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Reflectance of two-dimensional excitons in GaAs-AlGaAs quantum wells

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We report reflectance, excitation, and luminescence spectra of GaAs-AlGaAs quantum wells in the energy region of the light- and heavy-hole exciton transitions. The properties of the two-dimensional exciton are analyzed by using a dielectric model. The excellent agreement between experimental and theoretical reflectance spectra leads to an accurate determination of the excitonic eigenenergies, oscillator strength, and damping. The energy positions of the emission peaks of the excitation spectra are close to the eigenenergy as determined from reflectance, whereas the emission maxima of the luminescence spectra exhibit red shifts depending on the excitation intensity.

The optical properties of ultrathin semiconductor films strongly depend on their thickness. The energy states of electrons and holes (subbands) as well as of excitons formed by each subband become discrete in the direction perpendicular to the layer because of quantization due to the small dimensions. Experimentally, two-dimensional (2D) excitons have been investigated extensively in GaAs-AlGaAs quantum-well heterostructures by means of transmission, excitation, and luminescence spectroscopy.[1-8] The binding energy as well as the one-particle energy were found to depend on the well width L_z and the depth of the quantum well given by the Al content.[1,5-8] The experimental spectra so far have been analyzed on the basis of the energy positions of the respective peaks. However, a detailed comparison of experimental and theoretical spectra has not yet been attained. Reflectance experiments were commonly used to determine the excitonic parameters of three-dimensional (3D) excitons in bulk GaAs.[9,10]

In this Rapid Communication we report the first reflectance spectra of 2D excitons in GaAs-AlGaAs quantum wells. We compare the experimental data with theoretical spectra based on a dielectric model. The fitting procedure directly yields the excitonic eigen energies. In addition, we analyze the energy positions of the emission peaks of excitation and luminescence spectra observed in the vicinity of the heavy-hole (hh) and light-hole (lh) exciton transitions.

The experiments are performed on two different GaAs-AlGaAs quantum-well heterostructures grown by molecular beam epitaxy (MBE) on (100) oriented GaAs substrates. The well widths of the double (DQW) and single (SQW) quantum-well heterostructures are 14 and 5 nm, respectively. The Al content is 21% for the DQW and 18% for the SQW. The nominally undoped samples exhibit p-type conductivity in the high 10^{14}-cm^{-3} range due to residual C acceptors. Details of the MBE growth conditions have been reported elsewhere.[8] The samples are immersed in liquid He pumped below the λ point for the optical experiments. A tungsten iodine lamp is used for the reflectance experiments. The photoluminescence is excited with the 647.1-nm line of a Kr^+ laser. Excitation spectra are obtained with a tunable cw dye laser system (oxazine 750). The reflected or emitted light passes a double grating monochromator with a spectral resolution of 0.02 nm and is detected with a cooled photomultiplier (GaAs cathode). The spectra are recorded with a single photon counting system and digital data acquisition. A factor of 1 239.852 eV nm is used to convert energy to wavelength.[11] The refractive index of air is also included.[12]

The optical properties of a semiconductor with two excitonic resonances ($\pm$) and eigenfrequencies $\omega_\pm$ showing spatial dispersion are given for a wave vector $\vec{k}$ and frequency ω by a dielectric function $\epsilon(\omega,k)$:[13]

$$\epsilon(\omega,k)=\epsilon_\infty+\frac{1}{\epsilon_0}\frac{\alpha_+\omega_+^2}{[\omega_+ +(\hbar/2M_+)k^2]^2-\omega^2-i\omega\Gamma_+}+\frac{1}{\epsilon_0}\frac{\alpha_-\omega_-^2}{[\omega_- +(\hbar/2M_-)k^2]^2-\omega^2-i\omega\Gamma_-}\ , \quad (1)$$

where $\alpha_\pm$ are the polarizabilities of each excitonic resonance at $\omega=0$ and $k=0$; $\Gamma_\pm$ are the empirical damping constants ($i=\sqrt{-1}$); $M_\pm$ are the masses of the excitons; ϵ_0 is the permittivity of the vacuum, ϵ_∞ the background dielectric constant, and $\hbar$ Planck's constant divided by 2π. We describe the excitonic resonances in quantum wells by applying Eq. (1) to a layer of the quantum well. Spatial dispersion has to be considered only for a wave-vector component $k_\parallel$ within the direction of the quantum-well plane.

