Nanophotonics for solid state lighting

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1. Introduction

The Nobel Prize of Physics of this year has been awarded jointly to Isamu Akasaki, Hiroshi Amano and Shuji Nakamura *for the invention of efficient blue light-emitting diodes*. Blue LEDs based on GaN have enabled the development of bright white light sources, which are several times more efficient than traditional incandescent lamps. This technological breakthrough, achieved in the 90's thanks to the perseverant research programs of Akasaki at the University of Nagoya (Japan) and of Akamura in the small company Nichia Corporation (Japan), has led to the Solid State Lighting (SSL) revolution that is now being driven by big multinationals such as Philips Lighting and Osram. In spite of the quick expansion and implementation of this technology in our daily life (in a few years all incandescent and fluorescent lamps will be replaced by high efficiency LEDs), there are still many challenges to be solved and opportunities for new developments in which photonics plays a central role.

2. White LEDs

White light can be generated with two different approaches.[i] The first one uses several LEDs emitting different colors which are combined to produce white light. This method is limited by the low efficiency of green and yellow LEDs based on InGaN. As the indium concentration is increased to shift the emission to longer wavelengths, the efficiency drops significantly. The origin of this drop is in the accumulation of structural defects that lead to the non-radiative recombination of the injected carriers and the generation of heat instead of light. The second approach, which is prevailing nowadays for the generation of white light, uses high efficiency UV or blue LEDs to convert part of their emission to green and red by means of photoluminescent materials, which are known as phosphors. These are the so-called phosphor-converted LEDs (pcLEDs). Figure 1 shows a schematic representation of a pcLED. We will focus the rest of this article on these devices and how nanostructures can be used to mold and optimize their emission.

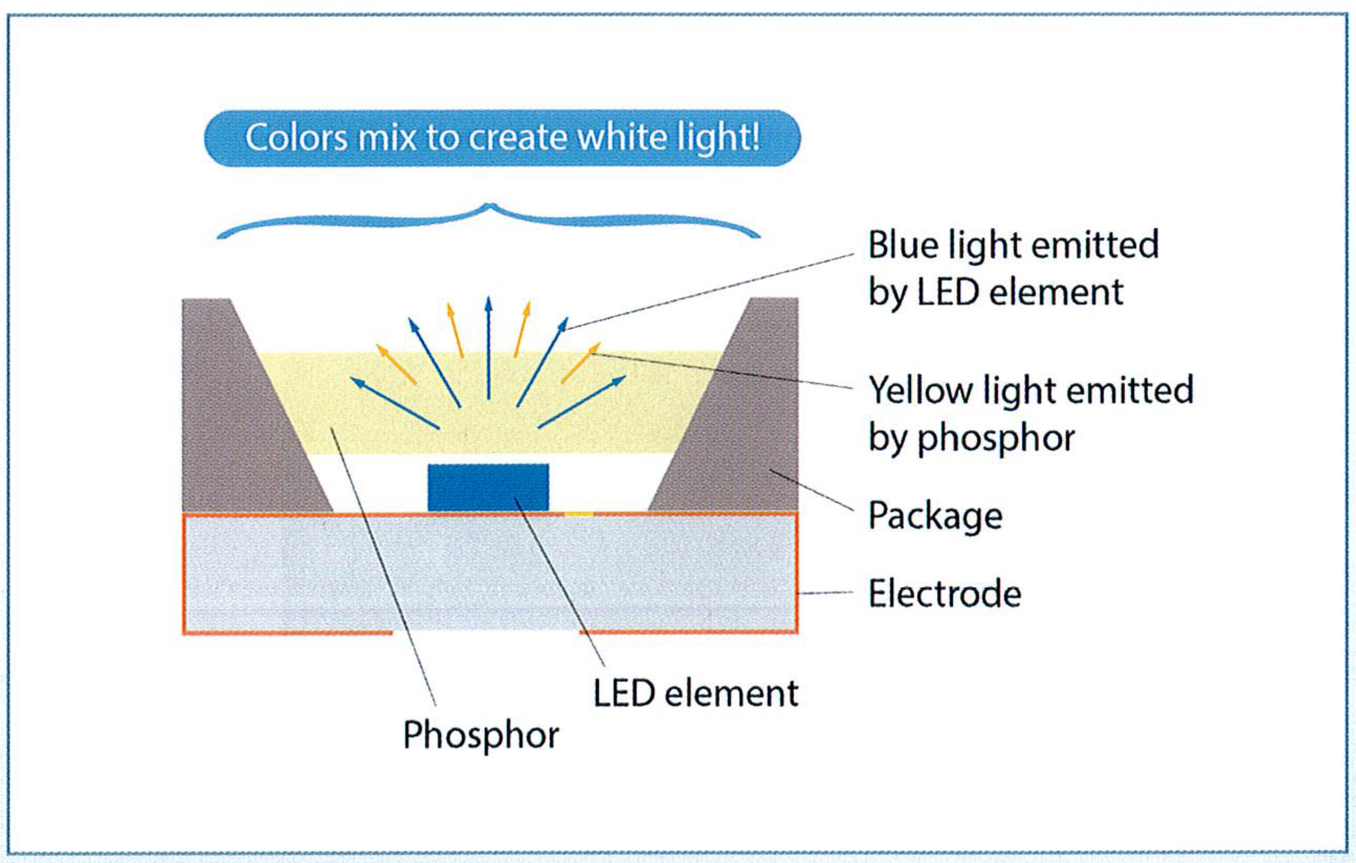
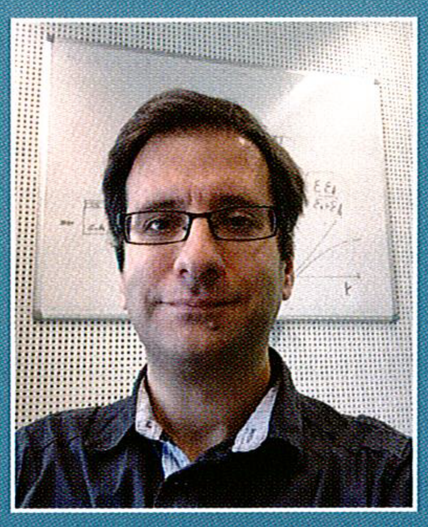


