

## 6, Quantum theory of Fluorescence

1. Quantum theory of Damping: Density operator
2. Quantum theory of Damping: Langevin equation
3. System-Reservoir Interaction
4. Resonance Fluorescence
5. Decoherence

### Ref:

Ch. 8, 9, 10 in *"Quantum Optics,"* by M. Scully and M. Zubairy.

Ch. 7 in *"Mesoscopic Quantum Optics,"* by Y. Yamamoto and A. Imamoglu.

Ch. 8 in *"The Quantum Theory of Light,"* by R. Loudon.

Ch. 14, 15 in *"Elements of Quantum Optics,"* by P. Meystre and M. Sargent III.

Ch. 8 in *"Introductory Quantum Optics,"* by C. Gerry and P. Knight.

# Damping via oscillator reservoir

- consider a single-mode field, with frequency  $\omega$ ,  $\hat{a}$  and  $\hat{a}^\dagger$  operators,
- the reservoir may be taken as any large collection of systems with many degrees (e.g. phonons, other photon modes, etc), with closely spaced frequencies  $\nu_k$ ,  $\hat{b}_k$  and  $\hat{b}_k^\dagger$  operators,
- the Hamiltonian for the field-reservoir system is

$$\hat{H} = \hbar\omega\hat{a}^\dagger\hat{a} + \sum_k \hbar\nu_k\hat{b}_k^\dagger\hat{b}_k + \sum_k \hbar g_k(\hat{b}_k^\dagger\hat{a} + \hat{a}^\dagger\hat{b}_k),$$

- the Heisenberg equations of motion for the operators are

$$\frac{d}{dt}\hat{a}(t) = \frac{i}{\hbar}[\hat{H}, \hat{a}] = -i\omega\hat{a}(t) - i\sum_k g_k\hat{b}_k(t),$$

$$\frac{d}{dt}\hat{b}_k(t) = -i\nu_k\hat{b}_k(t) - ig_k\hat{a}(t),$$

# Damping via oscillator reservoir

→ the closed equation for the field operator  $\hat{a}(t)$  is,

$$\begin{aligned}\hat{b}_k(t) &= \hat{b}_k(0)e^{-i\nu_k t} - ig_k \int_0^\infty dt' e^{-i\nu_k(t-t')} \hat{a}(t'), \\ \frac{d}{dt} \hat{a}(t) &= -i\omega \hat{a}(t) - \sum_k |g_k|^2 \int_0^\infty dt' e^{-i\nu_k(t-t')} \hat{a}(t') + \hat{f}_a(t),\end{aligned}$$

where

$$\hat{f}_a(t) = -i \sum_k g_k \hat{b}_k(0) e^{-i\nu_k t},$$

→ define slowly varying operator  $\hat{a}'(t) = \hat{a}e^{i\omega t}$ , then

$$\begin{aligned}\frac{d}{dt} \hat{a}'(t) &= - \sum_k |g_k|^2 \int_0^\infty dt' e^{-i(\nu_k - \omega)(t-t')} \hat{a}'(t') + \hat{F}_a(t), \\ \hat{F}_a(t) &= -i \sum_k g_k \hat{b}_k(0) e^{-i(\nu_k - \omega)t},\end{aligned}$$

→ we use the notation  $\hat{a}(t)$  to replace  $\hat{a}'(t)$  in the following formulations,

# Markovian white noise

- ➔ a single-mode field interacting with reservoir,

$$\begin{aligned}\frac{d}{dt}\hat{a}(t) &= -\sum_k |g_k|^2 \int_0^\infty dt' e^{-i(\nu_k - \omega)(t-t')} \hat{a}(t') + \hat{F}_a(t), \\ \hat{F}_a(t) &= -i \sum_k g_k \hat{b}_k(0) e^{-i(\nu_k - \omega)t},\end{aligned}$$

- ➔ with the Weisskopf-Wigner theory,  $G(t) = \frac{\Gamma}{2}\delta(t)$ ,

$$\sum_k |g_k|^2 \int_0^\infty dt' e^{-i(\nu_k - \omega)(t-t')} \hat{a}(t') \equiv \int_0^\infty dt' G(t-t') \hat{a}(t') \approx \frac{\Gamma}{2} \hat{a}(t),$$

where  $G(t-t') = \sum_k |g_k|^2 e^{-i(\nu_k - \omega)(t-t')} = \int_\nu D(\nu) |g(\nu)|^2 e^{-i(\nu - \omega)(t-t')}$ ,  
 $D(\nu)$  is the density of state for the reservoir, and the equation of motion for the field interacting with the reservoir is

$$\frac{d}{dt}\hat{a}(t) = -\frac{\Gamma}{2}\hat{a}(t) + \hat{F}_a(t),$$

# Commutation relation

- ➔ for a Markovian process

$$\frac{d}{dt}\hat{a}(t) = -\frac{\Gamma}{2}\hat{a}(t) + \hat{F}_a(t), \quad \hat{F}_a(t) = -i \sum_k g_k \hat{b}_k(0) e^{-i(\nu_k - \omega)t},$$

- ➔ if one dismiss the fluctuation term  $\hat{F}_a(t)$ ,

$$\frac{d}{dt}\hat{a}(t) = -\frac{\Gamma}{2}\hat{a}(t),$$

we have the solution  $\hat{a}(t) = \hat{a}(0)e^{-\Gamma/2t}$ ,

- ➔ for the non-interacting field, the commutation relation at  $t = 0$  is  $[\hat{a}(0), \hat{a}^\dagger(0)] = 1$ , but as time evolves to  $t \neq 0$ , the commutation relation is not satisfied,

$$[\hat{a}(t), \hat{a}^\dagger(t)] = \exp(-\Gamma t),$$

- ➔ the noise operator with appropriate correlation properties helps to maintain the commutation relation at all time,

# Thermal reservoir

- the noise operator is defined as,

$$\hat{F}_a(t) = -i \sum_k g_k \hat{b}_k(0) e^{-i(\nu_k - \omega)t},$$

- suppose the reservoir is in thermal equilibrium,

$$\langle b_k(0) \rangle_R = \langle b_k^\dagger(0) \rangle_R = 0$$

$$\langle b_k(0) b_{k'}(0) \rangle_R = 0 \quad \langle b_k^\dagger(0) b_{k'}^\dagger(0) \rangle_R = 0$$

$$\langle b_k^\dagger(0) b_{k'}(0) \rangle_R = \bar{n}_k \delta_{kk'}$$

$$\langle b_k(0) b_{k'}^\dagger(0) \rangle_R = (\bar{n}_k + 1) \delta_{kk'}$$

where

$$\bar{n}_k = \langle n \rangle = \sum_n n \rho_{nn} = \frac{1}{\exp(\hbar\nu_k/k_B T) - 1},$$

# Thermal reservoir

- the noise operator is defined as,

$$\hat{F}_a(t) = -i \sum_k g_k \hat{b}_k(0) e^{-i(\nu_k - \omega)t},$$

- the Langevin noise operators have *zero means*

$$\langle \hat{F}_a(t) \rangle_R = \langle \hat{F}_a^\dagger(t) \rangle_R = 0,$$

but non-zero variances,

$$\begin{aligned} \langle \hat{F}_a^\dagger(t) \hat{F}_a(t') \rangle_R &= \sum_k \sum_{k'} g_k g_{k'} \langle \hat{b}_k^\dagger \hat{b}_{k'} \rangle \exp[i(\nu_k - \omega)t - i(\nu_{k'} - \omega)t'], \\ &= \sum_k |g_k|^2 \bar{n}_k \exp[i(\nu_k - \omega)(t - t')], \\ &= \Gamma \bar{n}_{th} \delta(t - t'), \end{aligned}$$

where  $\bar{n}_{th} = \bar{n}_{(\nu_k - \omega)}$ ,

# Fluctuation-dissipation theory

- for a single-mode field interacting with thermal reservoir,

$$\frac{d}{dt}\hat{a}(t) = -\frac{\Gamma}{2}\hat{a}(t) + \hat{F}_a(t),$$

- the Langevin noise operators have *zero means* but non-zero variances,

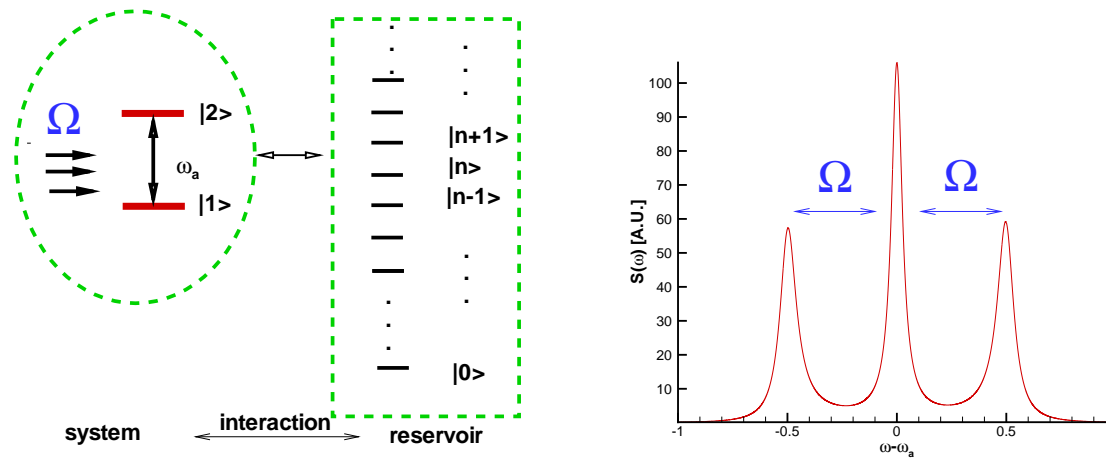
$$\begin{aligned}\langle \hat{F}_a(t) \rangle_R &= \langle \hat{F}_a^\dagger(t) \rangle_R = 0, \\ \langle \hat{F}_a^\dagger(t) \hat{F}_a^\dagger(t') \rangle_R &= \langle \hat{F}_a(t) \hat{F}_a(t') \rangle_R = 0, \\ \langle \hat{F}_a^\dagger(t) \hat{F}_a(t') \rangle_R &= \Gamma \bar{n}_{th} \delta(t - t'), \\ \langle \hat{F}_a(t) \hat{F}_a^\dagger(t') \rangle_R &= \Gamma (\bar{n}_{th} + 1) \delta(t - t'),\end{aligned}$$

