Spectral Properties of Localized Exciton in Deformable Lattice

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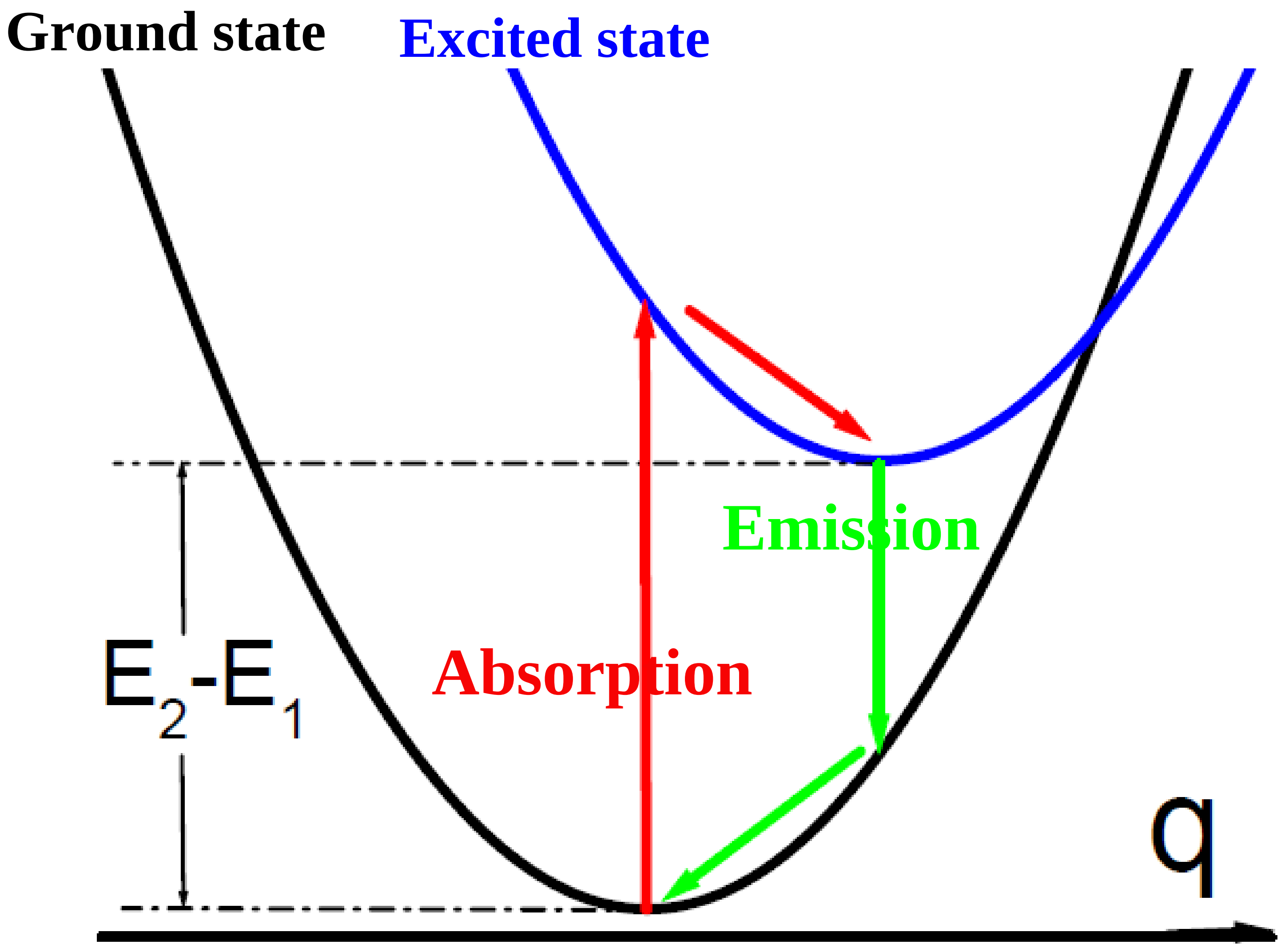
Outline

1. **Motivation.**
2. **Our method: How to improve the perturbation treatment.**
3. **Equilibrium dynamics, optical absorption**
4. **Non-equilibrium dynamics, occupation amplitude of the excited state.**
5. **A brief summary.**

Quantum transition and lattice relaxation

An old problem



Quantum transition and lattice relaxation

$$H = (E_2 + c_2 q)B_2^+ B_2 + (E_1 + c_1 q)B_1^+ B_1 + \frac{p^2}{2m} + \frac{1}{2} m\omega_0^2 q^2$$

$$+ \sum_k \frac{g_k}{2}(b_k^+ + b_k)\left(B_2^+ B_1 + B_1^+ B_2\right) + \sum_k \omega_k b_k^+ b_k$$

In solid, *q* is some configuration

Photo-induced molecular structure trans.

