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Luminescence of quantum-well exciton polaritons from microstructured $Al_xGa_{1-x}As$-GaAs multiple quantum wells

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Periodic multiple-quantum-well wires have been prepared by etching five-layer quantum-well structures through a holographically prepared mask. The periodicity was 380 nm, the lateral confinement 180 nm, and the quantum-well width 13 nm. The luminescence from these microstructured systems in the frequency regime of the one-electron–one-heavy-hole transition was strongly polarized with the electric field perpendicular to the periodic structure. This effect was caused by the resonantly enhanced emission of quantum-well-exciton (QWE) polaritons. Excitation of QWE polaritons was also observed in reflection measurements on the microstructured samples.

Currently, there is great interest in the fabrication and investigation of low-dimensional systems. Novel systems have been successfully realized by employing microlithographic techniques on two-dimensional (2D) and layered quantum-well structures. Luminescence measurements are a very useful technique to study these systems.[1-3] An unexpected highly efficient photoluminescence, which could not be explained by a confinement-induced increase of radiative recombination, was observed.[2] In very small microstructures, it was possible to detect a shift of the quantum-well-exciton energy due to a lateral 1D and 0D confinement.[3] The interest of our investigations here are quantum-well-exciton (QWE) polaritons. Because of the periodically modulated surfaces in these microstructured systems, coupling of QWE polaritons with photons becomes allowed and the strong-field enhancement,[4] which is inherent for these electrodynamic excitations, should lead to a significant influence on the optical spectra. We have prepared periodic quantum-well wire structures of periodicity $a=380$ nm and lateral confinement $w=180$ nm. For these dimensions, confinement effects on the electronic wave functions are expected to be small; thus, electrodynamic effects dominate. So far, QWE polaritons have only been treated theoretically, i.e., the dispersion has been calculated by Nakayama *et al.*[5,6] Luminescence of exciton polaritons, which propagate at the boundary of a semi-infinite solid with a statistical roughness, has been calculated in Ref. 7.

Let us briefly recall some aspects of electrodynamic surface excitations.[4] The dynamic dielectric response of excitons can be described by a Lorentzian oscillator model which is characterized, besides the oscillator strength and the damping constant, by a transversal (ω_T) and longitudinal (ω_L) frequency. We will neglect in the following the spatial dispersion of excitons which is not important for the small wave vectors considered here. Between ω_T and ω_L the dielectric function becomes negative which means that in this frequency regime at interfaces or in thin layers resonant electrodynamic excitations exist. In the following, we will refer to these excitations, if used in a general sense, as "surface" polaritons, in the special case of quantum wells as QWE polaritons. The dispersion of these modes is shown schematically in Fig. 1(a). We will discuss the dispersion of QWE polaritons in laterally microstructured systems in more detail below in the course of the interpretation of our experimental data. On smooth interfaces, the surface polaritons will not contribute to the luminescence since the polariton wave vector k_p is larger than ω/c. However, a lateral structure of periodicity a, i.e., a periodic wire structure, will lead to their radiative decay. The x component of the wave vector for the emitted photons, $k_x^{(-1)}$, is governed by the "grating coupler" condition $k_x^{(-1)}=(\omega/c)\sin\delta=k_p-n2\pi/a$, $n=\pm 1,2,\ldots$. [We consider polaritons of wave vector k_p propagating in the x direction; the wire structure is parallel to the y direc-

