Abstract
Spontaneous emission lifetime orientation distributions of a two-level quantum emitter in metallic nanorod structures are theoretically investigated by the rigorous electromagnetic Green function method. It was found that spontaneous emission lifetime strongly depended on the transition dipole orientation and the position of the emitter. The anisotropic factor defined as the ratio between the maximum and minimum values of the lifetimes along different dipole orientations can reach up to $10^3$. It is much larger than those in dielectric structures which are only several times usually. Our results show that the localized plasmonic resonance effect provides a new degree of freedom to effectively control spontaneous emission by the dipole orientation of the quantum emitters.

Keywords: Surface plasmons; Spontaneous emission; Lifetime distribution; Nanorod

PACS: 78.67.Qa; 73.20.Mf; 42.50.-p
reported before in photonic crystals and dielectric sphere structures [11,31,32].

**Methods**

In this manuscript, we only consider the case of weak QE-field coupling regime. In this regime, the SE decay lifetimes for both homogeneous and inhomogeneous environment are calculated by the formula [32-34]

\[
\tau_{\text{vac}}(\omega) = \frac{2\omega^2}{\hbar \varepsilon_0 c^2} \frac{\text{Im} \left[ G\left( \vec{r}, \vec{r}, \omega \right) \right]}{\vec{\mu}} \quad \text{and} \quad \eta\left( \vec{r}, \omega, \vec{\mu} \right) = \frac{\tau_{\text{max}}\left( \vec{r}, \omega, \vec{\mu} \right)}{\tau_{\text{min}}\left( \vec{r}, \omega, \vec{\mu} \right)}
\]

where \(\omega\) is the angular frequency, \(c\) is the speed of light in vacuum, \(\vec{\mu}\) is the unit vector of the dipole moment, \(\text{Im} \left[ G\left( \vec{r}, \vec{r}, \omega \right) \right]\) stands for the imaginary part of Green’s tensor, and \(\vec{r}\) is the position of the QE. Notice that the SE lifetime depends on the dipole orientation. As is known that the quantity \(\text{Im} \left[ G\left( \vec{r}, \vec{r}, \omega \right) \right]\) in vacuum equals \(\omega^7/(6\pi c)\), where \(\vec{I}\) is a unit tensor. We can easily deduce the SE lifetime \(\tau_{\text{vac}}(\omega) = \left[ \omega^3 d^2/(3\pi \hbar \varepsilon_0 c^3) \right]^{-1}\) of QE embedded in vacuum according to Equation 1.

Then, the normalized orientation-dependent SE lifetime could be defined as \(\eta\left( \vec{r}, \omega, \vec{\mu} \right) = \tau_{\text{max}}\left( \vec{r}, \omega, \vec{\mu} \right)/\tau_{\text{vac}}(\omega)\). To evaluate the difference degree of the lifetime orientation distribution, we define the anisotropic factor as

\[
\eta\left( \vec{r}, \omega, \vec{\mu} \right) = \frac{\tau_{\text{max}}\left( \vec{r}, \omega, \vec{\mu} \right)}{\tau_{\text{min}}\left( \vec{r}, \omega, \vec{\mu} \right)}
\]

The Green tensor in Equation 1 satisfies

\[
\left[ \nabla \times \nabla \times -\varepsilon(\vec{r}, \omega) \frac{\omega^2}{c^2} \right] G(\vec{r}, \vec{r}, \omega) = \vec{I} \delta(\vec{r}, \vec{r})
\]

where \(\varepsilon\) is the relative permittivity. It could be calculated from the electric field of a dipole source as [35,36]
\[ \mathbf{E}(\mathbf{r}, \omega) = \frac{\omega^2}{\varepsilon_0 c^2} \mathbf{G}(\mathbf{r}, \mathbf{r}, \omega) \cdot \mathbf{d}(\mathbf{r}, \omega) \]  

\[ \mathbf{d}(\mathbf{r}, \omega) = \mathbf{d}(\omega) \delta(\mathbf{r} - \mathbf{r}_0) \]  

is a dipole source at position \( \mathbf{r}_0 \). The whole elements of the Green tensor could be attained after setting the dipole source with \( x \), \( y \), and \( z \) polarizations in turn.

