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Surface plasmon enhanced UV emission in AlGaN/GaN quantum well

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The surface plasmon (SP) energy for resonant enhancement of light has shown to be modified by the epitaxial substrate and the overlying metal thin film. The modification of SP energy in AlGaN/GaN epitaxial layers is studied using spectroscopic ellipsometry for enhanced UV-light emission. Silver induced SP can be extended to the UV wavelength range by increasing the aluminum concentration in $Al_xGa_{1-x}N$ epilayer. A threefold increase in the UV-light emission is observed from AlGaN/GaN quantum well due to silver induced SP. Photoluminescence lifetime measurements confirm the resonant plasmon induced increase in Purcell factor as observed from the PL intensity measurements. © *2010 American Institute of Physics*. [doi:10.1063/1.3515419]

GaN/InGaN nitride quantum well based light emitters were one of the first III-V semiconductor material systems that were employed to demonstrate resonant exciton-surface plasmon polariton (SPP) coupling,[1,2] leading to the enhancement of light emission in the visible wavelength regime.[3,4] Surface plasmon (SP) coupled to excitons or free carriers in AlGaN/GaN system offers an attractive alternative to enhance the light emission in the UV wavelength range. The internal quantum efficiency of AlGaN/GaN emitters is rather low compared to the InGaN system,[5] and AlGaN/GaN emitters are therefore ideal for plasmonic enhancement. The efficiency of UV emission is currently restricted by the lattice mismatch of AlGaN epitaxial layers to sapphire substrate or the cost effectiveness of AlGaN substrates.[6] In the present paper, we study the factors influencing the SP coupling in AlGaN/GaN quantum well system and demonstrate SP induced enhancement of light emission in the UV wavelength region.

SPP induced light emission depends on the resonant interaction of the SP modes with the exciton emission at a certain wavelength. In nanoscale emitters, the resonant interaction can be achieved by using an appropriate metal with a SP energy resonant to the emission wavelength of the light emitter, which can be tuned by changing the dimension of the nanoscale emitter such as the width of a quantum well (QW) or wire or the diameter of a quantum dot.[7] The SP energy at the interface of a metal thin film and a semiconductor can also be influenced by the dielectric constant of the substrate.[8] Silver induced SPP has been reported as the most effective means for visible light enhancement in nitride semiconductors. The surface plasmon energy at the Ag-GaN interface has been reported[1] to be 2.95 eV, which renders Ag induced plasmon ineffective for the UV wavelength regime. However, as the refractive index of AlN reduces by nearly 19.5% compared to GaN,[9,10] a corresponding change in the SP energy is expected for GaN nitride quantum well based UV-light emitters capped with AlGaN layers. We therefore investigate the change in the surface plasmon energy for Ag metal film with various Al concentrations in $Al_xGa_{1-x}N$ epilayer and the effect of various metals on GaN for the design of UV-light emitters.

The details of the epitaxially grown wurtzite AlGaN alloy films used to study the compositional variation of the surface plasmon energy are described elsewhere.[11] The AlGaN layers were coated with 10 nm thin Ag metal layer using electron beam evaporation. The SP energy at the metal semiconductor interface was estimated from the effective dielectric constant measured using a Woollam spectroscopic ellipsometer. The measurements were carried out at room temperature at an incident angle of 75° for the 300–450 nm wavelength range. The dielectric constants were measured using a conventional multilayer model[9] including the surface and interface roughness of the metal layer after deposition and the semiconductor epilayer before the metal deposition. From Fig. 1, it is observed that the SP energy at the Ag/AlGaN interface increases from 2.97 to 3.7 eV as the Al mole fraction increases from 0% to 100%. Therefore this increase in the SP energy with Al concentration provides us an option to couple high energy photons from QWs in the UV range to the SPP modes that normally cannot be coupled in the case of Ag-GaN system.

