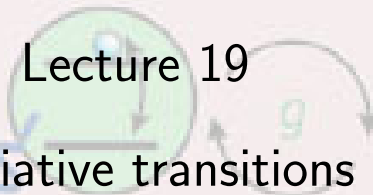


Lecture 19

Radiative transitions



Radiative transitions: background

- Previously, we have formulated a quantum theory of atoms (matter) coupling to a classical time-independent electromagnetic field, cf. Zeeman and Stark effects.
- To develop a fully quantum theory of light-matter systems, we have to address both the quantum theory of the electromagnetic field and formulate a theory of the coupling of light to matter.
- In the following we will address each of these components in turn, starting with light-matter coupling.
- Our motivation for developing such a consistent theory is that it:
 - (a) provides a platform to study radiative transitions in atoms (which will address)
 - (b) forms the basis of **quantum optics** (which will not address – but which is well-represented in subsequent courses).

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Radiative transitions: outline

- Coupling of matter to electromagnetic field
- Spontaneous emission, absorption and stimulated emission
- Einstein's A and B coefficients
- Selection rules
- Theory of the laser and coherent states

Coupling of matter to the electromagnetic field

- For a single-electron atom in a time-dependent external EM field, the Hamiltonian takes the form,

$$\hat{H}_{\text{atom}} = \frac{1}{2m} (\hat{\mathbf{p}} + e\mathbf{A}(\mathbf{r}, t))^2 - e\phi(\mathbf{r}, t) + V(\mathbf{r})$$

(with a straightforward generalization to multi-electron atoms).

- Previously, we have seen that it is profitable to expand Hamiltonian in \hat{A} , $\hat{H}_{\text{atom}} = \hat{H}_0 + \hat{H}_{\text{para}} + \hat{H}_{\text{dia.}}$, where $\hat{H}_0 = \frac{\hat{\mathbf{p}}^2}{2m} + V(\mathbf{r}) - e\phi(t)$

$$\hat{H}_{\text{para}}(t) = \frac{e}{m} \mathbf{A}(t) \cdot \hat{\mathbf{p}}$$

the paramagnetic term describes coupling of the atom to the EM field, and $\hat{H}_{\text{dia}} = (e\mathbf{A})^2/2m$ represents diamagnetic term.

- Since we will be interested in absorption and emission of single photons, influence of diamagnetic term is (as usual) negligible.

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Coupling of matter to the electromagnetic field

- When quantized, EM field is described by the photon Hamiltonian,

$$\hat{H}_{\text{rad}} = \sum_{\mathbf{k}, \lambda=1,2} \hbar \omega_{\mathbf{k}} \left(a_{\mathbf{k}\lambda}^\dagger a_{\mathbf{k}\lambda} + \frac{1}{2} \right), \quad \omega_{\mathbf{k}} = c|\mathbf{k}|$$

where $a_{\mathbf{k}\lambda}^\dagger/a_{\mathbf{k}\lambda}$ create/annihilate photons with polarization λ .

- These operators obey (bosonic) commutation relations,

$$[a_{\mathbf{k}\lambda}, a_{\mathbf{k}'\lambda'}^\dagger] = \delta_{\mathbf{k},\mathbf{k}'} \delta_{\lambda,\lambda'}, \quad [a_{\mathbf{k}\lambda}, a_{\mathbf{k}'\lambda'}] = [a_{\mathbf{k}\lambda}^\dagger, a_{\mathbf{k}'\lambda'}^\dagger] = 0$$

and act on photon number states, $|n_{\mathbf{k}\lambda}\rangle = \frac{1}{\sqrt{n_{\mathbf{k}\lambda}!}} (a_{\mathbf{k}\lambda}^\dagger)^{n_{\mathbf{k}\lambda}} |\Omega\rangle$, as

$$a_{\mathbf{k}\lambda} |n_{\mathbf{k}\lambda}\rangle = \sqrt{n_{\mathbf{k}\lambda}} |n_{\mathbf{k}\lambda} - 1\rangle, \quad a_{\mathbf{k}\lambda}^\dagger |n_{\mathbf{k}\lambda}\rangle = \sqrt{n_{\mathbf{k}\lambda} + 1} |n_{\mathbf{k}\lambda} + 1\rangle$$

- With these definitions, the vector potential is given by

$$\hat{\mathbf{A}}(\mathbf{r}) = \sum_{\mathbf{k}\lambda=1,2} \sqrt{\frac{\hbar}{2\epsilon_0 \omega_{\mathbf{k}} V}} \left[\hat{\mathbf{e}}_{\mathbf{k}\lambda} a_{\mathbf{k}\lambda} e^{i\mathbf{k}\cdot\mathbf{r}} + \hat{\mathbf{e}}_{\mathbf{k}\lambda}^* a_{\mathbf{k}\lambda}^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}} \right]$$

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- To determine radiative transition rates, we will exploit **Fermi's Golden rule**. To prepare for this, it is convenient to transfer time-dependence to operators (Heisenberg representation).
- As with any operator, the field operators obey equations of motion,

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Coupling of matter to the electromagnetic field

- Putting together all of these components, the total Hamiltonian is then given by $\hat{H} = \hat{H}_{\text{atom}} + \hat{H}_{\text{para}} + \hat{H}_{\text{rad}}$ where (with $\phi = 0$)

$$\hat{H}_{\text{atom}} = \frac{\hat{\mathbf{p}}^2}{2m} + V(\mathbf{r}), \quad \hat{H}_{\text{rad}} = \sum_{\mathbf{k}\lambda} \hbar\omega_{\mathbf{k}} \left(a_{\mathbf{k}\lambda}^\dagger a_{\mathbf{k}\lambda} + \frac{1}{2} \right) \quad \text{and}$$

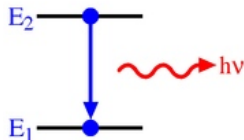
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- In the following, we will apply this Hamiltonian to the problem of radiative transitions in single electron atoms.

