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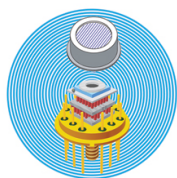
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Enhanced photoluminescence and directional white-light generation by plasmonic array

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White light-emitting diodes (LEDs), light sources that combine blue LEDs and yellow phosphors, are equipped with bulky optics such as lenses, mirrors, and/or reflectors to shape the light into the required directions. The presence of bulky optics causes optical loss and limits the design. Here, a periodic array of metallic nanocylinders, which exhibits a high scattering efficiency owing to the excitation of localized surface plasmon resonance, is proposed as an alternative means of achieving a directional output without the limitations of bulky optics. A prototype of a directional light emitter is fabricated consisting of an Al nanocylinder array on a yellow phosphor plate and a blue laser. The array shapes the yellow luminescence into the forward direction and generates directional quasi-white light (correlated color temperature of 4900 K). The intensity enhancement reaches a factor of five in the forward direction and is further improved up to a factor of seven by the deposition of a multilayer dichroic mirror on the back side of the phosphor plate, resulting in conversion efficiencies as high as 90 lm/W. Our results pave the way toward the development of efficient and compact directional white-light-source devices without any bulky optics. © 2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). <https://doi.org/10.1063/1.5050993>

I. INTRODUCTION

Various light sources have been invented to satisfy the demands of the telecommunication, mechanics, and electronics industries, among others. The existing light sources can be classified into four categories in terms of directionality and coherence, as shown in Fig. 1.¹ Incandescent bulbs and fluorescence tubes are light sources with an incoherent and omnidirectional output because incoherent light sources emit light in all directions. Light-emitting diodes (LEDs), organic LEDs, and white LEDs are emerging incoherent light sources included in this category.² Lasers produce the coherent output via stimulated emission, with cavities that predefine the output direction based on their geometry. Fabry–Perot, distributed feedback, and photonic crystal lasers fall into the category of light sources with a coherent and directional output.^{3–5} Random and microsphere lasers are classified as coherent but omnidirectional as random lasers do not possess cavities,^{6,7} and the cavities of microsphere lasers are spatially isotropic.⁸ For directional and incoherent light sources, which are of particular interest as illumination devices, combinations of incoherent and omnidirectional light sources with conventional optics are used. For LEDs, although the p-n heterostructures give some directionality to the emission, the bulky optics are still required to shape and give directionality to the emission as desired. The output radiating in unwanted directions is redirected using mirrors

and reflectors or absorbed by absorbers. These processes inevitably involve optical losses, decrease the output intensity, and limit the design of illumination devices.

The need for directional illumination devices is clearer in the context of next-generation high-power illumination, in which laser diodes (LDs) are used instead of LEDs as blue light sources to generate quasi-white light. Because LDs are directional light sources, conventional yellow phosphors, which provide omnidirectional radiation, cause a mismatch between the spatial distributions of blue and yellow light. Several attempts have been made to develop new incoherent and directional light sources by fabricating one- and two-dimensional periodic gratings or random nanostructures^{9–19} and microstructures^{20,21} composed of dielectrics on light-emitting layers. Although the output intensity is enhanced because of surface scattering, the directionality of the output remains insufficient for illumination applications because of the insufficient scattering efficiency.

Our intention is to produce new incoherent and directional light sources which are sufficient for illumination applications by applying plasmonic nanostructures to conventional phosphors. For this purpose, we place a periodic array of Al nanocylinders on a yellow phosphor plate. Periodic arrays of metallic nanocylinders, i.e., plasmonic arrays, have proven useful as directors and enhancers of photoluminescence (PL) for phosphors.^{22–26} For the overview of a plasmonic array, see Refs. 27 and 28. Up to a 60-fold intensity enhancement was reportedly achieved using a system consisting of a plasmonic array embedded in a thin phosphor layer (thickness = 650 nm).²⁵ Although a preceding work demonstrated a notable

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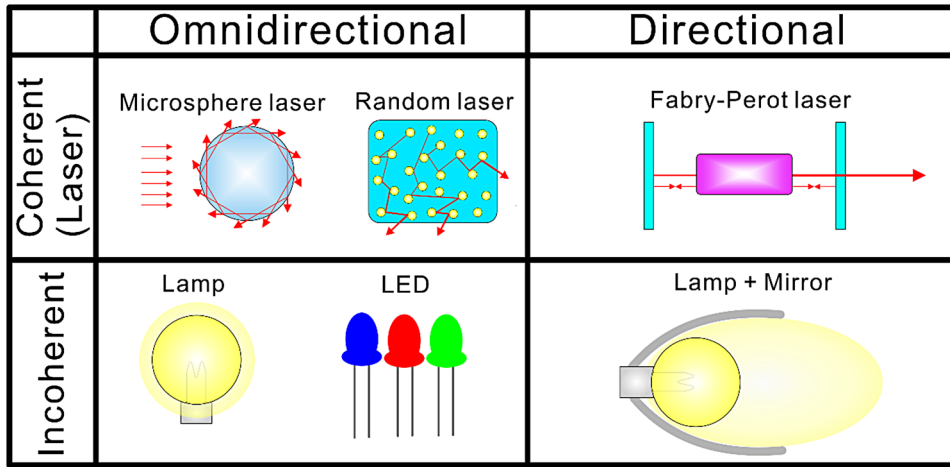


FIG. 1. Four categories of light sources classified according to coherence and directionality.

