Dependence of resonant coupling between surface plasmons and an InGaN quantum well on metallic structure

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The authors demonstrate the metallic-structure dependent surface plasmon (SP) coupling behaviors with a blue-emitting InGaN/GaN quantum well (QW), which is 10 nm away from the metallic structures. The SP-QW coupling behaviors in the areas of semiconductor surface coated with silver thin film and silver nanoparticles are compared. It is found that both the suppression of photoluminescence (PL) intensity and the reduction of time-resolved PL (TRPL) decay time strongly depend on the metallic morphology. A phenomenological model of carrier relaxation in the SP-QW coupling process is built to fit the TRPL decay profiles for calibrating the reasonable decay time constants of carrier and SP. © 2006 American Institute of Physics. [DOI: 10.1063/1.2390639]

The enhancement of electron-hole recombination rate in a semiconductor quantum well (QW) through the coupling between the dipoles in the QW and the surface plasmons (SPs) on a nearby metallic structure has been reported for the potential application to the increase of the emission efficiency in a GaN-based light-emitting device. The strength of such a coupling process is determined by the SP electric field distribution around the QW and the SP density of states, following Fermi’s golden rule. The SP field distribution, either propagating or localized, is controlled by the supporting metallic structure. In the coupling process, excited dipole energy inside a QW is transferred into the SP modes. In this situation, the radiative emission of the semiconductor can be enhanced or suppressed, depending on the phase-matching condition for SP radiation. A rough surface or a grating structure at the metal/semiconductor interface significantly modifies the SP dispersion relation and hence controls the phase-matching condition.

In either case of emission enhancement or suppression, the recombination rate is increased. Such an increase can be confirmed with the measurement of the time-resolved photoluminescence (TRPL) spectroscopy.

In this letter, we report and interpret the observation of the dependence of the coupling between SP and an InGaN QW on the metallic structure. The variation of silver morphology across the boundary of a finite-area silver film coated on an InGaN/GaN QW episturcure is used for the study of the metallic-structure dependence. Nanoparticles of different densities are distributed in the transition zone between the silver film and the bare semiconductor area. Different coupling behaviors are observed in different areas from the thin film, through the nanoparticles, to the bare semiconductor surface.

The InGaN/GaN single-QW epitaxial sample was grown on (0001)-oriented sapphire substrate with metal organic chemical vapor deposition. After the growth of a buffer layer and a 2 μm GaN layer, a 3 nm InGaN QW layer was deposited. The QW was covered by a 10 nm GaN cap layer. A 50 nm silver film was then coated on the sample with a glass plate covering part of the sample and serving as the mask. Because the glass plate did not tightly mask the sample, diffusive silver atoms migrate into the area beneath the glass plate to form nanoparticles on the surface of the sample. The density of nanoparticles is expected to diminish gradually along the distance from the glass plate edge. The sample was focused with an ×50 objective of 0.42 in numerical aperture. Figure 2 shows the PL spectra in the four areas. The spectral FWHMs in areas A, B, and C are broader than that in area D, indicating the nature of frequency-dependent SP-QW coupling.

FIG. 1. Schematic drawing of the sample structure. A silver film of 50 nm in thickness is coated to cover part of the top surface. Silver nanoparticles exist at the boundary of the silver coating region to form the four areas denoted by A through D.
the dashed lines in Figs. 3(a)–3(d). One can see that area D is quite flat with 0.3 nm in the standard deviation of height. The standard deviations of height in areas A, B, and C are 2.5, 5, and 4.5 nm, respectively. The nanoparticle density gradually decreases when the probe moves away from the silver thin film edge.

Figure 4 shows the scanning results of the integrated PL intensity and the normalized transmission when the probe moves away from the silver-film area, through the nanoparticle areas, to the bare semiconductor area. The transmission intensities in areas B, C, and D are almost the same, with their average normalized to unity. At the boundary between the silver film and the dense nanoparticle area, the transmission drops abruptly by 80%, showing the low transmission of the silver film. However, the integrated PL intensity shows four different levels in the four areas. The highest PL intensity is observed in the bare semiconductor area, followed by that in the area of silver film, and then by the area of less dense nanoparticles. The weakest PL intensity is observed in the area of dense nanoparticles. It is interesting to see the sharp boundary between areas B and C.

PL results in Fig. 4 show the suppression of emission through the SP-QW coupling. The assertion of SP-QW coupling is supported by the observations of smaller and negligible PL suppressions and TRPL decay rate changes in the similar samples of 20 and 100 nm GaN spacers, respectively, when compared with the above-mentioned sample of 10 nm GaN spacer. The increasing nanoparticle density, when the probe moves toward the silver thin film edge, implies the increasing spatial frequency of the rough surface morphology. Therefore, the number of localized surface plasmon modes increases discretely along the gradual increase of nanoparticle density. Areas B and C simply represent two neighboring regions of different surface plasmon mode numbers, which effectively couple to the QW. The difference in coupling mode number led to a sharp change of PL intensity between areas B and C.

Figure 5 shows the room-temperature TRPL profiles, with the excitation power at 10 mW and spot diameter at 50 μm, in the four areas with the decay time constants of the first-stage decays labeled next to the corresponding curves. Non-single-exponential decays are observed in all the curves. Such a decay is due to the recovery of the quantum-confined Stark effect (QCSE) from carrier screening. The QCSE results in the reduction of oscillator strength and hence weaker SP-QW coupling and radiative recombination such that the TRPL decay becomes slower. A stronger SP coupling leads to a faster carrier density decrease and hence an earlier recovery of the QCSE. Hence, the turning point of the TRPL profile appears the earliest in area B. Also, the contrast between the first-stage fast decay and the second-stage slower decay is the largest in area B.
TABLE I. Decay time constants used to fit the TRPL profiles in Fig. 5 for the four areas.

