Noisy metamolecule: strong narrowing of fluorescence line

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Collaborators:

- E. S. Andrianov
- A. A. Pukhov

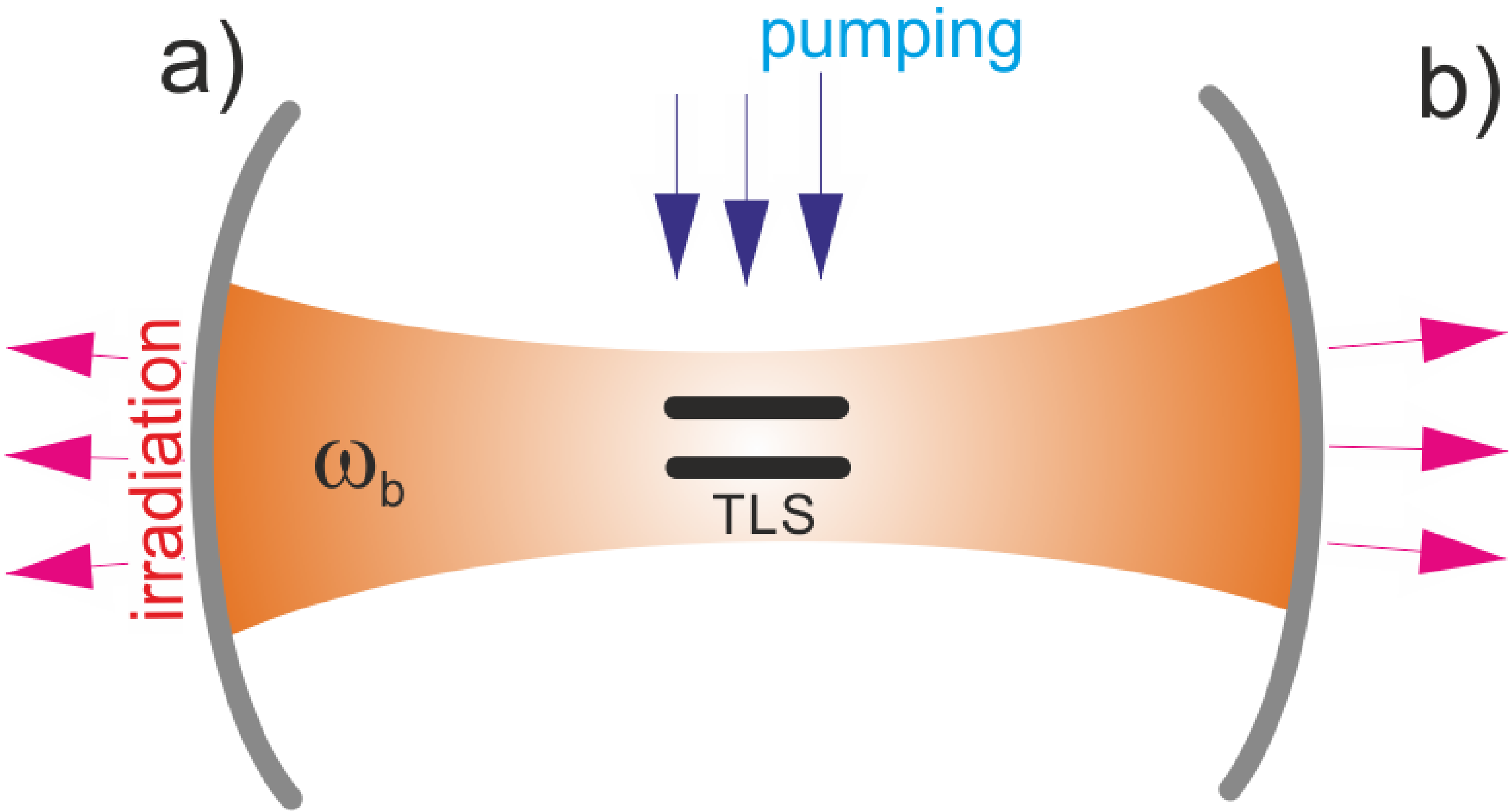
Acknowledgments:

Юрий Ефремович Лозовик

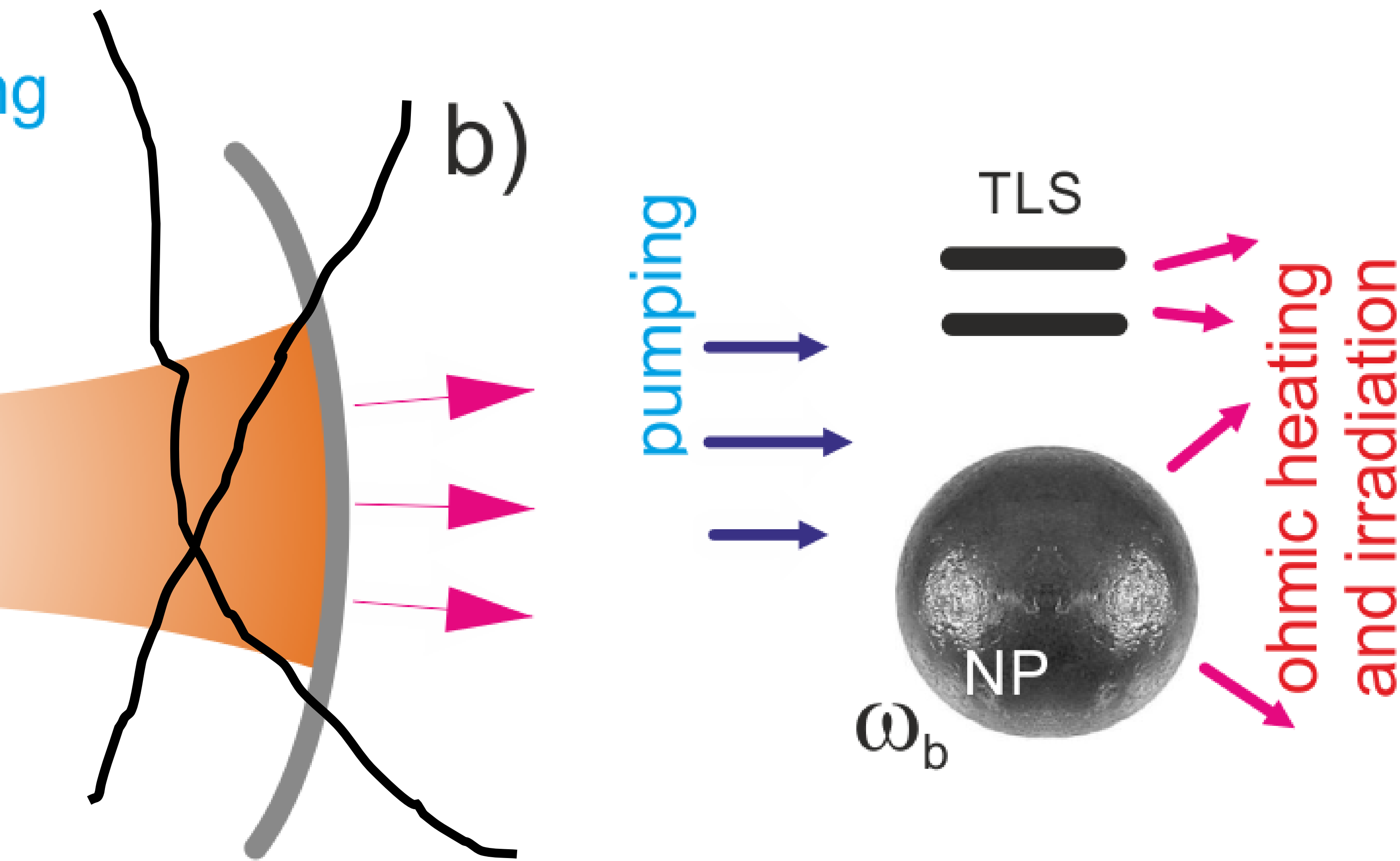

Нанолазер...

D. J. Bergman and M. I. Stockman,
“Surface Plas-
mon Amplification by Stimulated Emission of
Radiation: Quantum Generation of Coherent Surface
Plasmons in Nanosystems,”
Phys. Rev. Lett. 90, 027,402 (2003).