Figure 1. Schematic representation of a phosphor converted LED.



Jaime Gómez Rivas studied Astrophysics in Madrid (Spain) and Luik (Belgium) getting his Master degree in 1995. From 1995 until 1997 he worked at SRON (Utrecht) developing superconducting detectors. He did a PhD (1997-2002) at the UvA (Amsterdam) investigating the propagation of light in multiple scattering samples. From 2002 he worked as postdoctoral researcher at the University of Aachen (Germany) and in 2005 he was appointed as group leader at the FOM Institute AMOLF. One of his research topics is the emission of light from nanostructured surfaces. In 2009 Gómez Rivas was awarded with an ERC grant, initiating research on THz plasmonics with semiconductors. He was appointed in 2010 as part time professor at the TU/e.

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There are only a limited number of photoluminescent materials that are suitable for color (wavelength) conversion in SSL, being the development of novel emitters with properties suited to LED integration a subject of extensive research in materials science. Phosphors for color conversion must show: i) a close-to-one emission efficiency in order to maximize the overall electrical-to-optical conversion efficiency; ii) excellent chemical and temperature stability; iii) moderate thermal quenching at temperatures over 100°C; iv) an absorption spectrum that overlaps with the emission wavelength of the UV or blue LED, and large absorption cross section to efficiently absorb this light in the smallest possible volume, and v) an emission spectrum that leads to high-quality white light emission. A material intensively used in pcLEDs is cerium doped yttrium aluminum garnet (Ce:YAG). The broad emission spectrum of Ce:YAG , with a significant fraction of these emission in the infrared, limits the efficiency of Ce:YAG pcLEDs. Therefore, intensive research towards the development of narrow band emitter such as quantum dots or photo-stable dye molecules is taking place.

A phosphor layer can be described as a continuous distribution of randomly oriented dipoles over a layer of a given thickness near a planar interface. The surface is typically roughened to maximize the light extraction. As a result, light is emitted by the phosphor in all directions leading to a broad angular distribution of this emission in the far field. For many applications it is important to beam light in defined directions, which is currently achieved by means of inefficient optical components such as lenses and parabolic mirrors. The large size of these components sets a limit to the integration of LEDs. Moreover, approximately half of the converted light is emitted in the backward direction, i.e., towards the exciting blue LED. This emission pattern results in the need of incorporating additional optical elements (such as mirrors or scattering materials) to recycle as much as possible of this light. Nanostructures with designed properties may offer a solution to these problems.