The excited state may be of a longer life time

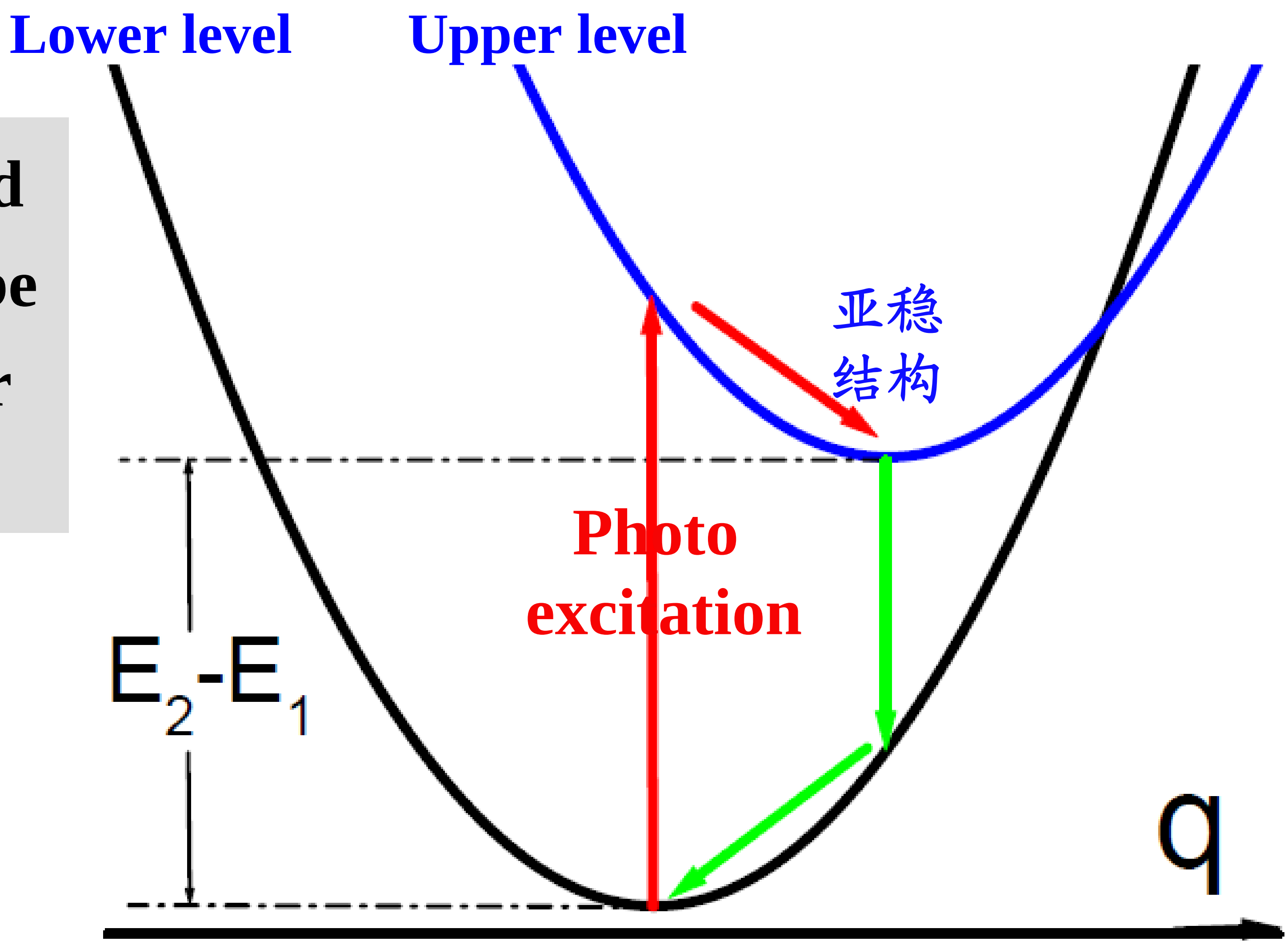


Photo-induced molecular structure trans.

$$H = (E_2 + c_2 q)B_2^+ B_2 + (E_1 + c_1 q)B_1^+ B_1 + \frac{p^2}{2m} + \frac{1}{2} m\omega_0^2 q^2$$

$$+ \sum_k \frac{g_k}{2}(b_k^+ + b_k)\left(B_2^+ B_1 + B_1^+ B_2\right) + \sum_k \omega_k b_k^+ b_k$$

Two-level molecule with structure-dependent level

Photo-induced structure transition, photoisomerization

Local exciton of cold atom in optic lattice

$$H=(E_2+c_2q)B_2^+B_2+(E_1+c_1q)B_1^+B_1+\frac{p^2}{2m}+\frac{1}{2}m\omega_0^2q^2$$

$$+\sum_k\frac{g_k}{2}(b_k^++b_k)\left(B_2^+B_1+B_1^+B_2\right)+\sum_k\omega_k b_k^+b_k$$

q **is for the harmonic trapping potential.**
Properties of excited state may by controlled by the trapping potential.

c_1, c_2, ω_0 **may be adjusted by the laser**

The model

$$H=(E_2+c_2q)B_2^+B_2+(E_1+c_1q)B_1^+B_1+\frac{p^2}{2m}+\frac{1}{2}m\omega_0^2q^2$$

$$+\sum_k\frac{g_k}{2}(b_k^++b_k)\left(B_2^+B_1+B_1^+B_2\right)+\sum_k\omega_kb_k^+b_k$$

$$=\frac{\Delta}{2}\sigma_z+\frac{g_0}{2}(a^++a)\sigma_z+\omega_0a^+a+\sum_k\frac{g_k}{2}(b_k^++b_k)\sigma_x+\sum_k\omega_kb_k^+b_k$$

$$\Delta=E_2-E_1,\quad \sigma_z=B_2^+B_2-B_1^+B_1,\quad \sigma_z=B_2^+B_1+B_1^+B_2$$

$$g_0=(c_2-c_1)\sqrt{\frac{1}{2m\omega_0}}\qquad q=\sqrt{\frac{1}{2m\omega_0}}(a^++a)\qquad p=i\sqrt{\frac{m\omega_0}{2}}(a^+-a)$$

What we calculate

$$H = \frac{\Delta}{2}\sigma_z + \frac{g}{2}(a^+ + a)\sigma_z + \omega_0 a^+ a + \sum_k \frac{g_k}{2}(b_k^+ + b_k)\sigma_x + \sum_k \omega_k b_k^+ b_k$$