нанолазер

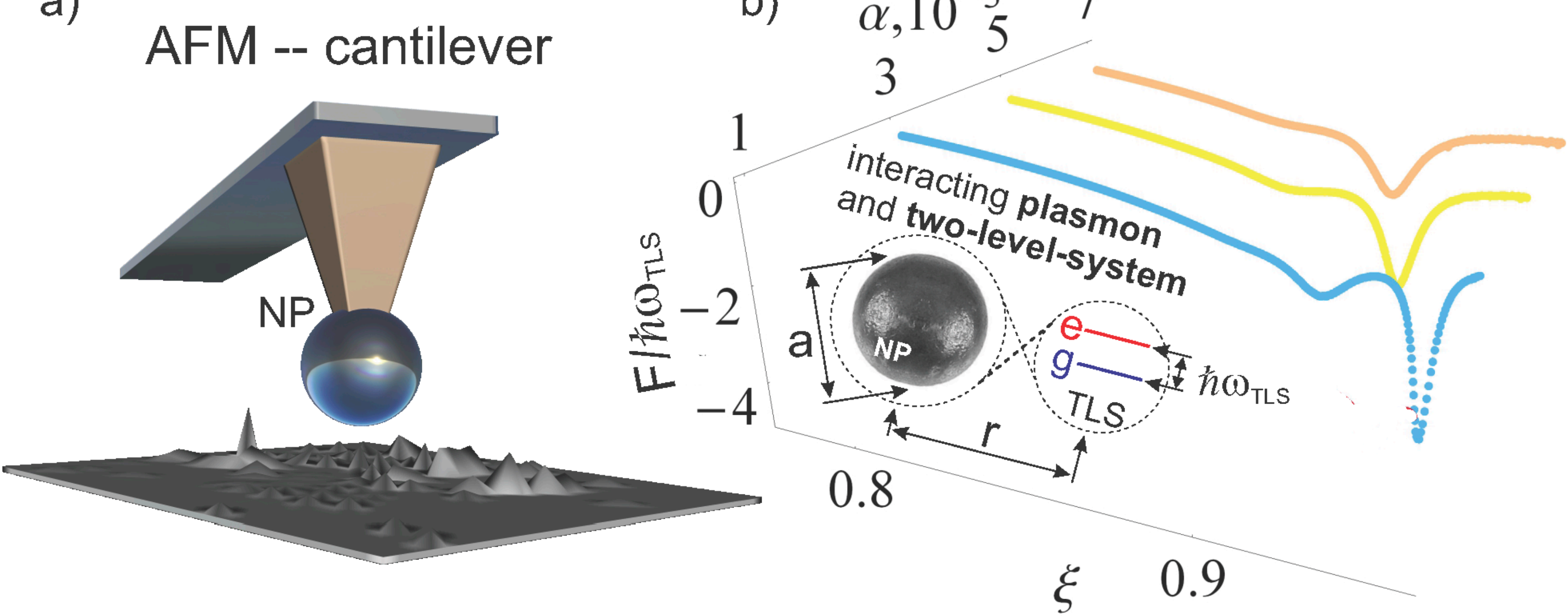


нанолазер



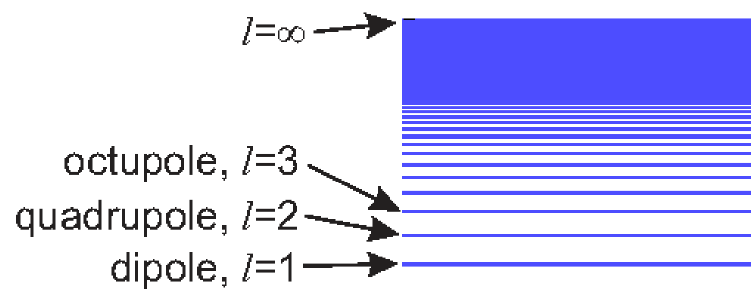
Плазмонный нанолазер (спазер)

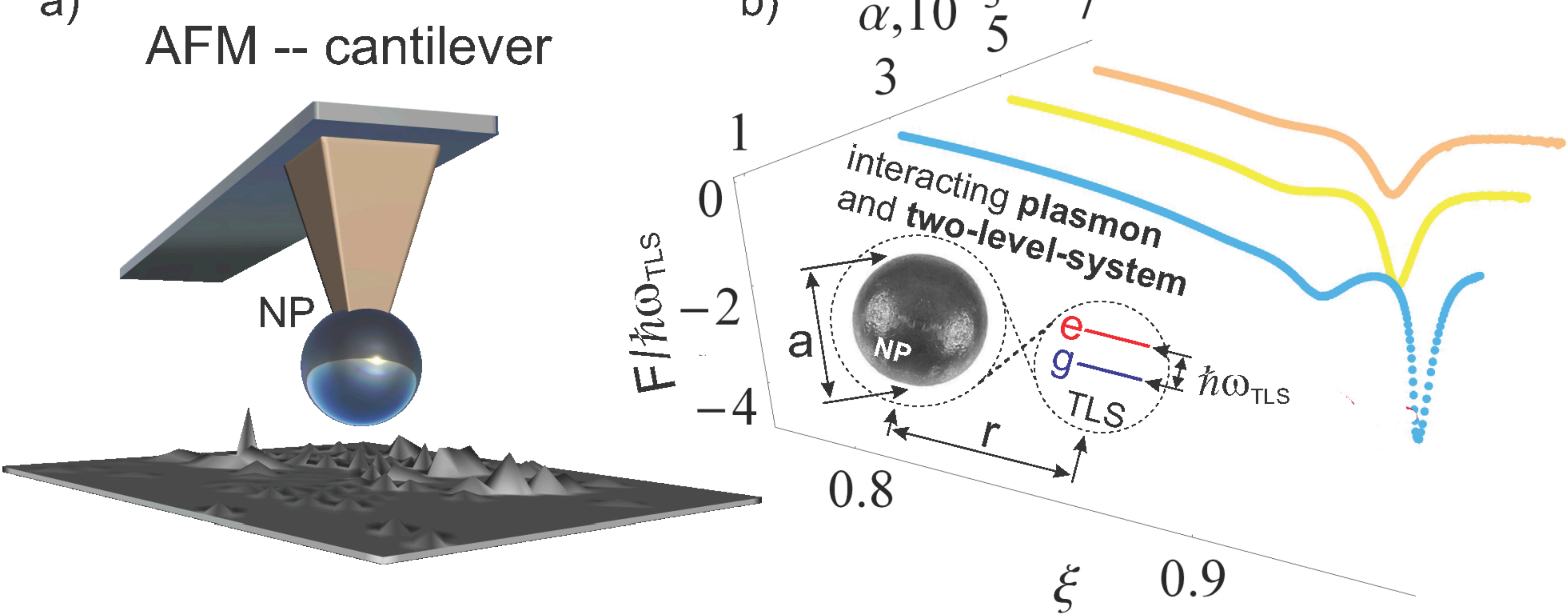
Плазмонная наночастица -- резонатор



Спектр плазмонных мод в сферической наночастице:

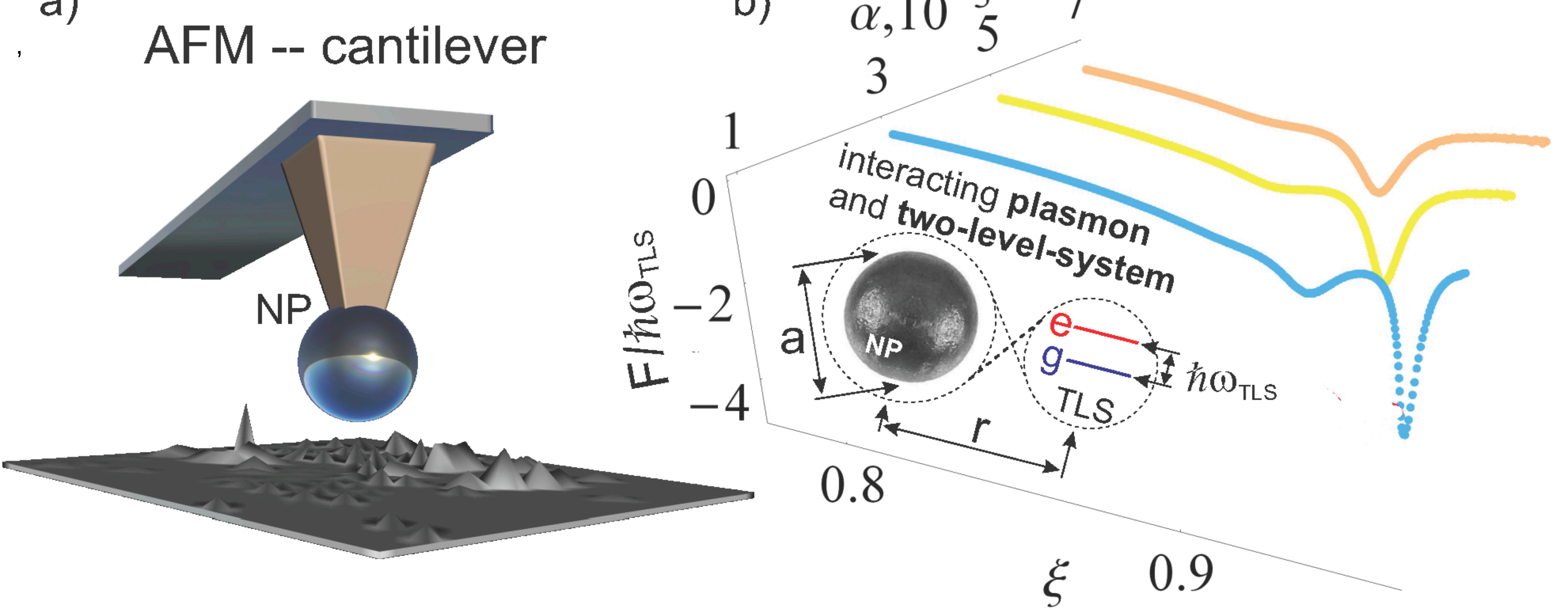
$$\omega_l = \omega_{\mathrm{pl}} \sqrt{\frac{l}{2l+1}}$$





