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Type II band alignment and exciton wave functions in Si/Si$_{1-x}$Ge$_x$ quantum wells

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Abstract

Photoluminescence experiments in high magnetic fields are presented which reveal diamagnetic shifts consistent with a type II CB offset for Si$_{0.76}$Ge$_{0.24}$ of at least 13 meV. From the magnetoluminescence data evidence for localized and free exciton recombination is found, which would not be separable from each other without magnetic field. © 2000 Elsevier Science B.V. All rights reserved.

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Si/Si$_{1-x}$Ge$_x$ heterostructures have attracted a lot of interest in recent years because of their potential for electronic devices but also because of the interesting properties related to strain and the many-valley conduction band. Despite important improvements in the material quality [1,2], fundamental questions like the type of the band alignment in Si/Si$_{1-x}$Ge$_x$ heterostructures could not be settled. Although it is known that strained Si$_{1-x}$Ge$_x$ layers confine heavy holes, the situation in the conduction band (CB) remained unclear. Because of the built-in strain in the Si$_{1-x}$Ge$_x$ layer the Δ_4-valleys in the directions parallel to the interfaces are lowered with respect to the Δ_2-valleys in the direction parallel to the growth direction. The question is whether the Δ_4 levels in the Si$_{1-x}$Ge$_x$ layer lie energetically above or below the Si CB edge, which corresponds to a type II or type I band alignment, respectively. Recent attempts to solve this problem utilized photoluminescence (PL) measurements under externally applied strain [3]. Although it has been claimed that such experiments would be able to distinguish unambiguously between types I and II alignment [4], the principle information obtained is whether the electrons involved in the luminescence occupy Δ_4 or Δ_2-valleys. In this context, the main conclusion from Ref. [3] was that the ground-state recombination in a 3 nm wide Si$_{0.7}$Ge$_{0.3}$ quantum well occurs via Δ_2 electrons. In our paper numerical calculations of the ground-state exciton wave functions

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will be used to give the binding energies for Δ_2-hh and Δ_4-hh excitons, which are essential for the interpretation of this experimental result with respect to CB offsets. Furthermore, we performed photoluminescence measurements on $Si_{1-x}Ge_x$ single quantum wells (SQWs) in magnetic fields up to 25 T, since the diamagnetic shift observed in such experiments should give additional information on the CB offset. This information can be extracted by modeling the diamagnetic shift with calculations considering excitonic recombination. To our knowledge only one such study was reported so far [5], in which very high excitation power densities had to be applied to detect rather weak luminescence signals.

The investigated $Si_{0.76}Ge_{0.24}$ SQW samples were grown pseudomorphically on Si by molecular beam epitaxy at 450°C and capped by 300 nm Si at the same temperature. PL was excited with the 488 nm line of an Ar^+ ion laser and detected with a liquid-nitrogen-cooled Ge detector. Laser light and luminescence light were guided through a silica multi-mode glass fiber pointing directly onto the sample surface. The sample chamber was cooled by liquid He and thermally contacted to the sample by He exchange gas. The magnetic field was applied along the growth direction of the samples.

The calculations were performed in an effective mass model allowing for different in-plane (parallel to the interfaces) and perpendicular (parallel to the growth direction) masses of the electrons and holes. The method, parameters and approximations are described in Ref. [6]. Although the parameters given there are valid for Ge contents x=30%, the modifications for x=24% are straightforward. The ground-state wave function of the exciton can be written as $\psi(\rho, z_e, z_h)$, where ρ is the relative in-plane distance between electron and hole,

$$\rho = \sqrt{(x_e - x_h)^2 + (y_e - y_h)^2}.$$

The positions of the electron and hole are given by (x_e, y_e, z_e) and (x_h, y_h, z_h), respectively, with the z-direction being parallel to the growth direction. The magnetic field, which was not treated in Ref. [6], is considered by an additional term in the Hamilton operator,

