# Exciton/plasmon polaritons in $GaAs/Al_{0.93}Ga_{0.07}As$ heterostructures near a metallic layer

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We report on the strong coupling between inorganic quantum well excitons and surface plasmons. For that purpose a corrugated silver film was deposited on the top of a heterostructure consisting of GaAs/GaAlAs quantum wells. The formation of plasmon/heavy-hole exciton/light-hole exciton mixed states is demonstrated with reflectometry experiments. The interaction energies amount to 21 meV for the plasmon/light-hole exciton and 22 meV for the plasmon/heavy-hole exciton. Some particularities of the plasmon-exciton coupling were also discussed and qualitatively related to the plasmon polarization.

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Since its first demonstration by Weisbuch *et al.*,[1] the strong photon-exciton coupling regime has been extensively studied in inorganic semiconductor planar microcavities.[2] In this regime the excitons hybridize with the microcavity photons to form polaritons. Epitaxial inorganic semiconductors are materials of choice for these studies since their well-mastered growth yields layers of high crystalline quality. In particular, quantum wells (QWs) exhibit intense excitonic transitions with narrow linewidth suitable for reaching the strong-coupling regime. More recently, polariton nonlinearities[3] and Bose-Einstein condensation have been claimed in semiconductor QWs coupled to planar microcavities,[4] as well as polariton lasing in GaN microcavities at room temperature[5] and in micropillar GaAs/GaAlAs cavities.[6]

The strong-coupling regime has been also demonstrated with surface plasmons (SPs) as predominant optical modes.[7] This regime has been obtained in organic semiconductors, with very large Rabi splitting (several hundreds of meV). During the past decade, the improvement of metal nanostructuration has lead to a renewed interest in SPs.[8] For example, SPs photonics bandgaps,[9] low losses SP guides, and interferometric systems[10,11] have been successfully developed. The interactions between SP and emitters have been also intensively studied. In the weak-coupling regime, a 2-orders-of-magnitude enhancement of the spontaneous emission rate was reported, leading to improved emitting devices,[12,13] and efficient coupling between single-quantum boxes and metallic wires has been demonstrated.[14] In the strong-coupling regime, the metal structuration has been used to tailor the properties of the plasmon/exciton mixed states[15–17] and for efficient polariton extraction.[18] Until now, in the strong-coupling regime, the experiments were restricted to organic materials, *J*-aggregated dyes, or hybrid organic/inorganic materials (perovskites).[19] These materials have high oscillator strength and can be used at room temperature. They are however extremely sensitive to the environment, easily deteriorated by light irradiation, and their structural properties are less controlled than those of inorganic semiconductors.

In this paper we show that the strong-coupling regime can be achieved between SPs and GaAs/AlAs QW excitons. The wide possibilities of plasmon mode engineering coupled to the highly controlled epitaxial heterostructures make this type of structures promising for future developments. In the first part of the paper, the sample and the experiment are described. The dispersion relations deduced from the reflectometry experiments are analyzed in the second part. The formation of coupled plasmon/exciton mixed states is demonstrated. In particular, we show that these polariton states are an admixture of both heavy- and light-hole excitons and SPs. Finally typical behaviors of plasmon/exciton polaritons are qualitatively discussed, considering the polarization of the plasmon modes in interaction with anisotropic structures.

The sample is formed by a corrugated silver film deposited on a GaAs/GaAlAs QW structure. The heterostructure was grown by molecular-beam epitaxy on a GaAs [001] substrate. It consists of five GaAs QWs (10 nm thick) separated by $Al_{0.93}Ga_{0.07}As$ barriers (15 nm). The last QW was covered by a 27-nm-thick $Al_{0.93}Ga_{0.07}As$ barrier and a 3 nm GaAs cap. The upper barrier is larger to prevent nonradiative recombinations induced by surface states or by the semiconductor-metal interface.[20] A luminescence spectrum of the heterostructure without the silver layer is shown in Fig. 1. The QWs were excited with the 488 nm line of an argon laser and maintained at a temperature of 77 K. Two

