Chapter 9

Atomic Coherence Effects

When certain atomic systems interact with a coherent electromagnetic field provided by, for example, monochromatic, continuous wave (CW) laser beams, the resulting dynamics can lead to novel and unexpected behavior. These coherent, quantum behavior leads to opportunities for utilizing these atomic states for useful applications, such as quantum sensors and qubits for quantum information processing. In this Chapter, we discuss some basic coherence effects that is widely used in quantum sciences and technology today.

9.1 Optical Pumping and Coherent Dark States

We consider a three-level atomic system, with two ground states ($|0\rangle$ and $|1\rangle$) and one excited state ($|2\rangle$) coupled with two coherent laser beams $\mathcal{E}_0$ and $\mathcal{E}_1$ with frequencies ω_0 and ω_1, respectively. This type of a laser system is often referred to as a "lambda" system, as the visual representation mimics the Greek letter Λ. We assume that the polarization of $\mathcal{E}_0$ and $\mathcal{E}_1$ are such that they can drive the $|0\rangle \leftrightarrow |2\rangle$ and $|1\rangle \leftrightarrow |2\rangle$ transitions, respectively. We further allow for the laser frequency ω_1 be detuned from the energy difference between the levels $(E_2 - E_0)/\hbar$ by an amount Δ, and the frequency difference between the two laser beams $\omega_0 - \omega_1$ can be detuned from the energy difference between the ground states $(E_1 - E_0)/\hbar$ by another amount δ. We also allow spontaneous emission from the excited state $|2\rangle$ to the two ground states $|0\rangle$ and $|1\rangle$, but we can safely ignore a spontaneous decay between the two ground states $|0\rangle$ and $|1\rangle$.

9.1.1 Optical pumping

The simplest situation is when we only have one optical field $\mathcal{E}_0$ that is on resonance with the $|0\rangle \leftrightarrow |2\rangle$ transition ($\Delta = 0$). Physically, we can predict what would happen under this condition: the probability of the electron being in $|0\rangle$ will be shifted to the $|2\rangle$ state, and the $|2\rangle$ state can spontaneously decay back down to either of the two ground states based on the Clebsch-Gordon coefficients between the excited state and the ground states. When the electron decays down to the $|0\rangle$ state, it will get re-excited by the applied laser field to the $|2\rangle$ state. Then, it can decay back into one of the two ground states. This process will

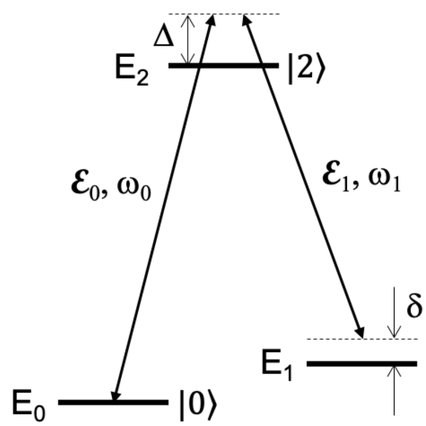

Figure 9.1: Schematic of a 3-level atomic system driven by two fields. There are two ground state energy levels ($|0\rangle$ and $|1\rangle$) and one common excited state energy level ($|2\rangle$), and two laser fields $\mathcal{E}_0$ and $\mathcal{E}_1$, with frequencies ω_1 and ω_2, driving the transition between the state $|0\rangle$ and $|1\rangle$ and the excited state $|2\rangle$, respectively. The frequencies of the laser beams can be detuned from the atomic transition by an amount Δ, and the difference between the two laser frequencies can be detuned from the energy difference $E_1 - E_0$ of the ground states by another amount δ. This type of a level structure is sometimes called a "lambda" system.

continue to repeat. When the electron decays down to the $|1\rangle$ state, then it stays there, as there is no mechanism for the electron population in $|1\rangle$ to transition to either $|0\rangle$ or $|2\rangle$ state. Eventually, all of the population will be shifted to $|1\rangle$ state under this condition. This is known as the "optical pumping" process.

This optical pumping comes with two more interesting consequences. First, if you consider a glass cell containing a lot of atoms with this structure and monitor the transmission of the laser field $\mathcal{E}_0$ through the atomic gas, you note that initially, the laser field is absorbed by the atom (to excite the electrons from the $|0\rangle$ to the $|2\rangle$ state). However, when all the atoms in its path are pumped to the $|1\rangle$ state, this laser field does not interact with the atoms any more (there are no more atoms that can absorb the field), and the field now propagates through the glass cell filled with the atomic gas unattenuated, *i.e.*, the atomic cell becomes "transparent" to the laser field. This is the first example of "induced transparency," where the optical behavior of an atom changes after some interaction with the field.

