

# Chapter 4: Quantum Optical Control

## 1 Laser cooling

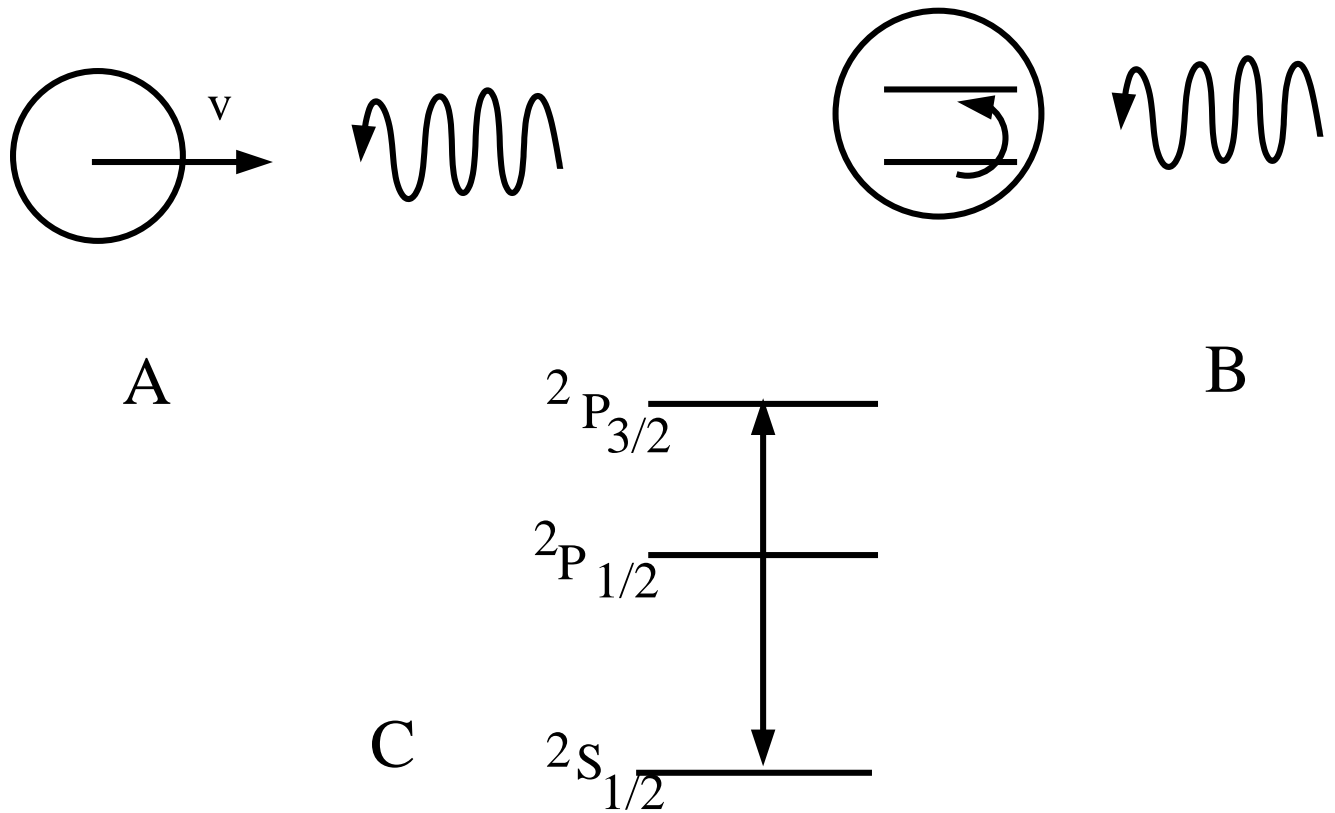


Figure 1: Figure A shows how an atom is hit with light with momentum  $\hbar k$  and slows down. Figure B shows atom will absorb light if frequency  $\hbar\omega$  matches the difference of energy between the internal energy levels. Figure C shows these energy levels for sodium. These are the electronic states with  $n = 3$ .

Many experiments in physics require slow atoms. At room temperatures, we have atoms moving at say 300 m/s. We like to slow them down to say 10 m/s. Slow atoms are used to make Bose Einstein condensates (velocity as low as cm/s). How do we slow atoms. We can do it by hitting them with light of the right frequency. Atoms have internal energy levels.

