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Surface Plasmon Polariton Cross-Coupling Enhanced Forward Emission from Insulator–Metal-Capped ZnO Films

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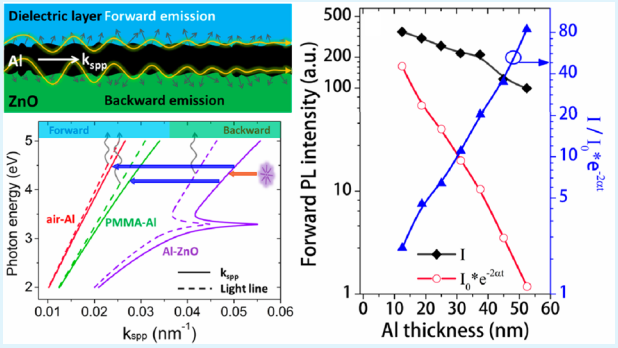
ABSTRACT: Increasing light extraction efficiency in the forward direction is being extensively pursued due to its crucial role in realizing top-emitting organic and inorganic light emitting devices. Various surface plasmon polariton (SPP)-based strategies for emission enhancement and light extraction have been developed to improve the top-emitting efficiency of these devices. However, the role of surface roughness of both semiconductor film and metal electrode in improving the emission efficiency of a practical device has not been thoroughly studied yet. In this work, the influence of surface roughness of a top metal electrode on the photoluminescence enhancement of a ZnO thin film is investigated experimentally and numerically based on an insulator–metal–semiconductor system. It is found that the generic surface roughness of the metal electrode plays an encouraging role in increasing the forward-emission intensity by facilitating cross-coupling of SPPs on the two opposite sides of the metal layer. More importantly, the forward emission can be further enhanced by capping a high-index polymer layer on the metal electrode to bridge the momentum mismatch between the two SPPs modes. The experimental observations are well explained by the SPPs cross-coupling mechanism that models the radiation power of a dipolar emitter underneath the metal electrode as a function of the metal surface roughness. Our work opens up the possibility of using cross-coupling of SPPs as an effective means to fabricate high-brightness top-emitting devices without the need of complicated nanoscale patterning.



KEYWORDS: *ZnO film, photoluminescence, surface plasmon polariton, roughness, cross-coupling, momentum matching*

INTRODUCTION

Recently, zinc oxide (ZnO) has been considered as one of the most promising ultraviolet (UV) light emission materials due to its wide band gap (3.37 eV) and large excitonic binding energy (60 meV).[1,2] Many emission enhancement and light extraction strategies have thus been developed to increase the emission efficiency of ZnO-based light emitting devices (LEDs), and they mainly focused on enhancing the backward light emission through a transparent substrate into free space. For an opaque substrate or a top-emitting device, however, only the forward emission is preferable, which has stimulated extensive research interests in increasing the light extraction efficiency in the forward direction. Surface plasmon polaritons (SPPs) have been widely used to magnify the energy conversion efficiency in various light emission processes, such as photoluminescence (PL), fluorescence, and Raman spectrum.[3−7] It has been demonstrated that SPPs were able to enhance the emissions of organic and inorganic semiconductors by more than one order of magnitude.[8,9] In particular, several works have also demonstrated forward-emission enhancements via SPPs in metallic nanostructures.[10−13] Alternatively, cross-coupling between SPPs at two interfaces of a thin metallic film can be used to achieve the same purpose yet without the need of complicated nanoscale patterning of the semiconductor or metal layer.[9,14,15] For example, surface roughness, an unavoidable, generic feature of metal films, is often preferred to be erased since roughness-induced scattering will accelerate the decay of SPPs,[16,17] whereas it could become favorable to energy extraction in thin film emitting systems.[18] Yet, the role of surface roughness in light emission enhancement has not been thoroughly investigated, especially in the case of forward emission.

