

Plasmonic off-axis unidirectional beaming of quantum-well luminescence

Jeff DiMaria, Emmanouil Dimakis,^{a)} Theodore D. Moustakas, and Roberto Paiella^{b)}

Department of Electrical and Computer Engineering and Photonics Center, Boston University, Boston, Massachusetts 02215, USA

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Plasmonic off-axis unidirectional beaming of luminescence is demonstrated using nitride semiconductor quantum wells. The underlying mechanism involves the near-field excitation of surface plasmon polaritons on an ultrathin metal film, which are then diffractively scattered by an adjacent periodic array of asymmetric metallic nanoparticles. By tailoring the nanoparticles shape, we show that forward scattering can be suppressed in favor of backward diffraction (or vice versa), thereby enabling unidirectional beaming at geometrically tunable oblique angles. These nanostructures can be used to control the output light directionality of arbitrary planar luminescent devices, with a spatial resolution that would be unattainable with bulk optics.

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The far-field radiation pattern of an optical emitter, which describes the directional dependence of its output light intensity, is a fundamental property with important practical implications. In the case of a continuous distribution of randomly oriented dipoles near a planar surface, which provides an accurate description of many luminescent devices of interest, light is emitted isotropically in all directions, leading to a broad Lambertian (i.e., cosine-law) distribution in the far field. Enhanced directionality is commonly obtained using external optical elements (e.g., lenses, curved mirrors, and diffraction gratings), whose bulky nature however provides critical limitations, particularly with regards to spatial resolution. Metallic nanostructures directly fabricated on the emissive surface offer new opportunities to overcome these limitations, by virtue of the highly localized nature of their plasmonic excitations and their strong coupling to luminescence processes.^{1,2} Specifically, the radiative decay rate of light emitters located within the near field of these nanostructures can be strongly increased through the excitation and subsequent scattering of resonant plasmonic oscillations.^{3–10} Under these conditions, the far-field properties of the emitted light are then determined by the nature of the plasmonic excitations involved in the emission process, rather than of the original radiating species.

In this work, we investigate the use of plasmonic nanoantenna arrays to control the far-field radiation pattern of Lambertian light emitting samples of arbitrary spatial extent. Within this context, we demonstrate a key beam shaping functionality, namely, the ability to generate a well collimated optical beam that emerges from the sample surface along a single geometrically tunable direction (off-axis unidirectional beaming). The underlying physical mechanism involves the excitation of propagating surface plasmon polaritons (SPPs) on an ultrathin metal film, which are then diffractively scattered by an array of asymmetric metallic nanoparticles periodically arranged on the film. By tailoring the shape of these nanoparticles, we show that forward

diffractive scattering can be suppressed in favor of backward diffraction (or vice versa), thereby enabling the demonstration of single-beam emission along arbitrary predefined directions.

These results build on prior work where metallic or dielectric diffraction gratings were employed to directionally outcouple in-plane guided modes from light emitting diodes (LEDs), lasers, or even thermal emitters.^{11–17} However, in all these reports only symmetric radiation patterns were produced, consisting of either a single beam along the sample surface normal, or of two or more directional beams at equal and opposite angles. In contrast, in the present work we show that the shape of the unit cells can be designed to selectively enhance emission towards one side of the grating relative to the other, so as to produce (asymmetric) unidirectional beaming at oblique angles. This approach is also fundamentally different from recent reports of plasmonic beam shaping from nanoscale sources (such as individual molecules and quantum dots) or otherwise highly localized emissive areas (e.g., confined within a nanoscale aperture in a metal film), which rely instead on an optimized positioning of the emitter relative to a single antenna or metallic grating.^{18–23} In contrast, the present approach can be applied to a continuous dipole distribution over an arbitrarily wide sample area (including the characteristic dimensions of typical optoelectronic devices), thanks to the extended nature of the SPPs involved in the beamed emission.