Within this model we thus obtain an effective excitonic polarizability which is correlated with the microscopic nonhomogeneous oscillator strength by averaging over the spatial extent of the exciton. The excitonic wave functions are mainly confined to the well for L_z larger than 5 nm. The effective polarizability of Eq. (1) is thus smaller than given by the actual oscillator strength.

The reflectance spectra are calculated for the multilayer system consisting of the GaAs quantum wells and the Al-GaAs layers by matching the amplitudes of the plane waves propagating in each layer at the boundaries according to Maxwell's equations. Details about the sample geometry are depicted in the left part of Fig. 1. The refractive index of the GaAs substrate and of the AlGaAs layer are taken to $n_{\mathrm{sub}}=3.64+i\times0.08$ and $n_{\mathrm{AlGaAs}}=3.46$, respectively.[14] The background dielectric constant is $\epsilon_\infty=12.6$.[10]

The reflectance spectra are measured under normal incidence with respect to the quantum well. In Fig. 1(a) the results for the DQW with 14-nm well width are shown. The spectrum exhibits two reflectance dips of different strengths in the vicinity of the hh and lh subband transitions. The reflectance spectrum of the SQW with a well width of 5 nm is shown in Fig. 1(b). The observed weak (2%) reflectance dip is located at 1.6135 eV. A second dip due to lh transition is not observed because of the low signal-to-noise ratio.

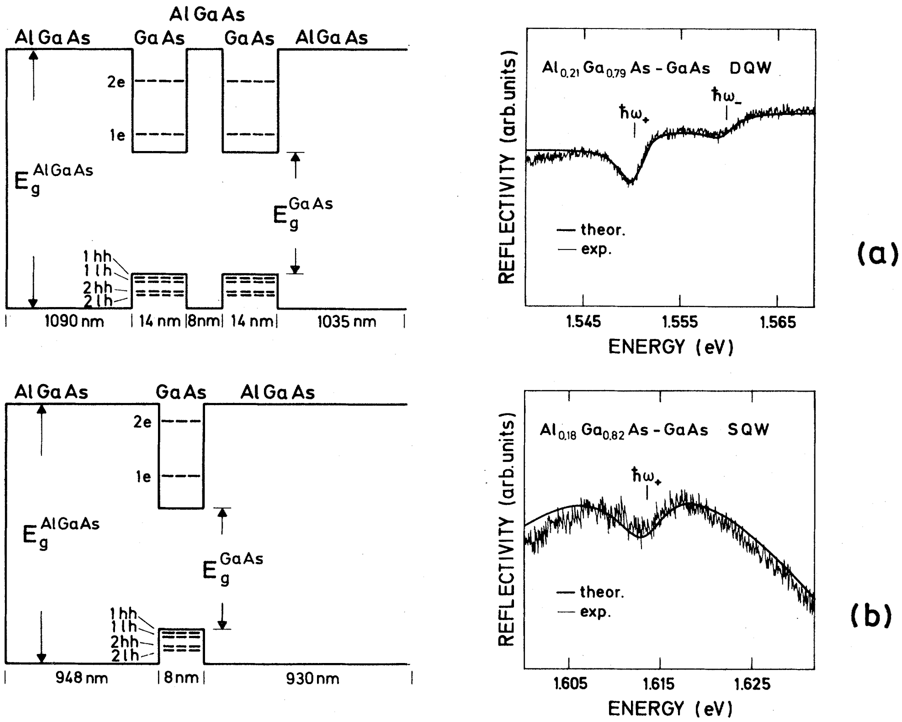

FIG. 1. Right: experimental (weak line) and theoretical (strong line) normal incidence reflectance spectra of GaAs-AlGaAs (a) DQW and (b) SQW . The eigenfrequencies $\omega_{\pm}$ obtained from the fit are listed in Table I. Left: schematic real-space energy-band diagram for the DQW and the SQW including the energy levels of the first ($n=1,2$) hh and lh hole subbands as well as electron (e) subbands of the quantum wells.

