}} - 1 | \hat{a}_{i,\vec{k}} | n_{i,\vec{k}} \rangle &= \sqrt{n_{i,\vec{k}}}\,. \end{aligned}$$

electromagnetic part

absorption

$$\boxed{\Gamma_{abs.} = \frac{\pi e^2}{V \omega \epsilon_0} n_{i,\vec{k}} \left| \langle m | \widehat{\vec{J}}_{-\vec{k}} . \hat{\epsilon}_{i,\vec{k}} | n \rangle \right|^2 \rho_f^-}$$

$E_m \quad |m; n_{i,\vec{k}} - 1\rangle$

$E_n \quad |n; n_{i,\vec{k}}\rangle$

2) Emission and absorption

emission

$$E_m \quad |m; n_{i,\vec{k}} - 1\rangle$$

$$E_n \quad |n; n_{i,\vec{k}}\rangle$$

$$\boxed{\Gamma_{em.} = \frac{\pi e^2}{V\omega\epsilon_0}(n_{i,\vec{k}}+1)\left|\langle m|\hat{\vec{J}}_{\vec{k}}.\hat{\epsilon}^*_{i,\vec{k}}|n\rangle\right|^2 \rho_f^+}$$

Most notable in these formulas is the presence of factors *n* and *n+1* derived from what the photon is a boson and are responsible for the **emission** and **absorption** of radiation **induced.**

The more photons in the initial state, the greater the process of emission or absorption is likely.

This is the principle of the **laser** effect.

2) Emission and absorption

$$\boxed{\Gamma_{em.} = \frac{\pi e^2}{V\omega\epsilon_0}(n_{i,\vec{k}}+1)\left|\langle m|\hat{\vec{J}}_{\vec{k}}.\hat{\epsilon}^*_{i,\vec{k}}|n\rangle\right|^2 \rho_f^+}$$

It is also remarkable that the emission of a photon can take place even in the absence of initial photon (*n = 0*), it means in the apparent absence of external stimulation.

Spontaneous emission

$$\Gamma_{em.} = \frac{\pi e^2}{V\omega\epsilon_0}\left|\langle m|\hat{\vec{J}}_{\vec{k}}.\hat{\epsilon}^*_{i,\vec{k}}|n\rangle\right|^2 \rho_f^+$$

It corresponds to the classical radiation of an accelerated charge.