

Localization of excitons in thermally annealed $\text{In}_{0.14}\text{Ga}_{0.86}\text{As}/\text{GaAs}$ quantum wells studied by time-integrated four-wave mixing

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 (Received 5 November 1997)

We have studied the effects of thermal annealing on the localization of excitons in $\text{In}_{0.14}\text{Ga}_{0.86}\text{As}/\text{GaAs}$ multiple quantum wells by means of linear spectroscopy and time-integrated four-wave mixing. The localization occurs due to interface roughness resulting from well width fluctuations, respectively, alloy fluctuations on a length scale comparable to the exciton Bohr radius. Due to lateral inhomogeneities of the diffusion of In out of the quantum well and of Ga into the quantum well during the annealing, we observe a structural transition of the localization potential for annealing temperatures greater than 850 °C. This transition is characterized by the development of disorder on a length scale larger than the exciton Bohr radius. [S0163-1829(98)02512-0]

I. INTRODUCTION

The subject of disorder and localization has attracted considerable interest since the pioneering work of Anderson.¹ In quantum-well (QW) systems, the localization is mainly caused by interface roughness. Variations in the well width of one or two monolayers will, in narrow QW's, lead to differences in the exciton (X) energy of several meV. Therefore the X 's can be localized in potential minima associated with regions of enhanced thickness on a length scale comparable to the Bohr radius.

The common picture of localization in QW's is based on the experimental work of Hegarty and co-workers²⁻⁴ on $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ QW's and was theoretically formulated by Takagahara.⁵ It predicts that X 's in the energetic region below the center of the absorption line are localized by the interface roughness, while X 's above the line center are mobile. The center of the absorption line was therefore interpreted as a mobility edge for the X motion.^{3,5}

This theory was successful to describe the experiments on GaAs QW's. On the other hand, in experiments on $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{InP}$ QW's no mobility edge could be observed.^{6,7} In this material system all X 's seem to be localized. This was interpreted as an enhanced localization due to alloy fluctuations in this material system. Similar results were found in $\text{ZnCdSe}/\text{ZnSe}$ QW's.⁸

Also in $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ QW's, recent experiments^{9,10} revealed a more complicated situation. Under certain experimental conditions, both a stimulated photon echo, originating from localized X 's, and a free polarization decay from free X 's were found, which had an identical spectral response. These results indicated that the picture of the mobility edge, separating localized and delocalized states, may be incomplete and that there may exist a mixture of localized and delocalized states at the same energy.

Also additional theoretical approaches were developed, based on numerical simulations of the growth process. The calculation of the exciton kinetics in these models^{11,12} showed that there were no really free, extended X states. In contrast, all X 's are localized to some degree. Nevertheless, one can define an effective mobility edge, which separates

between strongly localized X 's and X 's with a localization length of the order of the diffusion length.¹³ Depending on the degree of disorder, this effective mobility edge was shown to be energetically well defined or to result in a rather smooth transition across the absorption line.¹³ The latter case occurs in samples with large potential fluctuations, where X 's of the same energy may be localized within one part of the QW and delocalized within another part.¹³

In this paper, we have investigated the localization of X 's in $\text{In}_{0.14}\text{Ga}_{0.86}\text{As}/\text{GaAs}$ multiple QW's, which were technologically modified in a controlled way. For this, the QW's were thermally annealed for 1 min at different temperatures between 750 and 950 °C. The annealing causes an interdiffusion of In out of and of Ga into the QW and, therefore, the band gap in the QW increases.¹⁴ With the technique of time-integrated degenerate four-wave mixing (DFWM), we studied the energy dependence of the X homogeneous linewidth Γ , using ps pulses shifted across the absorption line. Additionally, we recorded absorption and photoluminescence (PL) spectra of the samples. For the as grown sample we observe a mobility edgelike behavior of Γ , i.e., Γ is small for energies below the absorption line center and rises monotonically above. This behavior is even more pronounced for the QW's annealed at low temperatures. We ascribe this to an enhanced X localization occurring in the annealed samples. This appears due to lateral inhomogeneities of the diffusion of In and Ga during the annealing, which causes enhanced interface roughness by alloy fluctuations. For annealing temperatures higher than 850 °C we find clear hints for a structural change, which is driven by the inhomogeneities of the diffusion and which is characterized by the development of deep, long-scale fluctuations of the QW potential. These long-scale fluctuations result in a weak localization of the whole X population, which is seen in a constant value of Γ across the absorption line.

In the following section we briefly discuss the sample preparation and the experimental technique. Section III is devoted to the results of the linear spectroscopy, while Sec. IV treats the DFWM experiments. In Sec. V we discuss the quantum beats occurring in the DFWM signals.

II. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUE

Our samples were grown by molecular-beam epitaxy on a semi-insulating GaAs substrate at a growth temperature of 520 °C.¹⁵ They consist of 20 $\text{In}_x\text{Ga}_{1-x}\text{As}$ QW's with an In content of 14% and a width of 3 nm, separated by 60-nm-wide GaAs barriers. The samples were processed by a rapid thermal annealing step. For this, they were heated for 1 min at different temperatures between 750 and 950 °C.

The QW's were characterized by linear and nonlinear spectroscopy, i.e., by photoluminescence, absorption, and time-integrated, degenerate four-wave mixing. The samples were mounted in a liquid-helium cryostat, allowing measurements at varying temperatures between 2 and 30 K. For the PL experiments, we used a helium-neon laser as an excitation source. The luminescence was dispersed by a 0.25-m monochromator and detected with a liquid-nitrogen-cooled charge-coupled-device (CCD) camera. Absorption spectra were taken, measuring the transmission of the light from a broad-band lamp through the samples.

The DFWM experiments were performed, using the standard two-pulse self-diffraction technique.^{16,17} Two pulse trains from a fs Ti-sapphire laser (wave vectors \vec{k}_1 and \vec{k}_2) with a certain, variable delay time τ between them, were focused upon the sample into a spot of approximately 100 μm diameter. The DFWM signal in the phasematched direction $2\vec{k}_2 - \vec{k}_1$ was detected with a slow photomultiplier, using standard lock-in technique. The Ti-sapphire pulses were spectrally compressed to a width of approximately 2 meV, using a grating and a slit. The energy of the exciting laser was varied across the heavy-hole X absorption line to measure the energy dependence of the dephasing time. The average excitation power in the DFWM measurements did not exceed 5 mW, corresponding to an X density of less than $5 \times 10^9 \text{ cm}^{-2}$. Exciton-exciton interaction is therefore negligible.

III. LINEAR SPECTROSCOPY

Figure 1(a) shows the absorption and PL spectra at 5 K of three typical samples, namely, of the as-grown QW and the QW's annealed at 750 and 950 °C, respectively. The PL line of the as-grown QW is centered at 1.4591 eV and has a full width at half-maximum (FWHM) of approximately 2.9 meV, while the absorption line has its maximum at 1.4606 eV and its half-width is 2.1 meV.¹⁸ Due to the thermally activated diffusion of In out of and of Ga into the QW, the band gap increases and the PL and absorption lines of the annealed samples shift to higher energies with increasing annealing temperature. For the highest annealing temperature of 950 °C, the PL line is shifted 14.7 meV to a value of 1.4738 eV, while the absorption line is shifted 14.4 meV to 1.4750 eV.

While the PL and absorption lines show a comparable blueshift with growing annealing temperature, the half-widths of these lines develop quite differently. This is shown in Fig. 1(b), where the FWHM's of absorption (filled dots) and PL (open circles) lines are plotted as a function of the annealing temperature. The FWHM of the PL line does not change much during the annealing, having a value of ap-

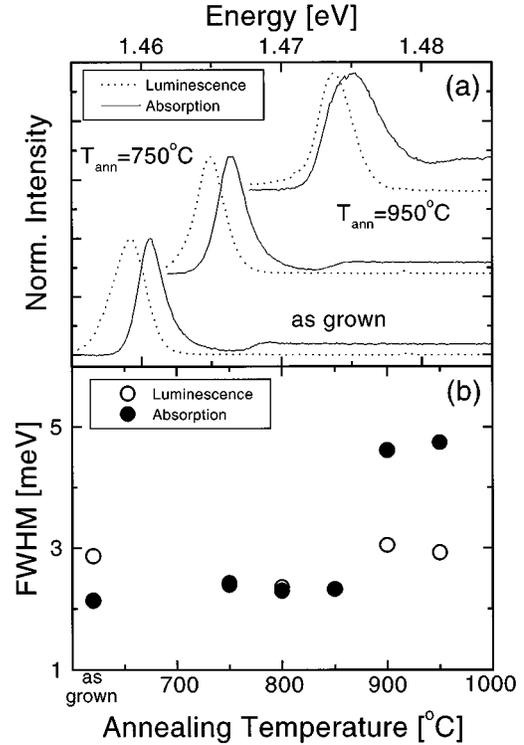


FIG. 1. (a) PL and absorption spectra of the as-grown QW and the QW's annealed at 750 and 950 °C. (b) shows the FWHM of the PL and absorption lines as a function of the annealing temperature.

proximately 2.5 meV for all samples. For temperatures below 850 °C also the absorption line FWHM is nearly constant and comparable to the half-width of the luminescence line. Above this temperature it increases quickly and reaches a value of 4.7 meV for $T_{\text{ann}}=950$ °C, i.e., it is now nearly twice as large as the corresponding PL line FWHM. Note that this broadening cannot be attributed to higher QW subbands collapsing into the ground subband due to the smoothing by annealing.