The reflectance structure of Fig. 1 is similar to the one observed in high-purity GaAs except for the modified energy position, which indicates excitonic transitions within the quantum well.[10]

We have calculated reflectance spectra with the model described by Eq. (1). The results are included in Fig. 1. The agreement with the experimental data is very good. The excitonic parameters of the 2D exciton obtained from the fits are listed in Table I. The actual thicknesses of the AlGaAs layers are fitted in order to reproduce the background slope of the reflectance spectra caused by interference (see Fig. 1). The eigenenergies obtained from the fit are 1.5503 and 1.5598 eV for the hh and lh transitions of the DQW and 1.6135 eV for the hh transition of the SQW. Inspection of Fig. 1(a) reveals that the reflectance minimum does not coincide with the resonance energy. The uncertainty in the determination of the absolute value of the resonance energy depends on the sharpness of the reflectance amplitude and is about 0.03 meV for the hh exciton transition of the DQW.

The polarizability α is a direct measure of the exciton-photon coupling and is given by the oscillator strength. The polarizabilities obtained from our fit are higher than the value of $2.1\times10^{-3}\epsilon_0$ obtained for the 3D exciton in GaAs.[9,10] The data of Table I indicate that the values for the oscillator strength of the 2D excitons increase with decreasing well width. This is indeed expected because the exciton Bohr radius shrinks with decreasing L_z, giving rise to an enhanced oscillator strength.[15] For a purely 2D exciton the oscillator strength is expected to be eight times the 3D value.

The damping constants obtained from the fits are larger by a factor of 50 than in bulk GaAs (Ref. 9) and also increase with decreasing well width. This indicates an addi-

TABLE I. Excitonic parameters of the double (DQW) and single (SQW) quantum wells obtained from fit to reflectance spectra (A) and from the emission peaks of excitation spectra (B).

	A		B	
	DQW	SQW	DQW	SQW
ω_+ (eV)	1.5503	1.6135	1.5507[a]	1.6136[a]
ω_- (eV)	1.5598	...	1.5610[a]	1.6323[a]
Γ_+ (meV)	3.0	7.0	3.0[b]	6.5[b]
Γ_- (meV)	4.0	...	3.6[b]	6.5[b]
$\frac{\alpha_+}{\epsilon_0}$ (10^{-3})	4.8	7.8	...	...
$\frac{\alpha_-}{\epsilon_0}$ (10^{-3})	2.4	...	...	...

[a]The experimental uncertainty is about 0.2 meV. [b]Determined from FWHM.

tional scattering mechanism which reduces the excitonic lifetime. Fluctuations of L_z are considered to cause inhomogeneous broadening of excitonic transitions observed in luminescence.[2] Additionally, an enhanced scattering rate due to fluctuations in L_z may also account for the increased damping with decreasing L_z. The higher damping of the lh exciton compared to the hh exciton may be explained by two mechanisms. First, the lh exciton is affected more by the fluctuations of L_z because of its larger spatial extent. Second, since the binding energy of the hh exciton exceeds the splitting of the two-hole subbands for a well width lower than $L_z = 15$ nm,[16,17] the lh exciton states are in resonance with the continuum states of the hh excitons, thus enabling an additional decay.

The excitation spectra obtained from the DQW and the SQW are depicted in Fig. 2. Detection was at the low-energy tail of the luminescence: 1.5378 eV for the DWQ and 1.569 eV for the SQW. The spectra exhibit two well resolved emission peaks attributed to the lh and hh exciton transitions.[4,6,7] The energy position and the full-width at half maximum (FWHM) of these peaks are also listed in Table I. The peak energies are close to the eigenenergies as obtained from the reflectance measurements. The excitation spectra in the excitonic energy region are thus mainly determined by the absorption strength rather than by a change of the efficiency of competing recombination channels. For the DQW an additional weak band is observed in 1.611 eV which can be attributed to the $n = 2$ hh exciton transition. The increase of the excitation spectra above 1.65 eV is caused by excitation of (e,C^0) transitions in the AlGaAs layers.[8,18]