- the damping of the system is determined from the fluctuating forces of the reservoir, in other words, the fluctuations induced by the reservoir give rise to the dissipation in the system,

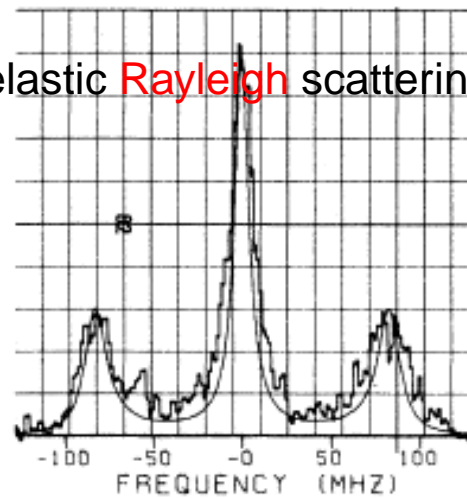
$$\Gamma = \frac{1}{\bar{n}_{th}} \int_{-\infty}^{\infty} dt' \langle \hat{F}_a^\dagger(t) \hat{F}_a(t') \rangle_R,$$



# Mollow's triplet: Resonance Fluorescence Spectrum



elastic **Rayleigh** scattering and inelastic **Raman** scattering

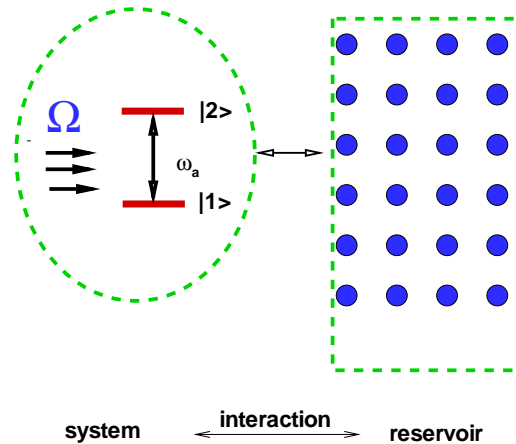
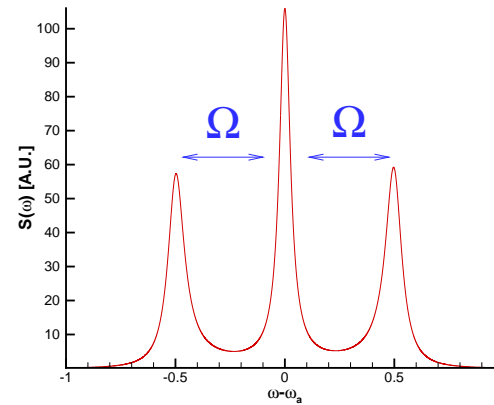
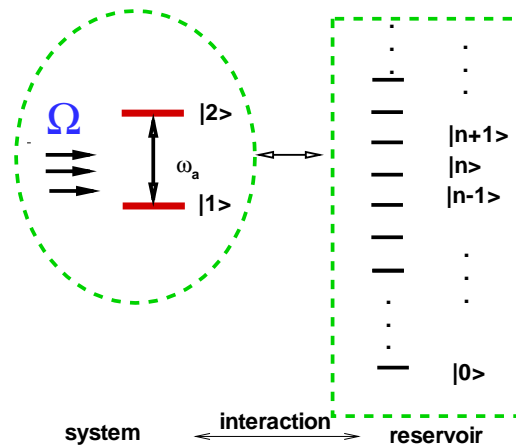


**Theory:** B. R. Mollow, *Phys. Rev.* **188**, 1969 (1969).

**Exp:** F. Y. Wu, R. E. Grove, and S. Ezekiel, *Phys. Rev. Lett.* **35**, 1426 (1975).

# Photon-Atom Interaction in PhCs

## Reservoir Theory



# Hamiltonian of our system: Jaynes-Cummings model

$$\begin{aligned}
 H &= \frac{\hbar}{2}\omega_a\sigma_z + \hbar \sum_k \omega_k a_k^\dagger a_k + \frac{\Omega}{2}\hbar(\sigma_- e^{i\omega_L t} + \sigma_+ e^{-i\omega_L t}) \\
 &+ \hbar \sum_k (g_k \sigma_+ a_k + g_k^* a_k^\dagger \sigma_-)
 \end{aligned}$$

And we want to solve the generalized Bloch equations:

$$\begin{aligned}
 \dot{\sigma}_-(t) &= i\frac{\Omega}{2}\sigma_z(t)e^{-i\Delta t} + \int_{-\infty}^t dt' G(t-t')\sigma_z(t)\sigma_-(t') + n_-(t) \\
 \dot{\sigma}_+(t) &= -i\frac{\Omega}{2}\sigma_z(t)e^{i\Delta t} + \int_{-\infty}^t dt' G_c(t-t')\sigma_+(t')\sigma_z(t) + n_+(t) \\
 \dot{\sigma}_z(t) &= i\Omega(\sigma_-(t)e^{i\Delta t} - \sigma_+(t)e^{-i\Delta t}) + n_z(t) \\
 &- 2 \int_{-\infty}^t dt' [G(t-t')\sigma_+(t)\sigma_-(t') + G_c(t-t')\sigma_+(t')\sigma_-(t)]
 \end{aligned}$$

## Remarks:

1. coupling constant:

$$g_k \equiv g_k(\hat{\mathbf{d}}, \vec{r}_0) = |d|\omega_a \sqrt{\frac{1}{2\hbar\epsilon_0\omega_k V}} \hat{\mathbf{d}} \cdot \mathbf{E}_k^*(\vec{r}_0)$$

2. memory functions:

$$G(\tau) \equiv \sum_k |g_k|^2 e^{i\Delta_k \tau} \Theta(\tau)$$

$$G_c(\tau) \equiv \sum_k |g_k|^2 e^{-i\Delta_k \tau} \Theta(\tau)$$

3. Markovian approximation:

$$G(t) = G_c(t) = \Gamma\delta(t)$$

# Quantum noise operators

$$n_{-}(t) = i \sum_k g_k e^{i\Delta_k t} \sigma_z(t) a_k(-\infty)$$

$$n_{+}(t) = -i \sum_k g_k^* e^{-i\Delta_k t} a_k^+(-\infty) \sigma_z(t)$$

$$n_z(t) = 2i \sum_k [g_k^* e^{-i\Delta_k t} a_k^+(-\infty) \sigma_{-}(t) - g_k e^{i\Delta_k t} \sigma_{+}(t) a_k^+(-\infty)]$$

where the mean and the correlation functions of the reservoir before interaction,

$$\langle a_k(-\infty) \rangle_R = \langle a_k^{\dagger}(-\infty) \rangle_R = 0$$

$$\langle a_k(-\infty) a_{k'}(-\infty) \rangle_R = 0$$

$$\langle a_k^{\dagger}(-\infty) a_{k'}^{\dagger}(-\infty) \rangle_R = 0$$

$$\langle a_k^{\dagger}(-\infty) a_{k'}(-\infty) \rangle_R = \bar{n}_k \delta_{kk'}$$

$$\langle a_k(-\infty) a_{k'}^{\dagger}(-\infty) \rangle_R = (\bar{n}_k + 1) \delta_{kk'}$$

# Heisenberg-Langevin equations

$$\begin{aligned}\dot{\sigma}_-(t) &= i\frac{\Omega}{2}\sigma_z(t)e^{-i\Delta t} + \int_{-\infty}^t dt' G(t-t')\sigma_z(t)\sigma_-(t') + n_-(t) \\ \dot{\sigma}_+(t) &= -i\frac{\Omega}{2}\sigma_z(t)e^{i\Delta t} + \int_{-\infty}^t dt' G_c(t-t')\sigma_+(t')\sigma_z(t) + n_+(t) \\ \dot{\sigma}_z(t) &= i\Omega(\sigma_-(t)e^{i\Delta t} - \sigma_+(t)e^{-i\Delta t}) + n_z(t) \\ &\quad - 2 \int_{-\infty}^t dt' [G(t-t')\sigma_+(t)\sigma_-(t') + G_c(t-t')\sigma_+(t')\sigma_-(t)]\end{aligned}$$

where the Langevin noise operators have *zero means* but non-zero variances,

$$\langle n_-(t) \rangle_R = \langle n_+(t) \rangle_R = \langle n_z(t) \rangle_R = 0$$