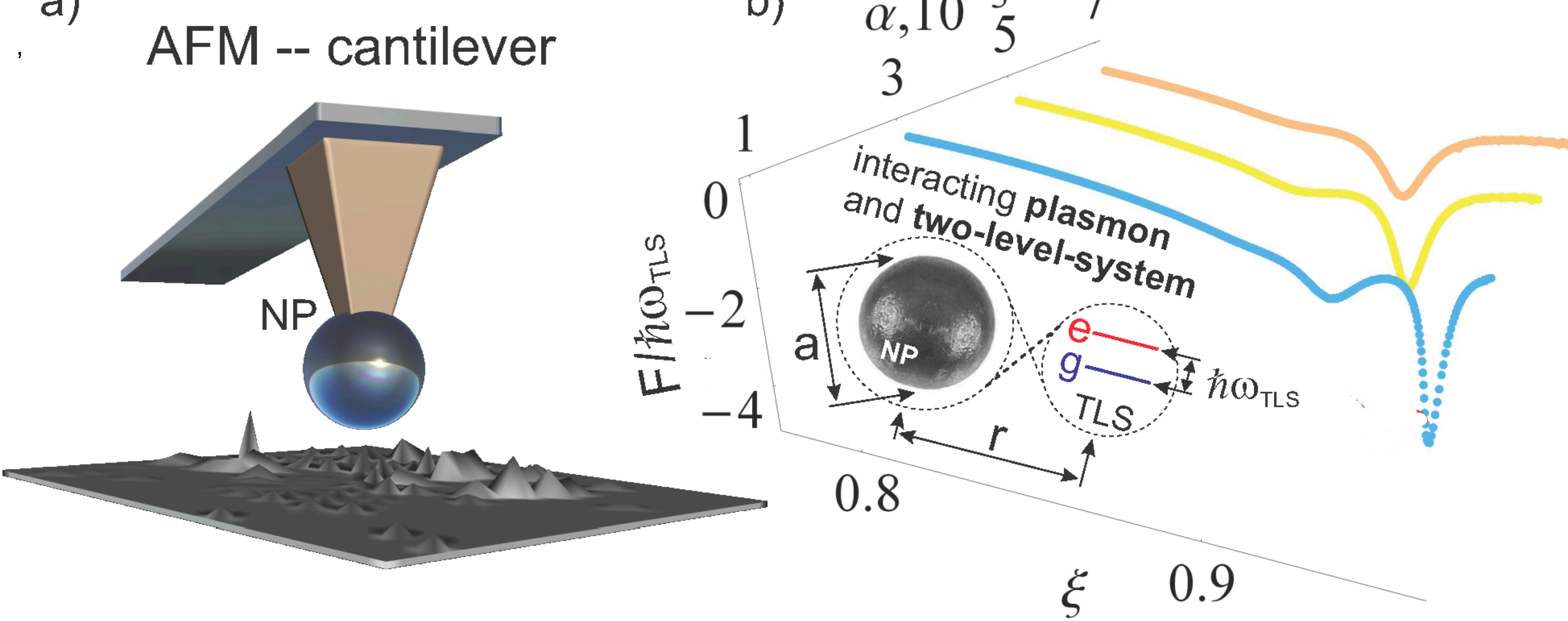
Электрическое поле тоже надо разложить по $Y_{lm}(\varphi,\theta)$

$$E_{lm} = \sqrt{4\pi\hbar\omega_l/2a(2l+1)}$$



electric field operator:

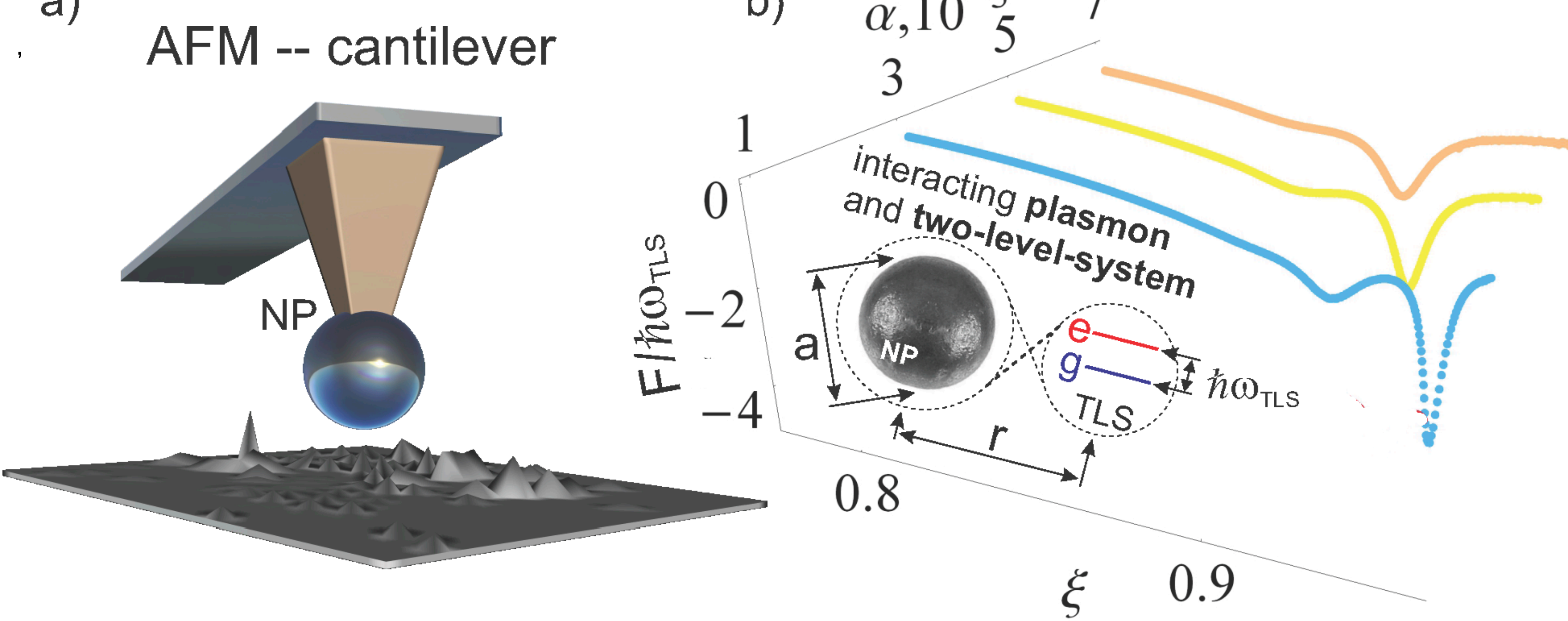
$$\hat{E}_{lm} = -E_{lm}\nabla\varphi_{lm}\left(\hat{a}_{lm}+\hat{a}_{lm}^{\dagger}\right)$$



electric field operator:

$$\hat{E}_{lm} = -E_{lm}\nabla\varphi_{lm}\left(\hat{a}_{lm} + \hat{a}^{\dagger}_{lm}\right)$$

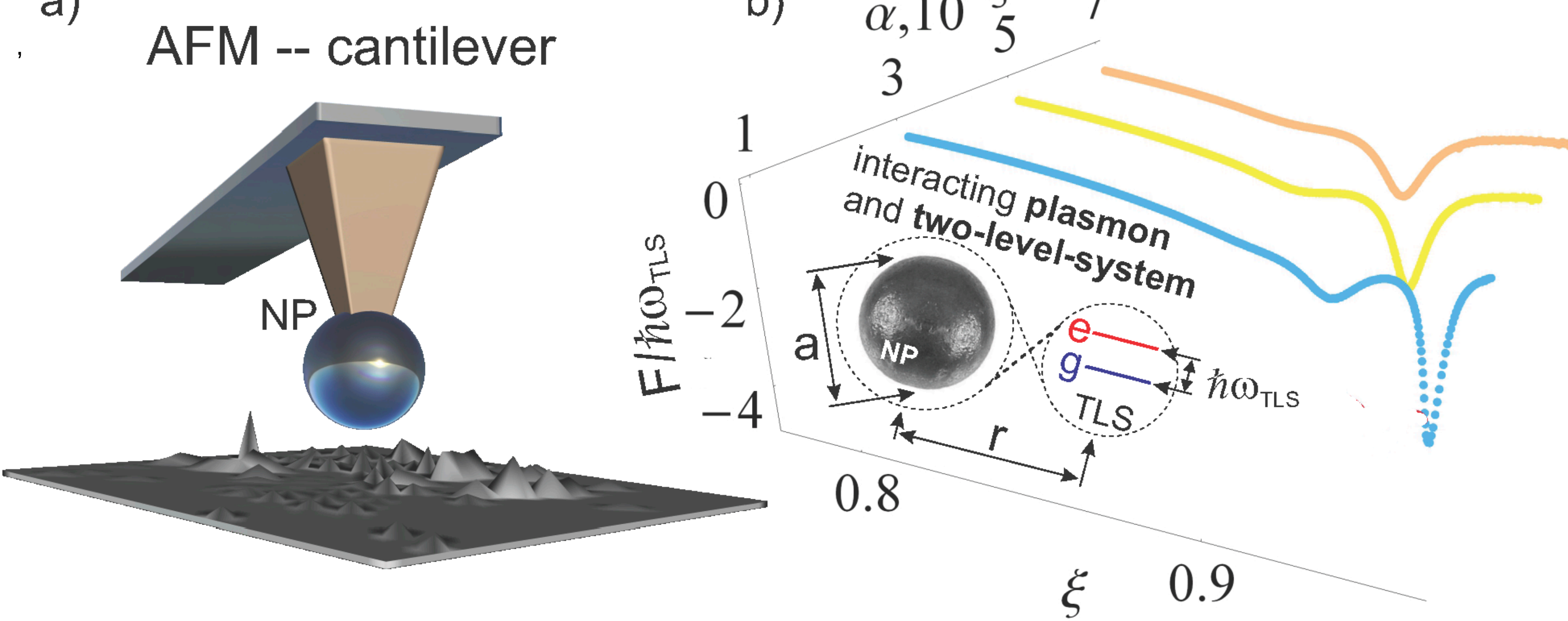
$$\boxed{\hat{H}_{\mathrm{NP}} = \sum_{lm}\hbar\omega_l\left(\hat{a}^{\dagger}_{lm}\hat{a}_{lm} + 1/2\right)}$$