3. Nanophotonics for Solid State Lighting

In the Center for Nanophotonics of the FOM Institute AMOLF we investigate nanostructured materials which modify the characteristics of light emitters. This modification is achieved by the strong light-matter interaction that is achieved with resonant structures with dimensions comparable to the wavelength of light. An example of these structures is shown in Figure 2. This sample is a periodic array of metallic nanoparticles, made of silver or aluminum, spaced by a distance comparable to the wavelength of light in the visible.[ii] On top of the array a layer of phosphor with a thickness of about 1 μm is deposited. The nanoparticles have dimensions and shapes such that they support resonances at optical frequencies. These resonances are called localized surface plasmon polaritons (LSPPs) and are the result of the oscillation of the electrons in the metal driven by the electric field of light. LSPPs are characterized by concentrating the incident light in small volumes close the particles. By reciprocity, a light emitter in the proximity of a particle can efficiently emit light by coupling this emission to LSPPs. This process of receiving or emitting electromagnetic waves is exactly what an antenna does at radio frequencies. Therefore, metallic nanoparticles are often referred as optical antennas or nanoantennas. [iii]

An important characteristic of antennas is the directivity of their capacity to radiate in defined directions. This directivity can be enhanced in antenna phased arrays, i.e., in arrangements of antennas which emit radiation with a

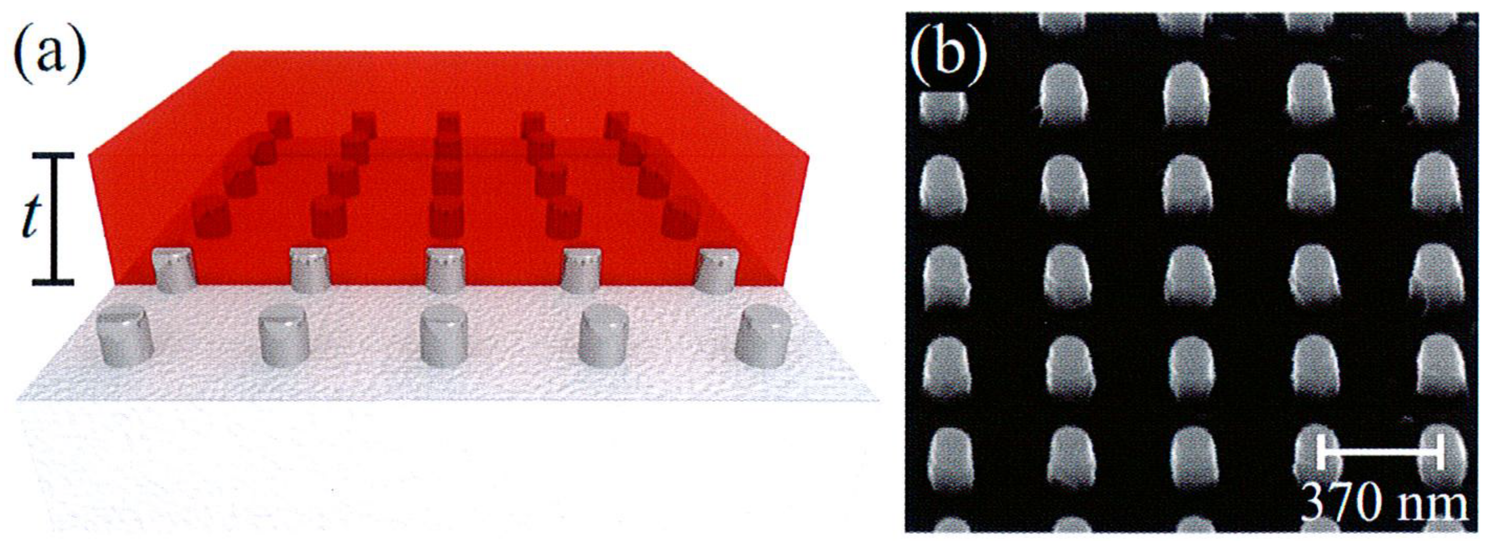

Figure 2. Schematic representation of a nanophotonic converting layer in a phosphor white LEDs. A phosphor layer (quantum dots or dye molecules) with a thickness t , typically around 1 μm (represented by the red layer), is on top of a periodic array of metallic nanoparticles on a glass substrate. The figure on the right corresponds to a scanning electron microscope image of a typical array made of aluminum particles.