<table>
<thead>
<tr>
<th></th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\tau_R) (ns)</td>
<td>1.45</td>
<td>3.9</td>
<td>1.96</td>
<td>1.55</td>
</tr>
<tr>
<td>(\tau_{NR}) (ns)</td>
<td>30.5</td>
<td>30.5</td>
<td>30.5</td>
<td>30.5</td>
</tr>
<tr>
<td>(\tau_{SP}) (ns)</td>
<td>0.5</td>
<td>0.24</td>
<td>1.03</td>
<td></td>
</tr>
<tr>
<td>(\tau_{SP-R}) (ps)</td>
<td>15.14</td>
<td>280</td>
<td>320</td>
<td></td>
</tr>
<tr>
<td>(\tau_{SP-NR}) (ps)</td>
<td>2</td>
<td>0.3</td>
<td>2</td>
<td></td>
</tr>
</tbody>
</table>

Note that the sequence of decreasing PL decay time among the four areas is different from that of decreasing PL intensity in Fig. 4. The different sequences of areas A and C between the PL intensity and the TRPL decay time imply different ratios of radiative over nonradiative SP decay rates between the two areas. To analyze the results in Fig. 5, particularly the first-stage decays, we built a phenomenological model for various decay rates of the QW carrier density \(n\), the SP density \(n_{SP}\), and the cumulative photon density \(n_{ph}\). We established three coupled differential equations to fit the TRPL profiles in Fig. 5 as

\[
\frac{dn(t)}{dt} = G(t) - \left( \frac{1}{\tau_R} + \frac{1}{\tau_{NR}} + \frac{1}{\tau_{SP}} \right) n(t),
\]

(1)

\[
\frac{dn_{sp}(t)}{dt} = \frac{1}{\tau_{SP}} n(t) - \left( \frac{1}{\tau_{SP-R}} + \frac{1}{\tau_{SP-NR}} \right) n_{sp}(t),
\]

(2)

and

\[
\frac{dn_{ph}(t)}{dt} = \frac{n(t)}{\tau_R} + \frac{n_{sp}(t)}{\tau_{SP-R}}.
\]

(3)

Here, \(G(t)\) is the near-band-edge carrier generation rate, which can be approximately obtained from the slow-decayed TRPL profile of intrinsic QW. We took the derivative of curve D in the rising range to obtain \(G(t)\), as shown with the curve of empty circles in Fig. 5. In Eqs. (1)–(3), \(\tau_R\) and \(\tau_{NR}\) represent the radiative and nonradiative decay times of the QW, respectively. Also, \(\tau_{SP}\) stands for the SP-QW coupling time constant. Meanwhile, \(\tau_{SP-R}\) and \(\tau_{SP-NR}\) denote the radiative and nonradiative decay time constants of SP, respectively. In the fitting process, we used a weakly excited TRPL profile in area D and the calibrated internal quantum efficiency of the QW to obtain \(\tau_{NR}\). This parameter was then used for fitting other curves to obtain other time constants. In Fig. 5, the continuous lines are plotted based on the calculations of Eqs. (1)–(3) for fitting the experimental data in the first-stage decay range. The fitting curves agree well with the experimental data. In Table I, we list the values of the time constants used for fitting. Here, one can see that SP coupling dominates the carrier relaxation process for all silver-coated areas (smaller \(\tau_{SP}\) compared with \(\tau_R\) and \(\tau_{NR}\) in areas A, B, and C). \(\tau_{NR}\) is assumed to be the same in all areas. However, the dipole radiative recombination rates \(1/\tau_{ph}\) are different among the four areas because of the different degrees of the screened QCSE.\(^9\) Note that \(\tau_{SP-R}\) in area A is significantly smaller than those of areas B and C, indicating that the transfer of SP energy into photon is more effective in area A. This fitting result explains the observation that the SP coupling in area A is stronger than that in area C, but the PL intensity in area A is also stronger than that of the other area. Note that \(\tau_{SP-R}\) in area B is smaller than that in area C. Three possible reasons can be used for explaining this difference: (1) more localized surface plasmon modes are coupled to the QW and hence more modes can radiate in area B, (2) the higher spatial frequency in the surface morphology of area B leads to a situation closer to the phase-matching condition for SP radiation (around 62 nm in period),\(^9\)\(^10\); and (3) a higher plasmon density usually results in a higher decay rate. Finally, the smaller \(\tau_{SP-NR}\) values in all SP-QW coupling areas indicate that the metal dissipation in all silver-coated areas dominates the energy loss in the carrier relaxation process.\(^5\)\(^11\)

In summary, we have demonstrated the metallic-structure dependent SP coupling behaviors with an InGaN/GaN QW, 10 nm away from the metallic structures. The SP coupling with the dipoles in the QW led to emission suppression because of the ineffective SP radiation. In the region of coated silver nanoparticles, the SP coupling was stronger in the area of higher particle density because the number of coupled SP modes was higher. The fitting to a set of TRPL data based on a phenomenological model led to the calibrations of various decay time constants, whose variation trends were consistent with the PL measurement.

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