Двухуровневая система:

$$\boxed{\hat{H}_{\mathrm{TLS}} = \hbar\omega_{\mathrm{TLS}}\hat{\sigma}^{\dagger}\hat{\sigma}}$$

$$\hat{\sigma} = |g\rangle\langle e|$$

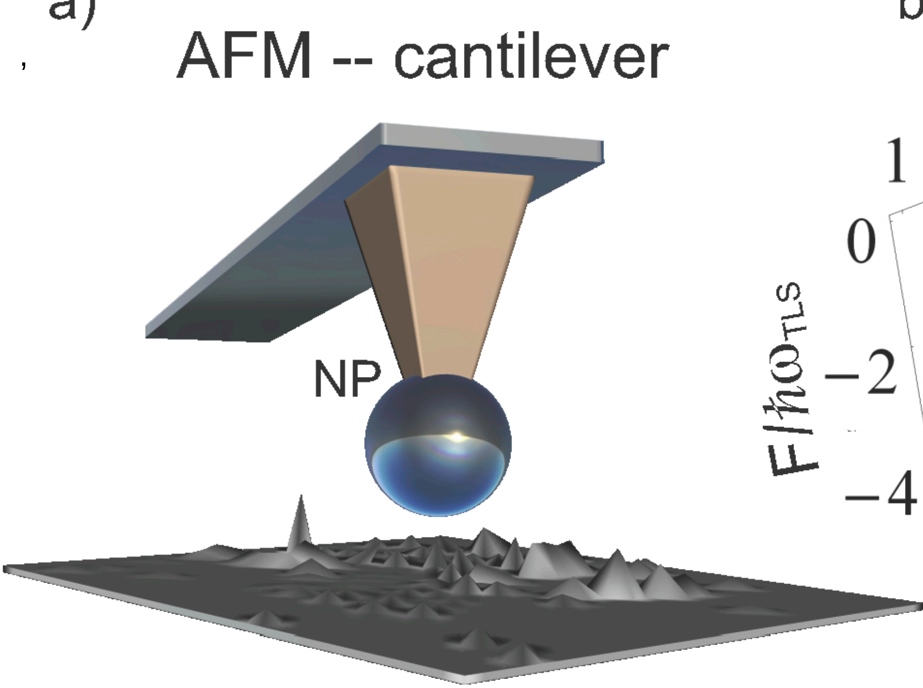


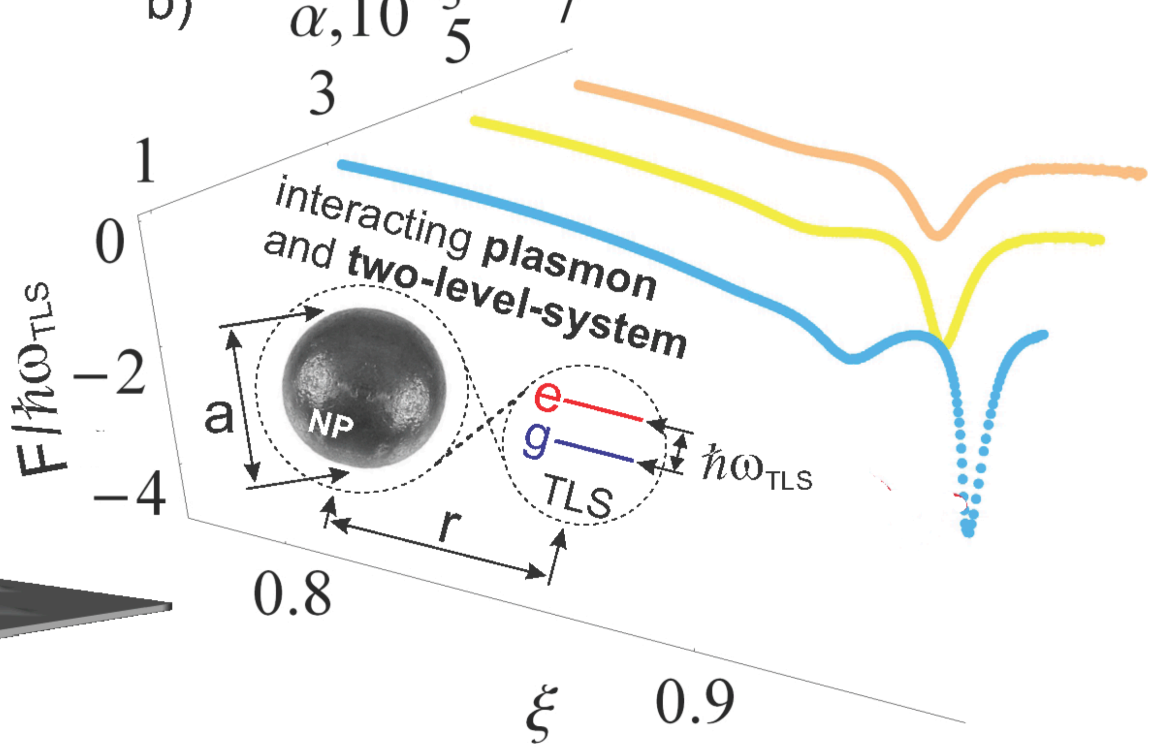
Двухуровневая система:

$$\boxed{\hat{H}_{\mathrm{TLS}} = \hbar\omega_{\mathrm{TLS}}\hat{\sigma}^{\dagger}\hat{\sigma}}$$

$$\hat{\sigma} = |g\rangle\langle e|$$

$$\boxed{\hat{\mathbf{d}}_{\mathrm{TLS}} = d_{eg}\left[\hat{\sigma}(t) + \hat{\sigma}^{\dagger}(t)\right]}$$





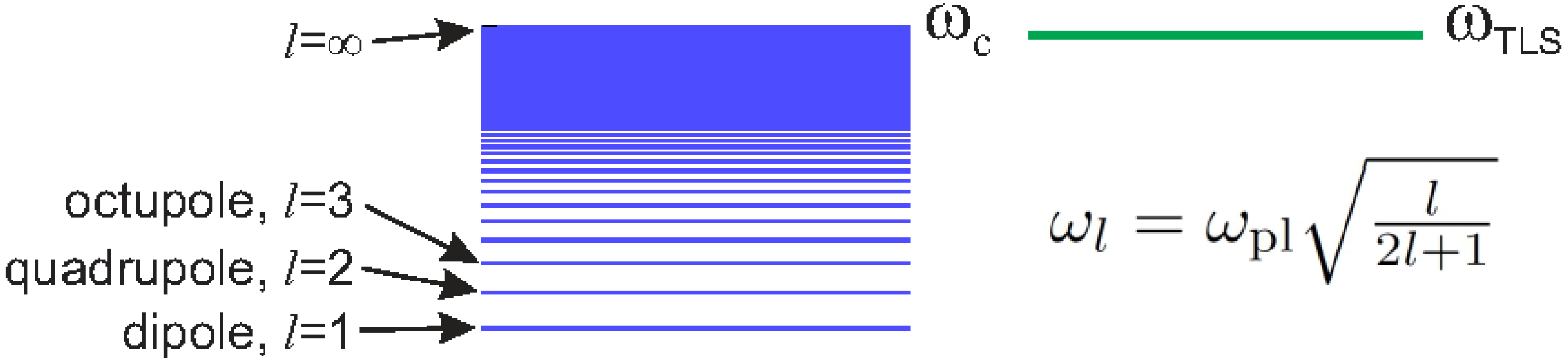
Двухуровневая система:

$$\boxed{\hat{H}_{\mathrm{TLS}} = \hbar\omega_{\mathrm{TLS}}\hat{\sigma}^{\dagger}\hat{\sigma}}$$

$$\boxed{\hat{V} = \hbar\sum\nolimits_{lm}\gamma_{lm}(\hat{a}^{\dagger}_{lm}\hat{\sigma} + \hat{\sigma}^{\dagger}\hat{a}_{lm})}$$

$$\left|\gamma_{l,m=0}\right|^2 = \alpha\omega_{\mathrm{pl}}^2\xi^{2(l+2)}(l+1)^2 l^{1/2}/(2l+1)^{1/2}, \qquad \alpha = \frac{|d_{eg}|^2}{2\omega_{\mathrm{pl}}\hbar a^3}.$$

$$\hat{H} = \hat{H}_{\mathrm{TLS}} + \hat{H}_{\mathrm{NP}} + \hat{V}$$



$$\omega_l = \omega_{\mathrm{pl}} \sqrt{\frac{l}{2l+1}}$$

Эксперимент

M. Noginov, G. Zhu, A. Belgrave, R. Bakker, V. Shalaev, E. Narimanov, S. Stout, E. Herz, T. Suteewong, and U. Wiesner, «Demonstration of a spaser-based Nanolaser»,

Nature 460(7259), 1110–1112 (2009).

