Recent progress in emittance-controlled optical metasurfaces

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Abstract. Emittance is one of the fundamental optical quantities characterizing optical metasurfaces. Optical metasurfaces with high emittance have been demonstrated to be very efficient for enhancing light emission of fluorescent molecules and rare-earth ions, strongly suggesting that the metasurfaces are useful to make light-emission materials more efficient. In particular, we found so far that stacked complementary metasurfaces are able to exhibit thousand-fold larger emission intensity in a quite uniform manner than non-enhancing bulk substrates like Si. We survey our recent progress in the high-emittance metasurfaces.

1. Introduction
Metasurfaces are very thin artificial single-layer or a-few-layer structures fabricated on substrates, and are attracting growing interest in their diverse functions such as ray control [1, 2], anti-reflection [3], thermal emission [4–7], polarization and phase control [8–10], holograms [11], enhanced spectroscopy [12], hot electrons [13, 14], and so on.

Here, we focus on recent progress in emittance-controlled optical metasurfaces, and address the strategy and achievements to date. The basic idea is similar, in part, to thermal radiation. It is widely known that blackbody (that is, perfect absorber) is the most efficient thermal emitter because the study on blackbody, so-called Planck radiation, was a trigger of quantum physics. Due to reciprocity, light absorptance is equivalent to emittance [15]; therefore, perfect light absorber is an essential criterion to design ideal emitters. Indeed, metasurface thermal emitters were produced, which met the criterion [7].

To compare the blackbody and optical metasurfaces of artificially designed resonances, let us consider a simple situation. Input energy $E_1$ is supplied by external light and output energy $E_2(<E_1)$ comes out from the objects. Figure 1 shows schematic energy diagrams of (a) blackbody and (b) discrete electromagnetic (EM) resonant states in optical metasurfaces. Figure 1(c) shows emittance spectrum corresponding to Figure 1(b).

In the blackbody, the input energy is entirely consumed by inner relaxation such as phonon and inelastic scatterings in the constituent materials. Therefore, resultant thermal emission exhibits broad spectrum, reflecting low density of photonic state, and it is, in principle, difficult to selectively emit photons of the particular energy $E_2$. In contrast, the optical metasurface is designed to have a discrete energy level at the $E_2$, where the emittance is large (almost 100% in Figure 1(c)). Therefore, it is possible to selectively emit photons at the $E_2$. This scheme is intuitively understood in thermal emissions; however, this strategy has been proved to be valid for luminescence emission in the metasurfaces [16–19].

We briefly survey recent progress in emittance-controlled optical metasurfaces [16–20] in the next section: fluorescence (FL) enhancement in section 2.1, photoluminescence (PL) from rare-earth Er ions...
in section 2.2, and unconventional superlinear PL responses from quantum dots coupling with plasmonic metasurfaces in section 2.3.

2. Recent progress

2.1. Fluorescence enhancement in high-emittance metasurfaces

Figure 2 shows a set of typical results for enhanced FL on a stacked complementary (SC) metasurface, which is illustrated in Figure 2(a) and consisted of SC Au layers and perforated silicon-on-insulator (SOI) layer [18]. FL molecules of IR783 (Sigma-Aldrich Inc., USA) were dispersed on the SC metasurface in a quite uniform manner.

Figure 2(b) shows enhanced FL-molecule signals with solid red and dashed green curves, which were measured on the SC metasurfaces with and without 10-carboxy-1-decanethiol (10-CDT) self-assembled monolayer (SAM) (Dojindo Laboratories, Japan), respectively. Reference FL signals are shown in Figure 2(c), which were measured on a flat Si wafer in an equivalent condition to Figure 2(b). From the comparison of the FL signals emitted from the SC metasurfaces and the reference, it is obvious that FL signals are highly enhanced on the SC metasurfaces. In Figure 2(b), for the Raman signals at about

![Figure 1](image1.png)

**Figure 1.** Schematics of energy diagrams of (a) blackbody and (b) EM resonant states of discrete levels in metasurfaces. (c) Emittance spectrum corresponding to (b).

