# Surface plasmon assisted photoluminescence in GaAs–AlGaAs quantum well microstructures

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Surface plasmon resonance has been investigated in a quantum well (QW) GaAs microstructure the photoluminescence (PL) of which is coupled via a submicrometer period grating with surface plasmons (SPs) propagating at $SiO_2$–Au-dielectric interfaces. Introduction of the $SiO_2$ layer allowed to increase both the propagation length and the penetration depth of SPs and, consequently, achieve their enhanced interaction with the QW PL signal. For a QW GaAs–$Al_{0.5}Ga_{0.5}As$ microstructure emitting at 822 nm, a modulated PL emission has been observed in agreement with the calculated resonance conditions expected for such a microstructure and the 375 nm period grating. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2798253]

Due to the inherent surface sensitivity related to the resonant nature of the surface plasmon resonance (SPR) effect, numerous SPR devices for bioanalytical applications have been developed and have become commercially available for almost 20 years.[1] Since the plasmons exist at the metal/dielectric interface, conditions of their excitation are extremely sensitive to the refractive index of the dielectric medium and surface chemistry. Most SPR biosensors use the Kretschmann prism geometry to direct *p*-polarized laser light through a glass prism and reflect it from a gold (Au) film deposited on its surface. The SPR effect leads to the appearance of a resonant dip in the reflected intensity at a defined angle ($\theta_{SPR}$). The bulky structure and elevated cost of conventional prism-based SPR biosensors[2,3] have not allowed the expansion of this technology into the out-of-laboratory space. However, recent advancements in manufacturing nanotechnology of waveguides, fibers, and new sources of optical signals based on nanocrystals, quantum wells, and quantum dots producing bright luminescence have opened an opportunity to develop integrated (compact) SPR biosensors,[4–6] which could potentially offset this deficiency. Grating-coupled SPR devices[7–10] are of particular interest due to their ability to offer real-time and simultaneous monitoring of relatively large areas of biofunctionalized surfaces using conventional imaging detectors.[11]

If a thin metal layer is deposited on a bulk semiconductor, a considerable amount of excitation light can still pass through the film, depending on the nature of the metal, its thickness, and the frequency used. Recombination of some electron-hole pairs excited close to the metal-semiconductor interface could be enhanced due to the coupling of the emitted electromagnetic field with surface plasmons (SPs). Thus, instead of direct emission of photons, this process will produce SPs and dramatically quench photoluminescence (PL). Many authors presented such cases concerning silver and GaN (Refs. 12–15) and Au and GaAs.[16] The interaction between semiconductor PL and SPs could be used to enhance PL emission in two ways. The first approach is to couple the incident light directly with SPs to raise the excitation energy density within the quantum well (QW) region. Such an enhancement, by increasing the excitation energy reaching the QW, has been reported in literature.[16–19] The second approach is to enhance PL emission by extracting the naturally generated SPs out of the surface. By coupling the QW electron-hole pair generation with SP, it becomes possible to increase the spontaneous recombination rate of the QW. The dipole-dipole coupling between SPs and electron-hole pairs a few orders of magnitude faster than radiative deexcitation since the strong electromagnetic field is induced by the high density of SP states.[13,15]

Here, we report on a study of SPs propagating at Au–GaAs and Au–$SiO_2$–GaAs interfaces located above PL emitting GaAs QWs, and we describe conditions leading to the modulated PL emission resulting from the SP-QW PL photon interaction. The ultimate goal of this study was to investigate the feasibility of constructing a monolithically integrated semiconductor-based biosensor with its output signal modulated by SPs.

The investigated GaAs–$Al_{0.5}Ga_{0.5}As$ heterostructure consists of a 7 nm thick GaAs QW buried with a 20 nm thick barrier of AlGaAs and a 10 nm thick cap of GaAs. The room-temperature QW peak emission for this heterostructure is observed at 822 nm (1.51 eV). Two separate SPR microstructures have been studied. The first one (sample A) comprised a 10 nm thick Au film deposited directly on the surface of the heterostructure. A 295 nm period Au grating was fabricated on top of that microstructure. In the second microstructure (sample B), a 350 nm thick layer of $SiO_2$ has been added between the surface of GaAs and a 10 nm thick Au film. A 375 nm period Au grating was fabricated on top of that microstructure. In both structures, the SPs cannot directly interact with the QW, as their penetration depths are far too short to reach the 30 nm deep GaAs well. Therefore, in both samples presented, the generation of SPs at the Au interfaces is solely due to PL emitted photons that couple with the Au layer through the grating.

