

Fig. 2.18. (a) Normalized transition probability as a function of the detuning ( $\omega - \omega_{ba}$ ) in the rotating-wave approximation; (b) probability of a transition to the upper level as a function of time for different detuning; (c)  $|b(t)|^2$  under broadband excitation and weak fields

Since  $\lim_{x \rightarrow 0} \frac{(\sin xt)^2}{x^2} = t^2$ , the transition probability at resonance ( $\omega \rightarrow \omega_{ba}$ ) is given by

$$|b(t)|_{\omega=\omega_{ba}}^2 = \left(\frac{\sqrt{2} \Omega_{ab}}{2}\right)^2 t^2, \quad (26)$$

which increases proportionally with  $t^2$ . Note that the conclusion from above derivation is only valid under the following conditions:

$$|b(t)|^2 \ll 1, \text{ i.e., } \sqrt{2} \Omega_{ab} t^2 \ll 1 \text{ or } t \ll T = \frac{\hbar}{\Omega_{ab} E_0} = \frac{1}{\sqrt{2} \Omega_{ab}}. \quad (27)$$

This small-signal approximation only holds if the maximum interaction time  $T$  of the field with the atom is restricted to  $t \ll T$ .

Recall Heisenberg uncertainty principle, the spectral analysis of a wave with the finite detection time  $T$  gives the spectral width  $\Delta \omega \approx \frac{1}{T}$ . Thus, we cannot assume monochromaticity, but have to take into account the frequency distribution of the interaction term.

### ③. Transition Probabilities with Broad-Band Excitation.

Let us consider a radiation source with broad bandwidth. Instead of using a single frequency  $\vec{E} = \vec{E}_0 \cos \omega t$ , we introduce the spectral energy density  $\rho(\omega)$  within the frequency range of the absorption line for the radiation field. We can generalize Eq. (25) to include the interaction of broadband radiation with our two-level system by integrating Eq. (25) over all frequencies  $\omega$  of the radiation field. This yields the total transition probability  $P_{ab}(t)$

Within the time  $t$ , if  $\vec{D}_{ab} \parallel \vec{E}_0$ .

$$P_{ab}(t) = \int |b(t)|^2 d\omega = \frac{(D_{ab})^2}{2\epsilon_0 \hbar^2} \int \rho(\omega) \left[ \frac{\sin(\omega_{ba} - \omega)t/2}{(\omega_{ba} - \omega)/2} \right]^2 d\omega. \quad (28)$$

For thermal light source or broadband laser,  $\rho(\omega)$  varies slowly over the absorption line profile, so it is essentially constant over the frequency range where the sinc function is large. So,

$$\rho(\omega) \rightarrow \rho(\omega_{ba}).$$

Also, consider the integration: 
$$\int_{-\infty}^{+\infty} \frac{\sin^2(xt)}{x^2} dx = 2\pi t. \quad (29)$$

For broadband excitation, the transition probability for the time interval between 0 and  $t$  is then given by

$$P_{ab}(t) = \frac{\pi}{\epsilon_0 \hbar^2} D_{ab}^2 \rho(\omega_{ba}) t. \quad (30)$$

Note that this probability is linearly dependent on  $t$ . Thus, for broadband excitation, the transition probability per unit time

$$\frac{d}{dt} P_{ab}(t) = \frac{\pi}{\epsilon_0 \hbar^2} D_{ab}^2 \rho(\omega_{ba}) \quad (31)$$

becomes independent of time!

