

Lecture 4. Interaction between atom and light: semiclassical approach

Semiclassical approach. Dipole approximation. Unperturbed atom. Perturbation theory. Monochromatic field. Two-level system. Rotating-wave approximation. Rabi oscillations.

1. Semiclassical approach.

In order to describe most effects in the interaction between light and an atom, it is sufficient to use semiclassical approach, in which light is described classically and only the atom is described quantum mechanically. Certainly, for some effects, such as the spontaneous emission of the atom, we will need quantum optical description of light. However, as the first step we will use the semiclassical approach since quantum description is usually called 'counter-intuitive'.

Then, the atom will be described by a wave function $\Psi(r,t)$, which will satisfy the Schrödinger equation

$$i\hbar \frac{d\Psi(r,t)}{dt} = H\Psi(r,t), \quad (1)$$

where the Hamiltonian is a sum of the one in the absence of light, H_0 , and the perturbation caused by light, V .

2. Dipole approximation.

Because the atom is much smaller than the wavelength, $a \ll \lambda$ (Fig.1), the electric wave in the vicinity of the atom has the form

$$E(\vec{r},t) = E(t)e^{i\vec{k}\vec{r}} \approx E(t)e^{i\vec{k}\vec{r}_0},$$

i.e., it can be considered rather as an oscillation in time than as a wave. Then, the interaction between light and the atom is a function of time only, and

$$\hat{H} = \hat{H}_0 + \hat{V}(t). \quad (2)$$

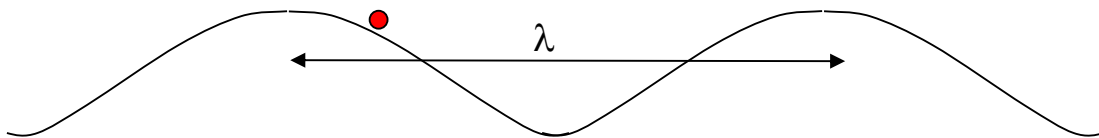


Fig.1

In the dipole approximation, moreover, the interaction is given by the product of the electric field and the dipole moment of the atom:

$$\hat{V}(t) = -\hat{d}\vec{E}(t). \quad (3)$$

3. Unperturbed atom.

In the absence of light, the atom wavefunction will satisfy the Schrödinger equation.

$$i\hbar \frac{d\Psi(r,t)}{dt} = \hat{H}_0\Psi(r,t),$$

And the solutions will be the so-called stationary solutions, consisting of the spatial part and temporal part (with trivial dependence on time):

$$\Phi_n(\vec{r}, t) = \varphi_n(\vec{r})e^{-i\varepsilon_n t/\hbar}, \quad (4)$$

where the energies ε_n satisfy $\hat{H}_0\varphi_n(\vec{r}) = \varepsilon_n\varphi_n(\vec{r})$. The eigenfunctions $\varphi_n(\vec{r})$ determine the shape of the 'electron cloud'. They form an orthonormal set of functions, i.e.,

$$\int d\vec{r}\varphi_n(\vec{r})\varphi_m^*(\vec{r}) = \delta_{mn}.$$

For instance, if the atom can be represented as a harmonic oscillator, these spatial eigenfunctions in one dimension are given by the Hermite functions.

In the absence of light, the atom, as a quantum system, can be in a superposition of these eigenfunctions (4), i.e.,

$$\Psi^{(0)}(\vec{r}, t) = \sum_n c_n \Phi_n(\vec{r}, t) = \sum_n c_n \varphi_n(\vec{r})e^{-i\varepsilon_n t/\hbar}, \quad (5)$$

with the normalization condition, $\int d\vec{r}|\Psi^{(0)}(\vec{r}, t)|^2 = 1$, leading to $\sum_n |c_n|^2 = 1$. The squared coefficients $|c_n|^2 \equiv P_n$ are probabilities of the atom to be on level n.