![Figure 2](image2.png)

**Figure 2.** (a) Schematic of optical configuration on the SC metasurface. (b) Enhanced FL spectra (solid red and dashed green curves) measured on SC metasurfaces with and without the SAM, respectively. We note that, on the SC metasurface without the 10-CDT SAM, substantial Raman signals are seen as narrow peaks. (c) Reference FL spectrum of IR783 on Si wafer, measured in an equivalent condition to (b). Adapted with permission from [18].
880 nm that are estimated to be 10000 counts, the Raman signal in the reference spectrum is less than detection limit, which means less than 5 counts; therefore, enhancement factor of the Raman signal was evaluated to be more than 2000. Also, for the FL signal at 1000 nm in Figure 2(b), the enhancement factor was estimated to be 1400. Thus, the FL-intensity enhancing capability of the SC metasurfaces is prominent, being one of the best among the numerous plasmonic structures reported to date [21, 22].

In particular, we stress that the enhanced FL and Raman signals appear at absorbance peaks, which are equivalent to emittance peaks due to reciprocity. Emittance is a macroscopic quantity of the metasurface. Thus, the experimental results support that highly enhanced optical signals from FL molecules are attained under emittance-controlled condition. Another successful example using a different FL molecule was reported on the SC metasurfaces [17], where enhancement factor was examined, in more details, in terms of electric-field intensity on the SC metasurfaces, lifetime of the FL, and the quantum yield of FL molecules.

2.2. Selective control of electric- and magnetic-dipole emissions from rare-earth ions

Figure 3(a) shows a schematic of one-dimensional periodic array of plasmon cavities, which are infinitely long along the y axis [19]. Figure 3(b) presents a set of EM-field intensity in the plasmonic cavity. Figure 3(c) shows reflection (solid green curve) and absorption (dashed black curve) spectra in the upper panel. The large absorption peak more than 90% was tuned to the emission wavelengths of Er ions around 1.53 \( \mu \)m. Figure 3(c) also shows electric- (red) and magnetic-field (blue) intensities in the lower panel. The intensities were taken at the positions A and B in Figure 3(b). Figure 3(d) shows scanning tunneling-electron-microscopy (STEM) images. Thin white lines indicated by red arrows represent Er-doped layer inside the plasmonic cavities. Red scale bars indicate 200 nm.

We set the Er-doped layers precisely at the maximum positions of electric- and magnetic-field intensities, and selectively enhanced electric-dipole (ED) and magnetic-dipole (MD) emissions in the Er ions [19]. The ED- and MD-emission-intensities were several-ten-fold enhanced, compared with Er-doped SiO\(_2\) films under an equivalent experimental condition. From the experimental data, we succeeded in extracting ED and MD emission spectra; moreover, Purcell effect was observed for both ED and MD emissions.
transitions. Recently, it was reported that outmost surface of a dielectric metasurface was controlled by liquid crystal to obtain PL of constituent material in a more efficient way [23]. The control was active while the resultant PL-intensity enhancement was limited to be about two-fold.

2.3. Plasmon–quantum-dot coupling systems

The strategy in the preceding subsections is considered to be valid for other luminescent materials such as semiconductors. However, highly prominent results similar to the FL molecules and rare-earth ions have not yet reported to date. This probably means that detailed and relevant designs for the metasurfaces including semiconductors have not yet been obtained and that interplay between metasurfaces and semiconductors is more complicated. Indeed, recent reports have shown that, in coupling systems of plasmonic metasurfaces and quantum dots (QDs), unconventional superlinear PL dynamics takes place due to hot electrons [14, 24].

We mention that efficient extraction of PL from semiconductor QDs has been attempted in many other platforms such as photonic crystals [25, 26], which can be regarded as a kind of metasurfaces when they are located at the surface of bulk substrates. The PL was prominently (or 200~1200-fold) enhanced at wavelengths of the large photonic density of states; broad PL spectrum coming from the Ge QDs was modified into PL spectrum of several narrow peaks [26]. In the configuration, there is no contribution from hot electrons, in contrast to the plasmon–QD coupling systems [14,24]; therefore, the enhancement mechanism of PL from the Ge QDs is ascribed to the photonic modes of large density of states.

3. Summary

We described our recent progress in emittance-controlled optical metasurfaces. It turns out that emittance, which is a macroscopic quantity featuring metasurfaces, plays a key role in significant enhancement effects on dipole emitters, irrespective of electric and magnetic components.

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