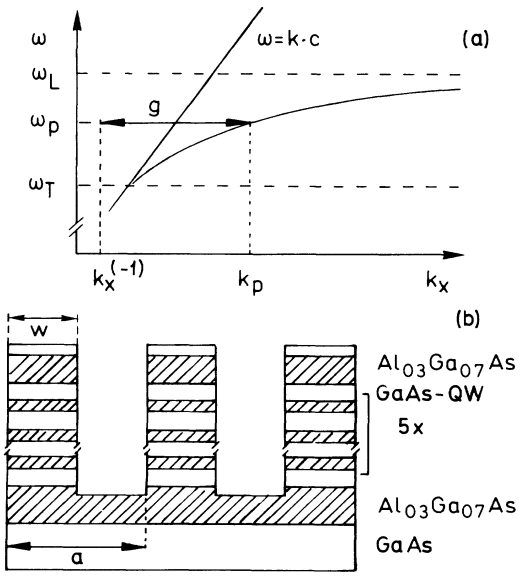

FIG. 1. (a) Schematic diagram of the QWE polariton dispersion. $g=2\pi/a$ is the reciprocal grating vector of the periodic microstructure that couples QWE polaritons of frequency ω_p and wave vector $k_p$with radiative photons of the same frequency and wave vector $k_x^{(-1)}<\omega/c$. (b) Schematic representation of a laterally microstructured multiple-quantum-well system.

tion. δ is the angle of emission with respect to the interface normal ($=z$ direction).] The interesting aspect of surface polaritons is that these excitations are associated with a largely enhanced field strength at the interface (or in the thin layer).[4] This leads to many interesting phenomena, e.g., the surface-enhanced Raman effect. For the QWE polaritons, we expect a resonantly enhanced photoluminescence intensity due to their radiative decay in the microstructured systems. A clear experimental indication of surface polaritons and the QWE polaritons is that these excitations are purely p polarized, i.e., for QWE polaritons propagating in x direction the electric field vector E is in the x-z plane. Thus, photons resulting from the radiative decay of surface polaritons have the same polarization. s polarization (E$\|y$) is only possible for direct exciton luminescence.

The samples were grown by molecular-beam epitaxy at 580°C employing the method of growth interruption at each interface. For the experiments described in detail here the samples had the following configuration: (100) GaAs substrates, 200-nm GaAs buffer layer, a short period GaAs/AlAs superlattice and a 50-nm $Al_{0.3}Ga_{0.7}As$ layer, five GaAs quantum wells of widths 13 nm, separated by 10-nm $Al_{0.3}Ga_{0.7}As$ barriers, and a cap layer consisting of 50-nm $Al_{0.3}Ga_{0.7}As$ and 2-nm GaAs. A mask consisting of periodic photoresist lines was prepared by holographic lithography. With reactive ion etching in a $SiCl_4$ plasma nearly rectangular grooves of depth 170 nm were etched all the way through the five multiple-quantum-well layers. The periodicity of the samples was $a=380$ nm, the lateral width w of the remaining multiple quantum-well structure was about 180 nm [see Fig. 1(b)]. Photoluminescence was excited with normally incident light at 647.1 nm from a Kr^+ laser. The luminescent light was collected normally to the sample in a solid angle of 30° opening. The spectral dependence of the luminescence was analyzed with a 1-m Jobin-Yvon monochromator and a photon-counting system. The resolution was set to 0.05 nm and the temperature was 4.2 K, if not otherwise stated.

The experimentally observed luminescence spectra are shown in Fig. 2. The spectrum (a) was obtained from an unstructured reference sample which was directly located near the microstructured sample on the wafer. Two peaks at 806.05 nm and at 807.05 nm and a shoulder at 807.5 nm are present. Applying the model of separated quantum wells with finite barriers, which is appropriate for our system, and boundary conditions according to Bastard,[8] we can unambiguously ascribe the two peaks to free-exciton transitions [one-electron–one-heavy-hole ($1e$-$1hh$)] in quantum wells with, respectively, m and $m+2$ monolayers ($m=46$). The very small half-width of the luminescence peaks (less than 0.5 nm) shows the excellent quality of our samples. There is also a small contribution of $(m+1)$-monolayer excitons hidden in the overlapping profile of the two luminescence peaks. This contribution was identified in excitation spectroscopy and reflection measurements on the same sample. The fact, that monolayer fluctuations could be resolved so clearly is the subject of further studies on exciton migration effects in photoluminescence which will be reported elsewhere. The

