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Absorption spectroscopy on Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As multi-quantum-well heterostructures.

I. Excitonic transitions

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The physical properties of excitons (linewidth, binding energy, oscillator strength, reduced effective mass) in Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As multi-quantum-well heterostructures (MQWH's) as a function of the well width and thus of the quasi-two-dimensional (2D) character of the respective MQWH were investigated by absorption and magnetoabsorption experiments. Samples lattice matched to InP with well widths ranging from 2.3 to 13.8 nm were grown by molecular-beam epitaxy. Double-crystal x-ray diffraction was used to determine the structural perfection and the individual layer thicknesses of the MQWH. The intrinsic statistical composition fluctuation of the ternary Ga$_{0.47}$In$_{0.53}$As well material is found to be the dominant contribution to the linewidth of the excitonic transition. With decreasing well width the binding energy of the electron–heavy-hole exciton increases up to 9.2 meV for a well width of 3.4 nm. Similarly we observe a strong increase of the excitonic oscillator strength and of the reduced effective mass. A quasi-2D carrier system in this MQWH is realized for well widths of 3 to 5 nm.

I. INTRODUCTION

Quantum-well heterostructures (QWH's) and superlattices (SL's) composed of III-V semiconductors have been studied in recent years both for their possible application in advanced photonic devices and for the fundamental physical properties of quasi-two-dimensional (2D) carrier systems. QWH's based on the Ga$_{0.47}$In$_{0.53}$As/InP and the Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As material systems emit in the near-infrared region at wavelengths around 1.55 and 1.3 µm, which is the optimal wavelength range for telecommunication systems based on silica fibers. The composition of the ternary materials are chosen in the way that they are lattice matched to the InP substrate material. In this paper we will focus on the Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As QWH system, which is up to now almost exclusively grown by molecular-beam epitaxy (MBE). The recent improvements in material quality, which now approaches the quality of the most commonly studied GaAs/Al$_x$Ga$_{1-x}$As QWH system, offer the possibility to investigate the intrinsic electronic properties of these structures. Excitonic transitions in absorption measurements have been reported at room temperature and even up to 500 K (Ref. 4) for the Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As material system. The high structural perfection of Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As multi-quantum-well heterostructures (MQWH's) with respect to chemical homogeneity, interface quality, and thickness fluctuations has been demonstrated by double-crystal x-ray diffraction measurements. The first direct evidence of excitonic recombination has been found in temperature-dependent photoluminescence and photoluminescence excitation spectroscopy experiments.

As yet, important physical parameters including the exciton binding energy, the exciton oscillator strength, or the effective carrier masses as a function of well width and thus of the quasi-2D character of Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As QWH's have not been determined. These three parameters are interrelated to each other because a change in the carrier effective masses influences the binding energy and also the oscillator strength of the excitonic transition. For the electron–heavy-hole binding energy $B_{1h}$ different estimates of $B_{1h} = 5.7$ meV ($L_z = 10$ nm) (Ref. 1) and of $B_{1h} = 16$ meV ($L_z = 8$ nm) (Ref. 4) have been published. The exciton oscillator strength is not known up to now for the Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As material system. The knowledge of this parameter is important for a possible application of these QWH's in devices, which make use of the unique properties of quasi-2D excitons. The quantum confined Stark effect and its device applications as demonstrated in the GaAs/Al$_x$Ga$_{1-x}$As system can be stated as an example.

We present the results of absorption and magnetoabsorption experiments on Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH's with well widths ranging from 13.8 to 2.3 nm. The evaluation of these experiments is divided into two parts. In this paper on excitonic transitions, the physical properties of excitons in this QWH system, i.e., linewidth, binding energy, oscillator strength, and reduced effective
masses, are determined. As structural parameters like the well width, the chemical homogeneity of the ternary layers, and fluctuations of these quantities strongly influence the experimental results, we also give the results of the structural characterization of our MQWH’s by double-crystal X-ray diffraction experiments. In the companion article[2] to this work on the subband structure of Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As MQWH’s, the experimentally determined subband spacings are compared to theoretical calculations using the envelope-function approximation (EFA). The influence of the input parameters in the theory, i.e., band offsets and carrier effective masses, are discussed.