Spontaneous emission

- Consider probability for an atom, initially in state $|i\rangle$ to make transition to $|f\rangle$ with emission of a photon of wavevector \mathbf{k} and polarization λ – spontaneous emission.



- If radiation field initially prepared in vacuum state, $|\Omega\rangle$, then final state involves one photon, $a_{\mathbf{k}\lambda}^\dagger|\Omega\rangle$.
- Therefore, making use of Fermi's Golden rule, with the perturbation

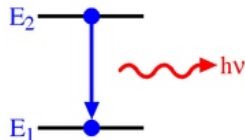
$$\hat{H}_{\text{para}}(t) = \frac{e}{m} \hat{\mathbf{A}}(\mathbf{r}, t) \cdot \hat{\mathbf{p}}$$

transition probability given by,

$$\Gamma_{i \rightarrow f}(t) = \frac{2\pi}{\hbar^2} |\langle f| \otimes \langle \mathbf{k}\lambda | \hat{H}_{\text{para}} | i \rangle \otimes |\Omega\rangle|^2 \delta(\omega_{if} - \omega)$$

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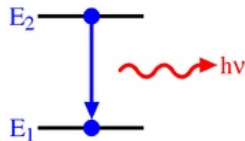
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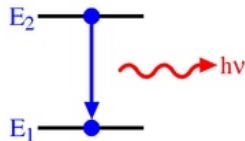
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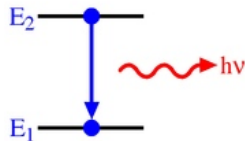
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- To determine transition rate, we must analyse matrix elements of the form $\langle f | e^{-i\mathbf{k} \cdot \mathbf{r}} \hat{\mathbf{e}}_{k\lambda}^* \cdot \hat{\mathbf{p}} | i \rangle$. For typical state, $\langle \hat{\mathbf{e}}_{k\lambda}^* \cdot \hat{\mathbf{p}} \rangle \sim p \sim Zm c \alpha$.
- But what about exponential factor? With $r \sim \hbar/p \simeq \hbar/mZc\alpha$, and $\omega_k = c|\mathbf{k}| \sim \frac{p^2}{2m}$ (for electronic transitions), we have

$$\mathbf{k} \cdot \mathbf{r} \simeq \frac{\omega_k}{c} \frac{\hbar}{p} \simeq \frac{\hbar p}{mc} \simeq Z\alpha$$

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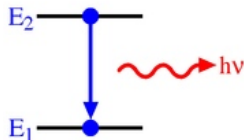
i.e. for $Z\alpha \ll 1$, we can expand exponential as power series in $\mathbf{k} \cdot \mathbf{r}$ with lowest terms dominant.

- Taking zeroth order term, and using $\hat{\mathbf{p}} = \frac{im}{\hbar} [\hat{H}_0, \mathbf{r}]$ (cf. Ehrenfest)

$$\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda}^* \cdot \hat{\mathbf{p}} | i \rangle = -im\omega_{\mathbf{k}} \langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda}^* \cdot \mathbf{r} | i \rangle$$

Spontaneous emission: electric dipole approximation

$$\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda}^* \cdot \hat{\mathbf{p}} | i \rangle = -im\omega_{\mathbf{k}} \langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda}^* \cdot \mathbf{r} | i \rangle$$



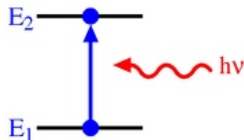
- This result, which emerges from leading approximation in $Z\alpha$, is known as **electric dipole approximation**: Effectively, we have set

$$\hat{H}_{\text{para}} = \frac{e}{m} \hat{\mathbf{A}}(\mathbf{r}, t) \cdot \hat{\mathbf{p}} \simeq e \hat{\mathbf{E}}(\mathbf{r}, t) \cdot \mathbf{r} = -\hat{\mathbf{E}}(\mathbf{r}, t) \cdot \hat{\mathbf{d}}$$

translating to the potential energy of a dipole, with moment $\hat{\mathbf{d}} = -e\mathbf{r}$, in an oscillating electric field.

Stimulated absorption and emission

- Consider now absorption of a photon. If we assume that, in the initial state, there are $n_{\mathbf{k}\lambda}$ photons in mode $(\mathbf{k}\lambda)$ then, after the transition, there will be $n_{\mathbf{k}\lambda} - 1$ photons.



- Then, if initial state of the atom is $|i\rangle$ and final state is $|f\rangle$,

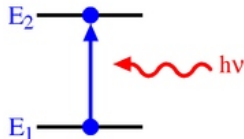
$$\begin{aligned} &\langle f| \otimes \langle (n_{\mathbf{k},\lambda} - 1)| \hat{H}_{\text{para}} |i\rangle \otimes |n_{\mathbf{k}\lambda}\rangle \\ &= \langle f| \otimes \langle (n_{\mathbf{k},\lambda} - 1)| \frac{e}{m} \sqrt{\frac{\hbar}{2\epsilon_0\omega_{\mathbf{k}}V}} \hat{\mathbf{e}}_{\mathbf{k}\lambda} a_{\mathbf{k}\lambda} e^{i\mathbf{k}\cdot\mathbf{r}} \cdot \hat{\mathbf{p}} |i\rangle \otimes |n_{\mathbf{k}\lambda}\rangle \end{aligned}$$

- Then, using the relation $a_{\mathbf{k}\lambda} |n_{\mathbf{k}\lambda}\rangle = \sqrt{n_{\mathbf{k}\lambda}} |(n_{\mathbf{k}\lambda} - 1)\rangle$,

$$\langle f| \otimes \langle (n_{\mathbf{k},\lambda} - 1)| \hat{H}_{\text{para}} |i\rangle \otimes |n_{\mathbf{k}\lambda}\rangle = \langle f| \frac{e}{m} \sqrt{\frac{\hbar n_{\mathbf{k}\lambda}}{2\epsilon_0\omega_{\mathbf{k}}V}} e^{i\mathbf{k}\cdot\mathbf{r}} \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{p}} |i\rangle$$