The broadening of the absorption line is connected with a change of the form of the absorption spectrum. The PL spectra have a symmetrical form for all measured QW's and can be well described by a Voigt line shape. The absorption line shows, in contrast, a more and more pronounced high-energy tail with increasing annealing temperature, as can be seen clearly in Fig. 1(a) for the case of $T_{\text{ann}}=950$ °C.

In general, the PL emission reflects the distribution of the local minima of the disordered QW potential, while the absorption contains information about the spectral distribution of all optically allowed X transitions.¹⁹ So the differences between the half-widths of PL and absorption lines that develop in the case of the annealed samples indicate that the annealing causes a change of the QW localization potential. The fluctuations in the potential increase and, therefore, the absorption linewidth is growing,²⁰ while the photogenerated X's can still relax to potential minima nearby and therefore the PL linewidth stays almost constant.

Another interesting feature in linear spectroscopy is the temperature development of the Stokes shift between the maxima of PL and absorption. For low temperatures, the Stokes shift is approximately 1.5 meV for all samples, as can be seen in Fig. 1(a). With increasing temperature, it develops

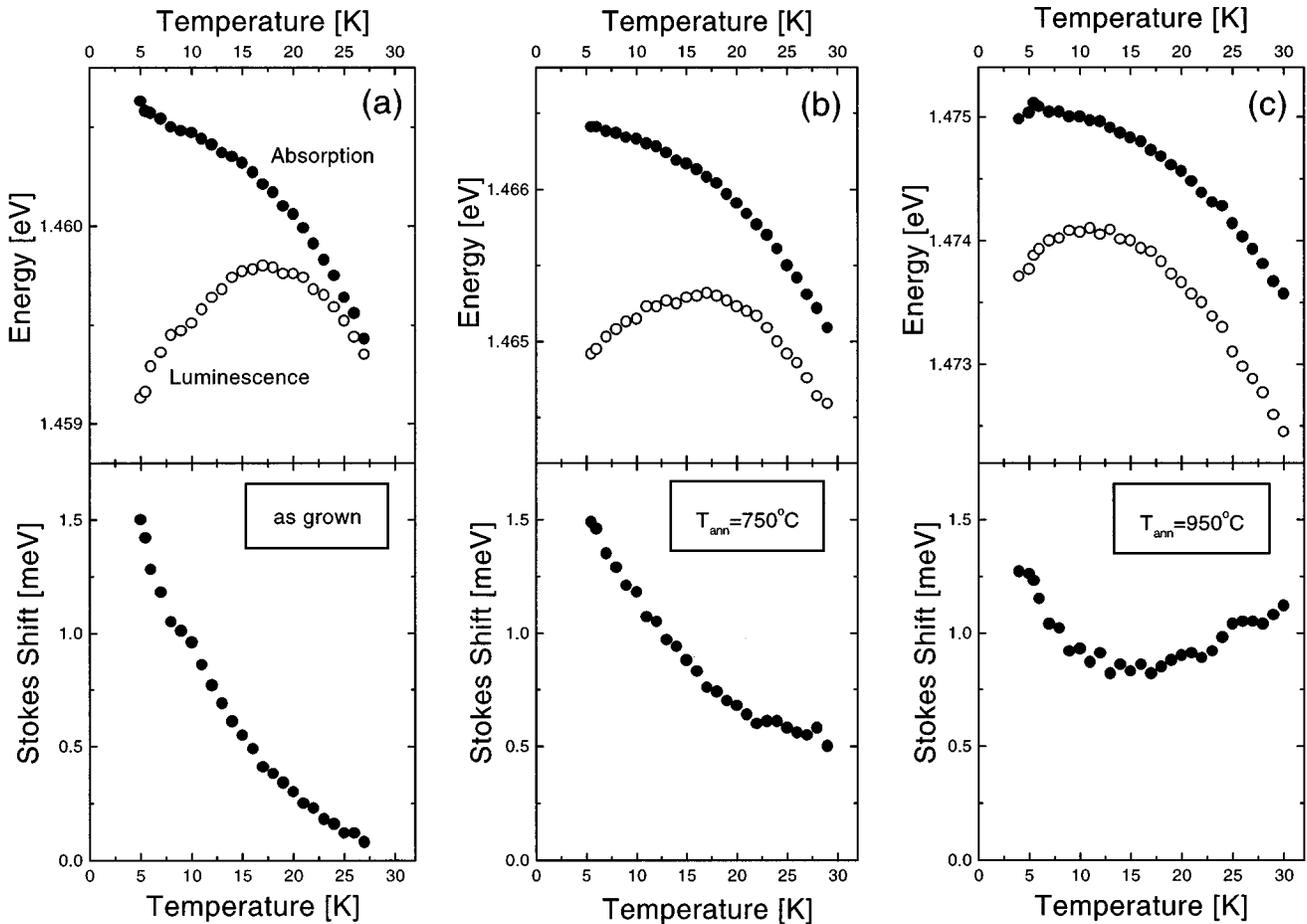


FIG. 2. Temperature development of the absorption and PL maxima for the as grown QW and the QW's annealed at 750 and 950 °C (upper graphs). The lower graphs show the development of the resulting Stokes shifts with temperature.

quite differently for the various annealing temperatures. The temperature dependence of the maxima of PL and absorption as well as of the Stokes shift is shown in Fig. 2 for three typical samples.

For the as grown sample [Fig. 2(a)], the Stokes shift has a value of 1.5 meV at 5 K. With increasing temperature, the absorption line shifts monotonically to lower energies, as expected from the temperature dependence of the band-gap energy. In contrast, the PL line shows, up to a temperature of about 15 K, a clear blueshift. This blueshift originates from the thermal activation of localized X 's to energetically higher states.¹² Above 20 K, the shrinkage of the band gap leads also to a redshift of the PL maximum, but it is weaker than the redshift of the absorption line. Therefore the Stokes shift between the maxima of PL and absorption lines is monotonically decreasing, as can be seen in Fig. 2(a), and finally vanishes for a temperature around 30 K.