When the frequency of the laser light matches this, the light is absorbed (optical transition). But light carries momentum  $\hbar k$  and therefore on absorption atom gets a kick which slows it down. The absorbed photon is spontaneously emitted but in a random direction. When we average over many such absorption and emission, the kick is always in the same direction while the recoil due to spontaneous emission is random and averages to zero and in the end atom slows down. If frequency of the laser light is  $\omega$  an atom moving towards the light source will see the frequency shifted to  $\omega(1 + \frac{v}{c})$ . Then if detune the laser frequency to be slightly less than the internal energy level, due to this doppler shift the atom will see just the right frequency and will absorb it. Atoms moving slowly will have negligible doppler shift and will not absorb light. Hence we will only cool fast atoms and not slow ones. This way we will bring all to same velocity. This is the basic idea of laser cooling. It has been successful in cooling atoms to very low velocities where they form a Bose Einstein condensate. Figure C shows these energy levels for sodium. These are the electronic states with principal quantum number  $n = 3$ . The transition shown is called the  $D_2$  line.

## 2 Infrared Spectroscopy

Consider a Diatomic molecule as shown in Fig. 2. When the two nuclei are too close they will repel. Their repulsion is shielded by electron cloud but when they come inside the cloud this shielding doesn't work well and there is strong repulsion. When we pull very far they practically donot talk to each other. There is zero attraction/repulsion. When we bring them close then as discussed in chapter 3, their electron clouds begin to overlap and they form molecular bond which lowers the energy and we get a negative energy. If we trace out the potential energy as function of interatomic distance  $r$ , we will find a surface like Fig. 2B. Repulsion at close distance meaning positive potential energy and negative energy at optimal separation  $r_0$ .

Now in chapter 1, on quantum mechanics, we studied Harmonic oscillator. where potential energy as function of distance had a similar surface like Fig. 2B. There we talked about wavefunctions  $\phi_n(x)$ , then we have same way wavefunctions here  $\phi_n(r - r_0)$  which describe interatomic separation. These are shown as ladder rugs in Fig. 2B. They are called vibrational levels. When we apply a electric field along say  $x$  direction, then there is transition element.

$$E_x \langle \phi_k | x | \phi_l \rangle,$$

which will induce transition between these wavefunctions. The energy difference between wavefunctions is in infrared range (THz). For instance, if  $\epsilon_0$  is energy of  $\phi_0$  and  $\epsilon_1$  energy of  $\phi_1$ , then let  $\hbar\omega_0 = \epsilon_1 - \epsilon_0$ . When we shine infrared light it will be absorbed. The frequency difference  $\hbar\omega_0$  is characteristic of the molecule. Therefore depending on which frequency is

