

# Plasmon-enhanced light emission based on lattice resonances of silver nanocylinder arrays

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Diffractive arrays of silver nanocylinders are used to increase the radiative efficiency of InGaN/GaN quantum wells emitting at near-green wavelengths. Large enhancements in luminescence intensity (up to a factor of nearly 5) are measured when the array period exceeds the emission wavelength in the semiconductor material. The experimental results and related numerical simulations indicate that the underlying mechanism is a strong resonant coupling between the light-emitting excitons in the quantum wells and the plasmonic lattice resonances of the arrays. These excitations are particularly well suited to light-emission-efficiency enhancement, compared to localized surface plasmon resonances at similar wavelengths, due to their larger scattering efficiency and larger spatial extension across the sample area. © 2011 Optical Society of America

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Metallic nanoparticles have unique optical properties arising from the excitation of plasmonic resonances, and, as a result, are finding an ever-increasing range of applications in nanoscience and nanotechnology [1]. These resonances produce strong extinction cross sections and large local field enhancements at wavelengths determined by the particle size, shape, and dielectric environment; furthermore, when the nanoparticles are arranged in periodic arrays, their optical response can also be influenced by near-field and/or diffractive coupling. A particularly interesting property is the ability of these nanostructures to increase the light-emission rate of nearby radiators through the direct, near-field excitation of plasmonic resonances [2]. Given the associated large field enhancements and high densities of optical modes, these processes can be extremely fast and, thus, compete favorably with nonradiative decay mechanisms, thereby leading to enhanced quantum efficiency. If, additionally, the emitted plasmonic excitations are efficiently scattered into radiation by the nanoparticles, as opposed to being dissipated via ohmic damping, the overall luminescence yield can also be enhanced [3].

In the field of optoelectronics, this basic idea is currently being investigated as a means to improve the efficiency of solid-state light-emitting materials including, most notably, the InGaN family of nitride semiconductors [4–9]. These alloys are particularly significant as they can potentially provide light emission over the entire visible spectrum by varying their composition. At the same time, however, their internal quantum efficiency strongly decreases with increasing In mole fraction, i.e., as the emission wavelength is extended into the green spectral region and beyond. Therefore, several approaches are currently being investigated to improve their radiative properties, including materials-based techniques, such as novel quantum-structure designs [10], and nanophotonic techniques, such as the use of plasmonic interactions. In recent years, enhanced luminescence from InGaN/GaN quantum wells (QWs) has in fact been reported based on the excitation of either propagating surface plasmon polaritons on rough metal films [4,9] or of localized surface plasmon resonances (LSPRs) in random arrays of spontaneously

formed nanoparticles [5–7]. In [8], nondiffractive square-periodic arrays of Ag nanocylinders (NCs) with highly controlled dimensions were employed, which allowed investigating the fundamental trade-offs associated with LSPR-enhanced light emission. The key limiting factor was identified in the limited scattering efficiency of these excitations, especially when relatively small NCs are used as needed to tune the resonance wavelength into the blue-green spectral region.

In the current work, similar arrays are investigated, except that their period  $P$  is now extended to values comparable to or larger than the QW emission wavelength in the semiconductor  $\lambda_{em}$ , so that strong diffraction of the emitted light can occur. Depending on the period and angle of illumination, some of the first-order diffracted light in these arrays can propagate at grazing angles (i.e., in the plane of the array). As a result, large plasmonic oscillations can be excited in each NC by the in-phase addition of the incident light and the light diffracted by the other NCs in the array. This phenomenon produces strong lattice resonances of mixed plasmonic and photonic character [11–16], which can be expected to be particularly favorable in the context of enhanced light emission due to their larger scattering efficiency and larger spatial extension compared to LSPRs. Indeed, the maximum enhancement in peak luminescence intensity measured in the current work (about 4.8) is significantly larger than the values obtained with the nondiffractive arrays of [8] (up to about 2.8) under otherwise identical conditions (i.e., same QW samples, equal NC height, etc.).

The light-emitting material consists of three pairs of nominally 2-nm-thick InGaN wells and 5-nm-thick GaN barriers, grown by molecular beam epitaxy on a GaN template on sapphire. Its emission spectrum is centered around 495 nm, near the technologically important green spectral region. Several square-periodic arrays of Ag NCs were fabricated on the top surface of the QW samples by electron-beam lithography and liftoff. An exemplary scanning electron microscopy (SEM) image of a fabricated array is shown in Fig. 1(a). To ensure strong near-field coupling between the QW excitons and the array plasmonic excitations, no additional cap layer

