Magnetoluminescence investigations of Si/Si_{0.76}Ge_{0.24} quantum wells

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Photoluminescence experiments in high magnetic fields reveal diamagnetic shifts consistent with a type-II conduction band offset of about 55 meV for Si/Si_{0.76}Ge_{0.24}. We point out that for such a type-II offset a magnetic field induced changeover from Δ_2 - to Δ_4 -like recombination is expected for an 8.5 nm wide quantum well, and that this changeover can explain our measurements on this well at low excitation power. The magnetoluminescence data provide evidence for localized and free exciton recombinations, which are hardly separable without the magnetic field. For interpretation of the data we employ calculations within an effective-mass model, obtaining two-particle ground-state wave functions for excitons in quantum wells at high magnetic fields.

I. INTRODUCTION

Si/Si_{1-x}Ge_x heterostructures have attracted much 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 such as the type of band alignment in Si/Si_{1-x}Ge_x heterostructures could not be settled for a long time. Although it was 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 was whether the Δ_4 levels in the Si_{1-x}Ge_x layer lie energetically above or below the Si CB edge, corresponding to a type-II or type-I band alignment, respectively. Recent photoluminescence (PL) measurements by Thewalt *et al.*³ under externally applied strain on a 3 nm wide Si_{0.7}Ge_{0.3} quantum well (QW) have proven a type-II band alignment. Based on considerations involving band bending it was later assumed that the magnitude of the type-II offset has to be of the order of few meV.⁴ However, one should bear in mind, that at very low excitation levels local fluctuations of the Ge content will play a role. Finally, in Ref. 5 it was shown that the experimental results³ are only consistent with a type-II CB offset of about 40 meV or more, if excitonic effects are properly taken into account. Further experimental confirmation of these findings seems highly desirable. The conduction band offset has considerable influence on the binding energy of excitons in a QW, which in turn affects the diamagnetic shift of the PL energy with magnetic field. We therefore performed PL measurements on a series of Si_{0.76}Ge_{0.24} single quantum wells (SQWs) in magnetic fields up to 25 T. Information on the CB offset can be extracted by modeling the diamagnetic shift

with calculations considering excitonic recombination. To our knowledge only one such study has been reported so far, in which very high excitation power densities had to be applied to detect rather weak luminescence signals.⁶

II. EXPERIMENTAL AND COMPUTATIONAL METHODS

The Si_{0.76}Ge_{0.24} SQWs were grown pseudomorphically on Si by molecular beam epitaxy at 450 °C and capped by 300 nm Si at the same temperature. The samples show the typical Si_{1-x}Ge_x luminescence signal consisting of the no-phonon (NP) line and various phonon replicas. PL was excited with the 488 nm line of an Ar⁺ ion laser and detected with a liquid-nitrogen-cooled Ge detector. Laser and luminescence radiation were guided through one and the same silica/silica multi mode glass fiber (core diameter 0.6 mm), which was pointing to the Si/Si_{1-x}Ge_x sample surface and directly contacting it. The lowest excitation power reported here, I_0 , corresponds to about 0.2 W/cm². The samples were mounted in a chamber which was cooled by liquid He and thermally contacted to the samples by He exchange gas. The magnetic field was applied along the growth direction of the samples. The measurements were performed at the High Field Magnet Laboratory in Nijmegen, either in a water-cooled Bitter-type magnet with a maximum magnetic field of 20 T, or in a hybrid magnet with a maximum field of 25 T.

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 (for $x=0.3$), and approximations are described in Ref. 5. Here we use $x=0.24$ to comply with our samples, and a reduced in-plane mass μ_ρ , which is defined in such a way as to reproduce the diamagnetic shift for anisotropic excitons quoted in Ref. 7, i.e.,

$$\frac{1}{\mu_x \mu_y} \left(\frac{1}{\mu_x} + \frac{1}{\mu_y} + \frac{1}{\mu_z} \right) =: \frac{1}{\mu_\rho^2} \left(\frac{2}{\mu_\rho} + \frac{1}{\mu_z} \right). \quad (1)$$

The z direction is chosen parallel to the $[001]$ growth direction, with $z=0$ in the center of the QW, and the reduced masses in the x , y , and z directions are denoted μ_x , μ_y , and μ_z . This method gives $\mu_p=0.1214m_0$, instead of $\mu_p=0.1306m_0$, which would be obtained according to Ref. 5. We propose that this decreased mass is the best approximation for describing diamagnetic shifts within our model. Using the mass according to Eq. (1) would yield a minimum Δ_4 CB offset of 33 instead of 37 meV for Si/Si_{0.7}Ge_{0.3}, which is no major modification.