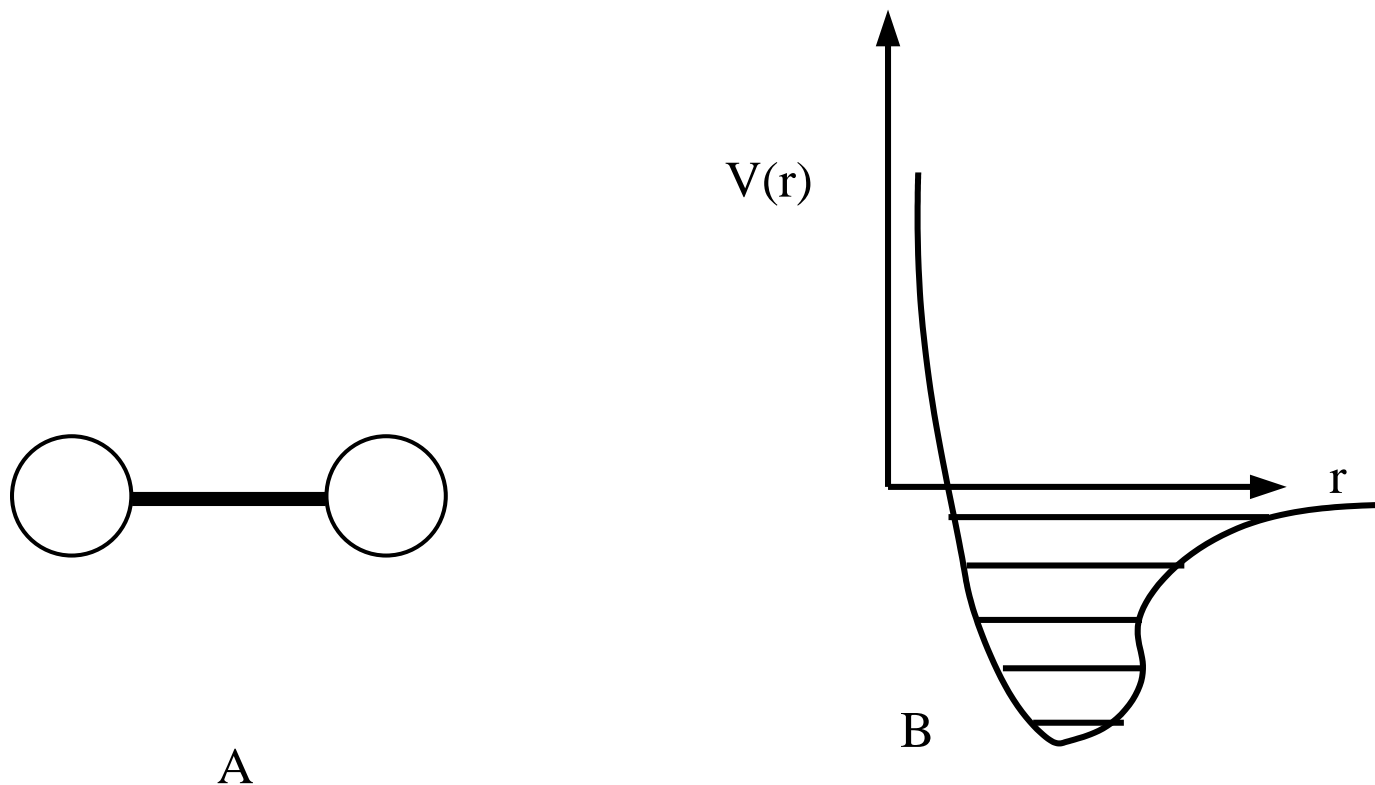


Figure 2: Figure A shows a diatomic molecule. Fig. B shows the potential energy as a function of distance.

absorbed, we get finger print spectra of the molecule. This is called vibrational spectroscopy or infrared spectroscopy.

### 3 Raman Spectroscopy

In last section we talked about how we can probe vibrational energy levels of a molecule using infra-red light and get finger print information of the molecule. There is another spectroscopic technique that does this, it also probes vibrational energy levels of a molecule but not with infra-red light but light in visible range i.e.  $10^{15}$  Hz. How is this possible, as we discussed that vibrational energies differ by tera-hertz frequency. The way experiment works is we shine light in visible range on the sample and it gets scattered of the sample and a light of different frequency comes out The difference of the frequencies of the input and out is the vibrational energy in tera-hertz frequency. The difference in the input and output tells us about the molecule as the difference probes the vibrational energy and we

get a fingerprint information of the molecule. How does all this work, to understand this we make a small digression.

We first describe three level atomic system so called  $\Lambda$  system. In a  $\Lambda$  system as shown in Fig. 3 we have two ground state levels  $|1\rangle$  and  $|3\rangle$  at energy  $E_1$  and excited level  $|2\rangle$  at energy  $E_2$ . The transition from  $|1\rangle$  to  $|2\rangle$  has strength  $\Omega_1$  and transition from  $|2\rangle$  to  $|3\rangle$  has strength  $\Omega_2$ . In the interaction frame of natural Hamiltonian of the system, we get a second order term connecting level  $|1\rangle$  to  $|3\rangle$  with strength  $\frac{\Omega_1\Omega_2}{(E_1-E_2)}$ . This term creates an effective coupling between ground state levels and drives transition from  $|1\rangle$  to  $|3\rangle$ . Scattering processes in QED can be modelled like this. Feynman amplitudes are calculation of second order term  $\mathcal{M} = \frac{\Omega_1\Omega_2}{(E_1-E_2)}$ .

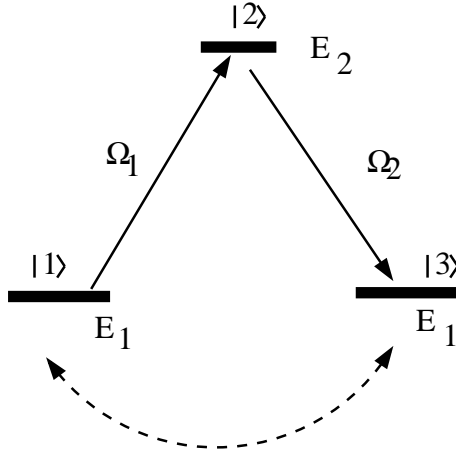


Figure 3: Above Fig. shows a three level  $\Lambda$  system with two ground state levels  $|1\rangle$  and  $|3\rangle$  and an excited level  $|2\rangle$ .