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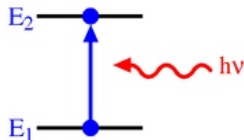
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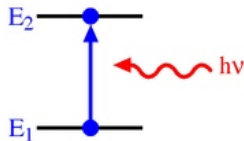
- Consider now absorption of a photon. If we assume that, in the initial state, there are $n_{\mathbf{k}\lambda}$ photons in mode $(\mathbf{k}\lambda)$ then, after the transition, there will be $n_{\mathbf{k}\lambda} - 1$ photons.
- As a result, using Fermi's Golden rule,

$$\Gamma_{i \rightarrow f}(t) = \frac{2\pi}{\hbar^2} |\langle f | \otimes \langle (n_{\mathbf{k}\lambda} - 1) | \hat{H}_{\text{para}} | i \rangle \otimes | n_{\mathbf{k}\lambda} \rangle|^2 \delta(\omega_{fi} - \omega)$$

we obtain the transition amplitude,

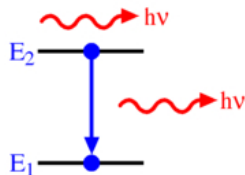
$$\Gamma_{i \rightarrow f, \mathbf{k}\lambda} = \frac{2\pi}{\hbar} \left| \langle f | \frac{e}{m} \sqrt{\frac{\hbar n_{\mathbf{k}\lambda}}{2\epsilon_0 \omega_{\mathbf{k}} V}} e^{i\mathbf{k} \cdot \mathbf{r}} \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{p}} | i \rangle \right|^2 \delta(E_f - E_i - \hbar\omega_{\mathbf{k}})$$

- In particular, we find that the absorption rate **increases** linearly with photon number, $n_{\mathbf{k}\lambda}$.



Stimulated absorption and emission

- Similarly, if we consider emission process in which there are already $n_{\mathbf{k}\lambda}$ photons in initial state,

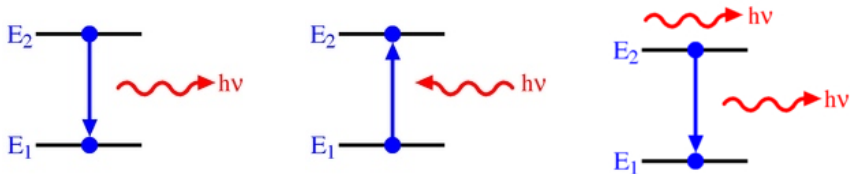


- using the relation $a_{\mathbf{k}\lambda}^\dagger |n_{\mathbf{k}\lambda}\rangle = \sqrt{n_{\mathbf{k}\lambda} + 1} |(n_{\mathbf{k}\lambda} + 1)\rangle$, we have revised transition rate,

$$\Gamma_{i \rightarrow f, \mathbf{k}\lambda} = \frac{2\pi}{\hbar} \left| \langle f | \frac{e}{m} \sqrt{\frac{\hbar(n_{\mathbf{k}\lambda} + 1)}{2\epsilon_0\omega_{\mathbf{k}}V}} e^{-i\mathbf{k}\cdot\mathbf{r}} \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{p}} | i \rangle \right|^2 \delta(E_f - E_i - \hbar\omega_{\mathbf{k}})$$

- Enhancement of transition rate by photon occupancy known as **stimulated emission**.

Radiative transitions: summary



- Altogether, in dipole approximation $\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{p}} | i \rangle \simeq -im\omega_{\mathbf{k}} \langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \mathbf{r} | i \rangle$

$$\Gamma_{i \rightarrow f, \mathbf{k}\lambda} \simeq \frac{\pi\omega_{\mathbf{k}}}{\epsilon_0 V} |\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{d}} | i \rangle|^2 \begin{cases} n_{\mathbf{k}\lambda} \delta(E_f - E_i - \hbar\omega_{\mathbf{k}}) & \text{absorption} \\ (n_{\mathbf{k}\lambda} + 1) \delta(E_i - E_f - \hbar\omega_{\mathbf{k}}) & \text{emission} \end{cases}$$

where $\hat{\mathbf{d}} = -e\mathbf{r}$ is electric dipole operator.

- If there are no photons present initially, $\Gamma_{i \rightarrow f, \mathbf{k}\lambda}$ reduces to result for spontaneous emission.
- The coincidence of $n_{\mathbf{k}\lambda}$ -independent coefficients for absorption and emission coincide is known as **detailed balance**.

Absorption and stimulated emission

$$\Gamma_{i \rightarrow f, \mathbf{k}\lambda} \simeq \frac{\pi \omega_{\mathbf{k}}}{\epsilon_0 V} |\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{d}} | i \rangle|^2 \begin{cases} n_{\mathbf{k}\lambda} \delta(E_f - E_i - \hbar\omega_{\mathbf{k}}) & \text{absorption} \\ (n_{\mathbf{k}\lambda} + 1) \delta(E_i - E_f - \hbar\omega_{\mathbf{k}}) & \text{emission} \end{cases}$$

- Integrated transition rate associated with a small solid angle $d\Omega$ in the direction \mathbf{k} given by

$$dR_{i \rightarrow f, \lambda} = \sum_{\mathbf{k} \in d\Omega} \Gamma_{i \rightarrow f, \mathbf{k}\lambda} = d\Omega V \int \frac{k^2 dk}{(2\pi)^3} \Gamma_{i \rightarrow f, \mathbf{k}\lambda}$$

- If we assume that the photon number, $n_{\mathbf{k}\lambda}$ is isotropic, independent of angle Ω , using the dispersion relation $\omega_{\mathbf{k}} = ck$, we obtain

$$\frac{dR_{i \rightarrow f, \lambda}}{d\Omega} = \frac{V}{c^3} \int \frac{\omega^2 d\omega}{(2\pi)^3} \frac{\pi\omega}{\epsilon_0 V} |\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{d}} | i \rangle|^2 \begin{cases} n_{\lambda}(\omega) \delta(E_f - E_i - \hbar\omega) \\ (n_{\lambda}(\omega) + 1) \delta(E_i - E_f - \hbar\omega) \end{cases}$$

where $\hbar\omega = |E_f - E_i|$.