PL enhancement for thin layers up to several micrometres thick,²⁹ the absolute amount of light is insufficient for illumination applications. In this study, we fabricated a prototype directional and compact quasi-white-light source by placing an Al nanocylinder array on a thick yellow phosphor plate (thickness = 200 μm) and exciting it with a blue LD. As the phosphor, we selected $\text{Y}_3\text{Al}_5\text{O}_{12}$ garnet doped with Ce^{3+} ions, YAG:Ce, which is a yellow phosphor used for commercial white LEDs.³⁰ We demonstrated that the periodic Al nanocylinder array could enhance the PL from the 200- μm -thick phosphor plate. This result is surprising given that the surface plasmon is a local phenomenon bound to the surface. As will be explained later, we do not use the local accumulation of light energy but utilize a large scattering cross section of Al nanocylinders associated with the localized surface plasmon resonances (LSPRs). Therefore, the plasmonic array works even for thick phosphor plates as an efficient filter that extracts the PL to the desired directions. We also verified numerically that the Al nanocylinder array works better compared to the dielectric (Al_2O_3 and TiO_2) nanocylinder arrays with the identical design. By using a blue LD as a directional excitation source, we realized a uniform mixture of directional PL and blue LD radiation, thus achieving a directional output of white light with a correlated color temperature of about 4900 K and a deviation from the blackbody curve (D_{uv}) of approximately 0.025. Thus, a new means of achieving incoherent and directional light sources was developed in this study.

II. MECHANISM OF PL ENHANCEMENT BY PLASMONIC ARRAY

The enhancement of PL by an array, $I (=I_{\text{array}}/I_0)$, involves three components, i.e., absorption, $A (=A_{\text{array}}/A_0)$, quantum yield, $\eta (= \eta_{\text{array}}/\eta_0)$, and outcoupling, $C (=C_{\text{array}}/C_0)$, where the subscripts *array* and *0* refer to the values obtained from the array and the reference values of the flat phosphor layer, respectively;³¹

$$I = A \cdot \eta \cdot C. \quad (1)$$

PL control by a plasmonic nanostructure has been a topic of extensive research in plasmonics,³² and the general conclusion

is that it can increase the PL efficiency dramatically for emitters with low η_0 through the absorption enhancement of excitation light and/or the increase in the local density of state.^{33–37} For emitters with high η_0 , in contrast, the net enhancement is small or even negative because energy dissipation to the metal decreases the quantum yield and could outweigh the gain in absorption.^{38,39} A nanocylinder array is an exceptional structure which could enhance the PL from emitters with high η_0 . The main contribution to the PL enhancement by the array is C , i.e., the PL light otherwise trapped inside the phosphor can be coupled out of the phosphor by the array. The array can strongly diffract the light into a direction predefined by the pitch of the array without increasing the nonradiative decay paths, and thus the intensified directional PL output can be attained.^{24–26,40} The scattering cross section hits a local maximum accompanied by the excitation of LSPRs, and the plasmonic array thus becomes an efficient light diffractor around the spectral region of the LSPRs. In this study, we designed the pitch of the array to be 400 nm in a triangle lattice so that the PL around 600 nm would be enhanced by C in the vertical direction while A and η remained almost unchanged by the array.

III. EXPERIMENTAL SECTION

A. Plasmonic array fabrication

An Al thin film (thickness = 150 nm) was deposited on a YAG:Ce flat plate (thickness = 200 μm) using electron beam deposition. A resist (TU2-170, thickness = 200 nm) was coated onto the Al thin film and prebaked for 5 min at 95 $^\circ\text{C}$. As a master mold for nanoimprint lithography, an Si mold consisting of a triangle array of nanocylinders (diameter d = 150–240 nm; height = 150 nm; periodicity = 400 nm) was fabricated by electron lithography (F7000s-KYT01, Advantest) and Si deep etching (RIE-800iPB-KU, Samco). Then, the surface structure of the Si mold was transferred to the resist on the Al thin film by nanoimprint lithography, and the transferred resist nanocylinder arrays were structured to expose the Al thin film by ashing. After removing the natural oxide layer from the exposed Al thin film, the plasmonic array was patterned by reactive ion etching (RIE) (RIE-101iPH, Samco) under a chamber pressure of 0.1 Pa, a radio frequency power

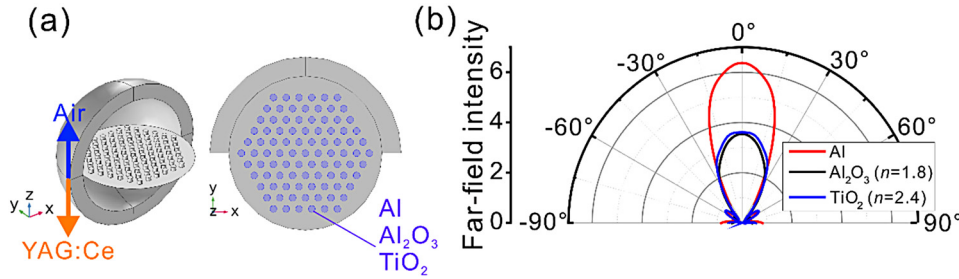


FIG. 2. (a) Far-field intensity calculation model. (b) Far-field intensity at $\lambda = 550$ nm from the substrate to the air with the presence of arrays of nanocylinders composed of various materials: Al ($n = 0.4 + 5.3i$, red line), Al_2O_3 ($n = 1.8$, black line), and TiO_2 ($n = 2.4$, blue line).

of 150 W, and both N_2 and Cl_2 gas flow rates of $5\text{ cm}^3/\text{min}$. After being processed by RIE, the residual resist was removed by O_2 plasma etching (RIE-10NR, Samco).

B. Characterization

The optical extinction ($1 - \text{transmittance}$) was measured as functions of angle of incidence θ_{in} and wavelength λ with an Si-based detector. The sample was placed on a rotation stage to vary θ_{in} on the sample surface. The zeroth-order extinction was obtained by normalizing the extinction of the sample to that of the YAG:Ce plate.