The state of the three level system evolves according to the Schrödinger equation

$$\dot{\psi} = \frac{-i}{\hbar} \begin{bmatrix} E_1 & \Omega_1^* & 0 \\ \Omega_1 & E_2 & \Omega_2^* \\ 0 & \Omega_2 & E_1 \end{bmatrix} \psi. \quad (1)$$

We proceed into the interaction frame of the natural Hamiltonian (system energies) by transformation

$$\phi = \exp\left(\frac{i}{\hbar} \begin{bmatrix} E_1 & 0 & 0 \\ 0 & E_2 & 0 \\ 0 & 0 & E_1 \end{bmatrix}\right) \psi. \quad (2)$$

This gives for  $\Delta E = E_2 - E_1$ ,

$$\dot{\phi} = \frac{-i}{\hbar} \underbrace{\begin{bmatrix} 0 & \exp(-\frac{i}{\hbar}\Delta E t)\Omega_1^* & 0 \\ \exp(\frac{i}{\hbar}\Delta E t)\Omega_1 & 0 & \exp(\frac{i}{\hbar}\Delta E t)\Omega_2^* \\ 0 & \exp(-\frac{i}{\hbar}\Delta E t)\Omega_2 & 0 \end{bmatrix}}_{H(t)} \phi. \quad (3)$$

$H(t)$  is periodic with period  $\Delta t = \frac{2\pi}{\Delta E}$ . After  $\Delta t$ , the system evolution is

$$\phi(\Delta t) = (I + \int_0^{\Delta t} H(\sigma)d\sigma + \int_0^{\Delta t} \int_0^{\sigma_1} H(\sigma_1)H(\sigma_2)d\sigma_2d\sigma_1 + \dots)\phi(0). \quad (4)$$

The first integral averages to zero, while the second integral

$$\int_0^{\Delta t} \int_0^{\sigma_1} H(\sigma_1)H(\sigma_2)d\sigma_2d\sigma_1 = \frac{1}{2} \int_0^{\Delta t} \int_0^{\sigma_1} [H(\sigma_1), H(\sigma_2)]d\sigma_2d\sigma_1. \quad (5)$$

Evaluating it explicitly, we get for our system that second order integral is

$$\frac{-i\Delta t}{\hbar} \begin{bmatrix} 0 & \frac{\Omega_1^*\Omega_2^*}{E_1-E_2} \\ 0 & 0 \\ \frac{\Omega_1\Omega_2}{\underbrace{E_1-E_2}_{\mathcal{M}}} & 0 \\ 0 & 0 \end{bmatrix}. \quad (6)$$

Thus we have created an effective Hamiltonian

$$\begin{bmatrix} 0 & \frac{\Omega_1^*\Omega_2^*}{E_1-E_2} \\ 0 & 0 \\ \frac{\Omega_1\Omega_2}{E_1-E_2} & 0 \end{bmatrix}, \quad (7)$$

which couples level  $|1\rangle$  and  $|3\rangle$  and drives transition between them at rate  $\mathcal{M} = \frac{\Omega_1\Omega_2}{(E_1-E_2)}$ .

Now lets come back to Raman spectroscopy. We have talked about vibrational energy levels. In 4A we plot them again from two configurations of the electrons. One in the ground state (recall bonding orbital from chapter 3) and one in excited sate (antibonding orbital). These two orbitals have energy difference in eV range corresponding to optical frequencies. For each electronic configuration we have vibrational energy levels.

Now lets simplify as in 4B, where two vibrational levels in bottom manifold with energy  $\epsilon_1, \epsilon_3$  and one in top at energy  $\epsilon_2$ .

Now suppose we start with level 1 and we bring in optical photon of frequency  $\omega_1$ , then lets say our initial energy is  $E_1 = \hbar\omega_1 + \epsilon_1$ , the atomic energy plus the photon energy. This photon induces a transition to level 2 which has energy  $E_2 = \epsilon_2$  and finally we can emit a photon of frequency  $\omega_2$  and make a transition to level 3. The total energy of the emitted photon plus atomic energy is  $E_3 = \hbar\omega_2 + \epsilon_3$ . Observe  $E_1 = E_3$  when  $\hbar(\omega_1 - \omega_2) = \epsilon_3 - \epsilon_1$ .

