**Russian Academy of Sciences** 





## Vasily Klimov New Optical properties of nanoholes and their applications

## Outline

1. Symmetry breaking of optical field in symmetrical structure

2. Fluorescence of molecules near nanoaperture: analytical results

3. Optical Tamm states and optical properties of nanoholes

Symmetry breaking of optical and plasmon fields in symmetrical structure? (Tsema et al OE 20 (2012)10538)





Field intensity or Field intensity or Plasmon pattern Plasmon pattern The distribution of difference of intensities for right- and left-hand circularly polarized illumination in plane of holes



 $2\pi h/\lambda=1$  Tsema et al OE 20 (2012)10538)

Handedness-sensitive emission of surface plasmon polaritons by elliptical nanohole ensembles



Tsema et al OE 20 (2012)10538)

## Symmetry breaking explanation

 Strong interaction between parts (in our case between holes)

2. Retardation effects

Analytical theory of Molecule fluorescence near nanoholes (Klimov, Guzatov, Treshin 2014)



## Theory of fluorescence in free space

# Model of molecule $\begin{array}{c} \text{Molecule dynamics} \\ \text{Molecule dynamics} \\ \text{Excitation} \\ 1 - e^{-t/\tau_1} \\ \text{Emission} \\ 1 - e^{-t/\tau_2} \\ e^{-t/\tau_2} \\ \end{array}$

Intensity of fluorescence



 $\boldsymbol{U}$  is electric energy density

$$m{\mathcal{C}}$$
 is speed of light

**Theory** of fluorescence near nanohole Near nanobodies  $\Gamma_{om}, U$ are modified  $\Gamma_{em}(\mathbf{r}) = \tilde{\Gamma}_{em}(\mathbf{r})\Gamma_{0}, \ U = \frac{|\mathbf{n}\mathbf{E}|^{2}}{8\pi} = K(\mathbf{r})\frac{|\mathbf{n}\mathbf{E}_{0}|^{2}}{8\pi}$ n is orientation of dipole momentum Fluorescence intensity into lower halfspace lower half-space full intensity  $\hbar\omega_{2}\Gamma_{0}\Gamma_{em}^{\downarrow}$  $\hbar\omega_{2}$ em <u>/(r</u>  $-\frac{1+\beta\tilde{\Gamma}(\mathbf{r})/K(\mathbf{r})}{1+\beta\tilde{\Gamma}(\mathbf{r})/K(\mathbf{r})}$ <u>rtotal</u>  $\left(\frac{\hbar\omega_{1}}{cU\sigma_{abs}}\right) +$ -total  $\frac{\hbar\omega_{1}\Gamma_{0}}{cU_{0}\sigma}$ 



(through spheroidal functions)

Decay rates for an arbitrary atom position (quasistatic limit, Klimov JETP Letters, 78(2003)471)



$$d_{z,tot}^{\pm} = \frac{a\sqrt{2}}{\pi} \left( \mathbf{d}_0 \nabla' \right) f^{\pm} \left( \boldsymbol{\xi}', \boldsymbol{\eta}' \right)$$

$$f^{\pm}(\xi',\eta') = \pm \frac{2\sin(\xi'/2)}{\sqrt{\cosh\eta' - \cos\xi'}} + \frac{\sqrt{2}(\pi/2 \pm \arcsin\left(\cos\left(\xi'/2\right)/\cosh\left(\eta'/2\right)\right)}{\cosh\eta' - \cos\xi'}$$

Toroidal coordinates

$$x = a \frac{sh\eta}{ch\eta - \cos\xi} \cos\psi, y = a \frac{sh\eta}{ch\eta - \cos\xi} \sin\psi, z = a \frac{\sin\xi}{ch\eta - \cos\xi}$$

#### SPONTANEOUS DECAY RATE OF AN ATOM NEAR A NANOAPERTURE

 $\frac{\gamma}{\gamma_0} = \frac{1}{2} \left( \frac{d_{tot}^{+2}}{d_0^2} + \frac{d_{tot}^{-2}}{d_0^2} \right) \quad d_{tot}^{\pm} \text{ is dipole momentum of radiation in upper or lower halfspace}$ 



$$\frac{d_{tot}^{\pm}}{d_0} = 1 \pm \frac{2}{\pi} \left( \frac{a}{\sqrt{\rho^2 - a^2}} + \arcsin \frac{\sqrt{\rho^2 - a^2}}{a\rho} \right) \qquad \frac{d_{tot}^{\pm}}{d_0} = \mp \frac{2}{\pi} \frac{\rho}{\sqrt{a^2 - \rho^2}}, \rho < a$$

#### Decay rates of an atom with the z-orientation of the dipole moment depending on its position on the system axis.



## Decay rates of an atom with the *z*-orientation of the dipole moment depending on its position on the system axis.



Decay rate of an atom with the z-orientation of the dipole moment depending on its position in the aperture plane z = 0.



