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Impact of spectral broadening on plasmonic enhancement with metallic gratings

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The Purcell factor (PF) of the propagating surface plasmon polariton (SPP) mode on metallic grating is evaluated with full integration formula of Fermi's golden rule, while both spontaneous emission linewidth of single quantum dot and the spectrum broadening of photonic density of state (DOS) due to the propagation loss of SPP mode are involved. It is found that the PF would be degraded by taking account of the emission linewidth. For emitters with narrow linewidth, the DOS broadening is dominant while it would be helpful to some degree for wide-linewidth emitters.
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The surface plasmon polariton (SPP) is a transverse-magnetic surface electromagnetic excitation that propagates in a wave like fashion along the metal and dielectric interface.[1] In particular, SPP is attractive to enhance the spontaneous emission (SE) from active materials due to its large density of state (DOS) and small mode volume, namely Purcell effect.[2] Till now, enhanced SE has been proposed and demonstrated not only for wide band gap semiconductors, e.g., GaN/InGaN[3,4] and ZnO,[5] but also for silicon nanocrystals (Si-NCs) or silicon quantum dots (Si-QDs).[6,7] Especially, SPP offers advantages of SE enhancement for Si emitters due to very low original radiative efficiency.[8] In order to obtain sufficient plasmonic enhancement for Si emitter, the SPP resonance should be tuned to match the central frequency of emission from Si-NCs, and metallic gratings or islands/particles were therefore introduced to Si-emitter.[6,7] In our previous work,[9,10] the SE enhancement due to SPP band gap effect on metallic gratings was investigated by calculating Purcell factor (PF), which is the most popular figure of merit to evaluate the SE modification. Usually, PF is calculated from the reduced form of Fermi's golden rule, where only the DOS and mode volume of photon (or SPP mode) are involved,[3,7–10] while the emission linewidth of emitter is assumed sufficiently narrow.[11,12] Obviously, the PFs calculated with the reduced form would exclude the influence of active material and only evaluate the effect of optical cavity or SPP waveguide. However, it is found that the PF would be degraded for emitter with broad emission linewidth.[13,14] Meanwhile, the absorption loss of metal material would introduce spectral broadening of DOS so that SE rate coupled into SPP mode is strongly influenced.[15,16] Since the DOS (or group velocity) is rather high (or low) at the band edge (BE) of a periodic metal structure or SPP resonant frequency, such DOS broadening would be more significant.[16] Thus, both the emission linewidth of emitter and the DOS broadening due to the propagation loss should be involved in PF, but the interaction between them has not yet been clarified.

In this paper, Purcell enhancement of SPP mode propagating on metallic gratings is evaluated with a full integration formula deduced from Fermi's golden rule, while emission linewidth and DOS degradation/broadening due to mode propagating loss are taken into account. As a representative example, the PF is calculated for Au-Si_3N_4 grating within frequency span of $\hbar\omega_0 = 1.6$–$1.9\,\text{eV}$, while the emission linewidth of single Si-QD is involved. For both lossless and lossy SPP modes, our calculations suggest a general reduction of PF along with increasing emission linewidth. Moreover, it is found that narrow- and wide-linewidth emitters would react differently when the DOS of SPP decreases and broadens. For emitters with sufficiently narrow linewidth ($\hbar\Delta\omega < 1\,\text{meV}$), the reduction and broadening of DOS collaborate in the degradation of PF. However, in the wide linewidth cases, the broadening of DOS spectrum tends to enlarge the entire integration span, counterbalance the impact of reduced DOS value, and could increase the PF to some degree. For instance, with the emission linewidth of 152 meV, the averaged PFs of lossless and lossy mode are 13.5 and 23.4, respectively. Furthermore, our results indicate that ultrahigh PF(>100) would be achieved if the emission linewidth is sufficiently narrow and the mode propagating loss is negligible, while for wide-linewidth emitters, higher SE enhancement would be obtained in lossy SPP modes especially when the emission linewidth and DOS linewidth of SPP mode are matched.

