

Chapter 1

Atom Light Interactions

1.1 Wave Function description

1.1.1 Basics

Time dependent Schrödinger equation:

$$\hat{H}\Psi(r, t) = i\hbar \frac{d\Psi(r, t)}{dt} \quad (1.1)$$

stationary solution

$$\hat{H}_{Atom}\psi_n(r) = E_n\psi_n(r) \quad (1.2)$$

with

$$\Psi_n(r, t) = \exp(-i\frac{E_nt}{\hbar})\psi_n(r) \quad (1.3)$$

in the following we will consider a 2-level system $|\psi_1\rangle$ and $|\psi_2\rangle$ with energy eigen values E_1 and E_2 . The energy difference is then related to the transition frequency $\omega_0/2\pi$

$$\hbar\omega_0 = E_2 - E_1 \quad (1.4)$$

Let us assume the atom is driven by an external electro magnetic field (frequency $\omega/2\pi$, interaction Hamiltonian $\hat{H}_i$). The total Hamiltonian is then:

$$\hat{H} = \hat{H}_{Atom} + \hat{H}_i \quad (1.5)$$

the solution to the Schrödinger equation 1.1 can then be written as:

$$\Psi(r, t) = C_1(t)\Psi_1(r, t) + C_2(t)\Psi_2(r, t) \quad (1.6)$$

with $|C_1(t)|^2 + |C_2(t)|^2 = 1$. Inserting Eq. 1.6 into Eq. 1.1 one obtains the following equations for the coefficients $C_1(t)$ and $C_2(t)$

$$\begin{aligned} C_1 M_{11} + C_2 \exp(-i\omega_0 t) M_{12} &= i \frac{dC_1}{dt} \\ C_1 \exp(i\omega_0 t) M_{21} + C_2 M_{22} &= i \frac{dC_2}{dt} \end{aligned} \quad (1.7)$$

where M_{mj} are the transition matrix elements

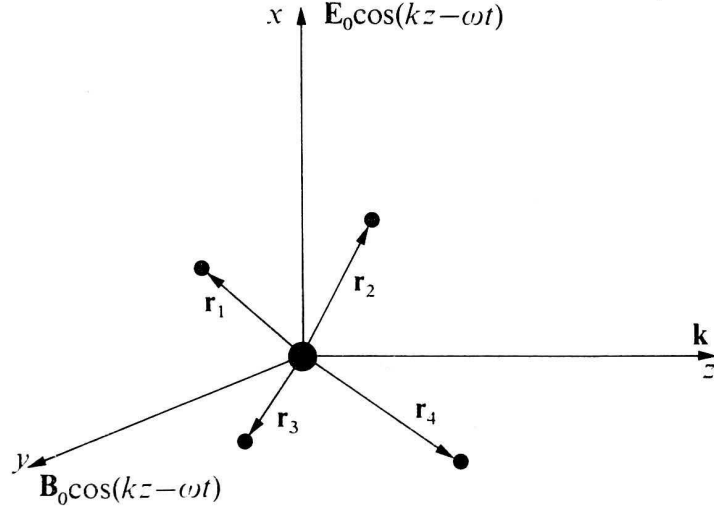


FIG. 2.1. Coordinate system for the atom and the electromagnetic wave.

Figure 1.1: Dipole approximation for the atom-light interaction

1.1.2 Dipole matrix elements

The transition matrix elements M_{mj} are given by:

$$\hbar M_{mj} = \int \psi_m^* \hat{H}_i \psi_j dV = \langle \psi_m | \hat{H}_i | \psi_j \rangle \quad (1.8)$$

with

$$\hat{H}_i = e\mathbf{D} \cdot \mathbf{E}_0 \cos(\omega t) \quad (1.9)$$

$e\mathbf{D}$ being the total electric dipole. From symmetry one immediately sees:

$$\begin{aligned} M_{11} &= M_{22} = 0 \\ M_{12} &= M_{21}^* = \frac{1}{\hbar} e E_0 X_{12} \cos(\omega t) \end{aligned} \quad (1.10)$$

where

$$X_{12} = \int \psi_1^* X \psi_2 dV = \langle \psi_1 | X | \psi_2 \rangle \quad (1.11)$$

is the dipole matrix element. For further on we define the Rabi frequency Ω_{Rabi} as

$$\Omega_{Rabi} = \frac{1}{\hbar} e E_0 X_{12} \quad (1.12)$$

1.1.3 Rabi oscillations

Using these matrix elements the above equations (Eq.1.7) can be now written as:

$$\begin{aligned} \Omega_{Rabi} \cos(\omega t) \exp(-i\omega_0 t) C_2 &= i \frac{dC_1}{dt} \\ \Omega_{Rabi}^* \cos(\omega t) \exp(i\omega_0 t) C_1 &= i \frac{dC_2}{dt} \end{aligned} \quad (1.13)$$

the time dependent terms $\cos(\omega t) \exp(-i\omega_0 t)$ can be rewritten using $\cos(\omega t) = \frac{1}{2}(e^{i\omega t} + e^{-i\omega t})$ as:

$$\cos(\omega t) \exp(-i\omega_0 t) = \frac{1}{2} (\exp(-i(\omega - \omega_0)t) + \exp(-i(\omega + \omega_0)t)) \quad (1.14)$$

for $|\omega - \omega_0| \ll \omega$ we can neglect the fast oscillating terms $(\omega + \omega_0)t$. The evolution will be governed by the slow oscillating terms. This approximation is called the **Rotating Wave Approximation (RWA)**.

$$\begin{aligned} \Omega_{Rabi} \frac{1}{2} \exp(-i(\omega - \omega_0)t) C_2 &= i \frac{dC_1}{dt} \\ \Omega_{Rabi}^* \frac{1}{2} \exp(i(\omega - \omega_0)t) C_1 &= i \frac{dC_2}{dt} \end{aligned} \quad (1.15)$$

for zero detuning: $\omega = \omega_0$ one finds then the well known **Rabi oscillations** between the ground and excited state of the driven two level system. With the starting conditions $|C_1|^2 = 1$ and $|C_2|^2 = 0$ one finds:

$$\begin{aligned} |C_1|^2 &= \cos^2(\Omega_{Rabi}t/2) \\ |C_2|^2 &= \sin^2(\Omega_{Rabi}t/2) \end{aligned} \quad (1.16)$$

Equations 1.13 provide an exact description of the state of a two level atom (without decay) interacting with an oscillating electric field. But for general solution, and because the interesting quantities are not the bare coefficients C_i but the probabilities $|C_i|^2$ they are best transformed into equations for the density matrix.

In addition it is not simple to include the decay of the excited state in the wave function description of the atom-light interaction.

1.2 Optical Bloch Equations

From the coefficients C_1 and C_2 we can form equations for the density matrix of the atom:

$$\begin{aligned} \rho_{11} &= |C_1|^2 = \frac{N_1}{N} \\ \rho_{22} &= |C_2|^2 = \frac{N_2}{N} \\ \rho_{12} &= C_1 C_2^* \\ \rho_{21} &= C_2 C_1^* \end{aligned} \quad (1.17)$$

with the diagonal elements ρ_{11} and ρ_{22} satisfying

$$\rho_{11} + \rho_{22} = 1 \quad (1.18)$$

and the off diagonal matrix elements are in general complex and they satisfy

$$\rho_{12} = \rho_{21}^* \quad (1.19)$$

1.2.1 Optical Bloch Equations without damping due to spontaneous emission

One can find the equations of motion for the density matrix easily from the equations of motion of the coefficients C_1 and C_2 equation 1.13

$$\begin{aligned} \frac{d\rho_{22}}{dt} = -\frac{d\rho_{11}}{dt} &= -i \cos(\omega t) [\Omega_{Rabi}^* \exp(i\omega_0 t) \rho_{12} - \Omega_{Rabi} \exp(-i\omega_0 t) \rho_{21}] \\ \frac{d\rho_{12}}{dt} = +\frac{d\rho_{21}^*}{dt} &= +i \Omega_{Rabi} \cos(\omega t) \exp(-i\omega_0 t) (\rho_{11} - \rho_{22}) \end{aligned} \quad (1.20)$$