The specific light emitting material used in this work consists of three 2-nm-thick InGa_N quantum wells (QWs) separated by 5-nm-thick Ga_N barriers, grown by molecular beam epitaxy on a Ga_N template on sapphire. This family of semiconductor materials in general has important optoelectronic device applications, such as visible LEDs, which could benefit strongly from the unique properties of plasmonic excitations. In fact, the use of metallic nanostructures to enhance the radiative efficiency of InGa_N LED materials has already been widely investigated.^{24–30} The samples employed in the present work provide emission centered around 495 nm in the blue-green spectral region. In the absence of plasmonic interactions, their radiation output is relatively unpolarized with an internal quantum efficiency of about 8%,²⁹ and their

^{a)}Present address: Institute of Ion-Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany.

^{b)}E-mail: rpaiella@bu.edu

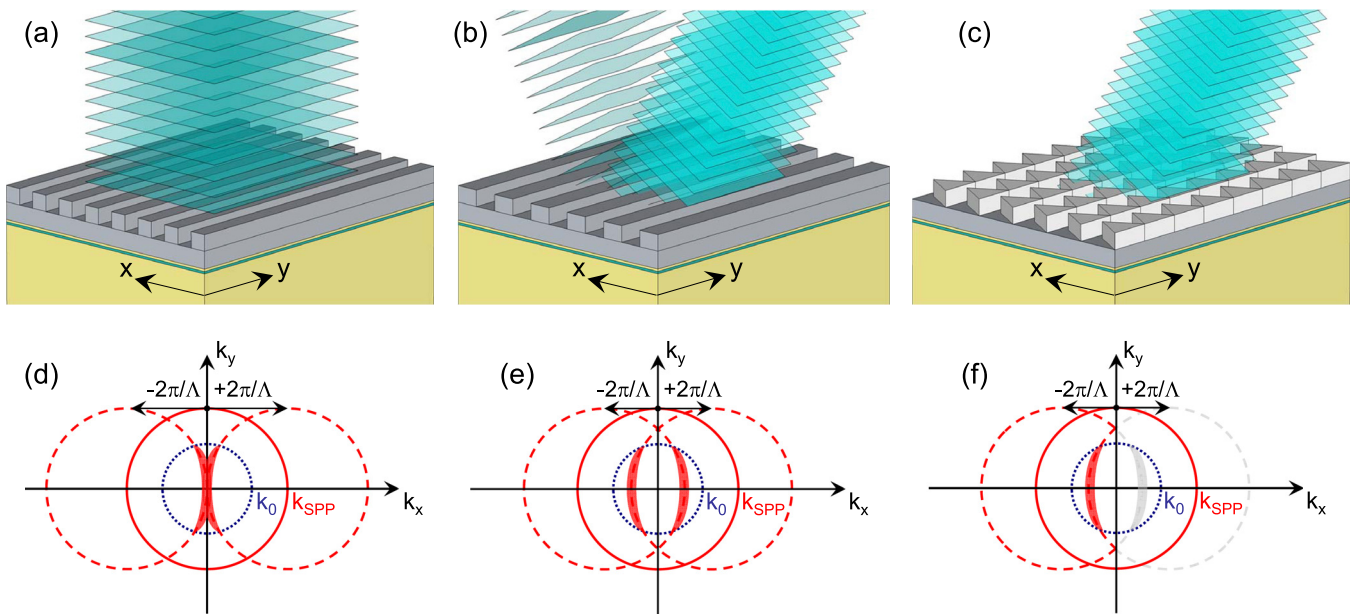


FIG. 1. Plasmonic nanostructures designed to provide beaming of luminescence (a) along the sample surface normal, (b) along two equal and opposite directions, and (c) along a single oblique direction. (d)–(f) SPP mode distribution on the k_x – k_y reciprocal plane (solid red circle) and its replicas created by first-order diffraction (dashed red circles) in the structures of (a)–(c), respectively. The thicker line segments indicate the SPP modes diffracted inside the light cone of free space (whose boundary is shown by the dotted blue line). The faint lines in (f) represent diffracted modes with negligibly small amplitude.