Fig. 1 shows the one dimensional metallic grating, where the interface shape is described as $z = s(x)$. The dielectric and metal are in the region $z > s(x)$ and $z < s(x)$, respectively. According to Fermi's golden rule, the SE rate Γ_{sp} is[12]

$$\Gamma_{sp} = \frac{2\pi}{\hbar^2}\int_0^\infty |\langle f|\boldsymbol{d}\cdot\boldsymbol{E}|i\rangle|^2 \cdot \rho(\omega)\cdot \ell(\omega)d\omega, \qquad (1)$$

where $\langle f|\boldsymbol{d}\cdot\boldsymbol{E}|i\rangle$ is the dipole emission matrix element, $\boldsymbol{d}$ is the electro-hole pair dipole moment, $\boldsymbol{E}$ is the electric field, $\rho(\omega)$ is the DOS for photon and $\ell(\omega)$ is the mode density of dipole transition (material emission spectrum) with the normalization condition $\int_0^\infty \ell(\omega)d\omega = 1$. Usually, the spectrum of the spontaneous emission of single QD could be described

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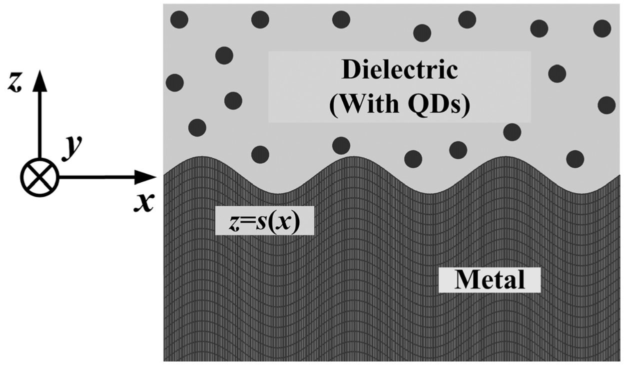

FIG. 1. Schematic metallic grating and reference coordinate system. Mass identical QDs are assumed to be uniformly distributed in dielectric material ($z > s(x)$).

as Lorentzian shape $\ell(\omega-\omega_0)=(\Delta\omega/2\pi)/[(\omega-\omega_0)^2+(\Delta\omega/2)^2]$, where ω_0 and $\Delta\omega$ denote the central frequency and linewidth.[12] If the ω dependence of $\rho(\omega)$ is much gentler than that of $\ell(\omega)$, Eq. (1) could be reduced to $\Gamma_{sp}(\omega_0)=\frac{2\pi}{\hbar^2}|\langle f|\boldsymbol{d}\cdot\boldsymbol{E}|i\rangle|^2\cdot\rho(\omega_0)$,which is widely used to evaluate the plasmonic enhancement.[3,7–10] However, for a practical emitter with finite emission linewidth, Eq. (1) should be employed.

It has been found that DOS spectrum of SPP mode would be broadened due to the ohmic loss of metal materials[15,16] and could also be described as Lorentzian shape $\rho_k(\omega)=(\omega_k/2\pi Q_k)/[(\omega-\omega_k)^2+(\omega_k/2Q_k)^2]$.[16] Here ω_k and Q_k are the frequency and quality factor corresponding to mode k, respectively. As a result, the SE rate should be a sum of all k-states $\Gamma_{sp}(\omega_0)=\int_0^\infty\sum_k \frac{2\pi}{\hbar^2}|\langle f|\boldsymbol{d}\cdot\boldsymbol{E}_k|i\rangle|^2\cdot\rho(\omega-\omega_k)\,\ell(\omega-\omega_0)d\omega$.

For simplicity, we start from the reduced form of Γ_{sp}. First, the polarization of $\boldsymbol{d}$ is considered as random directional to $\boldsymbol{E}$ so that an averaged factor of 1/3 is taken as $|\langle\boldsymbol{d}\cdot\boldsymbol{E}\rangle|^2=1/3(d^2|E|^2)$.[3] $\boldsymbol{E_k}$ is normalized to a half-quantum of zero point fluctuations by assuming a prepared space of $V=L_xL_yL_z$.[3] Thus, the stored field energy can be expressed as $1/8\pi\int^{L_x}\int^{L_y}\int^{L_z}[\partial(\varepsilon\omega)/\partial\omega]\cdot E_k^2(x,z)d_xd_yd_z$. For the considered SPP mode, the integration along z direction should be $L_z\to\pm\infty$. Since the boundary in x direction is periodic $E_k\ (x+a,z)=E_k\ (x,\ z)$, the integration can be re-expressed as $\int_0^{L_x}dx=L_x/a\cdot\int_0^a dx$. In the y direction, $\boldsymbol{E_k}$ is homogeneous so that the stored energy should be $L_xL_y/(8\pi a)\cdot\int_0^a\int_{-\infty}^{+\infty}[\partial(\varepsilon\omega)/\partial\omega]E_0^2(x,z)dzdx$. In the considered case of $k_x=k$, $k_y=0$, the reciprocal mode length of one SPP mode is $\Delta k=\Delta k_x=2\pi/L_x$. Then the PF could therefore be deduced with the well known SE rate in bulk material