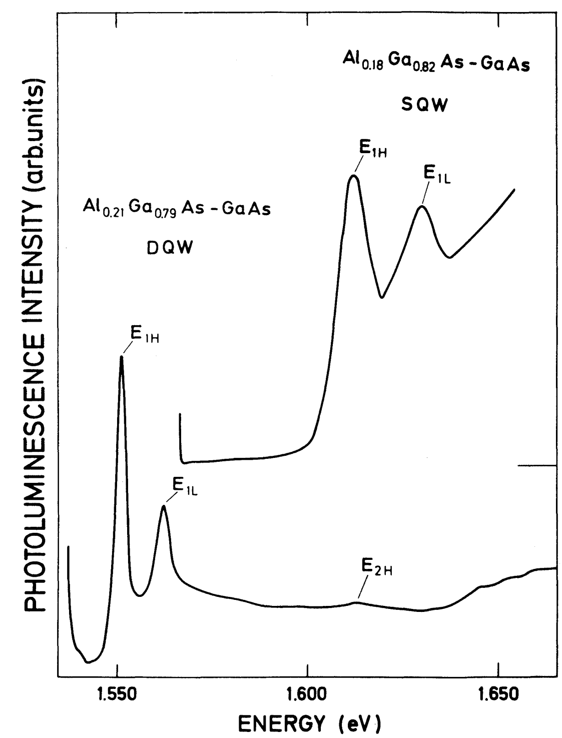

FIG. 2. Excitation spectra of the DQW and the SQW showing the $n = 1$ heavy- (E_{1h}) and light-hole (E_{1l}) exciton transitions. The spectrum of the DQW also exhibits the weak $n = 2$ heavy-hole (E_{2h}) exciton transition. Detection was at a photon energy of 1.5378 eV for the DQW and 1.569 eV for the SQW.

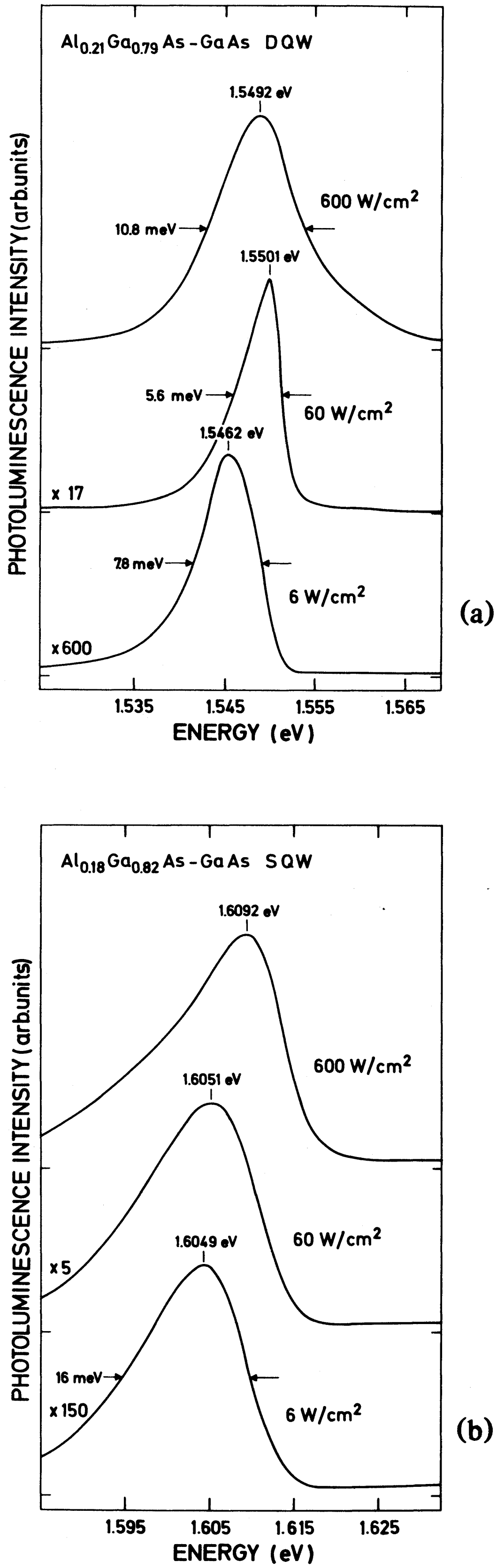

FIG. 3. Luminescence spectra of the (a) DQW and the (b) SQW at three excitation intensities.

