Heavy- and light-hole character of optical transitions in InAs/GaAs single-monolayer quantum wells

O. Brandt, H. Lage, and K. Ploog
Max-Planck-Institut für Festkörperforschung, D-7000 Stuttgart 80, Heisenbergstrasse 1, Germany
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We examine the nature of the optical transitions in InAs/GaAs single-monolayer quantum wells grown by molecular-beam epitaxy. The heavy- and light-hole character of the observed transitions is revealed by monitoring the linear polarization of the emitted luminescence and by selective excitation of the states by linearly polarized light. In addition, these experiments provide information about the band alignment between InAs and GaAs.

The optical properties of ultrathin InAs/GaAs quantum wells (QW's) are currently attracting great attention.[1-3] However, both the fundamental band alignment and the nature of the electronic states in these structures are still not well established. Recently, Ksendzov *et al.*[2] made an attempt to identify the nature of the optical transitions of 2–4-monolayer (ML) -thick InAs/GaAs QW's by using modulated absorption spectroscopy. This method is based on the different symmetry of heavy- and light-hole states and allows one to distinguish between light- and heavy-hole contributions to the optical transitions. The authors did not observe light-hole related features, but attributed the higher-energy peak in the absorption spectra to a transition between heavy-hole and unconfined electron states.

In this work, we examine the heavy- and light-hole character of the observed transitions in an InAs/GaAs single-monolayer quantum well. Polarization-dependent photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy are employed to identify the nature of each of the transitions. The heavy- and light-hole character of the transition is revealed by monitoring the emitted luminescence in the edge-emission and the edge-excitation mode. Additionally, we deduce information about the band alignment between strained InAs and GaAs.

The investigated InAs/GaAs structure is synthesized by conventional solid source molecular-beam epitaxy on semi-insulating (100) GaAs substrates. Details of the growth procedure have been described elsewhere.[4] The investigated sample consists of a single InAs layer with an average (integrated) thickness of 1.2 ML, capped with 200 nm GaAs. The integrated In coverage of the (100) GaAs plane as well as the strain state of the layer are determined by high-resolution double-crystal x-ray diffraction in connection with the dynamical x-ray-diffraction theory.[4] The sample studied here contains a coherently strained InAs layer; i.e., the whole lattice mismatch of 7.16% is accommodated by an elastic tetragonal distortion of the InAs unit cell. Moreover, these experiments demonstrate the existence of well-defined InAs/GaAs interfaces, meaning that fluctuations of the InAs film are restricted to monatomic steps. PL and PLE measurements in the conventional backscattering geometry are performed by using a tunable dye (Styril-9) laser pumped by an Ar^+ laser as excitation source. Another optical setup, equipped with an Ar^+ laser pumped Ti-sapphire laser as tunable excitation source, provides versatile detection and excitation geometries suitable for the polarization-dependent PL and PLE experiments. In either case, the emitted light is dispersed by a 1-m double monochromator and detected by a GaAs photomultiplier tube operating in the photon-counting mode.

In Fig. 1, we show the PL and PLE spectra of the 1.2-ML-thick InAs/GaAs single QW under consideration. The spectra are recorded in the usual backscattering geometry; i.e., without access to the polarization dependence of the optical transitions. The PL spectrum (dashed line) is dominated by a strong emission located at 1.438 eV. The low-energy tail of this emission can readily be saturated by increasing the excitation density and is thus related to an impurity. The weak emission at 1.4935

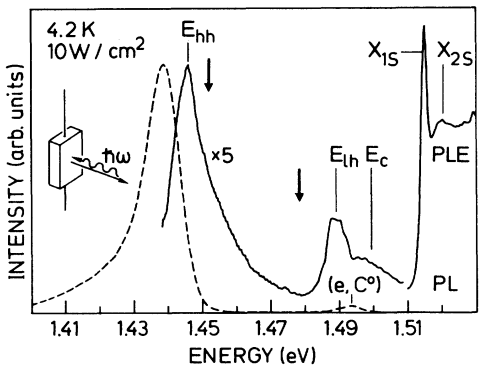

FIG. 1. Photoluminescence (dashed line) and photoluminescence excitation (solid line) spectra obtained at 4.2 K in standard backscattering geometry (as depicted in the illustrative scheme on the left) from a 1.2-ML-thick InAs/GaAs single quantum well. The PLE spectrum is obtained by detecting at 1.435 eV, i.e., 3 meV below the PL maximum. The peaks in the spectra are labeled according to the discussion in the text. The arrows indicate the spectral position calculated in the frame of the envelope-function method.

