TAMIONS-2
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Lecture Notes on
Correlations and Fluctuations in Quantum Optics and Open Quantum Systems

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PART A

1 A system in a weak field

$$H = H_0 + V \tag{1}$$

H_0 is the free Hamiltonian of a system (a particle or a many-particle system) and

$$V = -\mathbf{X} \cdot \mathbf{F}(t) \tag{2}$$

is the interaction of the system with the weak field $\mathbf{F}(t)$ where $\mathbf{X}$ is an operator of the system. For example, $\mathbf{X}$ may be an electric dipole or a magnetic dipole operator, and $\mathbf{F}(t)$ may be an electromagnetic field.

1.1 Linear response theory

We work in the Interaction picture.

$$U_0(t) = e^{-iH_0t/\hbar} \tag{3}$$

is the free evolution operator. We define operators in the interaction picture

$$\mathbf{X}(t) = U_0^\dagger(t)\mathbf{X}U_0(t) \tag{4}$$

where $\mathbf{X}$ and $\mathbf{X}(t)$ denote operators in the Schroedinger and interaction picture, respectively. Similarly, the density operator or density matrix $\rho^I(t)$ in the interaction picture is related to the density operator $\rho^S(t)$ in the Schroedinger picture by

$$\rho^I(t) = U_0^\dagger(t)\rho^S(t)U_0(t) \tag{5}$$

Thus one can write the interaction Hamiltonian in the interaction picture by

$$V^I(t) = U_0^\dagger(t)VU_0(t) = -\mathbf{X}(t) \cdot \mathbf{F}(t) \tag{6}$$

Note that the field $F(t)$ is treated classically. The density matrix equation in the interaction picture is given by

$$\frac{d\rho^I(t)}{dt} = -\frac{i}{\hbar}\left[V^I(t), \rho^I(t)\right] \tag{7}$$

which can be expressed in an integral form

$$\rho^I(t) = \rho - \frac{i}{\hbar}\int_{-\infty}^{t}\left[V^I(t'), \rho^I(t')\right] dt' \tag{8}$$

Note that here we have assumed that for time $t < 0$, $V(t) = 0$ and the interaction starts at $t = 0$. Here we have replaced $\rho(-\infty)$ by ρ for simplicity. ρ is the density matrix of an equilibrium or stationary system. The density matrix of a stationary or equilibrium system described by the Hamiltonian H_0 satisfies the equation

$$\dot{\rho} = -\frac{i}{\hbar}[H_0, \rho] = 0 \tag{9}$$

implying that

$$U_0^\dagger(t)\rho U_0(t) = U_0(t)\rho U_0^\dagger(t) = 0 \tag{10}$$

Next, we use first order perturbation theory by replacing $\rho^I(t')$ in the commutator on the RHS of by ρ

$$\rho^I(t) = \rho - \frac{i}{\hbar}\int_{-\infty}^{t}\left[V^I(t'), \rho\right] dt' \tag{11}$$

Let us calculate the expectation value of μ-th component X_μ of the operator $\mathbf{X}$, where μ denotes any component of a coordinate system (e.g., cartesian, spherical polar, etc). It is given by

$$\langle X_\mu(t)\rangle = \langle X_\mu\rangle - \frac{i}{\hbar}\int_{-\infty}^{t}\mathrm{Tr}\left[X_\mu(t)\left[V^I(t'), \rho\right]\right] dt' \tag{12}$$

where $\mathrm{Tr}[....]$ implies trace over the system's eigenstates, that is, the eigenstates of H_0. Here $\langle X_\mu\rangle = \mathrm{Tr}[X_\mu \rho]$. An eigenstate $\mid j\rangle$ is given by $H \mid j\rangle = E_j \mid j\rangle$ where E_j denotes the corresponding eigen energy. All the eigenstates of H_0 forms a complete orthonormal basis by which one can construct the identity operator

$$\mathbf{1} = \sum_n \mid n\rangle\langle n \mid \tag{13}$$

where the sum spans all the eigenstates. The stationarity condition (9) implies that $\rho = \sum_{nm} |n\rangle\langle m|\delta_{nm}$, that is, ρ is a diagonal matrix in terms of the eigenstates of H_0. Substituting

$$V^I(t') = -\mathbf{X}(t')\cdot\mathbf{F}(t') = \sum_\nu X_\nu(t')F_\nu(t') \tag{14}$$

on the RHS of Eq.(12) and setting $\langle X_\mu\rangle = 0$ by the assumption that the operator $\mathbf{X}$ couples only different eignstates, and using the property $\mathrm{Tr}[ABC] = \mathrm{Tr}[BCA] = \mathrm{Tr}[CAB]$, we obtain

$$\langle X_\mu(t)\rangle = \sum_\nu \int_{-\infty}^{t} dt' R_{\mu\nu}(t,t')F_\nu(t') \tag{15}$$

where

$$\boxed{R_{\mu\nu}(t,t') = \frac{i}{\hbar}\langle[X_\mu(t), X_\nu(t')]\rangle} \tag{16}$$

is called linear response function. Equation (15) manifestly fulfills the causality condition.

Exercise-1

Show that $R_{\mu\nu}(t,t')$ is a function of $(t-t')=\tau$ only due to the stationary of ρ.

Stationarity of ρ gives

$$\boxed{R_{\mu\nu}(t,t') = \frac{i}{\hbar}\langle[X_\mu(\tau), X_\nu(0)] = \frac{i}{\hbar}\langle[X_\mu(0)X_\nu(-\tau)]\rangle} \tag{17}$$

where $\tau = t - t' > 0$. This means $R_{\mu\nu}(t,t') = R_{\mu\nu}(\tau)$

1.2 Susceptibility

Consider

$$F_\nu(t) = \frac{1}{2}(F_\nu(\omega)e^{-i\omega t} + \text{C.c.}) \tag{18}$$

that is, the field is harmonically oscillating with angular frequency ω. Here C.c. stands for complex conjugate of the previous quantity. Substituting (18) in (15), we obtain

$$\langle X_\mu(t)\rangle = \frac{1}{2}\sum_\nu \left[\chi_{\mu\nu}(\omega)F_\nu(\omega)e^{-i\omega t} + \text{C.c.}\right] \tag{19}$$

where

$$\boxed{\chi_{\mu\nu}(\omega) = \int_0^\infty R_{\mu\nu}(\tau)e^{i\omega\tau}d\tau} \tag{20}$$

is called susceptibility.

Let us now explicitly calculate $R_{\mu\nu}(\tau)$ as defined by Eq.(17). It is written in terms of the difference between the two temporal correlation functions $\langle X_\mu(\tau)X_\nu(0)\rangle$ and $\langle X_\mu(0)X_\nu(\tau)$. Note that operators at two times do not commute. For simplicity, we calculate the auto-correlation for $X_\mu = X_\nu = X$. We have

$$\begin{aligned}\langle X(\tau)X(0)\rangle &= \text{Tr}\,[X(\tau)X(0)\rho] \\ &= \sum_n \langle n \mid U_0^\dagger(\tau)XU_0(\tau)\sum_k \mid k\rangle\langle k \mid X\rho \mid n\rangle \\ &= \sum_{nk} e^{-i(\omega_k-\omega_n)\tau}X_{nk}X_{kn}\rho_{nn} \end{aligned} \tag{21}$$

In the above, we have used the fact that ρ is a diagonal matrix. Similarly, we get

$$\langle X(0)X(\tau)\rangle = \sum_{nk} e^{i(\omega_k-\omega_n)\tau}X_{nk}X_{kn}\rho_{nn} \tag{22}$$

Since n and k are dummy indices, we can exchange $n \leftrightarrow k$ in the above expression (22) for $\langle X(0)X(\tau)\rangle$. Substituting the above two equations in Eq.(20), we obtain

$$\chi(\omega) = \frac{i}{\hbar}\sum_{nk}\int_0^\infty d\tau e^{i(\omega-\omega_{kn})\tau}|X_{nk}|^2(\rho_{nn} - \rho_{kk})$$

where $\omega_{kn} = \omega_k - \omega_n$. Multiplying $e^{i(\omega-\omega_k n)\tau}$ by a damping factor $e^{-\tau\gamma/2}$ where $\gamma \to 0$, we obtain

$$\boxed{\chi(\omega) = \frac{1}{\hbar}\sum_{nk}\frac{|X_{nk}|^2}{\omega_{kn} - \omega - i\frac{\gamma}{2}}(\rho_{nn} - \rho_{kk})} \tag{23}$$

It has pole at $\omega = \omega_{kn} - i\frac{\gamma}{2}$, that is in the lower half of the complex plane. Note that here we have introduced damping factor in a phenomenological or an ad hoc way. Later we will derive damping of a system by quantum-mechanical methods.