The fabrication of $1\times1$ mm$^2$ surface area Au gratings was accomplished using a standard electron beam lithography (EBL) technique and a polymethyl methacrylate based lift-off process. Other more efficient and rapid lithography methods could be employed for that purpose. However, the EBL technique offers the prototyping flexibility since various

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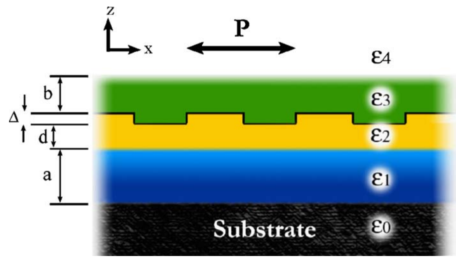


FIG. 1. (Color online) Cross section of the investigated SPR device microstructure. The substrate ($\varepsilon_0$) has a photoluminescence emitting heterostructure with a single quantum well buried about 30 nm below the GaAs surface. The introduction of a thin dielectric layer of $SiO_2$ ($\varepsilon_1$) allowed to increase significantly both the penetration depth and the propagation length of surface plasmons.

grating geometries could be fed directly into the scanning electron microscope (SEM) software, without requiring masks.

The PL measurements were carried out in a commercial mapping system (Philips SPM-200) with collinearly arranged both laser excitation beam (473 nm) and the PL collecting optics. Samples were mounted on a custom designed goniometer that allowed for the collection of the PL signal in the range of ±15° of the surface normal. The irradiation conditions were normalized to achieve angle-independent measurements. For each observed angle, the measured PL signal is an average over the $1\times 1$ mm$^2$ grating region, with uncertainty corresponding to the standard deviation of the measurements, and is divided by the average PL intensity over the nongrated area of the architecture.

A cross section of the investigated SPR microstructure is schematically shown in Fig. 1. The GaAs QW comprising the substrate, a dielectric adaptive layer (of thickness $a$), and a Au layer (of thickness $d$) has been indexed with their dielectric constants $\varepsilon_0$, $\varepsilon_1$, and $\varepsilon_2$, respectively. The layer $\varepsilon_3$ ($\geqslant 1$) has been used to model the biofunctionalized layer (of thickness $b$) surrounded by air ($\varepsilon_4$). The height of the Au grating's corrugation is indicated as $\Delta$. For both samples, $d=10$ nm, but in sample A, $a=0$ and $\Delta=5$ nm, while $a=350$ nm and $\Delta=20$ nm in sample B. Note that classical diffraction is prevented in those samples, as $P<\lambda_{QW}/2$.

The normalized PL intensity as a function of spectral density for sample A is shown in Fig. 2. The $x$ axis in this figure represents the spectral content, $s(\Delta k_x)$, obtained from the following expression for the SP wavevector:

$$k_{SP} = \frac{2\pi}{\lambda_{QW}} n_3 \sin(\theta) + s(\Delta k_x), \tag{1}$$

where $\lambda_{QW}$ is the wavelength of the QW emission, $n_3$ is the refractive index of the emerging media, and $\theta$ is the polar angle of the emerging photon. The choice to directly express the PL intensity dependence on the wavevector $s(\Delta k_x)$ allows to observe the possible solutions of Eq. (1). For a square grating of periodicity $P$ that has been investigated in this work, Eq. (1) can be approximated by

$$k_{SP} \approx \frac{2\pi}{\lambda_{QW}} n_3 \sin(\theta) + \frac{2\pi}{P} m, \tag{2}$$

where $m$ denotes grating diffraction order. This equation indicates that for sample A the SPs should be extracted in air at $k_G=63.8\pm 0.1$ $\mu$m$^{-1}$.