In this work, both backward and forward emissions of an insulator–metal-capped ZnO film are investigated by means of PL and transmission spectroscopy. Aluminum (Al) is chosen as the plasmonic metal due to its deep ultraviolate bulk plasmon energy.[19−22] It is found that the forward-emission intensity of this multilayer system far surpasses the prediction by the Beer–Lambert law. With systematic examinations of the emission intensities as a function of the metal film thickness and surface roughness, the unexpected forward-emission enhancement is attributed to the cross-coupling of SPPs existing at the

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insulator−metal and metal−ZnO interfaces. Significantly, surface-roughness-induced scattering plays a crucial role in extracting the energy carried by nonradiative SPP modes from near field to far field, which is consistent with the radiation power calculated for a dipolar emitter placed in the same dielectric environment (ZnO) underneath a rough metal film. This finding is firmly supported by the observation that the forward emission can be further enhanced by capping a high-index polymer layer on the Al film due to better momentum match between the SPP modes at the two interfaces of Al.

METHOD

ZnO films at a thickness 50 nm were first grown on fused silica substrate by using radio frequency (RF) magnetron sputtering at 550 °C and followed by Al film deposition at room temperature with thickness ranging from 12 to 48 nm.[9] To avoid the impurity AlO_x generated during deposition, the Al target was presputtered for 2 min to eliminate the residual oxide. All of the samples were then annealed at 500 °C in 5 Torr argon atmosphere. To investigate the influence of the top dielectric environment on forward emission, a layer of poly(methyl methacrylate) (PMMA) with thickness 360 nm was subsequently capped on the Al films by spin coating. The samples were then mounted on a manual rotation stage to measure both backward and forward emissions. A HeCd laser (325 nm) was used to excite the sample, and a CCD-coupled spectrometer was used to collect the PL signal. Transmission spectroscopic measurements were conducted with a Hitachi U350l UV−vis spectrophotometer. The surface roughness of the Al films was measured by atomic force microscopy (AFM). All of the measurements were carried out at room temperature.

Two-dimensional full-wave electromagnetic simulations were performed using the finite-difference time-domain (FDTD) method implemented in a commercial software package from LUMERICAL. The incoherent, isotropic spontaneous emission from ZnO was modeled by the radiation power from an unpolarized dipole source, which was generated by equally weighted averaging of the radiation powers from three discrete dipole sources polarized along *x*-, *y*-, and *z*-axes, respectively. In our calculations, the dipoles were positioned 25 nm away from the metal surface. Since the thickness of ZnO is much smaller than its emission wavelength, the phase difference between the direct radiation from the dipoles and the reflected power by the metal surface can be ignored. The surface-roughness values used in the simulations were obtained from the AFM measurements. For simplicity, the roughness at both interfaces of the Al film was set to be the same. The width of the simulation region was 20 μm. A power monitor was situated 10 nm away from the upper surface of the Al film to collect the forward radiation power. The dielectric permittivities of Al and ZnO were obtained from experimental data,[1,23] and the index of silica substrate was 1.45.

RESULTS AND DISCUSSION

In the insulator (air or PMMA)−Al-capped ZnO multilayer system, the SPPs at the Al−ZnO interface can be directly excited by the near-field component of the emission energy from ZnO and those at the Al−insulator interface can be excited via cross-coupling of near-field energy from the SPPs at the Al−ZnO interface.[24,25] When they propagate along the interfaces, surface-roughness-induced scattering could extract the energy carried by the nonradiative SPPs to far-field radiation,[26] as schematically shown in Figure 1a. To clearly understand the physical mechanisms involved in the SPP-mediated emission process, the calculated dispersion curves for air−Al−ZnO and PMMA−Al−ZnO multilayer systems are shown in Figure 1b. The emission energy from ZnO is coupled to the SPPs (solid purple line) at the Al−ZnO interface (orange arrow) rather than to other nonradiative channels, and