**Results and discussion**

In this paper, the dielectric constant of the gold nanorod is obtained by fitting the experimental data from Johnson and Christy with piecewise cubic interpolation [37]. The nanorod is placed upon the SiO\(_2\) substrate with refractive index of 1.5. Other parts are set as vacuum. We consider rectangular, cylinder, and capsule nanorods in the simulations. The corresponding schematic diagrams of the structures are shown in Figure 1a,b,c, respectively. The cross sections of each structure at \( x = 0 \) plane are shown in Figure 1d,e,f, respectively. The width of the rectangular nanorod is \( a = 20 \) nm, the length is \( L = 120 \) nm, and the height is \( h = 20 \) nm. The diameter of the cylinder nanorod is \( d = 20 \) nm and the length is also \( L = 120 \) nm. The capsule nanorod is modified from the cylinder shape nanorod by changing the two ends into a half-sphere shape. The total length of the capsule-shaped nanorod is still \( L = 120 \) nm. We perform the simulations by the Finite Element Method with the help of the software COMSOL Multiphysics. The coordinate origin is set at the center of the nanorod, and the nanorod is placed along the \( x \) axis. We adopt the perfectly matched layer (PML) for the absorption boundary.

In order to calculate for the plasmonic resonance frequency, we consider a planewave normal incident with \( x \) polarization as \( \mathbf{E} = \mathbf{E}_x \exp \{-ik_0z\} \), where \( k_0 \) is the wave number in vacuum. The extinction spectrums

| Table 1 Anisotropy factor \( \eta \) at different positions around gold nanorod |
|-----------------|---|---|---|
|                | A  | B  | C  | D  |
| Rectangular    | 206| 386| 361| 60.1|
| Cylinder       | 615| 858| 749| 126 |
| Capsule        | 1,016| 837| 794| 137|
of the rectangular, cylinder, and capsule-shaped nanorods are indicated in black, red-dashed, and blue-dotted curves in Figure 2a, respectively. We observe the peaks at wavelength of 1,013, 997, and 946 nm for the rectangular, cylinder, and capsule nanorods, respectively. The plasmonic resonance wavelengths shift and the peak values vary a little for different nanorods. The corresponding distributions of the $\mathbf{x}$ component of electric field at $z = 0$ plane are shown in Figure 2b,c,d, respectively. The $\mathbf{x}$ component of electric field retains the same sign in the nanorod, which means the charges between the two ends of the nanorod are opposite, indicating an electric dipole mode [38].

Then, we study the orientation-dependent lifetime distributions around the nanorods at the corresponding plasmonic resonance wavelengths. The orientation distributions around the rectangular, cylinder, and capsule nanorods at wavelengths of 1,013, 997, and 946 nm are shown in Figure 3a,b,c, respectively. The $\mathbf{x}$ component of electric field retains the same sign in the nanorod, which means the charges between the two ends of the nanorod are opposite, indicating an electric dipole mode [38].

The lifetime orientation distributions of the QE in the considered structures seem to be pancake-like with a sunken center but with different contours. It illustrates that the SE lifetime strongly depended on the direction of the transition dipole. This phenomenon is due to the localized surface plasmons which are longitudinal dipolar modes at these wavelengths. When the transition dipole moment of the QE is parallel to the electric field's direction of the longitudinal dipolar plasmon mode, the interaction between the QE and the plasmonic mode is the strongest, which leads to the shortest lifetime of the QE. The anisotropy of the lifetime distribution of the QE at point A around the capsule nanorod is larger than that around the rectangular and cylinder nanorods. This is because the end of the capsule nanorod is sharper than that of the other two nanorods, which results in the stronger field enhancement around the ends. At points B and C, the lifetime orientation distributions of the QEs are different for these nanorods. At point D, the lifetime orientation distributions of the QEs are similar for the cylinder and capsule nanorods, but different for the rectangular nanorod. This is because the sides of cylinder and capsule nanorods are round but the side of rectangular nanorod is flat.

As written in the Methods part, we define the anisotropy factor $\eta$ to evaluate the orientation anisotropy by the ratio of the maximum lifetime over the minimum lifetime in all dipole orientations. The results of rectangular, cylinder, and capsule nanorods are shown in Table 1. The lifetime differs hundreds of times around the end of the rectangular nanorod. The orientation anisotropy of the cylinder nanorod is much stronger than that of the rectangular nanorod. The orientation anisotropy of the capsule nanorod is the strongest, and the anisotropy factor reaches up to three orders of magnitude when the emitter is placed 10 nm to the end of the capsule nanorod.