$$PF(\omega_0|x,z)=1+\sum_k\frac{\Gamma_{sp}^k(\omega_0)}{\Gamma_0(\omega_0)}$$
$$=1+\frac{\pi c^3}{n\cdot\omega_0^2}\sum_k H(\omega_k)\cdot\rho(\omega_0-\omega_k)\Delta k, \quad (2)$$

where $H(\omega_k)=E_k^2(x,z)/(L_y/a)\int_0^a\int_{-\infty}^{+\infty}[\partial(\varepsilon\omega_k)/\partial\omega_k]E_k^2(x,z)$ $dzdx$. L_y denotes the width of the grating bar and is proportional to the mode volume of SPP mode defined as $V=L_y\int_0^a\int_{-\infty}^{+\infty}[\partial(\varepsilon\omega)/\partial\omega]E_0^2(x,z)dzdx/max\{[\partial(\varepsilon\omega)/\partial\omega]E_0^2(x,z)\}$.[7]

If the real-space is large enough, k can be considered as continuous variable, and the sum in Eq. (2) is approximated as integration. Then the full integration form of PF is

$$PF(\omega_0|x,z)=1+\frac{\pi c^3}{n\cdot\omega_0^2}\int_0^\infty\int_0^\infty H(\omega_k)\cdot\rho(\omega-\omega_k)\ell(\omega-\omega_0)\times\frac{dk}{d\omega_k}d\omega_k d\omega. \quad (3)$$

It should be noticed that the integration variable of dk is replaced by $d\omega_k$ by multiplying $dk/d\omega_k$ for a convenient calculation of PF since both $\rho(\omega)$ and $\ell(\omega)$ are explicit functions of ω. If the emission linewidth is sufficiently narrow, $\ell(\omega)$ would be Dirac's function $\delta(\omega-\omega_0)$ and Eq. (3) is reduced to $PF(\omega_0)=1+\pi c^3/(n\cdot\omega_0^2)\int_0^\infty H(\omega_k)\cdot\rho(\omega_0-\omega_k)(dk/d\omega_k)d\omega_k$, which is very similar to the formula reported in Ref. 16. For lossless mode, the DOS should also be $\delta(\omega-\omega_k)$ so that Eq. (3) would be further simplified to $PF(\omega_0)=1+\pi c^3/(n\cdot\omega_0^2)\cdot H(\omega_0)\cdot(dk/d\omega_0)$, which is consistent with our previous work.[10]

We consider a special case of Au-Si_3N_4 grating with sinusoidal shape ($s(x)=d\sin(2\pi x/a)$, $d=0.1\,a$). The parameters of Au are from Ref. 17 while the emitter is considered as the Si-QD embedded in Si_3N_4 with $\varepsilon_d=4$. To obtain the dispersion curve and electromagnetic field distribution required in calculating PF, we have proposed a combination method for lossless SPP mode.[18] Actually, this method could also serve for lossy SPP mode while introducing some necessary algorithms of complex operation. The calculated dispersion curves for lossless and lossy modes with $a=80$ nm are shown in Fig. 2(a) with the dispersion curve for flat surface and light line. For more clarity, only the curves for lower energy branch of lossy mode are shown. The quality factor of lossy mode is calculated as $Q_k=\omega_k/\alpha v_g$, where the energy decay rate α and group velocity v_g could be obtained as $\alpha=2\mathrm{Im}[k]$ and $v_g=\mathrm{d}\omega_k/\mathrm{d}(\mathrm{Re}[k])$.[16]

As shown in Eq. (3), PF is related to the position of QD in x axis and z axis. Since the QDs are assumed as uniformly

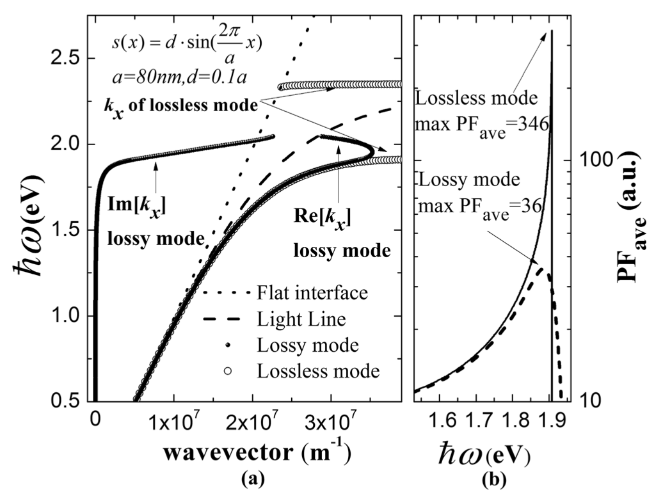

FIG. 2. (a) The calculated dispersion curves for Au-Si_3N_4 grating with $a=80$ nm and $d=0.1\,a$. The open and solid circles are for lossless and lossy mode, respectively. (b) The calculated PF spectrum for lossless and lossy mode. The PF is calculated at $z=s(x)+1$ nm and averaged within one period.