This paper is organized as follows. In Sec. II we present the growth conditions of the Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As MQWH’s by molecular-beam epitaxy (MBE), as well as the experimental conditions for the double-crystal x-ray diffraction and the absorption and magnetooabsorption measurements. The structural properties of the MQWH’s with respect to chemical and thickness homogeneity and interface quality and the determination of well and barrier width obtained by evaluation of double-crystal x-ray diffraction curves are given in Sec. III. The properties of excitonic transitions in the Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As MQWH as a function of the well width and thus of the quasi-2D character are studied in Sec. IV by use of absorption and magnetooabsorption experiments. Section V summarizes our results on excitonic transitions in Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As MQWH’s.

II. EXPERIMENT

The growth of the Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As MQWH’s by MBE was performed in a three-chamber ultrahigh-vacuum (UHV) system for separate sample loading, preparation, and growth. This system is equipped with an azimuthally rotating substrate holder. The (100) InP substrates were etched in (3:1:1) [H_{2}SO_{4}]:[H_{3}O_{2}]:[H_{2}O] and in 0.3% Br_{2}+CH_{3}OH solutions according to a procedure described by Nishitani and Kotani.[4] The samples were grown at a substrate temperature of 570–580°C under As-stabilized surface reconstruction. The reference temperature of 500°C was given by the desorption of the InP oxide. The layer structure consisted of a periodic sequence of Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As double layers, which were grown without any buffer layer directly lattice matched on InP. The layer thicknesses of the different samples are summarized in Table I (see Sec. III). Special care was taken to keep inhomogeneities in the ternary epitaxial layers as low as possible. The individual group-III element effusion cells were equipped with stable and fine-scaled temperature regulations, which allow us to control the ternary epitaxial layer composition precisely. The substrate rotation speed was synchronized to the shutter movement, i.e., an integer number of rotations per growth time for each constituent layer, and adjusted to the chosen growth rate of 1 μm/h (see also Ref. 9). In this way lateral thickness fluctuations and periodic composition changes in growth direction of the constituent ternary Ga_{0.47}In_{0.53}As and Al_{0.48}In_{0.52}As compounds are minimized.

The x-ray diffraction measurements were performed with a computer-controlled double-crystal diffractometer of high angular resolution. As first monochromator and collimator for the Cu Kα_{1} radiation an asymmetrically cut Ge(100) crystal was applied.\textsuperscript{10} Part of the absorption experiments without magnetic field were made in a Cary-17D apparatus with a large area focus of 4 × 8 mm² at sample temperatures of 5 and 300 K. The absorption measurements with magnetic field were performed in a superconducting, split magnet coil at magnetic fields up to 10 T at a sample temperature of 2 K. The light of a spectral lamp, dispersed by a 0.5-m grating monochromator and transmitted through the sample, was detected by a liquid-nitrogen-cooled PbS detector.

The absorption coefficient was determined by comparing the transmitted light intensity from a sample with a Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As MQWH (I_{1}) and without the MQWH epilayer (I_{2}). By definition of the transmission coefficient we obtain the following relation between these two quantities:

\begin{equation}
\frac{I_{2}}{I_{1}} = \frac{(1 - R^2)}{1 - R^2 \exp(-2ad)} = A \exp(-ad),
\end{equation}

with R being the reflection coefficient (0.36), \(\alpha\) the absorption coefficient (\(10^4\ cm^{-1}\)), and d the total thickness of the absorbing well layers (0.5 μm). The prefactor A in Eq. (1) ranges between 0.9 and 1 for the present typical values, given within parentheses, and is approximated by unity in the following. Thus the absorption coefficient takes the form

\begin{equation}
\alpha = \ln(I_{2}/I_{1})/d.
\end{equation}

III. STRUCTURAL PROPERTIES

OF Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As MQWH’s

Because of the great differences in the physical properties (lattice constant, band gap, etc.) of InAs and GaAs (AlAs), the structural properties of the all-ternary Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As MQWH’s which are lattice matched to InP for the given compositions, are far more important for the optical properties of this MQWH sys-

