Spontaneous Emission of Electric and Magnetic Dipole Transitions in Plasmonic Gratings and Strips Arrays

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Abstract: Spontaneous emission of Eu^{3+} is studied in the vicinity of gold gratings and nanostrip arrays. Radiation patterns of magnetic and electric transitions show features associated with plasmonic excitations and a narrow reflection anomaly in the strips.
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Spontaneous emission of an emitter is significantly modified in the modified optical environment as commonly discussed in terms of the Purcell effect [1], considering the changes in the local density of photonic modes affecting the dipole emission rates. Depending on a particular modification, electric and magnetic dipoles can be affected in a different manner. In the vicinity of flat metal, both electric and magnetic dipole transitions will experience an acceleration of the transition rates, with the effects being much stronger for electric dipoles [2]. In gold nanoholes exhibiting magnetic resonance at optical frequencies, the magnetic dipole emission is enhanced in comparison to the electric dipole transition [3]. Another important factor is related to coupling of an emitter to propagating waves. In close vicinity of flat metal, quenching of the emission is observed for the distances of 15-30 nm depending on the thickness of the metal film [2]. In this work, we experimentally study the emission of Eu^{3+} in the vicinity of the gold gratings and gold strips, which provide efficient decoupling of the surface plasmons at the particular angles determined by the sample periodicity.

Schematics of the experimental structures and results of the optical characterization are shown in Figure 1. Both samples were fabricated using interferometric lithography technique [4]. The arrays of gold strips on glass substrates were made by forming the pattern on the top of metal, and transferring it to the metal using reactive ion etching. Gold gratings were fabricated by forming a photoresist pattern on glass and depositing the metal onto the pattern.

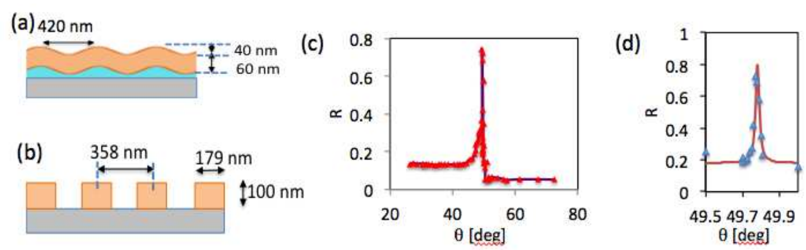

Fig. 1. Schematics of the gratings (a) and strip array (b). (c) Reflectivity peak observed in strips. (d) Narrow feature in the strip reflectivity observed at θ= 49.75 degree.

The continuous grating (Fig. 1(a)) supports SPPs at metal-air interface at $k_{spp} = -2\pi/d_g + k_0 \sin\theta$, where θ is the incidence angle, manifesting with a typical dip [5] in the reflectivity. According to the experiments, the ratio $\xi = k_{spp}/k_0$ is very close to what is expected in flat films, $\xi = [(\varepsilon_m \varepsilon_d)/(\varepsilon_m + \varepsilon_d)]^{1/2}$, where ε_m and ε_d are correspondingly the permittivities of metal and air. The strip array demonstrated a peak as expected [5] with an additional extra sharp feature (Fig. 2 (c)), with the angular width of ~ 0.04 degrees. This sharp feature corresponds to $k_0 = 2\pi/d_g - k_0 \sin\theta$ (bright-Rayleigh-Wood anomaly [6]).

In the emission measurements, we used a highly luminescent material, $Eu(NO_3)_3.Bpy_2$, synthesized in house [7]. This material can be efficiently excited at UV wavelengths, with a maximum of absorption around 330 nm, and demonstrates emission in visible wavelengths. The strongest line in the spontaneous emission spectrum of this material is observed at ~ 613 nm, corresponding to the electric dipole transition, while the transition at 590 nm is predominanetly magnetic dipole reated, Fig. 2 (a). A thin film of Eu^{3+} in polyvinylpyrrolidone (PVP) (10wt %), approximately 70 nm thickness, was deposited on the top of the structures using the spin coating technique. The sample was mounted on the rotating stage, and illuminated at λ = 325 nm through the optical fiber from a He-Cd laser. The emission intensity was recorded as a function of the observation angle with a photomultiplier detector. Narrow band interferometeric filters centered at 590 nm or 610 nm were placed before the detector in order to separately record the magnetic or electric dipole emission correspondingly. A polarizer was introduced in the detector setup to distinguish effects at different polarizations.