The second interesting property arises when you monitor the spontaneous emission from the atoms along the beam's path. Initially, the atoms (that start in the state $|0\rangle$) is excited to the excited state $|2\rangle$, and spontaneously emits photons. If one is monitoring the atomic emission (spontaneously emitted photons), the atoms are "bright," *i.e.*, they scatter photons due to excitation followed by spontaneous emission. However, once all the atoms are optically pumped into the $|1\rangle$ state, the atoms do not get excited any more, and therefore all spontaneous emission stops. The atoms turn "dark," in the sense that it does not scatter photons from the field $\mathcal{E}_0$ any more. This si the first example of a "dark state," formed by the interaction of the atoms with an optical field. It is interesting to note that the induced transparency and the formation of dark state originates from identical physical phenomena, and is the consequence of the same (optical pumping) process. In a sense, these two phenomena are two sides of the same coin.

Although the entire optical pumping process can be considered incoherent (mostly driven by the spontaneous emission process), it is the first example where a coherent interaction of the optical field with the atoms can create a non-trivial physical behavior. Since the process involves spontaneous emission, it cannot be fully explained by solving Schrödinger equation that describes the evolution of pure states. we must resort on the density matrix formulation to account for the spontaneous emission events to fully describe this behavior.

For the purposes of this Chapter, we can consider fully coherent interaction between the field and the atoms using Schrödinger equation (or fully coherent density matrix evolution) when the effects of the spontaneous emission is small. Then, we can consider the impact of incoherent process like spontaneous emission as necessary (in cases like optical pumping). The full treatment can be performed using the density matrix formulation, using the optical Bloch equations.

9.1.2 Coherent dark states

Next, we consider the case where we have two laser fields $\mathcal{E}_0$ and $\mathcal{E}_1$, which are fully resonant with the system ($\Delta = \delta = 0$). We consider the laser fields to be "classical," *i.e.*, we do not consider the quantization of the electromagnetic field. We further treat the interaction between the laser field and the atomic states as a perturbation. Under these assumptions, we have the Hamiltonian for the atom as

$$\hat{H}_0 = E_0 \left|0\right\rangle \left\langle 0\right| + E_1 \left|1\right\rangle \left\langle 1\right| + E_2 \left|2\right\rangle \left\langle 2\right|. \tag{9.1}$$

The coherent interaction between the field and the atomic state is discussed in Section 6.6, and the solution for the transition coefficient is derived in Eq. 6.119. There are two terms in this equation: one where the frequency difference between the two atomic levels and the frequency of the laser beams add (the first term), and the other where they subtract. Since the sum and the difference of the frequencies show up in the denominator, when we the laser is truly resonant with the atomic energy levels ($\omega_{fi} = \omega$), then the first term is going to be much smaller than the second term. So, in the "long-time limit," we can ignore the first term in favor of the second. This is called the "rotating wave approximation." Under the rotating wave approximation, the transition rate between the initial and the final state becomes constant, proportional to $V'_{fi} = \langle f|\vec{\mathcal{E}} \cdot \vec{d}|i\rangle$, which is proportional to the strength of the electric field and the dipole moment between the two states. This interaction Hamiltonian is captured as

$$\hat{H}'(t) = -\frac{\hbar}{2}\left[\Omega_0 e^{-i\omega_0 t} \left|2\right\rangle \left\langle 0\right| + \Omega_1 e^{-i\omega_1 t} \left|2\right\rangle \left\langle 1\right|\right] + h.c., \tag{9.2}$$

where $\Omega_i \equiv \vec{\mathcal{E}}_i \cdot \vec{d}_{2i}/\hbar$ is the Rabi frequency between the state $|i\rangle$ and $|2\rangle$ ($i = 0,\, 1$), $\vec{d}_{2i}$ is the dipole moment between the state $|i\rangle$ and $|2\rangle$, and *h.c.* indicates the Hermitian conjugate of the first term in the equation. This model of interaction between the field and a two-level system is called "Janyes-Cummings interaction."