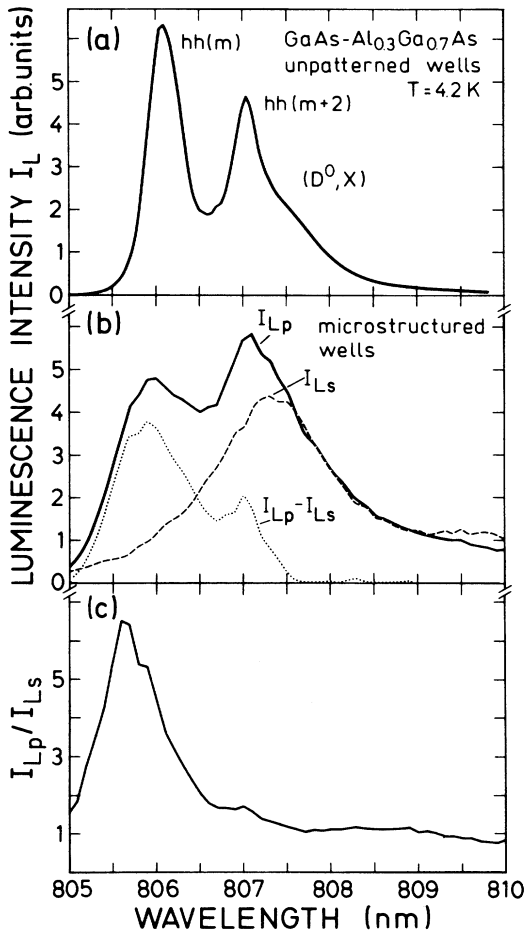

FIG. 2. Experimental luminescence spectra (a) of an unpatterned reference sample and (b) of microstructured quantum-well systems. I_{Lp} and I_{Ls} denote the p- and s-polarized luminescence intensity, respectively. The excitation intensity was about 10 mW/cm^2. (c) shows the ratio of I_{Lp}/I_{Ls}. The strongly enhanced p-polarized emission in the regime of free exciton transition energies is caused by the radiative decay of QWE polaritons.

shoulder at 807.5 nm is ascribed to a bound quantum-well exciton transition since the strength of this shoulder saturates with increasing excitation intensity and is quenched at higher temperatures. The energy separation to the 1hh $(m+2)$ transition of about 1 meV is typical for donor-bound excitons (D^0,X). Obviously, the (D^0,X) transition, which corresponds to the 1hh (m) transition, is too weak and cannot be resolved. We had polarizers both in the incident exciting beam (P_i) as well as in the emitted luminescent beam (P_e) and did not find any significant change in the photoluminescence intensity for different relative orientations of P_i with respect to P_e.

Figure 2(b) shows photoluminescence spectra measured on the microstructured sample. Exciting with the same intensity as in the case of the reference sample ($\sim$10 mW/cm^2), we see a total decrease of the luminescence efficiency of more than an order of magnitude. The s-polarized spectrum (dashed curve) consists mainly of the (D^0,X) transition which was also observed for the refer-

ence sample. The p-polarized spectrum shows an enhanced intensity with more structures. Temperature and excitation intensity-dependent measurements indicate that the peak at 807.1 nm consists predominantly of the (D^0,X) transition. The additional contribution at smaller wavelengths, has, like the peak at 806 nm, the temperature and intensity behavior of a free quantum-well exciton (see below). The strongly enhanced p polarization of the emission becomes apparent in Fig. 2(c) where we show the ratio I_{Lp}/I_{Ls}. It is important to note that this enhancement effect, which is not observed on samples without microstructures, does *not* depend on the polarization of the incident exciting radiation with respect to the wire structure.