Optical absorption:

$$\sigma(\omega) = \int d\omega e^{i\omega t} \frac{1}{2} \mathrm{Tr}\left\{ e^{-\beta H} \left[\sigma_x(t)\sigma_x - \sigma_x\sigma_x(t) \right] \right\} / Z$$

Initial state (t=0) is an excited state

$$x(t) = \langle \psi(0) | e^{-iHt} | \psi(0) \rangle$$

$$\sigma_z |\psi(0)\rangle = |\psi(0)\rangle$$

Unitary transformation

$$H = \frac{\Delta}{2}\sigma_z + \frac{g}{2}(a^+ + a)\sigma_z + \omega_0 a^+ a + \sum_k \frac{g_k}{2}(b_k^+ + b_k)\sigma_x + \sum_k \omega_k b_k^+ b_k$$

$$H' = \exp(S_1) H \exp(-S_1)$$

$$= \frac{\Delta}{2}\sigma_z + \omega_0 a^+ a - \frac{g^2}{4\omega_0} + \left(\sigma_- e^{-X} + \sigma_+ e^{X}\right)\sum_k \frac{g_k}{2}(b_k^+ + b_k) + \sum_k \omega_k b_k^+ b_k$$

$$S_1 = \frac{g}{2\omega_0}\sigma_z (a^+ - a)$$

$$X = \frac{g}{\omega_0}(a^+ - a)$$

Unitary transformation

$$H'' = \exp(S_2)H'\exp(-S_2) = H_0'' + H_1'' + H_2''$$

$$S_2 = \left(\sigma_- e^{-X} + \sigma_+ e^{X}\right)\sum_k \frac{g_k}{2\omega_k}\xi_k (b_k^+ - b_k)$$

$$\xi_k = \frac{\omega_k}{\omega_k + \eta\Delta}$$

$$H_0'' = \frac{\eta\Delta}{2}\sigma_z + \omega_0 a^+ a + \sum_k \omega_k b_k^+ b_k - \frac{g^2}{4\omega_0} - \sum_k \frac{g_k^2}{4\omega_k}(2\xi_k - \xi_k^2)$$

Level shift:

$$\eta = \exp\left(-\sum_k \frac{g_k^2}{2\omega_k^2}\xi_k^2\right)$$

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Unitary transformation

$$H'' = \exp(S_2)H'\exp(-S_2) = H_0'' + H_1'' + H_2''$$

$$S_2 = \left(\sigma_- e^{-X} + \sigma_+ e^{X}\right)\sum_k \frac{g_k}{2\omega_k}\xi_k(b_k^+ - b_k)$$

$$\xi_k = \frac{\omega_k}{\omega_k + \eta\Delta}$$

Single-phonon transition:

$$H_1'' = \sum_k V_k\left\{b_k^+\sigma_- e^{-X} + b_k\sigma_+ e^{X}\right\}$$

$$V_k = \eta\Delta\frac{g_k\xi_k}{\omega_k}$$

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Unitary transformation

$$H'' = \exp(S_2)H'\exp(-S_2) = H_0'' + H_1'' + H_2''$$

$$H_2'' = H'' - H_0'' - H_1'' = O(g_k^2)$$

H_2'' contain the terms of multi-boson transition

and will be omitted.

$$H'' \approx H_0'' + H_1''$$

Purpose of transformation

$$H \Rightarrow H'' = \exp(S_2)\exp(S_1) H \exp(-S_1)\exp(-S_2)$$

unitarity

$$S^{+} = -S$$

S is time-independent, the Schroedinger equation

$$i\frac{d}{dt}|\psi(t)\rangle = H|\psi(t)\rangle \Rightarrow i\frac{d}{dt}|\psi''(t)\rangle = H''|\psi''(t)\rangle$$

$$|\psi(t)\rangle = \exp(-S_1)\exp(-S_2)|\psi''(t)\rangle$$

Time correlation

$$\mathrm{Tr}\left(e^{-\beta H} e^{iHt} A e^{-iHt} B\right) = \mathrm{Tr}\left(e^{-\beta H''} e^{iH''t} A'' e^{-iH''t} B''\right)$$

Approximation we use

$$H'' \approx H_0'' + H_1''$$

$$H_0'' = \frac{\eta\Delta}{2}\sigma_z + \omega_0 a^+ a + \sum_k \omega_k b_k^+ b_k - \frac{g^2}{4\omega_0} - \sum_k \frac{g_k^2}{4\omega_k}(2\xi_k - \xi_k^2)$$

$$H_1'' = \sum_k V_k \left\{ b_k^+ \sigma_- e^{-X} + b_k \sigma_+ e^{X} \right\}$$

g_0 to all orders

g_k to second order

All three-body transition terms are omitted

$$g_0 g_k a^+ b_k^+ \sigma_- e^{-X}, \quad g_0 g_k a b_k^+ \sigma_- e^{-X}$$

$$g_0 g_k a^+ b_k \sigma_+ e^{X}, \quad g_0 g_k a^+ b_k \sigma_+ e^{X} \approx 0$$

Optical absorption

$$\sigma(\omega) = \int d\omega e^{i\omega t} \frac{1}{2} \mathrm{Tr}\left\{ \mathrm{e}^{-\beta \mathrm{H}} \left[\sigma_x(t)\sigma_x - \sigma_x \sigma_x(t) \right] \right\} / Z$$

$$G(t) = -i\theta(t) \left\langle \sigma_x(t)\sigma_x - \sigma_x \sigma_x(t) \right\rangle_H$$