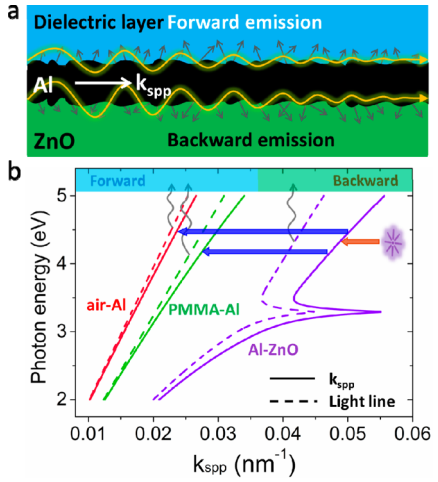

Figure 1. (a) Schematic of SPP excitation in an insulator−Al−ZnO multilayer system. The yellow arrows represent propagating SPPs, and the gray arrows represent energy extracted from SPPs to free space in either forward or backward direction. (b) Dispersion curves of surface plasmons (solid lines) at Al−ZnO (purple), PMMA−Al (green), and air−Al (red) interfaces and the corresponding light lines (dashed lines). Blue and orange arrows represent coupling of SPPs across Al and near-field coupling from ZnO to SPPs, respectively.

the SPPs propagating along this interface can either be scattered to far-field radiation (dashed purple line) by surface roughness or evanescently coupled to the SPPs (solid red and green curves) at the air−Al or PMMA−Al interface (blue arrows). On the one hand, such an SPP-mediated emission process is expected to significantly enhance the overall emission efficiency of ZnO due to the increased photonic density of states generated at the Al−ZnO interface. On the other hand, the near-field cross-coupling of SPPs from the Al−ZnO to air−-Al or PMMA−Al interface and subsequent scattering of the energy to the top side jointly contribute to the forward emission in addition to the direct transmission of the emission. One can see that at both interfaces surface-roughness-induced scattering plays an essential role in transforming the evanescent near-field energy of SPPs to far-field radiation energy. Thus, it is of paramount importance to study the influence of surface roughness on the emission enhancement in both directions.

To systematically investigate the influence of surface roughness on light extraction efficiency, Al films with thicknesses ranging from 12 to 48 nm were deposited on 50 nm thick ZnO films. Parts a and b of Figure 2 show the AFM images of two Al films at thicknesses of 21 and 48 nm, respectively. It is apparent that the film surface gets rougher and larger islands form in the thicker film. As plotted in Figure 2c, the measured root-mean-square (RMS) roughness ranges from 1 to 2.6 nm as Al thickness increases from 12 to 48 nm, while the averaged peak-to-valley value increases from 5.2 to 20.9 nm.

To verify the energy coupling and extraction processes shown in Figure 1b, both backward and forward PL emissions from the Al-capped ZnO films were measured. The backward PL spectra for Al films at different thicknesses are plotted in Figure 3a. The emission peak occurs at ∼3.25 eV (∼381 nm). The slight difference between the measured emission peak and the band edge emission of ZnO may arise from the roughness modification on the electronic and optical properties of the ZnO films.[27,28] It can be seen that a 15-fold enhancement, defined as the intensity ratio of the Al-capped ZnO film to bare

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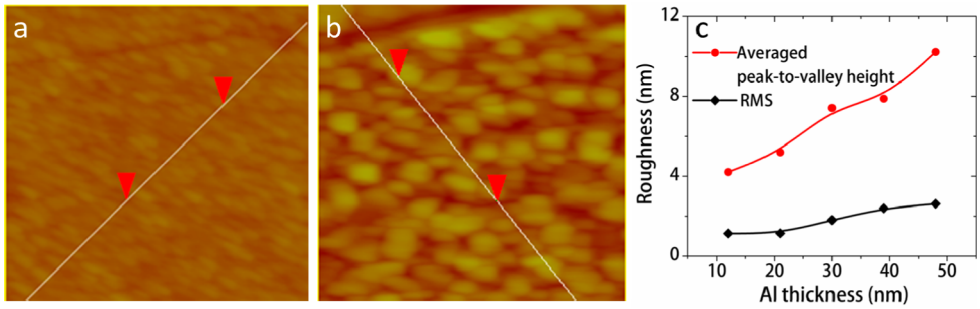

Figure 2. AFM images of two Al films at thicknesses (a) 21 and (b) 48 nm. (c) The RMS roughness and averaged peak-to-valley height against the thickness of Al film ranging from 12 to 48 nm. The solid lines are a guide to the eye.