4. Atom in an external light field.

In the presence of an external light field $E(t)$, the Hamiltonian will take the form (2) and be time-dependent. In this case the wavefunction of the atom can still be expanded as (5), due to the completeness of the set of wavefunctions $\{\varphi_n(\vec{r})\}$, but the coefficients c_n will be different each time, i.e., will be time-dependent:

$$\Psi(\vec{r}, t) = \sum_n c_n(t)\Phi_n(\vec{r}, t) = \sum_n c_n(t)\varphi_n(\vec{r})e^{-i\varepsilon_n t/\hbar}. \quad (6)$$

To this, we will apply the Schrödinger equation (1), and we will get (the dot denoting the time derivative)

$$\sum_n i\hbar[\dot{c}_n(t)\Phi_n(\vec{r}, t) + c_n(t)\dot{\Phi}_n(\vec{r}, t)] = (\hat{H}_0 + \hat{V}(t))\sum_n c_n(t)\Phi_n(\vec{r}, t),$$

But due to (4), the second term on the left and the first term on the right cancel each other, and we obtain

$$\sum_n [i\hbar\dot{c}_n(t) - c_n(t)\hat{V}(t)]\Phi_n(\vec{r}, t) = 0. \quad (7)$$

We will now left-multiply this equation by $\Phi_m^*(\vec{r}, t)$ and integrate over the coordinate, which will yield

$$i\hbar\dot{c}_m(t) = \sum_n c_n(t) \int d\vec{r}\Phi_m^*(\vec{r}, t)\hat{V}(t)\Phi_n(\vec{r}, t). \quad (8)$$

From (4),

$$\int d\vec{r}\Phi_m^*(\vec{r}, t)\hat{V}(t)\Phi_n(\vec{r}, t) = e^{i\omega_{mn}t} \int d\vec{r}\varphi_m^*(\vec{r})\hat{V}(t)\varphi_n(\vec{r}) \equiv e^{i\omega_{mn}t}V_{mn}(t),$$

where we introduced the Bohr frequency $\omega_{mn} \equiv \frac{\varepsilon_m - \varepsilon_n}{\hbar}$ and the matrix element

$V_{mn}(t) \equiv \int d\vec{r}\varphi_m^*(\vec{r})\hat{V}(t)\varphi_n(\vec{r}) = -E(t) \int d\vec{r}\varphi_m^*(\vec{r})\hat{d}(t)\varphi_n(\vec{r})$ of the transition from level n to level m.

Then we get

$$i\hbar\dot{c}_m(t) = \sum_n c_n(t)V_{mn}(t)e^{i\omega_{mn}t}. \quad (9)$$

This equation is just another form of the Schrödinger equation (1).

5. Perturbative approach.

Consider first that the light (field) is weak, hence the matrix element $V_{mn}(t)$ and the whole right-hand part of (9) can be treated as a weak perturbation. We can then write the coefficient $c_n(t) = c_n^{(0)}(t) + c_n^{(1)}(t) + \dots = \sum_s c_n^{(s)}(t)$, as a series expansion, so that $c_n^{(s)}(t) \sim V^s$, and

therefore each next term of the series is smaller than the previous one. Then, keeping in the left and right sides of (9) terms of the same order in V , we obtain

$$\begin{aligned} \dot{c}_m^{(0)}(t) &= 0, \\ i\hbar\dot{c}_m^{(1)}(t) &= \sum_n c_n^{(0)}(t)V_{mn}(t)e^{i\omega_{mn}t}, \\ i\hbar\dot{c}_m^{(2)}(t) &= \sum_n c_n^{(1)}(t)V_{mn}(t)e^{i\omega_{mn}t}, \\ &\dots \end{aligned}$$