The characteristic redshift of the absorption line with increasing temperature is also seen in the case of the QW annealed at 750 °C [Fig. 2(b)]. Also the shift of the PL line is qualitatively similar to that of the as-grown sample, going to higher energies for temperatures below 15 K and to lower energies above 20 K. The difference to the case of the as-grown sample is that the Stokes shift decreases much more slowly with increasing temperature. At a temperature of 30 K it still has a value of 0.5 meV, i.e., a part of the X 's is still localized in this QW, even at this elevated temperature. We

ascribe this to a roughening of the interfaces and therefore a stronger localization, caused by inhomogeneities along the QW in the diffusion of In and Ga.

For intermediate annealing temperatures ($T_{\text{ann}}=800$ and 850°C , not shown in Fig. 2) the behavior of the Stokes shift is still similar to that of the as-grown sample, except that it vanishes at even lower temperatures (approximately at 20 K for $T_{\text{ann}}=800^{\circ}\text{C}$ and approximately at 17 K for 850°C). This indicates a smoothing of the interfaces during annealing at such temperatures.

With further increasing annealing temperatures, we observe a qualitative change in the behavior of the Stokes shift, which is depicted for the case of $T_{\text{ann}}=950^{\circ}\text{C}$ in Fig. 2(c). Again, the absorption line shows the characteristic redshift with increasing sample temperature. The PL line reveals a blueshift up to a temperature of 10 K and shifts then back to lower energies. But now, for temperatures above 15 K, this redshift becomes stronger than the redshift of the absorption line and so the resulting Stokes shift is increasing from approximately 0.8 meV at 15 K to more than 1.1 meV at 30 K. An explanation for this unexpected, nonmonotonous behavior of the Stokes shift may be that X 's from shallow localized states of higher energy are thermally activated and can be trapped into deeper states.¹² This change in the behavior of the Stokes shift takes place under the same annealing conditions, for which also the absorption line form changes, as mentioned earlier. We suppose that an annealing at such high

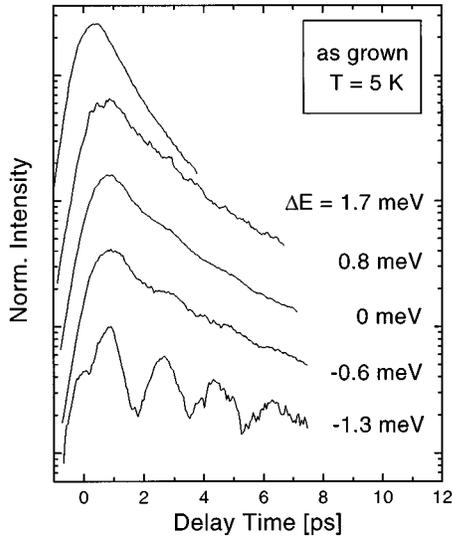


FIG. 3. DFWM signals of the as-grown QW, taken at different spectral positions ΔE of the exciting laser relative to the maximum of absorption.

temperatures causes a change of the localization potential, which is characterized by the development of deep potential fluctuations.