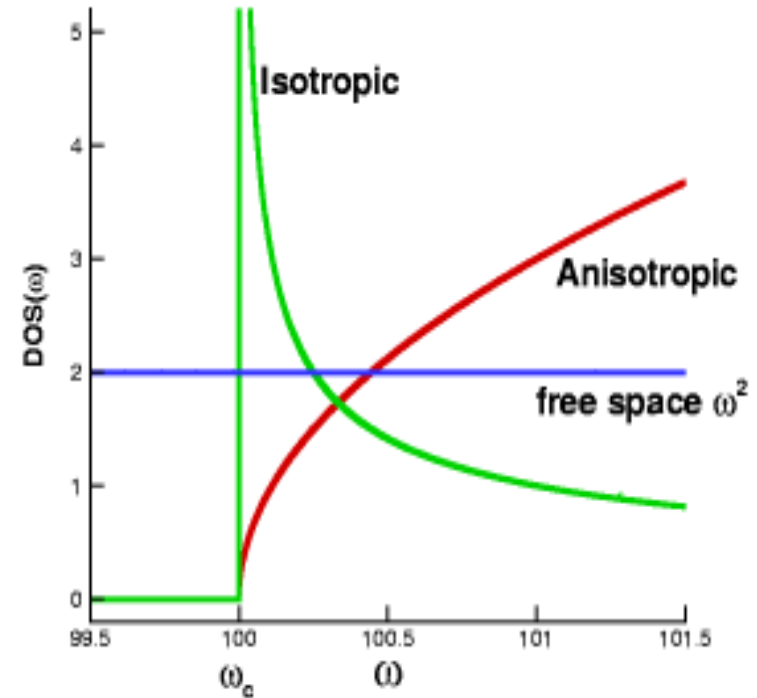
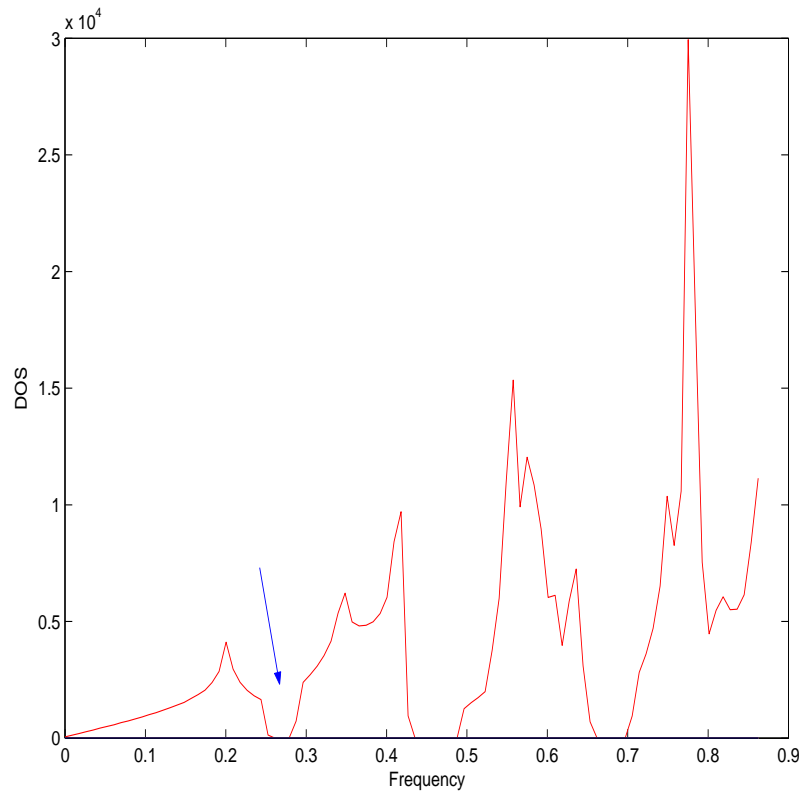
$$\langle n_-(t)n_-(t') \rangle_R = \langle n_+(t)n_+(t') \rangle_R = 0$$

$$\langle n_-(t)n_+(t') \rangle_R = \sum_k |g_k|^2 (\bar{n}_k + 1) e^{i\Delta_k(t-t')} \langle \sigma_z(t)\sigma_z(t') \rangle$$

$$\langle n_+(t)n_-(t') \rangle_R = \sum_k |g_k|^2 \bar{n}_k e^{-i\Delta_k(t-t')} \langle \sigma_z(t)\sigma_z(t') \rangle$$

$$\langle n_z(t)n_z(t') \rangle_R = 4 \sum_k |g_k|^2 [(\bar{n}_k + 1) e^{i\Delta_k(t-t')} \langle \sigma_+(t)\sigma_-(t') \rangle + \bar{n}_k e^{-i\Delta_k(t-t')} \langle \sigma_-(t)\sigma_+(t') \rangle]$$

# Modeling DOS of PBCs



**anisotropic model:**  $\omega_k = \omega_c + A|\mathbf{k} - \mathbf{k}_0^i|^2$

$$D(\omega) = \sqrt{\frac{\omega - \omega_c}{A^3}} \Theta(\omega - \omega_c)$$

S. Y. Zhu, et al., *Phys. Rev. Lett.* **84**, 2136 (2000).

# Memory functions of PBCs

- **anisotropic model:**  $\omega_{\mathbf{k}} = \omega_c + A|\mathbf{k} - \mathbf{k}_0^i|^2$

$$D(\omega) = \sqrt{\frac{\omega - \omega_c}{A^3}} \Theta(\omega - \omega_c)$$

- the memory functions under this anisotropic model also can be derived:

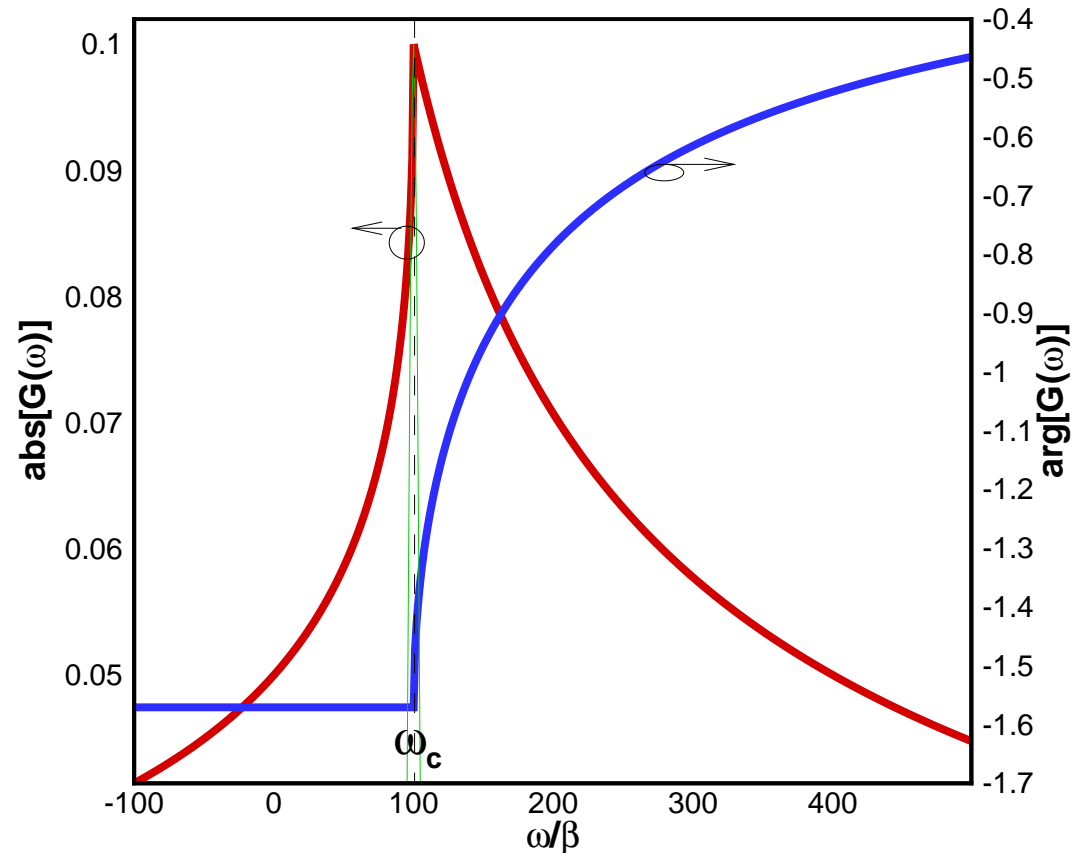
$$\tilde{G}(\omega) = \beta^{3/2} \frac{-i}{\sqrt{\omega_c + \sqrt{\omega_c - \omega_a - \omega}}}$$

$$\tilde{G}_c(\omega) = \beta^{3/2} \frac{i}{\sqrt{\omega_c + \sqrt{\omega_c - \omega_a + \omega}}}$$

where  $\beta^{3/2} = \frac{\omega_a^2 d^2}{6\hbar\epsilon_0\pi A^{3/2}} \eta$ , and we have used the space average coupling strength  $\eta \equiv \frac{3}{8\pi} \int d\Omega |\mathbf{d} \cdot \mathbf{E}|^2$  in the derivation.



# Memory functions of PBCs



Amplitude and phase spectrum of the memory function

with  $\omega_a = \omega_c = 100\beta$ .