To compare this result with the Einstein coefficient  $B_{ab}$  introduced phenomenologically in section 6.2, we must take into account that the blackbody radiation is isotropic, while the EM wave used in above derivation propagates into one direction. In case of the isotropic radiation, the interaction term  $D_{ab}^2 \rho(\omega_{ba})$  has to be divided by a factor of 3. Thus,

$$\frac{d P_{ab}(t)}{dt} = \frac{\pi}{3\epsilon_0 \hbar^2} \rho(\omega_{ba}) D_{ab}^2 = \rho(\omega_{ba}) B_{ab} \quad (32)$$

↑  
Einstein B coefficient

Therefore, the Einstein B coefficient is given by

$$B_{ab}^{\omega} = \frac{\pi}{3\epsilon_0 \hbar^2} D_{ab}^2 \quad (33).$$

Considering the definition of  $D_{ab}$  (Eq. (18)), the Einstein coefficient  $B_{ik}$  of induced absorption  $E_i \rightarrow E_k$  becomes

$$B_{ik}^{\omega} = \frac{\pi e^2}{3\epsilon_0 \hbar^2} \langle \Phi_i | \vec{r} | \Phi_k \rangle, \quad B_{ik}^{\omega} = B_{ik}^{\omega} / 2\pi \quad (34).$$

The Einstein coefficient  $B_{ik}$  for the transition  $E_i \rightarrow E_k$  between two degenerate levels  $|i\rangle$  and  $|k\rangle$  is therefore

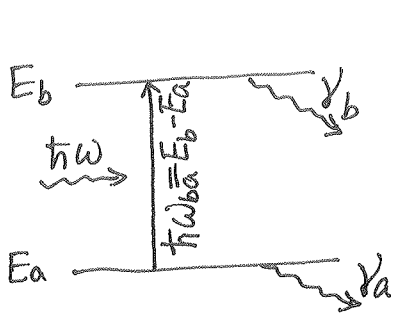
$$B_{ik} = \frac{\pi}{3\epsilon_0 \hbar^2} \frac{1}{g_i} \sum_{m=1}^{g_i} \sum_{n=1}^{g_k} |D_{imkn}|^2 = \frac{\pi}{3\epsilon_0 \hbar^2 g_i} S_{ik}. \quad (35)$$

Here the line strength  $S_{ik} = \sum_{m=1}^{g_i} \sum_{n=1}^{g_k} |D_{imkn}|^2 = |D_{ik}|^2$ .

#### ④ Phenomenological Inclusion of Decay Phenomena

Above derivation is made on a "closed" or "isolated" 2-level system, i.e.,  $|a\rangle$  and  $|b\rangle$  are not coupled with any other levels. Such system does not exist, because the states  $|a\rangle$  and  $|b\rangle$  are not only coupled by transition induced by the external radiation field, but also decay by spontaneous emission or by other relaxation processes such as collision-induced transitions, to couple with other energy levels.

We can include these decay phenomena in our formulas by adding phenomenological decay terms to Eq. (19), which can be expressed by the decay constants  $\gamma_a$  and  $\gamma_b$ . A rigorous treatment requires QED, i.e., quantization of the radiation field.



Two-level system with open decay channels into other levels interacting with an EM field.

$$\begin{cases} \dot{a}(t) = -\frac{1}{2}\gamma_a a(t) + \frac{i}{2}\sqrt{2}\gamma_{ab} e^{-i(\omega_{ba}-\omega)t} b(t) \\ \dot{b}(t) = -\frac{1}{2}\gamma_b b(t) + \frac{i}{2}\sqrt{2}\gamma_{ab} e^{+i(\omega_{ba}-\omega)t} a(t) \end{cases} \quad (36)$$

Note: Eq.(36) is obtained under the rotating-wave approximation (i.e., the  $(\omega_{ba}+\omega)$  term is neglected) for single frequency ( $\omega$ ) radiation field excitation.

When the radiation field amplitude  $E_0$  is sufficiently small, we can use the weak-signal approximation, i.e.,

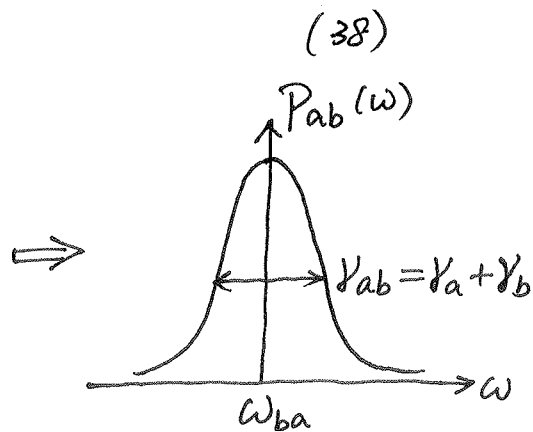
$$|a(t)|^2 = 1, \quad |b(t)|^2 \ll 1, \quad \text{and} \quad aa^* - bb^* \approx 1. \quad (37)$$