Separating center-of-mass and relative motion, 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 and $z_{e(h)}$ denotes the z coordinate of the electron (hole). The Schrödinger equation for the ground state, including a magnetic field in the z direction, is then given by

$$\left(-\frac{\hbar^2}{2\mu_p} \frac{1}{\rho} \frac{\partial}{\partial \rho} \rho \frac{\partial}{\partial \rho} - \frac{\hbar^2}{2m_{ez}} \frac{\partial^2}{\partial z_e^2} - \frac{\hbar^2}{2m_{hz}} \frac{\partial^2}{\partial z_h^2} + U_e(z_e) + U_h(z_h) - \frac{e^2}{4\pi\epsilon_0\epsilon} \frac{1}{\sqrt{\rho^2 + (z_e - z_h)^2}} + \frac{e^2 B^2}{8\mu_p} \rho^2 \right) \psi(\rho, z_e, z_h) = E \psi(\rho, z_e, z_h). \quad (2)$$

Here $m_{e(h)z}$ denotes the perpendicular electron (hole) mass, $U_{e(h)}$ the heterostructure potentials in the conduction (valence) band, e the electron charge, ϵ_0 and ϵ the permittivity of free space and the dielectric constant, B the magnetic field, and E the eigenenergy of the eigenstate ψ . For illustrating wave functions in this paper we will utilize the function $p(z_e, z_h)$, which gives the probability density of finding the electron at z_e and the hole at z_h , regardless of the in-plane distance ρ . In addition, we introduce the function $p_\rho(\rho)$, which is the probability density for an in-plane distance ρ , regardless of the z coordinates of the particles:

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

The CB offsets for the Δ_2 and Δ_4 CBs are denoted $U_e(\Delta_2)$ and $U_e(\Delta_4)$, respectively, with positive values indicating that the QW is not favorable for the electrons.

III. MAGNETOEXCITONS: RESULTS AND DISCUSSION

In the following the recombination processes responsible for the PL signal from the SQW samples are established. Figure 1 shows the NP part of spectra taken without applied field at different excitation powers for the 8.5 nm wide QW. The peak shifts to higher energies with increasing power, but it is evident from the peak shape that this is no band-bending effect,⁸ but rather occurs due to the occupation of a higher energy recombination channel. We propose that at the lowest excitation powers only recombination from localized excitons (LEs) is observed, and that at increased excitation power this process saturates and in addition recombination

FIG. 1. NP peak in PL spectra from the 8.5 nm wide Si_{0.76}Ge_{0.24} QW, recorded at $B=0$ T and at different excitation powers P . From the peak shape it is apparent that with increasing excitation power an additional recombination channel at higher energies becomes populated; the arrows indicate estimated peak positions.

from free excitons (FEs) occurs.^{9–11} This presence of more recombination channels at high excitation power is more apparent in measurements with applied magnetic field (left part of Fig. 2). At higher fields two peaks are clearly resolvable, with the low energy peak becoming more intense with increasing field. From 25 down to 18 T reliable positions of these peaks can be given from fits. Below these fields the situation becomes complicated by the appearance of addi-

FIG. 2. NP peaks in spectra from the 8.5 nm wide Si_{0.76}Ge_{0.24} QW, taken at different magnetic fields. The magnetic field (in T) is given as a parameter to the curves. The spectra in the left part correspond to an excitation power of $6I_0$ and are plotted to scale with offsets. The spectra in the right part correspond to excitation powers of I_0 (solid lines) and $1.5I_0$ (dashed lines); they have been normalized and then plotted with offsets. LE indicates localized exciton, FE free exciton peaks.

FIG. 3. (a) NP peak positions in PL spectra from the 8.5 nm wide $\text{Si}_{0.76}\text{Ge}_{0.24}$ QW as a function of magnetic field, for different excitation powers P . LE and FE denote localized and free exciton peaks, respectively. Different data points for one and the same situation are intended to account for uncertainties in the evaluation of peak positions. Also included are calculated curves for Δ_2 - and Δ_4 -hh excitons and for different CB offsets $U_e(\Delta_4)$. The energy scale for all calculated curves is shifted in such a way that the $U_e(\Delta_4) = 57 \text{ meV}$ calculation coincides with the FE peak position at 25 T. (b) NP peak position in spectra from the 4.8 nm wide $\text{Si}_{0.76}\text{Ge}_{0.24}$ QW. Also included is a calculated Δ_4 -hh curve; the energy scale for this curve is shifted in such a way that the calculation coincides with the LE peak position at 0 T.

tional recombination on the high energy side, which can be observed in the spectra taken at 12 and 14 T, and by the fact that the peaks become less well resolved. With increasing magnetic field the luminescence intensity is redistributed toward the lower energy peaks.