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TABLE I. Structural parameters of the investigated Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As MQWH’s.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Periods</th>
<th>(L_{z}) (nm)</th>
<th>(L_{0}) (nm)</th>
<th>d (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>200</td>
<td>2.3</td>
<td>11.4</td>
<td>460</td>
</tr>
<tr>
<td>2</td>
<td>150</td>
<td>3.4</td>
<td>11.4</td>
<td>510</td>
</tr>
<tr>
<td>3</td>
<td>50</td>
<td>4.8</td>
<td>21.7</td>
<td>240</td>
</tr>
<tr>
<td>4</td>
<td>70</td>
<td>8.0</td>
<td>11.4</td>
<td>560</td>
</tr>
<tr>
<td>5</td>
<td>50</td>
<td>10.3</td>
<td>10.3</td>
<td>515</td>
</tr>
<tr>
<td>6</td>
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<td>50</td>
<td>11.2</td>
<td>11.2</td>
<td>560</td>
</tr>
<tr>
<td>10</td>
<td>50</td>
<td>13.8</td>
<td>11.5</td>
<td>690</td>
</tr>
</tbody>
</table>
tem than they are in the case of the nearly-lattice-matched GaAs/Al$_{x}$Ga$_{1-x}$As material system. Thus, knowledge of the structural parameters such as the lattice mismatch, interface quality and layer thicknesses, and the fluctuations of these quantities is an important prerequisite for the determination of the intrinsic properties of excitonic transitions and the subband structure in the Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH system as a function of the well width and thus of the 2D character of this MQWH system.

In Fig. 1 we present the x-ray diffraction pattern of a 50-period Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH taken in the vicinity of the (400) reflection of InP on a semilogarithmic scale. In this all-ternary MQWH system the precise strain distribution in the epilayer is not a priori known, but varies strongly with the composition of the ternary layers. Using the symmetric (400) reflection we can monitor the change of the lattice constant perpendicular to the (100) substrate surface, which is most sensitive to the lattice mismatch between epilayer and substrate. The average lattice mismatch of the MQWH to the InP substrate is obtained from the angular distance between the substrate and the main epilayer peak by applying Bragg's equation. For the sample in Fig. 1 the substrate and the main epilayer peak coincide. Therefore we can conclude that the average lattice mismatch within the epitaxial layer and the substrate is zero for this sample. For the samples investigated here the lattice mismatch to the InP substrate is in the range of $10^{-3}$.

The angular separation $\Delta \theta^{\pm \pi}$ between the main epitaxial peak and the satellite peaks $(\pm n)$ determines the period length $T$ of the respective MQWH (Refs. 3 and 10) according to

$$T = \frac{n \lambda}{2 \Delta \theta^{\pm \pi} \cos \theta_B},$$

where $\lambda$ is the x-ray wavelength and $\theta_B$ the kinematic Bragg angle. Any fluctuation of either the period length or the chemical composition within the epitaxial MQWH should lead to a broadening of the satellite peaks. In contrast, the main epilayer peak reflects the integral structure.

al quality of the epitaxial layer and is less sensitive to the thickness and composition inhomogeneities of the individual Ga$_{0.47}$In$_{0.53}$As and Al$_{0.48}$In$_{0.52}$As layers. Typical linewidths of the main epitaxial peak are in the range of 15–25 sec full width at half maximum (FWHM), depending on the total epitaxial layer thickness. From the sharpness of the satellite peaks the fluctuations of the period length can be determined to be less than two monolayers.

The evaluation of the individual well and barrier thicknesses from the MQWH period length requires the knowledge of the growth rates of the two ternary materials. All samples in this study were grown with the one In diffusion cell kept at a constant temperature for both the Ga$_{0.47}$In$_{0.53}$As and Al$_{0.48}$In$_{0.52}$As epitaxial layers. This ensures that the growth rate of the two ternary materials, which have the same lattice constant if lattice matched to InP, are equal. This expected behavior has been verified by earlier measurements of the epitaxial layer thicknesses by transmission electron microscopy. The individual well and barrier thicknesses can thus be directly deduced from the period length and the respective growth time with a precision of about one to two monolayers. The structural parameters of the investigated samples, which were evaluated in the described way, are summarized in Table I, where also the total thickness $d$ of the absorbing Ga$_{0.47}$In$_{0.53}$As well layers is given.