- From this expression, we can obtain the power loss as $P_{\lambda} = \hbar\omega R_{\lambda}$.

Absorption and stimulated emission

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$$\frac{dR_{i \rightarrow f, \lambda}}{d\Omega} = \frac{\omega^3}{8\pi^2 \epsilon_0 \hbar c^3} |\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \mathbf{d} | i \rangle|^2 \begin{cases} n_{\lambda}(\omega) \\ n_{\lambda}(\omega) + 1 \end{cases}$$

where $\hbar\omega = |E_f - E_i|$.

- From this expression, we can obtain the power loss as $P_{\lambda} = \hbar\omega R_{\lambda}$.

Einstein's A and B coefficients

In fact, frequency dependence of spontaneous emission rate can be inferred using ingenious argument due to Einstein who showed that stimulated and spontaneous transitions must be related.

- Consider ensemble of atoms exposed to black-body radiation at temperature T . Let us consider transitions between states $|\psi_j\rangle$ and $|\psi_k\rangle$, with $E_k - E_j = \hbar\omega$.
- If number of atoms in two states given by n_j and n_k , transition rates per atom given by:

absorption	$j \rightarrow k$	$B_{j \rightarrow k} u(\omega)$
stimulated emission	$k \rightarrow j$	$B_{k \rightarrow j} u(\omega)$
spontaneous emission	$k \rightarrow j$	$A_{k \rightarrow j}(\omega)$

where $u(\omega)$ denotes energy density of radiation.

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- In thermodynamic equilibrium the rates must balance, so that

$$n_k [A_{k \rightarrow j}(\omega) + B_{k \rightarrow j} u(\omega)] = n_j B_{j \rightarrow k} u(\omega)$$

- At the same time, relative populations of two states given by Boltzmann factor,

$$\frac{n_j}{n_k} = \frac{e^{-E_j/k_B T}}{e^{-E_k/k_B T}} = e^{\hbar\omega/k_B T}$$

Thus we have:

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$$u(\omega) = \frac{\hbar\omega^3}{\pi^2 c^3} \bar{n}(\omega) = \frac{\hbar\omega^3}{\pi^2 c^3} \frac{1}{e^{\hbar\omega/k_B T} - 1}$$

- Since $A_{k \rightarrow j}$ is intrinsic (independent of temperature), T must cancel on right hand side, i.e.

$$B_{k \rightarrow j} = B_{j \rightarrow k} \quad \text{and} \quad A_{k \rightarrow j}(\omega) = B_{k \rightarrow j} \frac{\hbar\omega^3}{\pi^2 c^3}$$

- So, A and B coefficients are related, and if we can calculate B coefficient for stimulated emission from Fermi's Golden rule, we can infer A , and *vice versa*.

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Selection rules:

$$\Gamma_{i \rightarrow f, \mathbf{k}\lambda} \simeq \frac{\pi\omega_{\mathbf{k}}}{\epsilon_0 V} |\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{d}} | i \rangle|^2 \begin{cases} n_{\mathbf{k}\lambda} \delta(E_f - E_i - \hbar\omega_{\mathbf{k}}) & \text{absorption} \\ (n_{\mathbf{k}\lambda} + 1) \delta(E_i - E_f - \hbar\omega_{\mathbf{k}}) & \text{emission} \end{cases}$$

- Formulae for rates $\Gamma_{i \rightarrow f, \mathbf{k}\lambda}$ show that radiative transitions will not occur between states $|i\rangle$ and $|f\rangle$ unless at least one component of the dipole matrix element $\langle f | \hat{\mathbf{d}} | i \rangle$ is non-zero.
 - If matrix elements are zero for certain pairs, they are disallowed (at least in the electric dipole approximation) leading to **selection rules**.
 - Since dipole operator $\hat{\mathbf{d}} = -e\mathbf{r}$ changes sign under parity ($\mathbf{r} \rightarrow -\mathbf{r}$), matrix element $\langle f | \hat{\mathbf{d}} | i \rangle$ will vanish if $|f\rangle$ and $|i\rangle$ have same parity.
- ! The parity of the wavefunction must change in an electric dipole transition.

Selection rules: parity

$$\Gamma_{i \rightarrow f, \mathbf{k}\lambda} \simeq \frac{\pi\omega_{\mathbf{k}}}{\epsilon_0 V} |\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{d}} | i \rangle|^2 \begin{cases} n_{\mathbf{k}\lambda} \delta(E_f - E_i - \hbar\omega_{\mathbf{k}}) & \text{absorption} \\ (n_{\mathbf{k}\lambda} + 1) \delta(E_i - E_f - \hbar\omega_{\mathbf{k}}) & \text{emission} \end{cases}$$

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Selection rules: spin

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- Separating wavefunction into spatial and spin components, $|f\rangle = |\phi_f\rangle \otimes |\chi_f\rangle$, since dipole operator acts only on spatial part,

$$\langle f | \hat{\mathbf{d}} | i \rangle = -\langle \chi_f | \chi_i \rangle \int d^3r \phi_f^*(\mathbf{r}) \mathbf{e} r \phi_i(\mathbf{r})$$

i.e. spin term, $\langle \chi_f | \chi_i \rangle$, vanishes unless $|\chi_i\rangle$ and $|\chi_f\rangle$ are identical,

$$\Delta s = 0, \quad \Delta m_s = 0$$

- 2 The spin state is not altered in an electric dipole transition.