Similar behavior was also observed for a QW of 4.8 nm width. For this well the FE peak was visible only at the highest magnetic fields and it was never possible to resolve LE and FE positions reasonably. This seems to be consistent with Ref. 9, where it was observed that the separation between LE and FE peaks becomes smaller with decreasing QW width. Since the FE energies are of major importance for the interpretation of the measurements, in the following we will concentrate on the 8.5 nm wide well.

At low excitation power (right part of Fig. 2) essentially only the LE peak is observed, which again shifts to higher energies with increasing field and becomes more intense. The open symbols in Fig. 3(a) show the resolvable peak positions at different excitation powers as a function of the magnetic field. It can be seen that the LE positions for the investigated excitation powers coincide quite well at high fields.

Localization effects on a length scale comparable to the

lateral extension of the exciton should lead to a decrease of the diamagnetic shift,¹² which is not reproducible by our theoretical model. The model can therefore only be used to fit the diamagnetic shift of the FE peak. To obtain the experimentally observed shift of about 3 meV for a Δ_4 -hh recombination when increasing the magnetic field from 18 to 25 T, we have to assume a comparably large type-II CB offset of $U_e(\Delta_4) = 57 \text{ meV}$. Nevertheless, this offset is still consistent with Ref. 5, since it was demonstrated there that only a lower limit for the offset can be given from the experiment in Ref. 3. The resulting curve is shown in Fig. 3(a). Note that the energy scale for all calculated curves is shifted (by about 13 meV) such that the $U_e(\Delta_4) = 57 \text{ meV}$ calculation coincides with the FE peak position at 25 T, in order to facilitate the comparison between calculated and experimental diamagnetic shifts. A perfect agreement in *absolute* energies cannot be expected because of the uncertainty of parameters entering the calculation. In particular, only a plausible, but not proved value of 194 meV is used for the valence band (VB) offset.⁵ Using a slightly different offset will not change any conclusions concerning energy differences, such as diamagnetic shifts or relative energies between Δ_2 - and Δ_4 -hh excitons, since the holes are basically well confined to the $\text{Si}_{1-x}\text{Ge}_x$ layer. Of course, a different offset leads to a shift in absolute energies. Consequently, the shifted curves shown in Fig. 3(a) can represent calculations corresponding to a slightly increased VB offset of about 207 meV.

Next we want to consider the energy of Δ_2 -hh excitons. Since the energies of these excitons are virtually unaffected by $U_e(\Delta_4)$ for the values treated in this paper,⁵ in Fig. 3(a) only one calculated Δ_2 -hh exciton curve is included, for which an intermediate value of 20 meV was chosen for $U_e(\Delta_4)$ [this means $U_e(\Delta_2) = 182 \text{ meV}$]. This curve can be used as a reference for all calculated Δ_4 -hh curves. It can be seen that at $B = 0 \text{ T}$ the Δ_2 -hh exciton is favorable for $U_e(\Delta_4) = 57 \text{ meV}$, but at high fields the Δ_4 -hh exciton becomes favorable, since its energy rises more slowly than that of the Δ_2 -hh exciton. This means that our picture is consistent. We fitted the shift of a Δ_4 -hh exciton to the FE data at high magnetic fields, and from the calculated Δ_2 -hh and Δ_4 -hh energies it follows that in this field range indeed the Δ_4 -hh exciton forms the ground state.

It could be suspected that an alternative assignment of luminescence peaks can be obtained by assuming that the peak labeled “LE” is in fact the free exciton ground state. To account for this scenario, we tried to reproduce the high field shift of the lower energy peak by a calculated curve. This fit yields an offset of $U_e(\Delta_4) = 11 \text{ meV}$ [see Fig. 3(a), noting that with this offset the Δ_4 -hh exciton is always favorable], which is too small given the considerations of Ref. 5. Moreover, such a fit cannot explain the low field behavior and raises a question about the origin of the higher energy peak. One could try to attribute this peak to recombination of an excited exciton state (similar to a “ $2s$ ” state in a hydrogen atom), but this would imply that in Fig. 1 excited exciton states were observed at zero magnetic field, which has to our knowledge never been reported for $\text{Si}_{1-x}\text{Ge}_x$ QWs. Furthermore, the relative intensities between the two peaks observable in the left part of Fig. 2 are not in accordance with the fact that the oscillator strength of the “ $2s$ ” exciton should

be negligible compared to the oscillator strength of the “1s” exciton.

These experimentally observed relative intensities also do not agree with an alternative explanation of the higher energy peak as being due to Δ_2 -hh recombination. For this scenario oscillator strengths can be calculated, and it turns out that, e.g., at a magnetic field of 20 T the oscillator strength of the Δ_4 -hh exciton at $U_e(\Delta_4)=11$ meV is approximately three orders of magnitude larger than the one for the Δ_2 -hh exciton. This is completely incompatible with the experiment, where the two recombination peaks are of the same intensity at this field (cf. the left part of Fig. 2). Thus, no good explanation for the higher energy peak can be given under the assumption that the low energy peak was due to FE recombination.