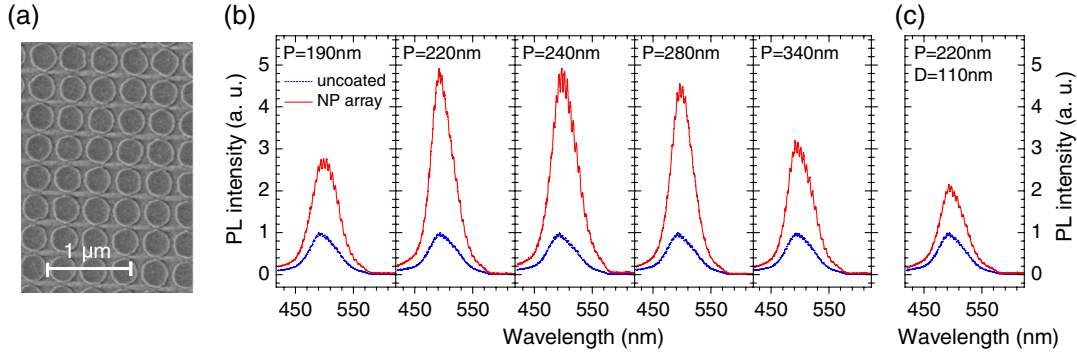


Fig. 1. (Color online) (a) SEM image of a Ag NC array. (b) PL spectra measured with arrays of different period  $P$  and NC diameter  $D = P - 40$  nm (solid curves), and from nearby uncoated regions of the same sample (dashed curves). (c) Same as (b) for  $P = 220$  nm and  $D = 110$  nm.

was grown over the QWs. The array-coated samples were characterized via photoluminescence (PL) measurements, with the QW emission both photoexcited and collected through the sapphire substrate. The pump light was provided by a diode laser emitting at 375 nm. It should be noted that all the arrays investigated in this work feature high transmittance at this wavelength ( $>75\%$ ) and, therefore, they cannot produce any significant increase in pump reflection into the QWs [8].

The solid curves in Fig. 1(b) are the PL spectra measured with five arrays of equal NC height  $H = 55$  nm and equal nearest-neighbor spacing  $S = 40$  nm, but different periods  $P$  in the range of 190 to 340 nm (and, therefore, different diameters  $D = P - S$ ). The dashed curves are simultaneously measured emission spectra from nearby uncoated regions of the same sample. In each panel, both spectra are normalized to the peak value of the dashed trace. At the lowest array period considered in the figure, the PL peak intensity is enhanced relative to the uncoated-QW case by a factor  $r_{\text{PL-I}}$  of about 2.8, consistent with the results of [8]. As  $P$  is increased to 220 nm,  $r_{\text{PL-I}}$  suddenly increases to a value of about 4.8 and then gradually decreases in larger-period arrays. If we assume a refractive index of 2.3 for the InGaN/GaN multiple-QW active medium, which is reasonable for this materials system at 495 nm including carrier-induced contributions [17], the peak emission wavelength in the semiconductor  $\lambda_{\text{em}}$  is 215 nm. As the array period is increased past this value, grazing diffraction of the QW luminescence becomes enabled [11]. Correspondingly, the QW light-emitting excitons can couple to the array lattice resonances, leading to the large measured increase in  $r_{\text{PL-I}}$ .

Additional information about the NC array geometries under study can be obtained from their transmission spectra. These are shown in Fig. 2, as measured with nominally identical arrays fabricated on bulk GaN films grown on sapphire (i.e., without the QWs that would otherwise provide strong optical absorption in the spectral range of interest). The main LSPR of the NCs manifests itself through a pronounced dip in each spectrum, which is found to redshift and broaden with increasing  $D$  (i.e., increasing  $P$  in these arrays) as expected for plasmonic dipolar resonances [1]. The large spectral separation between these dips and the QW emission wavelength confirms that the PL-intensity enhancements of Fig. 1(b) cannot be simply attributed to the excitation of such dipolar LSPRs. At the same time, additional resonances

on the short-wavelength side of the dipolar dip are also seen in each spectrum of Fig. 2, some of which closely overlap with the QW emission wavelength.

To further elucidate the PL measurement results, numerical simulations based on the finite-difference time-domain method were carried out. In Fig. 3(a), we show results obtained for the  $P = 220$  nm array geometry of Fig. 1(b), for the simple case of plane-wave illumination at normal incidence with linear polarization along a primitive lattice vector of the array. The trace plotted in this figure is the average electric-field-intensity enhancement spectrum produced by the NCs in the plane of the QWs, which is found to be dominated by a strong peak at a free-space wavelength of 510 nm. This value is very close to the array period multiplied by the substrate refractive index used in the simulations (2.3), again indicating that the peak is associated with a lattice resonance of the array. Additional simulations with varying values of  $P$  and/or varying angles of incidence confirm the presence of such resonances, while at the same time revealing a complex family of higher-order plasmonic excitations related to the large size of the NCs and their small spacing (which produces a topology resembling a periodic distribution of nanovoids). In the arrays under study, lattice resonances are, therefore, created through the strong