1.3 Dissipation-susceptibility connection

The rate of change of energy of a system in a field is given by $\frac{\partial\langle H\rangle}{\partial t}$ where the Hamiltonian H is given by Eq.(1). The explicit time dependence of H arises due to the field $\mathbf{F}(t)$, a component of which is harmonically oscillating as given by Eq.(18). We get

$$\frac{\partial\langle H\rangle}{\partial t} = -i\frac{1}{2}\sum_{\mu}\left[\omega\langle X_\mu F_\mu(\omega)\rangle e^{-i\omega t} - \text{C.c.}\right] \tag{24}$$

Note that here the quantity $\langle X_\mu\rangle$ is evaluated at time t, that is, after the field has acted upon the system for the duration of t. So, we substitute this quantity by Eq.(19) and obtain

$$\frac{\partial\langle H\rangle}{\partial t} = -i\frac{1}{4}\sum_{\mu\nu}\left[\omega\left\{\chi_{\mu\nu}(\omega)F_\nu(\omega)e^{-i\omega t} + \text{C.c.}\right\}F_\mu(\omega)e^{-i\omega t} - \text{C.c.}\right] \tag{25}$$

We now calculate the time-averaged value of the above quantity by averaging over the time period $T = 2\pi/\omega$ and thus obtain

$$\frac{\partial\langle H\rangle}{\partial t} = -i\frac{1}{4}\sum_{\mu\nu}\left[\omega\chi^*_{\mu\nu}(\omega)F^*_\nu(\omega)F_\mu(\omega) - \text{C.c.}\right] \tag{26}$$

If we assume that the field components F_α $(\alpha = \mu, \nu)$ are either real or all the components have the same phase, the quantity $F^*_\nu(\omega)F_\mu(\omega)$ is real. So we can write the above equation in the form

$$\frac{\partial\langle H\rangle}{\partial t} = i\frac{\omega}{4}\sum_{\mu\nu}\left[\chi_{\mu\nu}(\omega) - \chi^*_{\mu\nu}(\omega)\right])F^*_\mu(\omega)F_\nu(\omega) \tag{27}$$

which clearly implies

$$\frac{\partial\langle H\rangle}{\partial t} = -\frac{\omega}{4}\sum_{\mu\nu}\text{Im}\left[\chi_{\mu\nu}\right]F^*_\mu(\omega)F_\nu(\omega) \tag{28}$$

The above equation tells us that if $\text{Im}\left[\chi_{\mu\nu}\right] > 0$, $\frac{\partial\langle H\rangle}{\partial t}$ is the amount of energy lost per unit time, or energy dissipated per unit time. This is the connection between the dissipation and the imaginary part of the susceptibility. $\text{Im}\left[\chi_{\mu\nu}\right] > 0$ implies absorption of energy (by the system) supplied by the field resulting in the attenuation of the field. On the other hand, $\text{Im}\left[\chi_{\mu\nu}\right] < 0$ means gain of energy (supplied by the system) by the field, or in other words, amplification or lasing of the field. We will show later, after proving fluctuation-dissipation theorem, that such gain or amplification in thermal equilibrium is not possible.

1.4 Spectrum

Spectrum is defined defined as the Fourier transform of two-time correlation function of the operator X of a system. If the system is in a stationary state, then the spectrum is time-independent and given by

$$S(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\omega\tau}\langle X(\tau)X(0)\rangle d\tau \tag{29}$$

which can also be expressed as

$$\begin{aligned} S(\omega) &= \frac{1}{2\pi}\left[\int_{-\infty}^{0} e^{i\omega\tau}\langle X(\tau)X(0)\rangle d\tau + \int_{0}^{\infty} e^{i\omega\tau}\langle X(\tau)X(0)\rangle d\tau\right] \\ &= \frac{1}{2\pi}\left[\int_{0}^{\infty} e^{-i\omega\tau}\langle X(-\tau)X(0)\rangle d\tau + \int_{0}^{\infty} e^{i\omega\tau}\langle X(\tau)X(0)\rangle d\tau\right] \end{aligned} \tag{30}$$

By the property of stationary, $\langle X(-\tau)X(0)\rangle = \langle X(0)X(\tau)\rangle$. So

$$\begin{aligned} S(\omega) &= \frac{1}{2\pi}\left[\int_{0}^{\infty} e^{-i\omega\tau}\langle X(0)X(\tau)\rangle + \int_{0}^{\infty} e^{i\omega\tau}\langle X(\tau)X(0)\rangle\right] \\ &= \frac{1}{\pi}\mathrm{Re}\int_{0}^{\infty} e^{i\omega\tau}\langle X(\tau)X(0)\rangle d\tau \end{aligned} \tag{31}$$

Following the same procedure used in deriving the Eq. (22), we have

$$\begin{aligned} \langle X(\tau)X(0)\rangle) &= \sum_{nk} e^{-i\omega_{kn}\tau}|X_{nk}|^2\rho_{nn} \\ \langle X(0)X(\tau)\rangle &= \sum_{nk} e^{-i\omega_{kn}\tau}|X_{kn}|^2\rho_{kk} \end{aligned} \tag{32}$$

Eq.(29) or Eq.(31) can also be written as

$$S(\omega) = \frac{1}{4\pi}\int_{-\infty}^{\infty} e^{i\omega\tau}\left[\langle X(\tau)X(0)\rangle + X(0)X(\tau)\right] d\tau \tag{33}$$

Substituting (32) in (33), we obtain

$$S(\omega) = \frac{1}{4\pi}\sum_{nk}|X_{nk}|^2\int_{-\infty}^{\infty} e^{i(\omega-\omega_{kn})\tau}d\tau(\rho_{nn}+\rho_{kk})$$

Using $\int_{-\infty}^{\infty} e^{i(\omega-\omega_{kn})\tau}d\tau = \int_{0}^{\infty} e^{i(\omega-\omega_{kn})\tau}d\tau + \int_{0}^{\infty} e^{-i(\omega-\omega_{kn})\tau}d\tau$, and multiplying both integrals by a damping factor $e^{-\frac{\gamma}{2}\tau}$, we obtain

$$\int_{-\infty}^{\infty} e^{i(\omega-\omega_{kn})\tau}d\tau = -i\left[\frac{1}{\omega_{kn}-\omega-\frac{i\gamma}{2}} - \frac{1}{\omega_{kn}-\omega+\frac{i\gamma}{2}}\right]$$

Please note that in the limit $\gamma \to 0$, this integral reduces to $2\pi\delta(\omega-\omega_{kn})$. Thus we get

$$S(\omega) = \frac{1}{4\pi}(-i)\sum_{nk}|X_{nk}|^2\left[\frac{1}{\omega_{kn}-\omega-\frac{i\gamma}{2}} - \frac{1}{\omega_{kn}-\omega+\frac{i\gamma}{2}}\right](\rho_{nn}-\rho_{kk})\frac{(\rho_{nn}+\rho_{kk})}{(\rho_{nn}-\rho_{kk})} \tag{34}$$

suppose, the populations ρ_{nn}, ρ_{kk} are distributed according to thermal distribution, i.e.,

$$\rho_{nn} = \frac{e^{-\frac{\hbar\omega_n}{KT}}}{\mathrm{Tr} e^{-H_0/KT}}. \tag{35}$$

Therefore,

$$\frac{(\rho_{nn} + \rho_{kk})}{(\rho_{nn} - \rho_{kk})} = \frac{e^{\frac{\hbar\omega_{kn}}{KT}} + 1}{e^{\frac{\hbar\omega_{kn}}{KT}} - 1} \tag{36}$$

Since the part within the third bracket of Eq.(34) will contribute significantly when $\omega_{kn} \simeq \omega$ (usually $\gamma << \omega_{kn}$ for atomic transitions), we can approximate

$$\frac{(\rho_{nn} + \rho_{kk})}{(\rho_{nn} - \rho_{kk})} \simeq \frac{e^{\frac{\hbar\omega}{KT}} + 1}{e^{\frac{\hbar\omega}{KT}} - 1} = 1 + 2n_B \tag{37}$$

where

$$n_B = \frac{1}{e^{\hbar\omega/KT} - 1} \tag{38}$$

is the Bosonic occupation number of a quantum state.

1.5 Coherent and incoherent spectrum

The spectrum as defined by Eq.(29) or Eq.(31) has two parts: $S(\omega) = S_c(\omega) + S_{ic}(\omega)$, where $S_c(\omega)$ and $S_{ic}(\omega)$ stand for coherent and incoherent spectrum, respectively. Any operator $X(t)$ can be decomposed into two parts

$$X(t) = \langle X(t)\rangle + \Delta X(t) \tag{39}$$

where $\Delta X(t) = X(t) - \langle X(t)\rangle$ represents the deviation of $X(t)$ from its mean value $\langle X(t)\rangle$ or fluctuation of $X(t)$ around $\langle X(t)\rangle$. Clearly, $\langle \Delta X(t)\rangle = 0$. Consequently, the incoherent part of the spectrum is defined by

$$S_{ic}(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\omega\tau}\langle \Delta X(\tau)\Delta X(0)\rangle d\tau, \tag{40}$$

that is, by the Fourier transform of the two-time correlation function of the fluctuation operator ΔX. The coherent part of the spectrum is defined by

$$\begin{aligned} S_c(\omega) &= \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\omega\tau}\langle X(\tau)\rangle\langle X(0)\rangle d\tau \\ &= \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\omega\tau}\langle X(0)\rangle^2 d\tau = \langle X(0)\rangle^2\delta(\omega). \end{aligned} \tag{41}$$

The second equality in the above equation follows from the stationarity.