defined phase relation such that the emitted waves interfere constructively in the far-field in certain directions. The analog to an antenna phased array for visible light is the periodic array of nanoantennas shown on figure 2. The phase relation between the different nanoantennas is set by the periodic spacing between the particles with a distance that is comparable to the wavelength of light.

Light conversion by the phosphor in the proximity of these particle arrays takes place as follows: blue photons from the exciting source (a blue LED or a laser) are absorbed by the layer of emitters. This absorption can be enhanced by the presence of the antenna array if the angle of incidence and the blue wavelength are resonant with the structure. In this case the array operates as a receiving antenna. A resonant absorption leads to a brighter emission from the phosphor but also to an increase of the losses in the metal and a reduction of the wall-plug efficacy of the white LED (defined as the ratio between the total luminous flux emitted by the device and the total amount of electrical input power). The replacement of metal antennas by dielectric scatterers or the design of intelligent illumination patterns that minimize the blue light intensity in the metal can fully suppress these losses. The excited phosphor, after absorption of the blue light, decays emitting into surface modes of the nanoantenna array, i.e., modes that propagate on the surface of the array. These modes scatter with the nanoantennas and the scattered radiation interferes constructively in directions defined by the periodicity of the array. This produces a large enhancement of the emission in certain directions without the need of using lenses or mirrors to guide the emission in these directions.

Figure 3(a) illustrates the measurements that characterize the modes of the antenna array. The upper panel shows an extinction spectrum (defined as 1-transmission) of light incident normal to the sample surface. The broad resonance at 1.85 eV corresponds to the LSPPs in the individual nanoantennas, while the sharp resonances at higher energies (shorter wavelengths) correspond to the surface modes in the periodic array. These modes are known as Surface Lattice Resonances (SLRs) and are the result of diffraction in the plane of the periodic array of nanoantennas. The lower panel in Figure 3(a) shows the photoluminescence emission of the phosphor layer (Lumogen F Red 305 dye, BASF) on top of the array in the direction normal to the surface, normalized by the emission of phosphor layer without the particles. The emission is significantly enhanced (up to almost 60 times) at the wavelengths of at which the extinction is large due to the SLRs. This enhancement illustrates that the phosphor emits into SLRs, which in turn efficiently radiate to the far-field as an antenna phased array will do. It should be pointed out that the broad resonance in the extinction caused by the LSPPs does not produce a significant enhancement of the emission. Localized resonances only affect the emission of the phosphor in their very close proximity, while the phosphor layer extends over a larger volume to achieve a high brightness in a white LED. This measurement illustrates the relevance of antenna phased arrays in the emission enhancement.