It is reasonable to assume that at $t=0$, the atom is in some known state, for instance, $n=1$, ground state. Then, $c_n^{(0)}(t) = \delta_{n1}$, and the system can be 'decoupled':

$$i\hbar\dot{c}_m^{(1)}(t) = V_{m1}(t)e^{i\omega_{m1}t}, \dots$$

In the first order of the perturbation theory, we have the solution by integrating:

$$c_m(t) \approx c_m^{(1)}(t) = -\frac{i}{\hbar} \int_0^t dt' V_{m1}(t') e^{i\omega_{m1}t'}. \quad (10)$$

The probability of the transition from level 1 to level m is the squared modulus of this:

$$P_{1 \rightarrow m}(t) = |c_m(t)|^2.$$

At large times, the limits can be extended to infinities, hence (10) shows that the transition probability scales as the squared Fourier harmonic of the incident light field at the Bohr frequency ω_{m1} . In other words, $P_{1 \rightarrow m}(t) \propto S(\omega = \omega_{m1})$, the spectral density at ω_{m1} .

It also depends on the matrix element: if for this transition, $V_{m1} = 0$, which means that $d_{m1} = 0$, the transition is forbidden. The value of the matrix element depends on the symmetry of the electron clouds.

Similarly, we can calculate the probability of the transition back, from level m to level 1, and it will be exactly the same because the interaction Hamiltonian is a Hermitian operator, and $V_{mn} = V_{nm}^*$

Monochromatic field. Consider now the case of a monochromatic field. Recall that (see above) $V_{mn}(t)$ is determined by the matrix element of the atom dipole moment,

$$V_{mn}(t) \equiv -\vec{E}(t) \int d\vec{r} \varphi_m^*(\vec{r}) \hat{d}(t) \varphi_n(\vec{r}) = -\vec{E}(t) \vec{d}_{mn}.$$

For a monochromatic light, the real field is $E(t) = \frac{1}{2}E_0[e^{i\omega t} + e^{-i\omega t}]$, and if we also ignore the vectors, Eq. (9) becomes

$$i\hbar\dot{c}_m(t) = -\frac{1}{2}\sum_n c_n(t)d_{mn}E_0[e^{i(\omega_{mn}+\omega)t} + e^{i(\omega_{mn}-\omega)t}]. \quad (11)$$

We notice that one of the exponentials here can be, in principle, made a slowly varying function (or just a unity) provided that $\omega = \omega_{mn}$. This means a resonance of the incident radiation with the transition.

6. Atom as a two-level system.

An atom is, in the general case, not a two-level system. There are many energy levels, each of which can have sublevels. But in many cases only two levels are in resonance. Let us assume that these are levels $n=1$, $m=2$ and $\omega_{21} = \omega_0$ (Fig.2). Then the system of equations (11) takes the form

$$\begin{aligned} -2i\hbar\dot{c}_1(t) &= c_1(t)d_{11}E_0[e^{i\omega t} + e^{-i\omega t}] + c_2(t)d_{12}E_0[e^{i(\omega-\omega_0)t} + e^{i(\omega+\omega_0)t}], \\ -2i\hbar\dot{c}_2(t) &= c_1(t)d_{21}E_0[e^{i(\omega_0+\omega)t} + e^{i(\omega_0-\omega)t}] + c_2(t)d_{22}E_0[e^{i\omega t} + e^{-i\omega t}]. \end{aligned}$$

Rotating-wave approximation. All terms here will be fast oscillating except probably $e^{i(\omega-\omega_0)t}$, which has a chance to be slowly varying if the field frequency is resonant or nearly resonant. This is called the rotating-wave approximation because it is equivalent to taking into account only positive-frequency field. Then we obtain

$$\begin{aligned} \dot{c}_1(t) &= ic_2(t)\frac{d_{12}E_0}{2\hbar}e^{i(\omega-\omega_0)t}, \\ \dot{c}_2(t) &= ic_1(t)\frac{d_{21}E_0}{2\hbar}e^{i(\omega_0-\omega)t}. \end{aligned}$$