IV. NONLINEAR SPECTROSCOPY

Figure 3 shows DFWM signals of the as grown sample at 5 K for different energies of the exciting laser ΔE relative to the maximum of absorption, as denoted at the different curves. From the semilogarithmic representation it is seen that the DFWM signal decays to a good approximation exponentially with delay time. In addition, a periodic modulation of the signal is superimposed upon this decay for $\Delta E \leq 0$. We ascribe this modulation to quantum beats between the X and the biexciton (bi- X) state. The excitation energy dependence of the quantum beats and their origin will be discussed in Sec. V in more detail.

The decay time of the FWM signal depends strongly on the excitation energy. As the excitation energy is increased, the DFWM signal decays faster, i.e., the dephasing time T_2 decreases. The X transitions in our samples are mainly inhomogeneous broadened, and so we assume T_2 to be four times the decay time.²¹ At 5 K, the phase relaxation time T_2 , as determined from the exponential decay, varies from about 45 ps for $\Delta E = -1.3$ meV to 9 ps for $\Delta E = 1.7$ meV. As shown by Hegarty *et al.*,³ such a variation originates from the transition between strongly localized X states on the low-energy side of the absorption line ($\Delta E < 0$) and weakly localized X states on the high energy side ($\Delta E > 0$).

The energy dependence of the dephasing time can be seen more systematically in Fig. 4(a), where $T_2(\Delta E)$ is plotted for different temperatures between 2 and 30 K for the as-grown sample. The dephasing time changes as a function of both the excitation energy and the temperature, and ranges between 4 and 66 ps. For low temperatures, the variation with excitation energy is large, while for higher temperatures the differences between high and low excitation energies are small. This can be easily explained in the frame of localized

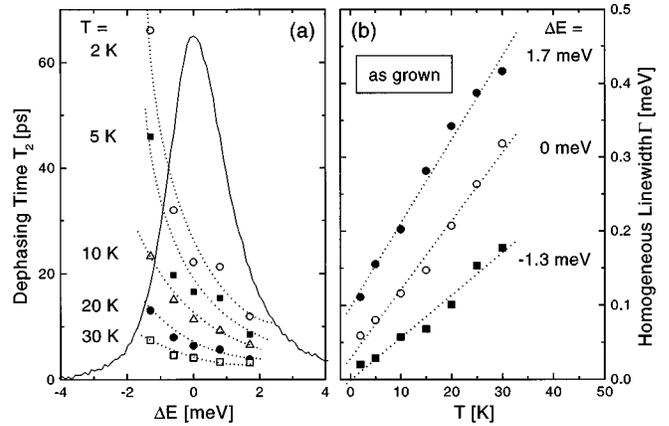


FIG. 4. Dependence of the dephasing time T_2 from the excitation energy ΔE for varying sample temperature T for the as-grown QW (a). The solid line shows the absorption spectrum. The dotted lines are to guide the eye. (b) shows the temperature dependence of the homogeneous linewidth Γ for different excitation energies. The dotted lines denote linear fits to the data.

X 's. With growing temperature, the phonon-assisted delocalization of the strongly localized X 's on the low-energy side of the absorption line leads to vanishing differences as compared to the "free" X 's on the high-energy side. This accounts for the stronger temperature dependence of the phase relaxation time in the case of $\Delta E < 0$, as compared to $\Delta E > 0$.

From the dephasing time T_2 we calculated the homogeneous linewidth Γ by $\Gamma = 2\hbar/T_2$. The temperature dependence of Γ for different excitation energies is shown in Fig. 4(b). Because it is inversely proportional to T_2 , Γ increases more weakly with increasing temperature for X 's on the low-energy side of the absorption line than on the high-energy side. A similar temperature dependence of the homogeneous linewidth for localized and free X 's was reported by Hellmann *et al.*²²

Figure 4(b) shows that Γ depends approximately linearly on the temperature for $\Delta E \geq 0$ and can therefore be written like $\Gamma(T) = \Gamma_0 + \gamma_{ac}T$. This behavior is typical for the scattering of free X 's by acoustic phonons in a one-phonon scattering process.⁵ For localized X 's, the phonon-assisted delocalization leads to a nonlinear temperature dependence $\Gamma \propto \exp(-E_a/kT)$, where E_a denotes the activation energy.⁵ For $\Delta E < 0$, we clearly observe deviations from the linear dependence. It is not possible to fit the data to an activationlike temperature dependence, because due to the finite spectral width of the exciting laser pulses, the activation energy varies across the laser line and the rise of $\Gamma(T)$ becomes multiexponential.