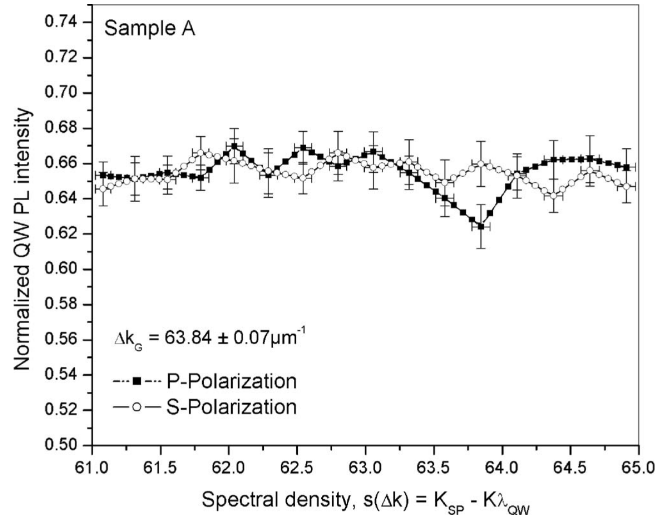


FIG. 2. Normalized QW PL intensity in the grating region of sample A as a function of the grating vector (angle of collected measurements) for a 295 nm grating with ridge height $\Delta=5$ nm.

A weak dip in $p$-polarized PL signal that is observed in Fig. 2 at $k_G=63.84\pm 0.1$ $\mu$m$^{-1}$ corresponds to the collection angle of 8°. This feature coincides with the expected resonant interaction between the QW PL emission and SPs. However, only a small minimum has been observed. To investigate this problem further, we have listed in Table I the values of propagation lengths and penetration depths of SP wavevectors at 1.51 eV expected for Au films of different thicknesses. For a 10 nm thick layer of Au, the SP propagation length is of 73 nm. This means that after propagating such a distance at the Au–GaAs interface, SPs have only $1/e$ (37%) of their original electric field intensity. This distance increases as the metal thickness is increased, but is still significantly shorter than the 295 nm period grating used in this case even for thicker Au films. Thus, a very weak coupling efficiency between QW PL signal and SPs could explain the small amplitude dip observed in Fig. 2. Shorter period gratings would likely improve the interaction with such SPs, but this would require the implementation of challenging and expensive technical processes. The penetration depth of the SPs into the substrate is also enhanced by increasing the metal thickness, but it goes down to 17 nm when the metal layer is 10 nm thick (the case investigated here). In such a case, only a tail of the SP electric wavevector will reach a QW buried about 30 nm below the surface. Note that the propagation length and penetration depth of SPs are strongly dependent on the dielectric constants of the materials that are in contact with the metal layer, and they will significantly increase if a low-dielectric constant material, such as $SiO_2$, is used instead of GaAs.

TABLE I. Calculated propagation length ($L$) and penetration depth ($\delta$) of surface plasmon wavevectors for Au films of different thicknesses ($d$) deposited directly on GaAs ($a=0$) or separated from GaAs by a $SiO_2$ film.

| | $a=0$ | | $a=350$ nm | |
|---|---|---|---|---|
| $d$ (nm) | $L$ ($\mu$m) | $\delta$ (nm) | $L$ ($\mu$m) | $\delta$ (nm) |
| 10 | 0.073 | 16.8 | 4.31 | 69.9 |
| 15 | 0.093 | 23.3 | 5.36 | 107 |
| 30 | 0.116 | 33.9 | 6.54 | 252 |