$$H_B = \frac{e^2B^2}{8m_\rho}\rho^2 \tag{1}$$

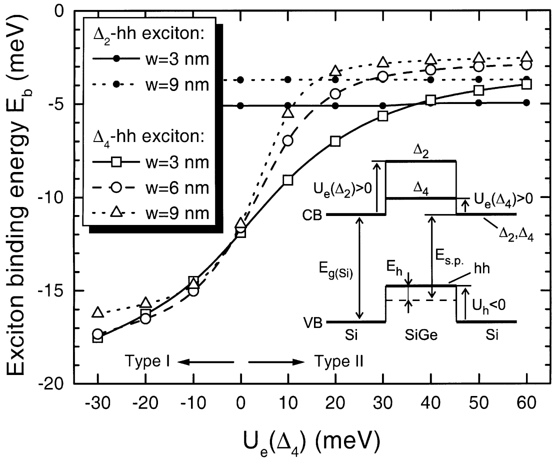

Fig. 1. The exciton binding energies for Δ_2- and Δ_4-hh excitons in 3, 6, and 9 nm wide $Si_{1-x}Ge_x$ quantum wells ($x = 30\%$) as a function of the Δ_4-CB offset $U_e(\Delta_4)$. The $w = 6$ nm curve for the Δ_2-hh exciton is omitted, but would fall in between the two shown curves. Lines connecting data points are guides to the eye. The inset schematically shows the band alignment in such quantum wells for a type II case.

with m_ρ denoting the reduced in plane mass. The following quantities will be utilized to illustrate wave functions in this paper:

$$p(z_e, z_h) = 2\pi \int_0^\infty d\rho\, \rho |\psi(\rho, z_e, z_h)|^2,$$

$$p_\rho(\rho) = \int_{-\infty}^{\infty} dz_h \int_{-\infty}^{\infty} dz_e |\psi(\rho, z_e, z_h)|^2.$$

The function $p(z_e, z_h)$ gives the probability density to find the electron at z_e and the hole at z_h, regardless of the in-plane distance ρ, and $p_\rho(\rho)$ is the probability density for an in-plane distance ρ, regardless of the z-coordinates of the particles.

The inset of Fig. 1 schematically shows the band structure in a $Si_{1-x}Ge_x$ SQW and defines all energies of interest here, especially the CB offsets $U_e(\Delta_2)$ and $U_e(\Delta_4)$ for the Δ_2- and Δ_4-CB, the single-particle confinement energy for the hole, E_h, and the single-particle recombination energy, $E_{s.p.}$, which is independent of the conduction band offsets as long as the band alignment is type II (i.e. $U_e(\Delta_4) > 0$). We define an exciton binding energy $E_b = E_X - E_{s.p.}$, where E_X is the exciton recombination energy. This definition is somewhat unusual since it yields negative binding energies, but is preferred here to illustrate their influence on recombination energies. Fig. 1 gives the binding energies for Δ_2-hh and Δ_4-hh excitons in $\mathrm{Si}_{0.7}\mathrm{Ge}_{0.3}$ SQWs with widths of $w = 3$, 6, and 9 nm as a function of the Δ_4-CB offset $U_\mathrm{e}(\Delta_4)$. The binding energy varies strongly for the Δ_4-hh excitons but is nearly constant for the Δ_2-hh excitons, since the Δ_2-CB is always far above the Si band edge. There is no doubt that Δ_4-hh excitons would be energetically favorable in the type I range. In the type II range the recombination energies of Δ_2 and Δ_4 excitons differ only by their binding energies, therefore the exciton with the larger amount of E_b will be favorable. If we concentrate for the moment on the $w = 3$ nm curves, which correspond to the sample investigated in Ref. [3], it can be seen that the Δ_4 exciton stays favorable up to $U_\mathrm{e} \cong 37$ meV, only at even higher offsets the Δ_2 exciton forms the ground state. This means that the observation of a Δ_2 ground state reported in Ref. [3] is only consistent with a type II offset of at least 37 meV. For the 9 nm wide well this changeover to Δ_2 behavior occurs at significantly lower offset (about 17 meV). The reason why the amount of the binding energy can become larger for the Δ_2 exciton in spite of the much higher CB barrier lies in the very different electron masses in z-directions, which cause that the Δ_4 electrons extend further into the surrounding Si layers [6].