# Correlations of noise operators at zero temperature

$$\begin{aligned}
 \langle \tilde{n}_-(\omega_1) \tilde{n}_+(-\omega_2) \rangle_R &= \pi N(\omega_1) \Theta(\omega_1 + \omega_a - \omega_c) \delta(\omega_1 - \omega_2) \\
 \langle \tilde{n}_z(\omega_1) \tilde{n}_z(-\omega_2) \rangle_R &= N(\omega_1) [4\pi \delta(\omega_1 - \omega_2) + \langle \tilde{\sigma}_z(\omega_1 - \omega_2) \rangle_R] \\
 &\quad \cdot \Theta(\omega_1 + \omega_a - \omega_c) \\
 \langle \tilde{n}_z(\omega_1) \tilde{n}_-(-\omega_2) \rangle_R &= 0 \\
 \langle \tilde{n}_-(\omega_1) \tilde{n}_z(-\omega_2) \rangle_R &= N(\omega_1) \langle \tilde{\sigma}_-(\omega_1 - \omega_2) \rangle_R \Theta(\omega_1 + \omega_a - \omega_c) \\
 \langle \tilde{n}_z(\omega_1) \tilde{n}_+(-\omega_2) \rangle_R &= N(\omega_1) \langle \tilde{\sigma}_+(\omega_1 - \omega_2) \rangle_R \Theta(\omega_1 + \omega_a - \omega_c) \\
 \langle \tilde{n}_+(\omega_1) \tilde{n}_z(-\omega_2) \rangle_R &= 0
 \end{aligned}$$

with  $N(\omega) \equiv 4\beta^{3/2} \frac{\sqrt{\omega_a + \omega - \omega_c}}{\omega_a + \omega}$

Quantum noises of the photonic bandgap reservoir are not only **color noises** but also exhibit **bandgap behaviour**.

# Liouville operator expansion

$$\sigma_{ij}(t) = e^{-i\mathcal{L}(t-t')} \sigma_{ij}(t') = \sum_{n=0}^{\infty} \frac{[-i(t-t')]^n}{n!} \mathcal{L}^n \sigma_{ij}(t')$$

For zero-th order Liouville operator expansion, we get

$$\dot{\sigma}_-(t) = i\frac{\Omega}{2}\sigma_z(t)e^{-i\Delta t} - \int_{-\infty}^t dt' G(t-t')\sigma_-(t') + n_-(t)$$

$$\dot{\sigma}_+(t) = -i\frac{\Omega}{2}\sigma_z(t)e^{i\Delta t} - \int_{-\infty}^t dt' G_c(t-t')\sigma_+(t') + n_+(t)$$

$$\begin{aligned} \dot{\sigma}_z(t) &= i\Omega(\sigma_-(t)e^{i\Delta t} - \sigma_+(t)e^{-i\Delta t}) \\ &\quad - \int_{-\infty}^t dt' [G(t-t') + G_c(t-t')](1 + \sigma_z(t')) + n_z(t) \end{aligned}$$

valid for the case of

atom with **longer lifetime** and under **weak pumping**

# Solve the optical Bloch equations

Since it is now a linear problem, by using Fourier transform the modified optical Bloch equations become:

$$\overline{\overline{\mathcal{M}}}(\omega) \cdot \vec{\mathcal{X}}(\omega) = \vec{\mathcal{X}}_0(\omega)$$

where

$$\overline{\overline{\mathcal{M}}}(\omega) = \begin{pmatrix} -i(\omega + \Delta) + \tilde{G}(\omega) & 0 & -i\frac{\Omega}{2} \\ 0 & -i(\omega - \Delta) + \tilde{G}_c(\omega) & i\frac{\Omega}{2} \\ -i\Omega & i\Omega & -i\omega + \tilde{G}(\omega) + \tilde{G}_c(\omega) \end{pmatrix}$$

$$\vec{\mathcal{X}}(\omega) = \begin{pmatrix} \tilde{\sigma}_-(\omega + \Delta) \\ \tilde{\sigma}_+(\omega - \Delta) \\ \tilde{\sigma}_z(\omega) \end{pmatrix}$$

$$\vec{\mathcal{X}}_0(\omega) = \begin{pmatrix} \tilde{n}_-(\omega + \Delta) \\ \tilde{n}_+(\omega - \Delta) \\ -2\pi[\tilde{G}(\omega) + \tilde{G}_c(\omega)]\delta(\omega) + \tilde{n}_z(\omega) \end{pmatrix}$$

where  $\tilde{n}_-(\omega)$ ,  $\tilde{n}_+(\omega)$ ,  $\tilde{n}_z(\omega)$ ,  $\tilde{G}(\omega)$ , and  $\tilde{G}_c(\omega)$  are Fourier transforms of  $n_-(t)$ ,  $n_+(t)$ ,

$n_z(t)$ ,  $G(t)$ , and  $G_c(t)$ , respectively.

# Solve the optical Bloch equations

The solutions are

$$\tilde{\sigma}_-(\omega + \Delta) = \frac{(2gh + \Omega^2) \tilde{n}_-(\omega) + \Omega^2 \tilde{n}_+(\omega) + i\Omega g \tilde{n}_z(\omega) - i2\pi\Omega g[\tilde{G}(\omega) + \tilde{G}_c(\omega)]\delta(\omega)}{\Omega^2(f + g) + 2fgh}$$

$$\tilde{\sigma}_+(\omega - \Delta) = \frac{\Omega^2 \tilde{n}_-(\omega) + (2fh + \Omega^2) \tilde{n}_+(\omega) - i\Omega f \tilde{n}_z(\omega) + i2\pi\Omega f[\tilde{G}(\omega) + \tilde{G}_c(\omega)]\delta(\omega)}{\Omega^2(f + g) + 2fgh}$$

$$\tilde{\sigma}_z(\omega) = \frac{2i\Omega g \tilde{n}_-(\omega) - 2i\Omega f \tilde{n}_+(\omega) + 2fg \tilde{n}_z(\omega) - 4\pi fg[\tilde{G}(\omega) + \tilde{G}_c(\omega)]\delta(\omega)}{\Omega^2(f + g) + 2fgh}$$

where

$$f(\omega) = -i\omega - i\Delta + \tilde{G}(\omega)$$

$$g(\omega) = -i\omega + i\Delta + \tilde{G}_c(\omega)$$

$$h(\omega) = -i\omega + \tilde{G}(\omega) + \tilde{G}_c(\omega)$$

# Fluorescence spectrum

- ➔ Because the two-time correlation function of the atomic dipole is proportional to the first order correlation function  $g^{(1)}(\tau)$ , we can obtain the fluorescence spectrum by taking the Fourier transform of the first order correlation function:

$$\begin{aligned} S(\omega) &= \int_{-\infty}^{\infty} d\tau g^{(1)}(\tau) e^{i\omega\tau} \\ &\propto \langle \tilde{\sigma}_+(\omega) \tilde{\sigma}_-(-\omega) \rangle_R \end{aligned}$$

- ➔ It should be noted that here we cannot directly apply the quantum regression theorem since it is invalid for non-Markovian process.

# Mollow's triplet

- ➔ As a check, we first use our formulation to calculate the free space case.
- ➔ At free space, one can assume the memory functions are delta functions since  $\sum_k |g_k|^2 e^{i\Delta_k t} = \Gamma \delta(t)$  with  $\Gamma$  being the decay rate of the excited atom.
- ➔ The noise correlation functions at zero temperature are also delta-function correlated (i.e., white noises).
- ➔ Therefore, the fluorescence spectrum at steady state is given by:

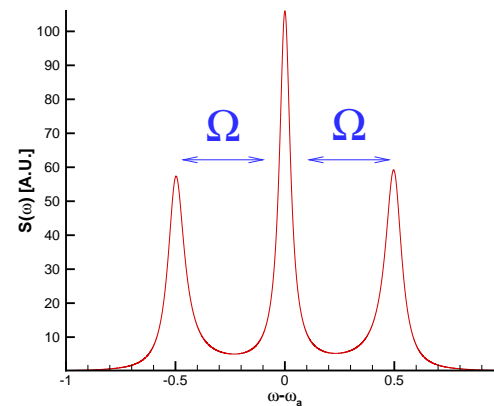
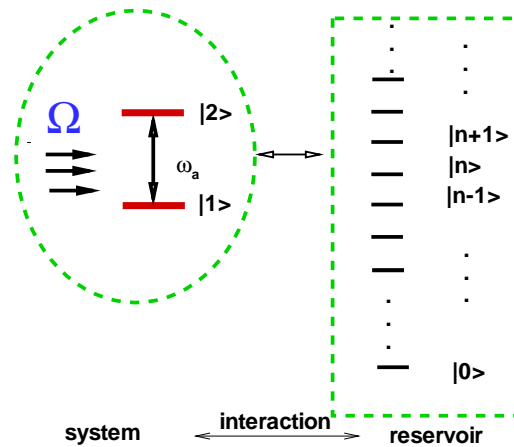
$$\langle \tilde{\sigma}_+(\omega) \tilde{\sigma}_-(-\omega) \rangle_R = \frac{\pi^2 \Omega^2 (\frac{\Gamma^2}{4} + \Delta^2)}{\frac{\Omega^2}{2} + \Delta^2 + \frac{\Gamma^2}{4}} \delta(\omega + \Delta)$$
$$+ \frac{\pi \Gamma \Omega^4 (\frac{\Omega^2}{2} + \Gamma^2 + (\omega + \Delta)^2)}{2(\frac{\Omega^2}{2} + \Delta^2 + \frac{\Gamma^2}{4}) [\Gamma^2 (\frac{\Omega^2}{2} + \Delta^2 + \frac{\Gamma^2}{4} - 2(\omega + \Delta)^2)^2 + (\omega + \Delta)^2 (\Omega^2 + \Delta^2 + \frac{5}{4} \Gamma^2 - (\omega + \Delta)^2)}$$