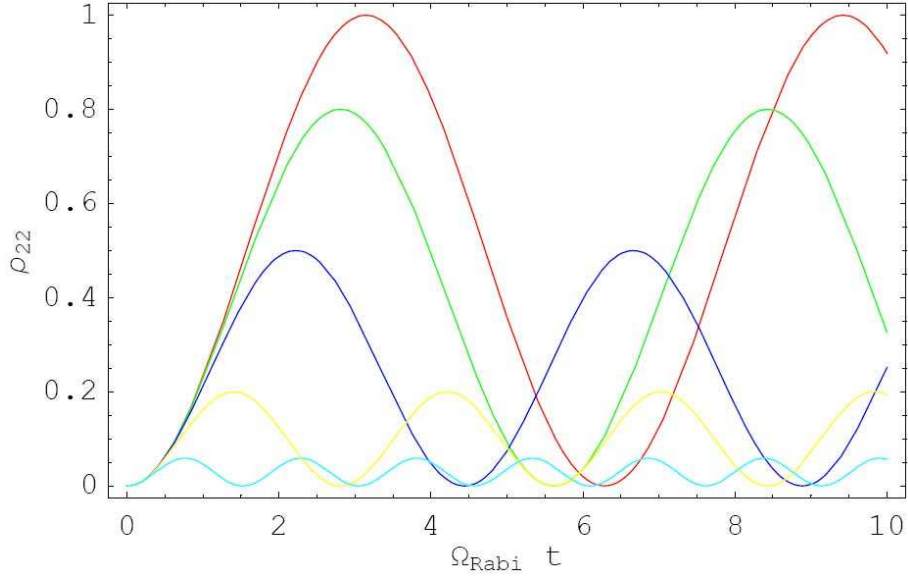


Figure 1.2: Driven Atom without spontaneous emission. At $t=0$ the atom is in the ground state ($\rho_{11}(0) = 1$, $\rho_{22}(0) = 0$). The probability to find the atom in the excited state is plotted for various detunings $\Delta = 0$, $\Delta = 0.5\Omega_{Rabi}$, $\Delta = 1\Omega_{Rabi}$, $\Delta = 2\Omega_{Rabi}$, $\Delta = 4\Omega_{Rabi}$

apply the **rotating wave approximation**: ($|\Delta| = |\omega - \omega_0| \ll \omega_0$)

$$\begin{aligned} \frac{d\rho_{22}}{dt} = -\frac{d\rho_{11}}{dt} &= -\frac{1}{2}i\Omega_{Rabi}^* \exp(i(\omega_0 - \omega)t)\rho_{12} + \frac{1}{2}i\Omega_{Rabi} \exp(-i(\omega_0 - \omega)t)\rho_{21} \quad (1.21) \\ \frac{d\rho_{12}}{dt} = -\frac{d\rho_{21}^*}{dt} &= +\frac{1}{2}i\Omega_{Rabi} \exp(-i(\omega_0 - \omega)t)(\rho_{11} - \rho_{22}) \end{aligned}$$

These can be solved using the Ansatz:

$$\begin{aligned} \rho_{11} &= \rho_{11}^{(0)} \exp(\lambda t) \\ \rho_{22} &= \rho_{22}^{(0)} \exp(\lambda t) \\ \rho_{12} &= \rho_{12}^{(0)} \exp(-i(\omega_0 - \omega)t) \exp(\lambda t) \\ \rho_{21} &= \rho_{21}^{(0)} \exp(-i(\omega_0 - \omega)t) \exp(\lambda t) \end{aligned} \quad (1.22)$$

which leads to the following equation:

$$\begin{bmatrix} -\lambda & 0 & \frac{1}{2}i\Omega_{Rabi}^* & -\frac{1}{2}i\Omega_{Rabi} \\ 0 & -\lambda & -\frac{1}{2}i\Omega_{Rabi}^* & \frac{1}{2}i\Omega_{Rabi} \\ \frac{1}{2}i\Omega_{Rabi} & -\frac{1}{2}i\Omega_{Rabi} & i(\omega_0 - \omega) - \lambda & 0 \\ -\frac{1}{2}i\Omega_{Rabi}^* & \frac{1}{2}i\Omega_{Rabi}^* & 0 & -i(\omega_0 - \omega) - \lambda \end{bmatrix} \cdot \begin{bmatrix} \rho_{11}^{(0)} \\ \rho_{22}^{(0)} \\ \rho_{12}^{(0)} \\ \rho_{21}^{(0)} \end{bmatrix} = 0 \quad (1.23)$$

and the following eigen value equation:

$$\lambda^2[\lambda^2 + (\omega_0 - \omega)^2 + |\Omega_{Rabi}|^2] = 0 \quad (1.24)$$

with the solutions:

$$\begin{aligned}\lambda_1 &= 0 \\ \lambda_2 &= i\Omega \\ \lambda_3 &= -i\Omega\end{aligned}\tag{1.25}$$

thereby we used

$$\Omega = \sqrt{(\omega_0 - \omega)^2 + |\Omega_{Rabi}|^2}\tag{1.26}$$

The general solution is then:

$$\rho_{ij} = \rho_{ij}^{(1)} + \rho_{ij}^{(2)} \exp(i\Omega t) + \rho_{ij}^{(3)} \exp(-i\Omega t)\tag{1.27}$$

For the special initial conditions:

$$\begin{aligned}\rho_{11} &= 1 \\ \rho_{22} &= 0 \\ \rho_{12} &= \rho_{21} = 0\end{aligned}\tag{1.28}$$

one finds:

$$\begin{aligned}\rho_{22}(t) &= \frac{|\Omega_{Rabi}|^2}{\Omega^2} \sin^2\left(\frac{1}{2}\Omega t\right) \\ \rho_{12}(t) &= \exp(-i(\omega_0 - \omega)t) \frac{\Omega_{Rabi}}{\Omega^2} \sin\left(\frac{1}{2}\Omega t\right) \left(-(\omega_0 - \omega) \sin\left(\frac{1}{2}\Omega t\right) + i\Omega \cos\left(\frac{1}{2}\Omega t\right)\right)\end{aligned}\tag{1.29}$$

and for resonant light ($\omega_0 = \omega$) the solutions become even simpler:

$$\begin{aligned}\rho_{22}(t) &= \sin^2\left(\frac{1}{2}\Omega_{Rabi}t\right) \\ \rho_{12}(t) &= \frac{\Omega_{Rabi}}{|\Omega_{Rabi}|} \sin\left(\frac{1}{2}\Omega_{Rabi}t\right) \cos\left(\frac{1}{2}\Omega_{Rabi}t\right)\end{aligned}\tag{1.30}$$

1.2.2 Optical Bloch Equations with damping due to spontaneous emission

Apply the rotating wave approximation: ($|\Delta| = |\omega - \omega_0| \ll \omega_0$)

$$\begin{aligned}\frac{d\rho_{22}}{dt} = -\frac{d\rho_{11}}{dt} &= -\frac{1}{2}i\Omega_{Rabi}^* \exp(i(\omega_0 - \omega)t)\rho_{12} + \frac{1}{2}i\Omega_{Rabi} \exp(-i(\omega_0 - \omega)t)\rho_{21} - 2\gamma\rho_{22} \\ \frac{d\rho_{12}}{dt} = +\frac{d\rho_{21}^*}{dt} &= +\frac{1}{2}i\Omega_{Rabi} \exp(-i(\omega_0 - \omega)t)(\rho_{11} - \rho_{22}) - \gamma\rho_{12}\end{aligned}\tag{1.31}$$

only in the case of resonant light ($\omega_0 = \omega$) we can give a general solution: for the special initial conditions:

$$\begin{aligned}\rho_{11} &= 1 \\ \rho_{22} &= 0 \\ \rho_{12} &= \rho_{21} = 0\end{aligned}\tag{1.32}$$

one finds:

$$\rho_{22} = \frac{\frac{1}{2}|\Omega_{Rabi}|^2}{2\gamma^2 + |\Omega_{Rabi}|^2} \left(1 - \left(\cos(\lambda t) + \frac{3\gamma}{2\lambda} \sin(\lambda t)\right) \exp\left(-\frac{3\gamma}{2}t\right)\right)\tag{1.33}$$