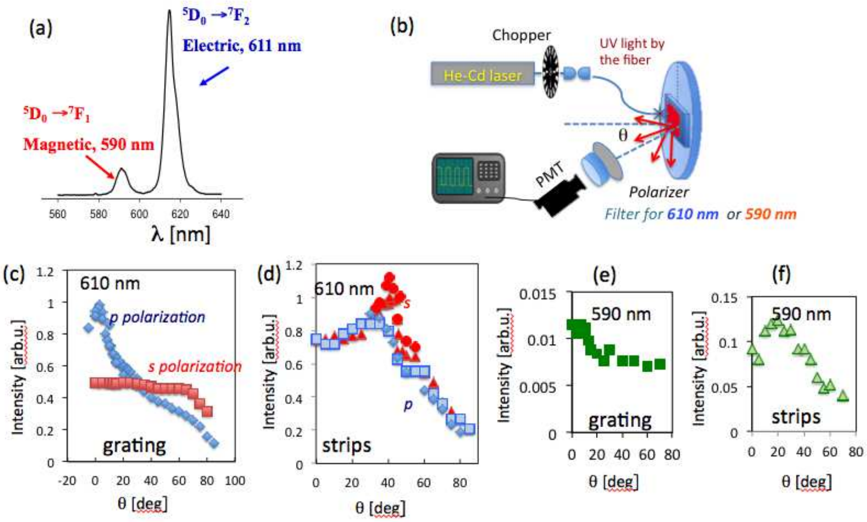

Fig. 2. (a) Spectrum of emission in $Eu(NO_3)_3.Bpy_2$; (b) Setup of the experiment; (c)-(f) Angular dependences of the emission at electric dipole and magnetic dipole transitions as indicated.

The results are shown in Fig. 2(c)-(f). In gratings, the p-polarized emission at 610 nm demonstrates a maximum at the SPP decoupling angle, while s-polarization is almost constant for a broad range of angles. In strips, peaks are observed for both polarizations, with the sharp maximum at s-polarization with the observation angle of ~ 42 degrees, which is close to the range of the narrow feature observed in the reflection spectrum. An enhancement of the magnetic dipole emission is observed at the SPP decoupled angle for both polarizations.

The results are analyzed considering modifications of the transition rates, coupling of emitters to collective plasmonic excitations and decoupling to propagating waves.

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References

[1] E. M. Purcell, “Spontaneous emission probabilities at radio frequencies”, Phys. Rev. **69**, 681 (1946).

[2] R. Hussain, D. Keene, N. Noginova, M. Durach, “Spontaneous emission of electric and magnetic dipoles in the vicinity of thin and thick metal”, Optics Express **22**, 7744 (2014).

[3] R. Hussain et al., “Enhancing Eu^{3+} magnetic dipole emission by resonant plasmonic nanostructures”, Opt. Lett. **40**, 1659 –1662 (2015).

[4] W. Fan, S. Zhang, K. J. Malloy, and S. R. J. Brueck,"Large-area,infrared nanophotonic materials fabricated using interferometric lithography," J. Vac. Sci. & Technol. **B 23**, 2700 (2005).

[5] N. Noginova, S. Mashhadi, V. Rono, M. LePain, M. Durach, “Collective plasmonic oscillations in nanostrips arrays and continuous sine-wave gratings. Comparative study”, CLEO: QELS-Fundamental Science, FW4B. 6 (2016).

[6] H. Gao, J. M. McMahon, M. H. Lee, J. Henzie, S. K. Gray, G. C. Schatz, and T. W. Odom, “Rayleigh anomaly-surface plasmon polariton resonances in palladium and gold subwavelength hole arrays”, Opt. Express **17**, 2334 (2009).

[7] N. Noginova, Y. Barnakov, H Li, M.A. Noginov, “Effect of metallic surface on electric dipole and magnetic dipole emission transitions in Eu^{3+} doped polymeric film”, Optics express **17**(13), 10767-10772 (2009).