eV (e,C^0) originates from the band-to-acceptor recombination in the surrounding bulklike GaAs matrix. In the PLE spectrum (solid line), besides the strong 1*S*- and 2*S*-exciton resonances of the GaAs matrix, two resonances (E_{hh} and E_{lh}) and an additional bump (E_C) below the fundamental band edge of GaAs are observed. The two resonances originate from the selective excitation of excitons localized by the monolayer potential well.[5] By comparing the spectral positions of these transitions with the results of envelope-function calculations,[6] the resonances at 1.445 and 1.489 eV are attributed to the heavy-hole and to the light-hole exciton, respectively. The high-energy shoulder of the light-hole transition (E_C) has been attributed in a previous work to a transition involving electrons belonging to the InAs conduction-subband and unbound holes belonging to GaAs valence-band states.[7]

The heavy- and light-hole character of the optical transitions is experimentally accessible on the basis of their polarization dependence.[8,9] In general, any optical anisotropy in semiconductors having zinc-blende crystal structure originates from a reduction of the initial crystal symmetry, which removes the fourfold degeneracy of the Γ_8 valence-band multiplet of the bulk crystal. In the particular case of a thin strained film on a (001) surface, the growth axis defines a unique direction in the crystal since both the quantizing axis and the tetragonal distortion of the strained unit cell introduce an uniaxial perturbation along [001]. This uniaxial perturbation splits the heavy- and light-hole states at the center of the Brillouin zone. The polarization dependence at the zone center then originates from the specific symmetry character of the heavy- and light-hole states, giving rise to characteristic selection rules with respect to the direction of the electric-field vector of the incident light.[10] In particular, the oscillator strength of heavy- and light-hole related transitions is predicted to be $\frac{3}{4}$ and $\frac{1}{4}$ for the electric-field vector $\hat{e}$ parallel to the plane and 0 and 1 for $\hat{e}$ perpendicular to the plane, respectively.[11] Heavy-hole related transitions are thus not allowed with the light polarized parallel to the privileged direction.

In Fig. 2, we present the results of the polarization-dependent measurements. Figure 2(a) shows the polarization-dependent PL spectra obtained in the edge-emission mode.[9] The laser light is incident normal to the sample surface with its polarization parallel to the (100) plane. Luminescence emitted normal to the cleaved edge is analyzed by a polarizer in front of the entrance slit of the monochromator. Under these conditions, a clear polarization dependence of the InAs-related emission is observed. Note, that the (e,C^0) transition does not exhibit any polarization dependence, serving thus as a built-in reference for the response of the system. The InAs-related emission is polarized parallel to the plane, demonstrating the heavy-hole character of this transition. The polarization, however, is not complete. We point out that this partial polarization is not necessarily an indication for valence-band mixing. Light initially emitted towards the surface can be reflected and refracted by the surface and can so reach the detector. This light, however, is unpolarized with respect to the layer plane, thus representing a background contribution superimposed to

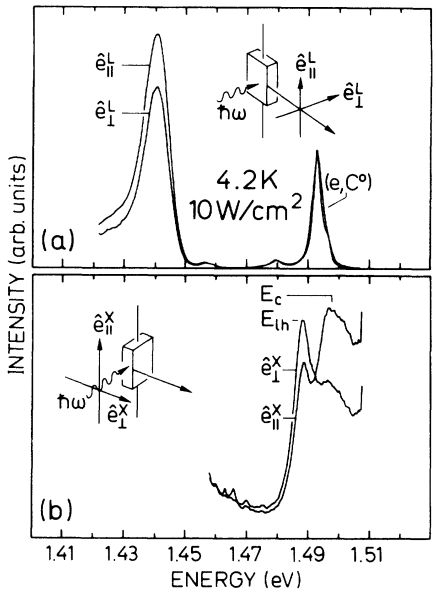

FIG. 2. Polarization-dependent photoluminescence (a) and photoluminescence excitation (b) spectra obtained at 4.2 K in the edge-emission and edge-excitation mode, respectively, of the sample of Fig. 1. The detection wavelength for the PLE experiment is set to 1.435 eV. The detection and excitation geometries are depicted by the schemes near the respective curves.

the polarized luminescence emitted perpendicular to the growth axis.[12]

In Fig. 2(b), we show the polarization-dependent PLE spectra obtained in the edge-excitation mode.[13] Here, the exciting light is focused right on the cleaved edge of the sample. Its polarization is set either parallel or perpendicular to the plane. The emission is detected from the sample surface close to the edge of the sample. In this geometry, we can selectively excite heavy- and light-hole-related transitions. As evident from the spectra, the transition assigned to the light-hole exciton does in fact exhibit a distinct light-hole character. The striking observation in these spectra, however, is the clear heavy-hole character of the E_C band. This observation demonstrates that the E_C band involves states exhibiting an optical anisotropy with respect to the privileged direction.