In order to underline the effect of the localized surface plasmon, we consider dielectric nanorods with the same geometrical parameters but without plasmonic modes.
The material of the dielectric nanorod is chosen as Si with refractive index of 3.4. The orientation distributions around the rectangular, cylinder, and capsule dielectric nanorods at wavelengths 1,013, 997, and 946 nm are shown in Figure 3d,e,f, respectively. The green area is the cross section of the Si nanorod at $z = 0$ plane. We select the four typical points as before. We observe that the maximum of the color bar can be larger than 1. So in some dipole directions, the lifetimes of QEs will be longer than those of the vacuum. They are different from the lifetimes of the QE around the metallic nanorod. The anisotropy factors of the rectangular, cylinder and capsule-shaped dielectric nanorod are shown in Table 2. The lifetime differs only several times. The lifetime orientation anisotropy factors are much smaller than the metallic nanorod case.

In the following, we further study the detailed lifetime orientation distributions of the QE near the end of the capsule gold nanorod. The orientation distributions at distance $g = 10, 15, 20, 25,$ and $30$ nm to the end of the capsule nanorod at wavelength 946 nm is shown in Figure 4a,b,c,d,e, respectively. The orientation anisotropy factors are shown in Figure 4f. The orientation anisotropy factor reduces as the distance increases. This is because the plasmonic resonance is weakly excited when the QE is far from the nanorod.

Next, we consider the frequency dependence of the orientation anisotropy. We still take the capsule nanorod as example. The QE is set at ($-70,0,0$) nm, 10 nm apart from the end of the nanorod. The orientation distributions of the QE at wavelengths 946, 1,000, 1,050, and 1,100 nm are shown in Figure 5a,b,c,d, respectively. The orientation anisotropy factors are shown in Figure 5e. We find that the orientation anisotropy factor reduces as the wavelength moves farther away from the peak wavelength. The reduction of the orientation anisotropy factor
is because the plasmon mode is weakly excited when the wavelength is moving away from the central peak frequency.

At last, we study the nanorod length dependence of orientation anisotropy. The orientation distributions of the QE at the distance 10 nm apart from the end of the capsule nanorod with length $L = 120, 90, 60$, and $20$ nm are shown in Figure 6a,b,c,d, respectively. In the case of $L = 20$ nm, the nanorod turns into a sphere. The dipole plasmonic mode of nanorods with length $L = 120, 90, 60$, and $20$ nm are at wavelengths 946, 791, 644, and 389 nm, respectively. The extinction spectrums of different nanorod lengths are not shown here. The orientation anisotropy factors are shown in Figure 6e. The orientation anisotropy is reduced rapidly as the nanorod length reduced.

Conclusions

In summary, we have studied the SE lifetime orientation distributions around a metallic nanorod by using the rigorous electromagnetic Green function method. Rectangular, cylinder, and capsule nanorods are considered. The anisotropic factor near the end of the gold capsule nanorod can reach up to $10^3$. By comparing the results of a dielectric nanorod, we point out the importance of localized plasmonic resonance to the lifetime orientation anisotropy distributions. The factors of QEs position, frequency, and the length of nanorod are investigated in detail. Our results show that the localized plasmonic resonance effect provides a new degree of freedom to effectively control spontaneous emission by the dipole orientation of the QEs.

Abbreviations

SE: spontaneous emission; QE: quantum emitter.

Competing interests

The authors declare that they have no competing interests.

Authors’ contributions

JML participated in the derivation of equations, performed the numerical simulations, interpreted the simulation results, and drafted the manuscript. JFL participated in the derivation of the equation and revised the manuscript. YCY participated in the analysis of the simulation results and revised the manuscript. LYX conceived of the study and revised the manuscript substantially. All authors had read and approved the final manuscript.

Acknowledgements

This work was financially supported by the National Basic Research Program of China (2010CB923200), the National Natural Science Foundation of China (Grant U0934002), and the Ministry of Education of China (Grant 2000801). Jingfeng Liu thanks the National Natural Science Foundation of China (Grant 11204089, Grant 11334015) for their financial support.

Author details

1 State Key Laboratory of Optoelectronic Materials and Technologies, School of Physics and Engineering, Sun Yat-sen University, Guangzhou 510275, China. 2 College of Science, South China Agriculture University, Guangzhou 510642, China. 3 Department of Physics, Foshan University, Foshan 528000, China.