Fig. 2 shows the no-phonon (NP) peak in PL spectra from the 4.8 nm wide $\mathrm{Si}_{0.76}\mathrm{Ge}_{0.24}$ SQW, recorded at magnetic fields from 0 to 20 T. A clear shift to higher energies and an increase in intensity with increasing magnetic field can be seen. In the lower inset the shift of the peak position with respect to its zero-field position is plotted as a function of the magnetic field. The peak positions were obtained from fitting Gaussian peaks to the spectra. The solid line in the inset is a fit to the diamagnetic shift which was obtained from our numerical calculations when assuming a type II conduction band offset $U_\mathrm{e}(\Delta_4) = 12.8$ meV and that the recombination stems from the recombination of Δ_4-hh excitons. This latter assumption is justified, since it was shown in Ref. [3] that the observation of Δ_2-like recombination requires extremely low excitation powers (about 1 $\mu\mathrm{W/cm}^2$), which are not used in our experiments. The fit reproduces the data points very well. To facilitate the interpretation of this diagram, shifts at $B = 20$ T which would correspond to different offsets are also included in the inset. It can be seen that the diamagnetic shift would be higher for higher offsets and therefore smaller amounts of binding energy.

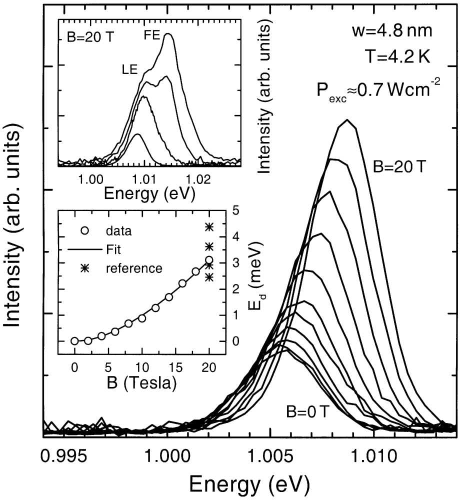

Fig. 2. No-phonon peak in PL spectra from a 4.8 nm wide $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ quantum well ($x = 24\%$) in magnetic fields from 0 to 20 T (2 T steps). The lower inset shows the analysis of peak positions together with a calculated fit using $U_\mathrm{e}(\Delta_4) = 12.8$ meV. As a reference, also shifts at 20 T which would correspond to offsets of 0, 10, 20, and 30 meV are included (shifts increase with increasing offset). The upper inset gives spectra at 20 T for successively increasing the excitation power by factors of 2.

The data are clearly in favor of a type II band alignment for $x = 24\%$, even more so since it is likely that the experimental diamagnetic shift is indeed somewhat reduced because of exciton localization effects [7], as will be pointed out below. Our calculation does not take into account such effects, it should therefore in principle be compared with the shift of a free exciton, which would be larger. To obtain a larger shift it would be necessary to choose a higher offset, therefore, the experiment indicates that the offset is *at least* 12.8 meV for $x = 24\%$.

The calculated exciton wave functions for the 4.8 nm wide well at magnetic fields of 0 and 20 T are visualized in Fig. 3. The main diagram shows the functions $p_\rho(\rho)$ and it can be seen how the lateral extension of the exciton is decreased by the magnetic field. This is clear from Eq. (1), since a non-zero magnetic field adds a harmonic potential in ρ to the Hamilton operator. The inset of Fig. 3 gives contour plots of the corresponding functions $p(z_\mathrm{e}, z_\mathrm{h})$. It is probably easier to interpret these plots when one is

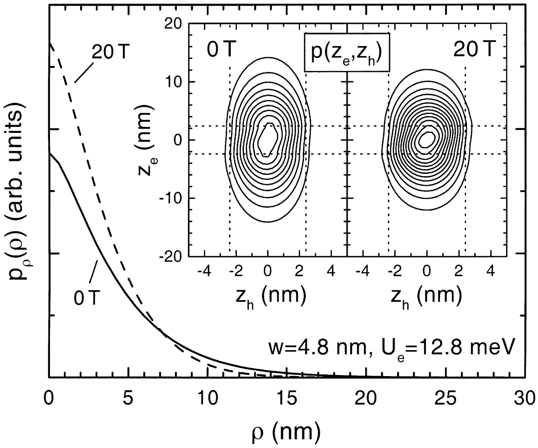

Fig. 3. The probability distributions $p_\rho(\rho)$ for the Δ_4-hh exciton wave functions at magnetic fields of 0 and 20 T, calculated for a 4.8 nm wide $\mathrm{Si}_{0.76}\mathrm{Ge}_{0.24}$ quantum well with a CB offset of 12.8 meV. The inset shows the corresponding distributions $p(z_e, z_h)$, the horizontal and vertical dotted lines indicate the quantum well borders for electrons and holes, respectively.