The PL spectra were measured as follows: a diode-pumped solid-state laser ($\lambda = 445$ nm, Shanghai Dream Lasers Technology, incident laser power = 13 mW) was incident from the back side of the sample at an angle of 5° from the normal and an optical fiber coupled to the spectrometer was rotated around the front side of the sample to obtain the PL spectrum as a function of emission angle θ_{em} .

In addition, the PL intensity was collected using a calibrated integrating sphere [diameter = 10 in. (0.254 m)] (FM-9025, Otsuka Electronics). An LD ($\lambda = 440$ nm) was incident on the sample placed on the input port of the sphere from the substrate side, and the output light was integrated throughout the sphere and collected by a fiber-coupled spectrometer. The output power was monitored as a function of the electricity input into the blue LD to calculate the conversion efficiency. The signal between $\lambda = 360$ and 830 nm was used to calculate the conversion efficiency. The color coordinates were calculated using CIE 1931 XYZ chromaticity coordinates.

C. Simulation

The forward-direction scattering by the triangle nanocylinder array was simulated using the finite element method (COMSOL). As shown in Fig. 2(a), the outer frame of the simulation model consists of a sphere (diameter = $5.4\text{ }\mu\text{m}$) surrounded by a perfectly matched layer (PML) (thickness = $0.6\text{ }\mu\text{m}$). The sphere model was divided into two parts: an upper part composed of air ($n = 1.00$) and a lower part consisting of YAG:Ce (refractive index estimated from ellipsometry measurements = 1.82). The 91 unit cells of the 400 nm-pitch triangle array of nanocylinders (height = 150 nm, diameter $d = 150$ nm, refractive index = n_{cyl}) in the xy -plane were located on the YAG:Ce. The lumped port (square: 670×670 nm) was placed in the YAG:Ce, 900 nm below its surface. The incident light ($\lambda = 550$ nm) from the port was allowed to propagate from the YAG:Ce to the air side at

$\theta_{in} = 0^\circ$. To compare the forward scattering efficiencies of the Al and dielectric (Al_2O_3 and TiO_2) nanocylinders, the refractive index of the nanocylinders was varied as follows: $n = 1.8$ (representing Al_2O_3), $n = 2.4$ (TiO_2), and the n and k values of Al ($0.4 + 5.3i$) obtained from ellipsometry measurements. Al_2O_3 and TiO_2 were chosen as representative dielectric materials that have relatively high n . The far-field intensity was integrated along the boundary edge between the PML and air in the xz -plane.

IV. COMPARISON TO DIELECTRICS BY SIMULATION

Metallic nanostructures exhibit high scattering and absorption efficiencies associated with the excitation of LSPRs, whereas dielectric nanostructures exhibit lower scattering and virtually no absorption in the visible region.^{41,42} We simulated the forward scattering strengths of periodic arrays of nanocylinders composed of metals or dielectrics. In the model [Fig. 2(a)], the light source was embedded in the substrate, and the plane wave ($\lambda = 550$ nm corresponding to the central emission wavelength of YAG:Ce) was projected to the array from the substrate side. The far-field intensity scattering onto the air side was monitored as a function of the emission angle θ_{em} . Figure 2(b) displays the scattering strengths of the periodic arrays of Al_2O_3 , TiO_2 , and Al nanocylinders. The periodic array of Al nanocylinders exhibits a scattering intensity higher than those of the dielectrics. The results numerically verify that metallic nanocylinder arrays extract light more effectively, i.e., C in Eq. (1) is larger than their dielectric counterparts in the present geometry and the wavelength of $\lambda = 550$ nm. Our previous report²⁶ shows that the increase in the refractive index of the substrate decreases C . For the low-refractive-index substrate such as SiO_2 , the dielectric cylinders such as titania can show larger C than Al, while the C of dielectric cylinders is lower for the high-refractive-index substrate such as YAG:Ce in the present work and sapphire. A powerful competitor to Al would be Si, which possesses a very high refractive index in the visible. Experimental PL enhancement has been demonstrated by the Si nanocylinder array in the red spectral region,⁴³ while the enhancement in the blue to yellow regions where Si has fundamental absorption has not yet been verified.

We note that metallic structures also exhibit high backward scattering and absorption. The backscattered PL can be reflected back into the forward direction by placing a multi-layer dichroic mirror on the opposite side of the substrate, as was experimentally demonstrated in this work.

V. RESULTS AND DISCUSSION

A. Optical extinction

The Al nanocylinder arrays were fabricated on the substrate using nanoimprint lithography and RIE, as shown in the insets of Fig. 3. Figures 3(a) and 3(b) present optical extinction ($1 - \text{transmittance}$) spectra as a function of the angle of incidence θ_{in} for the nanocylinder arrays with $d = 150$ and 210 nm, respectively. The peaks in the color maps are due to the excitation of LSPRs on each nanocylinder, and they red-shift with d . The dotted lines represent the conditions in which the incident light is scattered in the plane of the array, i.e., the Rayleigh anomaly.²⁴ From the conservation of the parallel component of the wave vector at the surface of

the array, the Rayleigh anomaly satisfies the relation $\mathbf{k}_{\text{out}\parallel} = \mathbf{k}_{\text{in}\parallel} \pm \mathbf{G}$, where $k_{\text{out}\parallel} (=2\pi n/\lambda)$ and $k_{\text{in}\parallel} [=2\pi/\lambda \times \sin(\theta_{\text{in}})]$ are the parallel components of the diffracted and incident wave vectors, respectively, with n being the refractive index surrounding the array. We shall refer to the magnitude of $\mathbf{k}$ as k . $\mathbf{G} = (m_1 \mathbf{b}_1, m_2 \mathbf{b}_2)$ is a reciprocal lattice vector, where