surface emission features a characteristic Lambertian profile. To enable plasmonic collimation and beaming, the sample top surface is coated with an ultrathin Ag film deposited by electron-beam evaporation, which is then used as the substrate for the subsequent fabrication of the nanoparticle arrays via electron beam lithography, Ag evaporation, and liftoff. The InGaN QWs in the active layer are only capped by a very thin (5 nm) epitaxial layer (i.e., the topmost GaN barrier), so that they all lie within the near field of the overlaying Ag film.

In this basic geometry [illustrated schematically via three examples in Figs. 1(a)–1(c)], the available plasmonic excitations are derived from the SPPs guided at the two surfaces of the film and from the surface-plasmon resonances of the nanoparticles (although the latter modes here are spectrally detuned from the QW emission). In the samples investigated in this work, we use ultrathin (40-nm) Ag films with no dielectric spacer layer between the film and the overlaying array. As a result, the aforementioned basic excitations are strongly coupled to one another leading to the formation of hybrid plasmonic modes, as described in Refs. 31–33. Collimation and beaming in these samples relies on the excitation by the QW electron-hole pairs of plasmonic modes that are primarily derived from the SPPs at the Ag-air interface. Due to their strong spatial coupling with the array nanoparticles, these modes can be diffractively scattered into free-space radiation with high efficiency. At the same time, due to their hybridization with the SPPs at the Ag-GaN interface, the same modes also feature appreciable spatial overlap with the QWs and therefore can be efficiently excited by the radiating electron-hole pairs. As a result, they also provide an effective means for the extraction of luminescence across the metal film.¹⁵

The ability to obtain highly directional emission from these SPPs (as opposed to directly radiated photons) is

related to the in-plane orientation of their wavevector and its constant magnitude k_{SPP} at the QW emission wavelength λ_0 . To introduce the proposed nanostructures, it is instructive to consider first the simpler case of a one-dimensional grating, as shown in Figs. 1(a) and 1(b). If the grating period Λ is chosen so that $2\pi/\Lambda \approx k_{SPP}$, first-order diffraction of the excited SPPs results in (one-dimensional) beam collimation along the sample surface normal [Fig. 1(a)]. This process is detailed in Fig. 1(d), where k_x and k_y are the in-plane wavevector components, and the concentric circles of radii $k_0 = 2\pi/\lambda_0$ and k_{SPP} represent, respectively, the boundary of the free-space light cone and the set of excited SPP modes. Through first-order diffraction, the SPP wavevector can be shifted by the amount $\pm 2\pi/\Lambda$ along the x direction (perpendicular to the grating lines), as shown by the dashed circles in the figure. If $2\pi/\Lambda \approx k_{SPP}$, the SPP modes diffracted inside the light cone and therefore efficiently coupled to free-space radiation (drawn with a thick line) are all mapped near the k_y axis. As a result, the energy carried by these modes is radiated in a well collimated beam with negligibly small divergence with respect to the y - z plane. Vice versa, if the grating period is detuned from the SPP wavelength $2\pi/k_{SPP}$, the SPP modes diffracted into the light cone retain a non-negligible x component of their wavevector, as shown in Fig. 1(e). In this case [Fig. 1(b)], diffraction of forward and backward propagating SPPs leads to the emission of two output beams along equal and opposite angles with respect to the y - z plane, given by $\theta_{\pm m} = \pm \sin^{-1}(\frac{k_{SPP} - 2m\pi/\Lambda}{k_0})$ with $m=1$ for first-order diffraction. These ideas have also been investigated with the light emitting material used in this work.³⁴