The combination $\Omega_0 \equiv \frac{d_{12}E_0}{\hbar}$ is called the Rabi frequency and let us denote the mismatch $\omega - \omega_0 \equiv \delta$; then

$$\begin{aligned} \dot{c}_1(t) &= ic_2(t)\frac{\Omega_0}{2}e^{i\delta t}, \\ \dot{c}_2(t) &= ic_1(t)\frac{\Omega_0}{2}e^{-i\delta t}. \end{aligned} \quad (12)$$

7. Rabi oscillations.

Resonant case. Assume that we have exact resonance, $\delta = 0$. Then (13) becomes

$$\begin{aligned} \dot{\tilde{c}}_1(t) &= i\tilde{c}_2(t)\frac{\Omega_0}{2}, \\ \dot{\tilde{c}}_2(t) &= i\tilde{c}_1(t)\frac{\Omega_0}{2}. \end{aligned}$$

This can be easily solved by taking the second derivative in the first equation and substituting the second one, which yields

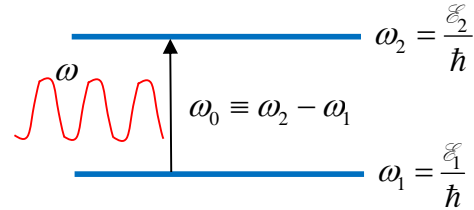


Fig.2

$$\ddot{\tilde{c}}_1(t) = -\frac{\Omega_0^2}{4}\tilde{c}_1(t),$$

$$\ddot{\tilde{c}}_2(t) = -\frac{\Omega_0^2}{4}\tilde{c}_2(t).$$

To integrate, we need the initial conditions. Let the atom state at $t=0$ be the ground one, 1. Then,

$$\tilde{c}_1(0) = 1, \quad \tilde{c}_2(0) = 0,$$

and the solutions are

$$\tilde{c}_1(t) = \cos\frac{\Omega_0 t}{2},$$

$$\tilde{c}_2(t) = i \sin\frac{\Omega_0 t}{2}.$$

Then the probabilities of the atom to be in the ground and excited states will be oscillating in time with the Rabi frequency:

$$P_1(t) \equiv |\tilde{c}_1(t)|^2 = \frac{1}{2}\{1 + \cos(\Omega_0 t)\},$$

$$P_2(t) \equiv |\tilde{c}_2(t)|^2 = \frac{1}{2}\{1 - \cos(\Omega_0 t)\}.$$

(14)

We see that the atom, starting from the ground state, will make transitions up and down – *stimulated transitions* – the faster, the stronger it is pumped by light. The probabilities of the atom to be on the two levels – one also says ‘populations of the levels’ – are shown in Fig.3.

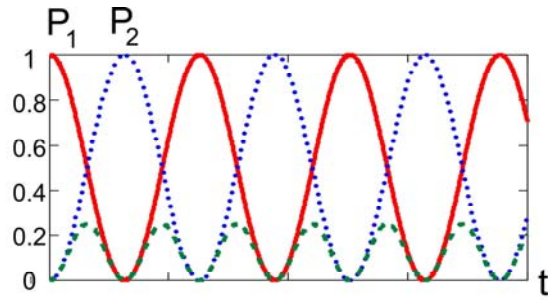


Fig. 3.

In the nonresonant case, both the frequency and the amplitude will change. For instance, the green dashed line in Fig.3 shows the situation with $\delta = \sqrt{3}\Omega_0$.