Теория

- ???

Теория

- there is no well defined small or large interaction parameter
- there is high level of dissipation
- there are large quantum fluctuations

Про диссипацию…

$$d\hat{\rho}/dt = -i\left[\hat{H},\hat{\rho}\right] + \frac{\gamma_b}{2}\left(2\hat{a}\hat{\rho}\hat{a}^{+} - \hat{\rho}\hat{a}^{+}\hat{a} - \hat{a}^{+}\hat{a}\hat{\rho}\right) +$$

$$+\frac{\gamma_a}{2}\left(2\hat{\sigma}\hat{\rho}\hat{\sigma}^{+} - \hat{\rho}\hat{\sigma}^{+}\hat{\sigma} - \hat{\sigma}^{+}\hat{\sigma}\hat{\rho}\right),$$

$$\hat{H} = \hat{H}_b + \hat{H}_a + \hat{V}_{a-w} + \hat{V}_{b-w} + \hat{V}_{a-b}.$$

Про диссипацию…

$$\boxed{\hat{H} = \hat{H}_b + \hat{H}_a + \hat{V}_{a-w} + \hat{V}_{b-w} + \hat{V}_{a-b}.}$$

$$\hat{H}_b = \omega_b \hat{a}^+ \hat{a}$$

$$\hat{H}_a = \omega_a \hat{\sigma}^+ \hat{\sigma}$$

$$\hat{V}_{b-a} = \Omega_R (\hat{a}^+ \hat{\sigma} + \hat{\sigma}^+ \hat{a})$$

$$\hat{V}_{a-w} = \Omega_a \left(\hat{\sigma} + \hat{\sigma}^+ \right)$$

$$\hat{V}_{b-w} = \Omega_b (\hat{a}^+ + \hat{a})$$

Про диссипацию...

Все плохо...

$$\hat{H} = \hat{H}_b + \hat{H}_a + \hat{V}_{a-w} + \hat{V}_{b-w} + \hat{V}_{a-b}.$$

$$\hat{H}$$

Все константы в Гамильтониане --- одного порядка

$$V_{b-a} = \Omega_R(\hat{a}^+\hat{\sigma} + \hat{\sigma}^+\hat{a})$$

Диссипативные константы такие же или еще больше

$$\hat{V}_{a-w} = \quad \hat{a}^+ + \hat{a})$$

For this system $\gamma_b \sim 10^{12} - 10^{14} s^{-1}$. Due to strong field localization in the plasmonic mode the Rabi interaction constant may become comparable with the damping in the plasmonic mode [36]. $\Omega_R \sim 10^{12} - 10^{13} s^{-1}$. We should note that in this experiment many TLS have been correlated with plasmonic bosonic mode and in our model — only one. The other difference is the external drive: in the experiment the system was driven by 5ns pulses while we consider coherent drive. We do not see however any problem with the realisation of coherent drive by an external laser and the use of a single quantum dot. The realistic amplitude of the laser field is $\sim 10^3 - 10^4 V/m$. For realistic semiconductor quantum dots typical relaxation rate is $\gamma_a \sim 10^9 - 10^{11} s^{-1}$ [8].

Для накачки берем источник

$$\simeq 10 - 100 V/m$$

Про диссипацию…

$$d\hat{\rho}/dt = -i\left[\hat{H},\hat{\rho}\right] + \frac{\gamma_b}{2}\left(2\hat{a}\hat{\rho}\hat{a}^{+} - \hat{\rho}\hat{a}^{+}\hat{a} - \hat{a}^{+}\hat{a}\hat{\rho}\right) + \\ + \frac{\gamma_a}{2}\left(2\hat{\sigma}\hat{\rho}\hat{\sigma}^{+} - \hat{\rho}\hat{\sigma}^{+}\hat{\sigma} - \hat{\sigma}^{+}\hat{\sigma}\hat{\rho}\right),$$

$$\hat{H} = \hat{H}_b + \hat{H}_a + \hat{V}_{a-w} + \hat{V}_{b-w} + \hat{V}_{a-b}.$$

$$\boxed{\hat{H} = \hat{H}_b + \hat{H}_a + \hat{V}_{a-w} + \hat{V}_{b-w} + \hat{V}_{a-b}.}$$

$$\hat{H}_b = \omega_b \hat{a}^+ \hat{a}$$

$$\hat{H}_a = \omega_a \hat{\sigma}^+ \hat{\sigma}$$

$$\hat{V}_{b-a} = \Omega_R (\hat{a}^+ \hat{\sigma} + \hat{\sigma}^+ \hat{a})$$

$$\hat{V}_{a-w} = \Omega_a \left(\hat{\sigma} + \hat{\sigma}^+ \right)$$

$$\hat{V}_{b-w} = \Omega_b (\hat{a}^+ + \hat{a})$$

$$\boxed{\Omega_b = 10\Omega_a \equiv \Omega}$$

$$5 \cdot 10^{12} s^{-1} - 1 \cdot 10^{13} s^{-1}$$

$$\boxed{\gamma_b \simeq \Omega_R \simeq \Omega \gg \gamma_a}$$

Time unit t0=10^{-14} c

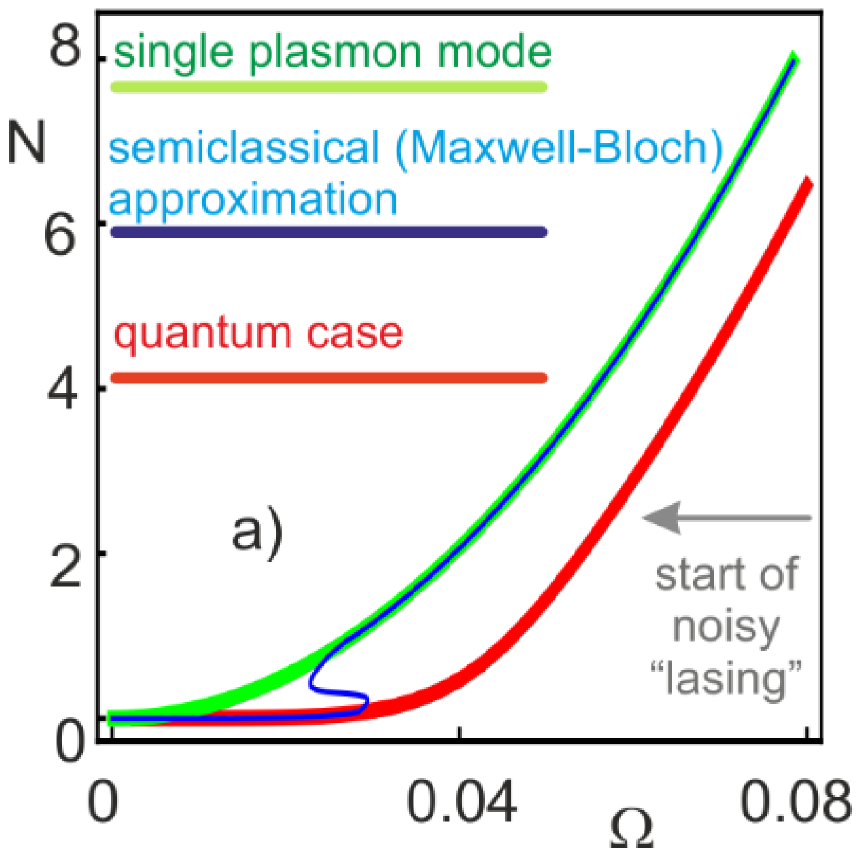
So, we imply the following values of constants: $\gamma_a = 10^{11}s^{-1}$ and $\Omega_R = 10^{13}s^{-1}$. The dumping rate of bosonic mode $\gamma_b \in (0.5 \cdot 10^{13}s^{-1}, 4 \cdot 10^{13}s^{-1})$ which corresponds to different nanocavity materials. Also we choose $\Omega_b = 10\Omega_a \equiv \Omega$ which corresponds to more intensive interaction between external field with nanocavity. It takes place when plasmonic mode is excited, e. g., see Ref. 8. The absolute value of interaction constant Ω with external field we will vary from $5 \cdot 10^{12}s^{-1}$ to $1 \cdot 10^{13}s^{-1}$ which corresponds to experimentally achievable laser field amplitude, $\simeq 10-100V/m$. In simulation we normalize all values on the $10^{-14}s$. So we consider

