Optical determinations of energy-band dispersion curves in novel compound semiconductor materials

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Abstract

We present magnetoluminescence data which provides a quantitative measure of the energy-band dispersion curves of novel compound semiconductor optoelectronic materials. Data for a n-type strained-layer InGaAs/GaAs (quantum-well width ~ 8 nm) and a n-type 4.5-nm-wide GaAs/AlGaAs lattice-matched single-quantum well are presented. We find that the conduction-bands are almost parabolic, with a mass of about $0.068m_0$ for InGaAs/GaAs and $0.085m_0$ for the GaAs/AlGaAs structure. The valence-bands are nonparabolic with wave vector dependent in-plane valence-band masses varying from about $0.1m_0$ at zone center to about $0.3m_0$ for 20 meV energies.

Keywords: semiconductor quantum wells, electronic structure, magnetoluminescence, photoluminescence characterization.

1. Introduction

The semiconductor quantum-well laser is the principle component for optoelectronic applications. The need for custom laser wavelengths, higher laser powers, and better beam quality or control has led to many different laser diode designs and configurations. The most common combination for semiconductor laser materials for these devices have been GaAs and AlGaAs. The desirability for light-hole valence-band masses for lasers, LED's, or other high-speed devices has been well documented.^{1,2}

Light-hole valence-band masses have been mainly achieved in layered semiconductor structures by the introduction of compressive biaxial strain in the active quantum layers. The biaxial strain is achieved by growing layered structures from materials with differing lattice constants, e.g., layers of $\text{In}_x\text{Ga}_{1-x}\text{As}$ and GaAs. However, because of level crossing repulsion between the in-plane heavy and light-hole valence-band states, the resulting in-plane valence-band energy dispersion curves (and masses) are nonparabolic.

For lattice-matched quantum well devices, the same conditions are true because of the quantum confinement splitting of the heavy and light-hole valence bands. For these structures, the in-plane heavy-hole light-hole mixing and hence valence-band nonparabolicity occurs at small values of the wave vector. For the devices based upon GaAs and AlGaAs, quantum confinement can be an effective method to alter the energy difference ΔE_{HL} between the heavy and light-hole valence bands. However, for wide (~ 15 nm) GaAs/AlGaAs structures, ΔE_{HL} is small ($\leq 5\text{meV}$) and thus the amount of heavy-hole light-hole mixing

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is large. For these wide quantum well structures, the in-plane valence-band ground state energy is also "heavy", i.e., for Fermi energies $E_f > 2$ meV, the valence-band mass $m_v \approx 0.35m_0$. By reducing the quantum-well width, the energy difference ΔE_{HL} can be increased by quantum confinement. For GaAs/AlGaAs lattice-matched quantum wells, a maximum $\Delta E_{HL} \approx 30$ meV occurs near a quantum-well width of about 4.5 nm.

Valence-band nonparabolicity, for lattice-matched or strained-layer optoelectronic devices, means that with increasing carrier densities the valence-band mass also increases, changing lasing thresholds, modulation frequency response, magneto-optic parameters, etc. Thus a laser structure which performs satisfactory at low power levels, i.e., low-carrier densities, may not perform as expected, under high power operation. Furthermore, device modeling codes predicting the optical response function should take into account any effects which can be attributed to valence-band mixing effects. As modulation frequencies or laser-power requirements increase, a knowledge of the energy-band dispersion curves is mandatory if these design criteria are to be modeled or achieved.

Recently, it was demonstrated that both the conduction-band and valence-band energy dispersion curves of modulation doped strained-single-quantum-well (SSQW)³⁻⁶ and lattice-matched single-quantum-well (LMSQW)⁷ structures can be simultaneously determined by magnetoluminescence measurements from a single sample. These papers discuss the importance of the energy difference ΔE_{HL} between the heavy and light-hole valence-bands in determining the degree of mixing between these two valence bands. Large values for ΔE_{HL} not only give rise to the smallest ground-state in-plane light-hole masses, but also reduce the valence-band nonparabolicity.

In this paper, we present results of magnetoluminescence measurements of band structure dispersion curves for two representative modulation-doped n-type structures, an $\text{In}_{0.20}\text{Ga}_{0.80}\text{As}/\text{GaAs}$ SSQW, and a $\text{GaAs}/\text{Al}_{0.25}\text{Ga}_{0.75}\text{As}$ LMSQW.