Selection rules: orbital angular momentum

$$\Gamma_{i \rightarrow f, \mathbf{k}\lambda} \simeq \frac{\pi\omega_{\mathbf{k}}}{\epsilon_0 V} |\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{d}} | i \rangle|^2 \begin{cases} n_{\mathbf{k}\lambda} \delta(E_f - E_i - \hbar\omega_{\mathbf{k}}) & \text{absorption} \\ (n_{\mathbf{k}\lambda} + 1) \delta(E_i - E_f - \hbar\omega_{\mathbf{k}}) & \text{emission} \end{cases}$$

- From the operator identity, $[\hat{L}_i, r_j] = i\hbar\epsilon_{ijk}r_k$, it follows that

$$[\hat{L}_z, z] = 0, \quad [\hat{L}_z, x \pm iy] = \pm(x \pm iy)\hbar$$

- We therefore obtain,

$$\langle \ell', m' | [\hat{L}_z, z] | \ell, m \rangle = (m' - m)\hbar \langle \ell', m' | z | \ell, m \rangle = 0$$

- Similarly, since $\langle \ell', m' | [\hat{L}_z, x \pm iy] | \ell, m \rangle = \pm\hbar \langle \ell', m' | x \pm iy | \ell, m \rangle$,

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- 3 Therefore, to get non-zero component of dipole matrix element, require $\Delta m_\ell = 0, \pm 1$.

Selection rules: orbital angular momentum

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Selection rules: orbital angular momentum

- Using operator identity $[\hat{\mathbf{L}}^2, [\hat{\mathbf{L}}^2, \mathbf{r}]] = 2\hbar^2(\mathbf{r}\hat{\mathbf{L}}^2 + \hat{\mathbf{L}}^2\mathbf{r})$, we have

$$\begin{aligned}\langle \ell', m' | [\hat{\mathbf{L}}^2, [\hat{\mathbf{L}}^2, \mathbf{r}] | \ell, m \rangle &= [\ell'(\ell' + 1) - \ell(\ell + 1)]^2 \langle \ell', m' | \mathbf{r} | \ell, m \rangle \\ &= 2[\ell'(\ell' + 1) + \ell(\ell + 1)] \langle \ell', m' | \mathbf{r} | \ell, m \rangle\end{aligned}$$

i.e. $(\ell + \ell')(\ell + \ell' + 2)[(\ell' - \ell)^2 - 1] \langle \ell', m' | \mathbf{r} | \ell, m \rangle = 0$. Since $\ell, \ell' \geq 0$, dipole matrix element non-vanishing only if $\ell' = \ell \pm 1$.

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- One may summarize the selection rules for ℓ and m_ℓ is by saying that the photon carries off (or brings in, in an absorption transition) one unit of angular momentum.
- N.B. it is possible, though much less likely in the case of an atom, for EM field to interact with magnetic dipole or electric quadrupole moment with different selection rules.

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Selection rules: polarization

$$\Gamma_{i \rightarrow f, \mathbf{k}, \lambda} = \frac{\pi \omega_{\mathbf{k}}}{\epsilon_0 V} |\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \mathbf{d} | i \rangle|^2 \begin{cases} n_{\mathbf{k}, \lambda} \delta(E_f - E_i - \hbar \omega_{\mathbf{k}}) & \text{absorption} \\ (n_{\mathbf{k}, \lambda} + 1) \delta(E_i - E_f - \hbar \omega_{\mathbf{k}}) & \text{emission} \end{cases}$$

- For transitions with $\Delta m_\ell = 0$, the dipole matrix element $\langle f | \mathbf{d} | i \rangle \sim \hat{\mathbf{e}}_z$ – and there is no component of polarization along z-direction.
- Similarly, for electric dipole transitions with $m' = m \pm 1$,
 $\langle \ell', m' | x \mp iy | \ell, m \rangle = 0 = \langle \ell', m' | z | \ell, m \rangle$, and $\langle f | \mathbf{d} | i \rangle \sim (1, \mp i, 0)$.
 - (a) If the wavevector of photon lies along z, the emitted light is circularly polarized with a polarization which depends on helicity.
 - (b) If the wavevector lies in xy plane, the emitted light is linearly polarized, while in general it is elliptically polarized.

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Selection rules: LS coupling

- In the presence of spin-orbit coupling, stationary states labelled by quantum numbers J, m_J, ℓ, s where $\hat{\mathbf{J}} = \hat{\mathbf{L}} + \hat{\mathbf{S}}$.
- The selection rules in this case can be inferred by looking for the conditions for non-zero matrix elements $\langle J', m_{J'}, \ell', s' | \mathbf{r} | J, m_J, \ell, s \rangle$.
- By expanding states $|J, m_J, \ell, s\rangle$ in basis states $|\ell, m_\ell\rangle \otimes |s, m_s\rangle$, one may uncover the following set of selection rules:

• For dipole transitions to take place, we require that

$$\Delta m_J = 0, \pm 1$$

$$\Delta J = 0, \pm 1 \quad \text{not } 0 \rightarrow 0$$

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Radiative transitions: recap