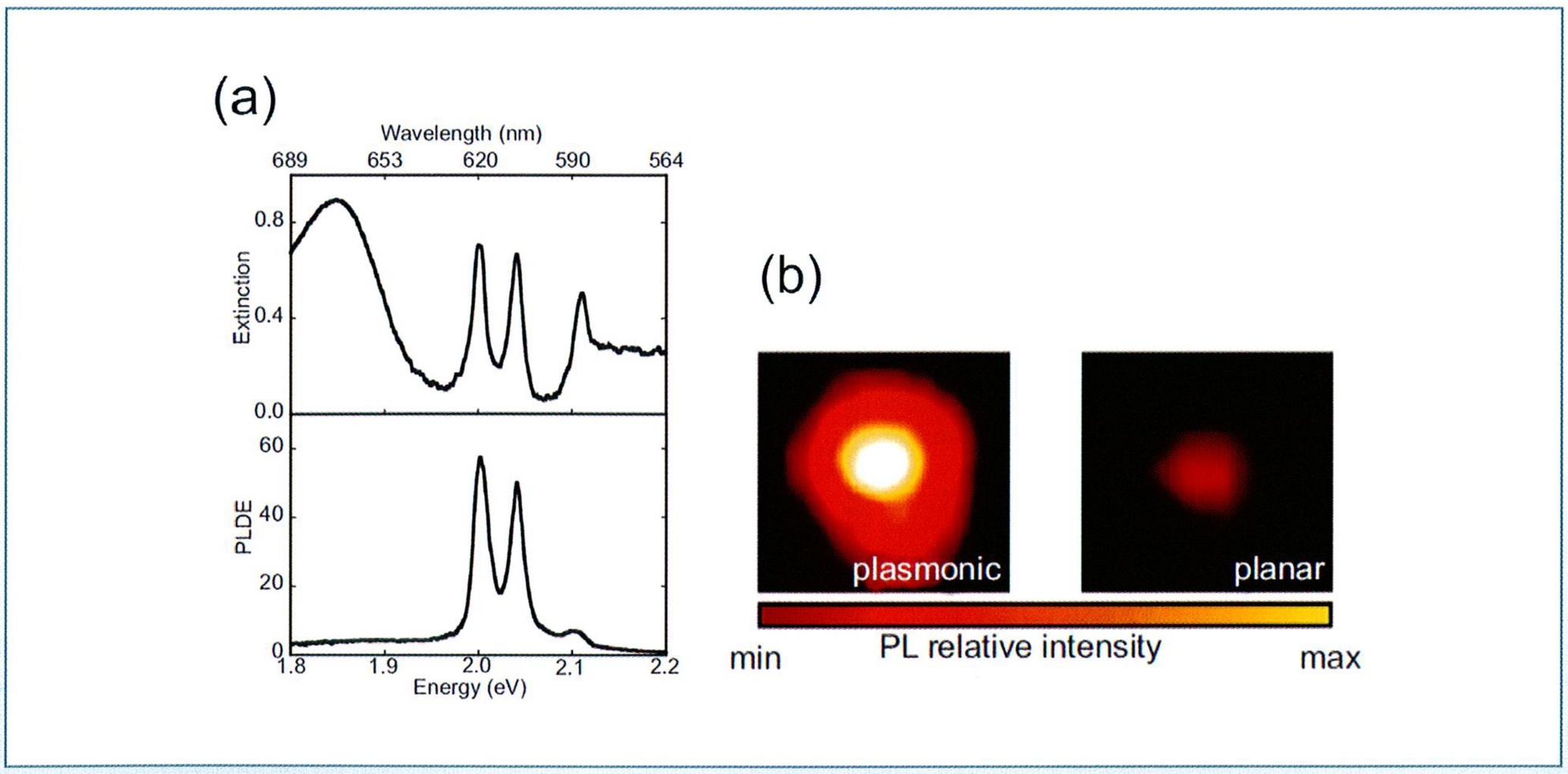

Figure 3. (a) Extinction (upper panel) and photoluminescence directional enhancement (lower panel) of an aluminum nanoparticle array covered by a thin phosphor layer. (b) False-color image of the emitted light intensity by the phosphor layer on top of the nanoparticle array (left side) and by the bare phosphor layer (right side).