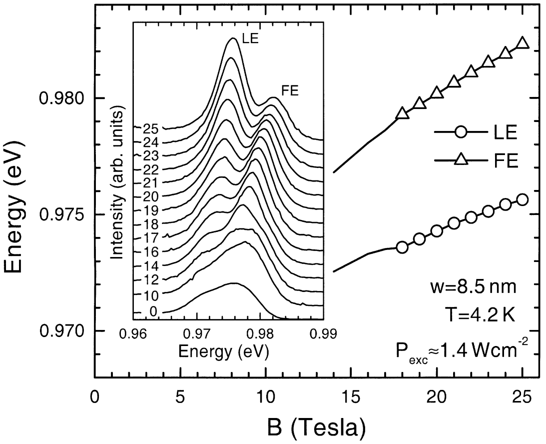

Fig. 4. Shift of the LE and FE no-phonon peak positions with magnetic field in the spectra from a 8.5 nm wide $\mathrm{Si}_{0.76}\mathrm{Ge}_{0.24}$ quantum well. Symbols are only used as long as the data are reliable. The inset shows the corresponding spectra taken at magnetic fields from 0 to 25 T. The spectra are plotted to scale with offsets.

aware that integrating the probability density over one coordinate yields one-particle distribution functions. It can be seen that the hole is well confined to the $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ layer, whereas the electron spreads considerably into the Si layers. The magnetic field narrows the electron distribution, since the decreased lateral extension of the exciton leads to an increased Coulomb interaction, which then also decreases the vertical extension of the exciton. Finally, the type II conduction band offset of about 13 meV is obviously not sufficient to separate the electron and the hole, for both carrier types the most probable location is in the center of the well.

The upper inset in Fig. 2 shows spectra from the 4.8 nm wide SQW at a magnetic field of 20 T and at different excitation powers. It can be seen that at higher excitation a second peak becomes visible at higher energies, whereas no such peak seems to appear when the excitation power is increased at zero field. We believe that at low excitation only a peak corresponding to localized excitons (LE) is present [8], which shifts slightly to higher energies with excitation because of the filling of localized states, until finally delocalized levels become accessible and a free exciton (FE) peak shows up. This interpretation is corroborated by measurements on a sample with an 8.5 nm wide SQW, where the distinct peaks are better resolvable. The inset of Fig. 4 shows spectra from this sample at a fixed excitation power but different magnetic fields. At higher fields two peaks are clearly resolvable, with the low-energy peak becoming more intense with increasing field. Fig. 4 itself gives the peak positions as a function of magnetic field. From 25 down to 18 T reliable positions can be given from fits, below that the situation becomes complicated by the appearance of an unidentified third peak, which can be well seen in the spectra taken at 12 and 14 T. In addition, the peaks become less resolved, and no separate analysis is possible anymore below 14 T. In the high-field range the two peaks shift almost parallel to each other, which excludes an explanation as resulting from different Landau levels, since they should exhibit clearly different slopes in such a diagram. The high-energy peak shifts only slightly more, which should be expected from a free exciton peak compared to a localized exciton peak [7]. A simple extrapolation of the shifts towards zero magnetic field would suggest separate peaks even without field, and indeed the spectrum taken at 0 T looks like the superposition of at least two broad peaks. Therefore, the separation of the peaks is probably not generated by the magnetic field and we assume that actually we deal with the presence of localized and free exciton recombination, which are almost indistinguishable without magnetic field applied. The $w = 4.8$ nm sample behaves qualitatively similar, therefore also the peaks in the upper inset of Fig. 2 are attributed to LE and FE recombination, and in particular the peak for which the diamagnetic shift was evaluated is assumed to correspond to localized excitons.

In conclusion, it was shown that recent results of PL measurements under external strain can only be explained with a quite large type II CB offset of about 40 meV for $Si_{0.7}Ge_{0.3}$. Our PL experiments in high magnetic fields on $Si_{0.76}Ge_{0.24}$ yielded diamagnetic shifts consistent with a type II CB offset of at least 13 meV. However, in fact significantly higher offsets are probable since the diamagnetic shift appears to be diminished by exciton localization effects. In addition, the measurements in magnetic field revealed the presence of both localized and free recombination channels, which are usually overseen when only measurements without magnetic field are examined.

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