When this Hamiltonian acts on the atom, we can solve for the evolution of the quantum state

$$|\psi(t)\rangle = c_0(t)\,|0\rangle + c_1(t)\,|1\rangle + c_2(t)\,|2\rangle\,. \tag{9.3}$$

The Schrödinger equation reads

$$i\hbar\frac{d}{dt}\,|\psi(t)\rangle = \left[\hat{H}_0 + \hat{H}'(t)\right]|\psi(t)\rangle\,, \tag{9.4}$$

from which we derive three equations

$$\dot{c}_0(t) + \frac{iE_0}{\hbar}c_0(t) = \frac{i}{2}\Omega_0 e^{i\omega_0 t}c_2(t), \tag{9.5}$$

$$\dot{c}_1(t) + \frac{iE_1}{\hbar}c_1(t) = \frac{i}{2}\Omega_1 e^{i\omega_1 t}c_2(t), \tag{9.6}$$

$$\dot{c}_2(t) + \frac{iE_2}{\hbar}c_2(t) = \frac{i}{2}\left[\Omega_0 e^{-i\omega_0 t}c_0(t) + \Omega_1 e^{-i\omega_1 t}c_1(t)\right]. \tag{9.7}$$

Defining $c_i'(t) = c_i(t)e^{iE_it/\hbar}$ (for i = 0, 1, 2) and $\omega_{2j} = (E_2 - E_j)/\hbar$ (for j = 0, 1) leads to three equations for the $c_i'(t)$ as

$$\dot{c}_0'(t) = \frac{i}{2}\Omega_0 e^{i(\omega_0-\omega_{20})t}c_2'(t), \tag{9.8}$$

$$\dot{c}_1'(t) = \frac{i}{2}\Omega_1 e^{i(\omega_1-\omega_{21})t}c_2'(t), \tag{9.9}$$

$$\dot{c}_2'(t) = \frac{i}{2}\left[\Omega_0 e^{-i(\omega_0-\omega_{20})t}c_0'(t) + \Omega_1 e^{-i(\omega_1-\omega_{21})t}c_1'(t)\right]. \tag{9.10}$$

When the frequency of the optical fields are resonant with the atomic transitions ($\omega_j = \omega_{2j}$), the equations simplify to

$$\dot{c}_0'(t) = \frac{i}{2}\Omega_0 c_2'(t), \tag{9.11}$$

$$\dot{c}_1'(t) = \frac{i}{2}\Omega_1 c_2'(t), \tag{9.12}$$

$$\dot{c}_2'(t) = \frac{i}{2}\left[\Omega_0 c_0'(t) + \Omega_1 c_1'(t)\right]. \tag{9.13}$$

Taking the time derivative of Eq. 9.13, and inserting the other two leads to

$$\ddot{c}_2'(t) = -\frac{1}{4}(\Omega_0^2 + \Omega_1^2)c_2'(t). \tag{9.14}$$

Letting $\Omega = \sqrt{\Omega_0^2 + \Omega_1^2}$, we find the solution $c_2'(t) = A\cos\Omega t/2 + B\sin\Omega t/2$. Inserting this solution back into Eqs. 9.11 and 9.12 yields the solutions

$$c_0'(t) = iA\frac{\Omega_0}{\Omega}\sin\frac{\Omega t}{2} - iB\frac{\Omega_0}{\Omega}\cos\frac{\Omega t}{2} + C_0, \tag{9.15}$$

$$c_1'(t) = iA\frac{\Omega_1}{\Omega}\sin\frac{\Omega t}{2} - iB\frac{\Omega_1}{\Omega}\cos\frac{\Omega t}{2} + C_1. \tag{9.16}$$

Using the initial conditions $c_i'(0)$, we can express A and B in terms of the initial conditions as

$$\begin{aligned} A &= c_2'(0), & (9.17) \\ B &= i\left[\frac{\Omega_0}{\Omega}(c_0'(0) - C_0) + \frac{\Omega_1}{\Omega}(c_1'(0) - C_1)\right]. & (9.18) \end{aligned}$$