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Decay rate of an atom with the radial orientation of the dipole moment depending on its position in the aperture plane



## Decay rate for horizontal dipole in the center of aperture



Excitation field behind aperture (Bouwcamp 1954, Klimov Letokhov 1992)

$$\mathbf{E}_{t} = \frac{ika}{3\pi} \left\{ \left[ \mathbf{n}_{z} \tilde{\mathbf{H}}_{0} \right] A(\mathbf{r}, z) + \frac{\left( \tilde{\mathbf{H}}_{0} \mathbf{r} \right) \left[ \mathbf{n}_{z} \mathbf{r} \right]}{r^{2}} B(\mathbf{r}, z) \right\}$$
$$E_{z} = -\frac{ika}{3\pi r} \left[ \tilde{\mathbf{H}}_{0} \mathbf{r} \right]_{z} C(\mathbf{r}, z)$$

 $H_{0}-\text{ magnetic field in standing wave (no hole)}$   $A = R^{-} \left( \frac{2a^{2}}{R^{*}} + 2 - \frac{z^{2}}{r^{2}} \right) + za \left( \frac{R^{+}}{r^{2}} - \frac{3}{a^{2}} \operatorname{arctg} \left( \frac{1}{R^{+}} \right) \right)$   $R^{*} = \left( \left( R^{2} - a^{2} \right)^{2} + 4a^{2}z^{2} \right)^{1/2}; R^{\pm} = \left( \frac{R^{*} \pm \left( R^{2} - a^{2} \right)}{2a^{2}} \right)^{1/2}$   $r^{2} = x^{2} + y^{2}; R^{2} = r^{2} + z^{2}$ 

#### Electric field behind aperture (Klimov Letokhov 1992)



 $\mathbf{M} = -\frac{2a^3}{3\pi}\mathbf{H}_0 \qquad \mathbf{E} = \frac{2a^3ik}{3\pi R^3}[\mathbf{R}\mathbf{H}_0]$ 



Decay Rate into lower halfspace















into lower halfspace



0.5 0.4 0.3 0.2 0.1



0.5 0.4 0.3 0.2 0.1





Optical Tamm state and extraordinary light transmission through nanoaperture

#### Anomalous light transmission through array of nanoholes



Transmission through single nanohole (Bethe,1944)  $T \approx \frac{64}{27\pi^2} (ka)^4 \sim 10^{-3}$ 



#### **Electron Tamm states**



What happens if our crystal is finite?  $\psi_{\mathbf{k}}(\mathbf{r}) = u_{\mathbf{k}}(\mathbf{r})e^{i\mathbf{k}\mathbf{r}}$ 

We could have real and complex k!

I.E. Tamm showed first the existence of surface states in 1932.

I. Y. Tamm, Phys. Z. Sowjetunion 1, 733 (1932)

#### **Optical Tamm states**

PHYSICAL REVIEW B 74, 045128 (2006)

(Received 13 March 2006; revised manuscript received 20 June 2006; published 31 July 2006) Surface state peculiarities in one-dimensional photonic crystal interfaces

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#### New mechanism of extraordinary transmission (Balykin et al 2011, Treshin et al 2013)



## New mechanism of extraordinary transmission



#### Hole about 60nm, transmission about 20%

## Hole shape influence on transmission (Klimov Treshin 2012)





#### Theory of extraordinary transmission in the presence of Tamm state (Treshin Klimov et al 2013)





Qualitative explanation of effect  $(\lambda = 796 nm, d = 100 nm)$ 

Gold film without Bragg mirror

12%

num

$$T_{Bethe} = \frac{64\pi^2}{27} \left(\frac{d}{\lambda}\right)^4 \approx 0.5\%$$
$$T_{num} = 0.2\%$$

Bragg mirror  

$$T = T_{Bethe} * G = \frac{64\pi^2}{27} \left(\frac{d}{\lambda}\right)^4 \cdot \left(\frac{|\mathbf{H}|^2}{|\mathbf{H}_0|^2}\right) \approx 30\%$$

$$T = 15\%$$

(Normalization on aperture area)



## Transmission spectrum (illumination from gold side)



Nonreciprocity of the system with Tamm state <u>Absorption in Gold film:</u>

100% when illuminated from Bragg mirror

0% when illuminated from gold side

Can we construct such nonreciprocity for transmission or Is Optical diode effect possible?



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Samples for measurements of optical diode are ready and preliminary proof of optical diode effect is observed



10 x 10 array of nanohles with diameter from 50 to 500 nm

## Conclusions

- 1. The effect of asymmetric electromagnetic and plasmon fields in symmetric structures is found
- 2. Analytical solution of dipole radiation near aperture is found in quasistatic and retardation cases. On the base of this solution the Fluorescence images of single molecule found.
- 3. Extraordinary light transmission and optical diode effect are found for nanoaperture in metal film on Bragg mirror. The effects are due construction and destruction of optical Tamm states.

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