8. Pulse area.

We see that if the atom was initially in the ground state, its further time evolution depends on the electric field strength. Consider again the exact resonance. After a certain time τ , such that $\Omega_0\tau = \pi$ [see Eqs. (13)], the atom will be definitely in the excited state. One says that the area of the pulse is

$$A = \Omega_0\tau = \frac{d_{12}E_0}{\hbar}\tau.$$

Hence, a π pulse brings the atom from the ground state into the excited one. Another interesting pulse is a $\pi/2$ pulse: at

$$\Omega_0\tau = \pi/2,$$

$$\tilde{c}_1 = \frac{1}{\sqrt{2}}, \quad \tilde{c}_2 = \frac{i}{\sqrt{2}},$$

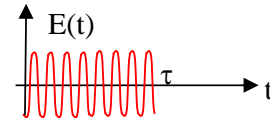


Fig.4

and the atom will be in the superposition of the ground and excited state – a so-called *coherent state*.

One can show that the shape of the pulse actually does not matter. Fig.4 shows a rectangular pulse, for which the area is given by the product $E_0\tau$, but one can define the area as $\int dtE(t)$.

9. Einstein's coefficients.

In Section 5, we obtained that the transition probability scales as the light spectral density at the transition Bohr frequency,

$$P_{1\rightarrow 2}(t) \propto S(\omega_{21}).$$

It is useful to introduce its time derivative, the transition rate, $W_{12} = \dot{P}_{1\rightarrow 2}$. For this rate, we have exactly

$$W_{12} = \frac{2\pi}{\hbar^2} |d_{21}|^2 S(\omega_{21}). \quad (15)$$

Instead of the field amplitude spectral density $S(\omega)$, one often uses the spectral energy

density, $\rho(\omega) = \frac{3}{2\pi} S(\omega)$, differing by just a numerical factor. Then, the proportionality

coefficient between the transition rate and the energy density is called the Einstein coefficient of stimulated emission:

$$W_{12} = \left(\frac{2\pi}{\hbar}\right)^2 \frac{|d_{21}|^2}{3} \rho(\omega_{21}) \equiv B_{12} \rho(\omega_{21}),$$

$$B_{12} = \frac{1}{3} \left(\frac{2\pi |d_{21}|}{\hbar}\right)^2 \quad (16)$$

The transition rates up and down should be the same, $W_{12} = W_{21}$, hence the Einstein coefficients are the same, $B_{12} = B_{21} \equiv B$. Then, if the numbers of atoms on levels 1,2 are N_1, N_2 , $N_1 + N_2 = N_0 = \text{const}$ one can write

$$\dot{N}_1 + \dot{N}_2 = 0,$$

$$\dot{N}_2 = B\rho(N_1 - N_2). \quad (17)$$

If we assume that the temperature is nonzero, then there is equilibrium (Planck) radiation, whose spectral energy density ρ is determined by the temperature:

$$\rho = \hbar k^3 \mathcal{S}(\omega) / \pi^2, \quad \mathcal{S}(\omega) = [e^{\hbar\omega/kT} - 1]^{-1}.$$

It will eventually balance the populations of the levels. However, the populations should be given by Boltzmann distribution, $N_1 / N_2 = e^{\hbar\omega/kT}$. It looks like a paradox.

As a remedy, one adds to (17) the spontaneous transitions down:

$$\dot{N}_2 = B\rho(N_1 - N_2) - AN_2. \quad (18)$$

In the stationary case, there is no time dependence, $\dot{N}_2 = 0$, and we get

$$A/B = \rho(N_1/N_2 - 1).$$

This makes sense, and the result is

$$A/B = \hbar k^3 / \pi^2.$$

For the A coefficient, we then get

$$A_{12} = \frac{4k^3}{3\hbar} |d_{21}|^2. \quad (19)$$

However, this was sort of a trick. A consistent derivation of spontaneous transitions can be only done with quantized electromagnetic field.

Home task:

Estimate the duration of a π pulse for $d_{21} = 1D$ and a 1 W laser beam focused into a waist of 10μ .

Books:

1. Scully, Zubairy, Quantum Optics, Sec. 5.2
2. I. Bloch, Licht-Atom Wechselwirkung im Zwei-Niveau System, Sec. 3
3. Klyshko, Physical Foundations of Quantum Electronics, Sections 2.4.3-2.5.1