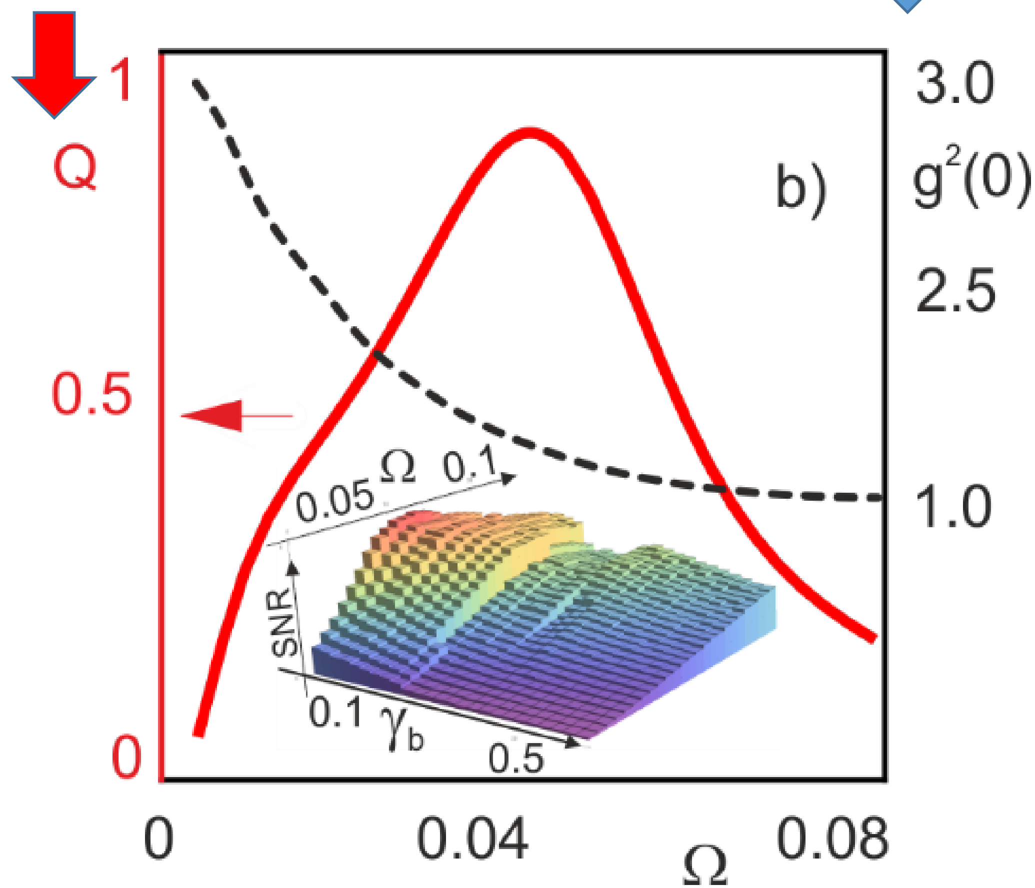
$$Q = (\langle \hat{N}^2 \rangle - \langle \hat{N} \rangle^2) / \langle \hat{N} \rangle - 1 = \langle \hat{N} \rangle (g^{(2)}(0) - 1)$$

$$g^{(2)}(\tau) = \langle \hat{a}^+(t+\tau)\hat{a}^+(t+\tau)\hat{a}(t)\hat{a}(t) \rangle$$

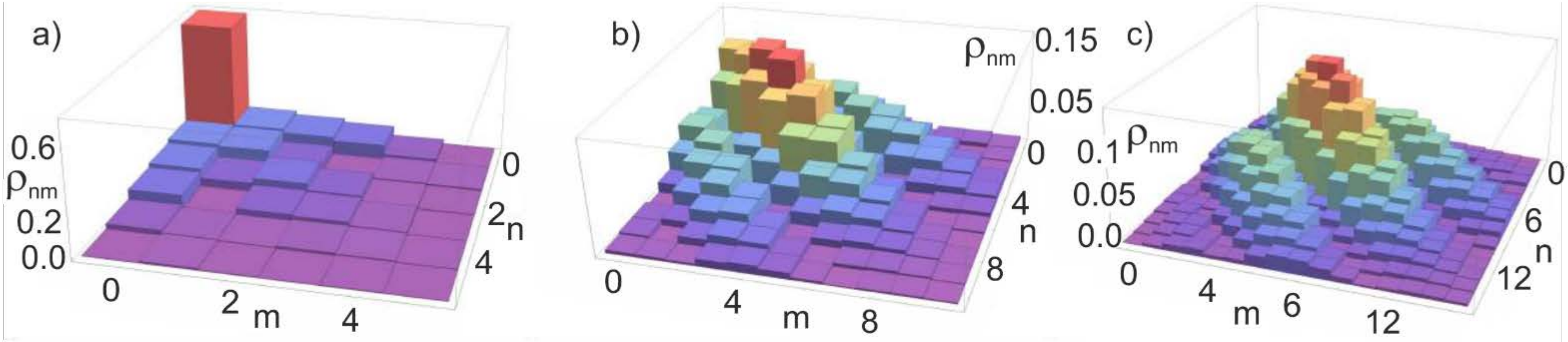
Параметр Манделя Q –
мера квантовых флуктуаций

Излучение:

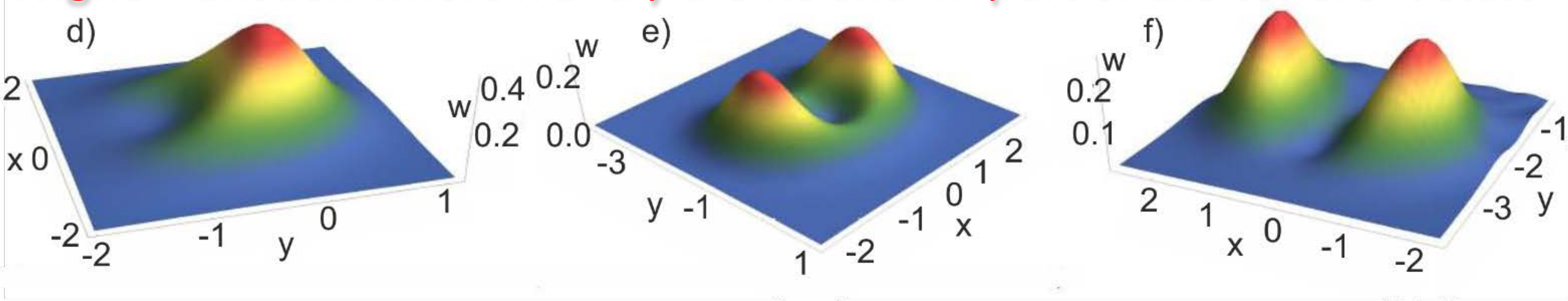


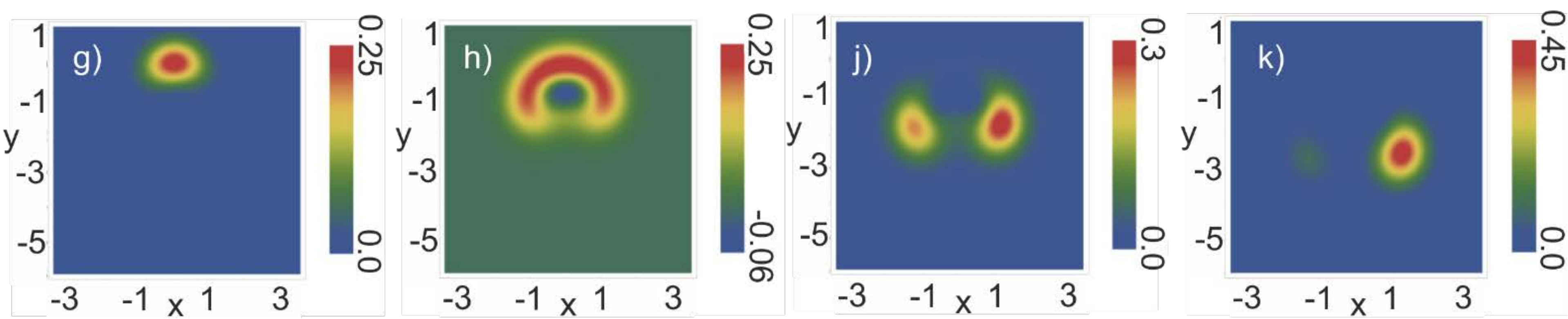


Бозонная матрица плотности



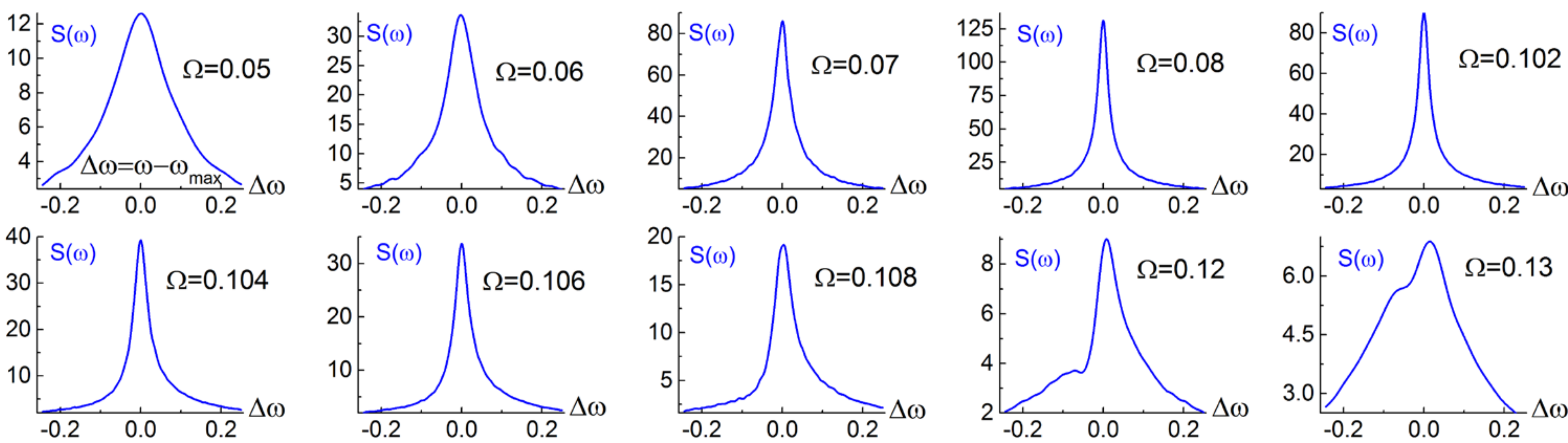
Wigner function where x and y are Re and Im part of the coherent state