The inset of Fig. 2(a) shows the schematic of the sample that has been used to study the SPP enhanced light interaction in the UV regime. The AlGaN/GaN QW structure was synthesized using molecular beam epitaxy and capped with

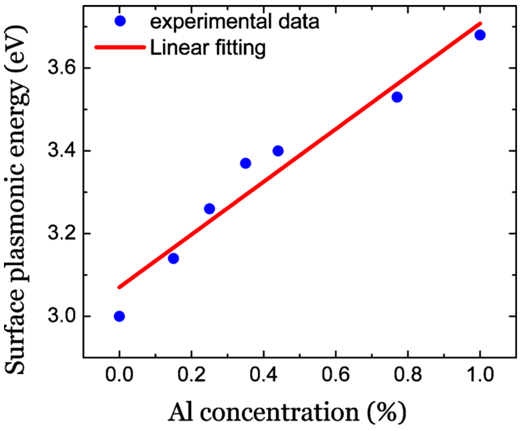

FIG. 1. (Color online) Variation of silver induced surface plasmon with Al mole fraction in AlGaN epilayer measured by spectroscopic ellipsometry.

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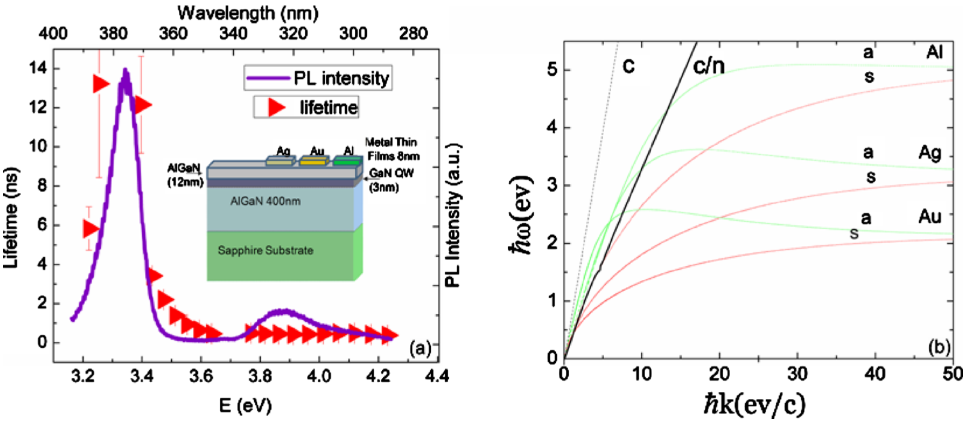

FIG. 2. (Color online) (a) The schematic of AlGaN/GaN quantum well structure used for studying the effect of surface plasmon enhancement (inset). PL spectrum and recombination lifetime measured using time-resolved PL spectroscopy measured at 10 K. (b) Dispersion relations for 8 nm Al, Ag, and Au layers on AlGaN/GaN/AlGaN quantum well showing the normal/antisymmetric (A) and the tangential/symmetric branches (S).

various metals for exciting SPP. A 3 nm GaN QW layer with 3 periods has been grown on a $\mathrm{Al_{0.23}Ga_{0.67}N}$ buffer layer with a 12 nm $\mathrm{Al_{0.23}Ga_{0.67}N}$ cap layer to enable SPP coupling. Photoluminescence (PL) emission intensity measurements were performed using a continuous wave (cw) 18 mW HeCd laser at 325 nm. The PL lifetime was measured using a tripled Ti:Sapphire femtosecond laser excitation at 267 nm wavelength (4.64 eV) with 60 mW average power. The PL signal was monodispersed using a spectrometer, and the lifetime was measured using a Hamamatsu Streak camera with an effective resolution of 15 ps. The time-integrated PL spectrum and the corresponding lifetime of the carriers within the GaN/AlGaN QW measured at 10 K are shown in Fig. 2(a). The pump laser excites the carriers into the AlGaN barrier, which results in an emission at $\sim 3.8-4.0$ eV, whereas the emission from the GaN quantum well is observed at ~ 3.34 eV. The lifetime of the carrier in the quantum well is relatively longer than that in the AlGaN barrier layer, which is dictated by the carrier recombination due to the carrier capture into the GaN well and nonradiative recombination at the interface.