Received: 14 March 2014 Accepted: 17 April 2014

References

1. Dang X, Qi J, Klug MT, Chen PY, Yun DS, Fang NX, Hammond PT, Belcher AM: Tunable localized surface plasmon-enabled broadband light-harvesting enhancement for high-efficiency panchromatic dye-sensitized solar cells. Nano Lett. 2013, 13:637–642.

2. Tagliabue G, Eghidi H, Poulikakos D: Facile multifunctional plasmonic sunlight harvesting with tapered triangle nanopatterning of thin films. Nanoscale. 2013, 5:9577–9582.

3. Koller DM, Hohnerau A, Dittbacher H, Galler N, Reif F, Aussenegg FR, Leitner A, List EW, Krenn JR: Organic plasmonic-emitting diode. Nat Photonics. 2008, 2:684–687.

4. Wiener JJ, David A, Megens MM: Ill-nitride photonic-crystal light-emitting diodes with high extraction efficiency. Nat Photonics. 2009, 3:163–169.

5. Noginov MA, Zhu G, Belgrave AM, Bakker R, Shalaveh VM, Narimanov EE, Stout S, Herz E, Suteewong T, Wiener U: Demonstration of a spaser-based nanolaser. Nature 2009, 460:1110–1112.

6. Qulton RF, Senger VJ, Zenggraf T, Ma RM, Gladden C, Dai L, Bartal G, Zhang X: Plasmon lasers at deep subwavelength scale. Nature 2009, 461:629–632.

7. Schietinger S, Barth M, Alchele T, Benson O: Plasmon-enhanced single photon emission from a nanossembled metal-diamond hybrid structure at room temperature. Nano Lett. 2009, 9:1694–1698.

8. Esteban R, Teperek TV, Greffet JI: Optical patch antennas for single photon emission using surface plasmon resonances. Phys Rev Lett 2010, 104:026802.

9. Min B, Ostby E, Sorger V, Ulin-Avila E, Yang L, Zhang X, Vahala K: High-Q surface-plasmon-polaronon whispering-gallery microcavity. Nature 2009, 457:455–458.

10. Xiao Y-F, Zou C-L, Li B-R, Li Y, Dong C-H, Han Z-F, Gong Q: High-Q exterior whispering-gallery modes in a metal-coated microresonator. Phys Rev Lett 2010, 105:153902.

11. Liu JF, Jiang HK, Jin CJ, Wang XH, Han ZS, Jin BH, Gu M: Orientation-dependent local density of states in three-dimensional photonic crystals. Phys Rev A 2012, 85:015802.

12. Chen GW, Liu JF, Jiang HK, Zhu XL, Yu YC, Jin CJ, Wang XH: Slab thickness tuning approach for solid-state strong coupling between photonic crystal slab nanocavity and a quantum dot. Nanoscale Res Lett 2013, 8:187.

13. Yamamoto T, Paschka Y, Astafoev O, Nakamura Y, Tsai JS: Demonstration of conditional gate operation using superconducting charge qubits. Nature 2001, 425:941–944.

14. Muhlschlegel P, Ederer HT, Martin OJ, Hecht B, Pohl DW: Resonant optical antennas. Science 2005, 308:1607–1609.

15. Gersc C, Ebbesen TW: Light in tiny holes. Nature 2007, 445:39–46.

16. Castané E, Krahmalnoff C, Gáza A, Pierrot R, De Wilde Y, Carminati R: Distance dependence of the local density of states in the near field of a disordered plasmonic film. Opt Lett 2012, 37:3006–3008.

17. Chen X-W, Agio M, Sandoghdar V: Metalloidielectric hybrid antennas for ultrastrong enhancement of spontaneous emission. Phys Rev Lett 2012, 108:233001.

18. Diaz-Egea C, Sigle W, van Aken P, Molina S: Resonant optical antennas. Nature 2007, 445:39–46.

19. Sinev IS, Petrov MI, Samusev AK, Rutckaia VV, Lipovskii AA: Tunable localized surface plasmon-enabled broadband light-harvesting enhancement for high-efficiency panchromatic dye-sensitized solar cells. Nano Lett. 2013, 13:637–642.

20. Tagliabue G, Eghidi H, Poulikakos D: Facile multifunctional plasmonic sunlight harvesting with tapered triangle nanopatterning of thin films. Nanoscale. 2013, 5:9577–9582.

21. Koller DM, Hohnerau A, Dittbacher H, Galler N, Reif F, Aussenegg FR, Leitner A, List EW, Krenn JR: Organic plasmonic-emitting diode. Nat Photonics. 2008, 2:684–687.