- When coupled to a quantized electromagnetic field, the total Hamiltonian for atomic system given by $\hat{H} = \hat{H}_{\text{atom}} + \hat{H}_{\text{para}} + \hat{H}_{\text{rad}}$ where

$$\hat{H}_{\text{atom}} = \frac{\hat{\mathbf{p}}^2}{2m} + V(\mathbf{r}), \quad \hat{H}_{\text{rad}} = \sum_{\mathbf{k}\lambda} \hbar\omega_{\mathbf{k}} \left(a_{\mathbf{k}\lambda}^\dagger a_{\mathbf{k}\lambda} + \frac{1}{2} \right)$$

denotes the Hamiltonian of the isolated atomic and radiation field, and

$$\hat{H}_{\text{para}}(t) = \frac{e}{m} \hat{\mathbf{A}}(\mathbf{r}, t) \cdot \hat{\mathbf{p}}$$

denotes the coupling with

$$\hat{\mathbf{A}}(\mathbf{r}, t) = \sum_{\mathbf{k}\lambda} \sqrt{\frac{\hbar}{2\epsilon_0\omega_{\mathbf{k}}V}} \left[\hat{\mathbf{e}}_{\mathbf{k}\lambda} a_{\mathbf{k}\lambda} e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_{\mathbf{k}}t)} + \hat{\mathbf{e}}_{\mathbf{k}\lambda}^* a_{\mathbf{k}\lambda}^\dagger e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega_{\mathbf{k}}t)} \right]$$

Radiative transitions: recap

- The transition rate between an initial and final state of the atom and electromagnetic field can be estimated using Fermi's Golden rule

$$\Gamma_{i \rightarrow f} = \frac{2\pi}{\hbar^2} |\langle f | \hat{H}_{\text{para}} | i \rangle|^2 \delta(\omega_{if} - \omega)$$

where $\hbar\omega_{if} = E_i - E_f$.

- Crucially, since the photon creation/annihilation operators obey the relations, $a_{\mathbf{k}\lambda}^\dagger |n_{\mathbf{k}\lambda}\rangle = \sqrt{n_{\mathbf{k}\lambda} + 1} |(n_{\mathbf{k}\lambda} + 1)\rangle$ and $a_{\mathbf{k}\lambda} |n_{\mathbf{k}\lambda}\rangle = \sqrt{n_{\mathbf{k}\lambda}} |(n_{\mathbf{k}\lambda} - 1)\rangle$ the transition rate depends on the photon number, $n_{\mathbf{k}\lambda}$.
- When $Z\alpha \ll 1$, the effective range of the interaction of the atom with the field is small (i.e. $kr \sim Z\alpha$) and we can effect the dipole approximation,

$$\langle f | e^{-i\mathbf{k}\cdot\mathbf{r}} \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{p}} | i \rangle \simeq \frac{im\omega_{\mathbf{k}}}{e} \langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \mathbf{d} | i \rangle, \quad \mathbf{d} = -e\mathbf{r}$$

Radiative transitions: recap



- In the electric dipole approximation, the transition rate is given by

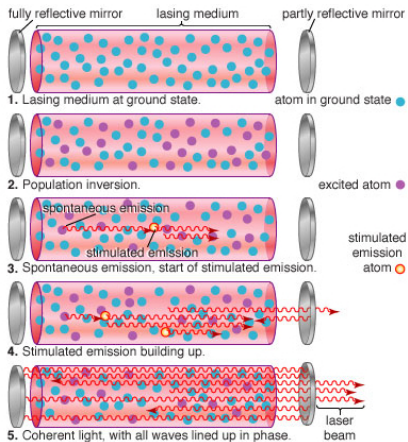
$$\Gamma_{i \rightarrow f, \mathbf{k}\lambda} \simeq \frac{\pi\omega_{\mathbf{k}}}{\epsilon_0 V} |\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{d}} | i \rangle|^2 \begin{cases} n_{\mathbf{k}\lambda} \delta(E_f - E_i - \hbar\omega_{\mathbf{k}}) & \text{absorption} \\ (n_{\mathbf{k}\lambda} + 1) \delta(E_i - E_f - \hbar\omega_{\mathbf{k}}) & \text{emission} \end{cases}$$

where $\hat{\mathbf{d}} = -e\mathbf{r}$ is electric dipole operator.

- The coincidence of $n_{\mathbf{k}\lambda}$ -independent coefficients for absorption and emission coincide is known as **detailed balance**.
- From these results, we turn now to consider the principle of the operation of an atomic laser.

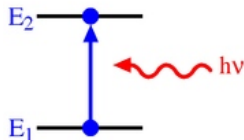
Theory of laser

- Principle of stimulated emission provides basis of laser operation: light amplification by stimulated emission of radiation.
- However, laser not only amplifies light, but provides source of **monochromatic** (single mode), **coherent** (spatial/temporal), **directional** and **intense** radiation.
- In atomic laser, the gain medium provided by a gas of atoms confined to a cavity and bound by highly reflective mirrors.



Theory of laser: rate equations

$$\Gamma_{k\lambda} \sim |\langle f | \hat{\mathbf{e}}_{k\lambda} \cdot \hat{\mathbf{d}} | i \rangle|^2 \begin{cases} n_{k\lambda} & \text{abs.} \\ (n_{k\lambda} + 1) & \text{emiss.} \end{cases}$$



- Consider gas of atoms in a cavity subject to an EM field of intensity $I \propto n(\omega)$ and angular frequency ω tuned to energy difference between two discrete energy levels of the atoms, i.e. $\hbar\omega = E_2 - E_1$.
- Taking into account stimulated absorption, atoms are transferred from level 1 to level 2 at a rate

$$\Gamma_{12} = WN_1 n(\omega)$$

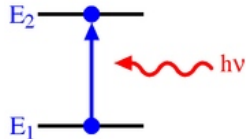
where N_1 atoms in level 1 and W includes matrix elements.