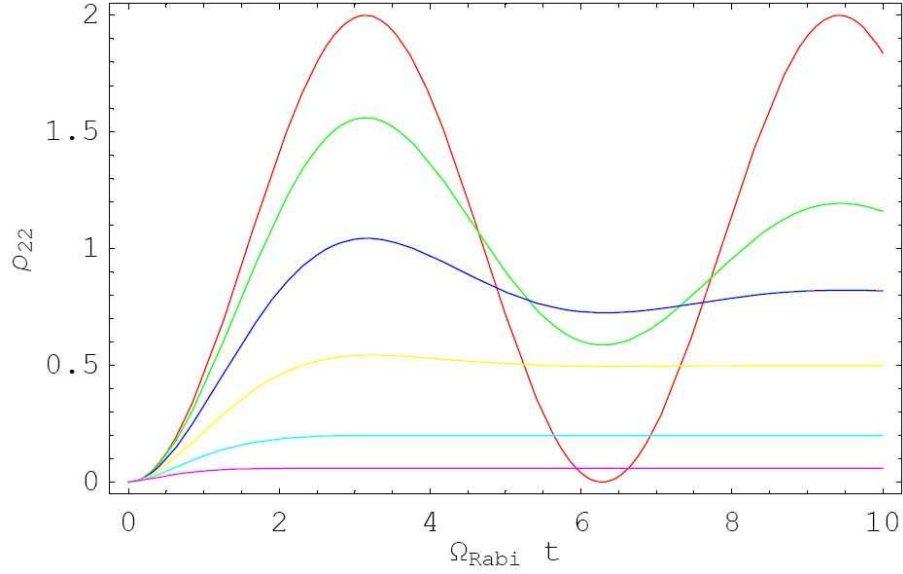


Figure 1.3: Driven Atom with spontaneous emission. At $t=0$ the atom is in the ground state ($\rho_{11}(0) = 1$, $\rho_{22}(0) = 0$). The probability to find the atom in the excited state is plotted for various ratios of $\frac{\gamma}{\Omega_{Rabi}}$, $\gamma = 0$, $\gamma = 0.1\Omega_{Rabi}$, $\gamma = 0.25\Omega_{Rabi}$, $\gamma = 0.5\Omega_{Rabi}$, $\gamma = \Omega_{Rabi}$, $\gamma = 2\Omega_{Rabi}$

with

$$\lambda = \sqrt{|\Omega_{Rabi}|^2 + \frac{1}{2}\gamma^2} \quad (1.34)$$

in general there are no closed solutions for these equations 1.31. So let's first look at the steady state solutions:

$$\begin{aligned} \rho_{22} &= \frac{\frac{1}{4}|\Omega_{Rabi}|^2}{(\omega_0 - \omega)^2 + \gamma^2 + \frac{1}{2}|\Omega_{Rabi}|^2} \\ \rho_{12} &= \exp(-i(\omega_0 - \omega)t) \frac{\frac{1}{2}|\Omega_{Rabi}|(\omega_0 - \omega - i\gamma)}{(\omega_0 - \omega)^2 + \gamma^2 + \frac{1}{2}|\Omega_{Rabi}|^2} \end{aligned} \quad (1.35)$$

Table 1.1: Table of Symbols

ρ_{ij}	density matrix element i,j
Ω_{Rabi}	Rabi frequency
ω	laser frequency
ω_0	atomic transition frequency
Δ	detuning : $\omega - \omega_0$
γ	line width
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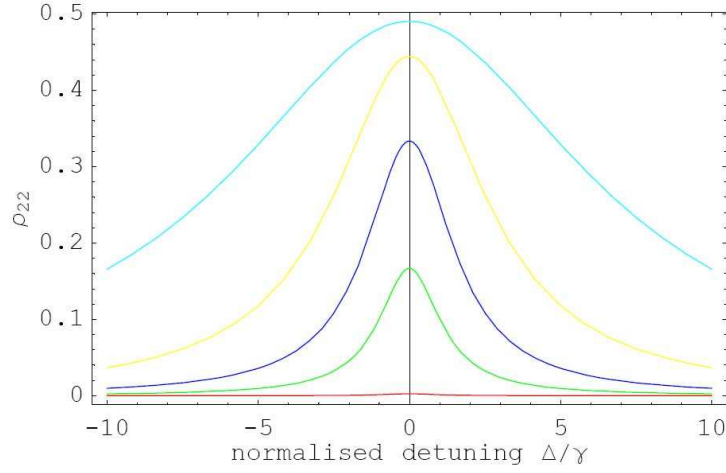


Figure 1.4: Driven Atom with spontaneous emission. The probability to find the atom in the excited state is plotted for various driving strength $\Omega_{Rabi} = \gamma$, $\Omega_{Rabi} = 0.1\gamma$, $\Omega_{Rabi} = 2\gamma$, $\Omega_{Rabi} = 4\gamma$, $\Omega_{Rabi} = 10\gamma$. One clearly sees the broadening and saturation of the line

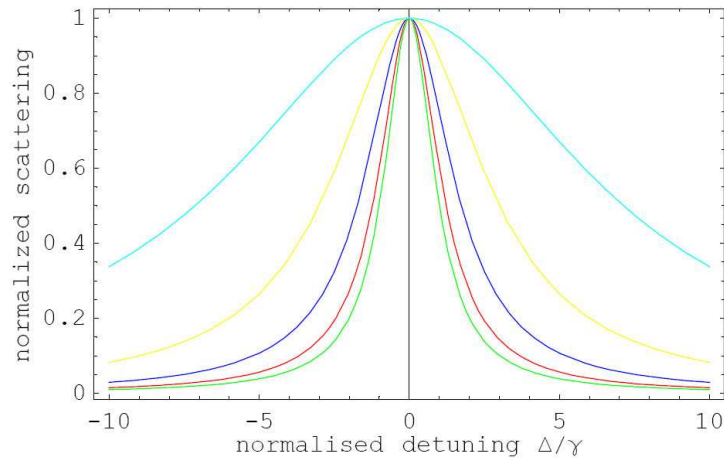


Figure 1.5: Driven Atom with spontaneous emission. Form of the scattering line $\rho_{22}(\Delta)/\rho_{22}(0)$ plotted for various driving strength $\Omega_{Rabi} = 0.1\gamma$, $\Omega_{Rabi} = \gamma$, $\Omega_{Rabi} = 2\gamma$, $\Omega_{Rabi} = 4\gamma$, $\Omega_{Rabi} = 10\gamma$. One clearly sees the broadening and saturation of the line

Chapter 2

The Two Level System: Resonance

2.1 Introduction

The cornerstone of contemporary Atomic, Molecular and Optical Physics (AMO Physics) is the study of atomic and molecular systems and their interactions through their resonant interaction with applied oscillating electromagnetic fields. The thrust of these studies has evolved continuously since Rabi performed the first resonance experiments in 1938. In the decade following World War II the edifice of quantum electrodynamics was constructed largely in response to resonance measurements of unprecedented accuracy on the properties of the electron and the fine and hyperfine structure of simple atoms. At the same time, nuclear magnetic resonance and electron paramagnetic resonance were developed and quickly became essential research tools for chemists and solid state physicists. Molecular beam magnetic and electric resonance studies yielded a wealth of information on the properties of nuclei and molecules, and provided invaluable data for the nuclear physicist and physical chemist. This work continues: the elucidation of basic theory such as quantum mechanics, tests of such quantum electrodynamics, the development of new techniques, the application of old techniques to more systems, and the universal move to even higher precision continues unabated. Molecular beams studies, periodically invigorated by new sources of higher intensity or new species (eg. clusters) are carried out in numerous laboratories - chemical as well as physical - and new methods for applying the techniques of nuclear magnetic resonance are still being developed.