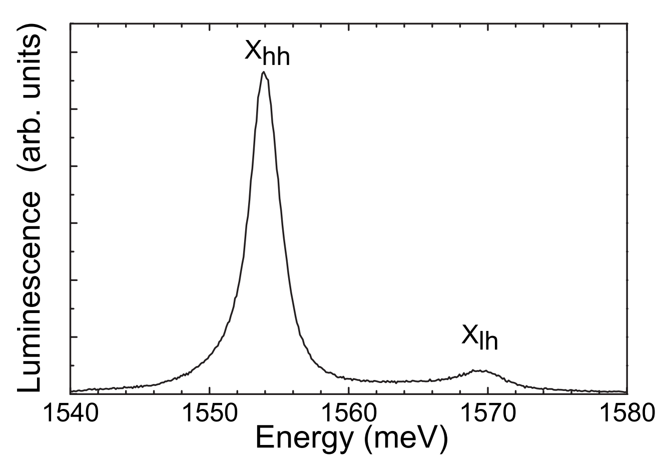


FIG. 1. Luminescence spectrum of the semiconductor heterostructure without silver film measured at 77 K.

 

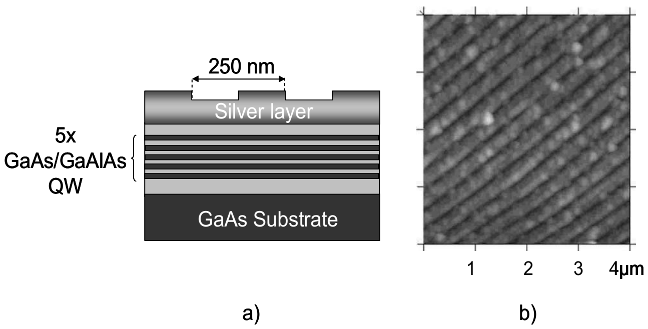


FIG. 2. (a) Schematic layout of the sample showing the heterostructure grown on a GaAs substrate and caped by an engraved silver film (not to scale). (b) Image of the surface topography of the engraved silver film recorded by atomic force microscopy.

peaks are observed at 1554 and 1570 meV with a full width at half maximum of 3 and 4 meV, respectively. They correspond, respectively, to the heavy-hole ($X_{hh}$) and the light-hole ($X_{lh}$) excitons.

Two distinct SP modes coexist on both interfaces of the silver film: Ag/semiconductor and Ag/vacuum. For a flat metal surface, these modes cannot couple to the free-space radiative modes. Therefore a grating has been engraved on the silver surface to make this coupling possible and hence to give access to the dispersion relations. The period of the grating was chosen, so that the Ag/semiconductor plasmon couples to free-space radiations via first-order scattering. For the Ag/vacuum plasmon, no diffracted order couples to free-space modes in the energy range investigated in this study. This geometry prevents the apparition of Ag/vacuum plasmon peaks in the optical spectra and possible interactions between Ag/semiconductor and Ag/vacuum plasmons.[21]

The fabrication of the engraved silver film was done as follows. A thin Ag film (60 nm thick) was deposited on the sample. The grating was then defined by *e*-beam lithography and partially engraved by Ar ion-beam etching. A layout of the sample is drawn in Fig. 2(a). The surface of the engraved silver film was imaged with an atomic force microscope and is shown in Fig. 2(b). The measured grating period is $250 \pm 10$ nm and the groove depth is 20 nm.

Angular-resolved reflectometry experiments have been performed with the sample mounted on the cold finger of a nitrogen cryostat cooled at 77 K. A white light source is focused on the sample. The reflected light is detected by a spectrometer coupled to a cooled Si detector with a lock-in amplifier. A goniometer is used to tune the reflection angle. Reflectometry spectra recorded at different angles are shown in Fig. 3. At $\theta = 29°$, three well-separated dips are present in the spectrum. The large dip at 1482 meV is related to the SP mode. The two sharp dips, well separated from the SP line and lying at 1555 and 1573 meV, correspond to the heavy- and light-hole excitons. The Stokes shift is 1 meV (3 meV) for the heavy- (light-) hole excitons, which is typical for good quality QWs.