We now discuss the E_{lh} and the E_C transitions and their implication for the band alignment at the InAs/GaAs interface. The polarization-dependent data shown above reveal (i) the light-hole character of the E_{lh} transition and (ii) the existence of an InAs-related transition with heavy-hole character occurring at higher energy than the light-hole exciton. In the following, we analyze these findings by using a heuristic model of the band alignment between the constituent materials at the interface, the results of which are depicted schematically in Fig. 3. Briefly, we take the valence-band offset determined by x-ray photoemission for the *unstrained* InAs/GaAs heterojunction (70 meV) (Ref. 14) and correct this value for the strain dependence of the band structure of InAs.[15] The hydrostatic component of the biaxial compressive strain is shared among the conduction band and the average valence-band multiplet accord-

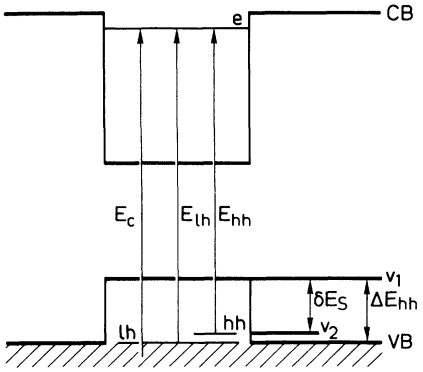

FIG. 3. Illustrative scheme of the real-space band alignment at the InAs/GaAs heterointerface obtained by the simple model described in the text. The potential wells formed in the conduction band (CB) and valence band (VB) are indicated by heavy horizontal lines. The compressive biaxial strain of the InAs unit cell induces a splitting between the two InAs valence-band doublets v_1 and v_2 of $\delta E_S = 247$ meV. For wave vectors perpendicular to the plane, the v_2 and v_1 states at the zone center essentially retain their heavy-hole ($|\frac{3}{2},\pm\frac{3}{2}\rangle$) and light-hole ($|\frac{3}{2},\pm\frac{1}{2}\rangle$) character, respectively. The heavy-hole band offset ΔE_{hh} is formed by the alignment between VB and v_2 and is estimated to be 282 meV. The bound electron (e), heavy- and light-hole (hh and lh) states are depicted by the horizontal lines in the potential step. The shaded area below the GaAs valence-band edge indicates the onset of the valence-band continuum. The vertical arrows illustrate the optical transitions observed in the photoluminescence excitation spectra.

ing to the respective intraband hydrostatic deformation potentials.[16] The shear component of the strain splits the heavy- and light-hole states via the uniaxial shear deformation potential for tetragonal distortions.[16] This splitting δE_S amounts to 247 meV. The heavy-hole band offset ΔE_{hh} is determined to be 282 meV. For this heavy-hole band offset, only one confined heavy-hole state exists. Furthermore, the light-hole band offset is small ($\Delta E_{hh} - \delta E_S = 35$ meV), causing the E_{lh} state to be quasiresonant with the GaAs valence band. Any feature in the spectra higher in energy than the light-hole exciton transition has to involve the valence-band continuum of GaAs.[7]

Most of the recent theoretical work[16,17] and also the only experimental determination[3] of the InAs/GaAs band offset for the strained (coherent) interface set the heavy-hole band offset to about 500 meV. We point out that also with this band offset the light-hole state is still very close (within 8 meV) to the band edge.[6] Furthermore, again only one confined heavy-hole state exists.[6] This larger band offset is thus also consistent with the interpretation of the E_C band.

The observed anisotropy of the E_C band reflects the anisotropic character of one of the participating states. However, the optical anisotropy cannot, in principle, originate from the electron states, as the cell-periodic part of the electron Bloch function is isotropic in space (s-like symmetry), but must be related to the GaAs valence-band states involved in the E_C transition, which are built up from p-type atomic orbitals. The valence-band states participating in the E_C band are thus actually not pure bulk states, but are affected by the InAs potential. This intriguing experimental finding is not yet understood in a quantitative way.