$$\begin{aligned} b_1 &= (2\pi/a)(\hat{x} + \hat{y}/\sqrt{3}), \\ b_2 &= (2\pi/a)(\hat{x} - \hat{y}/\sqrt{3}), \end{aligned} \quad (2)$$

and m_1 and m_2 are the pair of integers defining the diffraction order.²⁶ When $\mathbf{k}_{\text{in}\parallel}$ does not have a component in the y -direction, $k_{\text{out}\parallel}$ can be expressed as

$$\begin{aligned} k_{\text{out}\parallel}^2 &= k_{\text{in}\parallel}^2 + 2(2\pi/a)(m_1 + m_2)k_{\text{in}\parallel} + (2\pi/a)^2(m_1 + m_2)^2 \\ &\quad + (2\pi/a)^2(m_1 - m_2)^2/3, \end{aligned} \quad (3)$$

where m_1 and m_2 are equivalent and interchangeable. The $(m_1, m_2) = (1, 0), (-1, 0), (1, -1)$ diffraction orders are plotted in Figs. 3(a) and 3(b). The black and red dotted lines have been calculated using $n = 1.82$ and 1.00 , which are the refractive indices of the YAG:Ce substrate and air, respectively.²² In the absence of diffraction, simulation indicates that the Al nanocylinders with $d = 150$ and 210 nm show a broad LSPR centered at $\lambda = 450$ and 480 nm, respectively. It is clear that the LSPR band is modulated along the diffraction lines, which indicates the coupling of individual LSPRs through in-plane diffraction. The extinction increases with d because of the increased scattering cross section.³⁹ The angle-independent blue region at $\lambda < 475$ nm comes from the absorption of the YAG:Ce substrate, ascribable to the electron transitions from $4f$ ground state to $5d$ excited states of Ce^{3+} .^{30,44} In this region, the extinction of the substrate is more than 90% [see Fig. 3(c) for the extinction of this region at $\theta_{\text{in}} = 0^\circ$] and the spectral feature is difficult to see.

B. Directional PL

Figures 4(a) and 4(b) present the PL spectra as a function of θ_{em} for the reference YAG:Ce plate without the array and the YAG:Ce plate with the Al nanocylinder array ($d = 210$ nm), respectively. For the reference, $I_0(\lambda, \theta_{\text{em}})$ [Fig. 4(a)], the PL spectrum centered at $\lambda = 530$ nm, which is typical of YAG:Ce phosphors, is observable, and the spectral shape does not change with θ_{em} . In contrast, the PL from the YAG:Ce plate with the array, $I_{\text{array}}(\lambda, \theta_{\text{em}})$, changes drastically with θ_{em} in both intensity and spectral shape [Fig. 4(b)]. Note that the intensity scales in Figs. 4(a) and 4(b) are identical, and thus the difference in PL intensity is recognizable visually. The comparison at $\theta_{\text{em}} = 0^\circ$ in Fig. 4(c) clearly demonstrates the PL enhancement by the array as high as a factor of five. A kink is evident at $\lambda \sim 630$ nm, which corresponds to the in-plane diffraction condition. Also shown is the photopic luminosity function.⁴⁵ It is beneficial for lighting applications that the PL enhancement covers the spectral region where the human eye is most sensitive.

The angular profile can be better understood by plotting the PL enhancement, $I(\lambda, \theta_{\text{em}})$, defined by the PL intensity normalized to that of the reference. The angular profile of the

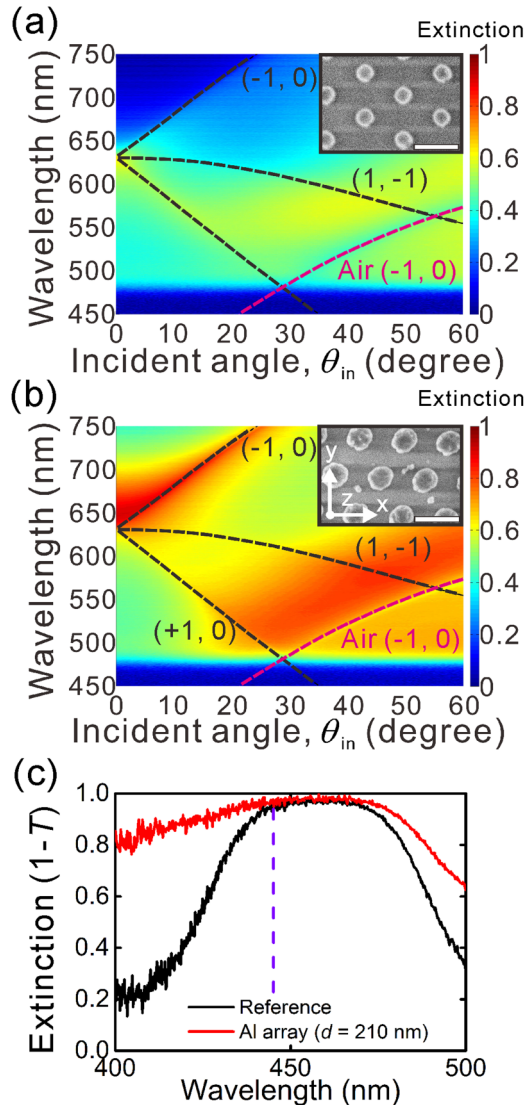


FIG. 3. Extinction, defined as $(1 - \text{transmittance})$, spectra as a function of θ_{in} for the arrays with (a) $d = 150$ and (b) $d = 210$ nm. The black and red dotted lines are the diffraction lines reflecting the refractive indices of YAG:Ce ($n = 1.82$) and air ($n = 1.00$), respectively. The insets are top-view scanning electron microscope images of the arrays with identical designs. The white bars in the insets represent 400 nm. The x , y , and z -axes used in this study are shown in the inset in (b). The incident angle was varied to give momentum in the x direction. (c) Extinction spectra at $\theta_{\text{in}} = 0^\circ$, where the transmittance is normalized to the intensity of incident light, for the YAG:Ce plate without and with the arrays with $d = 210$ nm. The vertical line is drawn at $\lambda_{\text{ex}} = 445$ nm.