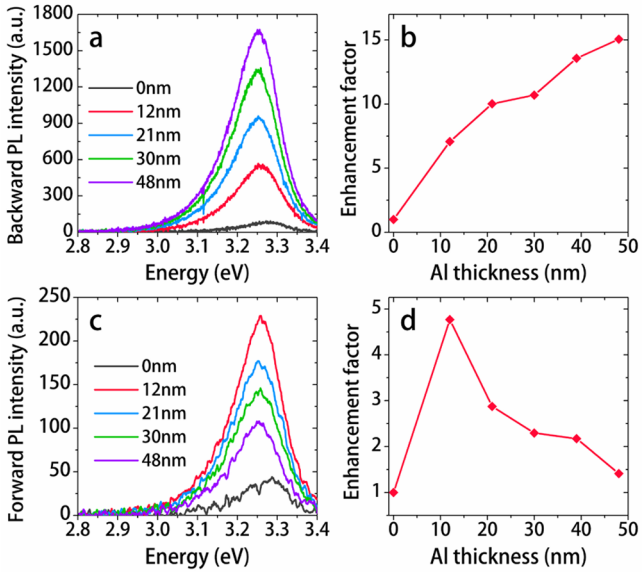

Figure 3. Backward PL emission (a) and its enhancement factors (b) of ZnO capped with Al at different thicknesses. The forward PL emission (c) and its enhancement factor (d). In b and d the red solid lines are a guide to the eye.

one at the emission peak, I_{Al-ZnO}/I_{ZnO}, is achieved at Al film thickness of 48 nm as shown in Figure 3b. Since the thickness of ZnO is much smaller than the emission wavelength, neither waveguide mode nor Fabry–Pérot interference exists in the system. As discussed earlier, in addition to direct radiation to far field contributing to the backward emission, the energy released from electron–hole pair recombination can also be coupled to the SPPs at the Al–ZnO interface through near-field coupling within the evanescent near-field zone at a much faster coupling rate than other nonradiative channels such as defect states.[29,30] When the SPP waves propagate along the Al–ZnO interface, they will be scattered randomly into free-space photons by surface roughness and thus lead to a strong emission enhancement in the backward direction.[8,11] By continuously increasing the thickness of Al film, the enhancement factor (EF) of the backward emission intensity increases monotonously as shown in Figure 3b. Such enhancement trend can be attributed to two factors, namely, larger light extraction efficiency due to stronger roughness scattering and more direct reflection both by thicker Al films. Notably, the former contribution dominates such enhancement.[14]

However, the forward emission shows some unexpected behaviors when varying the thickness of Al film. At the first glance, the forward PL intensity decreases monotonously as the Al thickness increases from 12 to 48 nm, as shown in Figure 3c, and the corresponding intensity EF follows the same trend in Figure 3d. Intuitively, the decrease in the forward emission intensity naturally arises from less transmission due to stronger absorption by thicker Al films. However, detailed analysis of the measured intensities shows an obvious deviation from the expectation by Beer–Lambert law, $I = I_0 e^{-2\alpha t}$, where I is the forward intensity, I_0 is the backward intensity, t is the thickness of ideally smooth Al films, and α is the absorption coefficient of Al at energy 3.25 eV.[24] The calculated intensities decrease much faster than the experimental counterparts as illustrated in Figure 4a. Furthermore, the ratio of the measured intensity to theoretical prediction gets larger for thicker Al films.