Within this framework, here we demonstrate the use of more complex array geometries to obtain single-beam plasmonic collimation along geometrically tunable oblique directions. Specifically, if grating lines with an asymmetric shape in the x direction are employed [e.g., as the lines of

triangular nanoparticles of Fig. 1(c)], backward diffractive scattering can be enhanced relative to forward diffraction (or vice versa), leading to predominant emission along a single direction. It should be emphasized that the array geometry here is designed explicitly to introduce a strong asymmetry in the scattering cross section of the individual unit cells (as opposed to selectively suppress specific diffraction orders as reported, e.g., in Ref. 35), which is essential in order to enable off-axis unidirectional beaming. The underlying mechanism is illustrated schematically in Fig. 1(f), where the faint lines represent diffractively scattered waves with negligibly small amplitude. These arrays could be described as an ultrathin planar implementation of a blazed grating, designed to operate with SPPs and suitable for integration with light emitting devices on a scale that could not be achieved with bulk optics. A similar geometry (i.e., an array of triangular holes in a metal film) has also been proposed recently as a means to enable the asymmetric excitation of SPPs with external light at normal incidence.³⁶

As illustrated by the scanning electron microscopy (SEM) image of Fig. 2(a), the arrays developed in this work consist of Ag nanoparticles having the shape of x -oriented isosceles triangles, partly overlapping in the y direction (patterned on an ultrathin Ag film on the light-emitting surface).

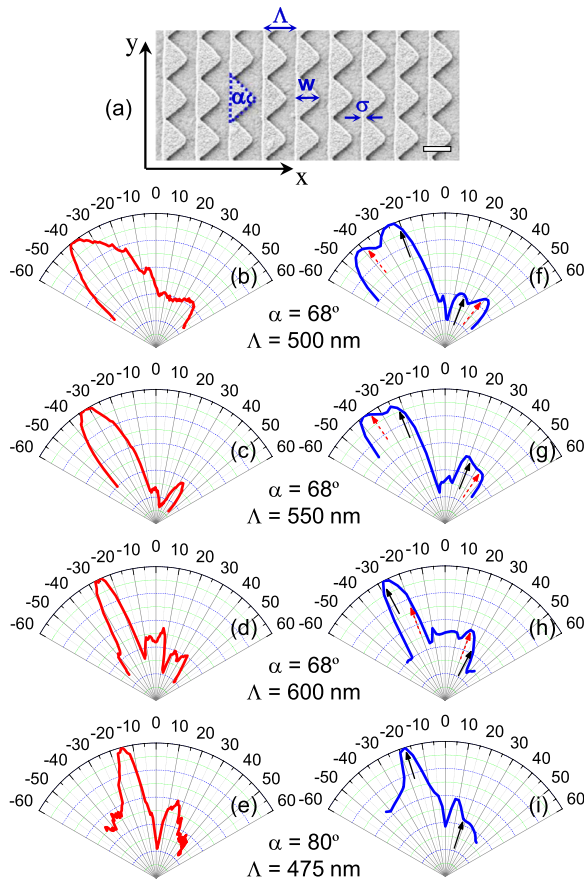


FIG. 2. Plasmonic off-axis unidirectional beaming with the Ag nanoantenna arrays of Fig. 1(c). (a) SEM image of a representative sample (the scale bar is 500 nm). (b)–(e) Experimental p -polarized far-field radiation patterns measured with four different arrays. (f)–(i) Numerical simulation results for the same array geometries of (b)–(e), respectively. The solid black and dashed red arrows indicate the diffraction angles $\theta_{\pm 1}$ and $\theta_{\pm 2}$, respectively, as obtained from numerical fits.