Properly practiced, resonance techniques controllably alter the quantum mechanical state of a system without adding any uncertainty. Thus resonance techniques may be used not only to learn about the structure of a system, but also to prepare it in a particular way for further use or study. Because of these two facets, resonance studies have lead physicists through a fundamental change in attitude - from the passive study of atoms to the active control of their internal quantum state and their interactions with the radiation field. This active approach is embodied in the existence of lasers and in the study and creation of coherence phenomena generally. One current frontier in AMO physics is the active control of the external (translational) degree of freedom.

The chief technical legacy of the early work on resonance spectroscopy is the family of lasers which have sprung up like the brooms of the sorcerer's apprentice. The scientific applications of these devices have been prodigious. They caused the resurrection of physical optics- now freshly christened quantum optics- and turned it into one of the liveliest fields in physics. They have had a similar impact on atomic and molecular spectroscopy. In addition they have led to new families of physical studies such as single particle spectroscopy, multiphoton excitation,

cavity quantum electrodynamics, and laser cooling and trapping, to name but a few of the many developments.

This chapter is about the interactions of a two state system with a sinusoidally oscillating field whose frequency is close to the natural resonance frequency of the system. The phrase “two level” (ie. two possibly degenerate levels with unspecified m quantum number) is less accurate than the phrase two state. However, its misuse is so widespread that we adopt it anyway - at least it correctly suggests that the two states have different energies. The oscillating field will be treated classically, and the linewidth of both states will be taken as zero until near the end of the chapter where relaxation will be treated phenomenologically.

2.2 Damped Resonance in a Classical System

Because the terminology of classical resonance, as well as many of its features, are carried over into quantum mechanics, we start by reviewing an elementary resonant system. Consider a harmonic oscillator composed of a series RLC circuit. The charge obeys

$$\ddot{q} + \gamma\dot{q} + \omega_0^2 q = 0 \quad (2.1)$$

where $\gamma = R/L$, $\omega_0^2 = 1/LC$. Assuming that the system is underdamped (i.e. $\gamma^2 < 4\omega_0^2$), the solution is a linear combination of

$$\exp\left(-\frac{\gamma}{2}t\right)\exp(\pm i\omega't) \quad (2.2)$$

where $\omega' = \omega_0\sqrt{1 - \gamma^2/4\omega_0^2}$. If $\omega \gg \gamma$, which is often the case, we have $\omega' \equiv \omega_0$. The energy in the circuit is

$$W = \frac{1}{2C}q^2 + \frac{1}{2}L\dot{q}^2 = W_0 e^{-\gamma t} \quad (2.3)$$

where $W_0 = W(t=0)$. The lifetime of the stored energy is $\tau = \frac{1}{\gamma}$.

If the circuit is driven by a voltage $E_0 e^{i\omega t}$, the steady state solution is $q_0 e^{i\omega t}$ where

$$q_0 = \frac{E_0}{2\omega_0 L} \frac{1}{(\omega_0 - \omega + i\gamma/2)}. \quad (2.4)$$

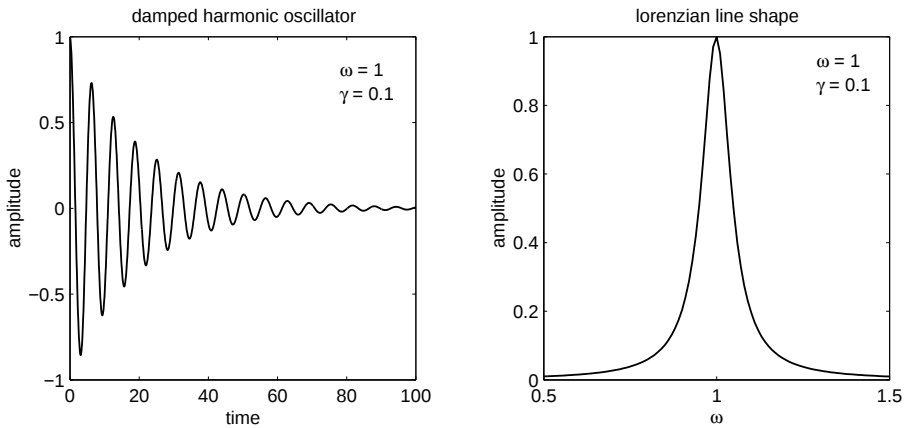


Figure 2.1: Damped harmonic oscillator

(We have made the usual resonance approximation: $\omega \approx 2\omega_0(\omega_0 - \omega)$.) The average power delivered to the circuit is

$$P = \frac{1}{2} \frac{E_0^2}{R} \frac{1}{1 + \left(\frac{\omega - \omega_0}{\gamma/2}\right)^2} \quad (2.5)$$

The plot of P vs ω (Fig. 1) is a universal resonance curve often called a "Lorentzian curve". The full width at half maximum ("FWHM") is $\Delta\omega = \gamma$. The quality factor of the oscillator is

$$Q = \frac{\omega_0}{\Delta\omega} \quad (2.6)$$

Note that the decay time of the free oscillator and the linewidth of the driven oscillator obey

$$\tau\Delta\omega = 1 \quad (2.7)$$

This can be regarded as an uncertainty relation. Assuming that energy and frequency are related by $E = \hbar\omega$ then the uncertainty in energy is $\Delta E = \hbar\Delta\omega$ and

$$\tau\Delta E = \hbar \quad (2.8)$$

It is important to realize that the Uncertainty Principle merely characterizes the spread of individual measurements. Ultimate precision depends on the experimenter's skill: the Uncertainty Principle essentially sets the scale of difficulty for his or her efforts.

The precision of a resonance measurement is determined by how well one can "split" the resonance line. This depends on the signal to noise ratio (S/N). (see Fig. 2) As a rule of thumb, the uncertainty $\delta\omega$ in the location of the center of the line is

$$\delta\omega = \frac{\Delta\omega}{S/N} \quad (2.9)$$

In principle, one can make $\delta\omega$ arbitrarily small by acquiring enough data to achieve the required statistical accuracy. In practice, systematic errors eventually limit the precision. Splitting a line by a factor of 10^4 is a formidable task which has only been achieved a few times, most notably in the measurement of the Lamb shift. A factor of 10^3 , however, is not uncommon, and 10^2 is child's play.

2.3 Magnetic Resonance: Classical Spin in Time-varying B-Field

The two-level system is basic to atomic physics because it approximates accurately many physical systems, particularly systems involving resonance phenomena. All two-level systems obey the same dynamical equations: thus to know one is to know all. The archetype two level system is a spin 1/2 particle such as an electron, proton or neutron. The spin motion of an electron or a proton in a magnetic field, for instance, displays the total range of phenomena in a two level system. To slightly generalize the subject, however, we shall also include the motion of atomic nuclei. In table 2.1 there is a summary of their properties.