- From spontaneous and stimulated emission processes, the rate of transfer of atoms from level 2 to level 1 is given by

$$\Gamma_{21} = WN_2(n(\omega) + 1)$$

Theory of laser: photon equations

$$\Gamma_{\mathbf{k}\lambda} \sim |\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{d}} | i \rangle|^2 \begin{cases} n_{\mathbf{k}\lambda} & \text{abs.} \\ (n_{\mathbf{k}\lambda} + 1) & \text{emiss.} \end{cases}$$



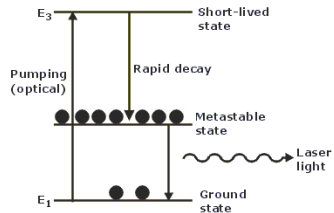
- Since transfer of particles from level 2 to 1 leads to creation of photons in cavity while from 1 to 2 they involve absorption, the rate of change of photon number is given by $\dot{n} = W(N_2(n + 1) - N_1n)$.
- However, to make use of cavity as a photon source, we have to allow photons to leak from the cavity through imperfect mirrors. Taking into account this and other loss processes, we have

$$\dot{n} = DWn + N_2W - \frac{n}{\tau_{\text{ph}}}$$

where $D = N_2 - N_1$ denotes population imbalance and $1/\tau_{\text{ph}}$ is the total loss rate.

Theory of laser: matter equations

$$\Gamma_{\mathbf{k}\lambda} \sim |\langle f | \hat{\mathbf{e}}_{\mathbf{k}\lambda} \cdot \hat{\mathbf{d}} | i \rangle|^2 \begin{cases} n_{\mathbf{k}\lambda} & \text{abs.} \\ (n_{\mathbf{k}\lambda} + 1) & \text{emiss.} \end{cases}$$



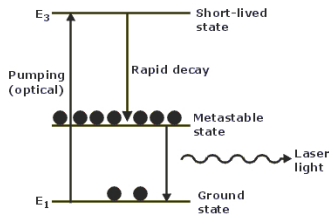
- Without further external processes, photons would escape from cavity and the system would relax into ground state – To create a steady-state photon population, energy must be pumped into the system in the form of excitations.
- Achieved by transferring atoms between 1 and 2 via level 3 by non-resonant optical pump. If lifetime of 3 is short, occupancy is effectively zero, rate of transfer of particles from 2 to 1,

$$\dot{N}_2 \simeq -w_{21} N_2 + w_{12} N_1 - (N_2 - N_1) W n \simeq -\dot{N}_1$$

where we have dropped small contribution from spontaneous emission, and w_{12}, w_{21} denote net non-resonant transition rates.

Theory of laser: stationary equations

$$\dot{N}_2 \simeq -w_{21}N_2 + w_{12}N_1 - (N_2 - N_1)Wn \simeq -\dot{N}_1$$



- Without cavity photons ($n = 0$), since $N_1 + N_2 \simeq N$, in steady state,

$$D^{(0)} = N_2^{(0)} - N_1^{(0)} = N \frac{w_{12} - w_{21}}{w_{12} + w_{21}}$$

denotes **unsaturated inversion**.

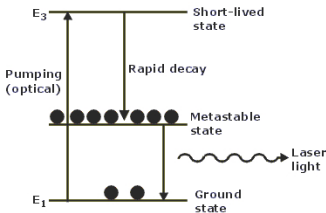
- Restoring the cavity photons, we have

$$\dot{D} = \dot{N}_2 - \dot{N}_1 = \frac{D^{(0)} - D}{T} - 2DWn$$

where $1/T = w_{12} + w_{21}$ represents typical relaxation rate.

Theory of laser: stationary equations

$$\dot{n} = DWn - \frac{n}{\tau_{ph}}, \quad \dot{D} = \frac{D^{(0)} - D}{T} - 2DWn$$



- In steady-state operation, $\dot{n} = \dot{D} = 0$, population imbalance

$$D \equiv N_2 - N_1 = \frac{D^{(0)}}{1 + 2TWn}$$

- From this result, we find the steady state photon number

$$n = \frac{D^{(0)}W - 1/\tau_{ph}}{2TW/\tau_{ph}}$$

- This result shows that the system will only start lasing when the unsaturated inversion exceeds a threshold, $D^{(0)} > 1/\tau_{ph}W$.

Theory of laser: coherence

- Although the analysis above addressed the threshold conditions for the laser, it does not provide any insight into the coherence properties of the radiation field.
- In fact, one may show that the radiation field generated by the laser cavity forms a **coherent or Glauber state**.
- The proof of this statement and the coherence properties that follow would take us on a considerable detour – see Part III quantum optics.
- However, we can gain some insight into the properties and physical manifestations of coherent states by looking at a toy example; but first we must define what we mean by a coherent state.

Coherent states

- A coherent state is defined as an eigenstate of the annihilation operator,

$$a|\beta\rangle = \beta|\beta\rangle$$

Since a is not Hermitian, β can take complex eigenvalues.

- The eigenstates are constructed from the harmonic oscillator ground state the by action of the unitary operator,

$$|\beta\rangle = \hat{U}(\beta)|0\rangle, \quad \hat{U}(\beta) = e^{\beta a^\dagger - \beta^* a}, \quad \hat{U}^\dagger(\beta)\hat{U}(\beta) = \mathbb{I}$$

- The proof follows from the identity (problem set I),

$$a\hat{U}(\beta) = \hat{U}(\beta)(a + \beta), \quad \text{i.e.} \quad a\hat{U}(\beta)|0\rangle = \beta\hat{U}(\beta)|0\rangle$$

i.e. \hat{U} is a translation operator, $\hat{U}^\dagger(\beta)a\hat{U}(\beta) = a + \beta$.

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$$|\beta\rangle = e^{-|\beta|^2/2} e^{\beta a^\dagger} |0\rangle$$

- With $|n\rangle = \frac{1}{\sqrt{n!}} (a^\dagger)^n |0\rangle$, we can write

$$|\beta\rangle = \sum_n e^{-|\beta|^2/2} \frac{\beta^n}{\sqrt{n!}} |n\rangle$$

showing that the probability of observing n excitations

$$P_n = |\langle n|\beta\rangle|^2 = e^{-|\beta|^2} \frac{|\beta|^{2n}}{n!}$$

is a Poisson distribution with average occupation, $\langle \beta|a^\dagger a|\beta\rangle = |\beta|^2$.