In Fig. 3(a) we show luminescence spectra of the DQW for various excitation intensities. A broad luminescence line with a FWHM of about 7.8 meV is observed at a low excitation intensity of 6 W/cm^2. The maximum of the emission is about 4.1 meV *below* the resonance energy of the hh exciton. An increase of the excitation level first causes a blue shift of the luminescence peak together with a decrease of the linewidth (5.6 meV). The luminescence peak at 1.5501 eV is only slightly below the hh exciton resonance energy. For an excitation intensity of 60 W/cm^2 emission of the lh exciton is observed (which is seen more clearly in Fig. 4). The emission line broadens again (10.8 meV) at the highest excitation intensity of 600 W/cm^2 and the luminescence maximum shifts slightly to lower energies. Similar results are obtained for the SQW. The energy separation between the luminescence maximum and the resonance energy is about 8.5 meV at 6 W/cm^2. Even at the highest excitation intensity, the peak energy is still 4 meV below the hh exciton transition. The FWHM remains nearly constant at 16 meV.

The evaluation of the luminescence line shape with increasing excitation intensity is more clearly demonstrated in a semilogarithmic plot, as shown for the DQW in Fig. 4. Intrinsic free exciton emission can occur only in the vicinity of the excitonic resonance within a spectral range given approximately by $kT_{\rm exc}$, where $T_{\rm exc}$ is the effective exciton temperature.[19] Previous luminescence investigations indicated that emission due to bound excitons may occur at the low-energy side of the excitonic resonance.[20] This interpretation is supported by the observed saturation of the low-energy emission band and the resulting blue shift of the luminescence maximum. The luminescence band below the eigenenergy of the hh exciton is extrinsic. The origin of the bound exciton emission might be due to residual C acceptors,[18] but also a binding of free excitons to the potential caused by the well width fluctuations has to be considered.[21]

Inspection of the spectra obtained at excitation intensities of 170 and 600 W/cm^2 in Fig. 4 reveals that the excitonic luminescence has nearly an exponential high-energy tail starting at the energy of the hh continuum states. The continuous density of states of the excitonic continuum is populated according to a Boltzmann distribution at quasithermal equilibrium. From the nearly straight slopes of the excitonic emission at 170 and 600 W/cm^2 we obtain excitonic temperatures of 38 and 50 K, respectively.

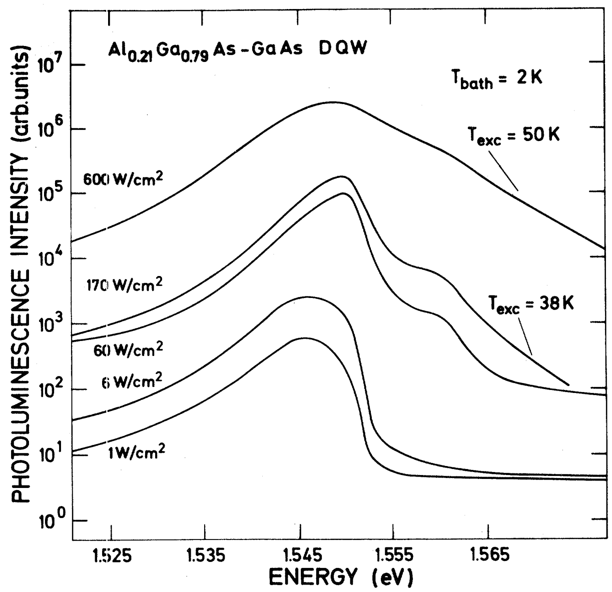

FIG. 4. Luminescence spectra of the DQW in a semilogarithmic plot for five excitation levels. The excitonic temperature $T_{\rm exc}$ is determined from the high-energy slope.

We conclude that we have resonantly excited 2D excitons in quantum wells by means of reflectance experiments. A dielectric model is applied to analyze the experimental data. The agreement with experiment is excellent, resulting in an accurate determination of the excitonic eigenenergies. The results of the reflectance measurement are compared to excitation and luminescence spectra. The resonance energies obtained by reflectance coincide with the emission peaks of the excitation spectra. The luminescence, however, is strongly affected by extrinsic emission particularly at a lower excitation intensity.

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