$\langle \ldots \rangle_H$ **means an average over equilibrium distribution**

density operator $\exp(-\beta H)$

Optical absorption

After unitary transformations

$$G(t) = -i\theta(t)\left\langle\left[e^{iH''t}\sigma_+ e^{X} e^{-iH''t}, \sigma_- e^{-X}\right]\right\rangle_{H''}$$

$$-\, i\theta(t)\left\langle\left[e^{iH''t}\sigma_- e^{-X} e^{-iH''t}, \sigma_+ e^{X}\right]\right\rangle_{H''}$$

$$H'' \approx H_0'' + H_1''$$

Optical absorption

Approximate decoupling

$$-i\theta(t)\left\langle\left[e^{iH''t}\sigma_{-}e^{-X}e^{-iH''t},\sigma_{+}e^{X}\right]\right\rangle_{H''}$$

$$\approx -i\theta(t)\left\langle\left[e^{iH''t}\sigma_{-}e^{-iH''t},\sigma_{+}\right]\right\rangle_{H''}\left\langle 0_a\left|e^{-X(t)}e^{X}\right|0_a\right\rangle$$

$$\left\langle 0_a\left|e^{-X(t)}e^{X}\right|0_a\right\rangle = e^{-\lambda}\exp\left(\lambda e^{-i\omega_0 t}\right)$$

$$\lambda = \frac{g^2}{\omega_0^2}$$

Optical absorption

$$-i\theta(t)\left\langle\left[e^{iH''t}\sigma_{-}e^{-iH''t},\sigma_{+}\right]\right\rangle_{H''}$$

$$=\frac{1}{2\pi}\int_0^{\infty}d\omega\frac{\gamma(\omega)e^{-i\omega t}}{[\omega-\eta\Delta-R(\omega)]^2+\gamma^2(\omega)}$$

$$R(\omega)=\sum_k\frac{V_k^2}{\omega-\omega_k}$$

$$\gamma(\omega)=\pi\sum_k V_k^2\delta(\omega-\omega_k)$$

Optical absorption

$$\lambda = \frac{g^2}{\omega_0^2}$$

$$\sigma(\omega) = \frac{e^{-\lambda}}{\pi}\sum_{l=0}\frac{\lambda^l}{l!}\frac{\gamma(\omega - l\omega_0)}{[\omega - l\omega_0 - \eta\Delta - R(\omega - l\omega_0)]^2 + \gamma^2(\omega - l\omega_0)}$$

When $\lambda = 0$

Optical absorption of two-level atom

$$\sigma(\omega) = \frac{1}{\pi}\frac{\gamma(\omega)}{[\omega - \eta\Delta - R(\omega)]^2 + \gamma^2(\omega)}$$

When $\alpha = 0$

Optical absorption of independent boson

$$\sigma(\omega) = e^{-\lambda}\sum_{l=0}\frac{\lambda^l}{l!}\delta(\omega - l\omega_0 - \Delta)$$

Optical absorption

$$\lambda = \frac{g^2}{\omega_0^2}$$

$$\sigma(\omega) = \frac{e^{-\lambda}}{\pi}\sum_{l=0}\frac{\lambda^l}{l!}\frac{\gamma(\omega - l\omega_0)}{[\omega - l\omega_0 - \eta\Delta - R(\omega - l\omega_0)]^2 + \gamma^2(\omega - l\omega_0)}$$

吸收光子能量 ω:

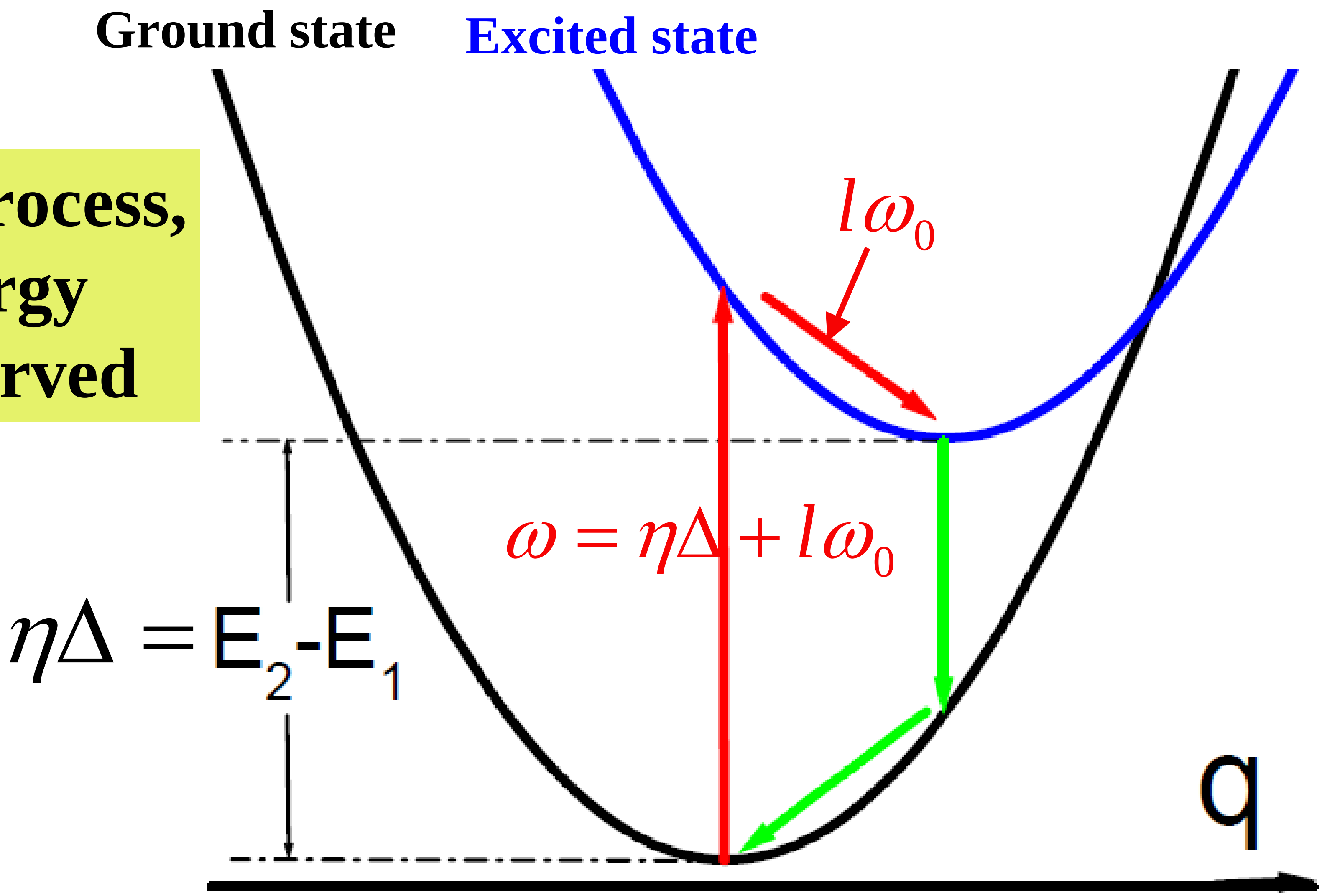
$$\omega = \eta\Delta + l\omega_0$$

$$\gamma(\omega - l\omega_0) = \gamma(\eta\Delta)$$

不同数目的多声子跃迁过程各自贡献的吸收谱的非相干相加

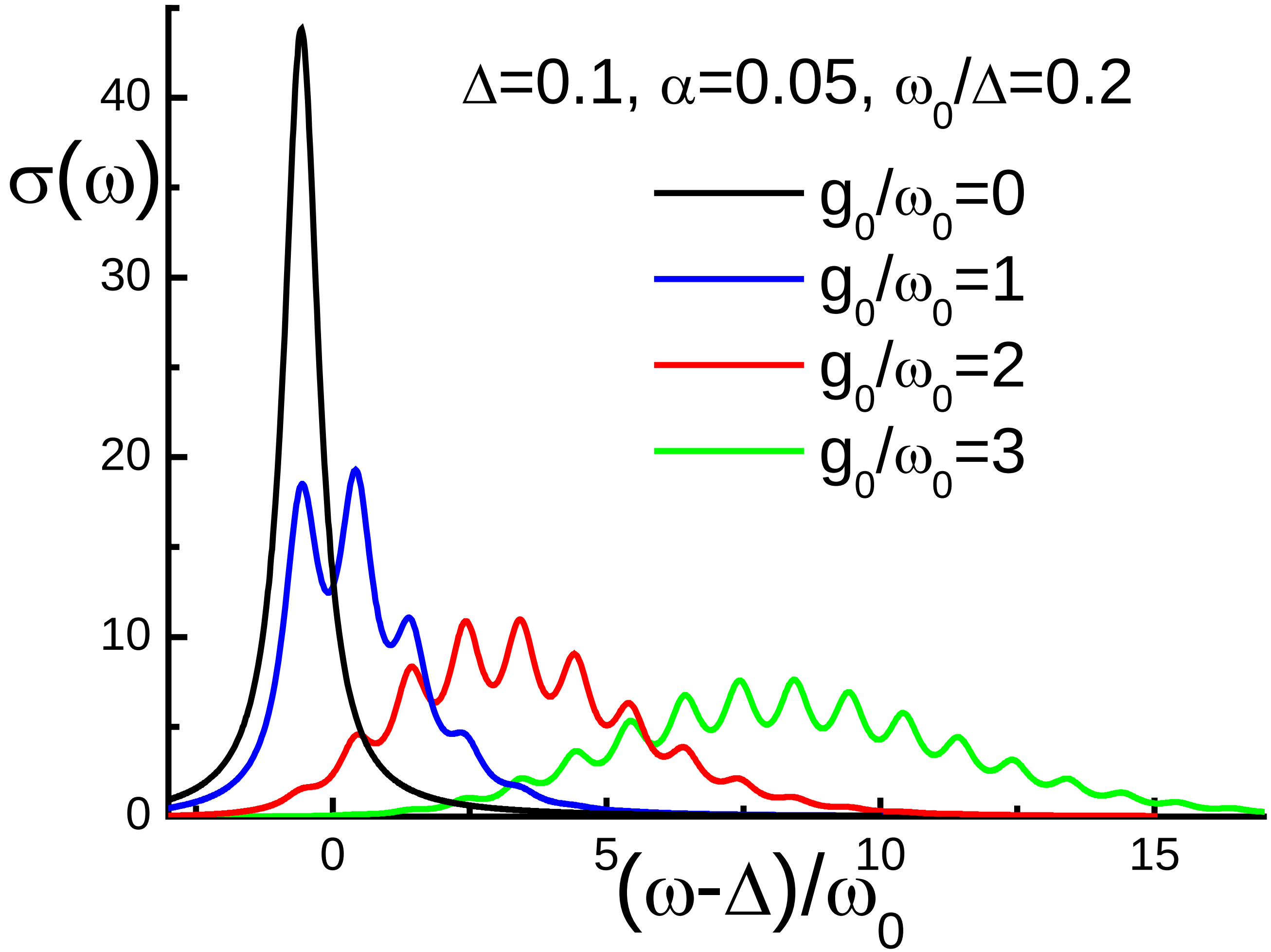
Optical absorption

Real process, energy conserved



Optical absorption

不同数目的多声子跃迁过程吸收谱非相干相加



Solution in interaction pic.