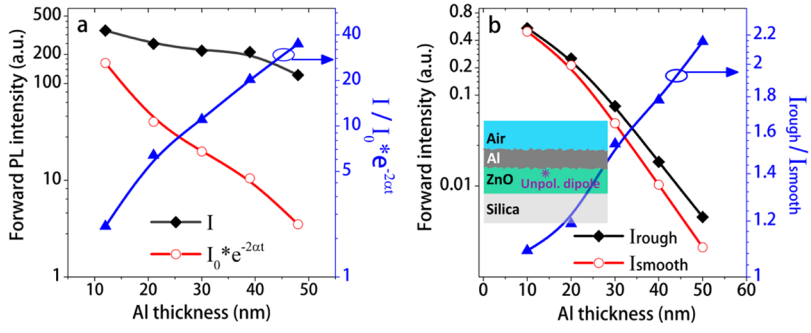

Figure 4. (a) Experiment measured (black diamonds) and calculated (red circles) forward PL intensities and their ratios (blue triangles) at emission peak 3.25 eV. (b) Simulated forward radiation powers for an unpolarized dipole emitter placed 25 nm underneath Al films of 10–50 nm thickness with (black diamonds) and without (red circles) considering surface roughness and their ratios (blue triangles). Inset schematically shows the system used in simulations. In both panels the solid lines are a guide to the eye.

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In order to elucidate the deviation between experiment and theory, we numerically calculated the forward radiation power of an unpolarized dipole emitter (oscillating frequency at 3.25 eV) placed 25 nm away from an Al film with thickness ranging from 10−50 nm at a step of 10 nm. To account for the influence of surface roughness, the forward emission intensities were calculated both in the presence and absence of surface roughness, where the roughness value was obtained by fitting AFM measurements shown in Figure 2c. As shown in Figure 4b, the simulated forward radiation intensity decreases with increasing of the Al film thickness in both cases while the intensity ratio of the rough and smooth surfaces increases, consistent with the experimental observations shown in Figure 4a. By fitting the forward radiation intensity with respect to Al film thickness, an absorption coefficient of 0.06 nm^{-1} is extracted in the presence of surface roughness and 0.07 nm^{-1} in the absence of surface roughness. It is evident that surface roughness does play a positive role in improving the forward radiation. In experiment, the fitted absorption coefficient is 0.015 nm^{-1}, which is much smaller than the absorption coefficient of 0.06 nm^{-1} obtained from the calculations.[1] Because the thickness of the Al film is larger than 10 nm in the experiment, Dexter or Förster resonance energy transfer is excluded in our case.[31] Therefore, the underlying mechanism is clear that the strong coupling of SPPs at the two opposite interfaces of the thin Al film substantially contributes to the enhancement of the forward emission. However, it is noticed that there is still a deviation between roughness-induced emission enhancements in the simulations and experiments; that is, the intensity ratio in Figure 4a is much larger than that in Figure 4b for the same Al film thickness. This may arise from some important factors that have been neglected in the ideal roughness-modified multilayer system in simulations, such as nonuniformity of each layer and grain boundaries of the Al films. It has been shown that grain boundaries in deposited metal films can increase the possibility of electron scattering at those inner boundaries and lead to an increased absorption decay rate,[32] which reduces the total external quantum efficiency. It is consistent with the larger enhancement ratio in experiment, especially for a thicker Al film with more grain boundaries.