Several spectral modifications occur as the detection angle increases. At small angles ($\theta \leq 39°$) the SP energy increases because of the SP dispersion, while the exciton energies remain almost constant. Above 44°, when the SP energy

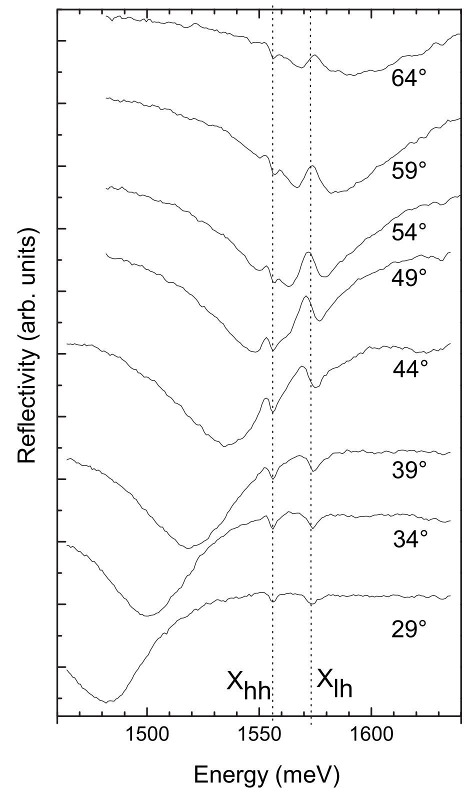


FIG. 3. Reflectometry spectra as a function of the incident light energy for different detection angles. The spectra are arbitrary translated for the clarity of the figure. The dashed lines at 1555 and 1573 meV indicate the energies corresponding to the heavy- and light-hole excitonic transitions, respectively.

is close to the QW excitons energies, the peak previously associated to the light-hole exciton at small angles (1573 meV) starts to move toward higher energies and progressively broadens. The evolution of the dip associated to the heavy-hole exciton is more complex. While a dip remains visible at 1555 meV at all detection angles, a second dip is observed remaining on the low-energy side of the 1573 meV dip up to 59°. A better understanding is gained when plotting the dip energies as a function of the in-plane wave vector of the detected light $[k_d = \frac{2\pi}{\lambda}\sin(\theta)]$, as in Fig. 4.

As mentioned above, a dip is always present at 1555 meV. On the opposite, the energy of the dip associated to the SP mode at small wave vector first increases, as expected for a bare SP mode, but then saturates at a value slightly below 1555 meV for large wave vector, whereas the energy of the dip at 1573 meV remains constant at $k \leq 5.5$ $\mu m^{-1}$ and increases at higher wave vector to follow the bare SP dispersion. Lastly, a fourth dip appears around $k = 6$ $\mu m^{-1}$, emerging from the 1555 meV dip and moving asymptotically toward the bare light-hole exciton energy. These behaviors are the signature of two successive anticrossings, unveiling the strong coupling between the plasmon mode and both excitons.

In order to analyze the experimental results, the coupled $X_{hh}/X_{lh}$/plasmon polariton dispersions were calculated using a coupled oscillator model. The wave vector in the layer plane is a good quantum number. The dispersions were ob-

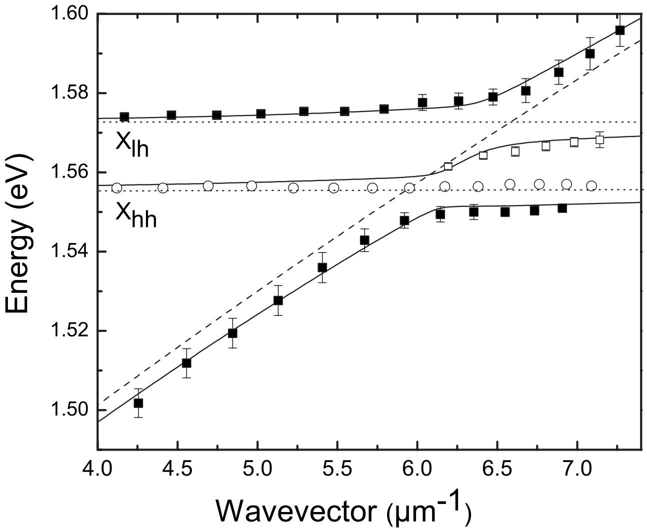


FIG. 4. Reflectometry dip energy (■, □, and ○) as a function of the in-plane wave vector of the detected light. The dashed line is the dispersion relation of a bare SP calculated with a transfer-matrix method. The dotted lines are the $X_{\mathrm{hh}}$ and $X_{\mathrm{lh}}$ bare energies. The calculated polariton energies are drawn in full lines.