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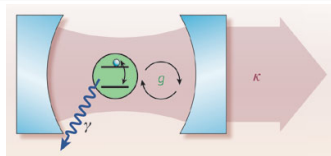
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Driven quantum harmonic oscillator

But how can we prepare a system in a coherent state?



- Consider a single two-level atom resonantly coupled to a single cavity mode – the quantum Hamiltonian of the coupled system,

$$\hat{H} = \frac{1}{2}\hbar\omega\sigma_z + \hbar\omega\left(a^\dagger a + \frac{1}{2}\right) + \hbar g(\sigma_- a + \sigma_+ a^\dagger)$$

- When excitations of two level system are driven by an external pump, it can behave as a classical dipole source for the cavity mode leading to the driven harmonic oscillator Hamiltonian,

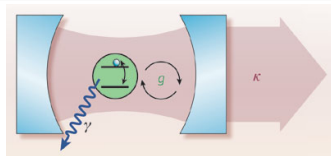
$$\hat{H} \simeq \hat{H}_{\text{rad}} + V(t) = \hbar\omega\left(a^\dagger a + \frac{1}{2}\right) + i\hbar(f(t)a^\dagger - f^*(t)a)$$

where $f(t) = f_0 e^{-i\omega t}$

Driven quantum harmonic oscillator

$$\hat{H} = \hbar\omega \left(a^\dagger a + \frac{1}{2} \right) + i\hbar (f(t)a^\dagger - f^*(t)a)$$

$$f(t) = f_0 e^{-i\omega t}$$



- If photon system is prepared in ground state, $|0\rangle$, the perturbation drives the system into a coherent state.

- To understand how, let us turn to the interaction representation, $i\hbar\partial_t|\psi(t)\rangle_I = V_I|\psi(t)\rangle_I$ where $|\psi(t)\rangle_I = e^{i\hat{H}_0 t/\hbar}|\psi(t)\rangle_S$. With $e^{i\omega t a^\dagger} a e^{-i\omega t a^\dagger} = e^{-i\omega t} a$,

$$V_I(t) = e^{i\hat{H}_0 t/\hbar} i\hbar (f(t)a^\dagger - f^*(t)a) e^{-i\hat{H}_0 t/\hbar} = i\hbar (f_0 a^\dagger - f_0^* a)$$

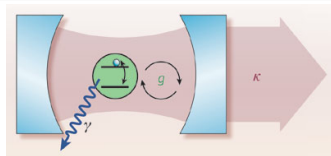
- Since $V_I(t)$ is time-independent, the time-evolution operator, defined by the equation $i\hbar\partial_t U_I(t) = V_I U_I(t)$, is given simply by

$$U_I(t) = \exp [(f_0 a^\dagger - f_0^* a)t]$$

Driven quantum harmonic oscillator

$$\hat{H} = \hbar\omega \left(a^\dagger a + \frac{1}{2} \right) + i\hbar (f(t)a^\dagger - f^*(t)a)$$

$$f(t) = f_0 e^{-i\omega t}$$



- If photon system is prepared in ground state, $|0\rangle$, the perturbation drives the system into a coherent state.
- To understand how, let us turn to the interaction representation, $i\hbar\partial_t|\psi(t)\rangle_I = V_I|\psi(t)\rangle_I$ where $|\psi(t)\rangle_I = e^{i\hat{H}_0 t/\hbar}|\psi(t)\rangle_S$. With $e^{i\omega t a^\dagger} a e^{-i\omega t a^\dagger} = e^{-i\omega t} a$,
$$V_I(t) = e^{i\hat{H}_0 t/\hbar} i\hbar (f(t)a^\dagger - f^*(t)a) e^{-i\hat{H}_0 t/\hbar} = i\hbar (f_0 a^\dagger - f_0^* a)$$
- Since $V_I(t)$ is time-independent, the time-evolution operator, defined by the equation $i\hbar\partial_t U_I(t) = V_I U_I(t)$, is given simply by

$$U_I(t) = \exp [(f_0 a^\dagger - f_0^* a)t]$$

Driven quantum harmonic oscillator

$$U_I(t) = \exp[(f_0 a^\dagger - f_0^* a)t]$$

- Therefore, if the system was prepared in the ground state $|0\rangle$ at $t = 0$, at later times,

$$|\psi(t)\rangle_I = \exp[(f_0 a^\dagger - f_0^* a)t]|0\rangle = e^{-|f_0|^2 t^2/2} e^{f_0 a^\dagger t}|0\rangle$$

- Reexpressed in the Schrödinger representation,

$$|\psi(t)\rangle_S = e^{-i\hat{H}_0 t/\hbar} |\psi(t)\rangle_I = e^{-|f_0|^2 t^2/2} e^{f_0 e^{-i\omega t} a^\dagger t}|0\rangle$$

- A classical oscillatory force drives a system prepared in the vacuum state into a coherent state with an excitation number which climbs as $|f_0|^2 t^2$.

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Synopsis: Lectures 16-19

12 **Field theory: from phonons to photons:**

From particles to fields: classical field theory of harmonic atomic chain; quantization of atomic chain; phonons. Classical theory of the EM field; waveguide; quantization of the EM field and photons.

13 **Time-dependent perturbation theory:**

Rabi oscillations in two level systems; perturbation series; sudden approximation; harmonic perturbations and Fermi's Golden rule.

14 **Radiative transitions:**

Light-matter interaction; spontaneous emission; absorption and stimulated emission; Einstein's A and B coefficients; dipole approximation; selection rules; †lasers.

15 Scattering theory

Elastic scattering; cross section; method of particle waves; Born approximation; scattering of identical particles.

16 Relativistic quantum mechanics:

Klein-Gordon equation; Dirac equation; relativistic covariance and spin; free relativistic particles and the Klein paradox; antiparticles; coupling to EM field: minimal coupling and the connection to non-relativistic quantum mechanics; \dagger field quantization.