We attribute this strongly p-polarized emission to the radiative decay of QWE polaritons. There are three arguments which confirm this interpretation. (1) Only QWE polaritons, which propagate in x direction perpendicular to the wire structure, can decay radiatively. Therefore, the additional QWE polariton luminescence can only be seen in the p-polarized spectrum, not in the s-polarized spectrum. (2) No polarization dependence is observed in the reference sample. (3) The additional photoluminescence is observed at energies about 3 meV higher than the free QWE photoluminescence. If one assumes that free QW-exciton transitions occur at the transversal frequency ω_T (which is actually known only for bulk GaAs so far),[9] then this energy position would be consistent with a polariton transition at $\hbar\omega > \hbar\omega_T$ [see Fig. 1(a)]. It should be noted that the energy position of the QWE polariton emission at frequencies $\omega > \omega_T$ indicates a higher value of the longitudinal-transverse splitting in quantum wells as compared to bulk GaAs (0.1 meV) of about 0.3 meV. This is not unexpected because of the enhanced oscillator strength in 2D confined systems.

The detailed features of the luminescence spectra on microstructured samples depends strongly on the fabrication process. For some samples, the free QWE luminescence is not so strongly reduced as in Fig. 2(b) and can be observed with a peaked intensity both in p and s polarization. The QWE polariton emission appears then for p polarization as a shoulder on the high-energy slope of the free-exciton transitions. For all these samples the free QWE transition shows only very small shifts of less than 0.1 nm with respect to the transitions of the reference samples. If we compare these energy shifts, with stress-induced shifts in Ref. 10, then we can conclude that a possible stress due to the microstructuring process is very small in our samples and *cannot* explain the strong polarization of the luminescence.

In detail, we interpret the spectra in Fig. 2(b) in the following way. The etching procedures and the lateral boundary interfaces of the microstructures create additional nonradiative decay channels which strongly reduce the direct free QWE luminescence of more than an order of magnitude. This leads in the s-polarized spectrum to a dominance of the extrinsic transition where the free QWE luminescence contributes only to the high-energy tail of the spectrum and cannot be resolved for this sample. For the p-polarized spectrum, in addition to the direct free QWE luminescence, which is assumed to be about the same as for the s-polarized emission, the radiative decay of QWE polaritons is possible. This leads to a strongly enhanced emission as compared with the direct QWE luminescence. In Fig. 2(b), we have also plotted the intensity difference $(I_{Lp}-I_{Ls})$ (dotted curve) of the p- and s-polarized luminescence spectra. This plot indicates the maxima of the additional photoluminescence which only occurs in the wavelength regime of the free QWE both at 805.9 nm for the m-monolayer exciton and, more weakly for the $(m+2)$-monolayer exciton at 807 nm. The bound exciton luminescence is not so strongly reduced as compared with the smooth reference sample. These excitons, which are localized at the impurity centers, cannot migrate to the etching-induced nonradiative decay centers, which are assumed to be located dominantly near the lateral interfaces.

The enhancement of the QWE polariton luminescence varies for different samples or for different spots on the same sample if the etching is not perfectly homogeneous. We attribute these variations to the very sensitive balance of "radiative" and nonradiative damping processes which are responsible for the grating coupler efficiency of surface polaritons. For the radiative decay of surface-plasmon polaritons on corrugated films which are, concerning electrodynamics, a very similar system, it was found[11] that for small modulation amplitudes H of the surface the emission from polaritons increases with increasing H. For a certain, "optimum" value of H one achieves a maximum emission rate. Then, for still larger modulations, the emission intensity *decreases*. The reason for the latter is that the emission of photons is an additional, i.e., "radiative" damping process which diminishes the resonant character, and thus the field-strength enhancement of the excitation. The optimized grating-coupler efficiency depends in a very sensitive way on the ratio of the "radiative" damping, which is induced by the grating coupler, and "internal" damping processes, which in the case of QWE polaritons depend predominantly on nonradiative recombination of excitons. We thus expect that the contribution of QWE polariton emission in the optical spectra depends strongly on the profile of the microstructures and the quality of the quantum well. The maximum polarization ratio of more than 6, which is observed in the spectrum of Fig. 2(c), is about the highest that we have found so far for our samples.