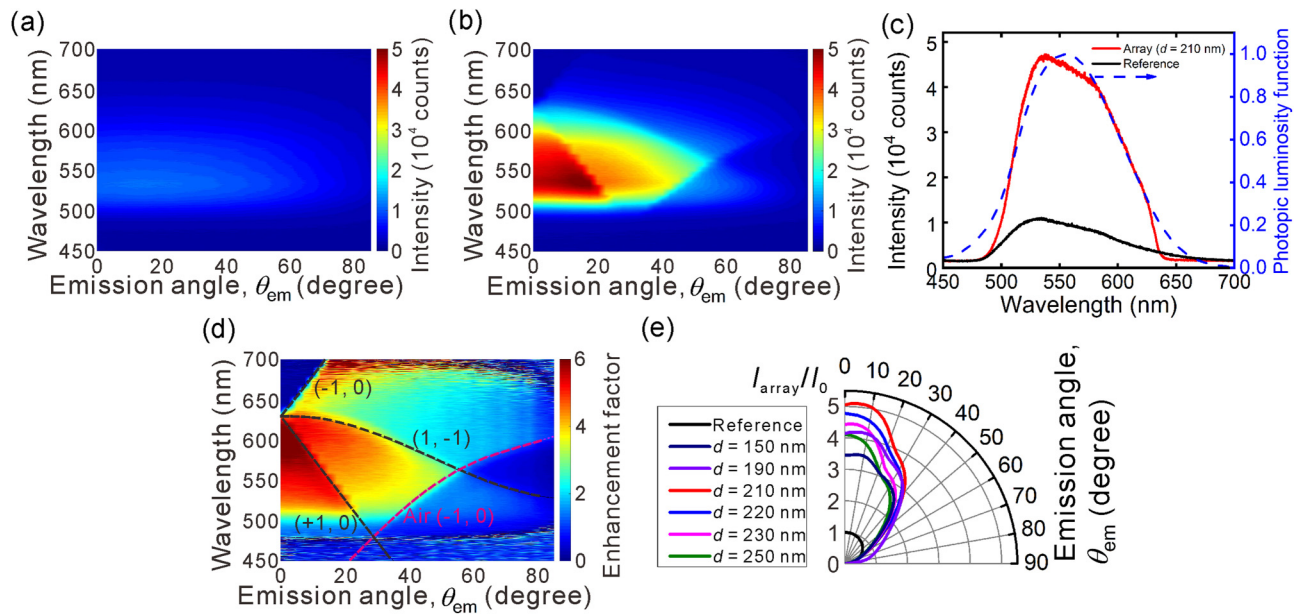


FIG. 4. PL spectra of YAG:Ce plates (a) without and (b) with an Al array ($d=210$ nm) as a function of θ_{em} . (c) PL spectra of the YAG:Ce plates at $\theta_{em}=0^\circ$ without and with an Al array plotted using black and red lines, respectively. Also shown is the photopic luminosity function. (d) PL enhancement spectrum obtained from the PL intensity of the YAG:Ce plate with an Al array divided by that of the YAG:Ce plate without an array as a function of θ_{em} . The black and red dotted lines are diffraction lines reflecting the refractive indices of YAG:Ce ($n=1.82$) and air ($n=1.00$), respectively. (e) Angular profiles of integrated PL enhancement over $\lambda=500$ – 700 nm normalized by the PL intensity of the reference at $\theta_{em}=0^\circ$ for the Al arrays with $d=150, 190, 210, 220, 230$, and 250 nm.

PL enhancement in Fig. 4(d) clearly depends on both λ and θ_{em} and presents domains with different enhancement values. Plotting the diffraction lines in the same way as in Figs. 3(a) and 3(b), the diffraction lines nicely overlap the boundaries between the domains. This coincidence indicates that the enhancement is related to the outcoupling of PL with diffraction, i.e., C in Eq. (1). At $\theta_{em}=0^\circ$, these domain boundaries can be characterized as follows:

$$\lambda < \sqrt{3}a/2, \quad (4-i)$$

$$\sqrt{3}a/2 < \lambda < n \cdot \sqrt{3}a/2, \quad (4-ii)$$

and

$$n \cdot \sqrt{3}a/2 < \lambda. \quad (4-iii)$$

We consider the situation in which the PL with λ_{em} travels inside the YAG:Ce plate and hits the surface. When λ_{em} is sufficiently short to satisfy Eq. (4-i), i.e., $\lambda_{em} < 346$ nm, the PL is scattered back into the plate, and thus no PL enhancement occurs. In contrast, when Eq. (4-ii) is satisfied (346 nm $< \lambda_{em} < 630$ nm), the PL is scattered out into the air so that directional and intensified outcoupling is realized. When λ_{em} is at a length that satisfies Eq. (4-iii) (630 nm $< \lambda_{em}$), no light diffraction occurs. Figure 4(d) also shows that the enhancement factor is largest at $\theta_{em}=0^\circ$ around $\lambda=630$ nm, because at this condition, three diffraction orders degenerate and contribute to the outcoupling.