2. Experimental

The samples were prepared using molecular beam epitaxy. The sample barrier materials were silicon-doped, with a spacing of about 8 nm between the ~3-nm-wide silicon-modulation layer and the quantum well. The SSQW structure (#BC042) consisted of a single 8-nm-wide $\text{In}_{0.20}\text{Ga}_{0.80}\text{As}$ strained quantum-well and unstrained GaAs barriers. Here, the energy difference ΔE_{HL} between the heavy and light-hole valence-bands, which includes contributions from both strain effects and quantum confinement, is about 60 meV. The two-dimensional carrier concentration N_{2d} and mobility μ at $T = 4$ K are respectively $5 \times 10^{11} \text{ cm}^{-2}$ and $1.2 \times 10^4 \text{ cm}^2/\text{Vsec}$. The GaAs/AlGaAs LMSQW structure (#G0260) has a 4.5-nm-wide GaAs quantum well with lattice-matched (unstrained) $\text{Al}_{0.25}\text{Ga}_{0.75}\text{As}$ barriers, and as previously mentioned, the heavy and light-hole valence-band energy difference $\Delta E_{HL} \approx 30$ meV. For the LMSQW sample at 4 K, $N_{2d} = 6.6 \times 10^{11} \text{ cm}^{-2}$ and $\mu = 2.2 \times 10^4 \text{ cm}^2/\text{Vsec}$. The magnetoluminescence measurements were made in the temperature range of 1.4 and 76 K, and the magnetic fields varied between 0 and 14 T. The sample was attached to the end of a 100 μm -core-diameter optical fiber. The luminescence measurements were made with an Argon-ion laser operating at 514.5 nm. The laser was injected into the optical fiber by means of a optical beam-splitter and the returning photoluminescence signal was directed to the optical monochromator and CAMAC-based data acquisition system.⁸ Typical laser power densities to the sample were less than $1 \text{ W}/\text{cm}^2$, therefore the photo-induced two-dimensional density of electrons and holes are very small, less than $1 \times 10^{10} \text{ cm}^{-2}$. Two types of photomultiplier tubes were used during the

course of the experiments, an S1-response RCA 7102 and an InGaAs-enhanced Hamamatsu R3310-02. The direction of the applied magnetic field is parallel to the growth direction, i.e., the resulting Landau orbits are in the plane of the quantum well. With this geometry, all measurements concerning the conduction and valence-band dispersion curves and masses refer to their *in-plane* values.

3. Discussion

A charged particle, in the presence of a magnetic field B , forms a series of quantized states, called Landau levels, with the quantized energy levels given by (cgs units) $\hbar\omega = (n + 1/2)(e\hbar B/mc)$, where n is the Landau index, e is the charge of an electron, $\hbar$ is Planks' constant over 2π , m is the mass of the carrier, and c is the velocity of light. The distribution function for a degenerate two-dimensional electron gas (conduction-band states for a n-type material) is determined by the Fermi-Dirac statistics, but because of the very small number of photo-induced two-dimensional hole states, the distribution for the valence-band holes are governed by Maxwell-Boltzmann statistics. At high temperatures, where kT is much larger than $\hbar\omega$, the $n_v = 0, 1, 2, 3, \dots$ valence-band Landau levels are populated and all magnetoluminescence transitions between the n_c and n_v Landau levels are *allowed*, obeying the $\delta n_{cv} \equiv (n_c - n_v) = 0$ selection rule. For these high temperatures, the interband luminescence transition energy E is given by

$$E(n) = E_{\text{gap}} + \left(n + \frac{1}{2}\right) \left(\frac{e\hbar B}{\mu c} \right), \quad (1)$$

where E_{gap} is the bandgap energy, μ is the reduced mass ($\mu^{-1} = m_c^{-1} + m_v^{-1}$) where m_c and m_v are respectively the conduction or valence-band effective masses expressed in terms of the free electron mass m_0 . In this paper we assume that the sample temperature is always less than the Fermi temperature, $kT_f = E_f$, of the degenerate two-dimensional electron gas.

A schematic showing these transitions for a n-type structure is shown in the right side of Fig. 1. The Fermi energy E_f , the bandgap energy E_{gap} , and the Landau level indices are also indicated in the figure. For large magnetic fields and low temperatures, i.e., $kT \ll \hbar\omega_v$, only the $n_v = 0$ valence-band Landau level is populated. Here, the transition between the $n_c = 0$ and $n_v = 0$ Landau level is allowed while transitions between the higher energy conduction-band Landau levels $n_c = 1, 2, 3, \dots$ and the $n_v = 0$ ground state valence-band Landau level are zeroth-order forbidden, but are observable due to higher order processes.^{9,10} The left side of Fig. 1 shows the energy level diagram for this case.