The asymmetry in their scattering cross-section along the x direction is governed by the triangles size and shape, which can be specified by means of the geometrical parameters α and w defined in Fig. 2(a). Their mutual overlap, quantified by the parameter σ , is advantageous for the purpose of increasing the overall diffraction strength. The far-field radiation patterns of these samples (i.e., emitted light intensity versus angle θ) were obtained via angle-resolved photoluminescence measurements, with the sample region underneath the array pumped at normal incidence through the backside using a 375-nm diode laser. The resulting photoluminescence signal was then measured from the metal-coated top surface, with the collection optics rotated about the excitation spot. The same radiation patterns were also computed with finite difference time domain (FDTD) simulations based on the principle of reciprocity.³⁴

In the polar plots of Figs. 2(b)–2(d) we show the measured p -polarized far-field intensity patterns of three samples featuring the same nanoantenna array geometry (with $w = 410$ nm, $\sigma = 50$ nm, $\alpha = 68^\circ$, 60-nm particle height, and 40-nm film thickness) except for different periods Λ in the range of 500–600 nm. In all samples, the s -polarized emission is substantially weaker than the p -polarized component, and as a result only adds a small pedestal to the overall intensity profile (see Fig. 3 below for more details). Strongly asymmetric radiation profiles are clearly obtained from these structures, with most of the output light emitted in a single beam at a nonzero angle, which can be tuned by varying Λ .

A more detailed understanding can be obtained from the corresponding FDTD simulation results plotted in Figs. 2(f)–2(h). These theoretical radiation patterns reveal the presence of several peaks associated with different diffraction orders (including weak contributions near $\theta = 0^\circ$ that also involve the array periodicity in the y direction). The dominant emission peak is generally due to first-order diffraction along x whereby the SPP wavevector increases by $2\pi/\Lambda$ in the positive x direction. At the same time, second-order diffraction along x whereby the SPP wavevector decreases by $4\pi/\Lambda$ also produces an emission peak of comparable size at a nearby angle. To confirm this interpretation, the solid and dashed arrows in Figs. 2(f)–2(h) indicate the diffraction angles $\theta_{\pm 1}$ and $\theta_{\pm 2}$, respectively, as obtained by fitting the peak-emission angles of these figures to the previously given expression for $\theta_{\pm m}$, with the SPP effective index k_{SPP}/k_0 used as the fitting parameter. As shown by these arrows, increasing the array period causes the first-order and

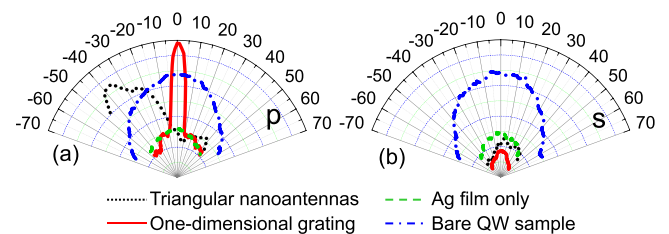


FIG. 3. Far-field radiation patterns with and without plasmonic beaming. (a) Experimental p -polarized radiation patterns measured with the plasmonic nanostructures of Fig. 1(a) (solid red line) and Fig. 1(c) (dotted black line), compared to those of the bare QW sample (dash-dotted blue line) and of the same sample coated only with a Ag film (dashed green line). (b) Same as (a) for s -polarized light.

second-order contributions to move to larger and smaller angles, respectively. For Λ close to 600 nm, the two peaks overlap, leading to particularly narrow beaming, which is also observed in the experimental data. The SPP effective index obtained from these fits (about 1.3) is relatively close to the refractive index of air, which confirms that plasmonic beaming in these structures involves SPP modes mostly guided at the Ag-air interface.

The beaming angles in these structures are ultimately determined by the interplay between the diffraction response of the array and the scattering cross section of the individual nanoparticles, which also features a strong angular dependence due to the particles asymmetric shape. Incidentally, the latter contribution (which is clearly not included in the expression above for $\theta_{\pm m}$) explains the small discrepancy between some of the arrows in Figs. 2(f)–2(h) and the corresponding angles of peak emission. In Figs. 2(b)–2(d), an experimental tuning range of about -38° to -25° is demonstrated based on the specific triangular geometry considered so far, by only varying the array period Λ . This range can also be extended by changing the shape of the triangles. To illustrate, in Fig. 2(e) we show data measured with nanoantennas having the same geometrical parameters as those of Figs. 2(b)–2(d), except for a larger value of α (80° instead of 68°). Asymmetric emission peaked at a substantially smaller angle (-15°) is obtained from this sample, consistent with the numerical simulation results plotted in Fig. 2(i).