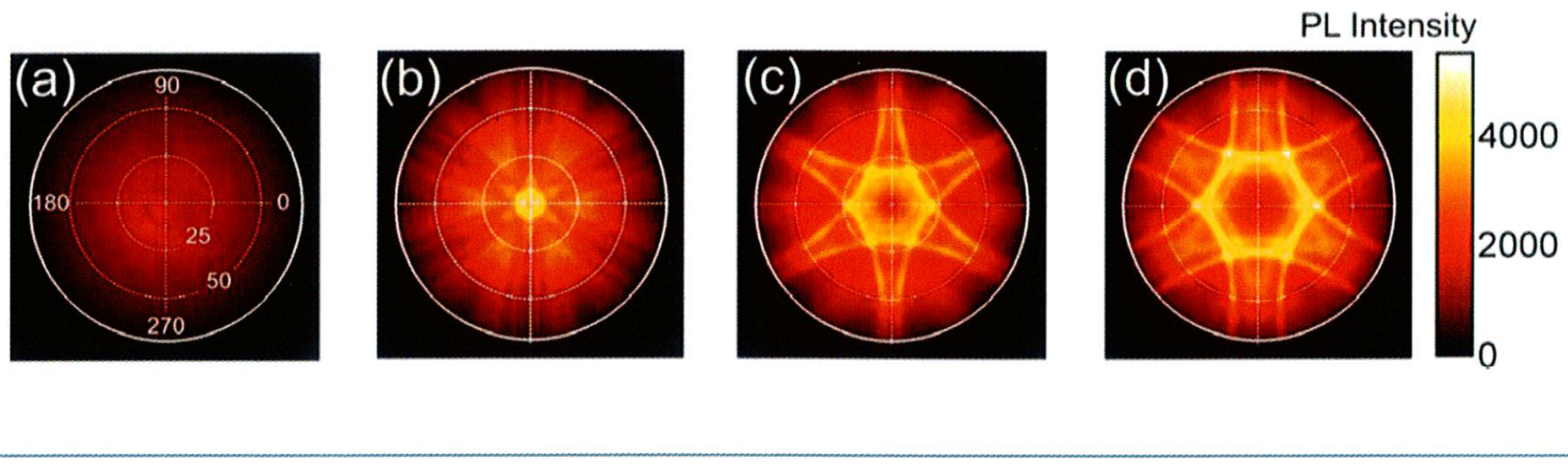

Figure 4. (a) Polar plot images of the red (620 nm) emission of a phosphor layer. (b)-(d) Emission of a similar layer on top of hexagonal arrays of Al nanoantennas with lattice constant (b) a=475 nm, (c) a=425 nm and (d) a=375 nm.

Figure 3(b) provides a clear visualization of the enhanced light emission from the phosphor layer on top of the nanoantenna array (left image) and without the array (right side). These photographs show the emitted intensity in false-color as function of the spatial coordinates. The presence of the nanoantenna array clearly enhances the intensity emitted in the direction of the camera. This enhancement corresponds to value of 17 when integrated over the full emission spectrum of the dye molecules compared to the layer without the nanoantenna array. [ii]