So, the final solution for the coefficients are

$$\begin{aligned} c_0'(t) &= c_0'(0)\left[\frac{\Omega_1^2}{\Omega^2} + \frac{\Omega_0^2}{\Omega^2}\cos\frac{\Omega t}{2}\right] + c_1'(0)\frac{\Omega_0\Omega_1}{\Omega^2}\left[\cos\frac{\Omega t}{2} - 1\right] + ic_2'(0)\frac{\Omega_0}{\Omega}\sin\frac{\Omega t}{2}, & (9.19) \\ c_1'(t) &= c_0'(0)\frac{\Omega_0\Omega_1}{\Omega^2}\left[\cos\frac{\Omega t}{2} - 1\right] + c_1'(0)\left[\frac{\Omega_0^2}{\Omega^2} + \frac{\Omega_1^2}{\Omega^2}\cos\frac{\Omega t}{2}\right] + ic_2'(0)\frac{\Omega_0}{\Omega}\sin\frac{\Omega t}{2}, & (9.20) \\ c_2'(t) &= c_2'(0)\cos\frac{\Omega t}{2} + i\left[\frac{\Omega_0}{\Omega}c_0'(0) + \frac{\Omega_1}{\Omega}c_1'(0)\right]\sin\frac{\Omega t}{2}. & (9.21) \end{aligned}$$

If we start from an initial condition where $c_0'(0) = \Omega_1/\Omega$, $c_1'(0) = -\Omega_0/\Omega$ and $c_2'(0) = 0$, then we see that we have a steady state, where $c_i'(t) = c_i'(0)$ for all t. This means that the linear superposition state

$$|D\rangle = \sin\theta\,|0\rangle - \cos\theta\,|1\rangle\,, \quad (9.22)$$

where $\cos\theta = \Omega_0/\Omega$ and $\sin\theta = \Omega_1/\Omega$, is a steady state of the Hamiltonian where the excited state is never populated. This linear superposition state is called the "coherent dark state," as it is formed by the coherent optical fields that is driving the atomic transitions. Similar to the optical pumping case, the presence of spontaneous emission will force any initial state of the system to eventually end up in this dark state, and the two optical fields are transparent against this state. Since there are two ground states that form the ground state manifold, the formation of a dark state by a linear superposition also means that there is an orthogonal state

$$|B\rangle = \cos\theta\,|0\rangle + \sin\theta\,|1\rangle\,, \quad (9.23)$$

which absorbs and scatters the photons by coupling maximally to the excited state $|2\rangle$. This state is called the coherent bright state.

This mechanism shows that certain atomic system, when driven with resonant fields, the population will be driven to the coherent dark state, so it is sometimes called "coherent population trapping." This usually is formed when the number of excited state is fewer than the ground states, such as the lambda system.

9.2 Applications of Atomic Coherence

9.2.1 Sub-Doppler cooling

The simplest laser cooling scheme is to use the Doppler effect. In this approach, a laser beam is red-detuned to an atomic transition ($\Delta < 0$). For an atom moving away from the laser beam, it experiences further red-detuning and the atom does not interact with the laser beam. For an atom moving towards the laser beam, it experiences blue-detuning, and the light comes closer to resonance with the atom. In this case, the atom will absorb a photon from the laser, getting a momentum kick $\hbar\vec{k}$ in the direction against its motion, where $\vec{k}$ is the wavevector of the photon. Once the photon is absorbed by the atom, the excitation is eventually released in the form of spontaneous emission, where the photon is emitted in random direction. The momentum recoil the atom experiences comes in units of $\hbar\vec{k}$ as well, only this time the direction of the recoil is random. On the average, the atom preferentially absorbs photons against the direction of its motion, and emits the photon in random directions. This means that the momentum kick it gets is preferential against its direction of motion. This effectively slows down the center of mass motion of the atom, leading to cooling. This mechanism is called Doppler cooling. For atoms moving in free space, one needs two counter-propagating red-detuned laser beams to cool the motion of the atoms in each dimension. For a three-dimensional cooling, one would need six counter-propagating beams. Doppler cooling can also be realized for atoms in a trap. When the atom is trapped in a potential well, it's motion tends to be oscillatory, and the atom moves back and forth. Under a single laser beam aligned along the direction of oscillation, the atom will experience Doppler cooling during the half-cycle period when it is moving towards the beam. In a three-dimensional trap, Doppler cooling in all three directions is achieved with just a single red-detuned laser beam, if it is aligned such that the $\vec{k}$-vector of the beam has projections in all three motional directions of the trap.