distributed, the PF should be averaged within one period along x axis. In z axis, the PF near boundary ($z = s(x) + 1\,\text{nm}$), namely $PF_{\text{ave}} = 1 + \frac{1}{a} \cdot \int_0^a [PF(x) - 1]dx|z = s(x) + 1\text{nm}$, is adopted. Fig. 2(b) shows the PF_{ave} calculated with reduced form for lossless and lossy SPP mode by setting $a = 80\,\text{nm}$. For lossless mode, the maximum value of $\sim$346 is achieved at the BE-frequency (1.91 eV) while that of lossy mode decreases to $\sim$36 at a lower frequency of 1.88 eV. As discussed in Ref. 16, it results from the propagation loss of SPP mode. Near the BE-frequency, the propagation loss (imaginary part of k) increases dramatically due to slow light effect, which would significantly degrade the DOS. At much lower frequency ($<$1.7 eV), the spectral broadening is small and would be averaged in the integration so that there is nearly no difference between such two modes. Such results are very similar to those for uniform metal-dielectric interface, and detailed discussions could be found in Ref. 16. Additionally, it should be mentioned that the PF would decay along z direction with $\boldsymbol{E}(x, z)$. For example, the calculated PF_{ave} for lossless/lossy SPP mode would be degraded from 346/36 to 271.5/28.5, 205/21.9 at the position of $z = s(x) + 5\,\text{nm}$ and $z = s(x) + 10\,\text{nm}$, respectively. However, the choice of z position would not significantly change the main results shown in the followed parts except for the particular PF values. Thus, we adopt the averaged PF at $z = s(x) + 1\,\text{nm}$ to evaluate the theoretically achievable enhancement.

To take the emission linewidth of emitter into account, we adopt a middle value of $\hbar\Delta\omega = 152\,\text{meV}$ measured at room temperature (RT).[19] By setting ω_0 of $l(\omega)$ consistent with that of the max$[PF_{ave}]$, the PF_{ave} for lossy and lossless mode are calculated with varied grating period of $a = 80\,\text{nm}$–$130\,\text{nm}$. In Fig. 3, the results obtained with reduced form and full integration are represented by solid and open symbols, respectively. In Fig. 3, it could be found that with the full integration form, the PF_{ave} of lossy mode would be higher than lossless mode (19.4–23.4 vs. 7–13.5), while PFs calculated with reduced form are on the contrary (24–35.7 for lossy mode vs.148–346 for lossless mode). Such results indicate that lossy mode is helpful to reduce the influence of wide emission linewidth.

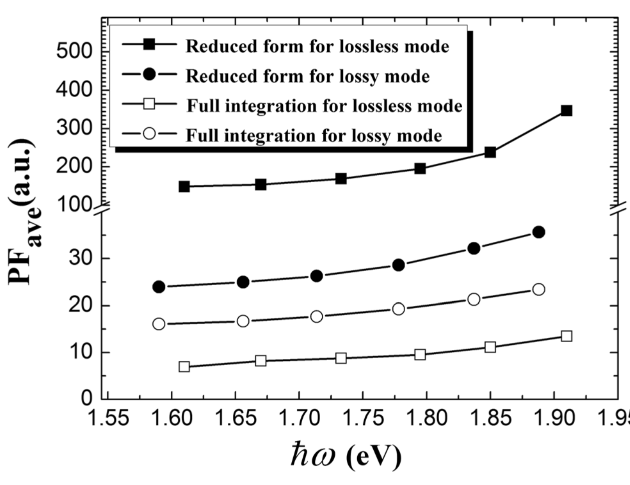

FIG. 3. The calculated PF_{ave} with reduced form (solid symbol) and full integration form (open symbol) for lossless (square) and lossy (circle) SPP mode with Au-Si_3N_4 grating period of $a = 80\,\text{nm}$–$130\,\text{nm}$.