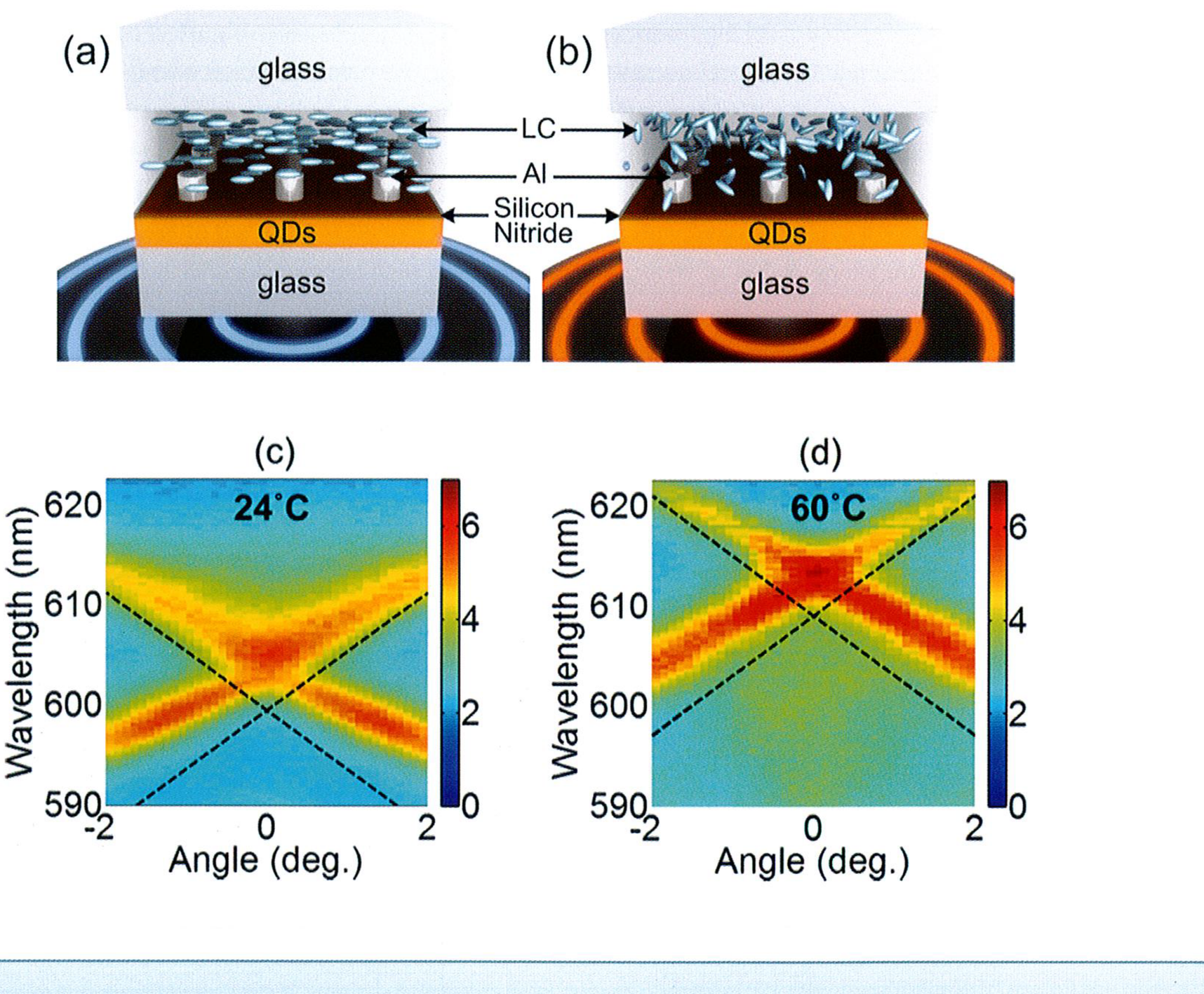

Figure 5: Schematic representation of a phosphor layer of quantum dots (QDs) beneath an array of Al nanoantennas an a liquid crystal below (a) and above (b) the critical temperature. Photoluminescence enhancement of the quantum dot layer (emission normalized by the emission of a QDs layer without the antenna array) below (c) and above (d) the critical temperature as a function of the angle of emission.

Figure 4 illustrates the tuneability of the enhanced directional emission in narrow angular ranges at a wavelength of 620 nm in antenna phased arrays formed a hexagonal arrangement of nanoparticles. [iv] A standard blue LED module was used to pump the phosphor layer. Figure 4(a) corresponds to the polar plot emission of a flat phosphor layer, while 4(b)-(d) are the emissions from similar phosphor layers on top of hexagonal arrays with a lattice constant varying from 475 to 375 nm. Note that the emission is enhanced for all directions compared to the phosphor alone. This is due to the enhanced absorption of the blue pump. At certain directions the enhancement is more pronounced due to the directivity of the emission of the antenna phased array. High symmetry lattices, such as the hexagonal arrays, facilitate a more homogeneous distribution of the emission over the azimuthal angle. Note that the emission of the array with lattice constant 475 nm (Fig. 4(b)) is predominant in a narrow cone along the normal direction. This high degree of directivity is solely achieved by the antenna array, without the need of additional collimating optics.

A characteristic of antenna phased arrays is their capacity of steering the beam of radiation by changing the relative phase of the emission between their antenna elements. A similar effect can be achieved with optical antennas, an example of which is shown in Fig. 5.[v] In this sample a layer of quantum dots (CdSe/CdS/ZnS core-shell) acting as phosphor is beneath a phased array of aluminum nanoantennas. On top of the antennas there is a liquid crystal (E7 Merck) that changes from an order (Fig. 5(a)) to a disordered phase (Fig. 5(b)) at a temperature of 58 C. This change is accompanied by a modification of the refractive index, which corresponds to a change in phase of the scattered light by the nanoantennas. The beam steering by the antenna array can be appreciated in Figs. 5(c) and (d) where the emission enhancement spectrum of the quantum dots is shown as a function of the angle of emission below (c) and above (d) the critical temperature of the liquid crystal. At a fixed color, the emission angle changes as a consequence of the difference in refractive index of the liquid crystal. This phenomenon can be used to control the direction of emission and the color of the emission.

In summary, after the discovery of the blue LED which lead to the efficient generation of white light and the solid state revolution that is currently taking place, the next generation of white LEDs will combine nanostructures to control the emission. Examples of these nanostructures are periodic arrays of nanoparticles that support surface modes. These structures behave as antenna phased arrays, significantly enhancing the emission and beaming the light in defined directions. Nanophotonics will make even brighter the future of solid state lighting.

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