1.6 Fluctuation-dissipation theorem

Substituting (37) in (34), we obtain

$$S(\omega) = \frac{1}{4\pi}(1 + 2n_B)(-i)\sum_{nk}|X_{nk}|^2\left[\frac{1}{\omega_{kn} - \omega - \frac{i\gamma}{2}} - \frac{1}{\omega_{kn} - \omega + \frac{i\gamma}{2}}\right](\rho_{nn} - \rho_{kk}) \tag{42}$$

Further, making use of the expression for susceptibility as given in Eq.(23), we obtain

$$S(\omega) \;=\; \frac{\hbar}{4\pi}(1+2n_B)(-i)\left[\chi(\omega)-\chi^*(\omega)\right] = \frac{\hbar}{2\pi}(1+2n_B)\mathrm{Im}[\chi(\omega)] \tag{43}$$

This is fluctuation-dissipation relationship since the $\mathrm{Im}[\chi(\omega)]$ is related to the dissipated power as given by Eq.(28) when $\mathrm{Im}[\chi(\omega)] > 0$

1.7 Negative temperature and its physical meaning

In our derivation of Eq.(42), $\omega_{kn} = (\omega_k - \omega_n) > 0$, that is, the state $\mid k\rangle$ has higher energy than the state $\mid n\rangle$. So, in thermal equilibrium $(\rho_{nn} - \rho_{kk}) > 0$. The quantity $(-i)$ times the bracketed part on the RHS of Eq.(42) is a positive quantity. So, the spectrum as defined by Eq.(35) is always a positive quantity in thermal equilibrium. Therefore, in order for $S(\omega)$ to be positive, $\mathrm{Im}(\chi)$ must be be positive. If $Im(\chi)$ becomes negative, then temperature T has to be negative. Because, the quantity

$$1 + 2n_B = \frac{e^{\hbar\omega/KT}+1}{e^{\hbar\omega/KT}-1} \tag{44}$$

will become negative only when $T < 0$. The physical meaning of negative temperature is that the system is driven far from equilibrium where the upper state $\mid k\rangle$ has higher population or higher probability of occupation than that of the lower state $\mid n\rangle$. In such a far-from-equilibrium situation, one can not really invoke the concept of an equilibrium temperature, since two subsystems consisting of the field and the matter (which are interacting) can have two different temperatures. So, equilibrium thermodynamics is not applicable in this case. Since $\mathrm{Im}(\chi) \propto (\rho_{\mathrm{nn}} - \rho_{\mathrm{kk}})$, $\mathrm{Im}(\chi) < 0$ means $(\rho_{nn} - \rho_{kk}) < 0$ implying population inversion. One example is the laser system which is driven far from equilibrium to attain population inversion.

Exercise-2

Suppose, X is a component of dipole moment operator of a two-level atom. Applying the susceptibility formula (23) to an ensemble of two-level atoms in thermal equilibrium, show that the susceptibility is proportional to $(1 - 2n_F)$, where

$$n_F = \frac{1}{e^{\hbar\omega/KT}+1} \tag{45}$$

is the Fermionic occupation number

Exercise-3

Derive classical expression of electric dipole susceptibility using Lorenz model. State under which conditions this classical expression reduces to the quantum-mechanical expression of electric dipole susceptibility of Eq.(23) (assuming X is the electric dipole operator) at zero temperature

Exercise-4

Following the earlier lecture by Professor Subhasish Dutta Gupta, write the optical Bloch equation (with phenomenologically introduced dissipation and decoherence terms) of a two-level atom interacting with a single-mode laser field, and derive electric polarization density of a medium of two-level atoms in steady-state. From this expression of the polarization density, derive susceptibility in the weak-coupling regime and show that this matches with that of Eq.(23) at zero temperature.

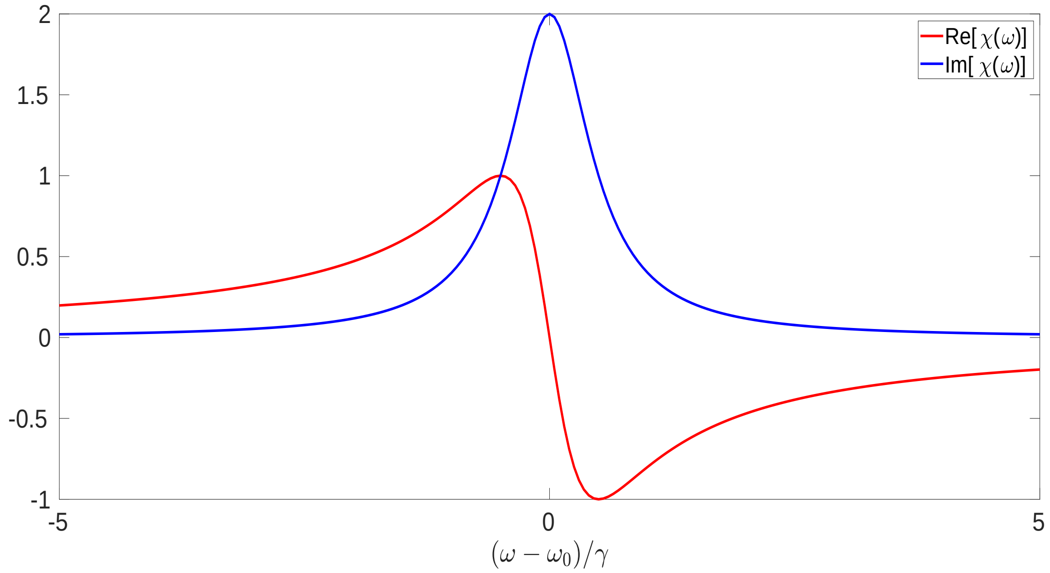

Figure 1: Real and imaginary parts of the susceptibility

1.8 Analyticity of susceptibility: Kramers-Kronig relations

We have seen earlier that the susceptibility $\chi(\omega)$ as defined in Eq. (23) has simple poles only in the lower half of the complex plane and there is no pole in the upper half plane. This means $\chi(\omega)$ is analytic in the upper half plane. So, by Cauchy integral theorem, it can be expressed as

$$\chi(\omega) = \frac{1}{2\pi i}\int d\omega' \frac{\chi(\omega')}{\omega'-\omega} \tag{46}$$

where ω is assumed to be complex with small positive imaginary part. Now, using the relation

$$\lim_{\epsilon\to 0}\frac{1}{\omega'-\omega-i\epsilon} = \mathcal{P}\left[\frac{1}{\omega'-\omega}\right] + i\pi\delta(\omega'-\omega) \tag{47}$$

where $\mathcal{P}$ stands for principal value integral, one finds

$$\chi(\omega) = \frac{1}{\pi i}\int d\omega' \mathcal{P}\left[\frac{\chi(\omega')}{\omega'-\omega}\right] \tag{48}$$

Now, writing $\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$ where $\chi'(\omega)$ and $\chi''(\omega)$ are the real and imaginary parts of $\chi(\omega)$, and equating the real and imaginary parts on the both sides of the above equation, we obtain

$$\chi'(\omega) = \frac{1}{\pi}\int d\omega' \mathcal{P}\left[\frac{\chi''(\omega')}{\omega'-\omega}\right] \tag{49}$$

$$\chi''(\omega) = -\frac{1}{\pi}\int d\omega' \mathcal{P}\left[\frac{\chi'(\omega')}{\omega'-\omega}\right] \tag{50}$$

These are called Kramers-Kronig relations.

Note that the principal value integral of a function like $f(x')/(x'-x)$ is given by

$$\int_{-\infty}^{\infty} dx' f(x')/(x'-x) = \lim_{\epsilon\to 0}\left[\int_{-\infty}^{x-\epsilon} dx' f(x')/(x'-x) + \int_{x+\epsilon}^{\infty} dx' f(x')/(x'-x)\right] \tag{51}$$

The variations of the real and imaginary parts of the susceptibility of a two-level atom as a function of the dimensionless detuning $(\omega-\omega_0)/\gamma$ are shown in Fig.1. Here ω and ω_0

are the laser and atomic frequencies, respectively.

Exercise-5

Show that the two Kramers-Kronig relations can be combined to form one integral of the form

$$\chi(\omega) = \frac{1}{\pi}\int d\omega' \frac{\chi''(\omega')}{\omega' - \omega} \tag{52}$$

where ω is complex with small positive imaginary part.

Exercise-6

Plot the real and imaginary parts of the susceptibility of a two-level atom as functions of ω. By graphical illustrations and numerical integration, verify the Kramers-Kronig relations for the real and imaginary parts of the susceptibility.