# Mollow's triplet

- In the limit of strong on-resonance pumping ( $\Omega \gg \Gamma$ ,  $\Delta = 0$ ), Eq.(1) can be reduced to:

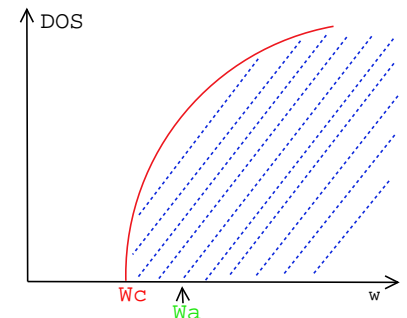
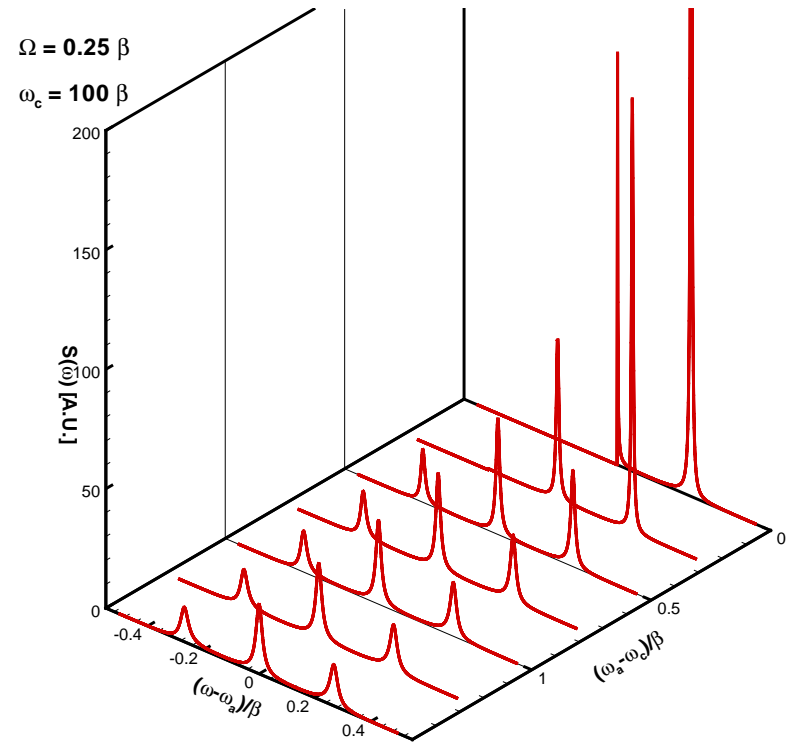
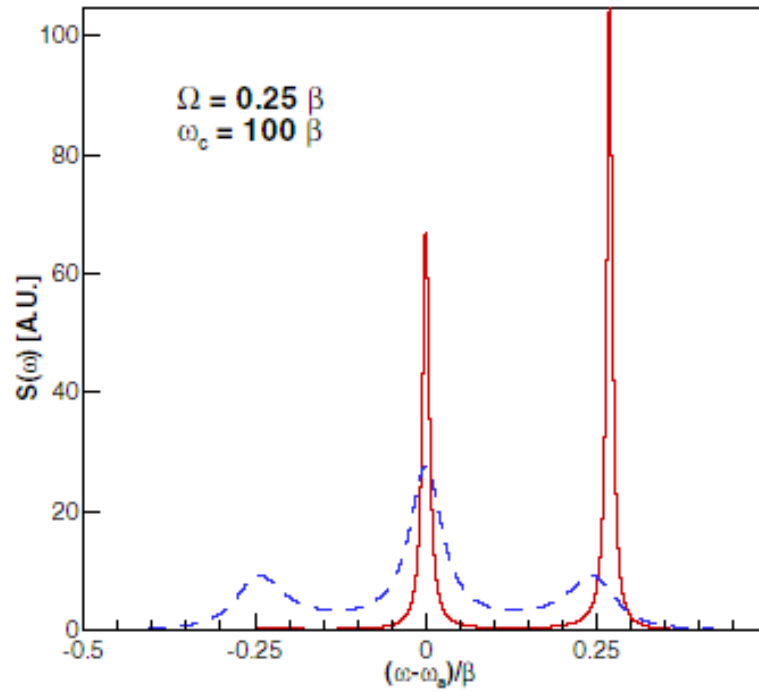
$$\langle \tilde{\sigma}_+(\omega) \tilde{\sigma}_-(-\omega) \rangle_R = 2\pi \left\{ 2\pi \frac{\Gamma^2}{4\Omega^2} \delta(\omega) + \frac{\frac{3}{16}\Gamma}{(\omega + \Omega)^2 + \frac{9}{16}\Gamma^2} + \frac{\frac{1}{4}\Gamma}{\omega^2 + \frac{1}{4}\Gamma^2} + \frac{\frac{3}{16}\Gamma}{(\omega - \Omega)^2 + \frac{9}{16}\Gamma^2} \right\}$$

- Then, the resonance fluorescence spectrum exhibits the Mollow triplets for white noise: three Lorentzian profiles with peaks in the ratio 1 : 3 : 1, and widths of  $\frac{3}{2}\Gamma$ ,  $\Gamma$ , and  $\frac{3}{2}\Gamma$ .





# Resonance fluorescence spectra near the band-edge



# Quadrature spectra

Define quadrature field operator as:

$$\hat{E}_\theta(t) = e^{i\theta} \hat{E}^{(+)}(t) + e^{-i\theta} \hat{E}^{(-)}(t)$$

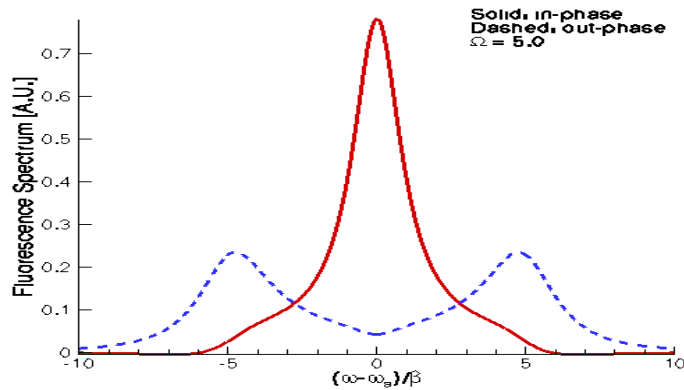
$\theta = 0$  ( $\frac{\pi}{2}$ ) are the *in-phase* (*out-of-phase*) quadrature fields.

Then the corresponding spectra with normally order variance is:

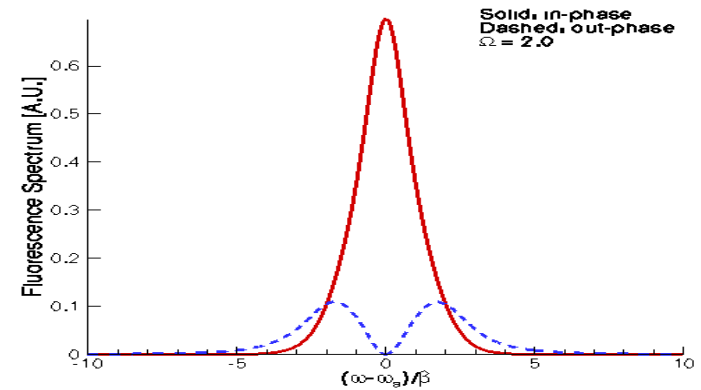
$$\begin{aligned} S_\theta(\omega) &\equiv \langle \tilde{E}_\theta(\omega), \tilde{E}_\theta(-\omega) \rangle \\ &\propto \frac{1}{4} [\langle \tilde{\sigma}_-(\omega) \tilde{\sigma}_-(-\omega) \rangle e^{-2i\theta} + \langle \tilde{\sigma}_+(\omega) \tilde{\sigma}_-(-\omega) \rangle \\ &\quad + \langle \tilde{\sigma}_+(-\omega) \tilde{\sigma}_-(\omega) \rangle + \langle \tilde{\sigma}_+(-\omega) \tilde{\sigma}_+(\omega) \rangle e^{2i\theta}] \end{aligned}$$