Interaction picture

$$i\frac{d}{dt}\left|\psi_I''(t)\right\rangle = H_1''(t)\left|\psi_I''(t)\right\rangle$$

$$H_1''(t) = \sum_k V_k \left\{ b_k^+ \sigma_- e^{i(\omega_k-\eta\Delta)t} e^{-X(t)} + b_k \sigma_+ e^{-i(\omega_k-\eta\Delta)t} e^{X(t)} \right\}$$

Perturbation to 2nd order in g_k

$$\left|\psi_I''(t)\right\rangle = \left|\psi_I''(0)\right\rangle - i\int_0^t dt' H_1''(t')\left|\psi_I''(0)\right\rangle$$
$$-\int_0^t dt_1 H_1''(t_1)\int_0^{t_1} dt_2 H_1''(t_2)\left|\psi_I''(t_2)\right\rangle$$

The initial state

$$\left|\psi_I''(0)\right\rangle = \left|\uparrow, 0_a, 0_k\right\rangle$$

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Solution in interaction pic.

Survival amplitude:

$$x(t) = \langle \psi_I''(0) | \psi_I''(t) \rangle$$

$$x(t) = 1 - \int_0^t dt_1 \int_0^{t_1} dt_2 \sum_k V_k^2 e^{-i(\omega_k - \eta\Delta)(t_1 - t_2)} \times \langle o_a | e^{X(t_1)} e^{-X(t_2)} | 0_a \rangle x(t_2)$$

$$x(t) = \frac{1}{2\pi i} \int_B \frac{e^{p\tau} dp}{p + e^{-\lambda} \sum_l \frac{\lambda^l}{l!} \sum_k \frac{V_k^2}{p + i(\omega_k - \eta\Delta + l\omega_0)}}$$

Occupation amplitude of excited st.

$$x(t)=\frac{i}{2\pi}e^{i\eta\Delta}\int_{-\infty}^{\infty}\frac{e^{-i\omega t}d\omega}{\omega-\eta\Delta-e^{-\lambda}\sum_{l}\frac{\lambda^{l}}{l!}[R(\omega-l\omega_0)-i\gamma(\omega-l\omega_0)]}$$

$$R(\omega)=\sum_k\frac{V_k^2}{\omega-\omega_k}$$

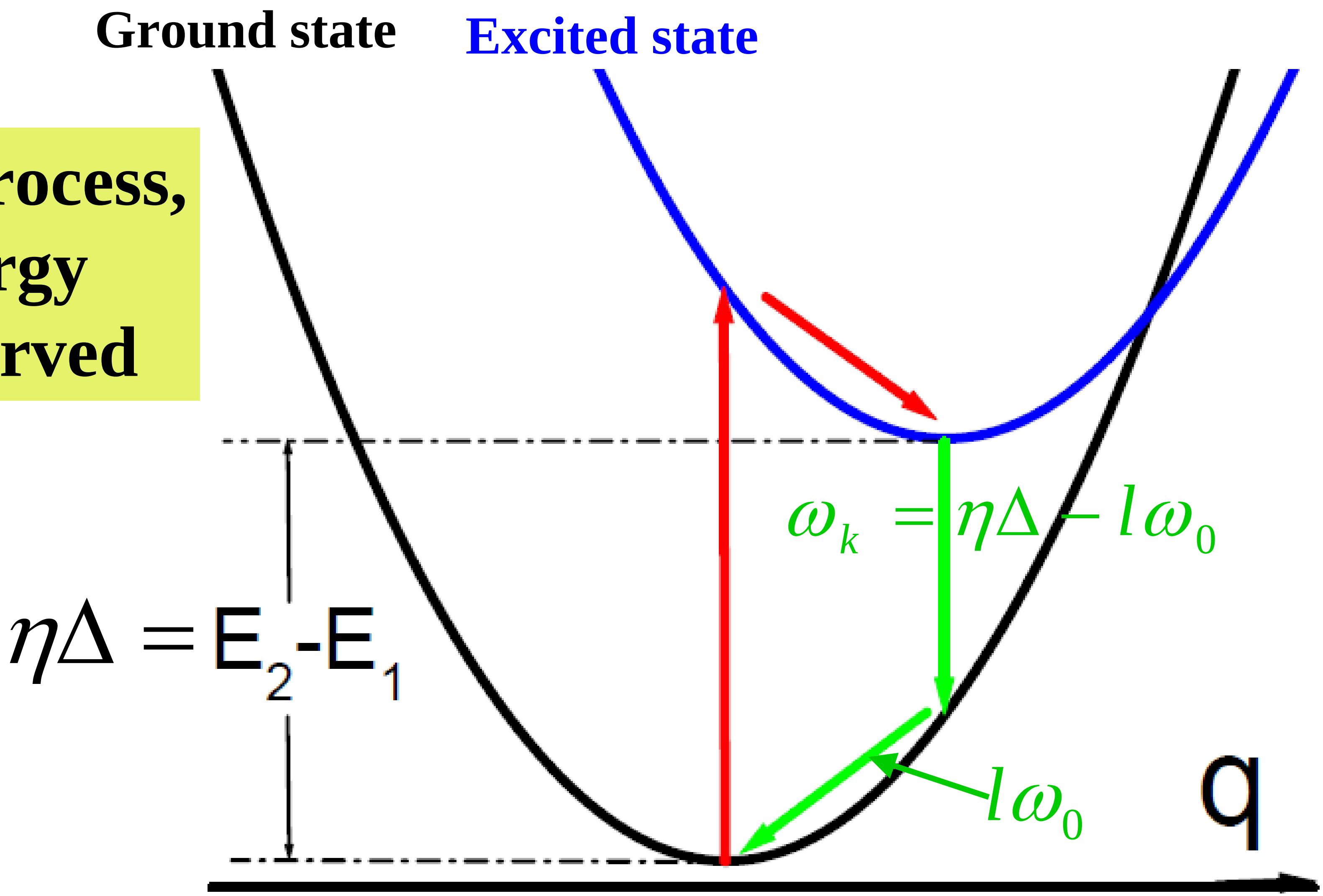
$$\gamma(\omega)=\pi\sum_k V_k^2\delta(\omega-\omega_k)$$