Finally, we address the question of how the sample luminescence intensity is affected by the plasmonic nanostructures developed in this work. In Figs. 3(a) and 3(b) we plot, respectively, the p - and s -polarized far-field radiation patterns of representative samples based on the geometries of Fig. 1(a) (solid line) and Fig. 1(c) (dotted), together with those of the same QW material coated only with a 40-nm-thick Ag film (dashed) and without any coating (dashed-dotted). All traces were measured from the same region of the QW wafer with the same procedure, and the vertical scale is the same in the two plots. In the absence of metallic nanostructures, a broad Lambertian profile is observed as expected. The Ag film by itself causes a substantial reduction in output intensity for both polarizations, since it provides no mechanism to scatter the excited SPPs into radiation (other than its unintentional surface roughness). In contrast, with the diffractive arrays of Fig. 1, the p -polarized light intensity in the direction of peak emission is even increased compared to the bare-sample case. Vice versa, the corresponding s -polarized signals are substantially weaker, as they originate from the in-plane components of the QW excitonic dipoles, whose coupling to the Ag-film SPPs is much weaker than that of the out-of-plane component.¹ In any case, in the direction of peak emission, the net output intensity (i.e., p plus s) produced by the beaming nanostructures developed in this work remains comparable to that of the bare sample. Therefore, the use of light extraction across a continuous metal film in these structures does not compromise their suitability to practical beaming applications.

This behavior is related to the interplay among several competing effects. In the absence of plasmonic nanostructures, the external quantum efficiency of the QW sample is limited by nonradiative recombination and light trapping

caused by total internal reflection. In the geometries of Fig. 1, excitation of the Ag-film SPPs provides a fast relaxation channel for the QW electron-hole pairs (particularly at the blue-green wavelengths used in this work²⁴) so that undesirable nonradiative recombination processes are largely suppressed. Furthermore, extraction of these SPPs via diffractive scattering is no longer limited by total internal reflection. Instead, the key limiting factor is provided by ohmic absorption losses in the metal, which can be significant for the modes that are predominantly guided at the Ag-GaN interface due to their limited overlap with the grating. The experimental results of Fig. 3 indicate that these competing effects are comparable in magnitude. As a result, directional effects of light emission across the metal film can be achieved without any significant loss in output power. In fact, the energy coupled into the SPPs at the Ag-GaN interface could also in principle be efficiently extracted, with the addition of a high-index dielectric coating on top of the film-array structure to promote phase matching and therefore hybridization of the SPPs at the two metal surfaces.³⁷ With this arrangement, plasmonic beaming accompanied by light-emission efficiency enhancement could therefore be envisioned.

In conclusion, we have demonstrated plasmonic unidirectional beaming of luminescence at geometrically tunable angles from Lambertian light emitting samples. This functionality may open the way for new applications in several areas of high technological significance. For example, the nanostructures of Fig. 1(c) could be used for the development of highly integrated multiple-pixel LEDs where each pixel emits light along a different direction and can be driven independently, with a level of integration that would not be possible with the use of external bulk optical elements for beam control. In turn, these devices would be attractive for applications in solid-state smart lighting, where LEDs are used to provide additional functionalities besides illumination,³⁸ such as wireless communications and light-field mapping for real-time monitoring of the illuminated space. Similarly, the same nanostructures could be used for highly multiplexed fluorescence sensing, where the directionality of the emitted light is used to discriminate between different sample regions. Similar geometries could also potentially be applied to all-dielectric systems (photonic crystals) although metallic nanostructures in general feature particularly large scattering cross sections and can simultaneously provide additional functionalities such as current injection.

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