# Quadrature spectra in free space

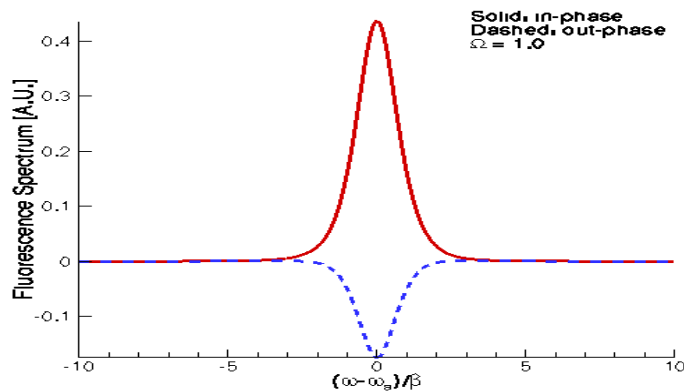
$$\Omega = 5.0\Gamma$$



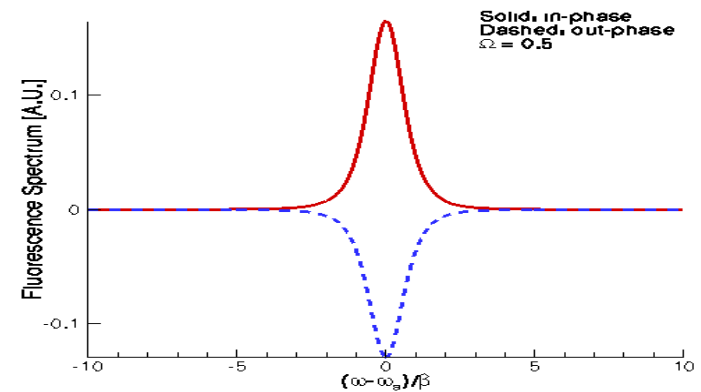
$$\Omega = 2.0\Gamma$$



$$\Omega = 1.0\Gamma$$



$$\Omega = 0.5\Gamma$$



Theory: D. F. Walls and P. Zoller, *Phys. Rev. Lett.* **47**, 709 (1981).

Theory: L. Mandel, *Phys. Rev. Lett.* **49**, 136 (1982).

# Observation of squeezing fluorescence spectra

VOLUME 81, NUMBER 17

PHYSICAL REVIEW LETTERS

26 OCTOBER 1998

## Observation of Squeezing in the Phase-Dependent Fluorescence Spectra of Two-Level Atoms

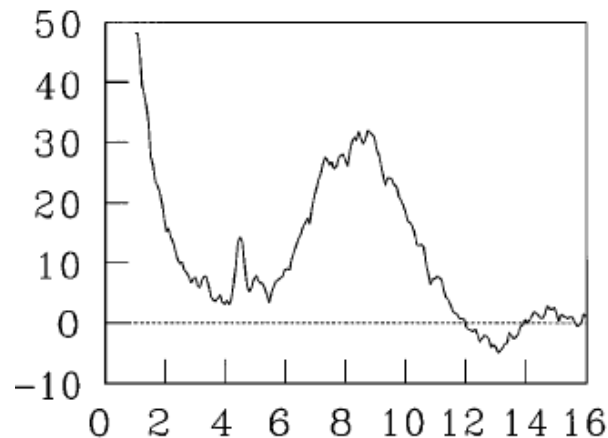
Z. H. Lu, S. Bali, and J. E. Thomas

*Physics Department, Duke University, Durham, North Carolina 27708-0305*

(Received 18 June 1998)

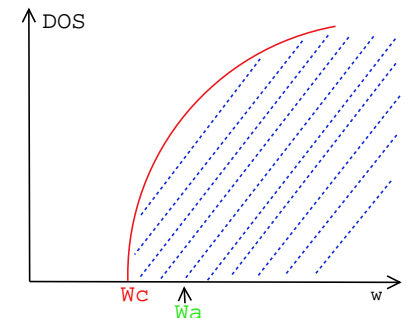
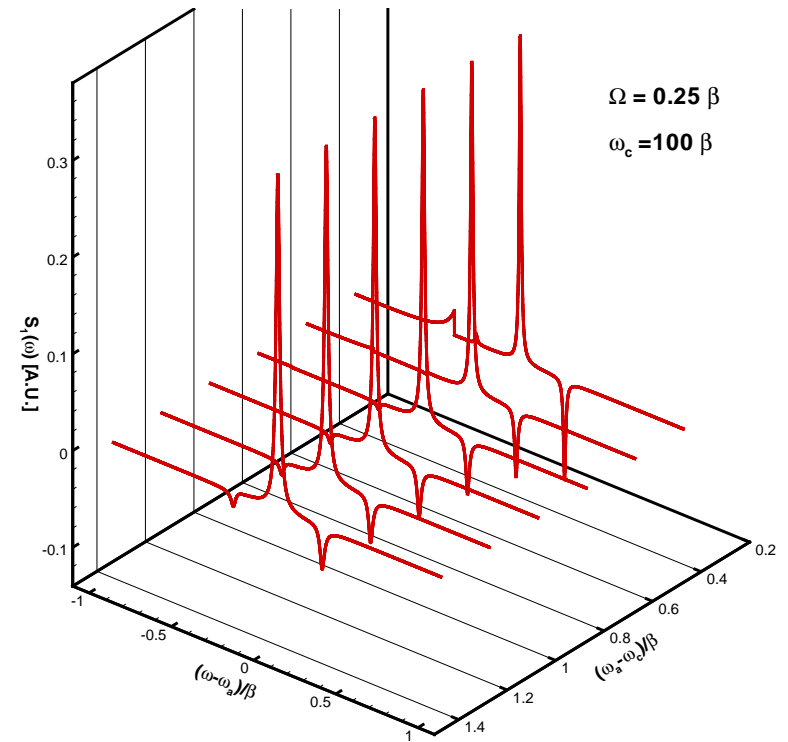
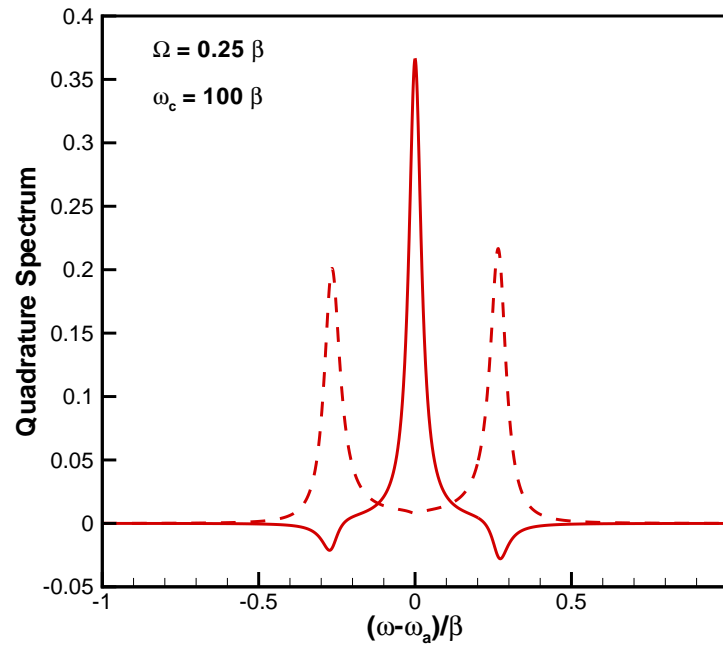
We observe squeezing in the phase-dependent fluorescence spectra of two-level atoms that are coherently driven by a near-resonant laser field in *free space*. In contrast to previous predictions that emphasized the in- and out-of-phase quadratures, we find that maximum squeezing occurs for homodyne detection at a phase near  $\pm 45^\circ$  relative to the exciting field. A new physical picture of phase-dependent noise is developed that incorporates quantum collapses into a Bloch vector model and yields a very simple form for the complete squeezing spectrum. [S0031-9007(98)07454-7]

PACS numbers: 42.50.Lc, 32.80.-t



with  $^{147}\text{Yb}$  atoms

# Fluorescence quadrature spectra near the band-edge



## Resonance fluorescence quadrature spectra near the band-edge

1. Suppression and enhancement of the relative fluorescence peak amplitudes varied at different wavelength offsets.
2. Squeezing occurs in the **out-of-phase** quadrature for free space when  $\Omega^2 < 4\Gamma^2$ .
3. Squeezing occurs in the **in-phase** quadrature for PhCs when  $\Omega^2 > 4\Gamma^2$ .
4. Resonance fluorescence squeezing spectra come from the interference between two sidebands of Mollow's triplet.

# Density operator method

- ➔ Heisenberg-Langevin equation is a directly correspondence to classical description of stochastic system,
- ➔ in many cases, Heisenberg-Langevin equation is *nonlinear*,
- ➔ in general it is extremely difficult to deal with the Heisenberg-Langevin equation,
- ➔ another method is to develop a Schrödinger or interaction picture analysis,
- ➔ in this way we want to use a linear deterministic differential equation for the *reduced system density operator*,
- ➔ naturally, as the quantum system is open, there is statistical as well as quantum uncertainty and a true wave function description is no longer possible,

## Ref:

**Ch. 7** in "*Mesoscopic Quantum Optics*," by Y. Yamamoto and A. Imamoglu.

"*Quantum Noise*," by C. W. Gardiner.