$$\lambda=\frac{g^2}{\omega_0^2}$$

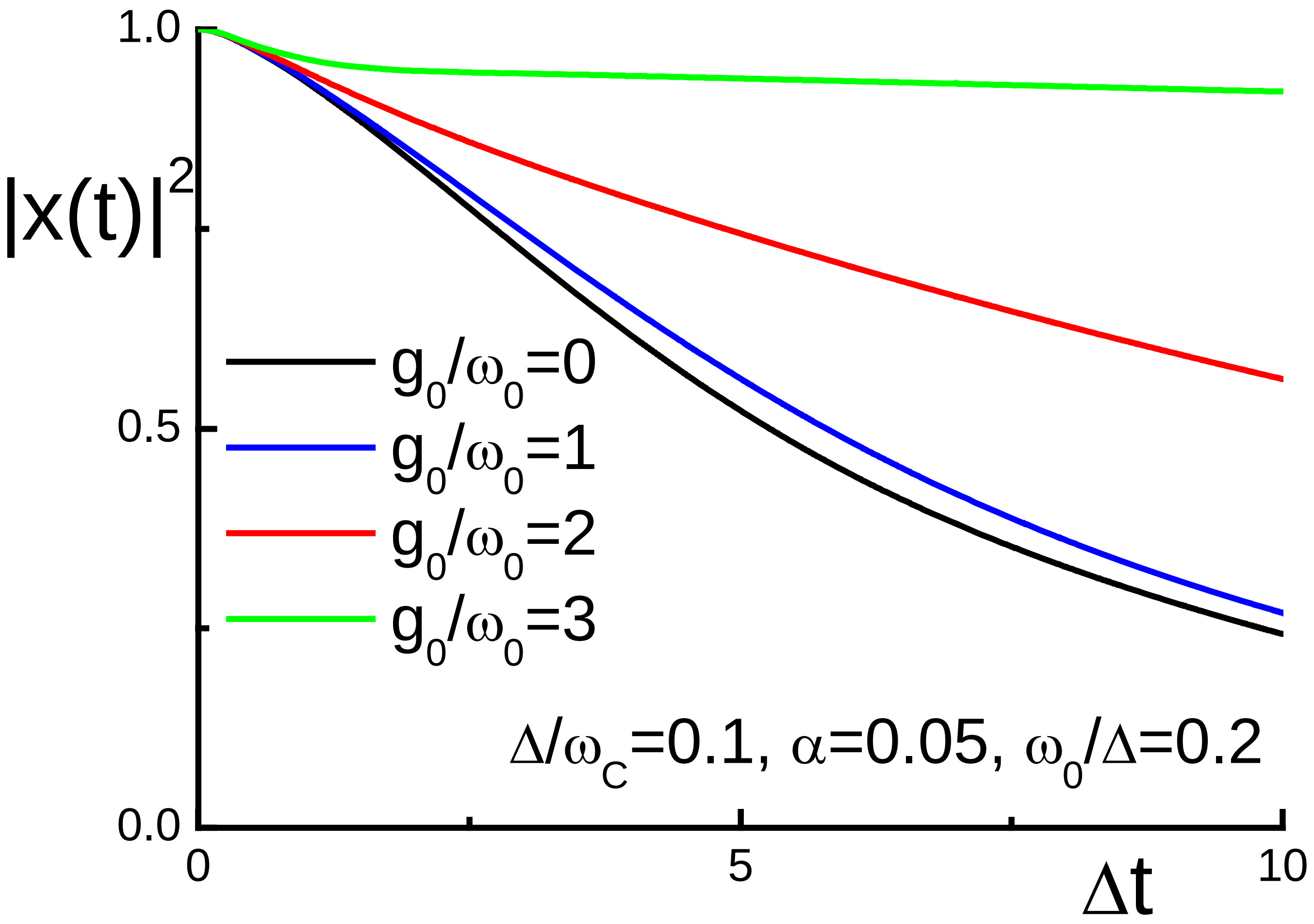
不同数目的多声子跃迁过程对振幅的贡献的相干相加

Occupation amplitude of excited st.

Real process, energy conserved



Occupation amplitude of excited st.