Exercise-7

Plot the real and imaginary parts of refractive index as functions of ω and demarcate the absorptive and dispersive regimes

1.9 Fluctuation-diffusion-dissipation connection

Consider a stationary random or fluctuating force $f(t)$ acts on a particle whose position and momentum at time t are $x(t)$ and $p(t)$, respectively. For simplicity we consider the motion of the particle in one dimension. Since a random force is acting on the particle, the variables of the particle such as $x(t)$ and $p(t)$ also become random variables. A random system or processes can only be described by its statistical properties, that is, by ensemble averaging of the physical quantities. We assume that the system in equilibrium is ergodic, meaning that the time-averaged value of an observable in a single realization of one ensemble is equal to its ensemble average. The stochastic equation of motion of the particle obeys the Langevin equation

$$\frac{dp(t)}{dt} = -\bar{\gamma}p(t) + f(t) \tag{53}$$

where $\bar{\gamma}$ is a damping constant. Since $f(t)$ is a fluctuating force, $\langle f(t)\rangle = 0$. Note that here $\langle\cdots\rangle$ implies classical averaging, that is, ensemble or time-averaging in the ergodic sense. Therefore the average value $\langle p(t)\rangle$ of $p(t)$ obeys the equation

$$\frac{d\langle p(t)\rangle}{dt} = -\bar{\gamma}\langle p(t)\rangle \tag{54}$$

The solution of the Eq.(52) can be written as

$$p(t) = e^{-\bar{\gamma}t}p(0) + \int_0^t dt' e^{-\bar{\gamma}(t-t')}f(t') \tag{55}$$

Obviously, $\langle p(t)\rangle = e^{-\bar{\gamma}t}p(0)$.

The equation of motion of the two-time correlation function $\langle p(t)p(t')\rangle$ can be readily written in the form

$$\frac{d}{dt}\langle p(t)p(t')\rangle = -\bar{\gamma}\langle p(t)p(t')\rangle + \int_0^{t'} dt'' e^{-\bar{\gamma}(t'-t'')}\langle f(t)f(t'')\rangle \tag{56}$$

where we have used the fact that $\langle p(0)f(t)\rangle = \langle p(0)\rangle\langle f(t)\rangle = 0$. Due to the stationarity of the force, the correlation function $\langle f(t)f(t'')\rangle$ is a function of $t' - t''$ only. So, one can write

$$\langle f(t)f(t'')\rangle = 2Dg(t' - t'') \tag{57}$$

where $g(t'-t'')$ is a symmetric function of the time difference $t'-t''$ and has the dimension of frequency. The pre-factor D is called diffusion coefficient. Instead of $p(t)$, one can consider the two-time correlation function of any other observable $O(t)$ of the system. The statistical nature of the fluctuating force is basically determined by the function $g(\tau)$. The width τ_c of the function $g(\tau)$ is the correlation time for the two-time correlation function of the force. The damping time of the particle is $1/\bar{\gamma}$ which is typically larger than τ_c by several orders of magnitude. Therefore, usually $g(\tau)$ can be approximated as a delta function $\delta(\tau)$. Stochastic dynamics with fluctuating force (or noise or any random function) having δ-correlation in time domain is called Markovian dynamics. Thus substitution of $\langle f(t)f(t'')\rangle = 2D\delta(t' - t'')$ in Eq.(56) yields

$$\begin{aligned}
\frac{d}{dt}\langle p(t)p(t')\rangle &= -\bar{\gamma}\langle p(t)p(t')\rangle + 2D, \qquad t < t' && (58)\\
&= -\bar{\gamma}\langle p(t)p(t')\rangle, \qquad t \geq t' && (59)
\end{aligned}$$

The conditions on the two time intervals t and t' as mentioned in the above two equations arise due to the existence of the delta function $\delta(t - t'')$ in the integrand with the specified limits of integration.

To know the physical meaning of D, we calculate $\sigma_p^2(t) = \langle (p(t) - \langle p(t)\rangle)^2\rangle$, that is, mean variance of $p(t)$. From Eq.(55), we get

$$\sigma_p^2(t) = \int_0^t dt' \int_0^t e^{-\bar{\gamma}(t-t')}\langle f(t')f(t'')\rangle e^{-\bar{\gamma}(t-t'')} \tag{60}$$

Since $\bar{\gamma} << \tau_c^{-1}$, the two exponentials in the above equation vary slowly in comparison to the time scale on which the correlation $\langle f(t')f(t'')\rangle$ decays. Therefore, again using $\langle f(t')f(t'')\rangle = 2D\delta(t' - t'')$, we obtain

$$\begin{aligned}
\sigma_p^2(t) &= 2D\int_0^t dt' e^{-2\bar{\gamma}(t-t')}\\
&= \frac{D}{\bar{\gamma}}\left(1 - e^{-2\bar{\gamma}t}\right) && (61)
\end{aligned}$$

For $\bar{\gamma}t << 1$, $\sigma_p^2(t) \simeq 2Dt$. For $t \to \infty$, $\sigma_p^2(t) = \langle p^2\rangle = D/\bar{\gamma}$. In the limit $t \to \infty$, the system reaches thermal equilibrium. So, D is equal to the variance (square of deviation or fluctuation) in equilibrium times the damping constant. We can thus replace $\langle p^2\rangle$ by mKT where m is the mass of the particle. Therefore,

$$\boxed{D = \bar{\gamma}mKT} \tag{62}$$

By fluctuation-dissipation theorem as discussed earlier, the rate of energy dissipation is proportional to the imaginary part of the susceptibility which is proportional to $\gamma/2$. Note that here $\bar{\gamma} \equiv \gamma/2$. Since D is proportional to $\bar{\gamma}$, it is also connected to dissipation.

Note that, at zero temperature D vanishes as Eq.(62) shows. We will see later that in quantum theory of fluctuation-dissipation-diffusion, D is finite at zero temperature.

PART B

2 Quantum theory of fluctuations and dissipation

The origin of dissipation of a system is its coupling or interaction with a much larger system which is often called an "environment" or "reservoir" or "bath". A reservoir consists of a large or an infinite number of quantum states that form a "quasi-continuum" or a "continuum" of states. The net effect of the reservoir is to impart "quantum noise" on the system's dynamics when the reservoir's degrees-of-freedom are treated quantum mechanically. Here we specifically consider quantized electromagnetic (EM) fields consisting of an infinite number of modes as a reservoir. A mode of an electromagnetic field is characterized by the polarization, frequency, propagation vector (or wave number). If the reservoir does not contain any photon (a photon is an excitation of a quantized EM field), then the reservoir is said to be in "vacuum" meaning that the fields are in their ground states. Before we present system-reservoir theory in density matrix picture that leads to master equation, we first briefly discuss how the introduction of fluctuating quantum force terms or quantum noise along with the damping terms in the Heisenberg equation of motion of the system's operators can correctly describe the system's quantum dynamics. The resulting Heisenberg equation is called quantum Lengevin equation.

2.1 Quantum Lengevin equation

Here we consider quantum Lengevin equation of lowering (annihilation) and raising (creation) operators a and $a^\dagger$, respectively, of a system interacting with a bath. The x and p operators (or any pair of canonically conjugate hermitian operators) of a system are related to these raising and lowering operators by

$$x(t) = \frac{x_0}{2}\left(ae^{-i\omega_0 t} + a^\dagger e^{i\omega_0 t}\right) \tag{63}$$

$$p(t) = \frac{\hbar}{2ix_0}\left(ae^{-i\omega_0 t} - a^\dagger e^{i\omega_0 t}\right) \tag{64}$$

where $\hbar\omega_0$ is an eigen energy of the system and x_0 is a characteristic length scale of the system. For instance, if the system is a harmonic oscillator of mass m, $x_0 = \sqrt{\hbar/(m\omega_0)}$. Similarly, a single-mode quantized field $F_j(t)$ oscillating with frequency ω_j can be expressed as

$$F_j(t) = \frac{1}{2}\left[A_j b_j e^{-i\omega_j t} + A_j^* b_j^\dagger e^{i\omega_j t}\right] \tag{65}$$

where b_j and $b_j^\dagger$ are the annihilation and creation operator of a quanta of the field. A_j and A_j^* are called amplitudes of the positive-frequency (or absorptive) and negative-frequency (or emissive) components of the field.

The quantum Langevin equation of a lowering operator α of the system is given by

$$\frac{d\alpha}{dt} = (-i\omega_0 - \kappa)\alpha + f(t) \tag{66}$$

where κ is the damping constant and $f(t)$ is quantized noise or force term that contains annihilation operators of the bath or reservoir ($f(t)$ may also contain system operators).