# Master equation

- we consider a system  $S$  interacting with a reservoir  $R$  via the interaction Hamiltonian  $\hat{V}$ ,
- the combined density operator is denoted by  $\hat{\rho}(t)$ ,
- assume that at an initial time  $t = 0$ , the two systems are uncorrelated,

$$\hat{\rho}(t = 0) = \hat{\rho}_S(0) \otimes \hat{\rho}_R(0),$$

- in the interaction picture, the dynamics of  $\hat{\rho}(t)$  is

$$\frac{d}{dt}\hat{\rho}(t) = \frac{1}{i\hbar}[\hat{H}_I(t), \hat{\rho}(t)],$$

- since the number of degrees of freedom of the reservoir is very large, it is impossible to keep track of its quantum evolution,
- we can only focus on the system with a reduced density operator, by tracing over the reservoir degrees of freedom,

$$\frac{d}{dt}\hat{\rho}_S(t) = \frac{1}{i\hbar}\text{Tr}_R([\hat{H}_I(t), \hat{\rho}(t)]),$$



# Master equation

- we can only focus on the system with a reduced density operator,

$$\frac{d}{dt}\hat{\rho}_S(t) = \frac{1}{i\hbar}\text{Tr}_R([\hat{H}_I(t), \hat{\rho}(t)]),$$

where

$$\frac{d}{dt}\hat{\rho}(t) = \frac{1}{i\hbar}[\hat{H}_I(t), \hat{\rho}(t)],$$

- without any approximation, the master equation for the reduced density operator is,

$$\frac{d}{dt}\hat{\rho}_S(t) = \left(\frac{1}{i\hbar}\right)^2 \int_0^t dt' \text{Tr}_R([\hat{H}_I(t), [\hat{H}_I(t'), \hat{\rho}(t')]]) + \frac{1}{i\hbar}\text{Tr}_R([\hat{H}_I(t), \hat{\rho}(0)]),$$

- since  $\text{Tr}_R([\hat{H}_I(t), \hat{\rho}(0)])$  vanish for all the interaction Hamiltonians of interest in quantum optics, we have the master equation,

$$\frac{d}{dt}\hat{\rho}_S(t) = \left(\frac{1}{i\hbar}\right)^2 \int_0^t dt' \text{Tr}_R([\hat{H}_I(t), [\hat{H}_I(t'), \hat{\rho}(t')]]),$$

# Born-Markov approximation

- the master equation for the reduced density operator, without any approximation,

$$\frac{d}{dt}\hat{\rho}_S(t) = \left(\frac{1}{i\hbar}\right)^2 \int_0^t dt' \text{Tr}_R([\hat{H}_I(t), [\hat{H}_I(t'), \hat{\rho}(t')]]),$$

- four key approximations used in the following,

1. Rotating-wave approximation,
2. Born approximation,

$$\hat{\rho}(t') = \hat{\rho}_S(t') \otimes \hat{\rho}_R(t'),$$

3. the initial radiation field density operator commutes with the free Hamiltonian and the reservoir is not affected by the interaction with the system,

$$\hat{\rho}_R(t) = \text{Tr}_S[\hat{\rho}(t)] = \hat{\rho}_R(0),$$

4. Markov approximation,

$$\hat{\rho}_S(t') \approx \hat{\rho}_S(t),$$

# Master equation

- the master equation for the reduced density operator, without any approximation,

$$\frac{d}{dt}\hat{\rho}_S(t) = \left(\frac{1}{i\hbar}\right)^2 \int_0^t dt' \text{Tr}_R([\hat{H}_I(t), [\hat{H}_I(t'), \hat{\rho}(t')]]),$$

- with Born-Markov approximation, the master equation becomes

$$\frac{d}{dt}\hat{\rho}_S(t) = \left(\frac{1}{i\hbar}\right)^2 \int_0^t dt' \text{Tr}_R([\hat{H}_I(t), [\hat{H}_I(t'), \hat{\rho}_S(t) \otimes \hat{\rho}_R(0)]]),$$

- atom damping by field reservoirs,
- field damping by field reservoirs,
- field damping by atomic reservoirs,

# Atom damping by field reservoirs

- consider a two-level atom damped by a field reservoir in free space,
- the interaction Hamiltonian is

$$\hat{H}_I = \sum_k \hbar(g_k \hat{\sigma}_- \hat{a}_k^\dagger e^{-i(\omega - \omega_k)t} + \text{H. C.}),$$

- assume the reservoir density operator is a multimode thermal field,

$$\hat{\rho}_R = \prod_k \sum_n \frac{\exp(-\frac{\hbar\omega_k n}{k_B T})}{1 - \exp(-\frac{\hbar\omega_k n}{k_B T})} |n\rangle_k \langle n|,$$

- the equation of motion for the reduced density operator  $\text{Tr}_R[\hat{\rho}(t)] \equiv \hat{\rho}_a(t)$  is,

$$\begin{aligned} \frac{d}{dt} \hat{\rho}_a(t) &= \left(\frac{1}{i\hbar}\right)^2 \int_0^t dt' \text{Tr}_R([\hat{H}_I(t), [\hat{H}_I(t'), \hat{\rho}_a(t) \otimes \hat{\rho}_R(0)]]), \\ &= -\frac{1}{2} \Gamma \{ n_{th} [\hat{\sigma}_- \hat{\sigma}_+ \hat{\rho}_a - \hat{\sigma}_+ \hat{\rho}_a \hat{\sigma}_-] + (n_{th} + 1) [\hat{\sigma}_+ \hat{\sigma}_- \hat{\rho}_a - \hat{\sigma}_- \hat{\rho}_a \hat{\sigma}_+] \} + \text{H} \end{aligned}$$

# Field damping by field reservoirs

- ➔ consider a single-mode field in a cavity with a finite leakage rate,
- ➔ assume the reservoir density operator is a multimode thermal field,

$$\hat{\rho}_R = \prod_k \sum_n \frac{\exp(-\frac{\hbar\omega_k n}{k_B T})}{1 - \exp(-\frac{\hbar\omega_k n}{k_B T})} |n\rangle_k \langle n|_k,$$

- ➔ the equation of motion for the reduced density operator  $\text{Tr}_R[\hat{\rho}(t)] \equiv \hat{\rho}_f(t)$  is,

$$\begin{aligned} \frac{d}{dt} \hat{\rho}_f(t) &= \left(\frac{1}{i\hbar}\right)^2 \int_0^t dt' \text{Tr}_R([\hat{H}_I(t), [\hat{H}_I(t'), \hat{\rho}_f(t) \otimes \hat{\rho}_R(0)]]), \\ &= - \int_{t_0}^t dt' \sum_k g_k^2 \{ n_{th} [\hat{a}\hat{a}^\dagger \hat{\rho}_f(t') - \hat{a}^\dagger \hat{\rho}_f(t') \hat{a}] e^{-i(\omega - \omega_k)(t-t')} \\ &\quad + (n_{th} + 1) [\hat{a}^\dagger \hat{a} \hat{\rho}_f(t') - \hat{a} \hat{\rho}_f(t') \hat{a}^\dagger] e^{i(\omega - \omega_k)(t-t')} \} + \text{H. C.}, \end{aligned}$$

# Field damping by field reservoirs

- the equation of motion for the reduced density operator  $\text{Tr}_R[\hat{\rho}(t)] \equiv \hat{\rho}_f(t)$  is,

$$\begin{aligned} \frac{d}{dt} \hat{\rho}_f(t) = & - \int_{t_0}^t dt' \sum_k g_k^2 \{ n_{th} [\hat{a} \hat{a}^\dagger \hat{\rho}_f(t') - \hat{a}^\dagger \hat{\rho}_f(t') \hat{a}] e^{-i(\omega - \omega_k)(t-t')} \\ & + (n_{th} + 1) [\hat{a}^\dagger \hat{a} \hat{\rho}_f(t') - \hat{a} \hat{\rho}_f(t') \hat{a}^\dagger] e^{i(\omega - \omega_k)(t-t')} \} + \text{H. C.}, \end{aligned}$$

- again, by replacing  $\sum_k g_k^2$  with the integral  $\int d\omega_k D(\omega_k) g(\omega_k)^2$ , and

$$\begin{aligned} \int_{t_0}^t dt' \sum_k g_k^2 e^{\pm i(\omega - \omega_k)(t-t')} &= \int_{t_0}^t dt' \int d\omega_k D(\omega_k) g(\omega_k)^2 e^{\pm i(\omega - \omega_k)(t-t')}, \\ &\approx \int d\omega_k D(\omega_k) g(\omega_k)^2 \pi \delta(\omega - \omega_k), \\ &\approx \pi D(\omega) g(\omega)^2 \equiv \frac{1}{2} \left( \frac{\omega}{Q_e} \right), \end{aligned}$$

where  $\omega/Q_e$  is the cavity photon decay rate due to leakage (output coupling) via a partially reflecting mirror,

# Field damping by field reservoirs

- the equation of motion for the reduced density operator  $\text{Tr}_R[\hat{\rho}(t)] \equiv \hat{\rho}_f(t)$  is,

$$\frac{d}{dt}\hat{\rho}_f(t) = -\frac{1}{2}\left(\frac{\omega}{Q_e}\right)\{n_{th}[\hat{a}\hat{a}^\dagger\hat{\rho}_f(t') - \hat{a}^\dagger\hat{\rho}_f(t')\hat{a}] + (n_{th} + 1)[\hat{a}^\dagger\hat{a}\hat{\rho}_f(t') - \hat{a}\hat{\rho}_f(t')\hat{a}^\dagger] + \text{H. C.},$$

- compared to the case of atom damping by field reservoirs,

$$\frac{d}{dt}\hat{\rho}_a(t) = -\frac{1}{2}\Gamma\{n_{th}[\hat{\sigma}_-\hat{\sigma}_+\hat{\rho}_a - \hat{\sigma}_+\hat{\rho}_a\hat{\sigma}_-] + (n_{th} + 1)[\hat{\sigma}_+\hat{\sigma}_-\hat{\rho}_a - \hat{\sigma}_-\hat{\rho}_a\hat{\sigma}_+]\} + \text{H. C.},$$