Figure 2(b) shows the theoretical estimate of SPP dispersion[12] for 8 nm metal films on a multilayer structure composed of AlGaN (12 nm)/GaN (3 nm)/AlGaN (400 nm)/sapphire. For the 8 nm thin metal layer, the SPs at the air-metal and the metal-AlGaN interfaces couple together, generating symmetric and antisymmetric plasma oscillations. The dielectric constants used in the calculation were interpolated from tabulated[10,13] values. For large wave vectors, the symmetric modes asymptotically approach an energy, which agrees closely with the spectral position of the plasmon resonance in a system.[1,2] The portions of the antisymmetrical plasmon branch between the c and c/n light lines must have a complex wave vector. The antisymmetric branches are "leaky" plasmon modes, which are not confined to the interface, but can propagate into the AlGaN layer due to the small wave vector to the left of the c/n light line, which can lead to an enhancement in plasmon enhanced coupling to excitons in QWs.

Figure 3(a) shows the surface plasmon induced modification of the cw PL emission from the GaN quantum well due to Au, Ag, and Al thin films with 8 nm thickness. The cw PL measurements were made using the 325 nm (3.81 eV) excitation of a HeCd laser with an average incident power of 18 mW. The metal films were deposited using electron beam evaporation, and a top excitation geometry (with the source and detector being above the metallic side of the sample) was used[1,2] to study the effect of surface plasmon coupling. It is observed that for an excitation below the barrier level, the Al and Ag thin films show an enhancement in PL emission compared to bare sample, whereas the Au coated QW shows a slight quenching at 300 K. From the theoretical estimates in Fig. 2(b), it is observed that the leakage of the SP modes for the Ag/Air interface into the AlGaN/GaN QW results in coupling with the exciton in the quantum well and results in the enhancement in light emission as observed in certain quantum confined structures.[14] The SPP induced enhancement in PL emission is similar to that observed in the InGaN system,[3] though in the present case, the samples were excited from the top instead of the back illumination geometry applied by Okamoto *et al.*[7] In case of the InGaN/GaN QWs excited from the top, the emission from the wells is actually quenched due to the resonant plasmon coupling in the presence of Ag thin film.[2] So to investigate the origin of enhancement, the recombination lifetime has been measured as the spontaneous emission rate of carriers are enhanced due to SPP coupling.[2,3]

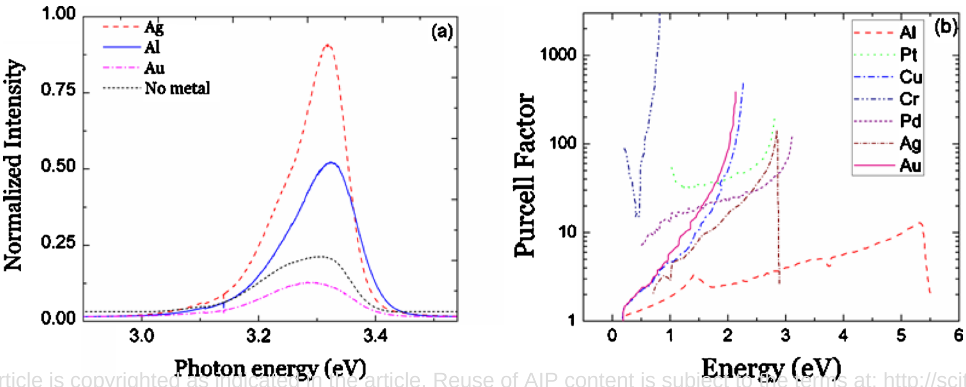

FIG. 3. (Color online) (a) The comparison of room temperature PL emission from AlGaN/GaN quantum well in the presence of Al, Ag, and Au induced metal surface plasmon at 300 K (excitation wavelength at 325 nm). (b) An estimation of Purcell enhancement in AlGaN/GaN quantum well due to surface plasmon interaction induced by various metals on the GaN cap layer.