There are two mechanisms that dictate the limits of Doppler cooling. The first limit arises from the natural linewidth of the atomic transition. If the atomic resonance has the natural linewidth of γ, a photon can be absorbed with a frequency uncertainty of $\sim \gamma$ near its transition frequency. This translates to the velocity uncertainty over which the momentum kick from the beam can happen. The effective temperature of the final atomic motional state can be estimated from the velocity distribution. The equivalent temperature that can be reached by the Doppler cooling mechanism is called the "Doppler temperature," and is given by

$$T_{Doppler} = \frac{\hbar\gamma}{2k_B}, \tag{9.24}$$

where k_B is the Boltzmann constant. The Doppler temperature is reached when the laser frequency is red-detuned by about half the linewidth of the transition from resonance. One can reach a lower Doppler temperature if the natural linewidth of the transition is narrower. For a transition with a typical dipole transition, $\gamma \sim 10^7$Hz, and the corresponding Doppler temperature is $\sim 40\mu$K.

The second mechanism that limits the temperature of the atom(s) is the recoil limit. This arises due to the random momentum kick that the atom experiences due to spontaneous emission of the absorbed photons. The temperature limit due to this recoil effect is called the "recoil temperature," and is given by

$$T_{recoil} = \frac{\hbar^2 k^2}{2Mk_B}, \tag{9.25}$$

where M is the mass of the atom being cooled. In a typical atom, the recoil temperature limit is much lower than Doppler temperature. For example, ^{87}Rb atom with transition wavelength of 780nm, the recoil limit is about 180nK. For $^{171}Yb^+$ ion with a transition wavelength of 370nm, the recoil limit is about 407nK.

Cooling the motion of atoms below Doppler temperature is often referred to as "sub-Doppler cooling." There are many mechanisms discovered so far for sub-Doppler cooling, such as the polarization gradient cooling (also known as the Sisyphus cooling), sideband-resolved Raman cooling, and cooling using atomic coherence effects. The coherent population trapping effect discussed in this chapter can be used to achieve sub-Doppler cooling.

Basically, any resonant absorption features with an effective linewidth narrower than the natural linewidth of the atomic transition provides an opportunity for the Doppler-like cooling effect to feature final temperatures that are lower than what's expected of the natural linewidth. One example is the sub-Doppler cooling due to velocity-selective coherent population trapping, first demonstrated by A. Aspect *et al.* [Phys. Rev. Lett. 61, 826 (1988)]. Here, we note that the population trapping discussed in previous section is only exact if there is no Doppler effect present. In this experiment, they arrange the two laser fields $\mathcal{E}_0$ and $\mathcal{E}_1$ in a counter-propagating geometry. For those atoms that have zero velocity along the propagating direction of the beams, the coherent population trapping is perfect and those atoms do not interact with the laser beams at all. However, when the atomic velocity is non-zero, the Doppler effect leads to a small detuning δ between the two counter-propagating beams experienced by the atom, where the detuning is proportional to the velocity of the atom. As the velocity of the atom approaches zero, the atom spends more and more time in the coherent dark state, and scattering less and less photons. Depending on the velocity of the atom, the state of the atom can be decomposed into a linear superposition of the dark state $|D\rangle$ and the bright state $|B\rangle$. The absorption rate, which is proportional to the probability of the atom being in the bright state, is on the order of $\sim \gamma(kv/M)^2/\Omega^2$, where v is the velocity of the atom, k is the wavevector of the photons in the beam, and Ω is the Rabi frequency between the bright state $|B\rangle$ and the excited state $|2\rangle$. We note that this scattering rate is proportional to the velocity of the atom (thus the term velocity-selective coherent population trapping), and can be made small depending on the Rabi frequency Ω^2 which is proportional to the intensity of the laser beams. There is no limit to how narrow one can make this scattering dip, as long as there is sufficient laser power. Indeed, the paper demonstrates cooling to below the single photon recoil temperature. When the photon is indeed scattered due to the presence of the bright state component, the atom will get a momentum kick of $\pm\hbar k$. The fact that the two peaks corresponding to $\pm\hbar k$ is visible in the atomic momentum distribution indicates that the momentum uncertainty along that direction is smaller than the recoil limit of $\hbar k$.