# Field damping by atomic reservoirs

- ➔ consider the damping of an optical cavity mode by a two-level atomic beam reservoir,
- ➔ this is the reverse problem of a laser,
- ➔ the statistics of the atomic reservoir is determined by the Boltzmann distribution,

$$\hat{\rho}_{R=atom}(t=0) = \begin{pmatrix} \rho_{aa} & 0 \\ 0 & \rho_{bb} \end{pmatrix} = \rho_{aa}|a\rangle\langle a| + \rho_{bb}|b\rangle\langle b|,$$

where

$$\rho_{aa} = \frac{1}{1 + \exp(\hbar\omega_0/k_Bt)}, \quad \text{and} \quad \rho_{bb} = \frac{\exp(\hbar\omega_0/k_Bt)}{1 + \exp(\hbar\omega_0/k_Bt)},$$

- ➔ assume there is no quantum coherence between the upper and lower states,  
 $\rho_{ab} = \rho_{ba} = 0,$



# Field damping by atomic reservoirs

- the interaction Hamiltonian for a single atom is

$$\hat{H}_I = \hbar g(\hat{\sigma}_- \hat{a}^\dagger + \hat{\sigma}_+ \hat{a}) = \hbar g \begin{pmatrix} 0 & \hat{a} \\ \hat{a}^\dagger & 0 \end{pmatrix},$$

- at  $t = 0$ , the atom-field density operator is,

$$\hat{\rho}(t) = \hat{\rho}_f(t) \otimes \hat{\rho}_R = \begin{pmatrix} \rho_{aa} \hat{\rho}_f(t) & 0 \\ 0 & \rho_{bb} \hat{\rho}_f(t) \end{pmatrix},$$

- the terms for the commutator are

$$[\hat{H}_I, [\hat{H}_I, \hat{\rho}(t)]] = \hbar g^2 \begin{pmatrix} \hat{a} \hat{a}^\dagger \rho_{aa} \hat{\rho}_f(t) - \hat{a} \rho_{bb} \hat{\rho}_f(t) \hat{a}^\dagger & 0 \\ 0 & \hat{a}^\dagger \hat{a} \rho_{bb} \hat{\rho}_f(t) - \hat{a}^\dagger \rho_{aa} \hat{\rho}_f(t) \hat{a} \end{pmatrix} + \text{H. C.},$$

# Field damping by atomic reservoirs

- the equation of motion for the reduced density operator  $\text{Tr}_R[\hat{\rho}(t)] \equiv \hat{\rho}_f(t)$  is,

$$\frac{d}{dt}\hat{\rho}_f(t) = \left(\frac{1}{i\hbar}\right)^2 \int_0^t dt' \text{Tr}_R([\hat{H}_I(t), [\hat{H}_I(t'), \hat{\rho}_f(t) \otimes \hat{\rho}_R(0)]]),$$

- assume that  $r$  atoms are injected into the cavity per second,  
→ and they spend an average time of  $\tau$  seconds inside the cavity, i.e.

$$\int_0^\tau dt' rt' = \frac{1}{2}r\tau^2,$$

then,

$$\frac{d}{dt}\hat{\rho}_f(t) = -\frac{1}{2}R_e[\hat{a}\hat{a}^\dagger\hat{\rho}_f - \hat{a}^\dagger\hat{\rho}_f\hat{a}] - \frac{1}{2}R_g[\hat{a}^\dagger\hat{a}\hat{\rho}_f - \hat{a}\hat{\rho}_f\hat{a}^\dagger] + \text{H.C.},$$

where

$$R_e = r\rho_{aa}g^2\tau^2, \quad \text{and} \quad R_g = r\rho_{bb}g^2\tau^2,$$

# Field damping by atomic reservoirs

- the equation of motion for the reduced density operator  $\text{Tr}_R[\hat{\rho}(t)] \equiv \hat{\rho}_f(t)$  is,

$$\frac{d}{dt}\hat{\rho}_f(t) = -\frac{1}{2}R_e[\hat{a}\hat{a}^\dagger\hat{\rho}_f - \hat{a}^\dagger\hat{\rho}_f\hat{a}] - \frac{1}{2}R_g[\hat{a}^\dagger\hat{a}\hat{\rho}_f - \hat{a}\hat{\rho}_f\hat{a}^\dagger] + \text{H.C.},$$

where

$$R_e = r\rho_{aa}g^2\tau^2, \quad \text{and} \quad R_g = r\rho_{gg}g^2\tau^2,$$

- $R_e$  is the rate coefficient for photon emission by atoms per second,
- $R_g$  is the rate coefficient for photon absorption by atoms per second,
- the cavity photon decay rate  $\frac{\nu}{Q_0}$  and the thermal equilibrium photon number  $n_{th}$  are defined by

$$\frac{\nu}{Q_0} \equiv R_g - R_e, \quad \text{and} \quad R_e(1 + n_{th}) = R_g n_{th} \rightarrow n_{th} = \frac{R_e}{R_g - R_e} = \frac{1}{\exp(\hbar\omega/k_b T) - 1},$$

- the later one condition gives the thermal equilibrium photon number  $n_{th}$ ,

- $R_e(1 + n_{th})$  is the sum of *spontaneous* and *stimulated* emission rate per second,

- $R_g n_{th}$  is (stimulated) *absorption* rate per second,

# Field damping by atomic reservoirs

→ the equation of motion for the reduced density operator  $\text{Tr}_R[\hat{\rho}(t)] \equiv \hat{\rho}_f(t)$  is,

$$\begin{aligned}\frac{d}{dt}\hat{\rho}_f(t) &= -\frac{1}{2}R_e[\hat{a}\hat{a}^\dagger\hat{\rho}_f - \hat{a}^\dagger\hat{\rho}_f\hat{a}] - \frac{1}{2}R_g[\hat{a}^\dagger\hat{a}\hat{\rho}_f - \hat{a}\hat{\rho}_f(t)\hat{a}^\dagger] + \text{H.C.}, \\ &= -\frac{1}{2}\frac{\nu}{Q_0}\{n_{th}[\hat{a}\hat{a}^\dagger\hat{\rho}_f - \hat{a}^\dagger\hat{\rho}_f\hat{a}] + (n_{th} + 1)[\hat{a}^\dagger\hat{a}\hat{\rho}_f - \hat{a}\hat{\rho}_f(t)\hat{a}^\dagger]\} + \text{H.C.},\end{aligned}$$

where we use that  $R_e = \frac{\nu}{Q_0}n_{th}$  and  $R_g = \frac{\nu}{Q_0}(n_{th} + 1)$ ,

→ the diagonal elements of the reduced density matrix are

$$\begin{aligned}\frac{d}{dt}\rho_{n,n}(t) &= -\frac{\nu}{Q_0}\{[n_{th}(n+1) - (n_{th} + 1)n]\rho_{n,n} \\ &\quad - n_{th}\rho_{n-1,n-1} - (n_{th} + 1)(n+1)\rho_{n+1,n+1}\}, \\ &= [-R_e(n+1) - R_g n]\rho_{nn} + R_e n\rho_{n-1,n-1} + R_g(n+1)\rho_{n+1,n+1},\end{aligned}$$

# Detailed balance

- equilibrium is obtained when the net flow between all pairs of levels vanishes,

$$R_g n \rho_{n,n} = R_e n \rho_{n-1,n-1}, \quad \text{or} \quad \rho_{n,n} = \frac{n_{th}}{n_{th} + 1} \rho_{n-1,n-1},$$

- this condition is referred to as *detailed balance*,
- the solution for detailed balance is,

$$\rho_{n,n} = [1 - \exp(-\hbar\omega/k_B T)] \exp(-n\hbar\omega/k_B T),$$

with  $n_{th} = \frac{1}{\exp(\hbar\omega/k_B T) - 1}$ ,

- detailed balance in this case gives the *thermal (Bose-Einstein)* distribution with an average photon number,

$$\langle n \rangle = \sum_n \rho_{n,n} n = \frac{1}{\exp(\hbar\omega/k_B T) - 1} = n_{th}$$

# Detailed balance

- ➔ detailed balance in this case gives the *thermal (Bose-Einstein)* distribution with an average photon number,

$$\langle n \rangle = \sum_n \rho_{n,n} n = \frac{1}{\exp(\hbar\omega/k_B T) - 1} = n_{th}$$

- ➔ this is the result we use for the thermal radiation field,
- ➔ although the field  $\hat{\rho}_f$  may initially be in a pure state, the process of tracing over the (unobserved) atomic states leads to a field in a mixed state,  $\hat{\rho}_f = \sum_n \rho_{n,n} |n\rangle\langle n|$ ,
- ➔ the effect of the atomic beam is to bring the field to the *same temperature* as that of atoms,

# Reservoir, decoherence, and measurement

- the reservoir theory lies in the process of tracing over the reservoir coordinates,
- which induces *dissipation* and *decoherence* of the system,
- at the same time, this is an *irreversible* dynamics for the system,
- this process corresponds to the lack of measurement as to whether the atom is in the upper level or in the lower level after interaction with the field,
- if the initial and final states of the atom are known, i.e. if the information concerning the atomic beam is not discarded, the field remains in a *pure state*,
- the primary difference between the reservoir and quantum measurement theories is whether information stored in the environment (reservoir) that interacts with system is discarded or read out.