Two magnetoluminescence spectra ($B = 5.75$ T) at 4.2 and 76 K are shown in Fig. 2. The origins of the energy axes of the two spectra have been adjusted in order to allow a comparison of the spacing between the magnetoluminescence transitions. For 4.2 K, $E_{\text{gap}} = 1330.1$ meV while at 76 K, $E_{\text{gap}} = 1321.1$ meV. The indices $n_c \rightarrow n_v$ for each peak are identified in the figure. As can be seen in Fig. 2, all observed transitions for the 76 K data are "allowed", i.e., $\delta n_{cv} = 0$, while for the 4.2 K data, the only zeroth-order allowed transition is the $0 \rightarrow 0$ transition. Because of the Maxwell-Boltzmann statistics for the holes, the energy dependence of the 76 K magnetoluminescence peak-amplitudes are also governed by Maxwell-Boltzmann distribution function. An analysis of the energy dependence of the peak-amplitudes for this spectrum yields a temperature of about 80 K, which is in good agreement with the expected temperature of liquid nitrogen. A theoretical treatment of the energy dependence of the amplitudes of the zeroth-order forbidden transitions has been performed by Lyo¹⁰ and data shown in Fig. 2 is in good agreement with his calculation.

Figure 1. A schematic representation for the Landau levels in a n-type quantum well. The left side is the low temperature case, $\delta E_V \gg kT$, and the right side is the high temperature condition $\delta E_V \ll kT$.

Figure 2. Magnetoluminescence spectra ($B = 5.75\text{ T}$) at two temperatures, 4 and 76 K, for an n-type InGaAs/GaAs SSQW showing the allowed ($\delta n_{cv} = 0$) and the zeroth-order forbidden ($\delta n_{cv} \neq 0$) transitions. The energy axes of the two spectra have been shifted in order to compare the energy difference between the magnetoluminescence transitions.

As is obvious from the left-hand side of Fig. 1, where $kT \ll \hbar\omega_v$, the energy difference δE between the $E(n_c)$ and $E(n_c-1)$ magnetoluminescence peaks depends only on the conduction-band cyclotron energy $\hbar\omega_c$. Setting $n_v = 0$ and using (1), the magnetoluminescence transition energy $E(n_c)$ is given by in terms of the respective conduction and valence-band cyclotron energies as

$$E(n_c) = E_{\text{gap}} + \left(n_c + \frac{1}{2}\right)\hbar\omega_c + \frac{1}{2}\hbar\omega_v, \quad (2)$$

where $n_c = 0, 1, 2, 3, \dots$. Thus, utilizing low temperatures and measuring the energy differences as a function of magnetic field, (2) provides a method for obtaining all the information about the conduction band energy dispersion curves

Figure 3 shows the derived low-temperature-derived conduction-band dispersion curve for the InGaAs/GaAs SSQW. The method (and justification) used relating the magnetic field B to the wavevector k has been adequately discussed.^{11,12} The maximum value of the wavevector k of about 3% of the Brillouin zone is determined by the conduction band Fermi energy E_f of about 35 meV. The minimum wavevector determinations are limited by our ability to distinguish magnetoluminescence peaks at low magnetic fields. The conduction-band dispersion curve is found to have a small nonparabolic correction with the zone center conduction-band mass $m_c \sim 0.067m_0$ and at the Fermi energy $m_c \sim 0.069m_0$. Non-parabolic effects upon the cyclotron resonance measured conduction-band masses have been discussed in detail^{13,14} and the magnetoluminescence results presented here are in agreement with that derived by conduction-band cyclotron resonance.

Figure 3. Magnetoluminescence determined conduction-band dispersion curve for SSQW BC042. The line through the data is a parabolic ($m_c = 0.067m_0$) best-fit curve.

Figure 4. Valence-band dispersion curve for SSQW BC042. The lines through the data are best-fit of the nonparabolic curve given by (3). A $\mathbf{k} \cdot \mathbf{p}$ calculation is also indicated in the figure.