Combining the above considerations, we can deduce the range of band alignments consistent with the observation of both the E_{lh} and the E_C transition. A lower limit for the heavy-hole band offset, of course, is given by the bare presence of the former one, revealing that ΔE_{hh} is certainly larger than the strain-induced splitting of the valence-band doublets (247 meV). An upper limit for ΔE_{hh}, on the other hand, is given by the close vicinity of the E_C band, revealing that the light-hole state is close to the band edge. These considerations set the maximum depth of the potential well in the valence band to about 0.5 eV, above which the light-hole state becomes progressively confined in the potential well.

We finally comment on the apparent discrepancy between our results and the absence of the light-hole feature in the absorption spectra presented in Ref. 2. Owing to the close vicinity of the band edge, the light-hole state is strongly delocalized and thus very sensitive to scattering events. Scattering by, e.g., interface roughness will result in a strong damping of the light-hole exciton and may prevent its observation. Lack of the light-hole transition in optical spectra of this material system hence does not necessarily imply a type-II band alignment, but may rather be related to less well-defined interfaces. As the interface quality degrades with increasing thickness of the InAs layer,[2] a distinct light-hole exciton resonance may only exist for InAs films of single-monolayer thickness.

In summary, we have studied the nature of the optical transitions in InAs/GaAs single-monolayer quantum wells. The heavy- and light-hole character of the transitions was revealed by monitoring the linear polarization of the emitted luminescence and by selective excitation of the states by linearly polarized light. From these experiments, the heavy-hole band offset between GaAs and coherently strained InAs is deduced to lie between 0.3 and 0.5 eV.

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[1]S. Wilke and D. Hennig, Phys. Rev. B **43**, 12 740 (1991).
[2]A. Ksendzov, F. J. Grunthaner, J. L. Liu, D. H. Rich, R. W. Terhune, and B. A. Wilson, Phys. Rev. B **43**, 14 574 (1991).
[3]K. Hirakawa, Y. Hashimoto, K. Harada, and T. Ikoma, Phys. Rev. B **44**, 1734 (1991).
[4]O. Brandt, L. Tapfer, R. Cingolani, K. Ploog, M. Hohenstein, and F. Phillipp, Phys. Rev. B **41**, 12 599 (1990).
[5]R. Cingolani, O. Brandt, L. Tapfer, G. Scamarcio, G. C. La Rocca, Phys. Rev. B **42**, 3209 (1990); O. Brandt, H. Lage, and K. Ploog, *ibid.* **43**, 14 285 (1991).

[6]A simple particle-in-the-box scheme is applied for obtaining the energy positions of the bound electron and hole states. The integrated In coverage of 1.2 ML is interpreted as the average quantum well thickness, meaning that the lateral distance of atomic steps occurring at the top of the continuous InAs monolayer is assumed to be less than the exciton Bohr radius (15 nm). For both the GaAs matrix and the InAs layer we use the effective masses of GaAs, since the wave functions of the bound states extend entirely into the matrix (see Ref. 5). The total band-gap discontinuity is distributed among the conduction and valence band according to a ratio of 71:29 (see Fig. 3 and the corresponding discussion in the text).

[7]O. Brandt, R. Cingolani, H. Lage, G. Scamarcio, L. Tapfer, and K. Ploog, Phys. Rev. B **42**, 11 396 (1990).

[8]J. S. Weiner, D. S. Chemla, D. A. B. Miller, H. A. Haus, A. C. Gossard, W. Wiegmann, and C. A. Burrus, Appl. Phys. Lett. **47**, 664 (1985).

[9]K. Fujiwara, N. Tsukada, T. Nakayama, and T. Nishino, Appl. Phys. Lett. **51**, 1717 (1987).

[10]Y.-C. Chang and J. N. Schulman, Phys. Rev. B **31**, 2069 (1985).

[11]L. C. Andreani and A. Pasquarello, Phys. Rev. B **42**, 8928 (1990).

[12]A waveguide structure has to be used for a meaningful determination of the absolute oscillator strength of the transitions.

[13]J.-Y. Marzin, M. N. Charasse, and B. Sermage, Phys. Rev. B **31**, 8298 (1985).

[14]R. S. Bauer and G. Margaritondo, Phys. Today **40**, 27 (1987), and references therein.

[15]F. H. Pollak and M. Cardona, Phys. Rev. **172**, 816 (1968).

[16]C. G. van der Walle, Phys. Rev. B **39**, 1871 (1989).

[17]M. Cardona and N. E. Christensen, Phys. Rev. B **35**, 6182 (1987); C. Priester, G. Allan, and M. Lanoo, *ibid.* **38**, 9870 (1988).