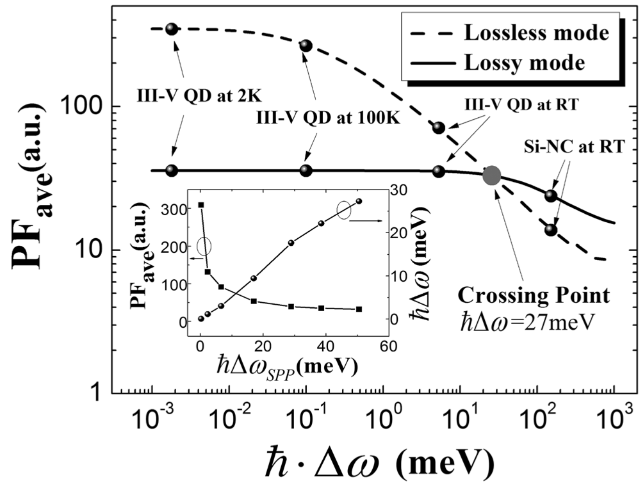

FIG. 4. The calculated PF_{ave} versus varied emission linewidth with constant central emission frequency $\hbar\omega_0 = 1.91$ eV (1.88 eV). The PF_{ave} and emission linewidth ($\hbar\Delta\omega$) corresponding to the crossing point versus varied linewidth of DOS spectrum ($\hbar\Delta\omega_{SPP}$) are also shown in the inset.

In order to clarify the impact of emission linewidth, the PF_{ave} is calculated with varied emission linewidth and fixed $\hbar\omega_0$ (1.91 eV/1.88 eV for lossless/lossy mode), as shown in Fig. 4. Furthermore, the PF_{ave} corresponding to some representative values of QD emission linewidth are marked as black dots. Besides Si-QD, the InGaAs/GaAs QD is considered as samples of narrow-linewidth emitter. Here, we adopted three values of $\hbar\Delta\omega = 1.8\,\mu\text{eV}$, $100\,\mu\text{eV}$, and 5.3 meV corresponding to temperature of 2 K, 100 K, and RT, respectively.[20]

In Fig. 4, the most important result is that there is a crossing point of the two curves at $\hbar\Delta\omega = \sim 27\,\text{meV}$. At the left side of this cross point, the PF_{ave} would approach the value predicted by the reduced form as the emission linewidth reduced. Thus, PF degradation is mainly due to DOS degradation/spectrum broadening for narrow-linewidth emitter (e.g., III-V QDs). However at the right side, PF_{ave} of lossy mode is larger than that of lossless mode. Typically, for $\hbar\Delta\omega = 152\,\text{meV}$, the PF_{ave}s for lossy and lossless mode are 23.4 and 13.5, respectively. Such result indicates that the SPP mode propagation loss is helpful to obtain higher SE enhancement for wide-linewidth emitter (e.g., Si-QDs). It could be understood that the broadening of DOS spectrum would enlarge the integration span and counterbalance the impact of reduced DOS value.

To clarify how this crossing point is influenced, the linewidth of DOS spectrum ($\hbar\Delta\omega_{SPP}$) is varied by multiplying the imaginary part of the metal permittivity with a factor of 1–10^{-3} and the corresponding emission linewidth ($\hbar\Delta\omega$), and PF_{ave} are calculated and shown in the inset in Fig. 4. As $\hbar\Delta\omega_{SPP}$ decreases, the $\hbar\Delta\omega$ of crossing point would be decreased with nearly a linear relation and the slope is $\sim$0.55. This result suggests that emission linewidth should be matched to the linewidth of DOS spectrum (nearly half of $\hbar\Delta\omega_{SPP}$) to obtain maximum PF. Otherwise, the SE enhancement would be degraded by the wide emission linewidth. Meanwhile, the corresponding PF_{ave} would be increased with decreasing $\hbar\Delta\omega_{SPP}$. Especially, PF_{ave} would be larger than 100 when $\hbar\Delta\omega_{SPP} < 1\,\text{meV}$. Thus, it could be concluded that both the emission linewidth and propagating loss of SPP mode are considerable limitations for ultra-high PF which account for the fact that the PF values of only a few to several tens are experimentally reported for metallic gratings[6,7,21] or other SPP systems.[3,4,22] Although only the case of metallic grating is considered in this work, there would be similar impact on other plasmonic structure. For the same reason, low temperature Si-emitter may be of interest to achieve very large PF since both the emission linewidth of single Si-QD and the ohmic loss of metal material could be reduced at low temperature.[16,23] We are expecting some experimental work related to it.

In conclusion, the plasmonic enhancement of one dimensional Au-Si_3N_4 grating is evaluated by the PF with full integration formula, in which both the linewidth of emission and DOS are involved (Eq. (3)). By comparing the evolutions of PF with varied emission linewidth for lossless and lossy SPP mode, it is found that both the emission linewidth of QD and propagating loss of SPP mode are physical limitations to obtain large plasmonic enhancement. For emitter with narrow linewidth, the DOS broadening due to propagation loss of SPP mode is dominant. But for emitter with wide linewidth, DOS broadening would be helpful to some degree.

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