2.2 Quantum Langevin equation for a harmonic oscillator

Consider a harmonic oscillator (or a single-mode cavity field) of frequency ω_0 interacting with a bosonic bath or reservoir. The Hamiltonian is given by

$$H = \left(a^\dagger a + 1/2\right)\hbar\omega_0 + \sum_j \left(b_j^\dagger b_j + 1/2\right)\hbar\omega_j + \sum_j \hbar\left[g_j b_j a^\dagger + g_j^* b_j^\dagger a\right] \tag{67}$$

where $a(a^\dagger)$ is the annihilation(creation) operator of the harmonic oscillator (or cavity field). $b_j(b_j^\dagger)$ represents the annihilation(creation) operator of a quanta of the bath in the mode j. g_j is the coupling constant between the bath and the system. Note that the Hamiltonian is written under rotating-wave approximation (RWA). The Heisenberg equation of motion of the operators are

$$\dot{a} = -i\omega_0 a - i\sum_j g_j b_j \tag{68}$$

$$\dot{b}_j = -i\omega_j b_j - i g_j^* a \tag{69}$$

Equation (69) may be formally integrated to obtain

$$b_j(t) = e^{-\omega_j t} b_j(-\infty) - i g_j^* \int_{-\infty}^{t} e^{-i\omega_j(t-t')} a(t')dt' \tag{70}$$

Substituting this into (68), one obtains

$$\dot{a} = -i\omega_0 a - \sum_j |g_j|^2 \int_{-\infty}^{t} e^{-i\omega_j(t-t')} a(t')dt' + f(t) \tag{71}$$

where

$$f(t) = -i\sum_j g_j e^{-\omega_j t} b_j(-\infty) \tag{72}$$

is quantized fluctuating force term or noise. Since the bath is assumed to be initially in thermal equilibrium, that is, described thermal density matrix, we have $\langle f(t)\rangle = \mathrm{Tr}_r[f(t)] = 0$ where we have done tracing over the initial bath or reservoir states. Since b_j and $b_j^\dagger$ obey bosonic algebra at the same time, Obviously $\langle f^\dagger(t)f(t')\rangle \neq 0$ at finite temperature while $\langle f^(t)f^\dagger(t')\rangle \neq 0$ for any temperature including zero temperature.

Let $\tilde{a} = ae^{i\omega_0 t}$. Then Eq.(71) can be written in the form

$$\dot{\tilde{a}} = -\sum_j |g_j|^2 \int_{-\infty}^{t} e^{-i(\omega_j-\omega_0)(t-t')}\tilde{a}(t')dt' + f(t) \tag{73}$$

To convert this equation into the standard form of quantum langevin equation as given in Eq.(66), we make Markoff approximation:

Only the present (and not the past history) determines the future.

and thereby substitute $\tilde{a}(t')$ in the integral on the RHS by $\tilde{a}(t)$. Substituting t' variable by $\tau = t - t'$ we thus obtain

$$\dot{\tilde{a}} = -\sum_j |g_j|^2 \int_{0}^{\infty} e^{-i(\omega_j-\omega_0)\tau}\tilde{a}(t')dt' + f(t) \tag{74}$$

Introducing small damping factor $e^{-\epsilon\tau}$ and letting $\epsilon \to 0$ after the integration is done, one obtains

$$\dot{\tilde{a}} = -i\omega_{LS} - -\kappa\tilde{a} + f(t) \tag{75}$$

where

$$\omega_{LS} = \int d\omega \mathcal{P} D(\omega)\frac{|g(\omega)|^2}{\omega - \omega_0} \tag{76}$$

is the Lamb shift of the system frequency and

$$\kappa = \pi \int d\omega D(\omega)|g(\omega)|^2 \delta(\omega - \omega_0) \tag{77}$$

is the damping constant. Here introducing the density of bath modes, we have converted the sum over j into an integration over the frequency. Now, Eq.(73) can be rewritten in the form

$$\dot{a} = \left[-i(\omega_0 + \omega_{LS}) - \kappa\right] a + f(t) \tag{78}$$

Usually, ω_{LS} is quite small and ignored. Note that if the system is a two-level atom or any spin, then the noise term $f(t)$ becomes a nonlinear operator.

Using Eq.(72), we calculate the two-time correlation function of the quantum noise term $f(t)$ in the following way.

$$\langle f^\dagger(t) f(t')\rangle = \int_0^\infty d\omega D(\omega)|g(\omega)|^2 e^{i\omega(t-t')}\bar{n}(\omega) \tag{79}$$

$$\langle f(t) f^\dagger(t')\rangle = \int_0^\infty d\omega D(\omega)|g(\omega)|^2 e^{-i\omega(t-t')}(\bar{n}(\omega) + 1) \tag{80}$$

where $\bar{n}(\omega) = \langle b_j^\dagger(-\infty) b_j(-\infty)\rangle$ is the average number of thermal photons in the jth mode of the bath with ω_j being replaced by ω. Usually, $|g(\omega)|^2|$ is a sharply peaked function near the system frequency ω_0. Therefore, the ω-integration may be effectively carried out within a short range around ω_0 by replacing $\omega \to (\omega - \omega_0)$ and letting the lower limit of integration to extend to $-\infty$. We then have

$$\langle f^\dagger(t) f(t')\rangle = 2\kappa\bar{n}(\omega_0)\delta(t - t') \tag{81}$$

$$\langle f(t) f^\dagger(t')\rangle = 2\kappa(\bar{n}(\omega_0) + 1)\delta(t - t') \tag{82}$$

Since f and $f^\dagger$ do not commute, one can define two diffusion constants in quantum theory: $D_N = 2\kappa\bar{n}$ and $D_A = 2\kappa(\bar{n} + 1)$ which are related to normal and anti-normal ordering of noise operators. The above two equations give relationship between quantum diffusion and quantum dissipation. At zero temperature $\bar{n}(\omega_0) = 0$. The important point here is that, unlike that in classical case, quantum diffusion does not vanish at zero temperature.

Exercise-8

Using quantum Langevin equation of Show that the bosonic operators $a(t)$ and $a^\dagger(t)$ at equal time t satisfy the bosonic algebra due to the existence of the quantum noise term in the equation.

2.3 Quantum regression theorem

In the classical theory of Markovian dynamics, the equation of the motion of two-time correlation function of p (or any other variable of the system) in the long time limit is given by Eq.(59) which has the same mathematical form as that of one-time average value of p (or any other variable). Similarly, in the quantum theory of Markovian dynamics, the equations of motion of the two-time correlation function $\langle\alpha^\dagger(t)\alpha(t')\rangle$ and $\langle\alpha(t)\alpha^\dagger(t')\rangle$ the operators α and $\alpha^\dagger$ of the system have the same form as that of the one-time average of the operators as $t \to \infty$. Since $\langle f(t)\rangle = 0$, Eq.(66) gives

$$\frac{d\langle\alpha\rangle}{dt} = (-i\omega_0 - \kappa)\langle\alpha\rangle \tag{83}$$

Quantum regression theorem dictates that

$$\frac{d}{dt}\langle\alpha^\dagger(t)\alpha(t')\rangle = (-i\omega_0 - \kappa)\langle\alpha^\dagger(t)\alpha(t')\rangle \tag{84}$$

$$\frac{d}{dt}\langle\alpha(t)\alpha^\dagger(t')\rangle = (-i\omega_0 - \kappa)\langle\alpha(t)\alpha^\dagger(t')\rangle \tag{85}$$

for $t > t'$. Quantum regression theorem is extensively used to calculate two-time correlation functions and spectrum.

2.4 Master equation

We begin with the Hamiltonian

$$H = H_S + H_R + H_{SR} \tag{86}$$

where H_S and H_R are the Hamiltonian for S (system)) and R(reservoir), respectively. H_{SR} is the system-reservoir interaction Hamiltonian. By taking partial trace on the total density matrix ρ on wither of the S or R, we obtain the reduced density matrices

$$\begin{aligned}\mathrm{Tr_R}\rho(\mathrm{t}) &= \rho_S(t)\\ \mathrm{Tr_S}\rho(\mathrm{t}) &= \rho_R(t)\end{aligned}$$

of S and R, respectively. We write the interaction-picture Hamiltonian and density matrix

$$\begin{aligned}H^{\mathrm{I}}_{SR}(t) &= e^{(i/\hbar)(H_R+H_S)t}H_{SR}e^{(-i/\hbar)(H_R+H_S)t}\\ \rho^{\mathrm{I}}(t) &= e^{(i/\hbar)(H_R+H_S)t}\rho(t)e^{(-i/\hbar)(H_R+H_S)t}\end{aligned}$$

So, in the interaction picture, the equation for ρ reads

$$\frac{\partial\rho^{\mathrm{I}}}{\partial t} = -\frac{i}{\hbar}[H^I_{SR}(t), \rho^I(t)] \tag{87}$$

Now, we rewrite Eq.(84) in the integral form

$$\rho^{\mathrm{I}}(t) = \rho(-\infty) - \frac{i}{\hbar}\int_{-\infty}^{t} dt'[H^I_{SR}(t'), \rho^I(t')] \tag{88}$$

$$\frac{\partial\rho^{\mathrm{I}}}{\partial t} = -\frac{i}{\hbar}[H^{\mathrm{I}}_{SR}(t), \rho(-\infty)] - \frac{1}{\hbar^2}\int_{-\infty}^{t} dt'[H_{SR}(t), [H^I_{SR}(t'), \rho^I(t')]] \tag{89}$$

Tracing over the reservoir in Eq.(86), one gets

$$\frac{\partial\rho^{\mathrm{I}}}{\partial t} = -\frac{1}{\hbar^2}\int_{-\infty}^{t} dt' tr_R[H_{SR}(t), [H^I_{SR}(t'), \rho^I(t')]] \tag{90}$$

where, for simplicity, we have eliminated the term $-\frac{i}{\hbar}[H^{\rm I}_{SR}(t), \rho(-\infty)]$ with the assumption $\text{Tr}_R[H_{SR}(t)\rho_R(0)] = 0$