To distinguish the effects of dephasing by acoustical phonons and thermal delocalization and to understand the development of the X localization in the annealed quantum wells, we have fitted the temperature dependence of Γ linearly and evaluated the homogeneous linewidth at zero temperature Γ_0 . Although this neglects nonlinearities of the temperature dependence in the case of the localized excitons, we suppose that Γ_0 still is a useful measure for the degree of localization.

The development of Γ_0 with the excitation energy ΔE is

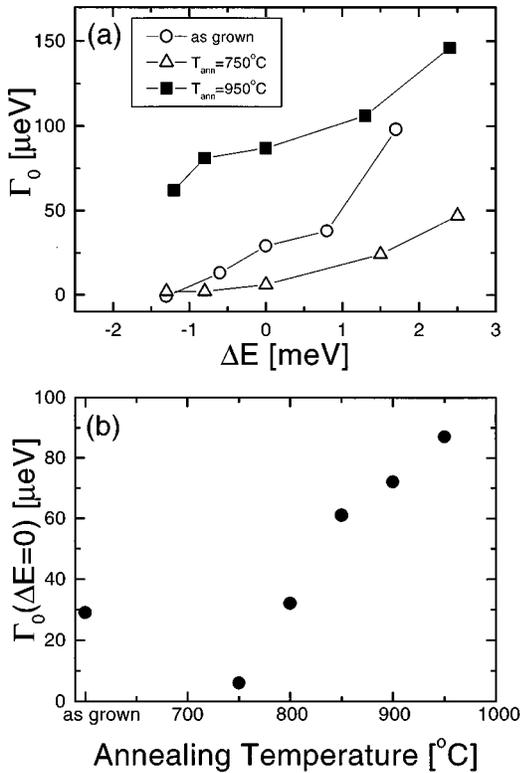


FIG. 5. Homogeneous linewidth at zero temperature Γ_0 as a function of excitation energy for three different QW's (a). The lower graph shows Γ_0 in the middle of the absorption line as a function of the annealing temperature.

shown in Fig. 5(a) for the as grown sample and the QW's annealed at 750 and 950 $^\circ\text{C}$, respectively. For the as-grown sample (open circles) we get the well-known transition from strong to weak localized X states. For $\Delta E < 0$, very low values of Γ_0 below 20 μeV are observed. In the middle of the absorption line Γ_0 reaches 29 μeV and for $\Delta E = 1.7 \text{ meV}$ it increases up to nearly 100 μeV . So in general, a mobility edgelike behavior, as reported by Hegarty *et al.*,³ is observed in the case of the as-grown sample.

The energy dependence of Γ_0 changes with the degree of localization, caused by the thermal annealing. For the QW annealed at 750 $^\circ\text{C}$ [open triangles in Fig. 5(a)], already the results of linear spectroscopy indicated an enhanced localization, as compared to the as-grown sample. This enhanced localization leads to a decrease of Γ_0 , as can be seen in Fig. 5(a). For the QW annealed at 750 $^\circ\text{C}$, Γ_0 is nearly zero for $\Delta E \leq 0$ and rises only slowly above the absorption center. This result again confirms our interpretation of an enhancement of localization that occurs during the annealing due to a roughening of the interfaces.

The QW's annealed at intermediate temperatures of 800 and 850 $^\circ\text{C}$ show an excitation energy dependence of Γ_0 that is similar to the as grown QW, but with slightly higher values of Γ_0 [not shown in Fig. 5(a)]. This is again consistent with the results of linear spectroscopy (see Sec. III) and shows that the interfaces are smoothed again with growing annealing temperature.

For annealing temperatures higher than 850 $^\circ\text{C}$, the dependence of Γ_0 from the excitation energy looks quite dif-

ferent. It is depicted in Fig. 5(a) for the example of $T_{\text{ann}}=950^\circ\text{C}$ (solid circles). Now, Γ_0 has already on the low-energy side of the absorption line a value that is comparable to Γ_0 for the "free" X 's in the as-grown sample. The variation of Γ_0 across the absorption line is small, compared to the as-grown QW. This is similar to the behavior observed for $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{InP}$ QW's.^{6,7} The relatively high value of Γ_0 for $\Delta E < 0$ shows that the X 's on the low-energy side of the absorption line are not strongly localized any more. This can be explained by a structural transition that takes place during the annealing at such high temperatures. It is connected with the development of fluctuations on a length scale larger than the X Bohr radius, in which the X 's are only weakly localized. Another hint for the development of such long-range fluctuations is the behavior of the observed quantum beats, which will be discussed in Sec. V.