**IV. EXCITONIC TRANSITIONS IN Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH**

The absorption spectra as a function of the photon energy are shown in Fig. 2 for three Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH's of different well widths. The steplike behavior of the absorption coefficient and thus of the density of states in these quasi-2D structures is clearly observed. The dependence of the different subband transitions, which are indicated by arrows in Fig. 2, as a function of the well width will be discussed in the second part of this work. In the present section we will focus on the strong resonances at the subband edges, which are assigned to discrete excitonic transitions using the results of magnetoabsorption experiments. The aim of this section is the determination of the physical properties of these excitonic transitions (linewidth, binding energy, and oscillator strength) as a function of the well width in the Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH system.

**A. Magnetoabsorption**

The application of a magnetic field in the $z$ direction perpendicular to the well layers leads to a complete quantization of the electron and hole subband levels in Landau levels. Assuming a parabolic subband dispersion in the $k_x$ and $k_y$ directions, the allowed energy levels of the first electron subband take, for example, the form

$$E_N = E_1 + (N + \frac{1}{2}) \hbar \omega_c, \quad N = 0, 1, \ldots$$

with $E_1$ being the energy eigenvalue caused by the quantization in the $z$ direction in the well, $\hbar$ Planck's constant divided by $2\pi$, and $\omega_c$ the cyclotron-resonance frequency.
The allowed optical transitions ($\Delta N = 0$) between the electron and the heavy-hole Landau levels have a transition energy $\Delta E_N$ of

$$\Delta E_N = E_{1h} + (N + \frac{1}{2}) \hbar \omega \mu_{1h}, \quad (5)$$

with $\mu_{1h}$ the reduced effective mass of the electron and heavy hole.

With application of a magnetic field the exciton continuum splits into discrete excited excitonic states, which are weakly bound to Landau levels and which can, therefore, be described by free-electron and hole states. Optical transitions between these states experience a linear energy shift with applied magnetic field according to Eq. (5). In contrast, the discrete ground-state exciton transition shows a weaker energy dependence on the magnetic field as long as $\gamma = \hbar \omega / 2B_{1h} \ll 1$ ($B_{1h}$ being the electron–heavy-hole exciton binding energy). The simultaneous observation of the magnetic field dependence of the bound and the continuum states allows a direct determination of the band-edge and the exciton-ground-state energies and thus of the exciton binding energy in the respective MQWH.

The absorption spectra of a Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH with a well width of $L_w = 8.0$ nm as a function of the wavelength of the incident light are shown in Fig. 3 at different values of the applied magnetic field. The spectra are not corrected for the spectral response of the experimental setup. In particular, the small structures in the absorption spectra in the wavelength range of 1350–1450 nm are caused by absorption of water vapor in the air environment. The dashed lines indicate the evolution of the absorption features with magnetic field. The energies of these optical transitions in this MQWH are summarized in Fig. 4 as a function of the magnetic field. For the Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH with a 8.0-nm well the electron–heavy-hole exciton binding energy can be evaluated to $B_{1h} = 7.5 \pm 2$ meV. The reduced effective mass, which follows from the slope of the energy of the continuum transitions with the magnetic field, will be discussed in the second part of this work in conjunction with the subband structure of this MQWH system.

**B. Semiempirical absorption model for a 2D system**

The 2D absorption model, which we apply to interpret the absorption spectra in the range of the lowest subband transition in Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH's in order to determine the binding energy, linewidth, and oscillator strength of the electron–heavy-hole exciton, has been presented by Iwamura et al. for GaAs/Al$_x$Ga$_{1-x}$As MQWH's. This model takes into account discrete excitonic transitions, the steplike varia-
tion of the 2D density of states, and the excitonic enhancement of the continuum absorption, which is described by the Sommerfeld factor of an ideal 2D system given by Shinada and Sugano.\textsuperscript{14} The statistical ternary-alloy structure of the Ga\textsubscript{0.47}In\textsubscript{0.53}As well material and monolayer fluctuations of the well width cause an inhomogeneous broadening of the absorption spectra. This broadening has been accounted for by using a Gaussian line profile with a broadening parameter $\Gamma$ (the FWHM is 2.36$\Gamma$). In Fig. 5 the theoretical spectra (solid line) obtained by a nonlinear least-squares fit to the experimental absorption spectra of three MQWH's in the energy range of the lowest subband transition are shown together with the experimental absorption spectra (crosses). The dashed line represents the 2D model without the contribution of the electron–heavy-hole exciton. These model calculations will be used in Sec. IVE to determine the oscillator strength of the electron–heavy-hole exciton transition.