We make Born approximation

$$\rho(t) \simeq \rho_S(t)\rho_R(-\infty)$$

that is, the system and the reservoir density matrices are separable (not entangled) and only the system's density matrix evolves while the reservoir's density matrix always remains in initial or equilibrium condition. Using this approximation, we get

$$\frac{\partial \rho^{\rm I}}{\partial t} = -\frac{1}{\hbar^2}\int_{-\infty}^{t} dt' tr_R[H_{SR}(t), [H^I_{SR}(t'), \rho^{\rm I}_S(t')\rho_R(0)]] \tag{91}$$

We next make Markoff approximation by replacing $\rho^{\rm I}_S(t')$ by $\rho^{\rm I}_S(t)$ and obtain the master equation

$$\frac{\partial \rho^{\rm I}}{\partial t} = -\frac{1}{\hbar^2}\int_{-\infty}^{t} dt' tr_R[H_{SR}(t), [H^I_{SR}(t'), \rho^{\rm I}_S(t)\rho_R(0)] \tag{92}$$

We now explicitly write the Hamiltonians

$$\begin{aligned} H_S &= \hbar\omega_0 \alpha^\dagger \alpha & (93)\\ H_R &= \sum_j \left(b_j^\dagger b_j + 1/2\right)\hbar\omega_j & (94)\\ H_{SR} &= \sum_j \hbar\left[g_j b_j \alpha^\dagger + g_j^* b_j^\dagger \alpha\right] & (95) \end{aligned}$$

where α ($\alpha^\dagger$) denotes the system's lowering (raising) operator. Here the system may be a bosonic or a fermionic particle or a spin. If it is a two-level atom, then $\alpha = S^- = \sigma^-$ is the Pauli lowering operator (or lowering operator of a spin-half particle) and ω_0 is the atomic transition frequency. The system-reservoir interaction Hamiltonian H_{SR} is written under RWA. But the reservoir or bath is modelled as a field of infinite number of bosonic modes, with jth mode being described by b_j and $b_j^\dagger$ operators. Having done the time-integration following the same procedure as in deriving quantum Langevin equation, the master equation of (92) under Born-Markoff approximation reduces to the standard Lindblad form

$$\frac{\partial \rho^{\rm I}}{\partial t} = -\frac{i}{\hbar}[H_S, \rho_S] + \mathscr{L}_{\rm diss}\rho_S \tag{96}$$

where $\mathscr{L}_{\rm diss}$ is a super-operator defined by

$$\begin{aligned} \mathscr{L}_{\rm diss}\rho_S &= -\kappa\left(\bar{n}(\omega_0) + 1\right)\left\{\alpha^\dagger\alpha, \rho_S\right\} + 2\kappa\alpha\rho_S\alpha^\dagger \\ &- \kappa\bar{n}(\omega_0))\left\{\alpha\alpha^\dagger, \rho_S\right\} + 2\kappa\alpha^\dagger\rho_S\alpha & (97) \end{aligned}$$

where $\{A, B\} = AB + BA$ is the anticommutator between A and B. The damping constant κ is defined earlier by Eq. (77). Note that at zero temperature, $\bar{n}(\omega) = 0$. For a two-level atom at zero temperature, κ is the half-width of the spontaneous emission line or half of Einstein's A coefficient. In the beginning while discussing linear response theory, we introduced phenomenological decay constant $\gamma/2$ which is the same as κ, so γ corresponds to full width.

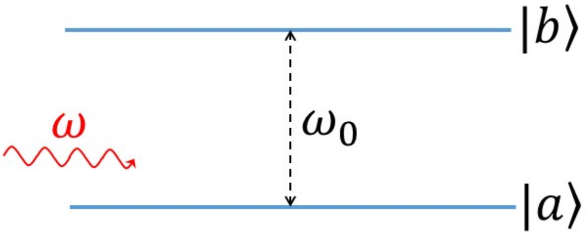

Figure 2: Two-level atom

2.5 Two-level atoms in an EM field: Dressed states

As shown in Fig.2, let $\mid a\rangle$ and $\mid b\rangle$ be the lower and upper states of a two-level atom (TLA), respectively; with E_a and E_b being the corresponding energies. Let us consider the interaction of the TLA with a single-mode classical laser field given by

$$\mathbf{E}(t) = \frac{1}{2}\left[\mathbf{E}_\omega e^{-i\omega t} + \mathbf{E}^*_\omega e^{-i\omega t}\right] \tag{98}$$

Under dipole and rotating-wave approximations, the semiclassical Hamiltonian of the (TLA) interacting with this single-mode laser field is given by

$$H = -\hbar\delta \mid b\rangle\langle b \mid + \frac{1}{2}\hbar\left(\Omega S^+ + \Omega^* S^-\right) \tag{99}$$

where $\delta = \omega - \omega_0$, $S^+ = \mid b\rangle\langle a \mid$ is the raising operator and $S^- = \mid a\rangle\langle b \mid$ is the lowering operator. To relate this model with a spin-half system, we can identify $S_z = \sigma_z/2 = (1/2)(\mid b\rangle\langle b \mid - \mid a\rangle\langle a \mid)$. Here ω and $\omega_0 = (E_b - E_a)/\hbar$ are the laser and atomic transition frequencies, respectively. Note that we have written this Hamiltonian in a frame rotating with ω. To write the Hamiltonian in this particular form, we have set the zero of the energy scale as $E_a = 0$ and $E_b = \hbar\omega_0$. Here Ω denotes the Rabi frequency defined by

$$\Omega = -\frac{1}{\hbar}\mathbf{d}_{ab} \cdot \mathbf{E}_\omega \tag{100}$$

is called Rabi frequency, where $\mathbf{d}$ and $\mathbf{E}_\omega$ are the transition dipole moment of the atom and the electric field of the laser, respectively; and $\langle\cdots\rangle$ implies averaging over the atomic states. One can easily diagonalize this Hamiltonian and obtain the dressed energies

$$E_\pm = -\hbar\frac{\delta}{2} \pm \frac{1}{2}\hbar\sqrt{\delta^2 + \Omega^2} \tag{101}$$

When $\Omega = 0$, they reduce to the bare energies $E_-^{\text{bare}} = -\delta$ and $E_+^{\text{bare}} = 0$. In the laboratory frame, they are $E_-^{\text{bare}} = \hbar\omega_0$ and $E_+^{\text{bare}} = \hbar\omega$.

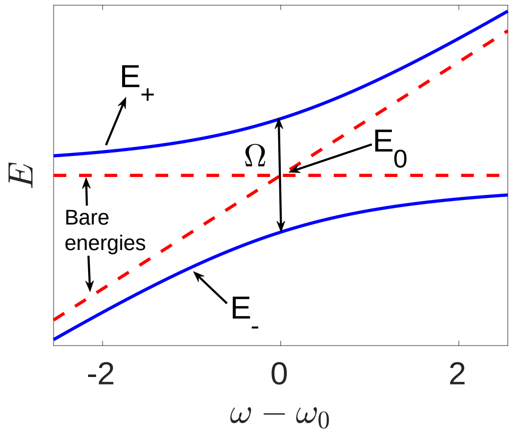

Figure 3: Dressed and bare energies. The horizontal dashed red line corresponds to the energy $E_0 = \hbar\omega_0$ of the upper bare state, and the other dashed red line indicates the energy of the lower bare state. The bare state energies cross at resonance, while the dressed state energies exhibit anti-crossing due to atom-field coupling.

The dressed and bare energies as a function of δ are shown in Fig.3.

The two eigenstates or dressed states are given by $\begin{bmatrix} |+\rangle \\ |-\rangle \end{bmatrix} = \begin{bmatrix} \cos\theta & \sin\theta \\ -\sin\theta & \cos\theta \end{bmatrix} \begin{bmatrix} |a\rangle \\ |b\rangle \end{bmatrix}$ that is,

$$
\begin{aligned}
|+\rangle &= \cos\theta|a\rangle + \sin\theta|b\rangle &(102)\\
|-\rangle &= -\sin\theta|a\rangle + \cos\theta|b\rangle &(103)
\end{aligned}
$$

where $\tan 2\theta = -\Omega/\delta$

2.6 Jaynes-Cummings model

Next, we consider the interaction of a TLA with a single-mode quantized EM field given by

$$\mathbf{E} = \frac{1}{2}\left[\mathbf{E}_1(\omega)ae^{-i\omega t} + \mathbf{E}_1^*(\omega)e^{i\omega t}\right] \tag{104}$$

where $\mathbf{E}_1(\omega) = \sqrt{\frac{2\hbar\omega}{\epsilon_0 V}}\hat{\epsilon}u(\mathbf{r})$ is the field associated with the annihilation of a photon described by the annihilation operator a, V is the quantization volume, $\hat{\epsilon}$ is the unit polarization vector, $u(\mathbf{r})$ is the mode function, for a plane wave (with periodic boundary condition) $u(\mathbf{r}) = \exp[\mathbf{k}.\mathbf{r}]$ with $\mathbf{k}$ being the propagation vector. The field $\mathbf{E}$ is derived from the vector potential $\mathbf{A}(\mathbf{r},t)$ by $\mathbf{E} = -\frac{\partial\mathbf{A}}{\partial t}$. In SI unit, the vector potential has the form

$$\mathbf{A} = \frac{1}{2}\left[-i\sqrt{\frac{2\hbar}{\epsilon_0\omega V}}u(\mathbf{r}\hat{\epsilon}ae^{-i\omega t} + \text{H.c}\right] \tag{105}$$

Note that for this choice of $\mathbf{A}$, $\mathbf{E}_1$ becomes real if $\hat{\epsilon}u(\mathbf{r})$ is real. The form of $\mathbf{A}$ without '$-i$' in front of $\sqrt{\frac{2\hbar}{\epsilon_0\omega V}}ae^{-i\omega t}$ also appears in the literature, in that case $\mathbf{E}_1$ or $\mathbf{E}_\omega$ (in case of classical field) becomes purely imaginary if $\hat{\epsilon}u(\mathbf{r})$ is real. The bottom line here is that whatever form one chooses for the vector potential, $\mathbf{A}$ and $\mathbf{E}_1$ or $\mathbf{E}_\omega$ differs by a phase difference of $\pi/2$.

