Modeling of emission and absorption distributions in a finite-size quantum molecular emitter

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ABSTRACT

A description of the model of a radiating distributed molecular emitter is given, taking into account the value of its intrinsic quantum yield QY, which is always strictly less than 1. The results of numerical calculations are presented and the features of the distributions of the fluorescent radiation power and the emitter absorption power are discussed. The zones of significant localization of the studied fields in the volume of the fluorophore molecule are determined.

Formulation of the problem

The differences of the proposed model for describing the processes of emission of a fluorescent molecule are in taking into account the following two factors:

a) the finite dimensions of the molecule (a cylinder with nanometer dimensions instead of a point source);

b) non-zero non-radiative losses in the volume of the molecule itself instead of a radiation source without attenuation of the radiation (intrinsic quantum yield Y=1).

Then we can derive the formula for power P and averaged over the period of optical oscillations flux density vector **S**:

$$P = P_{rad} + P_{nr} = \int_V Q dV$$

$$Q = -0.5\,\mathrm{Re}[\mathbf{E}_1(\mathbf{r}) \cdot \mathbf{j}_1^*(\mathbf{r})]$$

$$\mathbf{S} = 0.5\,\mathrm{Re}[\mathbf{E}(\mathbf{r}) \times \mathbf{H}^*(\mathbf{r})]$$

Here P_{rad} and P_{nr} are the radiation and non-radiation losses; Q is the specific volume absorption power; V is the volume of a cylindrical fluorescent molecule; $\boldsymbol{E}$ and $\boldsymbol{H}$ are the electric and magnetic field strength; $\boldsymbol{j}$ is the total current density; the subscript "1" denotes quantities related to the molecule.

For a solitary molecule we have:

$$P_{nr,0} = \int_V Q_{nr,0} dV,$$

$$P_{rad,0} = \int_V Q_{rad,0} dV,$$

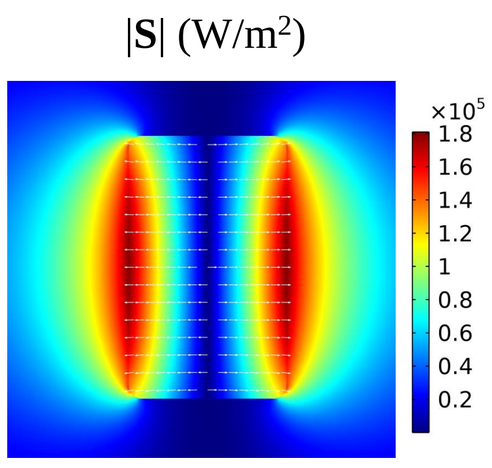
$$Q_{nr,0} = Q \le 0,$$

$$Q_{rad,0} = Q > 0,$$

Simulation results and discussion

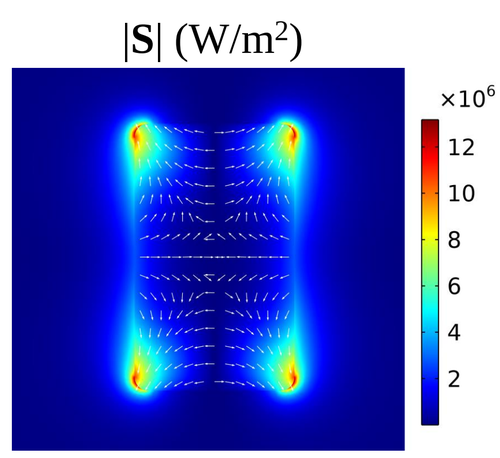
Model [*Sci. Rep.* **5**, 12956; doi: 10.1038/srep12956 (2015)]

ε″=0; QY_0=1

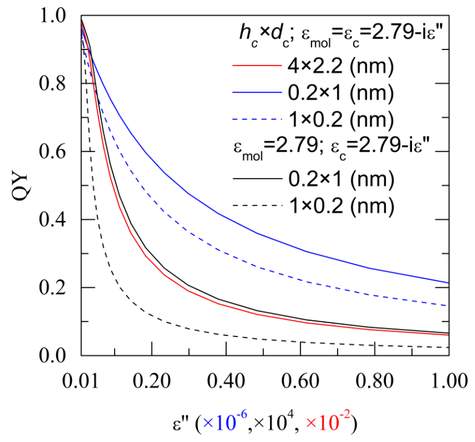


Suggested model

ε″>0; QY_0<1



The positive sign of the specific power density function Q in the cross-section of the model of a solitary distributed molecular quantum emitter corresponds to the regions of fluorescent radiation generation, the negative sign - to the regions in which radiation absorption prevails. Thus, localization zones in the volume of the fluorophore molecule are revealed. The pattern of fields in a lossless molecule (top row of fragments) changes radically when taking into account the real properties of the molecule (bottom row of fragments).



Quantum yield QY of a molecular emitter depending on the imaginary part of the permittivity of the material (modeling the main volume of the protein molecule) ε_{mol}=2.79-iε″ and the fluorescent core (chromophore) ε_c= 2.79-iε″; $h_c \times d_c$ is the size of the region in which the external current density $\mathbf{j_0}$ is specified.

CONCLUSIONS

1. **The proposed model of a distributed quantum emitter, taking into account both the finite dimensions of the fluorophore molecule and its optical properties, specifies the pattern of physical fields that arise during the generation of radiation.**
2. **Localization zones in which the processes of absorption and emission of the electromagnetic field prevail are identified.**

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