tained by diagonalizing the exciton-plasmon Hamiltonian[22]

$$H = \begin{pmatrix} E_{\mathrm{pl}}(k) - i\gamma_{\mathrm{pl}} & V_{X_{\mathrm{hh}}}/2 & V_{X_{\mathrm{lh}}}/2 \\ V_{X_{\mathrm{hh}}}/2 & E_{X_{\mathrm{hh}}} - i\gamma_{X_{\mathrm{hh}}} & 0 \\ V_{X_{\mathrm{lh}}}/2 & 0 & E_{X_{\mathrm{lh}}} - i\gamma_{X_{\mathrm{lh}}} \end{pmatrix},$$

where $E_{\mathrm{pl}}(k)$ is the bare surface-plasmon energy, $\gamma_{\mathrm{pl}}$ is the plasmon homogeneous broadening, $E_{X_{\mathrm{hh}}}$ and $E_{X_{\mathrm{lh}}}$ are the bare heavy- and light-hole exciton energies, $\gamma_{X_{\mathrm{hh}}}$ and $\gamma_{X_{\mathrm{lh}}}$ are the heavy- and light-hole exciton linewidth (3 and 4 meV, respectively), and $V_{X_{\mathrm{hh}}}$ ($V_{X_{\mathrm{lh}}}$) is the interaction energy between the plasmon and the heavy-hole exciton (light-hole exciton). In the considered wave-vector range, both exciton energies can be regarded as dispersionless. The energy $E_{\mathrm{pl}}(k)$ is calculated using a conventional transfer-matrix method for the semiconductor structure described above with a 50 nm flat silver layer (mean thickness value of the corrugated layer). The plasmon broadening deduced from the calculations is 25 meV and is taken as homogeneous width of the plasmon. The inhomogeneous broadening comes from variations in the grating period and in the metal interface in the large sample surface studied in reflectometry experiments (5 mm$^2$). The wave vector $k_d$ of the SP first diffracted order is given by

$$k_d = k - \frac{2\pi}{a},$$

where $a$ is the silver film grating period. A period of 245 nm was used. The bare plasmon dispersion curve fits well the experimental data far from the resonance where the interaction with the exciton is negligible. Three $k$-dependent polariton energies are obtained. The grating first diffracted order of the calculated polaritons dispersion lines is shown in Fig. 4 (energies as a function of $k-2\pi/a$). The interaction energies $V_{X_{\mathrm{hh}}}=22\pm 2$ meV and $V_{X_{\mathrm{lh}}}=21\pm 2$ meV have been obtained by fitting the reflectometry dips energies, and the calculated curves are in good agreement with the experimental curves. This demonstrates that the reflectometry dips result from the mixing of plasmon and excitons and that a strong-coupling regime can be obtained for inorganic quantum wells in the vicinity of a metal film. The interaction energies are of the same order of magnitude than in semiconductor microcavities in strong-coupling regime.[2]

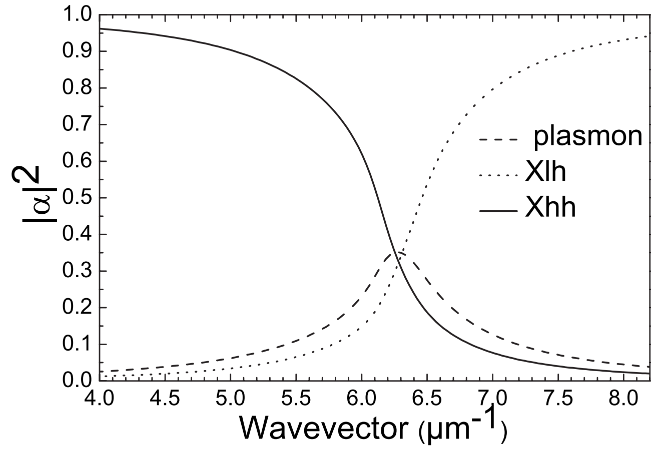


FIG. 5. Middle branch polariton wave-vector coefficient for the bare plasmon and excitons deduced from the coupled oscillator model.