2.3.1 The Classical Motion of Spins in a Static Magnetic Field

The interaction energy and equation of motion of a classical spin in a static magnetic field are given by

$$W = -\vec{\mu} \cdot \vec{B} \quad (2.10)$$

$$\vec{F} = -\nabla W = \nabla(\vec{\mu} \cdot \vec{B}) \quad (2.11)$$

$$\text{torque} = \vec{\mu} \times \vec{B} \quad (2.12)$$

In a uniform field, $\vec{F} = 0$. The torque equation ($\frac{d\vec{L}}{dt} = \text{torque}$) gives

$$\frac{d\vec{J}}{dt} = \gamma \vec{J} \times \vec{B} = -\gamma \vec{B} \times \vec{J} \quad (2.13)$$

To see that the motion of $\vec{J}$ is pure precession about $\vec{B}$, imagine that $\vec{B}$ is along $\hat{z}$ and that the spin, $\vec{J}$, is tipped at an angle θ from this axis, and then rotated at an angle $\phi(t)$ from the $\hat{x}$ axis (ie., θ and ϕ are the conventionally chosen angles in spherical coordinates). The torque, $-\gamma \vec{B} \times \vec{J}$, has no component along $\vec{J}$ (that is, along $\hat{r}$), nor along $\hat{\theta}$ (because the $\vec{J} - \vec{B}$ plane contains $\hat{\theta}$), hence $-\gamma \vec{B} \times \vec{J} = -\delta B |J| \sin \theta \hat{\phi}$. This implies that $\vec{J}$ maintains constant magnitude and constant tipping angle θ . Since the $\hat{\phi}$ -of component of $\vec{J}$ is $d\vec{J}/dt = |J| \sin \theta \frac{d\phi}{dt}$ it is clear that $\phi(t) = -\gamma B t$. This solution shows that the moment precesses with angular velocity

$$\Omega_L = -\gamma B \quad (2.14)$$

where Ω_L is called the **Larmor Frequency**.

For electrons, $\gamma_e/2\pi = 2.8$ MHz/gauss, for protons $\gamma_p/2\pi = 4.2$ kHz/gauss. Note that Planck's constant does not appear in the equation of motion: the motion is classical.

2.3.2 Rotating Coordinate Transformation

A second way to find the motion is to look at the problem in a rotating coordinate system. If some vector $\vec{A}$ rotates with angular velocity $\vec{\Omega}$, then

$$\frac{d\vec{A}}{dt} = \vec{\Omega} \times \vec{A} \quad (2.15)$$

If the rate of change of the vector in a system rotating at $\vec{\Omega}$ is $(d\vec{A}/dt)_{rot}$, then the rate of change in an inertial system is the motion *in* plus the motion *of* the rotating coordinate system.

$$\left(\frac{d\vec{A}}{dt}\right)_{inert} = \left(\frac{d\vec{A}}{dt}\right)_{rot} + \vec{\Omega} \times \vec{A} \quad (2.16)$$

The operator prescription for transforming from an inertial to a rotating system is thus

$$\left(\frac{d}{dt}\right)_{rot} = \left(\frac{d}{dt}\right)_{inert} - \vec{\Omega} \times \quad (2.17)$$

Applying this to Eq.2.13 gives

$$\left(\frac{d\vec{J}}{dt}\right)_{rot} = \gamma \vec{J} \times \vec{B} - \vec{\Omega} \times \vec{J} = \gamma \vec{J} \times (\vec{B} + \vec{\Omega}/\gamma) \quad (2.18)$$

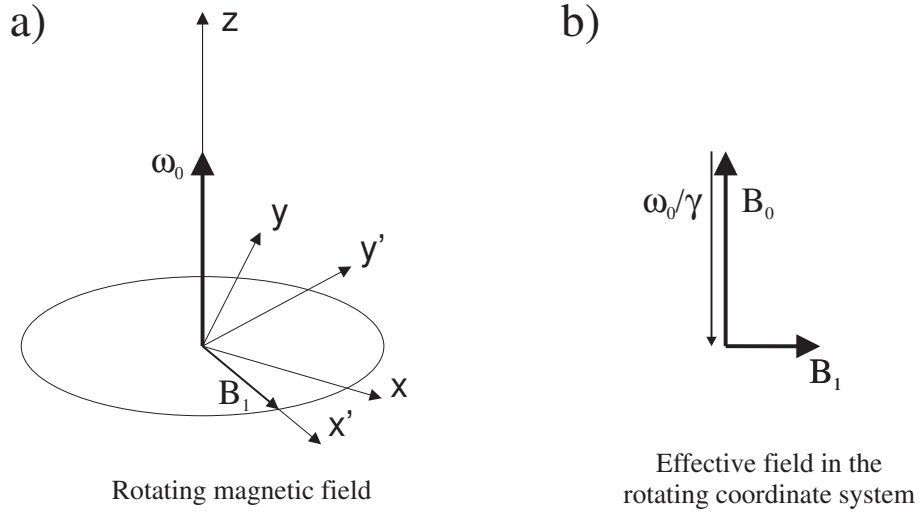


Figure 2.2: Effective magnetic field in a co-rotation coordinate system

If we let

$$\vec{B}_{eff} = \vec{B} + \vec{\Omega}/\gamma \quad (2.19)$$

Eq. 2.18 becomes

$$\left(\frac{d\vec{J}}{dt} \right)_{rot} = \gamma \vec{J} \times \vec{B}_{eff} \quad (2.20)$$

If $\vec{B}_{eff} = 0$, $\vec{J}$ is constant in the rotating system. The condition for this is

$$\vec{\Omega} = -\gamma \vec{B} \quad (2.21)$$

as we have previously found in Eq. 2.14.

2.4 Motion in a Rotating Magnetic Field

2.4.1 Exact Resonance

Consider a moment $\vec{\mu}$ precessing about a static field $\vec{B}_0$, which we take to lie along the z axis. Its motion might be described by

$$\begin{aligned} \mu_x &= \mu \sin \theta \cos \omega_0 t \\ \mu_y &= -\mu \sin \theta \sin \omega_0 t \\ \mu_z &= \mu \cos \theta \end{aligned} \quad (2.22)$$

where ω_0 is the Larmor frequency, and θ is the angle the moment makes with $\vec{B}_0$.

Now suppose we introduce a magnetic field $\vec{B}_1$ which rotates in the x - y plane at the Larmor frequency $\omega_0 = -\gamma B_0$. The magnetic field is

$$\vec{B}(t) = B_1(\hat{x} \cos \omega_0 t - \hat{y} \sin \omega_0 t) + B_0 \hat{z}. \quad (2.23)$$

The problem is to find the motion of $\vec{\mu}$. The solution is simple in a rotating coordinate system. Let system $(\hat{x}', \hat{y}', \hat{z}' = \hat{z})$ precess around the z-axis at rate $-\omega_0$. In this system the field $\vec{B}_1$ is stationary (and $\hat{x}'$ is chosen to lie along $\vec{B}_1$), and we have

$$\begin{aligned}\vec{B}(t)_{eff} &= \vec{B}(t) - \omega_0/\gamma \hat{z} \\ &= B_1 \hat{x}' + (B_0 - \omega_0/\gamma) \hat{z} \\ &= B_1 \hat{x}.\end{aligned}\tag{2.24}$$

The effective field is static and has the value of B. The moment precesses about the field at rate

$$\omega_R = \gamma B_1,\tag{2.25}$$

often called the **Rabi** frequency

This equation contains a lot of history: the RF magnetic resonance community conventionally calls this frequency ω_1 , but the laser resonance community calls it the **Rabi Frequency** ω_R in honor of Rabi's invention of the resonance technique.

If the moment initially lies along the z axis, then its tip traces a circle in the $\hat{y} - \hat{z}$ plane. At time t it has precessed through an angle $\phi = \omega_R t$. The moment's z-component is given by

$$\mu_z(t) = \mu \cos \omega_R t\tag{2.26}$$

At time $T = \pi/\omega_R$, the moment points along the negative z-axis: it has "turned over".

2.4.2 Off-Resonance Behavior

Now suppose that the field B_1 rotates at frequency $\omega \neq \omega_0$. In a coordinate frame rotating with B_1 the effective field is

$$\vec{B}_{eff} = B_1 \hat{x}' + (B_0 - \omega/\gamma) \hat{z}.\tag{2.27}$$

The effective field lies at angle θ with the z-axis, as shown. (Beware: there is a close correspondence between the resonance we are doing here and the dressed atom, but this θ , call it $\theta_{res} = 2\theta_{dressed}$.)