Occupation amplitude of excited st.

$$x(t) = 1 - \int_0^t dt_1 \int_0^{t_1} dt_2 \sum_k V_k^2 e^{-i(\omega_k - \eta\Delta)(t_1 - t_2)}$$

$$\times \langle o_a | e^{X(t_1)} e^{-X(t_2)} | 0_a \rangle x(t_2)$$

$x(t_2) \approx 1$ **for short time**

$$x(t) = 1 - \sum_k V_k^2 e^{-\lambda} \sum_l \frac{\lambda^l}{l!} \frac{2\sin^2\left[(\omega_k - \eta\Delta + l\omega_0)t/2\right]}{(\omega_k - \eta\Delta + l\omega_0)^2}$$

Occupation amplitude of excited st.

$$x(t) = 1 - \sum_k V_k^2 e^{-\lambda} \sum_l \frac{\lambda^l}{l!} \frac{2\sin^2\left[(\omega_k - \eta\Delta + l\omega_0)t/2\right]}{(\omega_k - \eta\Delta + l\omega_0)^2}$$

$$|x(t)|^2 = \exp[-\gamma(t)t]$$

The decay rate is modified by the lattice

$$\gamma(t) = 2\pi \sum_k V_k^2 e^{-\lambda} \sum_l \frac{\lambda^l}{l!} \frac{2\sin^2\left[(\omega_k - \eta\Delta + l\omega_0)t/2\right]}{\pi t(\omega_k - \eta\Delta + l\omega_0)^2}$$

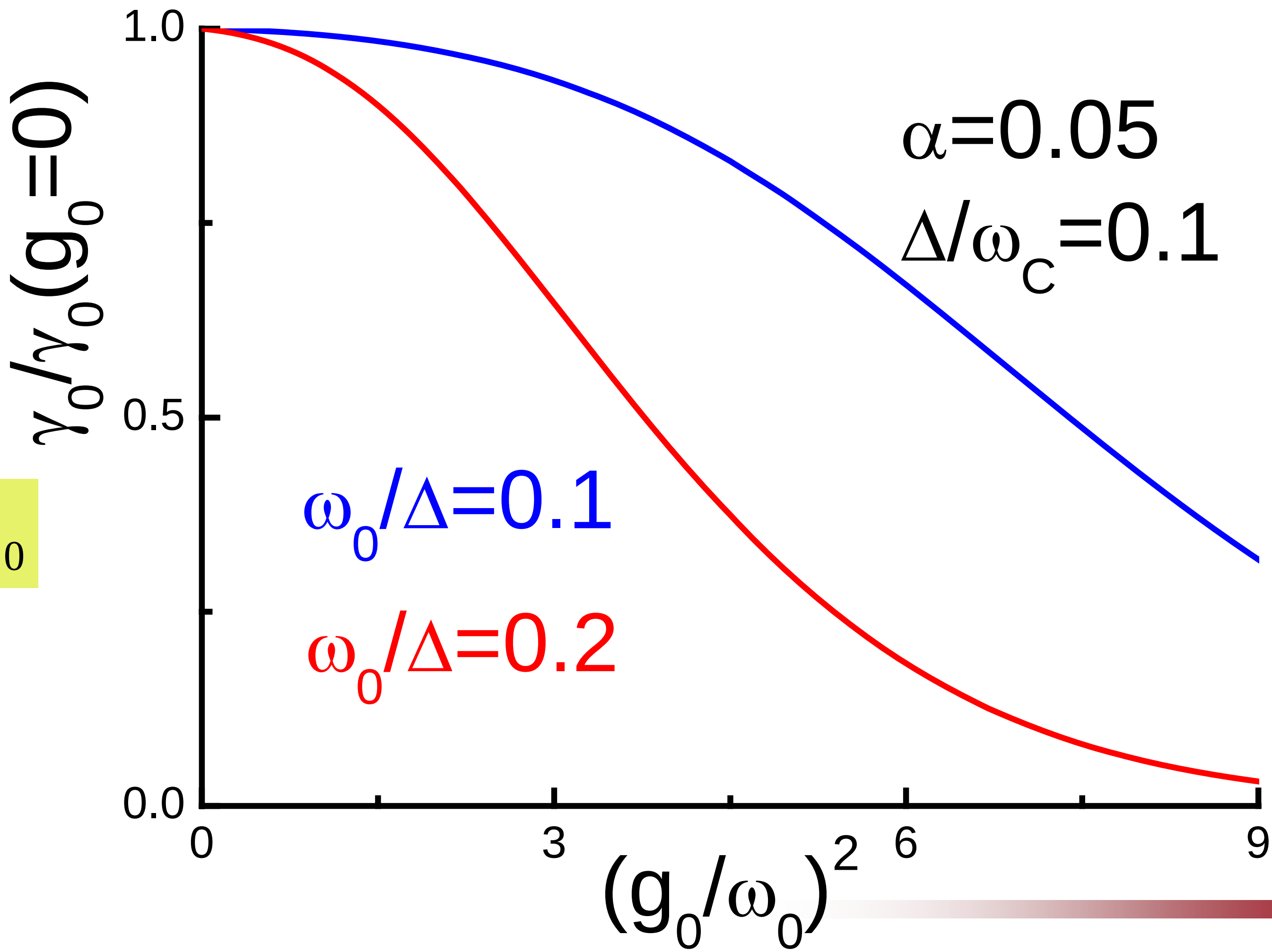
$$\gamma_0 = \gamma(t \to \infty) = 2\pi \sum_k V_k^2 e^{-\lambda} \sum_l \frac{\lambda^l}{l!} \delta(\omega_k - \eta\Delta + l\omega_0)$$

Occupation amplitude of excited st.

$$\gamma_0 = 2\pi \sum_k V_k^2 e^{-\lambda} \sum_l \frac{\lambda^l}{l!} \delta(\omega_k - \eta\Delta + l\omega_0)$$

实过程：

$$\omega_k = \eta\Delta - l\omega_0$$



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Summary

1. **A simple analytical approach is proposed.**

2. **Equilibrium dynamics, optical absorption $\sigma(\omega)$, which is an incoherent superposition of absorption spectra for all multi-phonon process.**

3. **Non-equilibrium dynamics, occupation amplitude x(t) and time-dependent decay γ(t), which are coherent superposition of all multi-phonon process with energy conserved.**

Thanks !