As a measure for the degree of localization in the QW's, Fig. 5(b) shows Γ_0 in the middle of the absorption line (i.e., at $\Delta E=0$), as a function of the annealing temperature. $\Gamma_0(\Delta E=0)$ decreases from 29 μeV for the as-grown sample to 6 μeV for the sample annealed at 750 $^\circ\text{C}$. For higher annealing temperatures it increases again monotonically and reaches a value as high as 87 μeV for $T_{\text{ann}}=950^\circ\text{C}$. First, the localization is enhanced by the roughening of the interfaces, which occurs during the annealing. This strong localization must occur in potential fluctuations, which are comparable to the X Bohr radius. At higher annealing temperatures, these short-range potential fluctuations, which cause the strong localization, are smoothed out again and so the X localization is again comparable to or slightly weaker than in the as-grown QW. For $T_{\text{ann}} > 850^\circ\text{C}$, deep potential fluctuations develop, which cause only a weak localization of the X 's and which, therefore, occur on a length scale larger than the X Bohr radius.

V. QUANTUM BEATS

As already mentioned in the discussion of Fig. 3, the DFWM signals exhibit periodic modulations. Now we will discuss these quantum beats more carefully, because they also show a characteristic dependence on the annealing temperature. Figure 6 shows DFWM signals at 5 K of three QW's, namely of the as grown QW and the QW's annealed at 800 and 950 $^\circ\text{C}$ for different excitation energies. The periodic modulation of the DFWM signal is clearly seen for all three samples.

We suppose that the modulation arises due to quantum beats between the X and the bi- X state. This assumption is based on the following facts. First, the phase of the modulation does not change with excitation energy. This can be seen most clearly in the case of the QW annealed at 950 $^\circ\text{C}$ in the upper part of Fig. 6. It shows that we have real quantum beats rather than polarization interferences.^{23,24} Therefore these quantum beats cannot occur between bound and free X 's or between X 's from different QW islands. Second, the modulation vanishes if we excite the samples with co-circular polarized laser beams,²⁶ as expected for quantum beats between the X and the bi- X , because the creation of bi- X is strongly suppressed in this case. Moreover, the quantum beats always start with a minimum at zero delay, as expected for quantum beats between the X and the bi- X under excita-

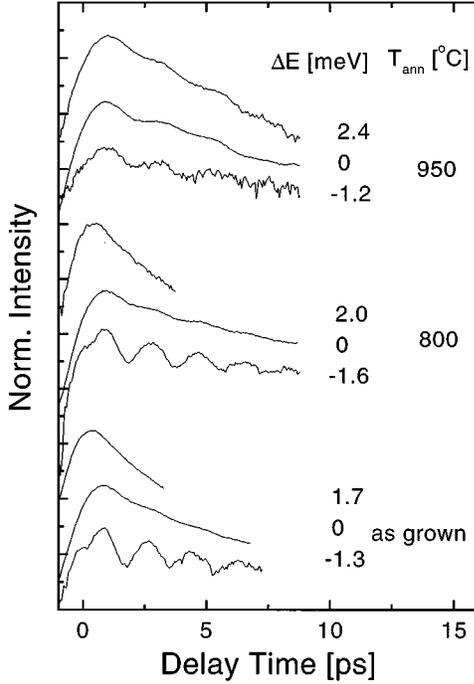


FIG. 6. DFWM signals of the as-grown QW and the QW's annealed at 800 and 950 °C for different excitation energies.

tion with collinear polarized laser pulses.²⁵

The period of the modulation Δ_M is connected with the bi-X binding energy E_B like $\Delta_M = h/E_B$.²⁷ For the as grown sample Δ_M is approximately 1.9 ps, corresponding to a bi-X binding energy of 2.2 meV. For the samples annealed at low temperatures (≤ 800 °C), the modulation period remains unchanged. For annealing temperatures greater than 850 °C, the modulation period increases and reaches a value of 2.2 ps for the QW annealed at 950 °C, corresponding to a bi-X binding energy of 1.9 meV. The decrease of the bi-X binding energy is a consequence of the decreasing localization at such annealing temperatures.

Another interesting feature is the excitation energy dependence of the quantum beats. For the as-grown QW we have already stated that the quantum beats are very pronounced for $\Delta E < 0$, while on the high-energy side and even for $\Delta E = 0$ the quantum beats are only barely visible. This behavior changes with growing annealing temperature. For $T_{\text{ann}} = 800$ °C, the modulation is clearly visible for $\Delta E = 0$, and for the QW annealed at 950 °C, the quantum beats are quite pronounced even for $\Delta E = 2.4$ meV. To obtain estimates for the energy dependence of the beating between the X and the bi-X state, we have fitted the DFWM decay curves to²⁷

$$I(\tau) \sim w_1^2 e^{-4\tau/T_{2,1}} + w_2^2 e^{-4\tau/T_{2,2}} + 2w_1 w_2 \cos(\Delta_M \tau) e^{-2\tau/T_{2,1} - 2\tau/T_{2,2}}, \quad (1)$$

where $w_{1(2)}$ and $T_{2,1(2)}$ are the spectral weights and the dephasing times of the X and bi-X state, respectively.