The field is static, and the moment precesses about it at a rate (called the **effective Rabi frequency**)

$$\begin{aligned}\omega_{R'} &= \gamma B_{eff} = \gamma \sqrt{(B_0 - \omega/\gamma)^2 + B_1^2} \\ &= \sqrt{(\omega_0 - \omega)^2 + \omega_R^2}\end{aligned}\tag{2.28}$$

where $\omega_0 = \gamma B_0, \omega_R = \gamma B_1$, as before.

Assume that $\vec{\mu}$ points initially along the +z-axis. Finding $\mu_z(t)$ is a straightforward problem in geometry. The moment precesses about B_{eff} at rate $\omega_{R'}$, sweeping a circle as shown. The radius of the circle is $\mu \sin \theta$, where

$$\sin \theta = B_1 / \sqrt{(B_0 - \omega/\gamma)^2 + B_1^2}\tag{2.29}$$

$$= \omega_R / \sqrt{(\omega - \omega_0)^2 + \omega_R^2}.\tag{2.30}$$

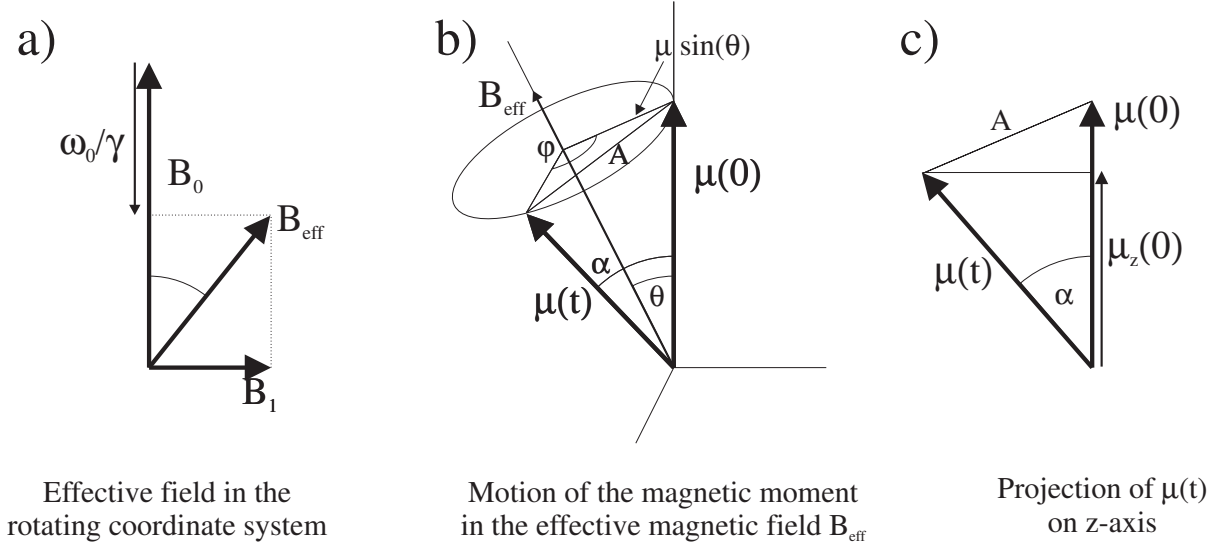


Figure 2.3: Motion of the magnetic moment driven by a rotating field.

In time t the tip sweeps through angle

$$\phi = \omega_R t.$$

The z-component of the moment is

$$\mu_z(t) = \mu \cos \alpha$$

where α is the angle between the moment and the z-axis after it has precessed through angle ϕ . As the drawing shows, $\cos \alpha$ is found from

$$A^2 = 2\mu^2(1 - \cos \alpha).$$

Since

$$A = 2\mu \sin \theta \sin(\omega'_R t/2)$$

we have

$$4\mu^2 \sin^2 \theta \sin^2(\omega'_R t/2) = 2\mu^2(1 - \cos \alpha)$$

and

$$\begin{aligned}
 \mu_z(t) &= \mu \cos \alpha = \mu(1 - 2 \sin^2 \theta \sin^2 1/2 \omega'_R t) \\
 &= \mu \left[1 - 2 \frac{\omega_R^2}{(\omega - \omega_0)^2 + \omega_R^2} \sin^2 \frac{1}{2} \sqrt{(\omega - \omega_0)^2 + \omega_R^2} t \right] \\
 &= \mu \left[1 - 2(\omega_R/\omega'_R)^2 \sin^2(\omega'_R t/2) \right]
 \end{aligned} \tag{2.31}$$

The z-component of $\vec{\mu}$ oscillates in time, but unless $\omega = \omega_0$, the moment never completely inverts. The rate of oscillation depends on the magnitude of the rotating field; the amplitude of oscillation depends on the frequency difference, $\omega - \omega_0$, relative to ω_R . The quantum mechanical result is identical.

2.5 Larmor's Theorem

Treating the effects of a magnetic field on a magnetic moment by transforming to a rotating co-ordinate system is closely related to Larmor's theorem, which asserts that the effect of a magnetic field on a free charge can be eliminated by a suitable rotating co-ordinate transformation.

Consider the motion of a particle of mass m , charge q , under the influence of an applied force $\vec{F}_0$ and the Lorentz force due to a static field $\vec{B}$:

$$\vec{F} = \vec{F}_0 + \frac{q}{c} \vec{v} \times \vec{B} \quad (2.32)$$

Now consider the motion in a rotating coordinate system. By applying Eq. 2.16 twice to $\vec{r}$, we have

$$(\ddot{\vec{r}})_{rot} = (\ddot{\vec{r}})_{inert} - 2\vec{\Omega} \times \vec{v}_{rot} - \vec{\Omega} \times (\vec{\Omega} \times \vec{r}) \quad (2.33)$$

$$\vec{F}_{rot} = \vec{F}_{inert} - 2m(\vec{\Omega} \times \vec{v}_{rot}) - m\vec{\Omega} \times (\vec{\Omega} \times \vec{r}) \quad (2.34)$$

where $\vec{F}_{rot}$ is the apparent force in the rotating system, and $\vec{F}_{inert}$ is the true or inertial force. Substituting Eq. 2.32 gives

$$\vec{F}_{rot} = \vec{F}_0 - \frac{q}{c} \vec{v} \times \vec{B} + 2m\vec{v} \times \vec{\Omega} - m\vec{\Omega} \times (\vec{\Omega} \times \vec{r}) \quad (2.35)$$

If we choose $\vec{\Omega} = -(q/2mc)\hat{z}$, we have

$$\vec{F}_{rot} = \vec{F}_0 - m\Omega^2 B^2 \hat{z} \times (\hat{z} \times \vec{r}) \quad (2.36)$$

where $\vec{B} = \hat{n}B$. The last term is usually small. If we drop it we have

$$\vec{F}_{rot} = \vec{F}_0 \quad (2.37)$$

The effect of the magnetic field is removed by going into a system rotating at the Larmor frequency $qB/2mc$.

Although Larmor's theorem is suggestive of the rotating co-ordinate transformation, Eq. 2.18, it is important to realize that the two transformations, though identical in form, apply to fundamentally different systems. A magnetic moment is not necessarily charged- for example a neutral atom can have a net magnetic moment, and the neutron possesses a magnetic moment in spite of being neutral - and it experiences no net force in a uniform magnetic field. Furthermore, the rotating co-ordinate transformation is exact for a magnetic moment, whereas Larmor's theorem for the motion of a charged particle is only valid when the Ω^2 is neglected.