Thus, the transition probability for single frequency ( $\omega$ ) radiation field excitation and decay/relaxation is

$$\begin{aligned} P_{ab}(\omega) &= |b(t, \omega)|^2 = \int \gamma_{ab} e^{-\gamma_{ab}t} |b(t)|^2 dt \\ &= \frac{1}{2} \frac{\sqrt{2}\gamma_{ab}^2}{(\omega_{ba}-\omega)^2 + (\frac{1}{2}\gamma_{ab})^2} \end{aligned} \quad (38)$$

where  $\gamma_{ab} = \gamma_a + \gamma_b$ .

This  $P_{ab}(\omega)$  is a Lorentzian line profile with a full width at half maximum  $\gamma_{ab}$ .



### ⑤ Interaction with Strong Fields

When intense laser beams are used for the excitation of atomic transitions, the weak-field approximation is no longer valid. Therefore, we introduce Rabi's strong-field theory for 2-level system.

We consider a monochromatic field of frequency  $\omega$  and start from Eq. (19) for the probability amplitudes in the rotating-wave approximation:

$$\begin{cases} \dot{a}(t) = \frac{i}{2} \Omega_{ab} e^{-i(\omega_{ba}-\omega)t} b(t) \\ \dot{b}(t) = \frac{i}{2} \Omega_{ab} e^{+i(\omega_{ba}-\omega)t} a(t) \end{cases} \quad (39)$$

Insert the trial solution:  $a(t) = e^{i\mu t}$  into Eq. (39).

$$b(t) = \frac{2}{i\Omega_{ab}} e^{i(\omega_{ba}-\omega)t} \cdot i\mu e^{i\mu t} = \frac{2\mu}{\Omega_{ab}} e^{i(\omega_{ba}-\omega+\mu)t}$$

$$\Rightarrow \frac{2\mu}{\Omega_{ab}} e^{i(\omega_{ba}-\omega+\mu)t} = \frac{i\Omega_{ab}}{2} e^{i(\omega_{ba}-\omega+\mu)t}$$

$$\Rightarrow 2\mu(\omega_{ba}-\omega+\mu) = \Omega_{ab}^2/2 \Rightarrow \mu^2 + (\omega_{ba}-\omega)\mu - \Omega_{ab}^2/4 = 0$$

$$\Rightarrow \mu_{1,2} = -\frac{1}{2}(\omega_{ba}-\omega) \pm \frac{1}{2}\sqrt{(\omega_{ba}-\omega)^2 + \Omega_{ab}^2} \quad (40)$$

The general solutions for the amplitudes  $a$  and  $b$  are then

$$\begin{cases} a(t) = C_1 e^{i\mu_1 t} + C_2 e^{i\mu_2 t} \\ b(t) = (2/\Omega_{ab}) e^{i(\omega_{ba}-\omega)t} (C_1 \mu_1 e^{i\mu_1 t} + C_2 \mu_2 e^{i\mu_2 t}) \end{cases} \quad (41)$$

With the initial conditions  $a(0)=1$  and  $b(0)=0$ ,

$$C_1 + C_2 = 1 \quad \text{and} \quad C_1 \mu_1 = -C_2 \mu_2$$

$$\Rightarrow C_1 = -\frac{\mu_2}{\mu_1 - \mu_2}, \quad C_2 = \frac{\mu_1}{\mu_1 - \mu_2}$$

$$\text{Define } \Omega \equiv \mu_1 - \mu_2 = \sqrt{(\omega_{ba}-\omega)^2 + \Omega_{ab}^2} = \sqrt{(\omega_{ba}-\omega)^2 + (\vec{D}_{ab} \vec{E}_0 / \hbar)^2} \quad (42)$$

$\Omega$  is called the general Rabi "flopping frequency" for the nonresonant case  $\omega \neq \omega_{ba}$ .