Figure 7 shows the ratio of the spectral weights of the bi-X and the X state w_2/w_1 as a function of the excitation energy for the three QW's already displayed in Fig. 6. The solid lines denote the corresponding absorption spectra. In

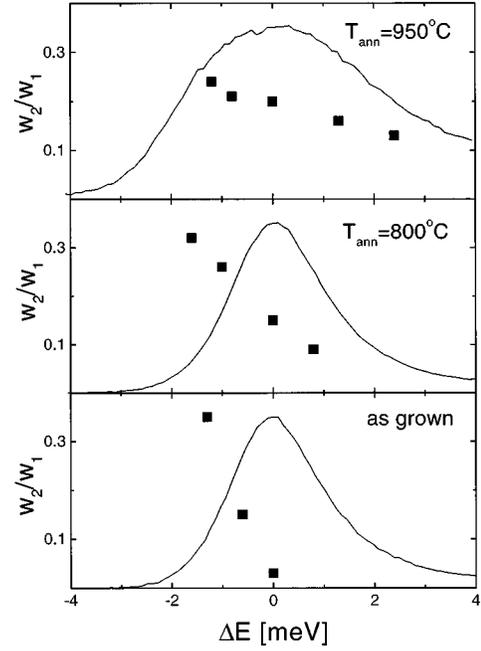


FIG. 7. Ratio of the spectral weights w_2/w_1 of the X and bi-X states for the as-grown QW and the QW's annealed at 800 and 950 °C as a function of excitation energy.

the case of the as-grown QW, w_2/w_1 has a value of 0.35 on the low-energy side of the absorption line, decreases very rapidly with increasing excitation energy, and vanishes completely for $\Delta E \geq 0$. For the QW annealed at 800 °C, w_2/w_1 has values comparable to that of the as-grown QW for $\Delta E \leq 0$, but has still a finite value for $\Delta E = 0$. For $T_{\text{ann}} = 950$ °C the variation of w_2/w_1 across the absorption line is only very small, i.e., the value for $\Delta E \leq 0$ is not as high as for the as-grown sample, but w_2/w_1 does not decrease very much for increasing excitation energy.

This different behavior of the ratio w_2/w_1 has also its origin in the localization of the X's. As we have seen before, in the as-grown sample X's below the absorption maximum are strongly localized, while X's with higher energies can move rather freely. The bi-X formation from localized X's in QW's was found to be enhanced remarkably, as compared to the formation from "free" X's.²⁸ Therefore, a clear modulation due to bi-X quantum beats is most likely expected if we excite localized X's on the low-energy side of the absorption line. For the weakly localized X's with $\Delta E > 0$ the bi-X formation is less efficient and, therefore, the modulation nearly vanishes.

We have seen in Sec. IV that the localization of the X's in the samples annealed at high temperatures is weaker as compared to the as grown sample and that there is no mobility edge to "free" X's anymore. This is consistent with the behavior of w_2/w_1 . The weakening of the localization is seen in the relatively low value of w_2/w_1 even for low energies. On the other hand, the beating is still present for $\Delta E \geq 0$, which is a clear hint that also the X's on the high-energy side of the absorption are, although weakly, localized.

VI. SUMMARY

In summary, we have investigated the influence of thermal annealing on the localization of X's in

$\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ QW's by linear and nonlinear spectroscopic methods, i.e., photoluminescence, absorption, and time-integrated, degenerate four-wave mixing. In linear spectroscopy we have measured the temperature dependence of the Stokes shift between absorption and PL. With DFWM we have especially investigated the energy dependence of the homogeneous linewidth. Moreover, the DFWM signal was clearly modulated by quantum beats between X and bi- X states.

The as-grown QW exhibits a mobility edgelike behavior, where the X 's below the absorption line center are strongly localized, while those above are only weakly localized. An annealing at relatively low temperatures causes an enhancement of the localization due to a roughening of the interface. With growing annealing temperature these short-range fluc-

tuations are smoothed again. For annealing temperatures greater than 850 °C, a structural transition of the localization potential occurs, which is characterized by the development of long-range fluctuations. In these deep, long-range fluctuations, all X 's, even those at the high-energy side of the absorption line, are weakly localized.

ACKNOWLEDGMENTS

We appreciate the expert technical support of E. Höfling and thank V. D. Kulakovskii for carefully reading the manuscript. The financial support by the Deutsche Forschungsgemeinschaft and the Volkswagen Foundation (L.V.K.) is gratefully acknowledged.

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