The Hamiltonian is given by

$$H = \hbar\omega_0 S^z + \hbar\omega a^\dagger a + \frac{1}{2}\left[\Omega_1 S^+ a + H.c\right] \tag{106}$$

where

$$\Omega_1 = -\frac{1}{\hbar}\mathbf{d}_{ab}\cdot\mathbf{E}_1 \tag{107}$$

is the Rabi frequency associated with one photon annihilation. In writing the Hamiltonian in this particular form, the zero of the energy scale is set by $E_a + E_b = 0$. The Hamiltonian can be decomposed into two parts: $H = H_1 + H_2$ where

$$H_1 = \hbar\omega a^\dagger a + \hbar\omega S^z \tag{108}$$

$$H_2 = -\hbar\delta S^z + \frac{1}{2}\left[\Omega_1 S^+ a + \text{H.c}\right] \tag{109}$$

Using the commutation relations $[S^z, S^\pm] = \pm S^\pm$ and $[a^\dagger a, a^\pm] = \pm a^\pm$ where $a^+ = a^\dagger$ and $a^- = a$, one can easily verify that $[H_1, H_2] = 0$.

Suppose, when the atom is in the lower state $\mid a\rangle$, the field is in the Fock state $\mid n+1\rangle$, that is, the field has $(n+1)$ photons. Because of RWA used in writing the Hamiltonian (109), the filed will be in $\mid n\rangle$ when the atom is in the upper state $\mid b\rangle$. So, one can rewrite the Hamiltonian in these two joint atom-field basis functions $\mid a, n+1\rangle$ and $\mid b, n\rangle$; and diagonalize the resulting Hamiltonian. Thus one obtains the two dressed energies

$$\begin{aligned} E_{n-} &= \langle a, n+1 \mid H_1 \mid a, n+1\rangle - \frac{1}{2}\hbar\sqrt{\delta^2 + \Omega_1^2(n+1)} \\ &= (n+1/2)\hbar\omega - \frac{1}{2}\hbar\sqrt{\delta^2 + \Omega_1^2(n+1)} \qquad (110) \\ E_{n+} &= \langle b, n \mid H_1 \mid b, n\rangle + \frac{1}{2}\hbar\sqrt{\delta^2 + \Omega_1^2(n+1)} \\ &= (n+1/2)\hbar\omega + \frac{1}{2}\hbar\sqrt{\delta^2 + \Omega_1^2(n+1)} \qquad (111) \end{aligned}$$

Corresponding eigenstates are given by

$$\mid n-\rangle = \cos\theta_n \mid a, n+1\rangle + \sin\theta_n \mid b, n\rangle \tag{112}$$

$$\mid n+\rangle = \cos\theta_n \mid b, n\rangle - \sin\theta_n \mid a, n+1\rangle \tag{113}$$

where

$$\tan 2\theta_n = -\frac{\Omega_1\sqrt{n+1}}{\delta} \tag{114}$$

The important point here is that when $n = 0$, that is, when the field is in vacuum, the two eigenvalues are separated by $\hbar\sqrt{\delta^2 + \Omega_1^2}$ which is called vacuum field Rabi splitting. Note that the these full quantum mechanical dressed energies and dressed states will reduce to those of semi-classical counterparts when we replace $(n+1)$ by an average photon number $\langle n\rangle$ and identify $\Omega_1\langle n\rangle$ with Ω

Exercise-9

Calculate the time-dependent probabilities P_{an+1} and P_{bn} to find the system in states $\mid a, n+1\rangle$ and $\mid a, n+1\rangle$, respectively. Plot these probabilities as a function of $\Omega_1 t$ for $n = 0$, $n = 1$ and $n = 10$, assuming that the system is initially in state $\mid a, n+1\rangle$.

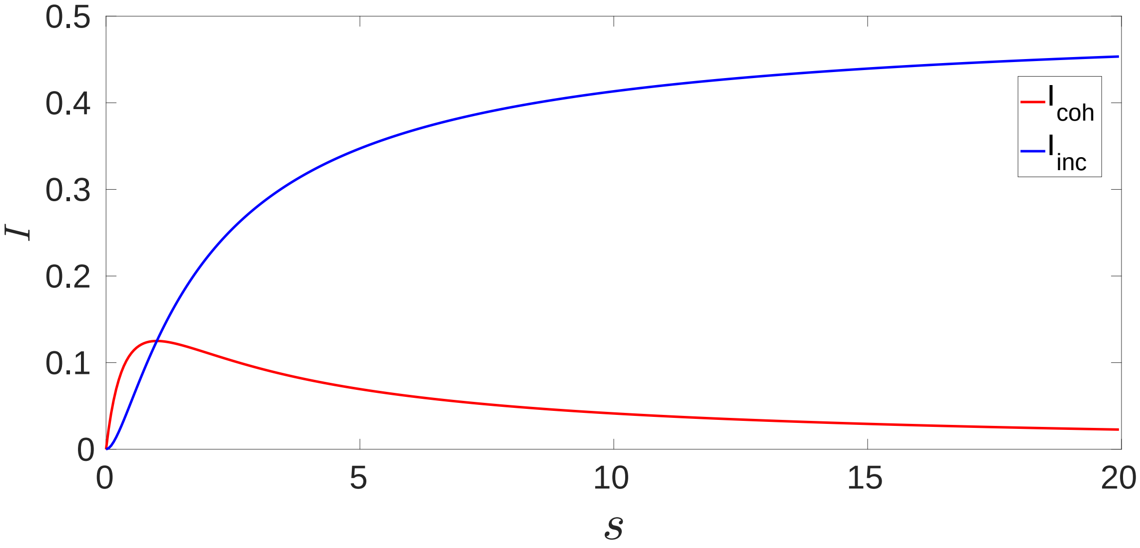

Figure 4: Coherent and incoherent intensity (for $\eta = 1$) as a function of saturation parameter s.

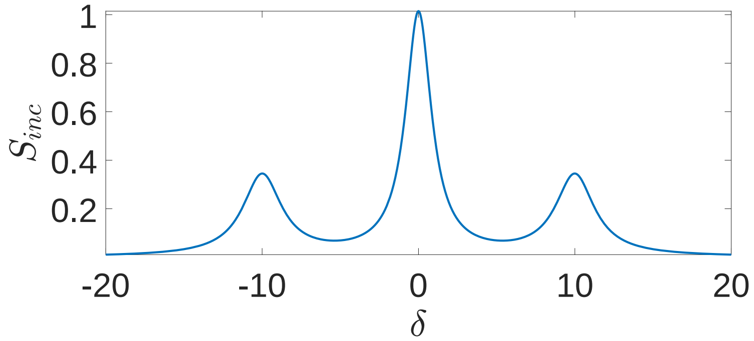

Figure 5: Resonance fluorescence spectrum for $\Omega = 5\gamma$. Note that the height of the side peaks is one third that of the central peak. Here the x-axis variable is $\delta = \omega - \omega_l$

2.7 Resonance fluorescence: Coherent and incoherent spectrum

Resonance fluorescence is the spectrum of light emitted by two-level atoms dressed by a laser field of frequency ω_l tuned on resonance to the atomic transition. In Sections 1.4 and 1.5, we discussed how spectrum is related to the Fourier transform of two-time correlation functions. Suppose, a photon is created by a dressed atom in mode i which was initially in vacuum $\mid 0\rangle$. The field correlation function is then given by

$$C(t,t') \propto \langle 0 \mid a_i(t) a_i^\dagger(t') \mid 0\rangle \tag{115}$$

The creation and annihilation operators $a_i^\dagger(t')$ and $a_i(t)$ of the generated field are obviously proportional to the atomic operators $S^-(t')$ and $S^+(t)$, respectively. So, the resonance fluorescence spectrum is given by

$$S(\omega) = \eta \frac{1}{2\pi} \int_{-\infty}^{\infty} \langle S^+(t) S^-(t-\tau)\rangle e^{i(\omega-\omega_l)\tau} d\tau \tag{116}$$

where η is a proportionality constant and $\tau = t - t'$. If the spectrum is obtained in steady-state at $t \to \infty$, then the spectrum is time-independent due to the stationarity.