The wave function of the polaritons formed is an admixture of bare plasmon heavy- and light-hole excitons with $|\phi\rangle = \alpha_p|\mathrm{plasmon}\rangle + \alpha_{\mathrm{hh}}|X_{\mathrm{hh}}\rangle + \alpha_{\mathrm{lh}}|X_{\mathrm{lh}}\rangle$. In order to quantify this mixing, the fraction of bare modes of the middle polariton line (open squares in Fig. 4) is shown in Fig. 5. A large mixing between $X_{\mathrm{hh}}$ and $X_{\mathrm{lh}}$ excitons is induced by the coupling to the SPs. A state with comparable weight of plasmon heavy- and light-hole excitons is obtained at $k=6.3$ $\mu$m$^{-1}$ ($|\alpha_p|^2=0.36$, $|\alpha_{\mathrm{lh}}|^2=|\alpha_{\mathrm{lh}}|^2=0.32$).

The three polaritonic energies are well reproduced by the coupled oscillator model, but not the quasidispersionless line at the bare heavy-hole exciton energy (open circles in Fig. 4). The coexistence of the signature of polaritons and incoherent states (states not coherently coupled to plasmons[23]) has already been observed for organic semiconductors near metal layers in luminescence experiments.[7] In semiconductor microcavities, in the strong-coupling regime, the incoherent luminescence may appear at the bare exciton energy with low reflectivity mirrors[24] but not with high reflectivity Bragg mirrors.[25] In our case, the dispersionless line can be related to transparency modes through the silver film since a transmission of a few percents is expected.

The signature of the absorption of the heavy-hole exciton and light-hole exciton can be seen by transparency at an angle of 29° when the plasmon is well separated from the exciton. The plasmon/exciton polaritons and the transparency modes are detected in the same direction but do not have the same wave vector. The transparency mode has the same in-plane wave vector as the detected light whereas the polariton is first-order diffracted. When the detection angle increases, the behavior for the heavy- and light-hole exciton differs: the incoherent line remains present during the anticrossing for the heavy-hole exciton but disappears for the light-hole exciton. This particular feature of plasmon in strong coupling can come from the polarization of the SP mode. Indeed, this surface wave only exists in TM polarization and the electric field is mainly transverse (perpendicular to the layer plane).[26] The transparency modes propagate in a direction quasiperpendicular to the layer plane (10° in the semiconductor for a detection angle of 40°) and the corresponding electric field is in this plane. Since optical transitions are forbidden for heavy-hole excitons when the field is perpendicular to the layer plane,[27] the interaction is reduced with the plasmon (only the longitudinal electric field interacts) but not with the transparency mode. Therefore the dip related to the incoherent states is still present during the anticrossing. For the light-hole exciton, on the opposite, the transition is allowed for the electric fields both perpendicular and parallel to the layer plane, i.e., for both longitudinal and transverse components of surface plasmon and for the incoherent mode. In this case the incoherent dip is masked when the plasmon anticrosses the exciton.

Finally it has to be noticed that an adequate grating is necessary to observe the plasmon/exciton polariton. When a semiconductor heterostructure is close to a flat metal surface, the polariton cannot couple to free-space radiative modes. The quantum well excitons can be strongly coupled to SP, inducing coupling between the different excitons and dynamic modification, but would not be detectable directly without modification of the upper metal interface.

In conclusion we have shown that the strong coupling between a surface plasmon and the excitons of GaAs/GaAlAs quantum wells occurs when the quantum wells are located in the vicinity of the metal surface. Polaritons are formed by the mixing of plasmon heavy- and light-hole exciton states. The interaction energies are 22 and 21 meV for the heavy- and light-hole excitons, respectively. Finally we observed some specific behaviors in the exciton-plasmon coupling related to the polarization of the electric field associated with surface plasmons.

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