Figure 4(e) presents the PL intensity integrated over the PL spectra (from $\lambda=500$ to 700 nm) and normalized by the integrated PL intensity of the reference at $\theta_{em}=0^\circ$, as a function of θ_{em} . The reference exhibits a Lambertian angular profile, i.e., the PL intensity varies with $\cos(\theta_{em})$, which is typical for phosphors exhibiting ideal flat surfaces. In sharp

contrast, the angular profiles for the array samples deviate from the Lambertian profiles and are elongated toward the forward direction, indicating a directional output. The magnitude of the enhancement varies with the d values of the cylinders. As long as the pattern and pitch of the array remain the same, d does not change the diffraction conditions. However, it does change the efficiency of diffraction. When d increases from 150 to 210 nm, the integrated PL intensity increases monotonically. This increase corresponds to the increase in the scattering cross section. The intensity reaches a maximum at $d=210$ nm, and the further increase in d decreases the intensity, because increasing the geometrical diameter leads to a large coverage of the surface by the Al cylinders, thus hindering the outcoupling of PL from the substrate to the air.

Figure 5 depicts the effect of the cylinder height. The height of the cylinder is varied from 50 to 200 nm for a fixed

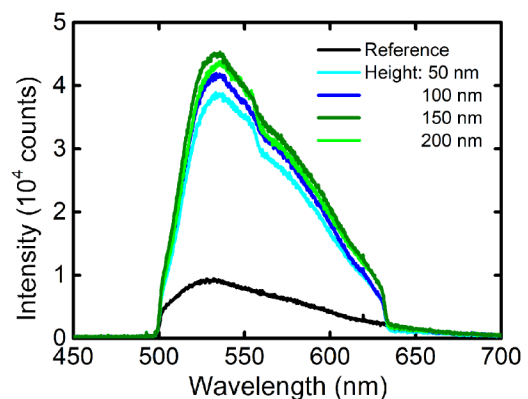


FIG. 5. PL spectra of the YAG:Ce plate without (black line) and with the Al arrays ($d=140$ nm) with the height of $50, 100, 150$, and 200 nm.

$d = 140$ nm, and the maximum enhancement is obtained when the height is 150 nm. This dependence of PL enhancement on the height is in agreement with the dependence of the intensity of LSPR on the aspect ratio of the cylinder in the array structures reported.⁴⁶

The lattice pattern and pitch dominate the outcoupling conditions via the diffraction conditions. Figure 6 compares the PL spectra at $\theta_{em} = 0^\circ$ for the triangle (pitch: 350 nm and 400 nm) and square (pitch: 350 nm and 400 nm) arrays. The spectral shape of the PL depends on the diffraction condition.²² The in-plane diffraction moves to shorter wavelengths when the pitch is reduced, and each spectrum is enhanced at the shorter wavelength region than the in-plane diffraction condition. A bump appeared around $\lambda = 550$ nm for the spectrum of the square array with pitch = 350 nm corresponds to the diffraction condition for triangle lattice with pitch = 350 nm: because all the arrays are on the same plate, the diffraction effect from the neighboring arrays also appears. The difference in lattice pattern affects not only the spectral shape but also the spatial distribution of the PL, as shown in Figs. 6(c) and 6(d). The triangle and square arrays show hexagonal and square radiation patterns, respectively.

We briefly describe the contributions of the other two factors (A and η) in Eq. (1) to the PL enhancement. Regarding A and η , the presence of the array increases the extinction of the excitation light ($\lambda = 445$ nm) from 94.5% to 96.7% [see Fig. 3(c)] and decreases the PL lifetime from 65.0 to 63.7 ns (see Fig. 7). The increase in extinction could be attributable to the increase in the absorption by the YAG:Ce plate, which leads to the PL enhancement up to 2% only. A very slight decrease in PL lifetime is intuitively understandable given the thickness of the YAG:Ce plate (200 μm) which is far thicker than the penetration depth of LSPRs.

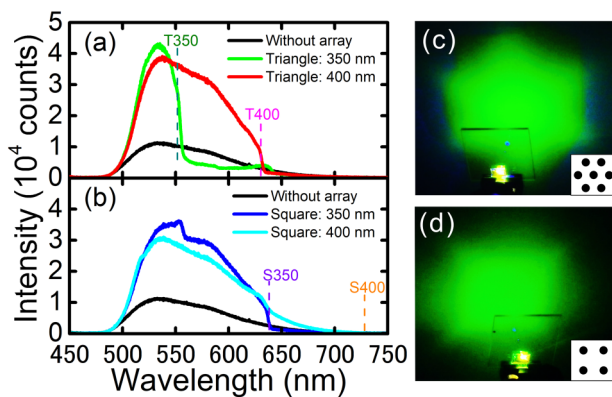


FIG. 6. PL spectra of the YAG:Ce plate with the Al nanocylinders ($d = 150$ nm, height = 150 nm) arranged in (a) triangle (pitch: 350 nm and 400 nm) and (b) square (pitch: 350 nm and 400 nm) lattices. Also shown in the figures are the PL spectra of the YAG:Ce plate without the array. The spectra were collected at normal direction ($\theta_{em} = 0^\circ$). The in-plane diffraction conditions for the triangle arrays with the pitches of 350 nm and 400 nm are denoted as T350 and T400, and those for square lattices with the pitches of 350 nm and 400 nm are denoted as S350 and S400, respectively. Spatial profiles of the PL from the YAG:Ce plate with triangle (c) and square (d) arrays with 400 nm pitches. The samples were excited by a blue LD from the substrate side and the PL in the forward direction was projected on the screen while the blue light was cut by a filter placed between the sample and the screen.