To calculate the spectrum, one has to use optical Bloch equation and quantum regression theorem. The optical Bloch vector $\mathbf{X}$ can be defined in two equivalent ways

$$\mathbf{X} = \begin{bmatrix} \langle S^{+} \rangle \\ \langle S^{-} \rangle \\ \langle S^{Z} \rangle \end{bmatrix} \tag{117}$$

Defining $u = \mathrm{Re}[\langle S^{+} \rangle]$, $v = \mathrm{Im}[\langle S^{+} \rangle]$ and $w = \langle S^{Z} \rangle$, one can also define an alternative form of Bloch vector $\tilde{\mathbf{X}}$ as

$$\tilde{\mathbf{X}} = \begin{bmatrix} u \\ v \\ w \end{bmatrix} \tag{118}$$

u and v are related to $S^{\pm}$ by $S^{\pm} = u \pm iv$.

The master equation for a TLA interacting with a laser field is given by

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar}\left[H, \rho\right] + \mathscr{L}_{\mathrm{diss}} \tag{119}$$

where H is given by Eq.(99) in the semiclassical picture, and the superoperator $\mathscr{L}_{\mathrm{diss}}$ has the same form as that of Eq.(97) with α ($\alpha^{\dagger}$) being replaced by $S^{-}(S^{+})$ and $\bar{n} = 0$. The equation of motion of the Bloch vector can be derived from the master equation by using the relation $\langle \frac{\partial S^{i}}{\partial t} \rangle = \mathrm{Tr}\left[S^{i} \frac{\partial \rho}{\partial t}\right]$, where the superscript i stands for $+$ or $-$ or Z. The equation of motion of $\mathbf{X}$ can be cast into the form

$$\frac{\partial X}{\partial t} = \mathbf{M}\mathbf{X} - \frac{\gamma}{2}\begin{bmatrix} 0 \\ 0 \\ 1 \end{bmatrix} \tag{120}$$

where $\mathbf{M}$ is a 3×3 matrix. In the steady-state, the elements of $\tilde{\mathbf{X}}$ are given by

$$u = \frac{1}{\Omega}\frac{s\delta}{s+1} \tag{121}$$

$$v = \frac{1}{\Omega}\frac{s(\gamma/2)}{s+1} \tag{122}$$

$$w = -\frac{1}{2}\left[1 - \frac{s}{s+1}\right] \tag{123}$$

where

$$s = \frac{\Omega^{2}/2}{\delta^{2} + (\gamma/2)^{2}} \tag{124}$$

is the saturation parameter which is a measure of the effective atom-field coupling. $s << 1$ means weak-coupling regime while $s >> 1$ implies strong-coupling regime.

The coherent part of the spectrum can be readily calculated as

$$\begin{aligned} S_{\mathrm{coh}}(\omega) &= \eta \frac{1}{2\pi} \int_{-\infty}^{\infty} \langle S^{+}(t \to \infty) \rangle \langle S^{-}(t \to \infty) \rangle e^{i(\omega - \omega_l)} d\tau \\ &= \eta \mid \langle S^{+}(t \to \infty) \rangle \mid^{2} \delta(\omega - \omega_l) \end{aligned} \tag{125}$$

$\langle S^{+}(t \to \infty) \rangle$ is the steady-state value of $\langle S^{+} \rangle$ which is $u + iv$. When the coherent spectral function is integrated over the entire range of frequencies, one obtains the total intensity of coherent radiation as

$$I_{\mathrm{coh}} = \eta(u^{2} + v^{2}) = \eta \frac{1}{2}\frac{s}{(s+1)^{2}} \tag{126}$$

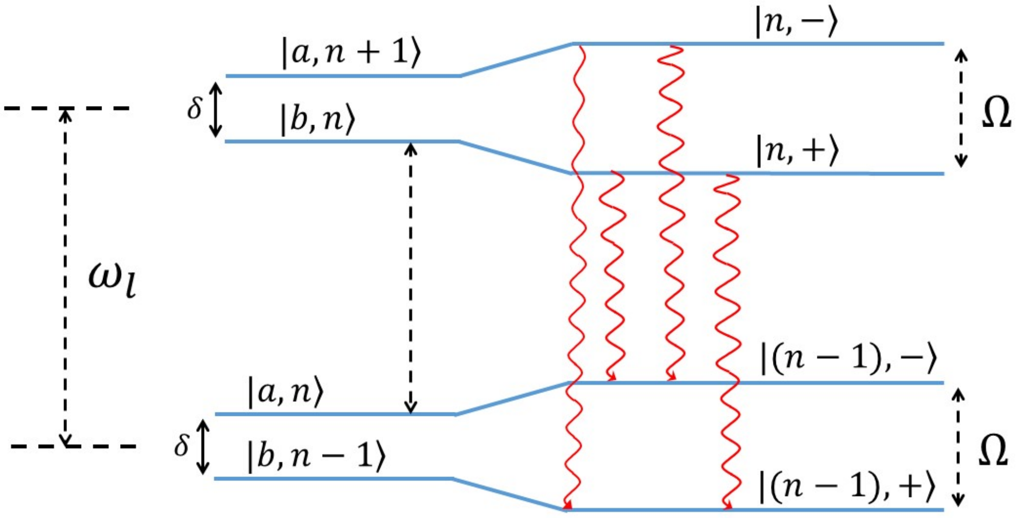

Figure 6: Transitions between dressed levels leading to resonance fluorescence

The total spectral intensity is given by

$$\begin{aligned} I &= \eta\int d\omega S(\omega) = \eta\langle S^{+}(t\to\infty)S^{-}(t\to\infty)\rangle \\ &= \eta\rho_{bb}(t\to\infty) = \eta\frac{1}{2}\frac{s}{s+1} \end{aligned} \tag{127}$$

So, the total incoherent intensity is $I_{\text{incoh}} = I - I_{\text{coh}}$. The variations of I_{coh} and I_{incoh} as functions of s are shown in Fig.4.

Although it is straightforward to calculate the coherent spectrum, calculating incoherent spectrum is tricky. One has to first write down the equation of motion of the two-time correlation function of the operators around steady-state. According to quantum regression theorem, the equations of motion of two-time correlation functions will be governed by the same mathematical form as that of one-time averages of the operators. So, the two-time correlation functions of the atomic operators will satisfy the same type of equations as in Eq. (120). In other words, the temporal evolution of the two-time correlation functions will be determined by the eigenvalues and eigenfunctions of the M matrix. However, one has to identify the time-independent part of the the two-time correlation functions, this part must be subtracted from the correlation function to obtain incoherent spectrum.

The matrix M has three eigenvalues which are $\delta + i\gamma/2$, $\delta \pm \Omega + i3\gamma/4$. Accordingly, in the strong-coupling regime, the incoherent spectrum as shown in Fig.5 has three-peak structure which is called Molllow triplet. The height of the two side peaks symmetrically shifted by $\pm\Omega$ from the central peak is one third the height of the central peak. The width of the side-peak spectral structure is 1.5 times larger than that of the central peak. All these features result from the transitions between the atom-photon dressed states as schematically shown in Fig.6.

3 Concluding remarks and further studies

We have so far discussed correlation functions of the product two operators at two different times. Higher order correlations involving product of multiple operators at different space-time points provide interesting and important information about the properties and dynamics of open quantum systems. For example, density-density correlations involve four operators at two different space-time points. In quantum theory of optical coherence and quantum statistical properties of radiation, higher order correlations play an important role. One of the important higher order correlations widely used in quantum optics is Hanbury Brown-Twiss (HBT) correlation and the physical effects associated with this correlation are called HBT effects. A brief description of HBT correlations and effects are discussed in a separate supplementary note.

Density matrix formalism is quite general and applicable to single- or multi-particle open quantum systems. It can be expressed in a compact Liovillian form

$$\frac{\partial \rho}{\partial t} = \mathscr{L} \rho \tag{128}$$

where the Liouvillian operator $\mathscr{L}$ has two parts: $\mathscr{L} = \mathscr{L}_{\text{coh}} + \mathscr{L}_{\text{diss}}$, the coherent part $\mathscr{L}_{\text{coh}}$ is given by

$$\mathscr{L}_{\text{coh}} = -\frac{i}{\hbar}\left[H, \rho\right] \tag{129}$$

where the Hamiltonian H may describe a driven one- or few- or many-body quantum system. The driving fields may be described classically. The dissipative part of the Liovillian operator $\mathscr{L}_{\text{diss}}$ can be written in a Lindbladian form as given in Eq.(97). The steady-state density matrix ρ^{ss} is given by

$$\mathscr{L} \rho^{\text{ss}} = 0 \tag{130}$$

that is, by the eigen matrix of $\mathscr{L}$ with zero eigenvalue. Except this zero eigenvalue, all the eigenvalues must have negative real parts. The long-time evolution of the system towards the steady-state is mainly governed by the Liovillian gap which is the nonzero eigenvalue with the minimum magnitude of the real part. The drives and dissipation may be engineered to obtain an interesting steady-state or multiple steady-states or non-equilibrium phase transitions which are of current research interest.

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