C. Linewidth of excitonic transitions

In contrast to the binary GaAs, statistical composition fluctuations are present in the ternary Ga\textsubscript{0.47}In\textsubscript{0.53}As material, which contribute to the linewidth of optical transitions. To evaluate this intrinsic linewidth contribution, we have applied the statistical model of Schubert et al.\textsuperscript{15} to quantum wells composed of ternary Ga\textsubscript{0.47}In\textsubscript{0.53}As. According to this model the statistical contribution to the exciton linewidth is given by

$$\Gamma_{\text{exc}} = \frac{dE_g}{dx} \left( \frac{x_0(1-x_0)}{K V_{\text{exc}}} \right)^{1/2},$$

where $K = 4a_0^{-3}$ is the density of the group-III elements in a zinc-blende-type crystal, $x_0 = 0.47$ the Ga concentration, and $dE_g/dx = 1.08$ eV for $x_0 = 0.47$, the band-gap

FIG. 4. Landau-level fan for a Ga\textsubscript{0.47}In\textsubscript{0.53}As/Al\textsubscript{0.48}In\textsubscript{0.52}As MQWH as a function of the magnetic field at 2 K.

FIG. 5. Comparison of experimental absorption spectra (crosses) of three Ga\textsubscript{0.47}In\textsubscript{0.53}As/Al\textsubscript{0.48}In\textsubscript{0.52}As MQWH's with different well widths with the theoretical absorption model (solid line). The dashed line indicates the theoretical model without the electron–heavy-hole exciton contribution.
variation of $\text{Ga}_{0.47}\text{In}_{0.53}\text{As}$ (after Ref. 16). The exciton volume has been corrected for the localization in the potential well to

$$V_{\text{exc}} = \frac{4}{3} \pi a_{\text{exc}}^3 L_z / 2.$$  \tag{7}

In Fig. 6 the results of the experimentally detected linewidths are compared with the theoretically calculated values. The dashed line represents the theory taking the 3D Bohr radius of 19 nm as the exciton radius in the well. If we use the exciton radius of an ideal 2D system ($a_{2D} = a_{3D} / 2$), the solid line results from the theory. Because of the continuous transition from a 3D to 2D system as a function of the well width, the intrinsic dependence of the linewidth follows the dashed line for large well widths (20 nm or greater), while for widths of about 5 nm the solid line should give a better description of the experimental situation. The comparison of the experimentally detected linewidths shows that the major contribution to the observed linewidths is caused by intrinsic composition fluctuations of the ternary $\text{Ga}_{0.47}\text{In}_{0.53}\text{As}$ material. In this evaluation we have not taken into account any nonparabolicity of the different bands which would increase the theoretical linewidth via the corresponding decrease in the exciton radius, caused by the larger carrier masses. This result again demonstrates the excellent quality of our MQWH layers.

In addition to this intrinsic broadening mechanism, fluctuations of the well width lead to a linewidth broadening of the excitonic transitions. According to the model of Singh and Bajaj they influence of well-width fluctuations depends on the step size and the lateral extent of the island with respect to the exciton radius in the well layer. The principal mechanism for this linewidth broadening is the shift $\Delta E$ of the subband energy with the variation of the well width $\Delta L_z$.

$$\Delta E = [d \langle E_{1h} - E_g \rangle / d L_z] \Delta L_z.$$  \tag{8}

As an indication for this model the energy shift $\Delta E$ for $\Delta L_z = 0.29$ nm is also shown as a dotted line in Fig. 6. This energy shift cannot be directly interpreted as a linewidth, because the microscopic interface structure has not yet been measured in the $\text{Ga}_{0.47}\text{In}_{0.53}\text{As}/\text{Al}_{0.48}\text{In}_{0.52}\text{As}$ material system. But this dotted line gives an indication that large linewidths can result for $L_z < 8$ nm, if monolayer fluctuations are present in the MQWH with a lateral island size which is comparable to the exciton radius.

The linewidths of the exciton resonance at low temperatures, reported by Weiner et al. and Kawamura et al., for the respective MQWH, are comparable or slightly larger than the values reported here. For the increase of the exciton linewidth with increasing temperature, caused by LO-phonon scattering, we observe a similar behavior as the previously mentioned authors.