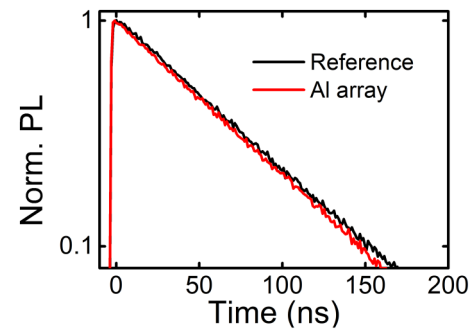


FIG. 7. Normalized PL decay curves of the YAG:Ce plate without (black line) and with (red) an Al nanocylinder array. The samples were excited upon $\lambda_{ex} = 470$ nm and the PL intensity at $\lambda_{em} = 525$ nm was collected.

Although the decrease in PL lifetime could indicate an increase in the radiative decay rate, this value alone cannot explain the fivefold enhancement demonstrated in Fig. 4(c). These results indicate the minor contributions of A and η to the enhancement observed here.

C. White light generation

Figures 8(a) and 8(b) show the photographs of the reference and YAG:Ce plate with the Al nanocylinder array ($d = 210$ nm) irradiated with the blue LD, respectively, with the PL and transmitted blue light projected onto a screen. Both plates have an identical thickness of 200 μm . For the reference in Fig. 8(a), the blue light dominates the transmission, although Fig. 3(c) indicates that the transmission of the YAG:Ce plate is less than 10% at $\lambda = 445$ nm. In contrast, for the plate with the array in Fig. 8(b), the yellow PL is enhanced, generating a quasi-white color, and a sixfold symmetry is evident owing to the diffraction by the triangle lattice. The top panel of Fig. 8(c) presents the intensities of the yellow PL and blue LD radiation onto the front side of the plate collected by the integrating sphere for the reference and sample ($d = 210$ nm). The plate with the array exhibits an up to threefold enhancement over the PL region and a decrease in the amount of blue light owing to outcoupling and reflection, respectively. These characteristics confirm that the PL intensity is enhanced not only in a specific direction, but also overall on the front side. The bottom panel of Fig. 8(c) shows the output intensity from the YAG:Ce plate with the array normalized to that from the reference without the array. The output intensity of blue light is reduced to 41% of that for the reference plate. This decrease means that 59% of blue light is lost by the presence of the array. The decrease in blue and the increase in yellow to red result in the drastic shift in color.

Figure 8(d) depicts the color coordinates calculated based on the spectra obtained using the integrating sphere. For the reference, the color is in the blue region, while the plate with the array exhibits enhanced yellow PL, so the light color is yellow. The color temperature of the array sample is as low as 4900 K. The deviation from the blackbody curve is $D_{uv} \sim 0.025$, indicating that the light color is close to that of blackbody radiation, i.e., it is close to the natural white color. Comparing the chromaticity coordinates of the reference and

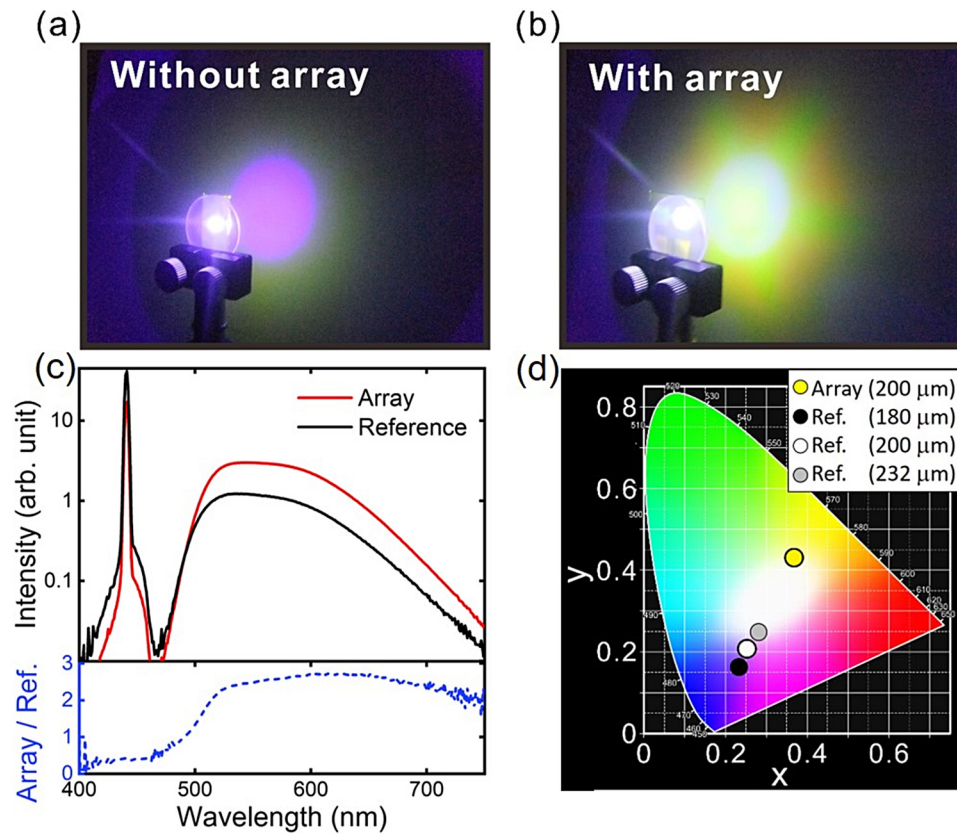


FIG. 8. Appearances of PL for YAG:Ce plates (a) without and (b) with an Al array with $d=210$ nm. The thicknesses of the samples in (a) and (b) were the same ($=200\mu\text{m}$). The spot size of the blue LD was increased by the optical diffuser placed in front of the YAG:Ce plate. (c) (Top panel) PL spectra collected by an integrating sphere for the reference and a sample with $d=210$ nm represented by black and red lines, respectively. (Bottom panel) The output ratio between the sample and the reference. (d) Chromaticity coordinates of the reference and a sample with $d=210$ nm calculated from the spectra shown in (c) using CIE 1931.