9.2.2 Electromagnetically-induced transparency (EIT)

Electromagnetically induced transparency (EIT) is a widely used phenomena in modern quantum technology. In this example, one considers the transmission of a weak probe beam $\mathcal{E}_1$ propagating through the atomic media, in the presence of a strong coupling beam $\mathcal{E}_0$. "Weak" beam implies that the coherent Rabi coupling (indicated by the Rabi frequency) of the beam is much lower than the spontaneous emission rate, so that that the incoherent effect dominates. On the other hand, "strong" coupling means that the Rabi frequency is either comparable to or larger than the spontaneous emission rate. We consider the case where the frequency ω_0 of the coupling field $\mathcal{E}_0$ is resonant with the transition $|0\rangle \leftrightarrow |2\rangle$ (*i.e.*, $\Delta = 0$), and we scan the frequency ω_1 of the probe beam across the resonance (*i.e.*, scan the detuning δ across zero).

In the absence of the coupling field, the atomic medium will be opaque to the weak probe beam, as the photons will be absorbed by the atom and spontaneously emitted. The transmission of the probe beam through the atomic medium will be very small, over a detuning scan δ over the linewidth of the $|1\rangle \leftrightarrow |2\rangle$ transition.

To understand the effect of the coupling beam, one can for the time being consider a two-level system coupled with a coherent field in the Jaynes-Cummings Hamiltonian, considered in Eq. 9.2, as

$$\hat{H}'(t) = -\frac{\hbar}{2}\Omega_0 e^{-i\omega_0 t} |2\rangle \langle 0| + h.c. \tag{9.26}$$

with the trial function

$$|\psi(t)\rangle = c_0(t) |0\rangle + c_2(t) |2\rangle . \tag{9.27}$$

The Schrödinger equation reduces to

$$\dot{c}_0'(t) = \frac{i}{2}\Omega_0 c_2'(t), \tag{9.28}$$

$$\dot{c}_2'(t) = \frac{i}{2}\Omega_0 c_0'(t). \tag{9.29}$$

The solution to these equations are given by

$$c_0'(t) = c_0'(0)\cos\frac{\Omega_0 t}{2} + ic_2'(0)\sin\frac{\Omega_0 t}{2}, \tag{9.30}$$

$$c_2'(t) = c_2'(0)\cos\frac{\Omega_0 t}{2} + ic_0'(0)\sin\frac{\Omega_0 t}{2}. \tag{9.31}$$

When the initial state is in $|0\rangle$ ($c_0'(0) = 1$ and $c_2'(0) = 0$), this solution shows an oscillation between the $|0\rangle$ and $|2\rangle$ states, as $|c_0'(t)|^2 = \cos^2 \Omega_0 t/2$ and $|c_2'(t)|^2 = \sin^2 \Omega_0 t/2$. it also shows that there are two stationary states, when $c_0'(0) = 1/\sqrt{2}$ and $c_2'/(0) = \pm 1/\sqrt{2}$, where

$$c_0'(t) = \frac{1}{\sqrt{2}}\left[\cos\frac{\Omega_0 t}{2} \pm i\sin\frac{\Omega_0 t}{2}\right] = \frac{1}{\sqrt{2}}e^{\pm i\Omega_0 t/2} \tag{9.32}$$

$$c_2'(t) = \frac{1}{\sqrt{2}}\left[\pm\cos\frac{\Omega_0 t}{2} + i\sin\frac{\Omega_0 t}{2}\right] = \pm\frac{1}{\sqrt{2}}e^{\pm i\Omega_0 t/2}. \tag{9.33}$$

The two resulting states

$$|\psi_+(t)\rangle = e^{i\Omega_0 t/2}\ \frac{|0\rangle + |2\rangle}{\sqrt{2}} \tag{9.34}$$

$$|\psi_-(t)\rangle = e^{-i\Omega_0 t/2}\ \frac{|0\rangle - |2\rangle}{\sqrt{2}} \tag{9.35}$$

are stationary states of the Hamiltonian which does not evolve in time. The eigenvalues corresponding to these states are $E_0 + E_2 \pm \hbar\Omega_0/2$.

When the strong coupling field is turned on, the field "dresses" the excited state $|2\rangle$ and "splits" its energy level into two by $\hbar\Omega_0$ its coupling to the $|0\rangle$ state. The size of this splitting is determined by the Rabi frequency of oscillation between the two states $|0\rangle$ and $|2\rangle$ induced by the coupling field. Now the probe beam sees two possible transitions to the two dressed states. It turns out that the transition coefficients from the $|1\rangle$ state to these two dressed states driven by the probe field have opposite sign, and they *destructively interfere* at exactly the middle point. Therefore, when the probe field is tuned to the exact resonance, the excitation probability becomes exactly zero, rendering the atomic medium transparent. This is the physical principle of EIT.