∴ The probability amplitude

$$b(t) = i(\sqrt{2}a_b/\sqrt{2}) e^{i(\omega_{ba}-\omega)t/2} \sin(\sqrt{2}t/2) \quad (43)$$

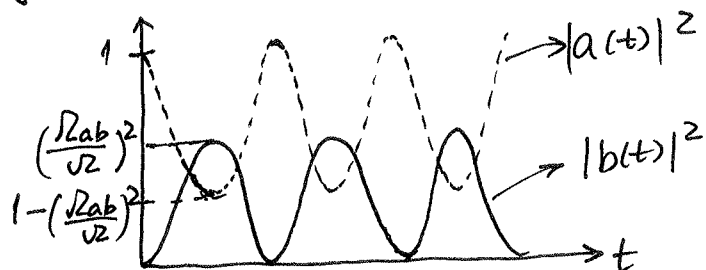
The probability of finding the system in level  $E_b$  is

$$|b(t)|^2 = (\sqrt{2}a_b/\sqrt{2})^2 \sin^2(\sqrt{2}t/2) \quad (44)$$

The probability of finding the system in level  $E_a$  is

$$|a(t)|^2 = 1 - |b(t)|^2 = 1 - (\sqrt{2}a_b/\sqrt{2})^2 \sin^2(\sqrt{2}t/2) \quad (45)$$

Eq. (44) and Eq. (45) show that the system oscillates with the frequency  $\sqrt{2}$  between the levels  $E_a$  and  $E_b$ .



At resonance  $\omega = \omega_{ba}$ ,  $\sqrt{2} \rightarrow \sqrt{2}a_b = D_{ab} E_0 / \hbar$ .

\*  $\sqrt{2}a_b$  is the Rabi flopping frequency for the resonance case  $\omega = \omega_{ba}$ , also called Rabi frequency.

At resonance  $\omega = \omega_{ba}$ ,

$$\begin{cases} |a(t)|^2 = \cos^2(\sqrt{2}a_b t/2) \\ |b(t)|^2 = \sin^2(\sqrt{2}a_b t/2) \end{cases} \quad (46)$$

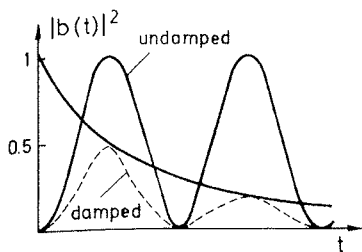


Fig. 2.20. Population probability  $|b(t)|^2$  of the levels  $E_b$  altering with the Rabi flopping frequency due to the interaction with a strong field. The resonant case is shown without damping and with damping due to decay channels into other levels. The decaying curve represents the factor  $\exp[-(\gamma_{ab}/2)t]$

On resonance,  $|a(t)|^2$  and  $|b(t)|^2$  oscillate between 0 and 1 sequentially. This means all atoms transit between  $E_a$  and  $E_b$  periodically, going from all on  $E_a$  to all on  $E_b$ , and vice versa.

We now include the damping terms  $\gamma_a$  and  $\gamma_b$ , and obtain the transition probability

$$|b(t)|^2 = \frac{\mathcal{J}_{ab}^2 e^{-\gamma_{ab}t/2} [\sin(\Omega t/2)]^2}{(\omega_{ba} - \omega)^2 + (\gamma/2)^2 + \mathcal{J}_{ab}^2} \quad (47)$$

This is the damped oscillation with the damping constant  $\frac{1}{2}\gamma_{ab} = (\gamma_a + \gamma_b)/2$ , with the Rabi flopping frequency

$$\Omega = \sqrt{(\omega_{ba} - \omega + \frac{1}{2}\gamma)^2 + \mathcal{J}_{ab}^2} \quad (48)$$

The spectral profile of the transition probability Eq. (47) is Lorentzian.