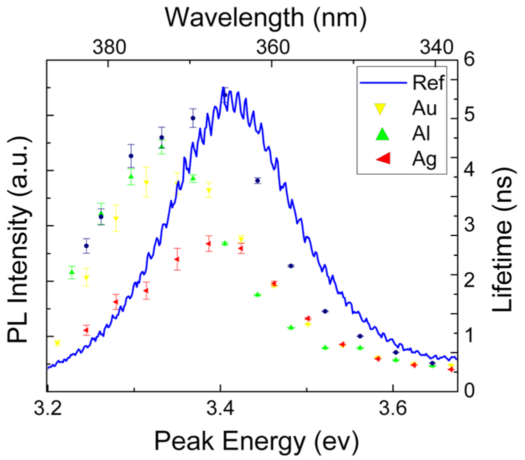

FIG. 4. (Color online) The comparison of time-integrated photoluminescence and PL recombination lifetime in the presence of metal induced surface plasmon polariton measured using time-resolved PL (excitation wavelength at 267 nm). Rectangles: reference sample (no metal); pyramid (green): Al; inverted pyramid (yellow): Au; and side-flipped pyramid (red): Ag.

As the surface plasmon energy of the Au-GaN layer is below the emission wavelength of QW, the surface plasmon modes due to Au does not play any role in the spontaneous emission process. A slight quenching of the PL in Fig. 3(a) is observed in the case of Au due to the absorption of the incident HeCd laser light as well as the PL emission by the metal layer. Though Al metal has a relatively higher energy SPP mode, which is more suitable for coupling to UV photons, in the present case, Ag is observed to be more effective at 3.34 eV as in the case of visible light enhancement from the InGaN/GaN QW system. We thereby also present a theoretical estimation of the Purcell enhancement factor for various metals coupled to GaN/AlGaN QW (in Fig. 2), which is regarded as the figure of merit for surface plasmon based emitters.[1] The Purcell enhancement factor is obtained from the ratio of the surface plasmon induced spontaneous recombination rate (Γ_p) to the radiative recombination rate (Γ_o) of the QW,

$$F_p(\omega) = \frac{\Gamma_p(\omega) + \Gamma_0(\omega) + \Gamma_{\mathrm{nr}}(\omega)}{\Gamma_0(\omega) + \Gamma_{\mathrm{nr}}(\omega)} \approx 1 + \frac{\Gamma_p}{\Gamma_0}, \qquad (1)$$

where Γ_{nr} is the nonradiative recombination rate and is considered to be negligible compared to the plasmon mediated radiative recombination process. The modified light emission depends on the density of the surface plasmon modes $\rho(\hbar\omega)$.

Utilizing the plasmon energy of various metals[15] and the optical constants of GaN/AlGaN,[16] the Purcell enhancement factor for various metal/GaN interface has been estimated using Eq. (1). It is observed that Al can couple to deep UV photons due to higher SP energy but has a relatively an order lower enhancement factor compared to Ag or Pt, which can enhance the light emission up to ~2.9 eV. Au and Cu can resonantly enhance visible wavelength emission at around 2.2–2.4 eV with a higher Purcell factor compared to Ag. Chromium has one of the highest Purcell enhancement factor and can be effective for resonant SP coupling, the near-infrared wavelength range including the communication wavelength intersubband devices at 1.55 μm.[17]

Figure 4 shows the time-resolved PL measurement for the AlGaN/GaN QWs in the presence of various metal layers. It is observed that below and around the emission edge of the QW, the PL recombination lifetime is not influenced by the Au or Al thin films, whereas the Ag surface plasmon induced recombination process is significantly fast implying radiative decay though SPP channels. At the QW emission edge, the surface plasmon induced contribution due to Al increases and the PL recombination lifetime becomes comparable to the silver surface plasmon induced recombination process. Within the width of the emission spectrum, the Ag induced SP decay rates are nearly three times faster than the spontaneous decay rate of the AlGaN/GaN quantum well. This enhancement in the decay rate is similar to the magnitude of the PL enhancement observed in the cw measurement [Fig. 3(a)].

In conclusion, we present the enhancement of UV-light emission from AlGaN/GaN quantum wells based on resonant surface plasmon interaction. We demonstrate that the surface plasmon energy of Ag/AlGaN material system can be increased to the ultraviolet wavelength regime by increasing the Al concentration in the AlGaN cap layer of the light emitter. We also study the applicability of various metals on GaN semiconductor for the surface plasmon enhanced light emission. The increase in the light emission from AlGaN/GaN quantum well due to being Ag and Al induced corresponds to the enhancement in the spontaneous emission rate.

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