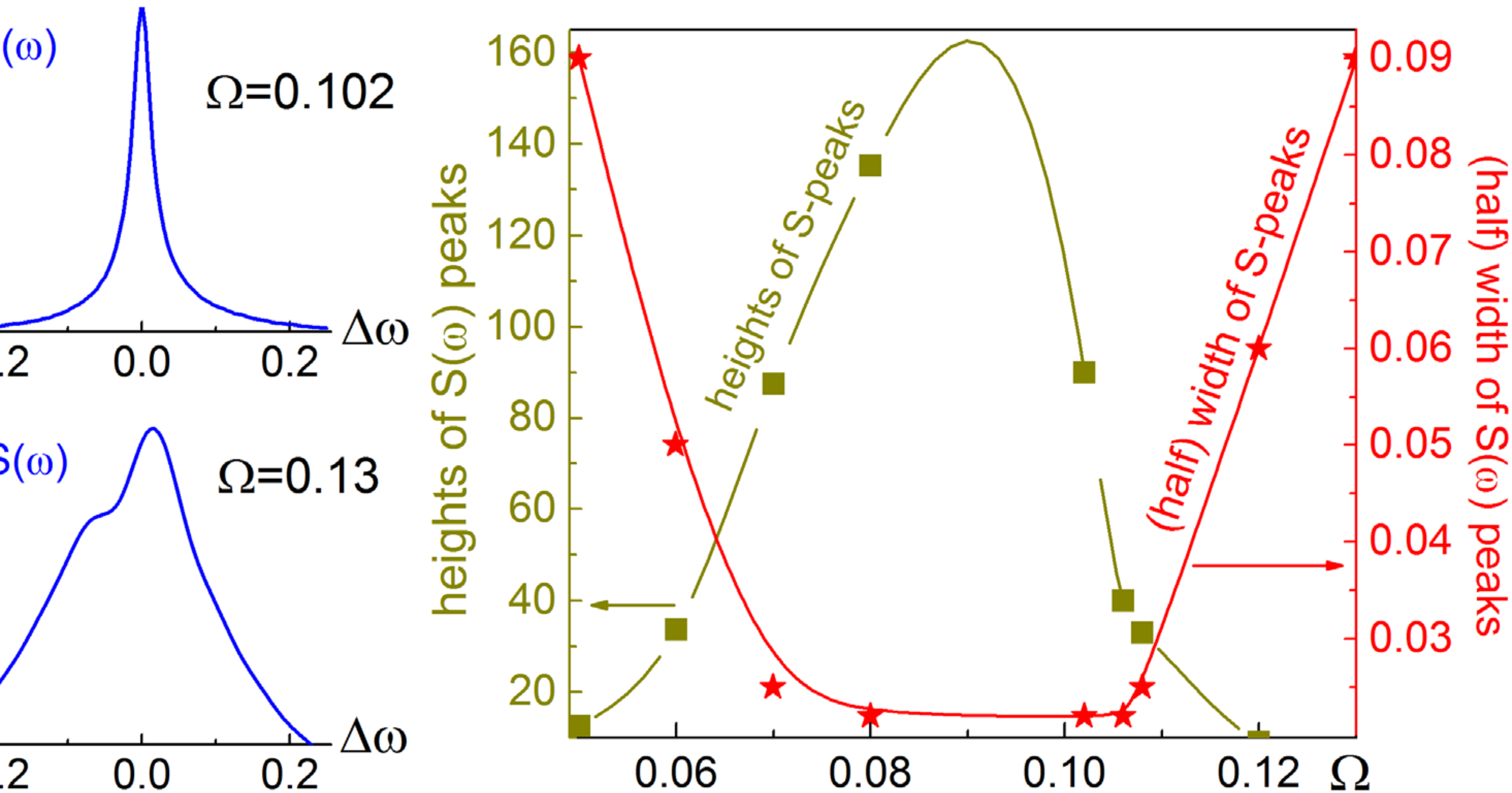


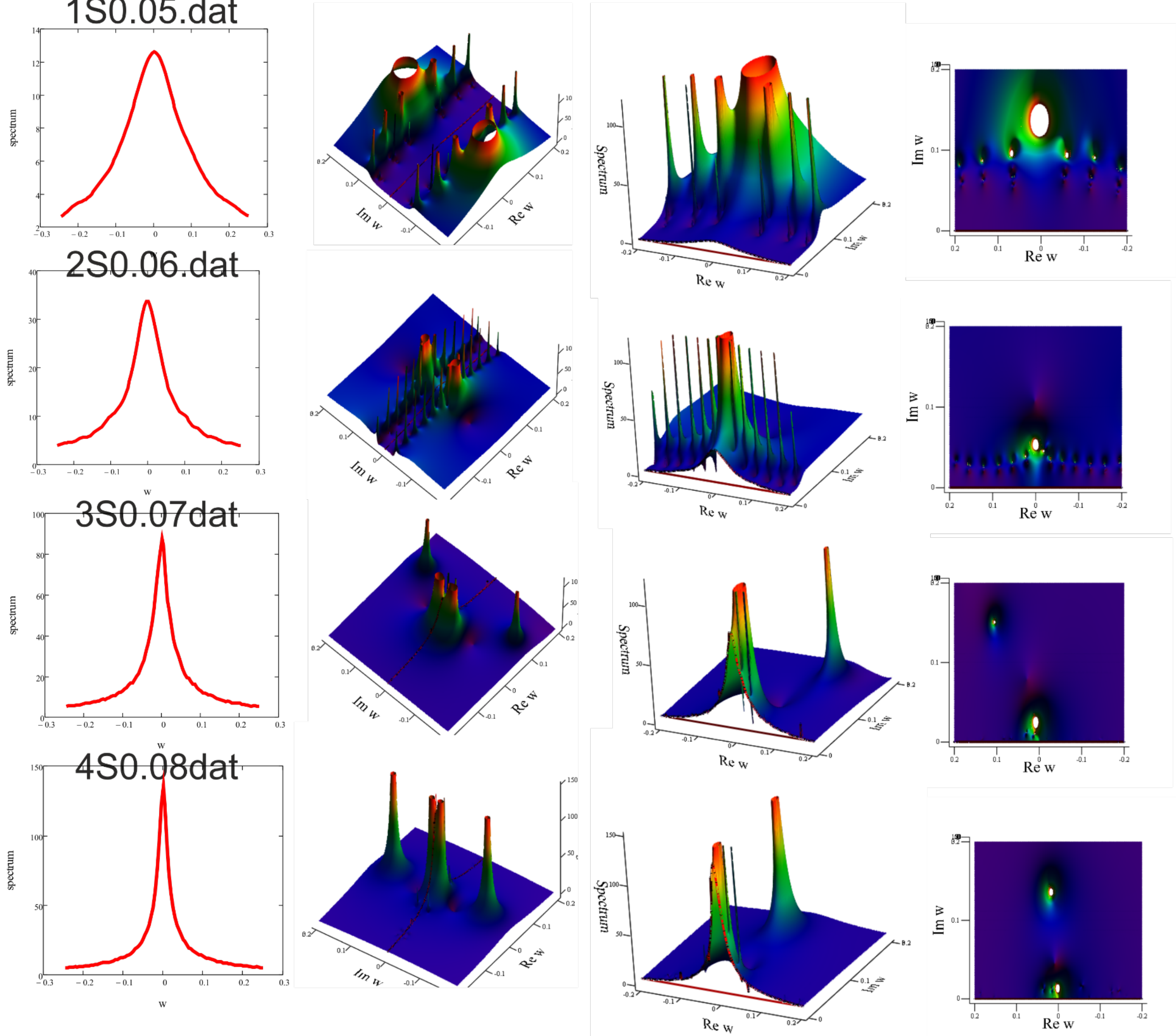
Спектральная линия

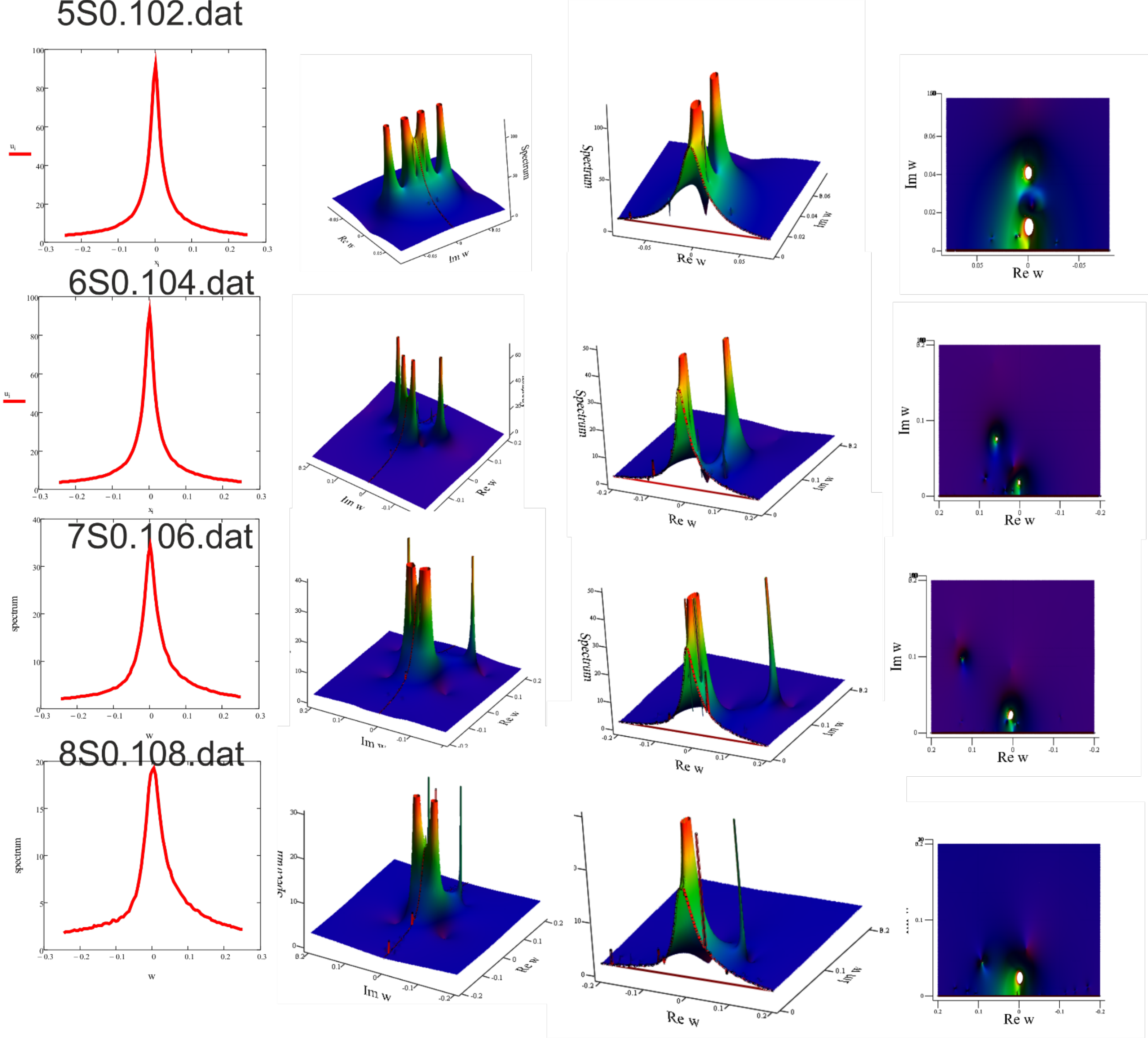
$$S(t) = \langle \hat{a}^{+}(t+\tau)\hat{a}(t) \rangle$$



Спектральная линия




1S0.05.dat
2S0.06.dat
3S0.07dat
4S0.08dat
spectrum
w
Im w
Re w
Spectrum


5S0.102.dat
6S0.104.dat
7S0.106.dat
8S0.108.dat
spectrum
Spectrum
Re w
Im w

Lasing criteria

-) transition to lasing should be accompanied by nonlinear dependence of the boson number on external drive,
- 2) emission linewidth should strongly decrease in the lasing regime and
- 3) the second order coherence function

$g^{(2)}(0)$ should tend to one which indicates the coherent output.

Как это считать?

- Кусочно-детерминистический квантовый процесс
- Piecewise deterministic processes

$$\frac{\partial}{\partial t}T(x,t|x',t') = -\frac{\partial}{\partial x_i}\left[g_i(x)T(x,t|x',t')\right] + \int dx''\left[W(x|x'')T(x'',t|x',t') - W(x''|x)T(x,t|x',t')\right] \quad (1.$$

$$\frac{d}{dt}\rho_S(t) = -i[H, \rho_S(t)] + \sum_i \gamma_i \left(A_i \rho_S(t) A_i^\dagger - \frac{1}{2} A_i^\dagger A_i \rho_S(t) - \frac{1}{2} \rho_S(t) A_i^\dagger A_i \right)$$

$$\frac{d}{dt}\psi(t) = -iG(\psi(t)),$$

$$\frac{d}{dt}\psi(t) = -iG(\psi(t)),$$

$$G(\psi) = \hat{H}\psi + \frac{i}{2}\sum_i \gamma_i \|A_i\psi\|^2\psi$$

$$\frac{d}{dt}\psi(t) = -iG(\psi(t)),$$

$$G(\psi) = \hat{H}\psi + \frac{i}{2}\sum_i \gamma_i \|A_i\psi\|^2\psi$$

$$\hat{H} = H - \frac{i}{2}\sum_i \gamma_i A_i^\dagger A_i$$

$$\frac{d}{dt}\psi(t) = -iG(\psi(t)),$$

$$G(\psi) = \hat{H}\psi + \frac{i}{2}\sum_i \gamma_i \|A_i\psi\|^2\psi$$

$$\hat{H} = H - \frac{i}{2}\sum_i \gamma_i A_i^\dagger A_i$$

$$\frac{d}{dt}\rho_S(t) = -i[H, \rho_S(t)] + \sum_i \gamma_i \left(A_i\rho_S(t)A_i^\dagger - \frac{1}{2}A_i^\dagger A_i\rho_S(t) - \frac{1}{2}\rho_S(t)A_i^\dagger A_i \right)$$

$$\frac{d}{dt}\rho_S(t) = -i[H,\rho_S(t)] + \sum_i \gamma_i \left(A_i \rho_S(t) A_i^\dagger - \frac{1}{2} A_i^\dagger A_i \rho_S(t) - \frac{1}{2}\rho_S(t) A_i^\dagger A_i \right)$$

$$F[\tilde{\psi},\tau] = 1 - \exp\left(-\int_0^\tau ds\, \Gamma[g_s(\tilde{\psi})] \right) = 1 - ||\exp(-i\hat{H}\tau)\tilde{\psi}||^2$$

Изменение волновой функции:

$$\tilde{\psi} \longrightarrow \psi = \frac{A_i\tilde{\psi}}{||A_i\tilde{\psi}||}$$

Вероятность скачка:

$$p_i = \frac{\gamma_i ||A_i\tilde{\psi}||^2}{\Gamma[\tilde{\psi}]}$$

Вычисление временных корреляторов → удвоение пространства

Вычисление спектральной линии

128 процессоров (mpi-нитей)
--- 6-12 часов счета
одного спектра

E. S. Andrianov, N. M. Chtchelkatchev, and A. A. Pukhov, "Noisy metamolecule: strong narrowing of fluorescence line," Opt. Lett.40, 3536-3539 (2015)

Выводы:

- To conclude, we describe the behavior of metamolcule driven by the external field.
- It is shown that fluorescence at dissipation larger than driving is accompanied by narrowing of the spectral line by the order of magnitude and splitting of the Wigner function of the emitted light.
- This effect may find applications in quantum systems where strong noise due to high losses is unavoidable problem like in plasmonics.