sample, the PL intensity is enhanced to an extent that the coordinates of the sample surpass the ideal white color at $(x, y) = (0.33, 0.33)$ and enter the yellow region. Therefore, ideal white light, e.g., a color temperature of 5500 K for lighting applications, can be obtained by tuning the yellow PL using the thickness of the YAG:Ce plate. We measured the output intensity of the reference plates with different thicknesses (180 and 232 μm). The results are plotted in Fig. 8(d). A simple linear extrapolation indicates that a 300 μm -thick plate would give white color ($=5500$ K) and a 350 μm -thick plate would give 4900 K color temperature that is achieved by the 200 μm -thick plate with the nanocylinder array.

We calculated the PL efficiency of the system from the dependence of the output intensity on the input power of the blue LD, as listed in Table I. The PL efficiencies of the reference and Al array ($d=210$ nm) are 31 and 72 lm/W,

respectively, implying that the Al array enhances the PL efficiency by up to a factor of 2.3. These values were calculated for the output emitted from the front side and integrated over all θ_{em} in the forward direction. Figure 4(c) indicates that the PL intensity is enhanced by a factor of five at $\theta_{\text{em}} = 0^\circ$. The increase in intensity by a factor of 2.3 indicates not only that the PL intensity increases at $\theta_{\text{em}} = 0^\circ$, but also that the total amount of light extracted from the front side of the sample and radiated in the forward direction increases.

To enhance the output intensity further, we combined the array with a dichroic multilayer mirror. The system is depicted in a schematic in Fig. 9, where the mirror is placed on the opposite side of the array, facing the blue LD. The mirror was designed to transmit blue light and reflect yellow light. Figure 9 presents the polar plots for the reference and samples with and without the mirror. The PL intensity is

TABLE I. Conversion efficiencies of the series of samples. The efficiency for the square array (pitch = 400 nm and $d=210$ nm) is also shown. Note that the samples denoted as “ $d=210$ nm” and “Without mirror ($d=210$ nm)” have the same geometry in design, but are the different samples prepared at different times.

Sample name	Reference	$d=210$ nm	$d=220$ nm	$d=230$ nm	$d=250$ nm	Without mirror ($d=210$ nm)	With mirror ($d=210$ nm)	Square (pitch = 400 nm and $d=210$ nm)
Conversion efficiency (lm/W)	31	72	70	61	56	68	90	67

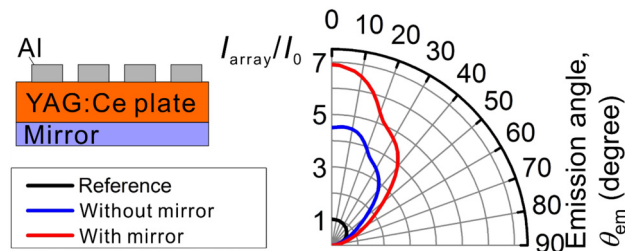


FIG. 9. Angular profiles of the PL intensity for the YAG:Ce plate (black line), YAG:Ce plate with an Al array ($d = 210$ nm) (blue), and YAG:Ce plate with an Al array ($d = 210$ nm) and a multilayer dichroic mirror (red). The PL intensities are integrated over the spectral range from $\lambda = 500$ to 700 nm and normalized by the intensity of the YAG:Ce plate at $\theta_{em} = 0^\circ$. The top-left illustration shows a schematic of the multilayer dichroic mirror.

seven times higher than that of the reference at $\theta_{em} = 0^\circ$. Furthermore, the intensity is 1.8 times higher than that of the array without the mirror, and the intensity over the integrating sphere is three times that of the reference. The PL efficiency increases from 72 to 90 lm/W (see Table I), which is comparable to the efficiencies of commercial fluorescent lamps.⁴⁷ It is noted that this value is for the prototype system, which was not optimized or packaged.

VI. CONCLUSION

In summary, we described a strategy of making directional white light sources using periodic arrays of nanocylinders on a phosphor plate. The periodicity and the pattern (square and triangle in the present case) of the array dominate the ranges of wavelengths and emission angles of PLs that are outcoupled from the plate. The array pattern also determines the spatial distribution of the PL. Simulation shows that the Al nanocylinder array possesses larger scattering cross section compared to dielectrics such as Al_2O_3 and TiO_2 in the present geometry and the wavelength, due to the excitation of LSPR at the spectral region of interest. The size (diameter and height) as well as the material of nanocylinders dominates the intensity of light scattering. Based on this strategy, we fabricated a prototype directional white light source by combining a plasmonic array on a YAG:Ce plate and a blue LD. For a triangle array (periodicity = 400 nm) of Al nanocylinders with the size being chosen to maximize the forward PL scattering, the preferential forward output was observed. The intensity was increased up to a factor of seven in the normal direction and a factor of three when integrated over the radiation in the forward direction, compared to the reference. The efficiency of the sample is 90 lm/W, which is comparable to the efficiencies of commercial fluorescent tubes, even though it has not yet been optimized. We expect that this plasmonic array-based technology will be especially important for next-generation directional light sources using LDs as blue sources and that it could change the design concept of lighting in various settings.

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