Table 2.1: Parameters for the magnetic moments of Electrons, Protons, and Nuclei

MASS

electron	$m = 0.91 \times 10^{-27} g$
proton	$M_p = 1.67 \times 10^{-24} g$
neutron	M_p
nuclei	$M = AM_p$
	$A = N + Z = \text{mass number}$
	$Z = \text{atomic number}$
	$N = \text{neutron number}$

CHARGE

electron	-e	$e = 4.8 \times 10^{-10} \text{ esu}$
proton	+e	
neutron	0	
nucleus	Ze	

ANGULAR MOMENTUM

electron	$S = \hbar/2$
proton	$I = \hbar/2$
neutron	$I = \hbar/2$
nuclei	even A: $I/\hbar = 0, 1, 2, \dots$
	odd A: $I/\hbar = 1/2, 3/2, \dots$

STATISTICS

electrons	Fermi-Dirac
nucleons:	odd A, Fermi-Dirac
	even A, Bose-Einstein

ELECTRON MAGNETIC
MOMENT

$$\mu_e = \gamma_e S = -g_s \mu_0 S / \hbar$$

$$\gamma_e = \text{gyromagnetic ratio} = e/mc = 2\pi \times 2.8 \text{ MHz/gauss}$$

$$g_s = \text{free electron g-factor} = 2 \text{ (Dirac Theory)}$$

$$\mu_0 = \text{Bohr magneton} = e\hbar/2mc = 0.9 \times 10^{-20} \text{ (erg/gauss)}$$

(Note that μ_e is negative.

We show this explicitly by taking g_s to be positive, and writing $\mu_e = -g_s \mu_0 S / \hbar$)

NUCLEAR MAGNETIC
MOMENTS

$$\mu_N = \gamma_I I = g_I \mu_N I / \hbar$$

$$\gamma_I = \text{gyromagnetic ratio of the nucleus}$$

$$\mu_N = \text{nuclear magneton} = e\hbar/2Mc = \mu_0(m/M_p)$$

proton	$g_p = 5.6, \gamma_p = 2\pi \times 4.2 \text{ kHz/gauss}$
neutron	$g_N = -3.7$

Chapter 3

Resonance line shapes

In this chapter, we will discuss several phenomena which affect the line shape. Our motivation for this is simple: No resonance line is infinitely narrow. Unless we understand the line shape, we cannot extract spectroscopic information with high accuracy.

3.1 Ideal line shape for Rabi resonance

The Rabi resonance transition probability,

$$P = \frac{\omega_R^2}{(\omega - \omega_0)^2 + \omega_R^2} \sin^2 \frac{\sqrt{(\omega - \omega_0)^2 + \omega_R^2} t}{2} \quad (3.1)$$

has the following properties:

- It is *exact*, it is not a perturbative result.
- P achieves a maximum value of 1 when the resonance condition is satisfied: $\omega = \omega_0$. At resonance P is periodic in the product $\omega_R t$.
- As $|\omega - \omega_0|$ is increased, P oscillates with increasing frequency but decreasing amplitude.

If the interaction time τ is fixed and the power is varied at resonance, then P achieves its maximum value when $\omega_R \tau = \pi$. Under these conditions the spin exactly “flips” under the influence of the applied field. If the frequency ω is then varied, P varies as a function of ω as shown in Fig. 3.1 (curve a). The resonance curve has a full width at half maximum (FWHM) of $\Delta\omega = 0.94\omega_R$, or, in units of Hz,

$$\Delta\nu_{\text{RABI}} = \frac{0.94\omega_R}{2\pi} = \frac{0.47}{\tau} \quad (3.2)$$

3.2 Line shape for an atomic beam

In practice the conditions described by the simple Rabi formula are rarely met exactly. In most cases, 2-level resonance involves averaging over some combination of interaction times, field strengths, and possibly resonance frequency. Fig. 3.1a shows the situation for atomic beam resonance. In this case the experimental resonance curve depends on the distribution of interaction times due to the various speeds of atoms in the thermal atomic beam.

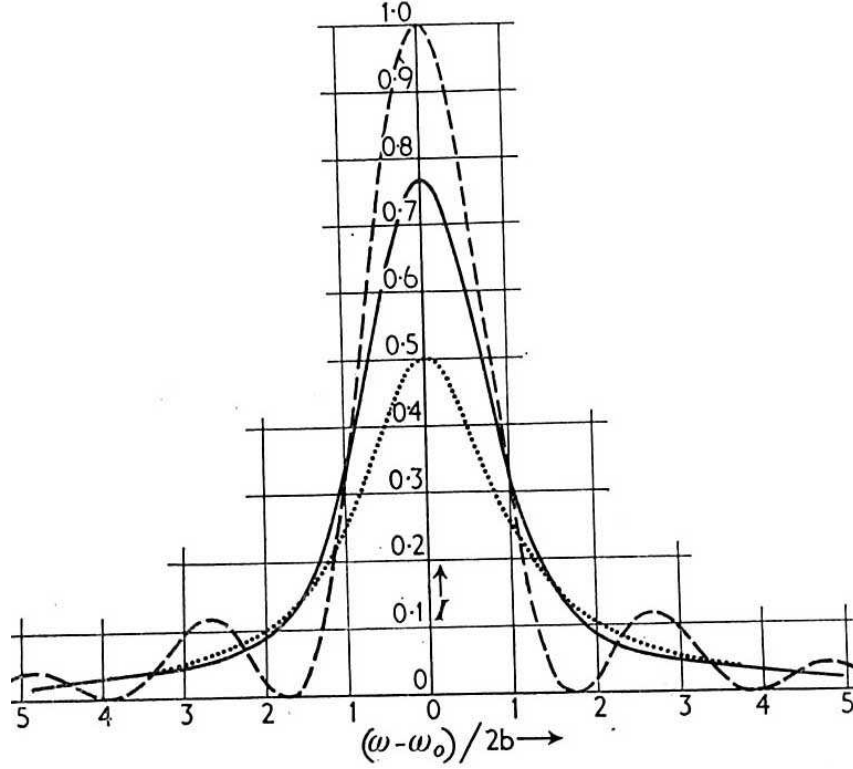


Figure 3.1: Calculated lineshapes: $\langle P \rangle$ plotted as a function of $(\omega - \omega_0)/\omega_R$. Curve a) single velocity: $\omega_R\tau = \pi$. Curve b) thermal atomic beam average; $\omega_R\tau = 1.200\pi$, where $\tau = L/\alpha$. Curve c) heavily saturated resonance, $\omega_R\tau \gg \pi$. From Ramsey, *Molecular Beams*.

In many cases the interaction time τ is not fixed, but is distributed according to a distribution of interaction times $f(t)$, where $\int_0^\infty f(t)dt = 1$. The observed quantity is the average transition probability which is given by

$$\langle P(\Delta\omega) \rangle = \int_0^\infty P(\Delta\omega, t) f(t) dt \quad (3.3)$$

where $\Delta\omega = \omega - \omega_0$.

In an atomic beam, for instance, the interaction time is L/v , where v is the speed. The distribution of speeds in an atomic beam can be found from the Maxwell-Boltzmann law and is

$$g(v) = \frac{2}{\alpha^4} v^3 \exp(-v^2/\alpha^2) \quad (3.4)$$

where $\alpha = \sqrt{2k_B T/M}$ is the most probable velocity of atoms of mass M in a gas at temperature T (cf Ramsey, *Molecular Beams*, p.20). Thus, for a Maxwell-Boltzmann speed distribution the average transition probability is

$$\langle P \rangle_{\text{MB}} = 2 \int_0^\infty \exp(-y^2) y^3 \frac{\omega_R^2}{a^2} \sin\left(\frac{aL}{2\alpha y}\right) dy, \quad (3.5)$$

where $a^2 = \Delta\omega^2 + \omega_R^2$, and $y = v/\alpha$.

Figure 3.2: Elementary atomic beam resonance experiment. O source of atoms; F_1 , F_2 , S collimation slits for atomic beam; A and B Stern-Gerlach magnets for deflecting atoms. These are inhomogeneous magnets which exert a force $\mathbf{F} = \nabla(\vec{\mu} \cdot \mathbf{B}) = \gamma\hbar\nabla(J_z B) = \gamma\hbar m \nabla B$. Magnet A, the “polarizer”, selects a given value of m ; magnet B, the “analyzer”, allows the atoms to pass to the detector D only if m remains unchanged. Resonance is induced in uniform field C by the oscillating field from the coil R . At the resonance a transition $m \rightarrow m'$ occurs; the atom can no longer pass through the analyzer, and the signal to the detector D falls. If the length of the coil is L , then atoms moving with speed of v interact with the oscillating field for time $t = L/v$. (from N.F. Ramsey, *Molecular Beams*.)