All the considerations presented above contradict the alternative assignment of peaks. Therefore our interpretation of the low energy peak as resulting from LE recombination is supported, and consequently also the high CB offset, which resulted from fitting the calculations to the shift of the higher energy FE peak, is endorsed.

For the shift of the LE peak our calculations are not directly applicable, but nevertheless a qualitative agreement in the shift could be expected when the decreased lateral extension of the exciton due to in-plane confinement potentials is modeled by a decreased type-II offset. For the Δ_4 -hh exciton such a decreased offset will result in a stronger penetration of the electron distribution into the $\text{Si}_{1-x}\text{Ge}_x$ layer. This will yield an increased Coulomb interaction between electron and hole, and therefore also a decreased lateral extension of the exciton.

Obviously, such a qualitative agreement cannot be achieved with a Δ_4 -hh curve, as can be seen from the discrepancy between the LE data and the $U_e(\Delta_4)=11$ meV calculation. But what has not been considered so far is that in the low magnetic field range, where the discrepancy occurs, the $U_e(\Delta_4)=57$ meV calculation predicts that the Δ_2 -hh exciton becomes favorable for FE recombination. This changeover from Δ_4 - to Δ_2 -like recombination is mainly related to differences in the electron distribution for Δ_4 -hh and Δ_2 -hh excitons, whereas the potentials leading to localization of the excitons affect the holes. It is therefore conceivable that for LE recombination, where additional potentials confine the hole laterally, a similar Δ_2/Δ_4 changeover will be observable, although not necessarily at exactly the same magnetic field.

The strong diamagnetic shift of the LE peak at low field could therefore be attributed to an initial Δ_2 -like recombination, which with increasing field is replaced by Δ_4 -like recombination. Further qualitative support for this interpretation can be found in the LE spectra shown in the right part of Fig. 2. In a field range below 10 T the PL spectra exhibit an asymmetry, which could be indicative of a coexistence of Δ_2 -like and Δ_4 -like recombination, and disappears for higher fields. To give an idea of the proposed changeover mechanism in Fig. 3(a) a calculation for $U_e(\Delta_4)=20$ meV is included. At this offset the changeover from Δ_2 - to Δ_4 -like FE recombination would occur at a magnetic field comparable to the one where the proposed changeover from Δ_2 - to Δ_4 -like LE recombination should take place.

In Fig. 3(b) peak positions from spectra taken for the 4.8

FIG. 4. The probability distributions $p_\rho(\rho)$ for the Δ_4 -hh exciton wave functions at magnetic fields of 0 and 25 T, calculated for an 8.5 nm wide $\text{Si}_{0.76}\text{Ge}_{0.24}$ quantum well with a CB offset of 57 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.

nm wide QW at different magnetic fields are given together with a calculated Δ_4 -hh curve, which was shifted in energy to coincide with the measurement at $B=0$ T. The spectra were recorded at an excitation power where only LE recombination was present. In contrast to the situation for the 8.5 nm wide well, here it is indeed possible to model the shift of the LE peak by a Δ_4 -hh curve with reduced CB offset. There is no indication for the appearance of Δ_2 -like recombination at low fields. This supports the statement in Ref. 5 that narrow wells are presumably less suited for the observation of Δ_2 -like recombination than wider ones.

Finally, Fig. 4 shows calculated Δ_4 -hh exciton wave functions at magnetic fields of 0 and 25 T for a CB offset of $U_e(\Delta_4)=57$ meV and a well width of 8.5 nm. 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. (2), since a nonzero magnetic field adds a harmonic potential in ρ to the Hamilton operator. The inset of Fig. 4 gives contour plots of the corresponding functions $p(z_e, z_h)$. It is apparent that the hole is well confined to the $\text{Si}_{1-x}\text{Ge}_x$ layer, whereas the electron is pushed 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. As can be seen from the contour plots in Fig. 4, this latter decrease is not very pronounced, due to the high type-II CB offset.

IV. CONCLUSION

In conclusion, it has been shown that PL experiments in high magnetic fields on $\text{Si}/\text{Si}_{0.76}\text{Ge}_{0.24}$ quantum wells yield diamagnetic shifts, which according to our model calculations are only consistent with a pronounced type-II CB offset of about 55 meV. Although this value is subject to some uncertainty, since it results from calculations based on an effective-mass model that neglects in-plane anisotropy, it

strongly supports a comparatively large type-II CB offset. The measurements reveal the presence of different recombination channels, attributed to localized and free exciton recombination, which are hardly noticeable in experiments at zero field. We interpret measurements at low excitation power by a magnetic field induced changeover from Δ_2 - to Δ_4 -like localized exciton recombination.

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