In Fig. 3, we show optical reflection measurements on the same microstructured samples. Light from a halogen lamp was monochromized by a 60-cm double monochromator and directed with 45° angle of incidence onto the sample. The wire structure was perpendicular to the plane of incidence. Radiation is called p-polarized if the electric field vector is parallel to the plane of incidence. The spectra in Fig. 3 show some broad features in the regime of the 1e-1lh (light hole) transition and the 1e-1hh transition at about 803 and 807 nm, respectively. These structures are residual "interference" effects due to the multilayer quantum-well system with different dielectric functions for the GaAs and the $Al_xGa_{1-x}As$ layers. This can be calculated using the Fresnel formulas for the multilayered system. Because of the non-normal incidence these spectra are different for p and s polarizations. Only

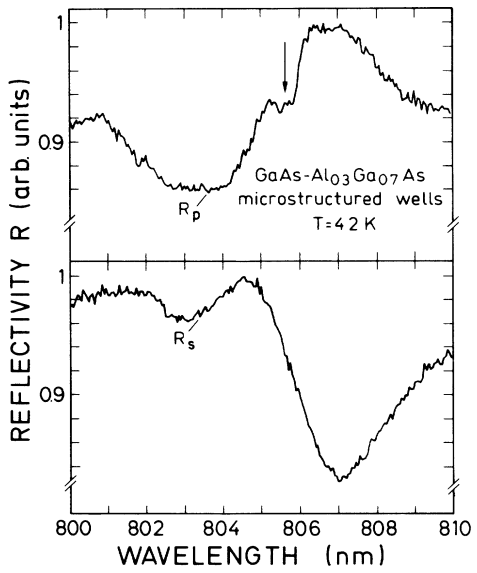

FIG. 3. 45° p- and s-polarized reflectivity (R_p and R_s, respectively) on wire structures. In the p-polarized spectrum grating coupler induced excitation of QWE polaritons is observed as a dip (see arrow) in the reflection.

for the p-polarized reflected light we observe at 805.7 nm a relatively sharp small dip (see arrow) which is not present in s polarization or for any polarization on unstructured reference samples. The position of this dip corresponds very well to the position of the QWE polariton emission in Fig. 2(b). We explain this dip in the following way. At wavelengths of about 806-nm p-polarized incident light can couple via the grating coupler effect of the microstructure to QWE polaritions, leading to a decrease of reflectivity.[4,11] The grating-coupler-induced QWE polariton excitation is the reverse process to the emission process described above. If we take the relative depth ($\Delta R/R = 3\%$) of the reflection minimum at 805.7 nm in our spectrum as a measure for the coupling efficiency of QWE polaritons here then this quantity is comparable with the efficiency that has been achieved for the excitation of surface exciton polaritons on semi-infinite ZnO crystals ($\Delta R/R = 2$–7%).[12]

Nakayama *et al.*[5,6] have calculated QWE polaritons in a laterally homogeneous quantum-well system in the limit of a quantum-well width which is small compared with the light wavelength in GaAs ($\lambda_{GaAs} = 2\pi c/\omega_T\sqrt{\epsilon_{GaAs}}$). The dispersion of QWE polaritons in microstructured multilayer quantum wells is actually not known so far. Additional effects as the coupling of the five quantum wells via Coulomb interaction (similar as for the case of 2D plasmons in layered system)[13] and lateral superlattice effects are expected to influence the details of the dispersion, the field enhancement, and the emission intensity. They will not, however, change the polarization selection rules which we have predominantly employed to identify the QWE polariton emission.

From the grating coupler condition $k_p = 2\pi/a + \omega\sin\delta/c$ (see above) we can, in principle, determine the dispersion $\omega_p(k_p)$ of QWE polaritons by scanning ω and the angle of emission δ. However, the dispersion is concentrated between ω_T and ω_L and the LT splitting is expected to be very small (about 0.3 meV, see above). Thus, the LT splitting is smaller than the linewidth of the exciton transitions and no details of the dispersion can be extracted so far.

In conclusion, we have demonstrated photoluminescence emission of quantum-well-exciton polaritons and their excitation in reflection on laterally microstructured quantum-well systems. The emission depends strongly on the profile and quality of the microstructured quantum wells and has to be considered as an essential contribution to the optical response in microstructured systems.

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