With a knowledge of the conduction-band dispersion curve, we can, using (1), derive the valence-band dispersion curve by performing 76-K magnetoluminescence measurements and analyzing the zeroth-order allowed transition peaks as a function of magnetic field. The valence-band data is shown in Fig. 4 and it is evident that the dispersion curve is nonparabolic. For the range of data shown in the figure, the valence-band effective mass m_v for the SSQW structure BC042 varies between a zone-center $0.11m_0$ to about $0.3m_0$ at $E(k) \sim 20$ meV. The solid line through the valence-band data is a best-fit nonparabolic curve to the data of the form

$$E(k) = \left(\frac{\hbar^2 k^2}{2m_v^0} \right) \left[1 - \frac{K}{E_{gap}} \left(\frac{\hbar^2 k^2}{2m_v^0} \right) \right], \quad (3)$$

where $m_v^0 = 0.11m_0$ is the zone center valence-band mass and $K \sim 16$ which is about an order of magnitude larger than measured for the conduction band.¹¹ A comparison with a $\mathbf{k} \cdot \mathbf{p}$ calculation for the valence-band dispersion, which includes strain and valence-band mixing, is also shown in Fig. 4. The agreement between the $\mathbf{k} \cdot \mathbf{p}$ calculation and the data is reasonable in view of the fact that the $\mathbf{k} \cdot \mathbf{p}$ calculation did not include the contributions from the conduction band or split-off valence band. Magnetoluminescence determined valence-band dispersion curves for other samples with varying indium concentration and hence, with differing values for the heavy-hole light-hole valence band splitting ΔE_{HL} , confirm that as ΔE_{HL} increases the valence-bands become more parabolic and that the ground state in-plane valence-band mass remains relatively constant.³

The conduction and valence-band dispersion curves for the GaAs/AlGaAs LMSQW sample were also determined by both low and high temperature magnetoluminescence measurements. The results of the magnetoluminescence data analyses are shown in Figs. 5 and 6, where comparisons with the SSQW

Figure 5. Conduction-band dispersion curve for SSQW BC042 and LMSQW G0260. The lines through the data are parabolic best-fit curve to the data. The conduction-band masses for the SSQW and the LMSQW are respectively $0.067m_0$ and $0.085m_0$.

Figure 6. Comparison of the valence-band dispersion curves for SSQW BC042 and LMSQW G0260. The two lines through the data are best-fit of the nonparabolic curve given by (3).

sample are presented. The conduction-band dispersion curve (Fig. 5) for the LMSQW structure is again nearly parabolic with respective zone-center conduction-band masses of $m_c \approx 0.085m_0$. The increase to m_c for the LMSQW conduction-band mass between the bulk GaAs $m_c \approx 0.065m_0$ to $0.085m_0$ is caused by the quantum confinement energy, i.e., increased bandgap energy.^{13,14} In order to verify these masses on our samples, the conduction-band masses for the SSQW and LMSQW structures were measured by far infrared cyclotron resonance techniques with the result that at their respective Fermi energies, $m_c \approx 0.069m_0$ for the SSQW sample and $m_c \approx 0.082m_0$ for the LMSQW structure in excellent agreement with the aforementioned magnetoluminescence data.

The LMSQW valence-band dispersion curve can be derived in the same manner as before, e.g., with a knowledge of the conduction-band dispersion curve the magnetic field dependent valence-band energy can be inferred from the high temperature (76 K) magnetoluminescence data using (1). The resulting valence-band dispersion curve for LMSQW sample is shown in Fig. 6. For the range of data shown in the figure, the valence-band mass m_v for the SSQW structure varied between $0.11m_0$ and $0.22m_0$. However for the LMSQW sample at small wavevectors, $m_v \approx 0.15m_0$ and for larger wavevectors, e.g., $E_v \approx 10$ meV, $m_v \approx 0.35m_0$. As previously mentioned, the heavy-hole light-hole energy separation ΔE_{HL} is about 60 meV for the SSQW structure and about 30 meV for the LMSQW sample. It is obvious from Fig. 6, that the valence-band dispersion curve for the SSQW structure is more parabolic than that of the LMSQW sample and this result is due to the increased heavy-hole light-hole energy separation ΔE_{LH} in the SSQW sample.

4. Conclusions

In conclusion, we have shown that magnetoluminescence measurement techniques is a powerful tool for performing single-sample simultaneous measurements of both the conduction and valence-band dispersion curves (and masses). We showed that for quantum-well devices the amount of heavy-hole light-hole splitting is the determining factor for valence-band nonparabolicities and that the larger the heavy-hole light-hole energy difference, the smaller the valence-band nonparabolicity. The experimental agreement with $k \cdot p$ calculations for the valence-band energies are good and with these dispersion curves, it is possible to model not only optoelectronic behavior, but also predict electronic phenomena which rely upon these dispersion curves. In particular, the amount of valence-band nonparabolicity can now be experimentally quantified and predictions for new material systems may be possible.

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