The integral must be evaluated numerically (Tables are given in Appendix D of Ramsey.) Results are shown in Fig. 3.1 $\langle P \rangle$ has a maximum for $\omega_R L/\alpha = 1.200\pi$. In contrast, in a monochromatic beam the maximum occurs for $\omega_R t = \omega_R L/v = \pi$. The maximum transition probably is 0.75. The FWHM is

$$\Delta\nu_{MB} = 1.07 \frac{\alpha}{L} \quad (3.6)$$

If we regard L/α as the mean time for an atomic pass through the coil, then compared to the line width for a monoenergetic beam, Eq. 3.2, the effect of the velocity spread is to broaden the resonance curve by approximately two. Furthermore, all traces of periodic behavior have been erased.

3.3 Method of Separated Oscillatory Fields

The separated oscillatory field (SOF) technique is one of the most powerful methods of precision spectroscopy. As the name suggests, it involves the sequential application of the transition-producing field to the system under study with an interval in between. This technique was originally conceived by Norman Ramsey in 1948 for application in RF studies of molecular beams using two separated resonance coils through which the molecular beams passed sequentially. It represents the first deliberate exploitation of a quantum superposition state. Subsequently it has been extended to high frequencies where the RF regions were in the radiation zone (i.e. source-free), to two photon transitions, to rapidly decaying systems, and to experiments where the two regions were temporally (rather than spatially) separated. It is routinely used to push measurements to the highest possible precision (eg. in the Cs beam time standard apparatus). Ramsey shared the 1990 Nobel prize for inventing this method. The following figure shows a typical configuration.

The atomic beam resonance region is composed of two oscillatory field regions, each of length ℓ , separated by distance L . The resonance pattern reveals an interference fringe structure with

Figure 3.3: Beam apparatus for separated oscillatory field experiments to search for a the electric dipole moment of the Neutron. (from N.F. Ramsey, *Molecular Beams*.)

characteristic width $\Delta\nu \approx \alpha / L$, where α is the most probable velocity for a thermal distribution of atoms at temperature T (see the figure below). Of course a single coil of the same length would produce approximately the same resonance width.

The transition probability for the Ramsey method can be calculated by straightforward application of the formalism presented earlier. Details are described in Ramsey's *Molecular Beams*, Section V.4.2. The result is that the transition probability for a two-state system is

$$P = 4 \frac{\omega_R^2}{a^2} \sin^2(a\tau/2) \times \left[\cos(\delta\omega T/2) \cos(a\tau/2) - \left(\frac{\omega - \omega_0}{a} \right) \sin(\delta\omega T/2) \sin(a\tau/2) \right]^2 \quad (3.7)$$

where $a = \sqrt{(\omega - \omega_0)^2 + \omega_R^2}$, and $\delta\omega = \omega - \bar{\omega}_0$, where $\hbar\bar{\omega}_0$ is the average energy separation of the two states along the path between the coil. $\tau = \ell/\alpha$ and $T = L/\alpha$.

The first line in this expression is just 4 times the probability of transition for a spin passing through one of the OFs (oscillating fields). All interference terms (which must involve T) are contained in the second term. The quantity $\delta\omega T$ is the phase difference accumulated by the spin in the field-free region relative to the OF in the first OF region. If the phase in the second OF region differs from the phase in the first OF, the above results must be modified by adding the difference to $\delta\omega T$ in the above equations.

The SOF technique is based on an interference between the excitations produced at two separated fields - thus it is sensitive to the phase difference (coherence) of the oscillating fields. The method is most easily understood by consideration of the classical spin undergoing magnetic resonance in SOF's.

To maximize the interference between the two oscillating fields, we want there to be a probability of 1/2 for a transition in each resonance region. This is achieved by adjusting both field intensities (ω_R above) so that $a\tau = \pi/2$ (an interaction with this property is termed a “ $\pi/2$ pulse” if the system is at resonance, in which case the spin's orientation is now along $\hat{y}$ in the rotating coordinate system).

During the field-free time T , the spin precesses merrily about the constant field B_0 between the two oscillating-field regions. When it encounters the second OF, it receives a second interaction equal to the first. If the system is *exactly* on resonance, this second OF interaction will just complete the inversion of the spin. If, on the other hand, the system is off resonance just enough so that $\delta\omega\tau = \pi$ (but $\delta\omega\tau \ll 1$) then the spin will have precessed about $\hat{z}_0$ an angle π

Figure 3.4: Transition probability by the separated oscillatory field method as a function of frequency. The “Ramsey fringe” is shown by the solid line: it is superimposed on the “Rabi pedestal”, dashed line, which is the resonance curve for a single end coil. (from N.F. Ramsey, *Molecular Beams*.)

less far than the oscillating field. It will consequently lie in the $-\hat{y}'$ direction rather than in the $\hat{y}'$ direction in the coordinate system rotating with the second OF, and as a result the second OF will precess the spin back to $+\hat{z}$, its original direction, and the probability of transition will be 0! A little more thought shows that the transition probability will oscillate sinusoidally with period $\Delta\omega = 2\pi/T$. The central maximum of this interference pattern is centered at ω_0 and its full width at half maximum (in ω -space) is π/T . The central maximum can be made arbitrarily sharp simply by increasing T . In fact, SOF can be used in this fashion to produce line widths for decaying particles which are narrower than the reciprocal of the natural line width! (This does not violate the uncertainty principle because SOF is a way of selecting only those few particles which have lived for time T .)

The separated oscillatory field method (the “Ramsey method”) has the following important properties compared to the single field method (“Rabi method”).

- The resonance frequency depends on the *mean* energy separation of the two states between the coils: instantaneous variations, for instance due to fluctuations in an applied field, either in space or in time, are averaged out.

- The Rabi method requires an applied oscillatory field having uniform phase: this is difficult to accomplish if the flight path is long compared to the wavelength. In contrast, the Ramsey method requires only that the phase be constant across the short coils of length ℓ . Thus, Doppler effects are to a large extent eliminated.
- By the Rabi method the resonance linewidth due to a thermal beam is $1.07 \alpha/\ell$, whereas by the Ramsey method it is only $0.65 \alpha/\ell$. The linewidth is reduced almost by a factor of two.
- In the presence of radiative decay or some other loss mechanism the Ramsey method can be used to selectively observe long-lived atoms: atoms which have decayed are simply absent from the interference pattern. This makes it possible to observe a resonance line which is narrower than the “natural” linewidth, though one must pay a price in reduced intensity.
- Numerous experimental effects can be measured or reduced by modulating the relative phase of the two fields.
- The method is not restricted to atomic beams: the important point is that the applied fields interfere in *time*. The method can be applied to a fixed sample by applying the fields in some desired sequence of pulses.

$$\begin{aligned}
\Omega_{Rabi} \frac{1}{2} \exp(-i(\omega - \omega_0)t) C_2 &= i \frac{dC_1}{dt} \\
\Omega_{Rabi}^* \frac{1}{2} \exp(i(\omega - \omega_0)t) C_1 - i\gamma C_2 &= i \frac{dC_2}{dt}
\end{aligned} \tag{3.8}$$

$$\begin{aligned}
i \frac{dC_1}{dt} &= \Omega_{Rabi} \frac{1}{2} \exp(-i(\omega - \omega_0)t) C_2 \\
i \frac{dC_2}{dt} &= \Omega_{Rabi}^* \frac{1}{2} \exp(i(\omega - \omega_0)t) C_1 - i\gamma C_2
\end{aligned} \tag{3.9}$$

$$\frac{dC_2}{dt} = -i\gamma C_2 \tag{3.10}$$