### D. Exciton binding energy

As mentioned in Sec. IV A, the sharp features in the absorption spectra (Fig. 2) are due to discrete excitonic transitions. The ground state of the exciton is lower than that of the free electron and hole by a small amount (the exciton binding energy). Due to the confinement of the exciton inside the well, the electron and hole, orbiting around each other, lose gradually one degree of freedom. This happens, of course, when the well width becomes smaller than the exciton Bohr radius. The loss of a degree of freedom leads to a shrinkage of the wave function of the exciton in the $z$ direction (due to the confinement), but also, although to a lesser extent, in the $xy$ plane (due to the reduction of the kinetic energy of electron and hole with respect to their mutual attraction). The direct effect of this reduced extension of the wave function is an increase in the excitonic oscillator strength (discussed in Sec. IV E) and an increase in the exciton binding energy. For very thin layers the exciton is expected to become completely two-dimensional, which corresponds to a fourfold increase of its binding energy if an infinite barrier height is assumed. In the intermediate well-width range, values between the 3D Rydberg energy $\mathcal{R}$ and $4\mathcal{R}$ are expected.

The binding energies $B_{1h}$ of the electron–heavy-hole excitons in $\text{Ga}_{0.47}\text{In}_{0.53}\text{As}/\text{Al}_{0.48}\text{In}_{0.52}\text{As}$ MQWHs, which result from the magnetooptical measurements discussed in Sec. IV A and from the 2D absorption model, are presented in Fig. 7 as a function of the well width of the respective MQWH. The error bars of the binding energies evaluated by the 2D absorption model follow from model calculations, in which the exciton binding energy was kept constant. We then tried to describe the spectral shape of the absorption coefficient by varying only the other parameters of the model. Within the error bars the two methods yield consistent results for the exciton bind-
ing energies. The solid line represents the theoretical exciton binding energies based on a variational calculation in an infinite potential well, which overestimates the exciton binding energy because of the infinite barrier height. For the 3D Rydberg energy we have assumed a value of $R = 2.7$ meV, following the effective masses given in the second part of this work. The large values of the experimentally determined exciton binding energy, compared to the overestimated theoretical values, indicate an increase in the carrier masses due to nonparabolicity of the conduction and valence bands, respectively.

The estimate of the exciton binding energy of 5–6 meV for a 10-nm MQWH reported by Weiner et al. is in accordance with the results presented here. The estimated value of $B_{1h} = 16$ meV for a 7.5-nm MQWH given by Kawamura et al., however, is in contradiction to our results. An exciton binding energy of 16 meV would have led to a clear distinction between the exciton resonance and the $E_{1h}$ subband edge for the linewidths observed here. Obviously, this is not the case for the absorption spectrum of the 8-nm MQWH shown in Figs. 2 and 5.

E. Excitonic oscillator strength

The oscillator strength determines excitonic transitions both in the absorption and in the recombination process. The knowledge of the excitonic oscillator strength is thus important for an understanding of optical transitions in MQWH systems. Masumoto et al. pointed out that the excitonic oscillator strength is directly proportional to the integrated area of the excitonic resonance in the absorption spectra, which can be determined by using the 2D absorption model described in Sec. IV B. For a precise description of the area of the excitonic resonance, see also Fig. 5. As for the GaAs/Al$_x$Ga$_{1-x}$As MQWH system the absorption coefficient of the 2D continuum, taken from Fig. 2 as indicated by the double arrows, is proportional to the inverse of the well width. The integrated area of the exciton resonance and thus the oscillator strength, however, shows a much stronger dependence on the well width. These results for the Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH system are summarized in Fig. 8. The solid lines represent the $1/L_z$ dependence both for the integrated area $S$ in the absorption spectrum and for the continuum absorption coefficient, shown in the inset. This strong enhancement of the oscillator strength of excitonic transitions is caused by different contributions. On the one hand, the exciton wave function shrinks directly due to the confinement as discussed before. On the other hand, because of band nonparabolicities the reduced effective mass of the exciton increases with decreasing well width, which causes an additional shrinkage of the exciton wave function. This shrinkage of the exciton wave function and thus the transition from a 3D to a quasi-2D exciton can be illustrated by plotting the inverse oscillator strength, which is directly proportional to the exciton volume, as a function of the well width (Fig. 9). The solid line in Fig. 9 serves as a guide to the eye. For large well widths, i.e., $L_z > 2a_{3D}$ (where $a_{3D} = 19$ nm, 3D Bohr radius of Ga$_{0.47}$In$_{0.53}$As), the exciton remains almost undisturbed in the well layer. By reducing the well width below this value the exciton will be squeezed in the $z$ direction by the Al$_{0.48}$In$_{0.52}$As barrier layers. Initially for $L_z > a_{3D}$, the exciton volume decreases roughly proportional to $L_z$. For $L_z = a_{3D}/2$ a drastic change in the exciton volume is observed as can be seen from Fig. 9. In this range the exciton wave function has shrunk substantially for the two reasons mentioned above. Simultaneously with the de-