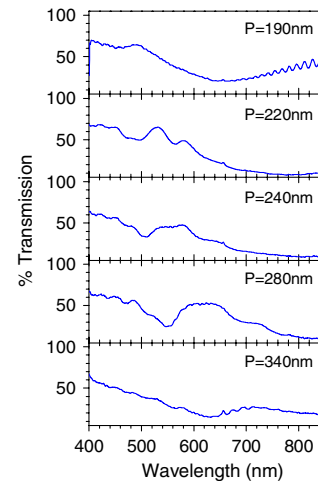


Fig. 2. (Color online) Transmission spectra of the same array geometries of Fig. 1(b) fabricated on a GaN film grown on sapphire, normalized to the transmittance of the bare GaN/sapphire substrate.

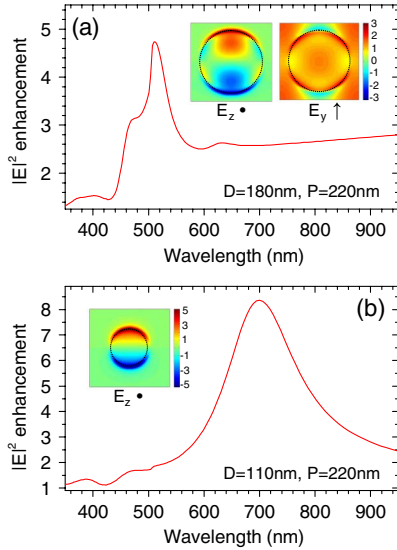


Fig. 3. (Color online) (a) Average electric-field-intensity enhancement spectrum produced in the QWs by an NC array with  $P = 220$  nm and  $D = 180$  nm, under plane-wave illumination at normal incidence with linear polarization in the  $y$  direction. Insets:  $z$  and  $y$  components of the electric field at the wavelength of maximum enhancement, plotted versus position in the plane of the QWs within a unit cell of the array. The dashed circles show the NC outline. The  $z$  and  $y$  directions are indicated by the dot and arrow, respectively. (b) Same as (a) for an NC array with  $P = 220$  nm and  $D = 110$  nm.

excitation via diffractive coupling of these (otherwise weak) higher-order plasmonic oscillations. Correspondingly, as illustrated in Fig. 3(a), large field enhancements are produced at wavelengths within the QW emission line shape, thereby increasing the QW radiative efficiency and output PL intensity.

The color maps in the insets of Fig. 3(a) show the leading components of the electric field at the lattice resonance wavelength (510 nm), calculated as a function of position in the plane of the QWs within a unit cell of the array, under the same excitation conditions as the main trace in the figure. These field distributions reveal a rather delocalized pattern with large enhancements both underneath and in between neighboring NCs. In contrast, dipolar LSPRs generally produce highly confined field enhancements near the edges of the NCs [an example is shown in the inset of Fig. 3(b)]. It should be noted that a relatively delocalized field distribution is advantageous in the current context, where we are dealing with a uniform ensemble of light-emitting excitons across the entire sample area. Furthermore, the lattice resonance of Fig. 3(a) is obtained with significantly larger NCs compared to LSPRs at similar wavelengths, which is also very desirable because of the increase in NC scattering efficiency with increasing diameter [1]. These considerations explain the particularly large PL-intensity enhancements observed in Fig. 1(b).

Further insight into the underlying enhancement mechanism can be obtained from a comparison between the array of Fig. 3(a) and one with the same period  $P$  (220 nm) but smaller NC diameter  $D$  (110 nm). On the one hand, the latter array can be expected to provide stronger first-order diffraction because of its 50% duty cy-

cle (i.e.,  $D = P/2$ ). On the other hand, as shown in Fig. 3(b), its electric-field-intensity enhancement spectrum is dominated by a broad peak at 700 nm whose field distribution (plotted in the inset) reveals a purely dipolar LSPR nature, whereas the lattice resonance near 510 nm has almost completely disappeared. This behavior is expected for arrays of small NCs lying on the interface between two different media, where Fresnel reflection of grazing diffracted light can strongly quench the lattice resonances via destructive interference [13–16]. In contrast, in the case of larger NCs, as in Fig. 3(a), phase variations across the NCs can partly prevent this quenching [16]. Experimentally, arrays based on the geometry of Fig. 3(b) have also been fabricated on the same QW samples and tested; as shown in Fig. 1(c), a relatively modest PL-intensity enhancement of only about 2.1 was correspondingly measured. We can therefore conclude that the large enhancements reported in this work cannot be ascribed to efficient diffraction alone (i.e., increased extraction of in-plane emitted light). Rather, the key attribute is a pronounced lattice resonance near the emission wavelength, which can strongly couple to the QW excitons and increase their light-emission efficiency. Proper choice of both NC diameter and array period is critical to maximize the strength of this process.

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