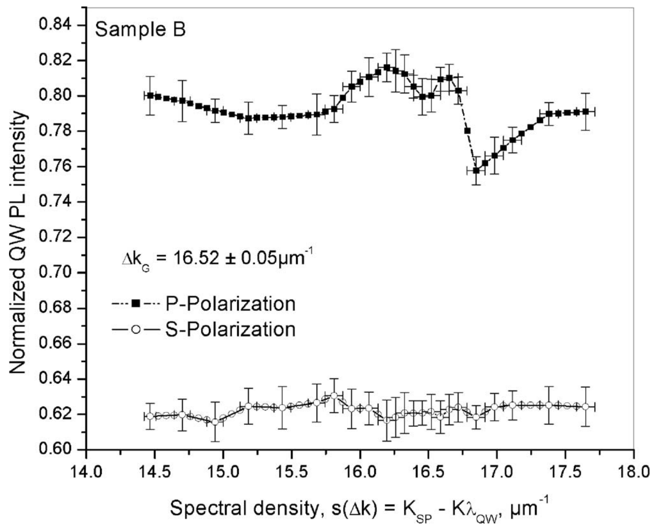


FIG. 3. Normalized QW PL intensity in the grating region of sample B as a function of the grating vector (angle of collected measurements) for a 375 nm grating with ridge height $\Delta = 20$ nm.

For $SiO_2$ and fixed values of $b$ and $d$ (see Fig. 1), we used scattering matrix formalism[20] to design an antitransmission microstructure for the QW PL and antireflecting for the excitation laser. We found that a 350 nm thick layer of $SiO_2$ would reduce the QW PL signal transmission to about 5% of its initial intensity, which implies a significant interaction with the metallic layer and hence the SP generation. At 1.51 eV, the SP theoretical wavevectors are found to be 18.32 and 14.82 $\mu m^{-1}$ for 10 and 15 nm metal thicknesses, respectively. This corresponds to the propagation lengths of 4.31 and 5.36 $\mu$m, respectively. Consequently, we could design a microstructure with the SP diffraction grating having periodicity larger than that used for sample A and still have the conditions for extracting SPs with the first order harmonic. For the 375 nm period grating, we expected to observe in air the SP extraction at the collection angle of 12°.

Figure 3 shows the normalized PL intensity as a function of spectral density for the microstructure with a 350 nm thick layer of $SiO_2$ separating the Au grating from the GaAs cap (sample B). For the $p$-polarized light, an absolute maximum is observed at $s(\Delta k) \approx 16.2\ \mu m^{-1}$ along with some modulations between 16.2 and 16.9 $\mu m^{-1}$ and local minima at 16.5 and 16.7 $\mu m^{-1}$. These values coincide with the grating vector $\Delta k_G = 16.52 \pm 0.05\ \mu m^{-1}$ that was evaluated using SEM measurements.

Our experiments and calculations showed that the grating groove height plays an important role in the photon-plasmon coupling. By increasing the groove height $\Delta$, we observed an increase in the depth of the PL signal modulation. In addition, it can be seen in Fig. 3 (the sample with $\Delta = 20$ nm) that for the $s$-polarized light, the PL intensity is constant over a broad range of scanned wavevectors. However, in comparison to the case discussed in Fig. 2, this component is significantly less intense than for the $p$-polarized light. This is due to the grating region now becoming an efficient wire-grid polarizer absorbing more efficiently the $s$-polarized field crossing the interface. Analogous results and mathematical treatment were presented by Heitmann for fast electrons on modulated surfaces[21] and the effects of surface roughness on SP diffraction were explored by Okamoto *et al.* and Vuckovic *et al.* in different setups.[13,22]

In summary, we have investigated mechanisms of interaction between the GaAs QW PL and the SP polaritons generated at the GaAs–Au and GaAs–$SiO_2$–Au interfaces. The PL emission originated from QWs buried about 30 nm below the GaAs cap and $Al_{0.5}Ga_{0.5}As$ barrier material. The extraction of SPs was achieved with Au gratings that were fabricated on top of the 10 nm thick film of Au. The introduction of a 350 nm thick layer of $SiO_2$ between the Au film and GaAs surface allowed to significantly increase the propagation length and penetration depth of SPs. The QW PL signal in the present experiment served as an energy source from the substrate interacting with the SP device. Recently, the SP induced enhanced PL from silicon nanocrystals has been reported for a back-side irradiated microstructure.[10] Our approach, however, is more attractive for biosensing applications as it offers the possibility of constructing a monolithically integrated SPR device.

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