![Figure 7](image.png)

**FIG. 7.** Electron–heavy-hole binding energy $B_{1h}$ in a Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH as a function of the well width. The solid line represents the variational calculation of the binding energy assuming an infinite barrier height.

![Figure 8](image.png)

**FIG. 8.** Integrated area $S$ of the electron–heavy-hole exciton resonance in absorption spectra of a Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH, which is proportional to the exciton oscillator strength, as a function of the well width. The inset shows the variation of the continuum absorption coefficient as a function of the well width $L_z$. The solid lines represent the $1/L_z$ dependence.
crease of the exciton volume an increase of its binding energy is observed in the same well-width range. In QWH's with a well width of $L_z = a_{3D}/4$ ($\approx 4$ nm) quasi-2D excitons are realized. The quasi-2D exciton volume is schematically shown in the left inset of Fig. 9, where the exciton extent in the $x$ or $y$ direction is much larger than the well width $L_z$. When the well width is further decreased to below $\approx 2$ nm an increasing penetration of the exciton wave function into the barrier material occurs which leads again to an increase of the exciton volume. In the limit of zero well width, this volume reaches the value of a 3D exciton in the respective barrier material.

For the well-width range between 20 and 10 nm ($a_{3D} < L_z < a_{3D}/2$), which is the range for the application of the quantum confined Stark effect, we assume an increase in the exciton oscillator strength by a factor of 3 to 6 as compared to bulk-type Ga$_{0.47}$In$_{0.53}$As epilayers.

V. CONCLUSION

The excellent structural quality of Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH's grown lattice matched to InP by MBE is confirmed by double-crystal x-ray diffraction experiments. These measurements demonstrate the chemical homogeneity of the ternary Ga$_{0.47}$In$_{0.53}$As well and the Al$_{0.48}$In$_{0.52}$As barrier layers. Because of the periodic layer sequence individual layer thicknesses can be evaluated from the appearance of satellite reflections in the diffraction pattern. Fluctuations of the well width in these structures are below one to two monolayers. The precise determination of these structural parameters of MQWH's, especially in this all-ternary material system, is the essential prerequisite for the evaluation of the intrinsic properties of excitonic transitions in Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH's.

Absorption experiments on MQWH's with well widths down to 2.3 nm clearly demonstrate the step-like variation of the 2D density of states. The observed resonances at the subband edges can be assigned to discrete excitonic transitions by magnetoabsorption experiments. The dominant contribution to the linewidth of these excitonic transitions is caused by the intrinsic statistical composition fluctuations of the ternary Ga$_{0.47}$In$_{0.53}$As well material. This again demonstrates the high quality of the investigated Ga$_{0.47}$In$_{0.53}$As/Al$_{0.48}$In$_{0.52}$As MQWH's.

The exciton binding energies, determined from the description of the absorption spectra by a semiempirical model, agree with the values resulting from magnetoabsorption experiments within the experimental error. With decreasing well width we observe an increase of the exciton binding energy to $9 \pm 2$ meV for $L_z = 4$ nm. For this well width a quasi-2D system, i.e., exciton extent in the $x$ and $y$ directions much larger than the well width $L_z$, is realized. This can be shown by evaluating the excitonic oscillator strength or the exciton volume, which is inversely proportional to the oscillator strength, as a function of the well width. For MQWH's with well widths of 20–10 nm, which are important for application of the quantum confined Stark effect in novel photonic devices, an increase of the exciton oscillator strength by a factor of about 3 to 6 is estimated as compared to bulk-type Ga$_{0.47}$In$_{0.53}$As material.

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