22. Wiener JJ, David A, Megens MM: Ill-nitride photonic-crystal light-emitting diodes with high extraction efficiency. Nat Photonics. 2009, 3:163–169.

23. Noginov MA, Zhu G, Belgrave AM, Bakker R, Shalaveh VM, Narimanov EE, Stout S, Herz E, Suteewong T, Wiener U: Demonstration of a spaser-based nanolaser. Nature 2009, 460:1110–1112.

24. Qulton RF, Senger VJ, Zenggraf T, Ma RM, Gladden C, Dai L, Bartal G, Zhang X: Plasmon lasers at deep subwavelength scale. Nature 2009, 461:629–632.
23. Beams R, Smith D, Johnson TW, Oh SH, Novotny L, Vamivakas AN: Nanoscale fluorescence lifetime imaging of an optical antenna with a single diamond NV center. Nano Lett 2013, 13:3807–3811.

24. Akimov AV, Mukherjee A, Yu CL, Chang DE, Zibrov AS, Hemmer PR, Park H, Lukin MD: Generation of single optical plasmons in metallic nanowires coupled to quantum dots. Nature 2007, 450:402–405.

25. Huck A, Kumar S, Shakoor A, Anderson UL: Controlled coupling of a single nitrogen-vacancy center to a silver nanowire. Phys Rev Lett 2011, 106:066801.

26. Barnard ES, Coenen T, Vesseur EJ, Polman A, Brongersma ML: Imaging the hidden modes of ultrathin plasmonic strip antennas by cathodoluminescence. Nano Lett 2011, 11:4265–4269.

27. de Leon NP, Shields BJ, Yu CL, Englund DE, Akimov AV, Lukin MD, Park H: Tailoring light-matter interaction with a nanoscale plasmon resonator. Phys Rev Lett 2012, 108:226803.

28. Chang DE, Sorensen AS, Derrler EA, Lukin MD: A single-photon transistor using nanoscale surface plasmons. Nat Phys 2007, 3:807–812.

29. Kolchin P, Oulton RF, Zhang X, Englund DE, Akimov AV, Lukin MD, Park H: Tailoring light-matter interaction with a nanoscale plasmon resonator. Phys Rev Lett 2012, 108:226803.

30. Taminiau TH, Stefani FD, van Hulst NF: Optical nanorod antennas modeled as cavities for dipolar emitters: evolution of sub- and super-radiant modes. Nano Lett 2011, 11:1020–1024.

31. Vos W, Koenderink A, Nikolaev I: Orientation-dependent spontaneous emission rates of a two-level quantum emitter in any nanophotonic environment. Phys Rev A 2010, 82:053802.

32. Liu J, Jiang H, Gan Z, Jia B, Jin C, Wang X, Gu M: Lifetime distribution of spontaneous emission from emitter(s) in three-dimensional woodpile photonic crystals. Opt Express 2011, 19:11623–11630.

33. Dung HT, Knoll L, Welsch D-G: Decay of an excited atom near an absorbing microsphere. Phys Rev A 2001, 64:013804.

34. Chen G, Yu YC, Zhuo XL, Huang YG, Jiang H, Jin C, Wang X: Ab initio determination of local coupling interaction in arbitrary nanostuctures: application to photonic crystal slabs and cavities. Phys Rev B 2013, 87:135138.

35. Tornai MS: Green function for multilayers: light scattering in planar cavities. Phys Rev A 1995, 51:2545–2559.

36. Novotny L, Hecht B: Principles of Nano-Optics. Cambridge: Cambridge University Press; 2006. ISBN 9780521832243.

37. Johnson PB, Christy RW: Optical constants of the noble metals. Phys Rev B 1972, 6:4370–4379.

38. Liu J, Lee T-W, Gray S, Guyot-Sionnest P, Pelton M: Excitation of dark plasmons in metal nanoparticles by a localized emitter. Phys Rev Lett 2009, 102:107401.

doi:10.1186/1556-276X-9-194
Cite this article as: Liu et al.: Strong anisotropic lifetime orientation distributions of a two-level quantum emitter around a plasmonic nanorod. Nanoscale Research Letters 2014 9:194.

Submit your manuscript to a SpringerOpen journal and benefit from:

- Convenient online submission
- Rigorous peer review
- Immediate publication on acceptance
- Open access: articles freely available online
- High visibility within the field
- Retaining the copyright to your article

Submit your next manuscript at ➤ springeropen.com