In general, momentum match facilitates more efficient energy exchange between SPP modes. As discussed earlier, the enhanced forward emission mainly arises from cross-coupling of the SPPs at the two interfaces of the Al film. However, such coupling effect cannot take place efficiently due to the large index difference between air and ZnO film that generates large momentum gap between the two SPP modes, as shown Figure 1b. In turn, capping the Al film with PMMA can shift the dispersion curve of SPP at the air−Al interface toward that at the Al−ZnO interface. The alleviated moment mismatch thus leads to an increase in forward emission.[31] In experiment, a layer of 360 nm thick PMMA with refractive index of 1.49 was spin-coated at 30 nm thick Al film and both backward and forward emission spectra of the PMMA-Al-ZnO sample were measured, with results shown in Figure 5a,b, respectively. It is clear that the introduction of PMMA on Al−ZnO has negligible effect on the backward emission since it is completely independent of the dielectric environment at the upper surface of Al. In sharp contrast, the forward emission from the PMMA−Al−ZnO sample is dramatically enhanced compared to that from the air−Al−ZnO sample. Furthermore, the ratio of the forward emission intensities for both systems at the emission peak plotted in Figure 5c increases monotonously with Al film thickness due to the increased surface roughness. This is further supported by the measured transmittance spectra for both systems which show an enhanced transmittance for the PMMA−Al−ZnO across the entire spectrum of interest as shown in Figure 5d. Therefore, it can be concluded that the stronger forward emission and higher transmittance can be attributed to the assistance of cross-coupling of SPPs arising from the two interfaces of the Al film. Due to the presence of surface roughness, the SPPs at both interfaces can couple and scatter nondispersively, as evidenced by the transmittance enhancement over the entire spectrum.

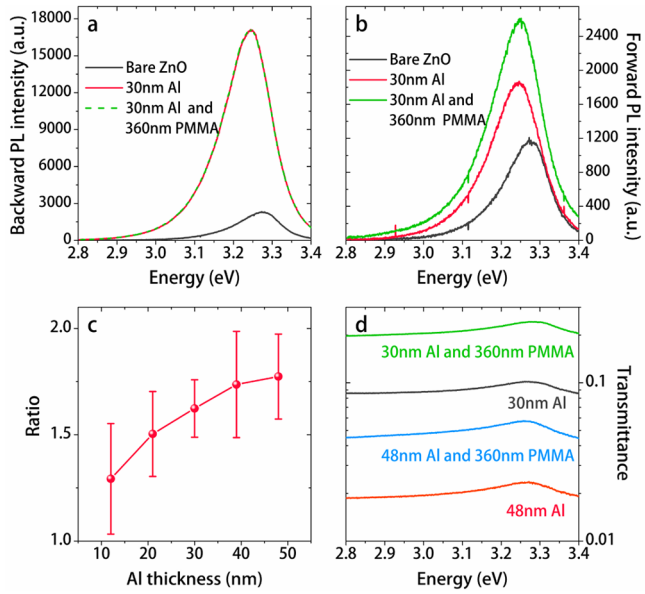

Figure 5. Measured backward (a) and forward (b) emission spectra of ZnO capped with 30 nm Al and 360 nm PMMA. (c) Calculated intensity ratio of forward emissions for the PMMA−Al−ZnO and Al−ZnO samples at 3.25 eV. The red solid line is a guide to the eye. (d) Measured transmittance spectra for two sets of PMMA−Al−ZnO and Al−ZnO samples having 30 and 48 nm thick Al films, respectively.

CONCLUSION

In summary, we have studied the SPP-mediated emission from insulator−metal-capped ZnO thin films by PL and transmission spectroscopy. By choosing Al as a plasmonic material to match its SPP resonance energy with the emission band of ZnO, we have experimentally and numerically shown that the forward emission can be substantially enhanced through coupling of SPPs across the Al layer. Significantly, our results demonstrate that the generic surface roughness of Al (in general top metal electrodes in light emitting devices) turns up to be favorable, rather than unwanted, in enhancing the PL emission of ZnO, particularly for the forward emission. By further capping Al−ZnO with a high-index polymer layer (also functioning as a protection layer for the metal electrode), the forward emission from Al−ZnO could be further enhanced. Our study provides an efficient means for realizing SPP cross-coupling mediated forward emission and could lead to the development of high-brightness top-emitting light emitting diodes and lasers.

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Author Contributions

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Notes

The authors declare no competing financial interest.

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