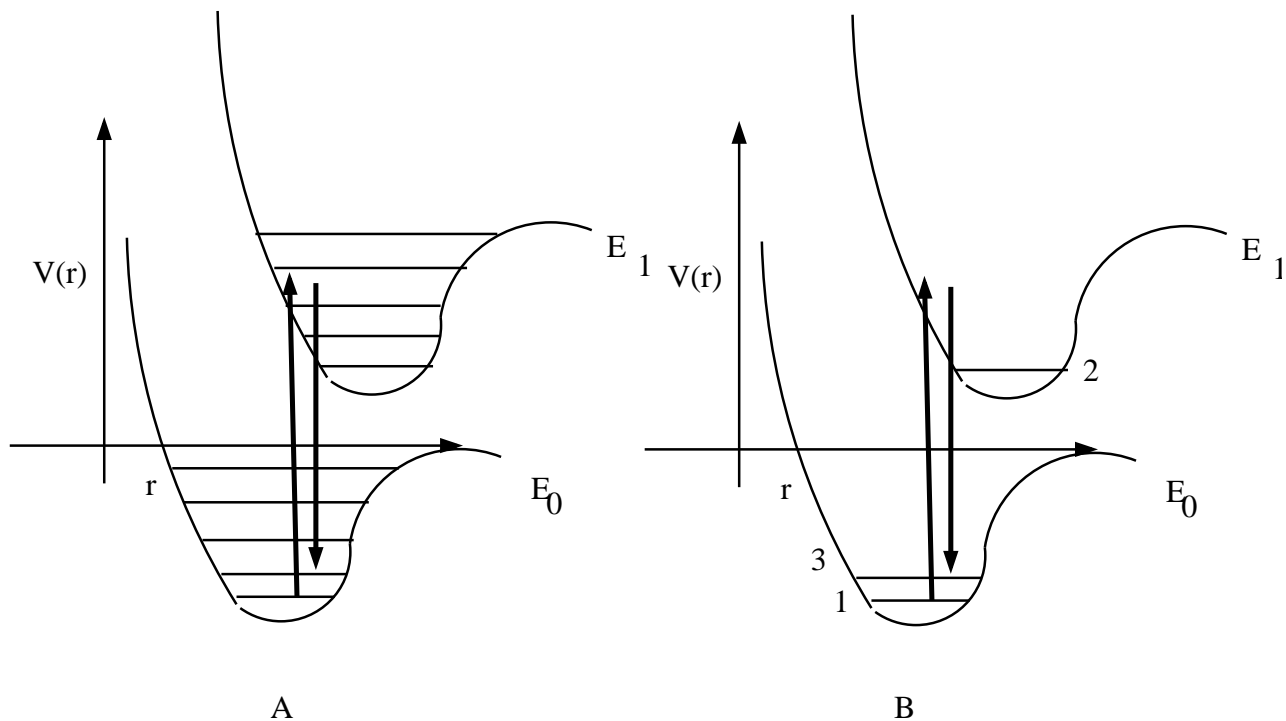


Figure 4: Above Fig. shows vibrational energy levels with electrons in ground state and vibrational energy levels with electrons in excited state.

Then we have constructed a three level system from joint atom-photon states. In this three level system that we have constructed we have a transition amplitude between initial state which is atom in level 1 and photon  $\omega_1$  in to final state, atom in level 3 and photon  $\omega_2$ . The transition goes as  $\mathcal{M} = \frac{\Omega_1\Omega_2}{(E_1-E_2)}$ , where  $E_1 - E_2 = \hbar\omega_1 - (\epsilon_2 - \epsilon_1)$ .

When  $\hbar\omega_1$  is close to  $\epsilon_2 - \epsilon_1$ , we get a big enhancement in  $\mathcal{M} = \frac{\Omega_1\Omega_2}{(E_1-E_2)}$ . This is called *Resonance Raman spectroscopy*.

## 4 Microwave Spectroscopy and Ovens

We have talked NMR spectroscopy done with radio frequency. Infrared spectroscopy done with terahertz frequency and Raman spectroscopy done with light. We will end the chapter with a brief discussion of microwave spectroscopy done with microwaves (GHz) for example as in microwave oven we use in our kitchen. The basic idea is to address the rotational states of molecules and make transitions between various rotational states. The energy difference between rotational states is in GHz frequency range. When we absorb radiation of this frequency we move between rotational states. Once gain this specific absorption frequency

is a fingerprint signature of the molecule. To make matters more concrete

If  $l$  is the angular momentum quantum number then the rotational kinetic energy is

$$E_r = \frac{\hbar^2 l(l+1)}{2I} = \hbar\omega, \quad (8)$$

where  $I$  is moment of inertia of the molecule. For a given  $l$ , there are  $L = 2l + 1$  states given by quantum number  $m$ , corresponding to angular momentum along say  $z$  direction, where  $m$  goes in steps of 1 and  $-l \leq m \leq l$ .

When we absorb a microwave photon, we make a transition between different  $l$  so that energy difference is the energy of the photon.  $m$  increments by 1 as angular momentum is conserved. In a microwave oven we excite water molecules to higher energy rotational states using microwaves. The oven is a cavity. The excited molecules dissipate energy to relax back in turn heating our food.